JAMES   ROBERT   HENDERSON

QUANTUM   MANY-BODY   THEORY

ABSTRACT

This thesis is a collection of theoretical investigations into different aspects of the broad subject of quantum many-body theory. The results are grouped into three main parts, which in turn are divided into separate self-contained sections. Some of the work is presented in the form of published papers and papers that have been submitted for publication.

The first section of Part A introduces some of the concepts involved in many-body problems, by developing methods to evaluate expectation values of the form $\langle f(r_{12}) \rangle$. In the rest of Part A I consider collective excitations of finite quantum systems. The calculations are confined to nuclei because the results can then be compared with the extensive investigations that have been made into collective nuclear modes. In Section AII, wavefunctions are proposed for rotational excitations of even-even nuclei. Both isoscalar and isovector nuclear modes are discussed. In particular, the $|2,M\rangle$ isoscalar states are investigated for both spherical and deformed even-even nuclei, and the simplest isovector wavefunction is shown to give a good description of the giant dipole resonance. In section AIII wavefunctions are proposed for compressional vibrational states of spherical nuclei. Section AIV discusses sum rules for nuclear transitions of a given electric multipolarity. It is found that the 2^+ and 1^- states investigated in section AII and all but one of the vibrational states discussed in AIII, each exhaust a large part of the appropriate sum rule.

In Part B I consider the problem of how to describe flow in quantum fluids. In particular, we want to be able to identify the

physical motion represented by any given many-body wavefunction.

Section BI derives a quantum mechanical velocity field for a

many-body system, paying special attention to the need for a quantum

continuity equation. It is found that when the wavefunction has the

usual time dependence $e^{-i\omega t}$, that the quantum velocity formula

averages over all oscillatory motion, so that much of the physical

nature of the flow field is lost. In section BII a particular wave-

function is proposed to represent the quantum excitation corresponding

to any given potential flow field. The results obtained by considering

specific examples are very encouraging.

In Part C I investigate the properties of surfaces. Section CI

presents a theoretical description of the tension, energy and thick-

ness of a classical liquid-vapour interface. In section CII the

classical results are extended to describe the surface of a quantum

system, namely superfluid helium four. Problems occur for the

quantum system if the correlations arising from the zero-point-motion

of the phonon modes are included in the ground state wavefunction.

Finally, in section CIII I discuss generalized virial theorems that

give the change in the free energy of a system undergoing an

infinitesimal deformation. For example, a particular deformation

gives the expression used in CII, for the surface tension of a plane

quantum surface.

CONTENTS

PAPERS

1) A collective rotational state of spherical nuclei, Aust. J. Phys.,
    (1976), 29, 363., [with J. Lekner]

2) The giant dipole resonance, to be submitted to Nucl. Phys. A.,
    [with J. Lekner]

3) Theory of surface excitations of liquid helium four, to be published in J. Phys. C : Solid st. Phys., [with J. Lekner]

4) Surface tension and energy of a classical liquid-vapour interface, to be published in Mol. Phys., [with J. Lekner]

5) The surface of liquid He^4 , based on the idea that $\prod_{i<j} f(r_{ij})$ describes a droplet, submitted to J. Phys. C : Solid st. Phys., [with J. Lekner]

NOTE : References are denoted by [ ] . Each section has a separate set of references, listed on the last page.

INTRODUCTION

There are two main themes which form the basis of the work reported in this thesis. Firstly, the importance of density correlation functions in many-body theory and secondly, the wish to understand the physical processes described by given many-body wavefunctions. It transpired (see below) that both of these considerations led me to investigate properties of classical and quantum liquids.

The mathematical formulations of a large number of problems in many-particle physics involve the use of density correlation functions. In particular, if an adequate description of the system can be given by neglecting three(or more)-particle interactions then it is sufficient to consider only the single-particle density $n(1)$ and the pair correlation function $g(12)$. Section AI begins with the definitions of these correlation functions, from which it follows that if the interparticle interactions are dependent only on the distance between pairs of particles (r), then the pair correlation function reduces to the simple radial form $g(r)$. So, the sytems most readily described by the many-body techniques under investigation are simple liquids, rather than solids or polar liquids, where the particles have preferred orientation towards one another.

The original impetus for investigating properties described by density correlation functions arose out of the progress reported in section AI, in simplifying double integrals over a spherical drop, in which the integrands involve the pair density. At the same time my supervisor (Dr J Lekner) had recently proposed a set of many-body wavefunctions suitable for describing collective excitations of self-bound quantum systems, and it was found that the results of AI are helpful in evaluating the energies of these proposed excitations. Sections AII-AIV report the investigation into the properties of these wavefunctions. The discussion is confined to excitations of nuclei because the results can be compared with the

large amount of experimental data on excited states of nuclei, in contrast
for example, with the lack of data on collective states of liquid helium
droplets. The work of Part A is dominated by attempts to understand the
physical processes described by the various wavefunctions considered and
the effect of pair correlations is at first argued to be small enough to
be neglected. However, by the time of completion of the thesis it had
become apparent that the behaviour of the nucleon pair correlation functions
(especially that due to Fermi statistics) has a crucial effect on the
calculated energies of the states. In particular, by including the effect
of pair correlations the variation of the energy with nucleon number A
changes from $A^{-2/3}$ to $A^{-1/3}$.

Part B, which arose out of a need to identify the flow fields corresp-
onding to the many-body excitations discussed in Part A, is a general
discussion on flow in quantum liquids. Although the main emphasis is on
the physics of wavefunctions the importance of density correlation functions
is again highlighted in section BII, when investigating surface excitations
of liquid helium four. Namely, it is found that the long range behaviour
of the pair correlation function determines the long wavelength (low energy)
part of the dispersion relation, in direct analogy with the situation for
bulk excitations.

Part C is concerned with the statistical mechanics of liquid surfaces,
and hence there arises the question of the behaviour of density correlation
functions in inhomogeneous systems. In section CI I discuss the liquid-
vapour interface of a simple classical flúid. The reader is referred to a
paper reprinted in this thesis, in which general expressions are derived
for the surface tension σ and the surface energy ϵ . The exact evaluation
of these expressions will require complete knowledge of the density profile
and the pair correlation function of the inhomogeneous system. Since this
is not yet available further analytic results are obtained by choosing
simple physical density profiles, and by making an approximation for the

pair correlation function, which it is argued should be reasonable at least near the triple point and in the critical region. From the results for σ and ϵ it is also possible to estimate the surface thickness.

In section CII the methods developed in CI are extended to describe the simplest corresponding quantum system, namely the surface of superfluid helium four. In particular the consequences of taking the zeroth order approximation for the ground state wavefunction are investigated. With this simple ground state the mathematical techniques used in the classical system are all that is required. Provided the long range phonon correlations are neglected, good results are obtained for σ, ϵ and the surface thickness. The problems that occur when these correlations are included led to a detailed investigation into the effect of the zero-point motion of phonon excitations on the ground state wavefunction. The work reported in section CIII arose from considering generalisations of the derivation of the expressions for σ, used in CI and CII.

The work in Parts A and B is concerned mainly with the second theme, that is, the wish to understand the physical processes described by certain many-body wavefunctions. The wavefunctions discussed in Part A are used to investigate collective excitations of nuclei, due to the availability of experimental data. The properties of a variety of wavefunctions are extensively investigated, in particular the energies and the contributions to the relevent sum rules are evaluated. It is worth pointing out that the methods of Part A treat the nucleus as a quantum fluid. That is, the wavefunctions are thought of as describing fluid flow and any shell structure effects are neglected. (The concept of treating the nucleus as a liquid drop when investigating collective excitations has been used previously in the "classical" liquid-drop model, which gives its best results in the description of giant resonances.) It was this flow analogy to collective nuclear excitations that led to the general investigation, reported in Part B, into the velocity fields corresponding to different types of many-

body wavefunctions. Many examples are considered, including wavefunctions discussed in Part A. In particular, the results of BII suggest that most of the ∧ wavefunctions correspond to what nuclear physicists call giant resonances, which in turn supports the conclusions of section AIV where the excitations are shown to exhaust large parts of the relevent sum rules. These results forced a revision of the original calculations of the energies, which were based on neglecting pair correlations. It is found that the inclusion of nucleon pair correlations crucially effects the calculated energies, and in particular that they are needed to obtain the correct variation of energy with nucleon number from the giant dipole wavefunction. Thus an understanding of the physical nature of the wavefunctions led to imformation about the importance of pair correlations in nuclei.

To sum up - this thesis can be looked at as a study in properties of liquids. Particular emphasis is placed on the physical interpretation of many-body wavefunctions describing fluid flow, and on the properties of density correlation functions in homogeneous and inhomogeneous liquids. Future work on problems raised in this thesis will probably rely heavily on a better understanding of the behaviour of pair correlation functions and density profiles in finite systems(for Part A) and in inhomogeneous systems (for Part C).

Finally, the reader will see from the papers reprinted at the end of the thesis that the work reported here is the result of an extensive collaboration between myself and my supervisor, Dr John Lekner. In particular, the published work is in all cases close to a 50-50 contribution from each of us. The unpublished part of the thesis is my own work and so represents my own ideas. However, it is inevitable that much of the motivation and technical progress at least partly arose out of the many enjoyable and stimulating discussions I have had with John Lekner over the past few years.

PART   A

FINITE   QUANTUM   SYSTEMS

ABSTRACT

The first section introduces some of the concepts involved in
many-body problems, by developing methods to evaluate expectation
values of the form $\langle f(r_{12}) \rangle$. The rest of the sections consider
collective excitations of finite quantum systems. The calculations
are confined to nuclei because the results can then be compared with
the extensive investigations that have been made into collective
nuclear modes. In Section II, wavefunctions are proposed for
rotational excitations of even-even nuclei. Both isoscalar and
isovector nuclear modes are discussed. In particular, the $|2,M\rangle$
isoscalar states are investigated for both spherical and deformed
even-even nuclei, and the simplest isovector wavefunction is shown
to give a good description of the giant dipole resonance. In
section III wavefunctions are proposed for compressional vibrational
states of spherical nuclei. Section IV discusses sum rules for
nuclear transitions of a given electric multipolarity. It is found
that the 2^+ and 1^- states investigated in section II and all but one
of the vibrational states discussed in III, each exhaust a large
part of the appropriate sum rule.

CONTENTS

I.   PAIR CORRELATIONS IN SELF BOUND SPHERICAL SYSTEMS.

In many-body physics it is often necessary to calculate integrals of the form

$$\int d1\, d2\, n(12) f(r_{12}) \tag{1}$$

where $f(r_{12})$ is a function of $r_{12} = |r_1 - r_2|$ and $n(12)$ is the two particle ground state density correlation function, defined by $n(1...S)$ for $S = 2$ where

$$n(1...S) = \frac{N!}{(N-S)!} \frac{\int d(S+1)....dN\, \Phi^2}{\int d1.....dN\, \Phi^2} \tag{2}$$

$\Phi(1....N)$ is the ground state wavefunction of an N- particle system. For example, the expectation value of the Hamiltonian of an N- particle Bose system

$$H = -\frac{\hbar^2}{2m} \sum_{i=1}^{N} \nabla_i^2 + \sum\sum_{1 \le i < j \le N} \upsilon(r_{ij}) \qquad , \tag{3}$$

when the wavefunction is of the form

$$\Phi(1...N) = \exp\left[\frac{1}{2} \sum_{i<j}^{N} \phi(r_{ij})\right] \qquad , \tag{4}$$

is of the form (1) with, [1],

$$f(r_{12}) = \frac{1}{2}\left[-\frac{\hbar^2}{4m} \nabla_{12}^2 \phi(r_{12}) + \upsilon(r_{12})\right] \tag{5}$$

To evaluate integrals of the type (1) we use an approximation for $n(12)$, namely

$$n(12) = n(1)\, n(2)\, g(r_{12}) \tag{6}$$

where $g(r_{12})$ is known as the pair correlation function. By noting that in the ground state, $n(1) dr_1$ is the probability of finding a particle in volume dr_1 at r_1 , $n(12) dr_1 dr_2$ is the probability of finding two particles, one in dr_1 at r_1 and the other in dr_2 at r_2 , then $n(2) g(r_{12}) dr_2$ is seen to be the probability in the ground state, of finding a particle in dr_2 about r_2 , if we already know there is a particle at r_1 . In Appendix AI(1) we discuss the weak correlation limit of $g(r)$, ($g(r)$ = constant for all r).

If we restrict ourselves to spherical systems so that $n(1)$ becomes $n(r)$, r the distance to the centre, then we can use bipolar coordinates. ([7] page 203.)

Let $|\underline{r}_1| = r, |\underline{r}_2| = s, r_{12} = t$
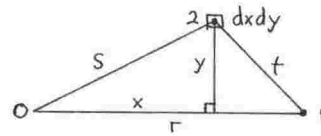
$$ \tag{7} $$

Then the Jacobian of the transformation $\left| \dfrac{\partial(s,t)}{\partial(x,y)} \right|$ yields

$$ \underline{dr_2} = 2\pi y \, dx \, dy = \left(\frac{2\pi}{r}\right) s.t. \, ds.dt \tag{8} $$

Hence

$$ \underline{d1}\,\underline{d2} = 8\pi^2 \, dr\,r \, ds\,s \, dt\,t \tag{9} $$

with $\qquad 0 \leq r, s \leq \infty \qquad$ and $\qquad |r-s| \leq t \leq r+s$

Using these coordinates we have

$$ \int \underline{d1}\,\underline{d2}\, n(12)\, f(r_{12}) = \int \underline{d1}\, n(1) \int \underline{d2}\, n(2)\, g(r_{12})\, f(r_{12}) $$
$$ = 8\pi^2 \int_0^\infty dr\,r\,n(r) \int_0^\infty ds\,s\,n(s) \int_{|r-s|}^{r+s} dt\,t\,h(t) $$

where $\quad h(t) = g(t) f(t)$

interchange s with t $\quad = \quad 8\pi^2 \int_0^\infty dr\,r\,n(r) \int_0^\infty dt\,t\,h(t) \int_{|r-t|}^{r+t} ds\,s\,n(s)$

interchange r with t $\quad = \quad 8\pi^2 \int_0^\infty dr\,r\,h(r) \int_0^\infty dt\,t\,n(t) \int_{|r-t|}^{r+t} ds\,s\,n(s)$ $\tag{10}$

Therefore interchanging s with t again, we have

$$ \int \underline{d1}\,\underline{d2}\, n(12)\, f(r_{12}) = 8\pi^2 \int_0^\infty dr\,r^2\,h(r)\,p(r) \tag{11a} $$

where $\quad h(r) = g(r) f(r) \quad$ and $\quad p(r) = \frac{1}{r} \int_0^\infty ds\,s\,n(s) \int_{|r-s|}^{r+s} dt\,t\,n(t) \tag{11b}$

From (2) and putting $f = 1$ in (11a) we have the normalization

$$ 4\pi \int_0^\infty dr\,r^2\,g(r)\,p(r) = \frac{N(N-1)}{2\pi} \tag{12} $$

As a first approximation we could use the cutoff density

$$ n(s) = n_0 \;,\; s \leq R \quad ; \quad n_0 = \frac{N}{V} \;,\; V = \frac{4}{3}\pi R^3 \tag{13} $$
$$ = 0 \;,\; s > R $$

Then as is shown in Appendix AI(2)

$$P_0(r) = \frac{n_0^2}{r} \int_0^R ds\, s \int_{|r-s|}^{r+s} dt\, t \qquad , \quad t \le R \qquad (14)$$

$$= n_0^2 \frac{2}{3} R^3 \left[1 - \frac{3}{2}\left(\frac{r}{2R}\right) + \frac{1}{2}\left(\frac{r}{2R}\right)^3 \right] \quad , \quad 0 \le r \le 2R \qquad (15)$$

$$= 0 \quad , \quad r > 2R$$

Therefore in the approximation of a cutoff density, where the self bound many body system becomes a sphere of radius R,

$$\int d\underline{1}\, d\underline{2}\, n(12) f(r_{12}) = \frac{N^2}{V} \int d\underline{r}\, h(r) \left[1 - \frac{3}{2}\left(\frac{r}{2R}\right) + \frac{1}{2}\left(\frac{r}{2R}\right)^3 \right] \qquad (16)$$

where $h(r) = g(r) f(r)$, $\int d\underline{r} = 4\pi \int_0^{2R} dr\, r^2$

We can get a conceptual feel for $P_0(r)$ by noting that the volume common to two sharp edged spheres of radius R, whose centres are separated by distance r
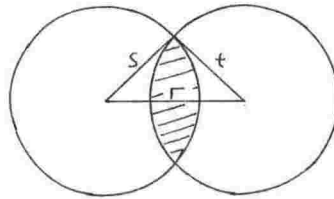
is, using (8)

$$\frac{2\pi}{r} \int_{r-R}^R ds\, s \int_{r-s}^R dt\, t$$

$$= \frac{4\pi}{3} R^3 \left[1 - \frac{3}{2}\left(\frac{r}{2R}\right) + \frac{1}{2}\left(\frac{r}{2R}\right)^3 \right] = \frac{2\pi}{n_0^2} P_0(r) \quad \text{(See Appendix AI(2))} \qquad (17)$$

Similarly the common volume of two overlapping fuzzy spheres (non zero surface width) each described by the general radial density $n(s)$, is

$$\frac{2\pi}{n_0^2} P(r) \qquad (18)$$

In some problems the weak correlation limit $g(r) = \frac{(N-1)}{N}$

(see Appendix AI(1) , or use $P_0(r)$ in the normalization condition (12)

for $g(r)$ = constant) may give good results. This weak correlation approximation, [3] , is such that the only correlation between particles is that due to the particles being constrained to move

within the sphere. This might be a good approximation for nuclei as the hard core of the nucleons only occupies about a percent of the total volume. [3]. In this case for a cutoff density

$$\int d\underline{1}\, d\underline{2}\, n(12) f(r_{12}) = N(N-1) \frac{3}{R^3} \int_0^{2R} dr\, r^2 \left[1 - \frac{3}{2}\left(\frac{r}{2R}\right) + \frac{1}{2}\left(\frac{r}{2R}\right)^3\right] f(r) \quad (19)$$

To find $P(r)$ exactly for a non cutoff radial density, we can try introducing the Fourier transform of the density function.

$$n(s) = \frac{1}{(2\pi)^3} \int d\underline{k}\, e^{i\underline{k}\cdot\underline{s}}\, n(\underline{k}) \quad (20)$$

where
$$n(\underline{k}) = \int d\underline{s}\, n(s) e^{-i\underline{k}\cdot\underline{s}} \quad (21)$$

Integrating over the angles we get

$$n(\underline{k}) = \frac{4\pi}{k} \int_0^\infty ds\, s\, n(s) \sin ks \quad (22)$$

It is shown in Appendix AI(3) that (20) and (21) give

$$\int d\underline{1}\, d\underline{2}\, n(12) f(r_{12}) = 8\pi^2 \int_0^\infty dr\, r^2 h(r) P(r) \quad ; \quad h(r) = g(r) f(r)$$

where
$$P(r) = \frac{1}{2\pi} \int d\underline{s}\, n(s)\, n(|\underline{s}+\underline{r}|)$$

$$= \frac{1}{4\pi^3} \cdot \frac{1}{r} \int_0^\infty dk\, k\, n^2(\underline{k}) \sin kr \quad (23)$$

For example, take $n(s)$ to be the cutoff density (13), then from (22),
$$n(\underline{k}) = \frac{4\pi}{k} n_0 \int_0^R ds\, s \sin ks$$

$$= \frac{4\pi}{k} n_0 R^2 j_1(kR) \quad (24)$$

where j_1 is the first order spherical Bessel function, (see Appendix AI(4)). Therefore (23) gives us,

$$P_0(r) = \frac{4}{\pi} n_0^2 R^4 \int_0^\infty dk\, j_1^2(kR)\, j_0(kr) \quad (25)$$

Equating (25) with (15) we see that we have proved

$$\int_0^\infty dx \, J_1^2(x) \, J_0(\alpha x) = \left[1 - \frac{3}{2}\left(\frac{\alpha}{2}\right) + \frac{1}{2}\left(\frac{\alpha}{2}\right)^3 \right] \frac{\pi}{6} \quad , \quad 0 \le \alpha \le 2$$

$$= 0 \quad , \quad \alpha > 2 \tag{26}$$

In general (22) and hence (23) are difficult to evaluate for a reasonable $n(s)$, although the $n(\underline{k})$ for some simple $n(s)'s$ are known from scattering theory where the form factor is of the form (22). $\left[9\right]$. For example a Gaussian $n(s) = n_0 e^{-cs^2}$, leads to a Gaussian $p(r) = \frac{n_0^2 \sqrt{2\pi}}{8 \, c^{3/2}} e^{-\frac{cr^2}{2}}$.

Because the above exact method is too difficult for a reasonable density function that will describe a system with a non zero surface width, we will revert to bipolar coordinates and calculate $p(r)$ for a simple exponential variation of the density, namely

$$n(s) = n_{R_1}(s) + \delta n_{R_1}(s) \tag{27}$$

where $n_{R_1}(s)$ is the cutoff function (13) with R replaced by R_1 and $\delta n_{R_1}(s)$ is

$$\delta n_{R_1}(s) = -\frac{n_0}{2} e^{-R_1/\lambda} e^{s/\lambda} \quad , \quad s < R_1$$

$$= \frac{n_0}{2} e^{R_1/\lambda} e^{-s/\lambda} \quad , \quad s > R_1 \tag{28}$$

This simple density variation has a continuous derivative and describes a surface whose thickness is approximately given by $\left| \frac{n_0}{n'(R_1)} \right| = 2\lambda$. The reason why R_1 and not R appears in (27) and (28), is because we must be careful in defining the position of the surface. The surface $(S = R)$ is chosen so that the total volume

$$V = \frac{1}{n_0} \int \underline{ds} \, n(s) \tag{29}$$

stays the same when $n(s)$ is changed from $n_R(s)$ to (27).

That is,

$$V = \frac{4}{3}\pi R^3 = \frac{1}{n_0}\int ds \left[n_{R_1}(s) + \delta n_{R_1}(s) \right]$$

$$= \frac{4}{3}\pi R_1^3 + \frac{4\pi}{n_0}\int_0^\infty ds\, s^2 \delta n_{R_1}(s)$$

$$= \frac{4}{3}\pi R_1^3 - 2\pi e^{-R_1/\lambda}\int_0^{R_1} ds\, s^2 e^{s/\lambda} + 2\pi e^{R_1/\lambda}\int_{R_1}^\infty ds\, s^2 e^{-s/\lambda}$$

$$= \frac{4}{3}\pi R_1^3 + 8\pi R_1 \lambda^2 \tag{30}$$

From (30) we find,

$$R_1 = R\left(1 - 2\left(\frac{\lambda}{R}\right)^2 + O\left(\frac{\lambda}{R}\right)^4\right) \tag{31}$$

Substituting (27) into (11b) and using (15), we have

$$p(r) = p_0'(r) + p_1(r) + p_2(r) \tag{32}$$

where, $p_0'(r)$ is given by (15) with R replaced by R_1 (33)

$$r p_1(r) = 2\int_0^\infty ds\, s\, \delta n_{R_1}(s)\int_{|r-s|}^{r+s} dt\, t\, n_{R_1}(t) \tag{34}$$

and $$r p_2(r) = \int_0^\infty ds\, s\, \delta n_{R_1}(s)\int_{|r-s|}^{r+s} dt\, t\, \delta n_{R_1}(t) \tag{35}$$

In Appendix AI(5) we show, neglecting terms of order $e^{-R_1/\lambda}$
and assuming that $h(r)$ decays fast enough so that when using $p(r)$
in (11a) we can drop all terms involving $e^{-|r-2R_1|/\lambda}$, that
evaluating (34) and (35) using (28) gives

$$r\, p_1(r) = -2n_0^2 \lambda^2 R_1^2\left[1 - 2\left(\frac{r}{R_1}\right) + \frac{1}{2}\left(\frac{r}{R_1}\right)^2 - e^{-r/\lambda}\right] +$$

$$2n_0^2 \lambda^3 r\, e^{-r/\lambda} - 6n_0^2 \lambda^4\left(1 - e^{-r/\lambda}\right) \quad , \quad 0 \le r \le 2R_1 \tag{36}$$

$$= 0 \quad , \quad r > 2R_1$$

$$r\, p_2(r) = \frac{n_0^2}{2} R_1^2 \lambda r\, e^{-r/\lambda} - \frac{n_0^2}{4}\lambda^4 e^{-r/\lambda}\left[\frac{1}{3}\left(\frac{r}{\lambda}\right)^3 + \left(\frac{r}{\lambda}\right)^2 + 3\left(\frac{r}{\lambda}\right)\right]$$

$$+ n_0^2 \lambda^4\left(1 - e^{-r/\lambda}\right) \quad , \quad 0 \le r \le 2R_1 \tag{37}$$

$$= 0 \quad , \quad r > 2R_1$$

Collecting (33), (36) and (37) together and using (31), we get

$$r^2 p(r) = R^3 \left(\frac{2}{3} n_o^2 r^2 \right) + R^2 n_o^2 \left[-\frac{r^3}{2} - 2\lambda^2 r + e^{-r/\lambda} \left(2\lambda^2 r + \frac{\lambda r^2}{2} \right) \right]$$

+ (terms independent of R)   + (terms $\lesssim 0 \left(\frac{1}{R} \right)$) , $0 \le r \le 2R$

$$\simeq 0 \quad , \quad r > 2R \tag{38}$$

(we have used $R \gg \lambda$ to put $2R_1 \simeq 2R$)

Then inserting (38) into (11a) we have

$$\int \underline{d1} \, \underline{d2} \, n(12) f(r_{12}) = \left(\frac{4}{3} \pi R^3 \right) 4\pi n_o^2 \int_0^{2R} dr \, r^2 h(r)$$

$$- (4\pi R^2) \pi n_o^2 \int_0^{2R} dr \, r \, h(r) \left\{ r^2 + \lambda^2 \left[4 - e^{-r/\lambda} (4 + r/\lambda) \right] \right\} \tag{39}$$

+ negligible terms (independent of R or $\lesssim 0 \left(\frac{1}{R} \right)$)

where $h(r) = g(r) f(r)$ and we have assumed $h(r)$ decays faster

than r^{-3} . So the integral (11a) splits up into a volume part

and a surface part, plus negligible terms independent of R. It is

interesting that there is no part proportional to R which would have

then been a curvature correction term, i.e. the integral has split

up into a volume term and a surface term as if the spherical system

had a plane surface of area $4\pi R^2$.

We can check that the above results are sensible by using (18),

namely that $\frac{2\pi}{n_o^2} p(r)$ is the common volume of two overlapping

spheres with densities described by (27). Firstly, $\frac{2\pi}{n_o^2} p_1(r)$ is

the difference between the overlap volume of two sharp edged spheres

(densities described by $n_{R_1}(s)$) and the overlap volume of a sharp

edged sphere and a fuzzy edged sphere (density described by

$n_{R_1}(s) + \delta n_{R_1}(s)$). So, $p_1(r)$ should be negative and should be

largest when $r \doteq 0$. With $\delta n_{R_1}(s)$ given by (28) an obvious

estimate for the volume $\frac{2\pi}{n_o^2} p_1(0)$ is $-4\pi \lambda R_1^2$, that is $p_1(0) \simeq -2n_o^2 \lambda R_1^2$

which is exactly what (36) gives to order R_1^2 as $r \to 0$. The

overlap volume of two spheres reasoning implies that $p_2(r)$ should

have a maximum for $\Gamma \doteq 0$, which is a result that agrees with (37).
Note, that the terms we would also expect to find in $P_1(r)$ and
$P_2(r)$ for $\Gamma \doteq 2R_1$, using the overlapping volume picture, do not
appear in (36) and (37) because we neglected them by assuming $h(r)$
decays fast enough. If $h(r)$ did not decay faster than Γ^{-3} then
the neglected terms could easily be evaluated as an extension to
Appendix AI(5), however in most applications of (11a) $h(r)$ does
decay fast enough. For example, if we use (5) with (11) to
calculate the expectation value of the Hamiltonian of a liquid
helium-four droplet, then the slowest decaying part of $h(r)$ comes
from the long range phonon part of the wavefunction; $\phi(r) \sim r^{-2}$,
that is $h(r) \sim r^{-4}$. (See section C.II.) Using (39) we can get an
approximation to the integral $\int d_1 d_2 \, n(12) f(r_{12})$ for any general
surface of the type (27), by adjusting the surface width parameter λ
in (28), to get the closest approximation to the $\delta n_{R_1}(s)$ by an
exponential form.

Finally, we will compare the result (39) with the expression
for the surface energy of a classical, plane, liquid-vapour
interface, derived in [10]. In [10] similar methods to those used
above are applied to a two phase classical system with a plane
liquid-vapour interface. The assumption made, that is analogous
to (6), is (for the surface lying in the xy plane),

$$n(12) = n(z_1) n(z_2) g(r_{12}, \bar{n}) \tag{40}$$

where $\bar{n}$ is some average density depending on the temperature, to be
chosen on physical grounds. In the above we considered a quantum
system at T = 0, so there was no vapour phase, while in [10] the
density was given by (for the surface at Z = D),

$$n(z) = n_v + (n_\ell - n_v) \left[f_D(z) + \delta f(z) \right] \tag{41}$$

where $f_D(z) = \frac{n_D(z)}{n_o}$; $n_D(z)$ is the cutoff function (13). The expression (41) is directly analogous to (27) except that $n_v \neq 0$. When the exponential density

$$\delta f(z) = -\frac{e^{-D/\lambda}\, e^{z/\lambda}}{2} \qquad , \quad z < D$$
$$= \frac{e^{D/\lambda}\, e^{-z/\lambda}}{2} \qquad , \quad z > D \tag{42}$$

was used, the expression for the surface energy became, ($U = \sum_{i<j}\sum u(r_{ij})$)

[10] eq. (78)

$$\epsilon = -\frac{\pi}{2}(n_\ell - n_v)^2 \int_0^\infty dr\, g(r,\bar{n})u(r)r\left\{r^2 + \lambda^2\left[4 - e^{-r/\lambda}\left(4 + \frac{r}{\lambda}\right)\right]\right\} \tag{43}$$

(42) is the plane surface analogue of (28), with the surface definition $\int dz\,\delta f(z) = 0$ replacing the spherical system surface definition (30). From (5) and (39) we can find the corresponding expression for the surface energy of a classical droplet, in the approximation $n_v = 0, n_\ell = n_o$, namely

$$\frac{\langle U\rangle}{4\pi R^2} = -\frac{\pi}{2}n_o^2 \int_0^{2R} dr\, g(r)u(r)r\left\{r^2 + \lambda^2\left[4 - e^{-r/\lambda}\left(4 + \frac{r}{\lambda}\right)\right]\right\} \tag{44}$$

So, comparing (43) with (44) we see that the expression for the surface energy per unit area is the same for a droplet and a plane surface. That is, the result (39) for the integral $\int d\underline{1}\,d\underline{2}\, n(12)f(r_{12})$, derived for a spherical system, also holds for a system with a plane surface with $\frac{4}{3}\pi R^3$ and $4\pi R^2$ replaced by the corresponding volume and surface area.

APPENDIX AI(1) : The Weak Correlation Limit of g(r).

The obvious first approximation for (6) is to put

g(r) = constant for all r. This is more than just a

noninteracting limit for some quantum systems, (for example,

noninteracting spin zero Bosons, [2] page 431.), and so I call

it the weak correlation limit, [3]. From the definition of

$n(1...s)$, (2), it follows that for a system of particles in the

weak correlation limit

$$n(12) = \frac{(N-1)}{N} n(1)n(2)$$

Hence from (6) we have for the weak correlation limit

$$g(r_{12}) = 1 - \frac{1}{N} \qquad \text{for all } r_{12}. \qquad (a)$$

We can also derive (a) from number conservation.

Using (2) and (6) we have

$$N(N-1) = \int d\underline{r}_1 \int d\underline{r}_2 \, n(12)$$

$$= \int d\underline{r}_1 \int d\underline{r}_2 \, n(1)n(2)\left\{ g(\infty) + \left[g(r_{12}) - g(\infty) \right] \right\}$$

So assuming that surface effects don't matter in the change of

variables $d\underline{r}_1 d\underline{r}_2$ to $d\underline{r}_{12}$, (Feynman and Cohen, [6] Appendix B.,

state that surface corrections cancel.), we get

$$N(N-1) = g(\infty)N^2 + n_0^2 V \int d\underline{r} \left(g(r) - g(\infty) \right) \qquad (b)$$

where $n(r_j) = n_0 \left(= \frac{N}{V} \right)$ when r_j is well inside the system.

(b) implies, $\quad n_0 \int d\underline{r} \left(g(r) - g(\infty) \right) = -1 + N\left(1 - g(\infty) \right)$

which is equivalent to the well known relation

$$n_0 \int d\underline{r} \left(g(r) - 1 \right) = -1 \qquad (c)$$

Putting g(r) = constant for all r, in (c), we get the weak correlation

limit, $g(r) = 1 - \frac{1}{N}$.

It should be noted that the definition (2), and hence the rest of the section, applies to a system of a fixed number of particles. That is, a canonical ensemble. For non zero temperature a relation similar to (c) is derived by considering a grand canonical ensemble within the total system, namely, (de Bohr page 364, [4], with [5] chapter 6, problem 11; and Feynman and Cohen, [6], Appendix B.)

$$n_o \int d\underline{r} \, (g(r) - 1) \ = \ -1 + n_o T \chi_{_T} \qquad \text{(d)}$$

where $\chi_{_T}$ is the isothermal compressibility at absolute temperature T (in units of energy). (c) and (d) fail to agree at $T > 0$ because (c) applies to the total canonical ensemble which cannot experience number fluctuations, while (d) applies to a sub-volume of the total system. In most experimental applications (d) is the relevant expression because the total system is very rarely measured as a single entity in an experiment.

APPENDIX AI(2). $P_o(r)$.

From (14) we have, with $t \leq R$, $V = \frac{4}{3}\pi R^3$

$$P_o(r) = \frac{N^2}{V^2} \frac{1}{r} \int_0^R ds\, s \int_{|r-s|}^{r+s} dt\, t$$

The two cases

are

$$r < R \ (R_2)$$
$$r > R \ (R_1)$$
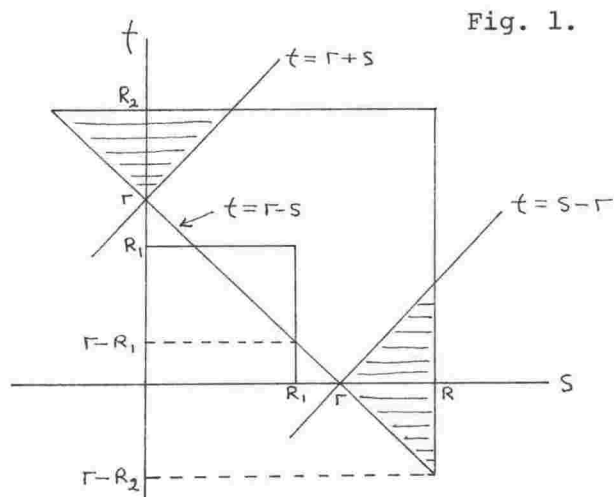
Fig. 1.

The integration over the shaded areas in figure 1. is zero

because

$$\int_{r-s}^{s-r} dt\, t = \int_{r-t}^{t-r} ds\, s = 0$$

Therefore the double integral is the same for both cases

giving

$$P_o(r) = \frac{N^2}{V^2} \frac{1}{r} \int_{r-R}^{R} dt\, t \int_{r-t}^{R} ds\, s \qquad , \quad 0 \leq r \leq 2R$$

$$= 0 \quad , \quad r > 2R$$

Evaluating the double integral we have for $\quad 0 \leq r \leq 2R$

$$P_o(r) = \frac{N^2}{V^2} \frac{1}{r} \frac{1}{2} \int_{r-R}^{R} dt\, t \left(R^2 - (r-t)^2 \right)$$

$$= \frac{N^2}{V^2} \left(\frac{r^3}{24} - \frac{rR^2}{2} + \frac{2}{3} R^3 \right)$$

$$= n_o^2 \frac{2}{3} R^3 \left[1 - \frac{3}{2}\left(\frac{r}{2R}\right) + \frac{1}{2}\left(\frac{r}{2R}\right)^3 \right]$$

where $\quad n_o = \frac{N}{V}$, $\quad V = \frac{4}{3}\pi R^3$.

APPENDIX AI(3). $p(r)$ as a function of $n(\underline{k})$.

Letting $h(r_{12}) = g(r_{12}) f(r_{12})$ we have using (6)

$$\int \underline{d1}\, \underline{d2}\, n(12) f(r_{12}) = \int \underline{dr_1}\, n(r_1) \int \underline{dr_2}\, n(r_2)\, h(r_{12})$$

Putting $\underline{r} = \underline{r_2} - \underline{r_1}$, $\underline{s} = \underline{r_1}$; $(r = |\underline{r_2} - \underline{r_1}| = r_{12},\, s = |\underline{r_1}|)$

we see that we can write

$$\int \underline{d1}\, \underline{d2}\, n(12) f(r_{12}) = \int \underline{dr}\, h(r) \int \underline{ds}\, n(s)\, n(|\underline{s} + \underline{r}|)$$

$$= 8\pi^2 \int_0^\infty dr\, r^2 h(r)\, p(r)$$

where

$$p(r) = \frac{1}{2\pi} \int \underline{ds}\, n(s)\, n(|\underline{s} + \underline{r}|)$$

Using (20), and noting that we can put $n(|\underline{s} + \underline{r}|) = n(\underline{r} + \underline{s})$,

we have

$$p(r) = \frac{1}{(2\pi)^7} \int \underline{ds} \int \underline{dk} \int \underline{dk'}\, e^{i(\underline{k} + \underline{k}').\underline{s}}\, e^{i\underline{k}'.\underline{r}}\, n(\underline{k})\, n(\underline{k}')$$

$$= \frac{1}{(2\pi)^4} \int \underline{dk}\, n^2(\underline{k})\, e^{i\underline{k}.\underline{r}}$$

where we have used $\dfrac{1}{(2\pi)^3} \int \underline{ds}\, e^{i(\underline{k} + \underline{k}').\underline{s}} = \delta(\underline{k} + \underline{k}')$

and also the fact that for spherical symmetry

$$n(\underline{k}) = n(k) = n(-\underline{k}) \quad , \text{ see (22).}$$

Integrating over the angular part of $\underline{dk}$ gives

$$p(r) = \frac{1}{4\pi^3}\, \frac{1}{r} \int_0^\infty dk\, k\, n^2(\underline{k})\, \sin kr$$

APPENDIX AI(4). Spherical Bessel Functions [8].

The regular solution to the differential equation

$$f''(r) + \frac{2}{r} f'(r) + \left(K^2 - \frac{\ell(\ell+1)}{r^2} \right) f(r) = 0 \tag{i}$$

is the spherical Bessel function of order ℓ, $j_\ell(Kr)$.

$$j_0(x) = \frac{\sin x}{x} \tag{ii}$$

$$j_1(x) = \frac{\sin x}{x^2} - \frac{\cos x}{x} = \int_0^1 dr\, r \sin xr \tag{iii}$$

$$j_{\ell-1}(x) + j_{\ell+1}(x) = \frac{(2\ell+1)}{x} j_\ell(x) \tag{iv}$$

Some useful properties of $j_\ell(x)$ are

$$\frac{d}{dx} j_\ell(x) = \frac{1}{(2\ell+1)} \left[\ell j_{\ell-1}(x) - (\ell+1) j_{\ell+1}(x) \right] = j_{\ell-1}(x) - \frac{(\ell+1)}{x} j_\ell(x) \tag{v}$$

$$\frac{d}{dx} \left(x^{\ell+1} j_\ell(x) \right) = x^{\ell+1} j_{\ell-1}(x) \tag{vi}$$

$$\frac{d}{dx} \left(x^{-\ell} j_\ell(x) \right) = -x^{-\ell} j_{\ell+1}(x) \tag{vii}$$

$$\int j_1(x)\, dx = -j_0(x) \tag{viii}$$

$$\int x^2 j_0(x)\, dx = x^2 j_1(x) \tag{ix}$$

APPENDIX AI(5) : $P_1(r)$ and $P_2(r)$

We first assume $R_1 \gg \lambda$ and define

$$x_1 = s/\lambda \;,\; x_2 = t/\lambda \;,\; y = r/\lambda$$

$$\overline{n(x_2)} = 1 \qquad,\quad x_2 \leq R_1/\lambda$$
$$= 0 \qquad,\quad x_2 > R_1/\lambda \qquad\qquad (a)$$

$$e(x) = -e^{-R_1/\lambda} e^{x} \qquad,\quad x < R_1/\lambda$$
$$= e^{R_1/\lambda} e^{-x} \qquad,\quad x > R_1/\lambda$$

Then from (27), (28), (34) and (35) we can write

$$f(y) \equiv \frac{r \cdot P_1(r)}{n_o^2 \lambda^4} = \int_0^\infty dx_1 x_1 e(x_1) \int_{|x_1-y|}^{x_1+y} dx_2 x_2 \overline{n(x_2)} \qquad\qquad (b)$$

$$J(y) \equiv \frac{4 r \cdot P_2(r)}{n_o^2 \lambda^4} = \int_0^\infty dx_1 x_1 e(x_1) \int_{|x_1-y|}^{x_1+y} dx_2 x_2 e(x_2) \qquad\qquad (c)$$

Now for $r \leq R_1 \left(y \leq \frac{R_1}{\lambda}\right)$ the lower limit of x_2 in both (b) and

(c) can be replaced by (x_1-y) . This is valid in (b) provided

we extend $\overline{n(x_2)}$ by defining $\overline{n(x_2)} = 1$ for $x_2 < 0$,

then for $y \leq R_1/\lambda$ the extra term in (b) includes (see figure 1.)

$$\int_{x_1-y}^{y-x_1} dx_2 x_2 = 0$$

Fig. 1.

$x_2 = x_1 + y$

$x_2 = x_1 - y$

} When $R_1 \gg \lambda$ $e(x_2)$ is only non negligible in a small range about R_1/λ .

$x_2 = y - x_1$

When $R_1 \gg \lambda$ $e(x_1)$ is only non negligible in a small range about R_1/λ .
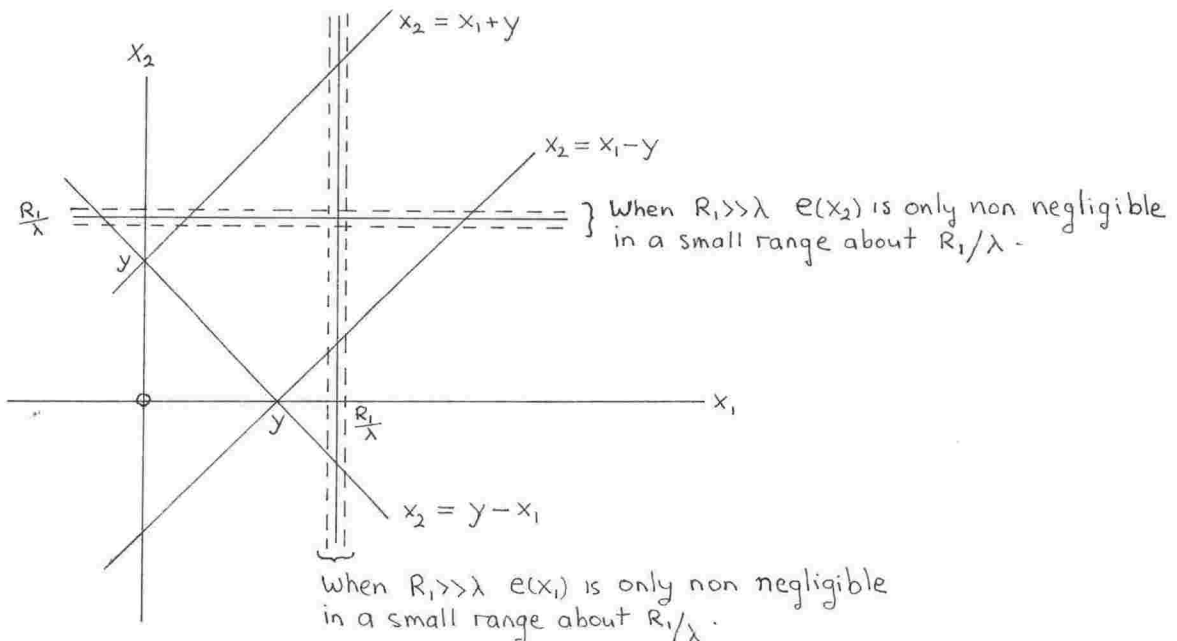
It is valid in (c) because $e(x)$ is non negligible only for

$x \doteq R_1/\lambda$ so that the extra term in (c) is (see figure 1.)

$$\int_0^y dx_1\, x_1\, e(x_1) \int_{x_1-y}^{y-x_1} dx_2\, x_2\, e(x_2) = 0 \text{ for } y \leq R_1/\lambda \text{ , neglecting terms } O\left(e^{-R_1/\lambda}\right)$$

For $R_1 \leq r \leq 2R_1$ $\left(R_1/\lambda \leq y \leq 2R_1/\lambda\right)$ we can still replace the lower

limit of x_2 in (b) and (c) by $(x_1 - y)$. This follows for (b)

provided we first subtract from (b) the contribution from the

shaded area in figure 2. with $\overline{n(x_2)}$ replaced by 1. Because $e(x_1)$

is non negligible only for $y \doteq 2R_1/\lambda$ we see from figure 2. that

this shaded area contribution is negligible except for $r \doteq 2R_1\left(y \doteq 2R_1/\lambda\right)$.

Similarly, because $e(x)$ is negligible except for $x \doteq R_1/\lambda$

the replacement of the lower limit of x_2 in (c) by $(x_1 - y)$

is valid except for terms non negligible only for $y \doteq 2R_1/\lambda$

(see figure 2.).

Fig. 2.

When $R_1 \gg \lambda$ $e(x_2)$ is only non negligible in a small range about R_1/λ.

when $R_1 \gg \lambda$ $e(x_1)$ is only non negligible in a small range about R_1/λ.
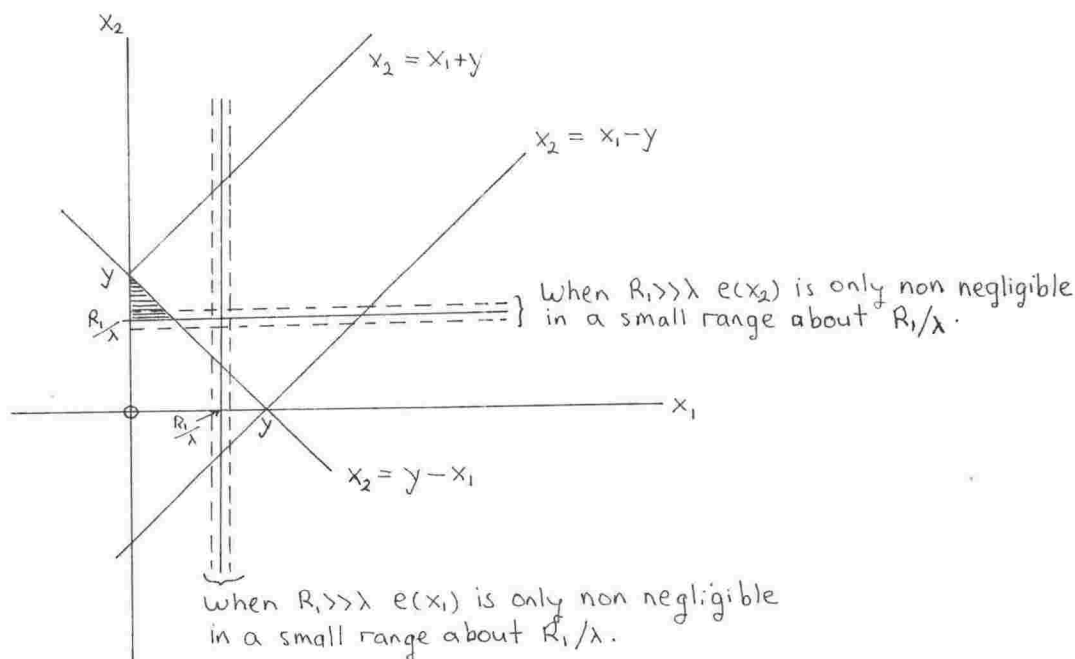
The calculation of these terms, which are negligible except when

$r \doteq 2R_1$, is straight forward and for $R_1/\lambda \leq y \leq 2R_1/\lambda$ yields

terms in (b) and (c) which are all proportional to $e^{Y - 2R_1/\lambda}$

Also we see from figure 2. that for $y > 2R_1/\lambda$ (b) and (c) will

only be non negligible for $y \doteq 2R_1/\lambda$. However, we will not

calculate any of these terms, but instead assume that in (11a) $h(r)$

decays with r fast enough so that we can neglect all the terms in

$p(r)$ that are non negligible only for $r \doteq 2R_1/\lambda$. Thus with this

proviso (b) and (c) become

$$
\begin{array}{l}
f(y) \\
{\scriptstyle (0 \le y \le 2R_1/\lambda)}
\end{array}
= \int_0^\infty dx_1 \, x_1 \, e(x_1) \int_{x_1 - y}^{x_1 + y} dx_2 \, x_2 \, \overline{n(x_2)}
\tag{d}
$$

$$
\begin{array}{l}
J(y) \\
{\scriptstyle (0 \le y \le 2R_1/\lambda)}
\end{array}
= \int_0^\infty dx_1 \, x_1 \, e(x_1) \int_{x_1 - y}^{x_1 + y} dx_2 \, x_2 \, e(x_2)
\tag{e}
$$

We now evaluate the derivatives of (d) and (e) with respect to y

$$
\frac{df(y)}{dy} = \int_0^\infty dx_1 \, x_1 \, e(x_1) \left[(x_1 + y) \, \overline{n(x_1 + y)} + (x_1 - y) \, \overline{n(x_1 - y)} \right]
$$

$$
= \int_0^\infty dx_1 \, x_1^2 \, e(x_1) \left[\overline{n(x_1 + y)} + \overline{n(x_1 - y)} \right] + y \int_0^\infty dx_1 \, x_1 \, e(x_1) \left[\overline{n(x_1 + y)} - \overline{n(x_1 - y)} \right]
$$

$$
= -e^{-R_1/\lambda} \int_0^{R_1/\lambda - y} dx \, x^2 e^x - e^{-R_1/\lambda} \int_0^{R_1/\lambda} dx \, x^2 e^x + e^{R_1/\lambda} \int_{R_1/\lambda}^{R_1/\lambda + y} dx \, x^2 e^{-x}
$$

$$
+ y \left\{ -e^{-R_1/\lambda} \int_0^{R_1/\lambda - y} dx \, x \, e^x + e^{-R_1/\lambda} \int_0^{R_1/\lambda} dx \, x \, e^x - e^{R_1/\lambda} \int_{R_1/\lambda}^{R_1/\lambda + y} dx \, x \, e^{-x} \right\}
$$

Neglecting terms of order $e^{-R_1/\lambda}$ we find

$$
\frac{df(y)}{dy} = 4 \left(\frac{R_1}{\lambda} \right) - 2y - e^{-y} \left[2 \left(\frac{R_1}{\lambda} \right)^2 + 2y + 4 \right]
\tag{f}
$$

From (e)

$$
\frac{dJ(y)}{dy} = \int_0^\infty dx_1 \, x_1 \, e(x_1) \left[(x_1 + y) \, e(x_1 + y) + (x_1 - y) \, e(x_1 - y) \right]
$$

$$
= \int_0^\infty dx_1 \, x_1^2 \, e(x_1) \left[e(x_1 + y) + e(x_1 - y) \right] + y \int_0^\infty dx_1 \, x_1 \, e(x_1) \left[e(x_1 + y) - e(x_1 - y) \right]
$$

$$
= e^y e^{-2R_1/\lambda} \int_0^{R_1/\lambda - y} dx \, x^2 e^{2x} - e^{-y} \int_{R_1/\lambda - y}^{R_1/\lambda} dx \, x^2 + e^{-y} e^{2R_1/\lambda} \int_{R_1/\lambda}^\infty dx \, x^2 e^{-2x}
$$

$$+ e^{-y} e^{-2R_1/\lambda} \int_0^{R_1/\lambda} dx\, x^2 e^{2x} - e^{-y} \int_{R_1/\lambda}^{R_1/\lambda+y} dx\, x^2 + e^{y} e^{2R_1/\lambda} \int_{R_1/\lambda+y}^{\infty} dx\, x^2 e^{-2x}$$

$$+ y \left\{ e^{y} e^{-2R_1/\lambda} \int_0^{R_1/\lambda-y} dx\, x\, e^{2x} - e^{-y} \int_{R_1/\lambda-y}^{R_1/\lambda} dx\, x + e^{-y} e^{2R_1/\lambda} \int_{R_1/\lambda}^{\infty} dx\, x\, e^{-2x} \right.$$

$$\left. - e^{-y} e^{-2R_1/\lambda} \int_0^{R_1/\lambda} dx\, x\, e^{2x} + e^{-y} \int_{R_1/\lambda}^{R_1/\lambda+y} dx\, x - e^{-y} e^{2R_1/\lambda} \int_{R_1/\lambda+y}^{\infty} dx\, x\, e^{-2x} \right\}$$

Neglecting terms of order $e^{-R_1/\lambda}$ we find

$$\frac{d J(y)}{dy} = e^{-y} \left[2\left(\frac{R_1}{\lambda}\right)^2 - 2y\left(\frac{R_1}{\lambda}\right)^2 + \frac{y^3}{3} + y + 1 \right] \tag{g}$$

From (d) and (e) we have $f(0) = J(0) = 0$, so that

integration of (f) and (g) gives

$$\underset{(0 \le y \le 2R_1/\lambda)}{f(y)} = -2\left(\frac{R_1}{\lambda}\right)^2 + 4\left(\frac{R_1}{\lambda}\right)y - y^2 + e^{-y}\left[2\left(\frac{R_1}{\lambda}\right)^2 + 2y\right] + 6\left[e^{-y} - 1\right] \tag{h}$$

$$\underset{(0 \le y \le 2R_1/\lambda)}{J(y)} = 2\left(\frac{R_1}{\lambda}\right)^2 y\, e^{-y} - e^{-y}\left[\frac{y^3}{3} + y^2 + 3y\right] + 4\left[1 - e^{-y}\right] \tag{i}$$

Therefore we have shown that for $R_1 \gg \lambda$ and assuming $h(r)$

decays fast enough so that we can neglect terms of order $e^{-|y-2R_1/\lambda|}$,

that the exponential surface (28) leads to

$$r\, p_1(r) = -2n_0^2 \lambda^2 R_1^2 \left[1 - 2\left(\frac{r}{R_1}\right) + \frac{1}{2}\left(\frac{r}{R_1}\right)^2 - e^{-r/\lambda}\right] + 2n_0^2 \lambda^3 r\, e^{-r/\lambda}$$

$$- 6n_0^2 \lambda^4 \left[1 - e^{-r/\lambda}\right], \quad 0 \le r \le 2R_1$$

$$= 0, \quad r > 2R_1 \tag{j}$$

$$r\, p_2(r) = \frac{n_0^2}{2} R_1^2 \lambda r\, e^{-r/\lambda} - \frac{n_0^2}{4} \lambda^4 e^{-r/\lambda}\left[\frac{1}{3}\left(\frac{r}{\lambda}\right)^3 + \left(\frac{r}{\lambda}\right)^2 + 3\left(\frac{r}{\lambda}\right)\right]$$

$$+ n_0^2 \lambda^4 \left[1 - e^{-r/\lambda}\right], \quad 0 \le r \le 2R_1$$

$$= 0, \quad r > 2R_1 \tag{k}$$

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II. ROTATIONAL STATES OF QUANTUM SYSTEMS.

In this section trial wavefunctions for collective rotational excitations of a quantum many-body system are investigated. The wave-functions considered are all of the form

$$\Psi = F \Phi \tag{1}$$

where Φ is the exact ground state wavefunction. The expectation value of the energy of the state (1) is easily calculated, provided the potential energy V commutes with F (for example V must not contain velocity dependent interactions). With this restriction it follows that

$$(H - E_0)\Psi = -\frac{\hbar^2}{2m} \Phi^{-1} \sum_k \nabla_k \cdot (\Phi^2 \nabla_k F) \tag{2}$$

where $H\Phi = E_0\Phi$ and H is the Hamiltonian

$$H = -\frac{\hbar^2}{2m} \sum_k \nabla_k^2 + V \tag{3}$$

Then by a simple integration by parts we have (cf. eqn 4 of [1])

$$\frac{\int d\tau\, \Psi^*(H-E_0)\Psi}{\int d\tau\, |\Psi|^2} = \frac{\hbar^2}{2m} \sum_k \frac{\langle |\nabla_k F|^2 \rangle}{\langle |F|^2 \rangle} \tag{4a}$$

where $d\tau = \prod_j d\underline{r}_j$ and $\langle\ \rangle$ is the ground state expectation value defined by

$$\langle B \rangle = \frac{\int d\tau\, B\, \Phi^2}{\int d\tau\, \Phi^2} \tag{4b}$$

The integrals in (4a) and (4b) can include a summation over spin variables as well as an integration over space. It is the simplicity of (4a) that makes trial wavefunctions of the form (1) so attractive.

This section is divided into two parts : The first part investigates certain isoscalar, rotational excitations of even-even nuclei. An isoscalar nuclear motion is one in which the proton and neutron fluids move in phase. (In fact for part (a), all differences between protons and neutrons are neglected when evaluating (4).) Most of the discussion is about a particular 2^+ state; spherical nuclei are discussed in a paper supplied with this thesis, [1] , and I consider a possible extension to deformed even-even nuclei.

Part (b) considers isovector analogues of the nuclear rotational excitations in part (a). An isovector motion (or polarization mode) is one in which the proton and neutron fluids move out of phase. It is noticed that the simplest wavefunction of the type proposed should give a good description of the resonance state of the 1^- giant dipole oscillation. The resonance energy of the giant dipole state for spherical nuclei is calculated using this wavefunction in a paper supplied with this thesis, [2] . The value obtained by including only those pair correlations due to Fermi statistics is close to experimental data.

Many of the results of this section apply to finite quantum systems other than nuclei. For example, the isoscalar excitations discussed in part (a) should also be rotational states of polyatomic molecules and droplets of superfluid He^4 . For nuclei the energies of these states are in the region of known experimental results, but in the case of liquid He^4 , it can be seen from the weak correlation approximation results of part (a), that the energies would be so low that they would probably be impossible to detect directly. For this reason I have restricted the quantitative results to nuclei.

{ aside: The problem of translational invariance (see below) is avoided if one considers rotational excitations of liquid He^4 in a

container. For a cylinder, the wavefunctions corresponding to the spherical states (9) and (12) follow by changing $Y_{LM}(\hat{r}) j_\ell(kr)$ to $e^{i\ell\phi} J_\ell(kr)$. These compressional states will have an energy that is of the order of N (or perhaps $N^{2/3}$) lower than the experimentally known vortex excitations, where N is the number of He^4 atoms. }

(a) Isoscalar Rotational States of Nuclei :

In reference [3] a set of three-body angular momentum eigenstates is generalized to obtain a set of many-body rotational wavefunctions of the form (1), with Φ assumed to be both translationally and rotationally invariant :

$$\Psi_{LM} = F_{LM} \Phi = \frac{1}{2} \sum_i \sum_j r_{ij}^L \, Y_{LM}(\hat{r}_{ij}) f(r_{ij}) \, \Phi \tag{5}$$

Since $Y_{LM}(-\hat{r}) = (-1)^L Y_{LM}(\hat{r})$ it follows that Ψ_{LM} is zero for odd L, i.e. L must be even. From this we also see that Ψ_{LM} has the same permutation symmetry (i.e. bose or Fermi) and inversion symmetry (parity) as Φ . An important property of these wave-functions, given particular attention to in [3] , is that they are translationally invariant. This is necessary because (i) the orbital angular momentum of a system of particles is independent of the choice of origin only in the rest frame of the system, (i.e. only if the wavefunction is translationally invariant), and (ii) the uncertainty principle insists that the centre of mass becomes indeterminate when the system is in its rest frame. That is, the true excitations of a self-bound many-body quantum system are translationally invariant and wavefunctions constructed on the basis of the centre of mass being fixed at the origin may describe

completely unphysical motions. It was shown in [3] that when

$\underline{L} \Phi = \underline{O}$ that Ψ_{LL} is an eigenstate of $\underline{L}^2$ and L_z with

eigenvalues $L(L+1)\hbar^2$ and $L\hbar$ respectively, and further, that for

harmonic pair interactions between N identical bose particles,

i.e. (3) with

$$V = \upsilon \sum_{i<j}^{A} \sum^{A} \left(\frac{r_{ij}}{a}\right)^2 + \text{constant} \quad , \tag{6}$$

that Ψ_{LL} with f = 1 is also an energy eigenstate with eigenvalue

$$E_L = E_0 + L\left(\frac{2\hbar^2 \upsilon A}{ma^2}\right)^{1/2} \tag{7}$$

(an erroneous factor of $\sqrt{2}$ in eqns 28 & 30 of [3] has been removed).

The particular rotational state Ψ_{2M} (with f = 1) for spherical

nuclei, is investigated in detail in reference [1] . By restricting

Φ to be the ground state of a non deformed even-even nucleus it

follows that $L = O$ as well as $J = O$ in the ground state. It is

assumed that for both Ψ_{2M} and Φ , that the spins are paired up

to give zero $\underline{S}$ so that spin coordinates can be ignored and the total

angular momentum is the orbital angular momentum. So Φ is a 0[+]

state and Ψ_{2M} is a 2[+] state. A consequence of taking Φ to be

spherically symmetric is that Ψ_{2M} is degenerate for all M, (-2,-1,0,1,2),

since the energy cannot depend on the azimuthal quantum number for a

spherical system. To evaluate the expression for the variational bound

to the rotational energy, $\Delta E_2 (\equiv E_2 - E_0)$, the weak correlation

approximation is introduced. In the weak correlation limit the

nucleons are correlated only by virtue of the finiteness of the

self-bound system, i.e. the correlations are characterized solely

by a number density n(r) (measured relative to the centre of mass).

Because the nucleon hard cores occupy less than 1% of the total volume,

it is assumed in [1] that the weak correlation approximation does not

introduce qualitative error. This is justified in section five of [1] using the particular pair correlation function $g(r_{12}) = 0$ for $r_{12} < a$ and $g(r_{12}) = 1$ for $r_{12} > a$. In the weak correlation approximation the variational energy of the 2^+ state is calculated to be

$$\Delta E_2 \lesssim \frac{7\hbar^2}{mR^2} \simeq 200\, A^{-2/3}\, MeV \qquad \text{(A is the number} \atop \text{of nucleons)} \quad (8)$$

where R is the nuclear radius (which is taken to be $1.2\, A^{1/3}\, fm$). It is noticed in section four of [1] that by fixing the centre of mass at the origin that the L = 2, f = 1, wavefunction of the class (5) can be written as

$$\Psi'_{2M} = F'_{2M}\, \Phi = \sum_j^A r_j^2\, Y_{2M}(r_j)\, \Phi \qquad (9)$$

Because the translationally <u>variant</u> wavefunction (9) originates from Ψ_{2M} , we can be sure that Ψ'_{2M} represents an internal excitation and not a spurious centre of mass motion. Using the simpler wavefunction (9) and the weak correlation approximation, the quadrupole moment and the transition rate to the ground state, are calculated to be

$$Q\,(M=2) \simeq -\frac{4}{9}\frac{Z}{A}e\,R^2 \qquad \text{(Z the number of protons)} \quad (10)$$

and

$$T_E\,(L=2,\,M=2) \simeq \frac{1}{175}\frac{Z^2}{A}e^2\frac{k^5}{\hbar}R^4 \qquad (11)$$

A comparison of (8) and (11) with experimental data produced only one possible identification, namely the first 2^+ state of Pb^{208} at 4.085 MeV .

In the last section of [1] it is concluded that Ψ_{2M} is a finite system angular momentum projection of Feynman's phonon

excitation proposed for liquid He^4 . This conclusion was arrived at by considering the generalization of (9) to $f \neq 1$:

$$\Psi'_{20} = \sum_j (3z_j^2 - r_j^2) f(r_j) \Phi \qquad (12)$$

In the weak correlation approximation and taking Φ to be spherically symmetric, it was found that the optimum f is $f(r) = r^{-2} j_2(kr)$, where $k^2 = 2m\Delta E_2 / \hbar^2$. With this f the wavefunction (12) becomes the L = 2 component of Feynman's wavefunction $\sum_j e^{i \mathbf{k} \cdot \mathbf{r}_j} \Phi$. From this interpretation it follows that Ψ_{2M} is a rotational compressional density wave carrying angular momentum L = 2. However, in note 7) of BII an argument based on the expected velocity field of wavefunctions of the form (12), strongly suggests that when f = 1 that the physical nature of (12) changes to become an incompressible surface oscillation, i.e. for f = 1 Ψ_{2M} should represent a rotational surface wave carrying angular momentum L = 2. For spherical nuclei the difference between the two physical interpretations is a minor one, provided the amplitude is small, but for deformed nuclei some of the small amplitude surface waves can also represent large amplitude rotational and vibrational modes. I shall make use of the surface wave interpretation a little later on, by assuming that Ψ_{2M} can also be applied to the deformed even-even nuclei. But first I should include a note of caution about the main assumption in [1] used to obtain the energy bound (8), namely that the weak correlation approximation does not introduce serious error.

1) The possibility that the weak correlation approximation introduces qualitative error:

In the calculation of the variational energy of the 2^+ state of spherical nuclei, in reference [1] , the weak correlation approximation implied that the four-particle part of $\langle |F_{2M}|^2 \rangle$ and the two-

particle part of $\langle |F'_{2M}|^2 \rangle$ were identically zero. However, both these terms are multiplied by an extra factor of A than the first non zero term in the energy denominator. So a large error in the result for ΔE_2 could have been introduced, unless the weak correlation approximation leads to errors of less than the order of $1/A$ in expectation values like $\langle r_{12}^2 \rangle$. In section 5 of [1] it is shown that the simple pair correlation function

$$g(r_{12}) = \begin{cases} 0 & r_{12} < a \\ 1 & r_{12} > a \end{cases}$$

(13)

leads to the result

$$\langle r_{12}^2 \rangle = \langle r_{12}^2 \rangle_0 \left\{ 1 + \left(\frac{a}{R}\right)^3 + O\left(\frac{a}{R}\right)^4 \right\}$$

(14)

where $\langle r_{12}^2 \rangle_0 \left(\simeq \frac{6}{5} R^2 \right)$ is the weak correlation limit. Taking a to be the experimental nucleon hard core we have $(a/R) \simeq 1/3 \, A^{-1/3}$. So the correction term in (14) is $\ll 1/A$, which supports the use of the weak correlation approximation in the case of nuclei. However, the particular pair correlation function (13) is misleading since when a is the nucleon hard core it fails to satisfy the number conservation identity (see eqn AI(2))

$$\int d\underline{r}_1 \int d\underline{r}_2 \, n(\underline{r}_1) n(\underline{r}_2) g(r_{12}) = A(A-1) ,$$

(15)

where the pair density has been approximated by (see eqn AI(6))

$$n(\underline{r}_1, \underline{r}_2) = n(\underline{r}_1) n(\underline{r}_2) g(r_{12})$$

(16)

{ aside: $n(\underline{r}_1, \underline{r}_2)$ is defined such that $n(\underline{r}_1, \underline{r}_2) d\underline{r}_1 d\underline{r}_2$ is the probability of finding particle one in a volume $d\underline{r}_1$ about $\underline{r}_1$, together with particle two in a volume $d\underline{r}_2$ about $\underline{r}_2$. So when particle one is fixed at $\underline{r}_1$ the integration over $d\underline{r}_2$ in (15) gives (A-1) instead of A. Thus the right hand side of (15) is A(A-1) and

not A^2 . $\Big\}$

In the notation of section five of $[1]$, we have

$$\langle \Gamma_{12}^2 \rangle = \langle \Gamma_{12}^2 \rangle_0 \left\{ 1 - \frac{8\pi^2}{A^2} K_0 + \quad \leq 0\left(\frac{1}{A^{4/3}}\right) \right\} \tag{17}$$

where, using (15)

$$K_0 \equiv \int_0^\infty d\vec{r_1}\, r_1\, n(r_1) \int_0^\infty d\vec{r_2}\, r_2\, n(r_2) \int_{|r_1 - r_2|}^{r_1 + r_2} dr_{12}\, r_{12} \left[g(r_{12}) - 1 \right]$$

$$= \frac{1}{8\pi^2} \left\{ \int d\underline{r_1} \int d\underline{r_2}\, n(r_1)\, n(r_2)\, g(r_{12}) - \left[\int d\underline{r_1}\, n(r_1) \right]^2 \right\}$$

$$= \frac{1}{8\pi^2} \left\{ A(A-1) - A^2 \right\}$$

$$= - \frac{A}{8\pi^2} \tag{18}$$

So the requirement of number conservation implies

$$\langle \Gamma_{12}^2 \rangle = \langle \Gamma_{12}^2 \rangle_0 \left\{ 1 + \frac{1}{A} + \leq 0\left(\frac{1}{A^{4/3}}\right) \right\} \tag{19}$$

The correct result (19), which replaces (14), suggests that errors of the order of 100% may have been introduced into the expression (8) for ΔE_2 .

There is also a possibility that the result for ΔE_2 could be completely incorrect, if the $1/A$ terms from expectation values like (19) were such that their contribution to the four-particle part of $\langle |F_{2M}|^2 \rangle$ (or the two particle part of $\langle |F_{2M}'|^2 \rangle$) happened to cancel with the weak correlation approximation terms in the energy denominator. If this was the case then ΔE_2 would be proportional to $A^{-1/3}$ instead of $A^{-2/3}$. In part (b) (i.e. $[2]$), the simplest isovector state ψ_{10} exhibits this type of cancellation. It is known that ψ_{10} represents the resonance state of the giant

dipole oscillation, which has an experimental energy

$$\Delta E_1 \simeq 80 A^{-1/3} \, MeV.$$

But even for arbitrary proton and neutron radial densities, the weak correlation approximation leads to $\Delta E_1 \approx 70 A^{-2/3} \, MeV$. The experimental variation of the energy with nucleon number A, is only obtained by including the pair correlation terms in the energy denominator, i.e. the terms analogous to the $1/A$ contribution in the expression (19). When this is done the contribution from the weak correlation approximation is cancelled exactly, leaving higher order terms that give $\Delta E_1 \sim A^{-1/3}$. In reference [2] the energy is calculated by including only those pair correlations due to Fermi statistics, but provided the non indistinguishability between protons and neutrons is first taken into account (i.e. Ψ_{1M} is an isovector state), then the $A^{-1/3}$ variation also follows from the same type of number conservation argument that led to (19). That is, the variation is independent of the nature of the short ranged correlations, which only determine the proportionality constant. The question is, could this be a general result for all of the isoscalar and isovector wavefunctions, Ψ_{LM} and Ψ_{LM} , when applied to spherical nuclei?

In particular, consider the 2^+ state Ψ_{2M} of reference [1] . What we would like to be able to do is to identify Ψ_{2M} with a known excitation of spherical nuclei. For this purpose the extensive work of reference [4] is very useful. I concluded above from the results of BII, that Ψ_{2M} (when f = 1) should represent a surface wave carrying angular momentum L = 2. However, this motion can equally be regarded as quadrupole shape oscillation about a spherical equilibrium. So perhaps Ψ_{2M} is the isoscalar giant quadrupole oscillation, in analogy to Ψ_{1M} which is the giant dipole oscillation. The discussion on quadrupole modes starting on page 507 of [4] is in the usual language of nuclear physics, but it has similarities with the many-body

approach used in this section. For example, reference [4] introduces the idea of the quadrupole field, which happens to be F_{2M}' (see the translationally variant wavefunction (9)). The spectrum of independent particle excitations produced by the field F_{2M}' is divided into two groups, one at low energy and the other at high energy (see page 466 of [4]). From this it is concluded that there are two different quadrupole modes. The strength of the low energy mode vanishes for closed shell configurations and it is identified with rotational and vibrational states of deformed nuclei (see below). The high energy mode is the giant quadrupole oscillation, which is found to have an energy

$$\Delta E_2 \simeq 58 A^{-1/3} \, MeV \qquad \text{(see [4] p 509)}.$$ An important property of the high energy quadrupole oscillation is that it uses up a large part of the $S_2^{(1)}$ sum rule (sum rules are discussed in section AIV). In Section AIV the contribution to the $S_2^{(1)}$ sum rule from the five degenerate Ψ_{2M} states (note, this degeneracy only holds for spherical nuclei), is calculated in the same approximation used to evaluate the energy in [1] , namely, the potential energy is assumed to commute with F_{2M}' , which implies the neglect of charge exchange and velocity dependent interactions. The result is (see AIV (17)), that the Ψ_{2M}' state in spherical nuclei exhausts the factor Z/A of the $S_2^{(1)}$ (class) sum rule, which strongly suggests that it represents the giant quadrupole resonance state.

From above it follows that the Ψ_{2M} state of spherical nuclei should have an energy proportional to $A^{-1/3}$, and not $A^{-2/3}$ as is found in the weak correlation approximation (i.e. (8)). If this is correct, then to get the true energy one must include the short ranged nucleon correlations. For the Ψ_{10} state of reference [2] the procedure is simple since the exact energy is expressed in terms of two-particle expectation values, but for Ψ_{2M} we need to evaluate

four-particle contributions. For Ψ'_{2M} we only need to evaluate two-particle terms but because of the broken translational invariance they are no longer of the form $\langle r_{12}^2 \rangle$. Because of these difficulties I will leave to the future, the problem of extending the work of [1] to include the contributions from short ranged internucleon correlations. It is worth pointing out that virtually all the collective states of spherical nuclei considered in reference [4] , have an energy varying as $A^{-1/3}$. For example, in the liquid drop model (App. 6A of [4]), every isoscalar and isovector compressional mode has an energy that varies as $A^{-1/3}$. Thus it is possible that the wavefunctions Ψ_{LM} and $\mathcal{L}_{LM}$ (see part (b)) represent these states ($\mathcal{L}_{10}$ is known to), but that the weak correlation approximation leads to an unexpected qualitative error, namely the weak correlation terms in the energy denominator may be cancelled by part of the contributions from the short ranged correlations. This speculation should be kept in mind when reading section AIII (which was written prior to this section), where the energies of collective vibrational modes are evaluated in the weak correlation approximation.

NOTE ADDED IN "PROOF" :

Consider the denominator term in the exact expression for the variational energy of the state Ψ_{2M} , namely (see eqn 31 of [1])

$$D = \left\langle \tfrac{9}{10} r_{12}^4 - r_{12}^2 r_{13}^2 \right\rangle + \frac{A}{10} \left\langle 3 r_{12}^4 - 6 r_{12}^2 r_{13}^2 + 2 r_{12}^2 r_{34}^2 \right\rangle + O(A^{-1})$$

In the weak correlation approximation

$$D = \left\langle \tfrac{9}{10} r_{12}^4 - r_{12}^2 r_{13}^2 \right\rangle_0 = \frac{4}{5} \overline{r^4}$$

where

$$\overline{f(r)} \equiv \frac{\int_0^\infty dr\, r^2 n(r) f(r)}{\int_0^\infty dr\, r^2 n(r)}$$

In the cutoff density approximation $\overline{\Gamma^n} = \frac{3}{n+3} R^n$,

(see (44) & (45)). So in the weak correlation approximation we have

$D \sim R^4$, which implies $\Delta E_2 \sim \hbar^2/mR^2 \sim A^{-2/3}$.

However, I will now show that the exact expression for D is such that

$D \sim A^{-1/3} R^4$, which implies $\Delta E_2 \sim A^{-1/3}$, as

was conjectured above. To evaluate the second term in the exact

expression for D we need to evaluate expectation values of the form

$$\langle h(\Gamma_1,...,\Gamma_s) \rangle = \int \underline{d1}....\underline{ds}\, n(1...s)\, h(\Gamma_1,..,\Gamma_s)$$

where $S \in (2,3,4)$ and (see AI(2))

$$n(1...s) = \frac{A!}{(A-s)!} \frac{\int \underline{d(s+1)}...dA\, \Phi^2}{\int \underline{d1}....dA\, \Phi^2}$$

In analogy with (16) let us define a generalized S-particle

correlation function g(1...S), by

$$n(1...s) = n(1)n(2)...n(s)\, g(1...s)$$

Then we can write

$$\langle h(\Gamma_1,..,\Gamma_s) \rangle = \frac{\int \underline{d1}\, n(1) \int \underline{d2}\, n(2)... \int \underline{ds}\, n(s)\, h(\Gamma_1,..,\Gamma_s)\left[1+(g(1...s)-1)\right]}{\int \underline{d1}\, n(1) \int \underline{d2}\, n(2)... \int \underline{ds}\, n(s)\left[1+(g(1...s)-1)\right]}$$

The weak correlation result, $\langle h \rangle_0$, is obtained by putting g = 1.

Now in the case of $h = \Gamma_{12}^2$, we found that the correction terms

coming from (g-1) $\neq$ 0 in the numerator were at least of order $A^{-1/3}$

smaller than the correction terms in the denominator (see (14) and

(17)). This is almost certainly true for all the other terms in D,

i.e. $\langle \Gamma_{12}^4 \rangle$, $\langle \Gamma_{12}^2 \Gamma_{13}^2 \rangle$ and $\langle \Gamma_{12}^2 \Gamma_{34}^2 \rangle$. (See [1] for a proof

for the case of $\langle \Gamma_{12}^n \rangle$ using the special model (13) for g.)

The reason for this lies in the nature of the integrands, which are

largest for maximum interparticle separations, while (g - 1) is

negligible except for small separations. We can find the correction

terms coming from the denominator, since from the definition of

$n(1....s)$ we have

$$\int \underline{d1}....\underline{ds}\, n(1...s) \;=\; \frac{A!}{(A-s)!}$$

and

$$A(A-1) \;=\; A^2(1- 1/A) \quad,\quad A(A-1)(A-2) \;=\; A^3\left(1- 3/A + O(1/A^2)\right)$$

$$A(A-1)(A-2)(A-3) \;=\; A^4\left(1- 6/A + O(1/A^2)\right)$$

so that the requirement of number conservation implies (cf. the previous result (19))

$$\langle \Gamma_{12}^4 \rangle \;=\; \langle \Gamma_{12}^4 \rangle_0 \left\{ 1+\tfrac{1}{A} + \leq O(\tfrac{1}{A^{4/3}}) \right\}$$

$$\langle \Gamma_{12}^2 \Gamma_{13}^2 \rangle \;=\; \langle \Gamma_{12}^2 \Gamma_{13}^2 \rangle_0 \left\{ 1+\tfrac{3}{A} + \leq O(\tfrac{1}{A^{4/3}}) \right\} \tag{19b}$$

$$\langle \Gamma_{12}^2 \Gamma_{34}^2 \rangle \;=\; \langle \Gamma_{12}^2 \rangle_0^2 \left\{ 1+\tfrac{6}{A} + \leq O(\tfrac{1}{A^{4/3}}) \right\}$$

Using these results we get

$$D = \left\langle \tfrac{9}{10}\Gamma_{12}^4 - \Gamma_{12}^2\Gamma_{13}^2 \right\rangle_0 + \tfrac{1}{10}\left\{ 3\langle\Gamma_{12}^4\rangle_0 - 18\langle\Gamma_{12}^2\Gamma_{13}^2\rangle_0 + 12\langle\Gamma_{12}^2\rangle_0^2 \right\}$$
$$+ O(A^{-1/3})$$

$$= \tfrac{2}{5}\left[(\overline{\Gamma^2})^2 - \overline{\Gamma^4} \right] \;+\; O(A^{-1/3})$$

(I have used equations 32 - 36 of [1] .)

So $D \sim R^4 A^{-1/3}$ if

$$\left[(\overline{\Gamma^2})^2 - \overline{\Gamma^4} \right] \;=\; 0 \tag{19c}$$

In the cutoff density approximation, (19c) is almost but not quite satisfied, instead I find

$$D \;=\; -\tfrac{2}{25}\left\langle \tfrac{9}{10}\Gamma_{12}^4 - \Gamma_{12}^2\Gamma_{13}^2 \right\rangle_0 \;+\; O(A^{-1/3})$$

For nuclei (i.e. $A \leq 250$), this result would give $\Delta E_2 \sim A^{-1/3}$ and not $\sim A^{-2/3}$, since the cancellation of the weak correlation limit terms is almost exact. However, for very large A the cutoff density approximation would lead to $D < 0$, i.e. $\Delta E_2 < 0$,

which is nonsense since ΔE_2 is positive definite by definition.
The explanation for this must be that the cutoff density approximation
is slightly inconsistent with the presence of short ranged correlations
(e.g. the surface thickness must be as large as the "hard" core diameter).
So it is quite possible that the real nucleon density will lead to (19c)
being satisfied exactly.

To sum up - the above argument (which is virtually a proof),
shows that the result derived in [1] , for the excitation energy of
Ψ_{2M} in the weak correlation approximation, is qualitatively
incorrect because of an unexpected error in the weak correlation
approximation. Namely, the inclusion of short ranged internucleon
correlations and the requirement of number conservation leads to the
weak correlation limit terms being cancelled, leaving ΔE_2
proportional to $A^{-1/3}$. The proportionality constant depends
on the physical nature of the correlations, which makes a calculation
of the true energy difficult, since four particle correlations are
involved.

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2) Rotational excitations of deformed even-even nuclei :

I will now consider the question of whether the isoscalar
wavefunctions (5) ($\Psi_{LM} = F_{LM} \Phi$), can represent rotational
states of deformed nuclei. That is, can we use wavefunctions of the
form $F\Phi$ when Φ is no longer spherically symmetric? This
discussion is based on speculation rather than rigor, but the results
obtained will lend substance to the speculation, which in turn will
make us think about the physical nature of rotational excitations
in deformed nuclei.

Deformed nuclei are found in regions away from closed shell configurations : (for $A > 30$) $150 \leq A \leq 188$ and $A \gtrsim 225$.

Odd A nuclei in these regions have large positive quadrupole moments (i.e. prolate or cigar shaped), and although even-even nuclei always have a zero ground state quadrupole moment (because the ground states of all even-even nuclei have the quantum numbers $I^{\pi} = 0^{+}$), the experimental results for the low lying states of even-even nuclei in the deformed regions imply an "intrinsic" deformation of the same order as for the neighbouring odd A nuclei. In particular, the low energy spectra of even-even nuclei in the deformed regions often consist of a sequence of states with $I^{\pi} = 0^{+}$, 2^{+}, 4^{+}, ... (where I is the total angular momentum and π is the parity) and energies approximating the formula

$$E (I) = C I (I + 1) \qquad \text{(C a constant)} \qquad (20a)$$

This formula is the well known expression for the quantized energy levels of a symmetric top rigid rotator (e.g. a diatomic molecule), rotating about an axis perpendicular to its symmetry axis. For the rigid rotator $C = \hbar^{2}/2\mathcal{J}_{rig}$, where $\mathcal{J}_{rig}$ is the rigid body value of the moment of inertia. Also, by requiring the symmetric top wave-functions to have positive parity it follows that the total angular momentum, relative to the rest frame, must be an even integer. So the experimental results for the low lying states of even-even nuclei in certain regions of A, strongly suggest the model of a prolate spheroid rotating about an axis perpendicular to its symmetry axis. The standard physical interpretation of a rotating deformed nucleus assumes that many-body correlations act to create a "slowly" varying rotation of a fixed shape, but that viewed from the body fixed frame the "fast" motion of the nucleons appears to be "erratic" and free of the many-body correlations (see p 385 of [5]). The collective nature

of these rotational states is verified by the extra large transition

rates for decay to the ground state. For a spherical system the

wavefunctions (5) apply only for even L and have the same parity as

the ground state Φ , and I will only be considering an extension to

deformed nuclei in which this continues to hold. Thus I am restricting

the discussion to the deformed even-even nuclei. Experimentally it is

found that $\mathcal{J}_{rig}$ is 2 to 5 times larger than the moments of inertia

calculated by assuming

$$E(I) = \frac{\hbar^2}{2\mathcal{J}} I(I+1) \qquad (20b)$$

So the deformed even-even nuclei are not rigid rotators, but instead

possess a non zero flow field in the body fixed frame. The opposite

extreme to rigid body flow is irrotational flow, which has a moment

of inertia ($\mathcal{J}_{irrot}$) given by equation 66 of section BII. The

irrotational flow field is better than rigid body flow in the sense

that $\mathcal{J}_{irrot}$ vanishes for zero deformation, as is found for nuclei,

but $\mathcal{J}_{irrot}$ turns out to be about four times smaller than the moment of

inertia of deformed even-even nuclei. That is, the amount of nuclear

matter transported during a single rotation is always found to lie

between the rigid body and irrotational flow values. However, from

the irrotational flow concept comes the interpretation of the rotation

as a large amplitude surface wave. As explained in note 7) of

section BII, a particular classical irrotational surface wave travelling

around the surface of a sphere, is equivalent to the rotation of a

prolate spheroid about an axis perpendicular to its axis of symmetry.

Furthermore, in section BII, arguments are given to show that the

(L,M) = (2,2) and (2,-2) states of the translationally variant wave-

functions (9), will describe such a surface wave travelling around

the Z axis. It was also argued that the (2,0) state can represent a

large amplitude vibration of a prolate spheroid, that preserves the

axial symmetry. The purpose of this section is to use the results of BII by assuming that the wavefunction $\Psi_{2M} = F_{2M} \Phi$ can be applied to deformed even-even nuclei, by taking the probability density Φ^2 to be that of a prolate spheroid.

The first problem that arises is that the ground states of all even-even nuclei are known to be 0^+ states, so that they must all be in some sense "spherical". In fact, as noted above, the experimentally determined quadrupole moment of the ground state of every even-even nucleus is zero. Some nuclear physicists (see for example [6] p 233), interpret the "spherical" properties of the deformed even-even nuclei as due to the uncertainty principle : the nucleus has an intrinsic quadrupole moment with respect to a symmetry axis but because of the uncertainty principle (orientation angle is conjugate to the angular momentum), the direction of the axis of symmetry is indeterminate, which means that an experiment to determine the quadrupole moment measures only the average over all space, i.e. zero. Consider the isoscalar wavefunctions (5), which are of the form

$$\Psi_{LM} = F_{LM} \Phi \tag{21}$$

If we continue to assume, as for the spherical case [1], that the spins are paired up so that $\underline{S}\,\Psi_{LM}$ and $\underline{S}\,\Phi$ are both zero, then the total angular momentum reduces to the orbital angular momentum and from [3] it follows that Ψ_{LM} is an eigenstate of angular momentum L provided $\underline{L}\,\Phi = 0$. However, it is not obvious that the uncertainty principle argument can be applied to a ground state such that $\underline{L}\,\Phi = 0$ as well as $\underline{I}\,\Phi = 0$. This is because the condition $\underline{L}\,\Phi = 0$ should imply that the ground state is rotationally invariant in space, i.e. not deformed. One possibility is that the ground states of even-even nuclei have $\underline{I}\,\Phi = 0$ but $\underline{L}\,\Phi = -\underline{S}\,\Phi \neq 0$.

To maintain a constant deformation throughout a rotational band we would need $\underline{S}\,\Psi_{LM} = \underline{S}\,\Phi$ and then perhaps the wavefunction (21) will still be an eigenstate of the total angular momentum as indicated by the subscripts (L,M). I shall avoid the problem by simply assuming that for even-even nuclei, Ψ_{LM} is an angular momentum eigenstate with eigenvalues (L,M), but that for the purposes of evaluating the energy for a deformed nucleus we can write

$$\Psi_{LM} = F_{LM}\,\Phi'$$
(22)

where Φ' describes a prolate spheroid. Another way of looking at this is to consider the question of orthogonality. For Ψ_{LM} to be a suitable trial wavefunction it must be orthogonal to the ground state Φ and this is guaranteed provided Ψ_{LM} has a different angular momentum eigenvalue than Φ. For example the Ψ_{2M} state is assumed to be a 2^+ state while Φ is of course a 0^+ state. However, the orthogonality condition can be expressed directly by

$$\langle F_{LM} \rangle = 0$$
(23)

where $\langle\ \rangle$ is the ground state expectation value defined by (4b). Equation (23) holds provided Φ^2 is spherically symmetric but when Φ is substituted by the deformed "ground" state Φ', (23) no longer holds, i.e.

$$\langle F_{LM} \rangle' \neq 0$$
(24)

where $\langle\ \rangle'$ is defined by

$$\langle B \rangle' = \frac{\int d1 \ldots dA\, B\, \Phi'^2}{\int d1 \ldots dA\, \Phi'^2}$$
(25)

The expression (24) together with equation (4a), implies a large drop in energy when Ψ_{LM} is applied to a deformed system in comparison with spherical systems.

The wavefunction (22) has some interesting similarities with the unified model of rotational collective motion. (The original theory is due mainly to Bohr and Mottelson, see [4], but I prefer to work from reference [5] p 386→.) The unified model is the main theory of nuclear rotations so far proposed by nuclear physicists, and is based on a Hamiltonian of the form

$$ H = H_{int} + T_{rot} + H_{coupl} \quad , \quad (26) $$

where H_{int} describes the intrinsic motion, T_{rot} describes the collective rotation and H_{coupl} represents the coupling between the two motions. In the zeroth order approximation, $H_{coupl} = 0$, the wavefunctions are of the form

$$ \Psi = F(\Theta_k) \, \Phi(\Gamma_i') \quad (27) $$

where Γ_i' are the coordinates with respect to the body fixed frame, which is defined by the angles Θ_k . $\Phi(\Gamma_i')$ is taken to be a stater determinant of single particle wavefunctions in a deformed potential whose orientation is given by Θ_k . So the wavefunctions (22) and (27) both assume a deformed "ground" state Φ' and it seems plausible to suggest that the F_{LM} of (22), which is a function of the particle coordinates in the non rotating frame, performs the same role as the $F(\Theta_k)$ of (27), which is a function of a set of collective coordinates. This idea is supported by the fact that $F(\Theta_k)$ (the eigenfunction of T_{rot}) is a symmetric top eigenfunction :

$$ F(\Theta_k) = \sqrt{\frac{2I+1}{8\pi^2}} \; D^{I\,*}_{MK}(\Theta_k) \quad (28) $$

$F(\Theta_k)$ is an eigenstate of I^2, I_z and $I_{z'}$ (z' is the rotating axis of symmetry), with eigenvalues $I(I+1)\hbar^2$, $M\hbar$ and $K\hbar$, respectively. For the lowest states, $K = 0$ (which will be the only case that I consider), (28) reduces to

$$F(\Theta_k) = \frac{1}{\sqrt{2\pi}} Y_{IM}(\Theta_k)$$,

which gives a direct connection with the wavefunctions (5) and (9), except that the collective coordinates Θ_k are replaced by a sum over the nucleon coordinates in the non rotating frame. Furthermore, the total angular momentum of $\Phi(r_i')$ is zero, because a rotation of the system does not change the numerical value of any of the internal coordinates. This is the corresponding statement to my assumption that $I \Phi' = 0$, for the wavefunction (22). Another interpretation of wavefunctions like (22) and (27) is to regard them as angular momentum projections of a deformed intrinsic state Φ' , where F_{LM} is the projection operator (see [4] p 90 and [5] p 461).

I will investigate the particular set of wavefunctions

$$\Psi_{2M} = F_{2M} \Phi'$$ (29)

where F_{2M} is given by (5) (with $L = 2$ & $f = 1$), or its translationally variant form (9), and Φ' is a deformed "ground" state, assumed to be a prolate ellipsoid. In [1] it was noted that for Φ' spherically symmetric, all the five states of the set (29) (i.e. M = -2,-1,0,1,2) are degenerate. However, when Φ' is deformed this degeneracy is broken. For example, if Φ' has axial symmetry (i.e. an ellipsoid of revolution), the five fold degeneracy splits up into two sets of two fold degenerate states, (2,2),(2,-2) and (2,1),(2,-1), plus the (2,0) state. I have previously noted from an argument in note 7) of section BII implying the nature of the irrotational flow field corresponding to wavefunctions of the type (9), that it follows that

the (2,2) and (2,-2) states represent a rotation of an ellipsoid of fixed shape, and the (2,0) state should represent a large amplitude vibration of an ellipsoid, that preserves the axial symmetry, while the (2,1) and (2,-1) states can only describe small amplitude surface waves. This broken degeneracy is associated with the fact that shape deformations of quadrupole symmetry exhibit both rotational and vibrational degrees of freedom (see [4] p 677 -). On page 682 of [4], it is stated that for an equilibrium shape with axial symmetry, the five quadrupole degrees of freedom separate into two rotational modes and three vibrational modes, one of which preserves the axial symmetry (β vibration) while the other two oscillate away from axial symmetry (γ vibrations). This appears to correspond exactly to the physical interpretation of the wavefunction (29), based on the argument of section BII.

To evaluate the variational energies of the states (29) I shall use the weak correlation approximation. This will lead to quantitative errors, but because of (24) there is no longer the possibility of the qualitative error that seems to have occurred for spherical nuclei. This is because the four body terms of $\langle |F_{2M}|^2 \rangle$, which are multiplied by an extra factor of A more than the other terms in the energy denominator, now dominate even in the weak correlation approximation, whereas for the spherical case they are approximated by zero. Another point to consider is whether or not the translationally invariant form of (29) is free of redundant coordinates when Φ' is not spherically symmetric. A translationally invariant wavefunction guarantees that the centre of mass motion is treated correctly but it is not obvious whether or not there are now redundant variables due to the broken rotational invariance. For example, in the unified model the introduction of the collective coordinates Θ_k , defining the body fixed frame, produces three redundant coordinates, which

cannot be completely removed because of the presence of the coupling term in the Hamiltonian (26), i.e. Coriolis forces. (See [5] pp 387-9.) { aside: From now on I will drop the dashed superscripts on the wavefunctions (9), so that Ψ_{LM} will refer either to the translationally invariant states (5) (with L = 2 & f = 1), or to the translationally variant states (9). It was shown in [1] for the spherically symmetric case, that in the weak correlation approximation, breaking translational invariance leads to an error in ΔE_2 of order $1/A$ only. }

(i) The (2,2) and (2,-2) states :

Consider the wavefunction $\Psi_{2 \pm 2} = F_{2 \pm 2} \Phi'$, where Φ' is the deformed ground state and

$$F_{2 \pm 2} = \sum_{j=1}^{A} (x_j \pm i y_j)^2 \qquad (30)$$

From the results of note 7) of section BII we expect Ψ_{22} and Ψ_{2-2} to represent an ellipsoid of revolution rotating about an axis (the Z axis) perpendicular to its symmetry axis (M = -2 is just the opposite rotation to M = 2). Because the rotational energy must be a constant of motion, I will assume that the energy of $\Psi_{2 \pm 2}$ can be calculated at the particular instant when the axis of symmetry coincides with the X axis. The variational bound to the rotational energy, $\Delta E_{2 \pm 2}$, is still given by the expression (4a) except that the expectation value is now over the deformed probability density Φ'^2 , i.e. $\langle \rangle'$ as defined by (25). From (30) we have

$$\langle |\nabla_1 F_{2 \pm 2}|^2 \rangle' = 8 \langle x_1^2 + y_1^2 \rangle' \qquad (31)$$

and

$$\langle |F_{2 \pm 2}|^2 \rangle' = A \langle (x_1^2 + y_1^2)^2 \rangle' + A(A-1) \langle (x_1^2 - y_1^2).$$
$$(x_2^2 - y_2^2) + 4 x_1 y_1 x_2 y_2 \rangle' \qquad (32)$$

In the weak correlation approximation (32) becomes

$$\langle |F_{\frac{2}{2} \pm 1}|^2 \rangle' \simeq A \langle (x_1^2 + y_1^2)^2 \rangle' + A(A-1) \langle x_1^2 - y_1^2 \rangle'^2 \qquad (33)$$

So in the limit of large A or large deformation or both, the last term of (33) dominates and from (4a), the variational energy is given by

$$\Delta E_{2 \pm 2} \lesssim \frac{4 \hbar^2}{Am} \frac{\langle x_1^2 + y_1^2 \rangle'}{\langle x_1^2 - y_1^2 \rangle'^2} \qquad (34)$$

$$= 2(2+1) \frac{\hbar^2}{2 \mathcal{I}_{2 \pm 2}}$$

where

$$\mathcal{I}_{2 \pm 2} = \frac{3}{4} Am \frac{\langle x_1^2 - y_1^2 \rangle'^2}{\langle x_1^2 + y_1^2 \rangle'} \qquad (35)$$

From equations (64) and (66) of BII we see that (35) is 3/4 times the irrotational moment of inertia of an ellipsoid of revolution symmetric about the X axis and rotating about the Z axis. Note that if the angular momentum was taken to be the "classical" value $4\hbar^2$ instead of $6\hbar^2$, then we would have $\mathcal{I}_{2 \pm 2} = \mathcal{I}_{irrot}$. The value of (35) is about five times smaller (i.e. $\Delta E_{2 \pm 2}$ is about five times larger) than the experimental results for the first 2^+ states of deformed even-even nuclei. It is plausible to suggest that the poor value of the energy is due to the weak correlation approximation. The inclusion of internucleon correlations (starting with pair correlations) will probably lower the energy but it remains to be proved whether this will explain the entire factor of five. On page 427 of reference [5] it is stated that for nuclei between closed shells, that the correlations due to statistics are responsible for the moment of inertia being much larger than the irrotational flow value. However, a pessimist would suggest that the wavefunction (30)

describes irrotational flow only. For comparison with (35), the rigid body moment of inertia is

$$\mathcal{J}_{rig} = Am \langle x_1^2 + y_1^2 \rangle' \qquad , \qquad (36)$$

which is two to five times larger than the experimental values for deformed even-even nuclei.

The simplest way to evaluate the deformed system expectation values is to use the scaling procedure described in section CIII. Following the method of equations (1 - 5) and (16) of CIII, we deform Φ into Φ' by scaling the coordinates :

$$\Phi' = \Phi (\mathbf{r}_1', \ldots, \mathbf{r}_A') \qquad (37)$$

where $\mathbf{r}'$ varies over the spherical system, so that $\mathbf{r}$ now varies over the deformed system, i.e. in the expression (25) for $\langle \; \rangle'$ the integration is over the deformed system. To deform a sphere into a prolate spheroid symmetric about the X axis, we write

$$x = \eta^2 x' \; , \quad y = \eta^{-1} y' \; , \quad z = \eta^{-1} z' \qquad (38)$$

where $\eta \geq 1$ (i.e. a large deformation is $\eta \to \infty$).
From (38) we have $dx\,dy\,dz = dx'\,dy'\,dz'$, so that

$$\langle B' \rangle' = \langle B \rangle \qquad (39)$$

where B' stands for B written in dashed coordinates and $\langle \; \rangle$ is the spherical system expectation value (i.e. $\langle x^2 \rangle = \langle y^2 \rangle = \langle z^2 \rangle$, etc.).

$\{$ aside: In ref. [5] pp 388-91, it is stated that for (x', y', z') to represent a body fixed frame of reference, we must have

$$\sum_j x_j' y_j' = \sum_j y_j' z_j' = \sum_j z_j' x_j' = 0 \qquad (40)$$

The unified model requires (38) to hold only on the average, i.e.

$$\langle x_i' y_i' \rangle' = \langle y_i' z_i' \rangle = \langle z_i' x_i' \rangle = 0 \qquad (41)$$

From equation (39) we see that (41) would hold for any deformation. Equation (39) also suggests an explanation as to what the orthogonality condition is for wavefunctions of the type (22). Namely, from (39) and (23) we have

$$\langle F_{LM}' \rangle' = \langle F_{LM} \rangle = 0 \qquad , \qquad (42)$$

which should be compared with (24). $\Big\}$

We can now use (38) and (39) to evaluate the expressions (35) and (36) in terms of the deformation parameter η. The calculations rely on the results of reference [1] for the values of expectation values like $\langle x_i^2 y_i^2 \rangle$, $\langle x_i^2 \rangle$, $\langle x_i^4 \rangle$, ... , etc., in terms of

$$\langle r^n \rangle \equiv \frac{\int_0^\infty dr\, r^2 n(r)\, r^n}{\int_0^\infty dr\, r^2 n(r)} \qquad , \qquad (43)$$

where $n(r)$ is the radial number density of the spherically symmetric system. Also, I will use the cutoff density approximation

$$n(r) = \begin{cases} A/V & r < R \\ 0 & r > R \end{cases} \quad , \quad V = \tfrac{4}{3}\pi R^3 \qquad (44)$$

to evaluate the expression (43), i.e. I take

$$\langle r^n \rangle = \frac{3}{n+3} R^n \qquad (45)$$

(Note, a good approximation for most nuclei is R = $1.2 A^{1/3} fm$.)

Then, using (38) and (39) it follows that

$$g_{rig} = Am \langle \eta^4 x_i^2 + \eta^{-2} y_i^2 \rangle = Am \frac{R^2}{5} \left(\eta^4 + \eta^{-2} \right) \qquad (46)$$

and

$$\mathcal{I}_{irrot} = Am \frac{\langle \eta^4 x_1^2 - \eta^{-2} y_1^2 \rangle^2}{\langle \eta^4 x_1^2 + \eta^{-2} y_1^2 \rangle} = Am\frac{R^2}{5} \frac{(\eta^4 - \eta^{-2})^2}{(\eta^4 + \eta^{-2})} \quad (47)$$

These results are another derivation of equations (66) and (67) of

section BII. It is also easy to evaluate the intrinsic quadrupole

moment :

$$Q_0 = Z \langle 2x_1^2 - y_1^2 - z_1^2 \rangle' = \frac{2}{5} Z R^2 (\eta^4 - \eta^{-2}) \quad (48)$$

where Z is the number of protons. For the purpose of plotting graphs

it is more convenient to define a deformation parameter that varies

from 0 to 1 instead of from 1 to ∞ , as the deformation increases.

For this reason I introduce

$$d \equiv 1 - \eta^{-3} \quad (49)$$

I choose this particular deformation parameter because for small

deformations it is close to the standard parameters δ and β

(see p 416 of reference [5]). δ is defined as the difference in

length between the single and the double axis of the prolate spheroid,

measured in units of R; i.e.

$$\delta = \eta^2 - \eta^{-1}$$
$$= d + O(d^2) \quad (50)$$

β is related to δ by the expression

$$\delta = \sqrt{\frac{45}{16\pi}} \, \beta \simeq 0.946\beta \quad (51)$$

Using (49 - 51) in the results (46 - 48) gives

$$\mathcal{I}_{rig} = \frac{2}{5} AmR^2 \left(1 + \frac{d}{3} + O(d^2)\right) = \frac{2}{5} AmR^2 \left(1 + 0.315\beta + O(\beta^2)\right) \quad (52)$$

$$\mathcal{I}_{irrot} = \frac{2}{5} A m R^2 d^2 \left(1 + O(d)\right) = \frac{9}{8\pi} A m R^2 \beta^2 \left(1 + O(\beta)\right) \tag{53}$$

and

$$Q_0 = \frac{4}{5} Z R^2 d \left(1 + O(d)\right) = \frac{3}{\sqrt{5\pi}} Z R^2 \beta \left(1 + O(\beta)\right) \tag{54}$$

Evaluating the expressions (31) and (33) by the above method, the complete expression for $\Delta E_{2 \pm 2}$ in the weak correlation approximation is

$$\Delta E_{2 \pm 2} \leq \frac{\hbar^2}{m R^2} \frac{4 \left(\eta^4 + \eta^{-2}\right)}{\left\{\frac{1}{7} \left(3\eta^8 + 2\eta^2 + 3\eta^{-4}\right) + \frac{(A-1)}{5} \left(\eta^4 - \eta^{-2}\right)^2\right\}} \tag{55}$$

In terms of the moment of inertia, the result (55) is

$$\mathcal{I}_{2 \pm 2} = 3 m R^2 \frac{\left\{\frac{1}{7} \left(3\eta^8 + 2\eta^2 + 3\eta^{-4}\right) + \frac{(A-1)}{5} \left(\eta^4 - \eta^{-2}\right)^2\right\}}{4 \left(\eta^4 + \eta^{-2}\right)} \tag{56}$$

I will now derive the variational energy bound for the translationally <u>in</u>variant form of $\Psi_{2 \pm 2}$, i.e.

$$F_{2 \pm 2} = \frac{1}{2} \sum_{i}^{A} \sum_{j}^{A} \left(x_{ij} \pm i y_{ij}\right)^2 \tag{57}$$

In the weak correlation approximation breaking the translational invariance (i.e. using (30) instead of (57)) leads to errors in $\Delta E_{2 \pm 2}$ of order $1/A$ only, but it is worth deriving the exact expression because this is the first step towards including the effect of the nucleon pair correlations (see the method used in [2] to include pair correlations in the calculation of the giant dipole resonance energy). Following the method of section two of reference [1]

we have

$$\left\langle |\nabla_1 F_{2\pm2}|^2\right\rangle' = \frac{4}{3} A(A-1)\left\langle r_{12}^2\right\rangle\left(\eta^4+\eta^{-2}\right) \tag{58}$$

and

$$\left\langle |F_{2\pm2}|^2\right\rangle' = \frac{1}{2} A(A-1)\left\langle\left(\eta^4 x_{12}^2+\eta^{-2}y_{12}^2\right)^2\right\rangle$$

$$+ A(A-1)(A-2)\left\langle\left(\eta^4 x_{12}^2-\eta^{-2}y_{12}^2\right)\left(\eta^4 x_{13}^2-\eta^{-2}y_{13}^2\right)+4\eta^2 x_{12}y_{12}x_{13}y_{13}\right\rangle \tag{59}$$

$$+\frac{1}{4} A(A-1)(A-2)(A-3)\left\langle\left(\eta^4 x_{12}^2-\eta^{-2}y_{12}^2\right)\left(\eta^4 x_{34}^2-\eta^{-2}y_{34}^2\right)+4\eta^2 x_{12}y_{12}x_{34}y_{34}\right\rangle$$

The expression (59) can be reduced to expectation values involving only r_{ij} terms, by using equations 14, 16, 17, 20, 21, 23, 28, and 29, of reference [1]. The final result for the variational energy is

$$\Delta E_{2\pm2} \leq \frac{\frac{\hbar^2}{m} A\left\langle r_{12}^2\right\rangle\left(\eta^4+\eta^{-2}\right)}{\left\{\frac{\left\langle r_{12}^4\right\rangle}{20}\left(3\eta^8+2\eta^2+3\eta^{-4}\right)+(A-2)\left[\frac{3}{20}\left\langle r_{12}^4\right\rangle\left(\eta^8+4\eta^2+\eta^{-4}\right)-\left\langle r_{12}^2 r_{13}^2\right\rangle\eta^2\right]\right.}$$
$$\left. +\frac{1}{20}(A-2)(A-3)\left[\left\langle r_{12}^4-2r_{12}^2 r_{13}^2\right\rangle\left(\eta^8+4\eta^2+\eta^{-4}\right)+\frac{1}{2}\left\langle r_{12}^2 r_{34}^2\right\rangle\left(3\eta^8+2\eta^2+3\eta^{-4}\right)\right]\right\}} \tag{60}$$

In the spherical limit, i.e. $\eta=1$, the expression (60) reduces to equation (31) of reference [1]. It is also easy to show that in the weak correlation approximation (i.e. $\left\langle r_{12}^2\right\rangle = 2\left\langle r_1^2\right\rangle$,

$$\left\langle r_{12}^4\right\rangle = 2\left\langle r_1^4\right\rangle + \frac{10}{3}\left\langle r_1^2\right\rangle^2, \quad \left\langle r_{12}^2 r_{13}^2\right\rangle = \left\langle r_1^4\right\rangle + 3\left\langle r_1^2\right\rangle^2 \quad \text{and}$$

$$\left\langle r_{12}^2 r_{34}^2\right\rangle = 4\left\langle r_1^2\right\rangle^2 \qquad , \text{ see [1]) and using (45), that}$$

(60) differs from (55) by terms of order $1/A$ only, for all values of η. The result (60) is a rigorous expression for the expectation value of the energy of the state $\Psi_{2\pm2}$, but to evaluate it exactly would require knowledge of the two-, three- and four-particle correlation functions. For this reason it may be simpler to use the translationally variant expression (see (31) and (32)), which has only two particle correlations, although they are

no longer of the form $\langle \Gamma_{12}^n \rangle$. The hope is, that if the nucleon

correlations are included, then $\Delta E_{2 \pm 2}$ will be lower by a factor

of about five and so reproduce the experimental results. An

important point to note about the above scaling method, is that the

energy of the deformed system is expressed in terms of expectation

values in the spherical system, so that a knowledge of the effect

of correlations in spherical nuclei is all that is required.

{ NOTE ADDED IN "PROOF" : The expressions (19b) are too small to

change the value of $\Delta E_{2 \pm 2}$ by more than about 10%, not 500%.

However, it is possible that the scaling method of evaluating

$\Delta E_{2 \pm 2}$ for deformed even-even nuclei is not capable of including

the correct correlations in deformed nuclei. That is, the result (60)

will be invalid if deformed nuclei have extra correlations that are

not taken into account by simply scaling the spherical system. }

(ii) The (2,0) state :

I will now discuss the wavefunction $\Psi_{20} = F_{20} \Phi'$,

where

$$F_{20} = \sum_{j=1}^{A} \left(2z_j^2 - x_j^2 - y_j^2 \right)$$

(61)

From the results of note 7) of section BII, we expect Ψ_{20} to

represent a vibration of a non rotating prolate spheroid, that

preserves the axial symmetry about the Z axis. However, it is also

worth investigating the result of applying F_{20} to the same deformed

"ground" state as we did with $F_{2 \pm 2}$. That is, I will consider

two possibilities; the first (labelled (Z)) regards Ψ_{20} as a

vibrating ellipsoid symmetric about the Z axis, while the second

case (labelled (X)) assumes Ψ_{20} to be an ellipsoid rotating about the

Z axis, at the instant at which the symmetry axis is the X axis. The second alternative is probably wrong but it may be equivalent to the unified model assumptions. For the second case (X), Φ' is defined by (38), while for the first case we write

$$(Z): \quad X = \eta^{-1} x' \;, \quad y = \eta^{-1} y' \;, \quad z = \eta^{2} z' \tag{62}$$

From (61) it follows that

$$\left\langle (\nabla_1 F_{20})^2 \right\rangle' = 4 \left\langle 4 z_1^2 + x_1^2 + y_1^2 \right\rangle' \tag{63}$$

and, in the weak correlation approximation

$$\left\langle F_{20}^2 \right\rangle' = A \left\langle (2z_1^2 - x_1^2 - y_1^2)^2 \right\rangle' + A(A-1) \left\langle 2z_1^2 - x_1^2 - y_1^2 \right\rangle'^2 \tag{64}$$

Using (45) together with (62) and (38), we have for the two cases

$$(Z) \begin{cases} \left\langle (\nabla_1 F_{20})^2 \right\rangle' = \dfrac{4}{5} R^2 \left(4\eta^4 + 2\eta^{-2} \right) & \text{(65a)} \\[3mm] \left\langle F_{20}^2 \right\rangle' = \dfrac{A}{35} R^4 \left(12\eta^8 - 8\eta^2 + 8\eta^{-4} \right) + A(A-1)\dfrac{4}{25} R^4 \left(\eta^4 - \eta^{-2} \right)^2 & \text{(65b)} \end{cases}$$

$$(X) \begin{cases} \left\langle (\nabla_1 F_{20})^2 \right\rangle' = \dfrac{4}{5} R^2 \left(\eta^4 + 5\eta^{-2} \right) & \text{(66a)} \\[3mm] \left\langle F_{20}^2 \right\rangle' = \dfrac{A}{35} R^4 \left(3\eta^8 - 2\eta^2 + 11\eta^{-4} \right) + A(A-1)\dfrac{R^4}{25} \left(\eta^4 - \eta^{-2} \right)^2 & \text{(66b)} \end{cases}$$

Defining a "moment of inertia" $\mathcal{J}_{20}$, by

$$\mathcal{J}_{20} \equiv \frac{3\hbar^2}{\Delta E_{20}} = \frac{6m \left\langle F_{20}^2 \right\rangle'}{A \left\langle (\nabla_1 F_{20})^2 \right\rangle'} \tag{67}$$

I find

$$(Z): \quad \mathcal{J}_{20} = \frac{6mR^2 \left\{ \tfrac{1}{7} \left(12\eta^8 - 8\eta^2 + 8\eta^{-4} \right) + \tfrac{4}{5}(A-1)\left(\eta^4 - \eta^{-2} \right)^2 \right\}}{4 \left(4\eta^4 + 2\eta^{-2} \right)} \tag{68a}$$

$$(x): \quad g_{20} = \frac{6mR^2\left\{\frac{1}{7}(3\eta^8 - 2\eta^2 + 11\eta^{-4}) + \frac{1}{5}(A-1)(\eta^2 - \eta^{-2})^2\right\}}{4(\eta^4 + 5\eta^{-2})} \qquad (68b)$$

For $\eta \to 0$ and $\eta \to \infty$ (68a) and (68b) become identical.

(iii) The (2,1) and (2,-1) states :

Consider the wavefunctions $\Psi_{2\pm 1} = F_{2\pm 1} \Phi'$, where

$$F_{2\pm 1} = \sum_{j=1}^{A} z_j (x_j \pm i y_j) \qquad (69)$$

The two body term of $\langle |F_{2\pm 1}|^2 \rangle'$ is

$$A(A-1)\langle z_1 z_2 (x_1 x_2 + y_1 y_2)\rangle' \qquad (70)$$

In the weak correlation approximation (70) reduces to

$$A(A-1)\left\{ \langle z_1 x_1 \rangle'^2 + \langle z_1 y_1 \rangle'^2 \right\} \qquad (71)$$

From (71) we see that for all deformations of the type (38) and (62), that in the weak correlation approximation the two body term in the energy denominator is zero. So the two states Ψ_{21} and Ψ_{2-1} remain of order A (or perhaps $O(A^{4/3})$) higher in energy than the first 2^+ states of deformed even-even nuclei. That is, $\Delta E_{2\pm 1}$ deviates only slightly from its value for spherical nuclei, as Φ' is deformed. This corresponds directly to the conclusion in note 7) of section BII, that $\Psi_{2\pm 1}$ could only represent small amplitude surface waves and not large amplitude modes like $\Psi_{2\pm 2}$ and Ψ_{20}. From the previous discussion on the Ψ_{2M} state of spherical nuclei, it seems plausible that the $\Psi_{2\pm 1}$ wavefunctions will represent the effect of a deformation on the giant quadrupole resonance.

In Fig. 1 (next page), I have used equations (36), (56), and (68) to plot $\mathcal{J}_{2M}/\mathcal{J}_{rig}$ for M = -2,0,2. For the small A dependent terms I took A = 170, which is the mean value of A for the main deformed region $150 \leq A \leq 188$. The experimental results for the deformed even-even nuclei in this region are taken from Bohr and Mottelson, [7]. There are two important features of Fig. 1. Firstly, the fact that $\mathcal{J}_{20}$ is greater than $\mathcal{J}_{rig}$ for very large deformations, supports the identification of Ψ_{20} as a vibrational state and not a rotational state, since rigid body rotation should have the lowest energy of all rotational modes. Interpreting Ψ_{20} as a vibration along the Z axis also explains why $\mathcal{J}_{20}(z)$ is always larger than $\mathcal{J}_{20}(x)$, since the lowest energy vibration of a prolate spheroid should be along the symmetry axis. I have already discussed the other main feature of Fig. 1, namely that if we assume that $\Psi_{2\pm2}$ represents a prolate spheroid rotating about an axis perpendicular to its symmetry axis, then $\mathcal{J}_{2\pm2}$ is about five times lower than that required to explain the experimental energies of the low lying states of the deformed even-even nuclei. The next step (which I will leave for the future) is to include the internucleon correlations (e.g. the effect of Fermi statistics), which hopefully will reduce the energy to the experimental values. One other possibility is that the wavefunctions could be improved by returning to the L = M = 2 state of the form (5) and minimising the energy with respect to a general $f(r_{ij})$ (the calculations of this section have taken f = 1). However, for the analogous translationally variant form, the arguments of section BII suggest that f = 1 is the optimum wavefunction, since f $\neq$ 1 should involve incompressible flow.

Fig. 1.
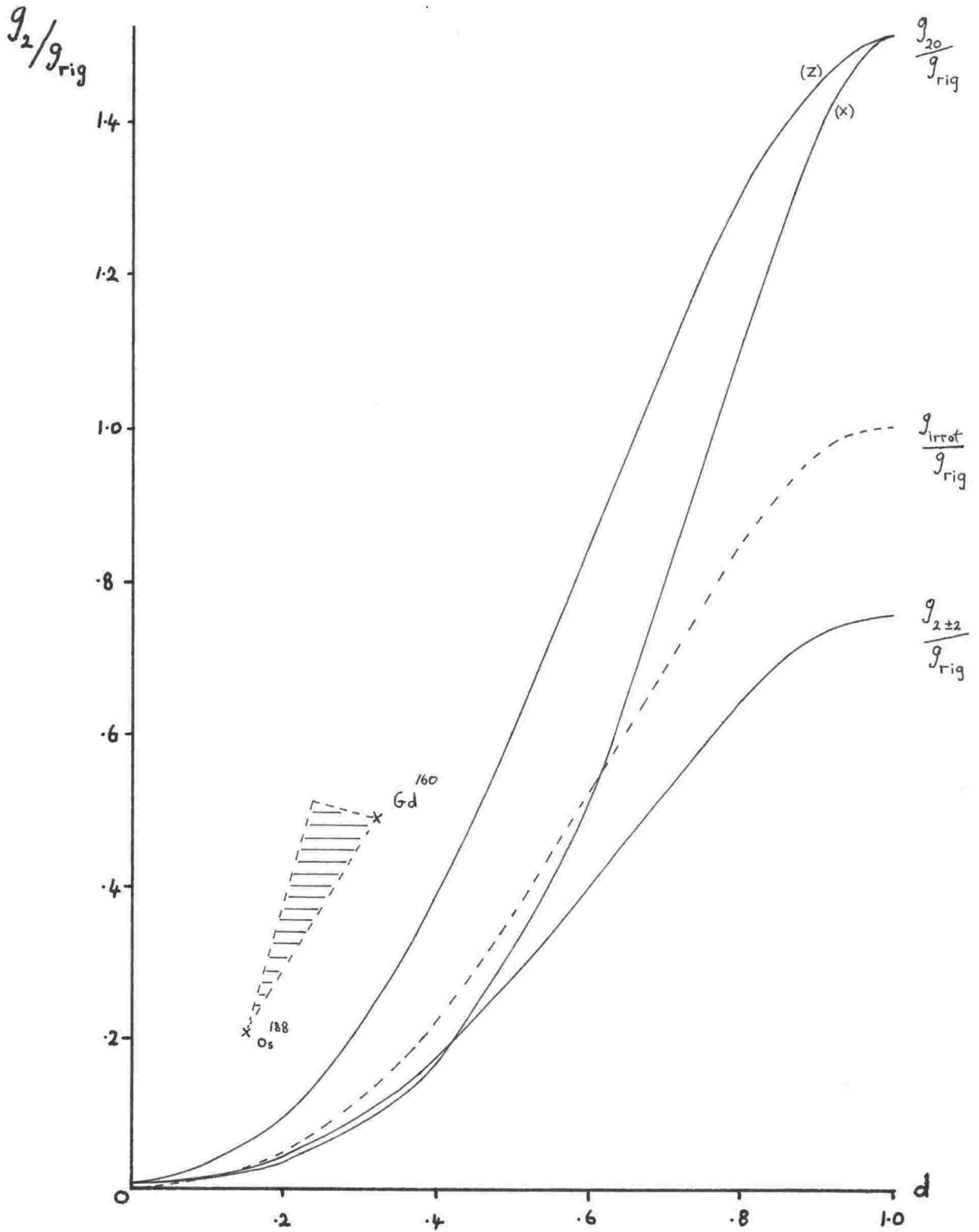
"Moments of inertia" for even-even nuclei in units of the rigid body value, plotted as a function of the nuclear deformation. The solid lines are defined by $\mathcal{J}_{2M} = 3\hbar^2 / \Delta E_{2M}$. The dashed line is the irrotational moment of inertia of a rotating spheroid. The shaded area covers the experimental results in the region $150 \leq A \leq 188$ (taken from Bohr and Mottelson (1955), [7]).

Transition Rates :

I will now use the weak correlation approximation to evaluate the transition rates for decay to the "deformed" ground state Φ' , and compare the results with the unified model prediction. Firstly, I note from equation 16 of section AIV, that the contribution to the $S_2^{(1)}$ sum rule from the five Ψ_{2M} states will each remain approximately $\frac{1}{5} \cdot \frac{Z}{A}$ of the total sum rule, for deformations applicable to nuclei, (ignoring velocity dependent and charge exchange interactions). Because the contributions to the sum rule are proportional to the transition rate times the excitation energy and since I have shown that for large deformations that $\Delta E_{2\pm1}$ is of the order of A (or perhaps $A^{4/3}$) larger than $\Delta E_{2\pm2}$ and ΔE_{20} , it follows that for the deformed even-even nuclei, that almost all of the transition rate strength will be taken up by the M = 2,0,-2 states. The transition rate probability, per unit time, for the emission of a photon of multipolarity (2,M) and energy $\hbar\omega = \hbar c k$, during which the nucleus decays from the Ψ_{2M} state to the ground state, is given by (cf. section four of [1])

$$ T_E(2,M) = \frac{4\pi}{75} \frac{k^5}{\hbar} \left| \langle f | Q_{2M} | i \rangle \right|^2 \tag{72} $$

where

$$ Q_{2M} = e \sum_{j=1}^{Z} r_j^2 Y_{2M}^*(\underline{r}_j) \tag{73} $$

$$ | i \rangle = N_i^{-1} F_{2M} \Phi' = N_i^{-1} \alpha_M^{-1} \sum_{j=1}^{A} r_j^2 Y_{2M}(\underline{r}_j) \Phi' \tag{74} $$

(For F_{2M} defined by (30) and (61) we have $\alpha_{\pm2} = \sqrt{\frac{15}{32\pi}}$ and $\alpha_0 = \sqrt{5/16\pi}$)

$$ | f \rangle = N_f^{-1} \Phi' \tag{75} $$

Φ' is the deformed "ground" state and N_i^{-1} and N_f^{-1} are normalization factors defined by

$$\langle i | i \rangle = \langle f | f \rangle = 1 \tag{76}$$

Using (73) - (76) I calculate

$$|\langle f | Q_{2m} | i \rangle|^2 = e^2 \frac{Z^2}{A^2} \frac{\alpha_m^2}{N_i^2 N_f^2} \left| \int d1 \dots dA \, |F_{2m}|^2 \, {\Phi'}^2 \right|^2 \tag{77}$$

$$= e^2 \frac{Z^2}{A^2} \alpha_m^2 \langle | F_{2m}|^2 \rangle'$$

The factor Z^2 / A^2 comes from the fact that Q_{2M} is a sum over the protons only, while the isoscalar wavefunction $|i\rangle$ is a sum over all the nucleons. From (72) and (77) we have

$$T_E(2,M) = \frac{4\pi}{75} \frac{k^5}{\hbar} \frac{Z^2}{A^2} e^2 \alpha_M^2 \langle | F_{2M}|^2 \rangle' \tag{78}$$

(i)   The (2,2) and (2,-2) States :

Comparing (30) with (74) we have $\alpha_{\pm 2} = \sqrt{\frac{15}{32\pi}}$. So

$$T_E(2, \pm 2) = \frac{1}{40} \frac{Z^2 e^2}{A^2} \frac{k^5}{\hbar} \langle | F_{2 \pm 2}|^2 \rangle' \tag{79}$$

In the weak correlation approximation and using (45), the result (79) becomes (see (55))

$$T_E(2, \pm 2) = \frac{1}{40} \frac{Z^2 e^2}{A} \frac{k^5}{\hbar} R^4 \left\{ \frac{1}{35} (3\eta^8 + 2\eta^2 + 3\eta^{-4}) + \frac{(A-1)}{25} (\eta^4 - \eta^{-2})^2 \right\} \tag{80}$$

In the limit $\eta \to 1$ (i.e. Φ' spherical), (80) reduces to equation (55) of [1] . For large deformations we can ignore the first term of (80), and from the expression (48) for the intrinsic quadrupole moment Q_o , I find

$$T_E(2, \pm 2) \simeq \frac{e^2}{160} \frac{k^5}{\hbar} Q_o^2 \tag{81}$$

For the experimental results plotted in Fig. 1. the maximum error involved in using (81) instead of the full expression (80), is about 10%.

(ii) The (2,0) State :

Comparing (61) with (74) we have $\alpha_o = \sqrt{\frac{5}{16\pi}}$. So

$$T_E(2,0) \;=\; \frac{1}{60}\,\frac{Z^2 e^2}{A^2}\,\frac{k^5}{\hbar}\,\left\langle F_{2o}^2 \right\rangle' \tag{82}$$

From equations (65b) and (66b), the results corresponding to (81) for the two deformations previously considered (i.e. (Z) and (X)), are

$$(Z): \qquad T_E(2,0) \;=\; \frac{e^2}{60}\,\frac{k^5}{\hbar}\,Q_o^2 \tag{83}$$

$$(X): \qquad T_E(2,0) \;=\; \frac{e^2}{240}\,\frac{k^5}{\hbar}\,Q_o^2 \tag{84}$$

The expressions (81), (83) and (84) are very similar to the unified model result (see [5] p 411, and [8])

$$T_E(2) \;=\; \frac{e^2}{300}\,\frac{k^5}{\hbar}\,Q_o^2 \tag{85}$$

This is a strong indication that the wavefunctions Ψ_{2M} are closely related to the unified model wavefunctions. It is worth pointing out that exact agreement is obtained with the unified model result (85), if we take the average transition rate for all of the five Ψ_{2M} states, when Φ' is assumed to be the rotating ellipsoid defined by (38), (i.e. case (X)). That is, $T_E(2,\pm1)$ are of order A (or perhaps $A^{4/3}$) smaller and so can be ignored, and $\frac{1}{5}\left(\frac{2}{60} + \frac{1}{240}\right) = \frac{1}{300}$. This suggests the possibility that the unified model result has ignored the broken degeneracy of the five Ψ_{2M} states. Since equation (85) is regarded as the total transition rate to the ground state, from the first

rotational level of a deformed even-even nucleus, it is quite possible that an average over non degenerate M levels has been included.

Remarks in Summary :

The isoscalar wavefunctions $\Psi_{2M} = F_{2M} \Phi'$, where Φ' is assumed to be a deformed "ground" state, are reasonably successful in describing the low lying 2^+ states of deformed even-even nuclei. The broken degeneracy is explained by the results of section BII, namely Ψ_{22} and Ψ_{2-2} represent the rotation of a nucleus about an axis perpendicular to its axis of symmetry, Ψ_{20} is identified as a large amplitude vibration that preserves the axial symmetry, while Ψ_{21} and Ψ_{2-1} are small amplitude high energy surface waves. The results for the transition rates for decay to the ground state are particularly encouraging, since they are very close to the prediction of the unified model. In the weak correlation approximation the rotational excitation energy $\Delta E_{2 \pm 2}$ is slightly higher than the irrotational flow value for a rotating ellipsoid, but there is some hope that the inclusion of internucleon correlations will lower the energy by the factor of five necessary to explain the experimental results.

A complete description of the low lying states of deformed even-even nuclei requires the identification of the entire band of rotational states, since the most important feature of the spectra of deformed even-even nuclei is the occurrence of sequences of states with energies approximating the formula (20), i.e.

$E(I) \sim I(I+1)$. At first sight one might be tempted to suggest that the I = 4,6,8,etc, wavefunctions are all of the form (5). For example, the 4^+ state would then be

$$\Psi_{4M} = F_{4M} \Phi' = \frac{1}{2} \sum_{i}^{A} \sum_{j}^{A} r_{ij}^4 \, Y_{4M}(r_{ij}) \, \Phi' \tag{86a}$$

The first problem with this is that it is no longer obvious that the translationally variant form of (86a), obtained by fixing the centre of mass at the origin, is

$$\sum_{j}^{A} r_j^4 \, Y_{4M}(r_j) \, \Phi' \tag{86b}$$

However, if we assume (86b) to be valid, then from section BII we can identify Ψ_{4M} as a surface wave carrying angular momentum $L = 4$, i.e. a surface oscillation of multipolarity four. Thus the states Ψ_{LM} probably involve deformations of multipolarity L and so can only describe a rotating ellipsoid when $L = 2$. For $L > 2$, these states will still represent real nuclear excitations (cf. [4] pp 137-41), but I do not regard them as being members of the ground state rotational bands of even-even nuclei.

From reference [4] pp 688-92, it appears much more likely that the correct method for constructing wavefunctions for members of a given rotational band is to take linear combinations of the wavefunctions of lower energy states in the same band. For example, instead of $\Psi_{44} = F_{44} \, \Phi'$, the wavefunction for the first 4^+ state is probably some linear combination of two Ψ_{22} states. Symbolically, we can write this wavefunction as $(F_{22})^2 \, \Phi'$. The correct combination would couple the two $L = 2$ quanta to form an $L = 4$ state in such a way that the shape of the rotating nucleus remained constant, namely a prolate spheroid with fixed quadrupole moment. It might also be possible to combine classical irrotational surface waves (see BII), to form analogous rotational modes of a classical prolate spheroid. By this process of angular momentum coupling a whole series of rotational and vibrational wavefunctions would be constructed and it should be possible to use group theory to identify and catalogue all the possible modes. Group theory

should also be able to identify for which values of M the general multipolarity wavefunctions Ψ_{LM} can represent rotational modes of fixed shape, or large amplitude symmetry preserving vibrations. (e.g. From the results of section BII I was able to identify $\Psi_{2\pm2}$ as being rotational modes and Ψ_{20} as a vibrational mode, of a prolate spheroid.) Then onto each of the Ψ_{LM} states it should be possible to construct rotational and vibrational bands by the process of angular momentum coupling.

Finally, I note that in order to extend the methods of this section to describe rotational states which have a non zero value of the quantum number K, we would need a generalization of the wavefunctions (5). From the unified model wavefunctions (27) and (28) we see that what is required is a change from $Y_{LM}'s$ to $D_{MK}^{L*}'s$, but the variables have to be the nucleon coordinates and not collective angular coordinates.

(b) Isovector Rotational States of Nuclei :

Consider the generalization of the isoscalar wavefunctions (5), obtained by explicitly distinguishing between protons and neutrons :

$$\underline{\Psi}_{LM} = \underline{F}_{LM} \Phi = \sum_{p}^{Z} \sum_{n}^{N} r_{pn}^{L} Y_{LM}(\hat{r}_{pn}) f(r_{pn}) \Phi \tag{87}$$

These wavefunctions are translationally invariant and it is straight-forward to extend the calculations of [3] to show that when $\underline{L}\,\Phi = \underline{0}$ that $\underline{\Psi}_{LL}$ is an angular momentum eigenstate of $\underline{L}^2$ and L_z with eigenvalues $L(L+1)\hbar^2$ and $L\hbar$ respectively, and further, that for a hypothetical system of two kinds of bosons interacting via the

harmonic pair potential (6), that Ψ_{LL} with f = 1 is also an energy

eigenstate with eigenvalue again given by (7). One difference between

(87) and (5) is that Ψ_{LL} is defined for all integer L, whereas Ψ_{LM}

is zero when L is odd. Note that for odd L Ψ_{LM} has the opposite parity

to Φ . Also, because we need only consider permutations between

indistinguishable particles, the wavefunctions (87) have the same

permutation symmetry as Φ . For example, when Φ is the ground state

of an even-even nucleus (i.e. a 0^+ state) then Ψ_{LM} is a $(-1)^L$ state,

assuming as in part (a) that the spins are paired up to give zero S .

Despite the very close similarity between the wavefunctions (87),

Ψ_{LM} , and the wavefunctions (5), Ψ_{LM} , there is an important physical

difference between them : Ψ_{LM} is an isovector state (protons and

neutrons move in opposite phase) while Ψ_{LM} is an isoscalar state

(protons and neutrons move in phase). It is obvious that Ψ_{LM}

describes isoscalar motion since it treats the protons and neutrons

as indistinguishable. Perhaps the best way to see that Ψ_{LM} describes

isovector motion is to note from (87) that the probability $|\Psi_{LM}|^2$

is largest when the proton and neutron fluids are separated

(note, $r^L Y_{LL}(\hat{r}) \sim (x + iy)^L$) implying that the protons and neutrons

move in opposite phase. For odd L this argument is especially clear,

since by comparing (5) with (87) we see that when the protons and

neutrons are evenly distributed that Ψ_{LM} for L odd is zero. For

example, the particular case of Ψ_{1M} with f = 1, is shown in ref. [2]

to give a good description of the giant dipole resonance, which is

known to be a simple oscillation of protons against neutrons (see

below). For even L the wavefunctions Ψ_{LM} describe the same type

of motion as the wavefunctions Ψ_{LM} , except that the protons and

neutrons now move in opposite phase. In particular, consider the

L = 2 isovector state Ψ_{2M} . In note 1) of part (a) the isoscalar

state Ψ_{2M} for spherical nuclei is identified as the high frequency

quadrupole mode discussed in ref. [4] , so it follows that in spherical

nuclei, $\mathcal{\Psi}_{2M}$ should be the isovector analogue of the giant quadrupole

resonance state. This conclusion is completely compatible with the

results of [4] , where the quadrupole field is shown to generate both

an isoscalar and an isovector quadrupole mode. Shell model calculations

lead to an energy $\Delta E_2(\text{isovector}) \simeq 135 A^{-1/3} \text{ MeV}$ for

the high frequency isovector quadrupole mode, (see [4] p 513), while

the liquid drop model yields $\Delta E_2(\text{isovector}) \simeq 127 A^{-1/3} \text{ MeV}$,

(see [4] p 671). This energy is much higher than the isoscalar

quadrupole mode ($\Delta E_2(\text{isoscalar}) \simeq 60 A^{-1/3} \text{ MeV}$), since the

isovector mode involves density variations whereas the isoscalar mode

is an incompressible shape oscillation.

In ref. [2] (which is supplied with this thesis), the wavefunction

$\mathcal{\Psi}_{1M}$ with f = 1, is investigated in detail. This particular wave-

function has been previously proposed by deShalit and Feshbach (see [5]

pp 501-2, 736) as a good approximation to the giant dipole resonance

state. The giant dipole resonance is perhaps the most important and

certainly the most investigated of all nuclear collective motions.

The most striking feature of the giant dipole resonance is that it

exhausts a very large part ($\approx \frac{1}{2}$) of the total absorption cross

section for El transitions, i.e. the $S_1^{(1)}$ sum rule (see section AIV).

The results of [2] prove that the deShalit and Feshbach wavefunction

gives a good description of the giant dipole state. Because we can

positively identify $\mathcal{\Psi}_{1M}$ with a known nuclear excitation, the

calculations in [2] have important implications for all of the

proposed related states Ψ_{LM} and $\mathcal{\Psi}_{LM}$. That is, we now have a

detailed understanding of the properties of one of a whole series of

related wavefunctions. Without unduly duplicating the results of

reference $[2]$, I will now discuss three particular features of the calculations involving the dipole state Ψ_{IM}' , that should be noted because they could be general features of all of the wavefunctions (5) and (87).

(1) Contribution to the $S_1^{(l)}$ sum rule :

In $[5]$ p 736, it is pointed out that since $\Psi_{IM}' \sim Q_{IM} \Phi$, where Q_{IM} is the translationally invariant dipole operator, that Ψ_{IM}' has total overlap with the dipole operator and so the three Ψ_{IM}' states must exhaust the entire $S_1^{(l)}$ sum rule. (Note, the three states are degenerate in spherical nuclei.) Thus if Ψ_{IM}' was an exact eigen-state then there would be only one 1^- level in the non deformed even-even nuclei, since only one E1 transition to or from the ground state would be possible. In section AIV this is proved explicitly in the approximation in which the potential energy is assumed to commute with F_{IM} (i.e. neglecting charge exchange and velocity dependent interactions). That is, it is shown that the three states together exhaust the entire $S_1^{(l)}$(class) sum rule. Experimentally it is found that the giant dipole resonance exhausts about $S_1^{(l)}$(class) of the total sum rule for medium to large nuclei. A large part of the rest of contributions come from the high energy region where charge exchange and velocity dependent forces are more important. It is because these interactions play a less important role in the region of the giant dipole state that Ψ_{IM}' gives such a good description of the dipole resonance.

All the states Ψ_{LM} and Ψ_{LM}' have a significant overlap with the electric multipole moment of multipolarity L, so it is likely that they will all be resonance states of spherical nuclei. For example, it is shown in section AIV that the five Ψ_{2M} states

exhaust Z/A of the $S_2^{(1)}$(class) sum rule.

(2) The weak correlation approximation :

It is found in [2] that the weak correlation approximation gives a completely unsatisfactory result for the giant dipole energy, in particular it leads to $\Delta E_1 \sim A^{-2/3}$ instead of $\sim A^{-1/3}$. However, the correct energy variation is achieved once the short ranged correlations are included. This is proved in a general way, by imposing the requirement of number conservation on the pair correlation functions. Because Ψ_{1M} is an isovector state it is necessary to first distinguish between protons and neutrons, so that there are three two particle number densities (i.e. n-n, p-p, p-n), instead of the single pair density (16). Once this is done, the requirement for number conservation to hold separately for the protons and neutrons (i.e. the isovector analogue of (19)), implies an exact cancellation of the weak correlation terms in the energy denominator, leaving $\Delta E_1 \sim A^{-1/3}$. That is, equation 14 of ref. [2] for the variational energy is really proportional to $1/R$ and not $1/R^2$ as it appears to be.

From equation (4a) it follows that the energy of all the states Ψ_{LM} and Ψ_{LM} will be proportional to $\hbar^2/mR^2$ (in the approximation in which the potential energy commutes with F_{LM} and F_{LM}) and since $R \simeq 1.2A^{1/3} fm$, it will always appear as if $\Delta E_L \sim A^{-2/3}$, i.e. it takes a cancellation of the leading order terms to produce an extra factor of $A^{1/3}$. I noted in part (a) that almost all of the collective states predicted for spherical nuclei in reference [4] have an energy varying as $A^{-1/3}$, so it is quite possible that the weak correlation approximation will lead to the same type of error for all of the Ψ_{LM} and Ψ_{LM} wavefunctions,

as occurred for the Ψ_{lm} state in [2] .

(3) Hard core correlations :

The dominant nucleon-nucleon interaction is the effect of Fermi statistics, i.e. Pauli repulsion (see [5] p 49). In [2] the result for ΔE_1 , calculated by using only those pair correlations due to Fermi statistics, is just 20% higher than the experimental energy. So it seems that the neglect of hard core correlations is a minor approximation for collective states like Ψ_{LM} and Ψ_{LM} . Support for this conclusion comes from comparing equation 64 of [1] , with equation (19), which suggests that the contribution to the pair correlations from the hard cores is of the order of $A\left(\frac{a}{R}\right)^3$ smaller than that from Fermi statistics, where a is the hard core diameter. For nuclei we have $\quad (a/R) \simeq \frac{1}{3} A^{-1/3}$.

Finally I will include for the future, some ideas and problems on two possible extensions of the work in ref. [2] .

(i) The giant dipole resonance in deformed nuclei :

Without repeating any of the extensive discussions in part (a), let us apply the method used to investigate the Ψ_{2M} states in deformed nuclei, to the Ψ_{lm} states, i.e. consider

$$\Psi_{lm} = F_{lm} \, \Phi'$$
(88)

where Φ' is a deformed ground state, and assume that (88) describes the giant dipole resonance in deformed nuclei. First of all, it follows that the three Ψ_{lm} states are no longer degenerate. From the scaling procedure of part (a) (cf. (62)), it follows that for a general deformation, (88) will split into three states, while for a prolate spheroid symmetric about the Z axis, Ψ_{ll} and Ψ_{l-l} will

describe two degenerate vibrations perpendicular to the symmetry axis, and ψ_{10} represents a lower energy vibration along the symmetry axis. Also, from section AIV we see that regardless of the deformation that each ψ_{1M} state continues to exhaust one third of the $S_1^{(1)}$ (class) sum rule. So for nuclei whose ground states can be regarded as prolate spheroids, one third of the dipole strength to the giant dipole resonance should be contained in the ψ_{10} mode, with the other two thirds taken up by the degenerate $\psi_{1\pm1}$ modes. Both of these qualitative features are known to be true experimentally (see p 490 of ref. [4]). However, there is a problem with the scaling method of part (a), associated with the fact that it affects the exact expression for the energy (eqn 14 of ref. [2]) in the same way as it affects the weak correlation limit result. That is, (62) describes the deformation with a single parameter that factors out of all of the expectation values. This results in a splitting between the (1,0) and (1,±1) modes that is about twice as large as is found experimentally. The experimental result can be derived by a scaling argument based on the energy variation $E_1 \sim A^{-1/3} \sim 1/R$. Namely, if we assume that the ψ_{1M} vibration has an energy $\Delta E_{1M} \sim \frac{1}{R_M}$, we have

$$\frac{E_{1\pm1} - E_0}{\overline{E}} = \frac{\frac{1}{R_{\pm1}} - \frac{1}{R_0}}{1/R} \simeq \delta \tag{89}$$

where δ is the deformation parameter (50), which is defined as the difference between the single and the double axis of the prolate spheroid, measured in units of R. The origin of the "error" in the method of part (a) is that it treats the dipole energy as if it were proportional to $1/R^2$ (i.e. the weak correlation limit result), so that instead of (89) it gives

$$\frac{E_{1\pm1} - E_0}{\overline{E}} = \frac{\frac{1}{R_{\pm1}^2} - \frac{1}{R_0^2}}{\frac{1}{R^2}} \simeq 2\delta \tag{90}$$

This value is always at least twice as large as the experimental data
on page 493 of ref. $\left[4\right]$). A possible explanation is that the scaling
method of part (a) may be too simple to describe the correct pair
correlations in deformed nuclei, i.e. if the scaling was also contained
within the pair correlation functions then the cancellation of the
weak correlation terms will result in a different energy splitting
than in the weak correlation approximation.

(ii) <u>Low lying 1⁻ single particle levels in even-even nuclei</u> :

In Fig. 2 and Fig. 3 below, I have plotted the known 1⁻ states
of even-even nuclei, together with the weak correlation approximation
result for the energy of ψ_{IM} , (i.e. $\Delta E_1(\text{w.c.limit}) \simeq 72A^{-2/3} \text{MeV}$).
Note that many of the low lying 1⁻ states of even-even nuclei have an
energy close to $72 A^{-2/3} MeV$. The main exceptions to this are
in the large A deformed regions ($150 \leq A \leq 190$ and $A \gtrsim 220$),
where there exist much lower 1⁻ states that usually belong to
vibrational or rotational bands. The question to be asked is,
whether the good agreement of the weak correlation approximation
result for the energy of the dipole resonance with so much of the
experimental data plotted in Fig. 2 and Fig. 3, is a coincidence
or not? For example, we could speculate that for a particular
$f(r_{pn}) \neq 1$ in the wavefunction (87), that the weak correlation
approximation becomes valid, while the energy remains close to
$72 A^{-2/3} MeV$. However, the experimental data for the transition
rates seems to rule this out. Namely, for the 14 states in Fig. 2
that the transition rates to the ground state are known (note, 10 of
these are within 15% of $72A^{-2/3} MeV$), all have lifetimes four
orders of magnitude larger than the result calculated for ψ_{IM} when
f = 1. So it is very unlikely that the transition rate data can be
explained without including a radial node in $f(r_{pn})$, which would

increase the energy well beyond the low lying states we wish to explain. In fact, from the transition rate data it follows that these states cannot be collective, i.e. they are single particle states. Still, it could be possible that Ψ_{lm} can describe single particle excitations under certain conditions. Another speculative suggestion is that perhaps these single particle excitations involve charge exchange processes, so that the number conservation argument of section four of [2] would no longer hold and the weak correlation result might then be a good approximation. But it remains virtually impossible to see how Ψ_{lm} could ever describe levels with very low transition rates, since it has total overlap with the dipole operator.

So the results of Fig. 2 and Fig. 3 may very well be a misleading coincidence. It is worth pointing out that the low lying 0^+ states discussed right at the end of section AIII, also follow an $A^{-2/3}$ trend. Thus, for spherical nuclei, it may be a general property that the energies of single particle states follow $A^{-2/3}$ variations, while the energies of collective states vary as $A^{-1/3}$.

Figure captions for Fig. 2 and Fig. 3.

Fig. 2 : The energy of the 1^- states of even-even nuclei ($6 \leq A \leq 44$). Full line: The energy of Ψ_{lm} , in w.c. limit: $72A^{-2/3}$ MeV. Crosses: Definite assignments. Circles: Tentative assignments. Arrows: The known 1^- states directly above the base of an arrow have been left out. The data is from reference [9] .

Fig. 3 : The energy of the 1^- states of even-even nuclei $(A > 50)$. This Fig. follows on from Fig. 2 (the lowest A state comes from Fig. 2) and is plotted in the same way. The data is from reference [10] .
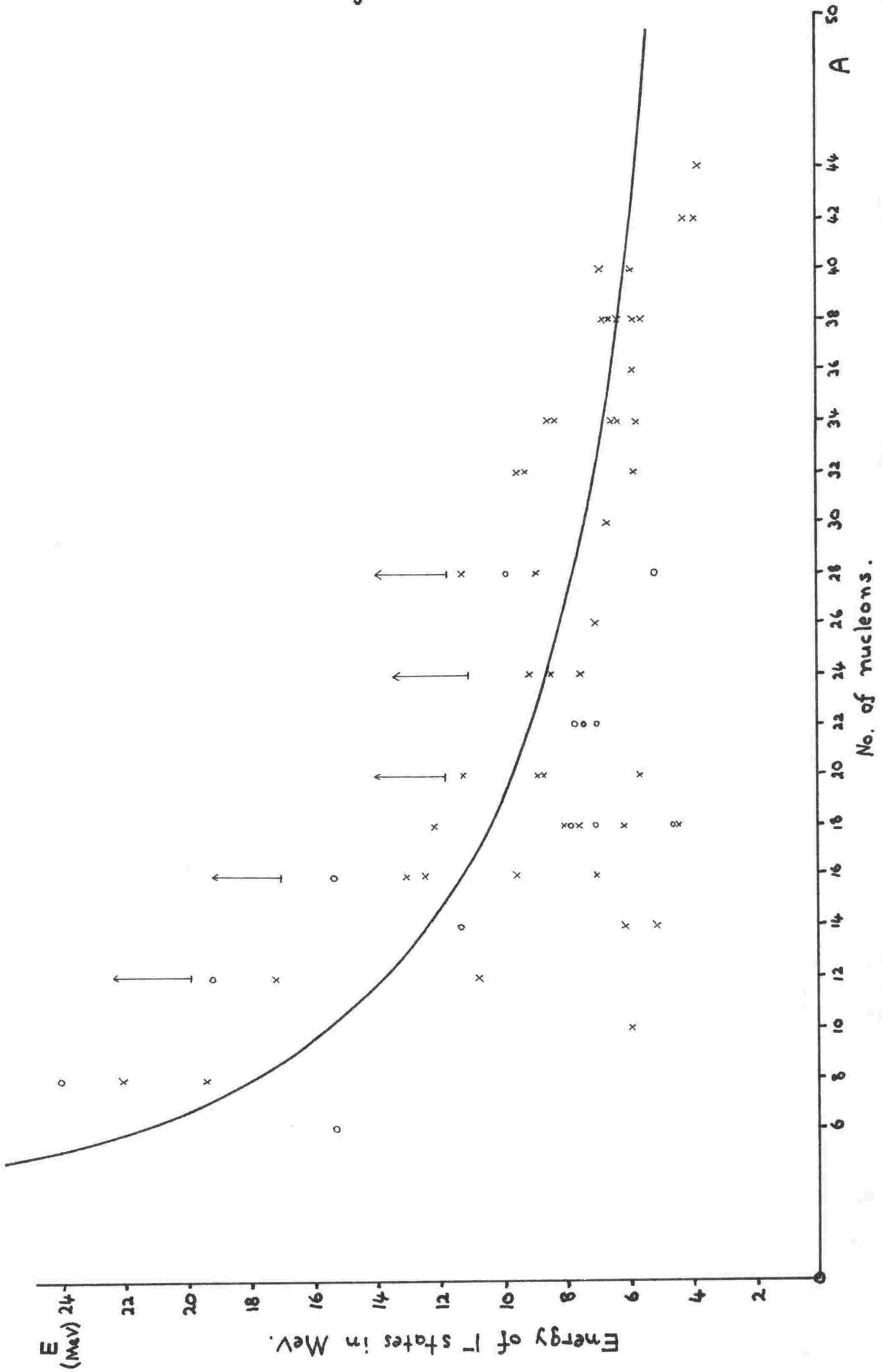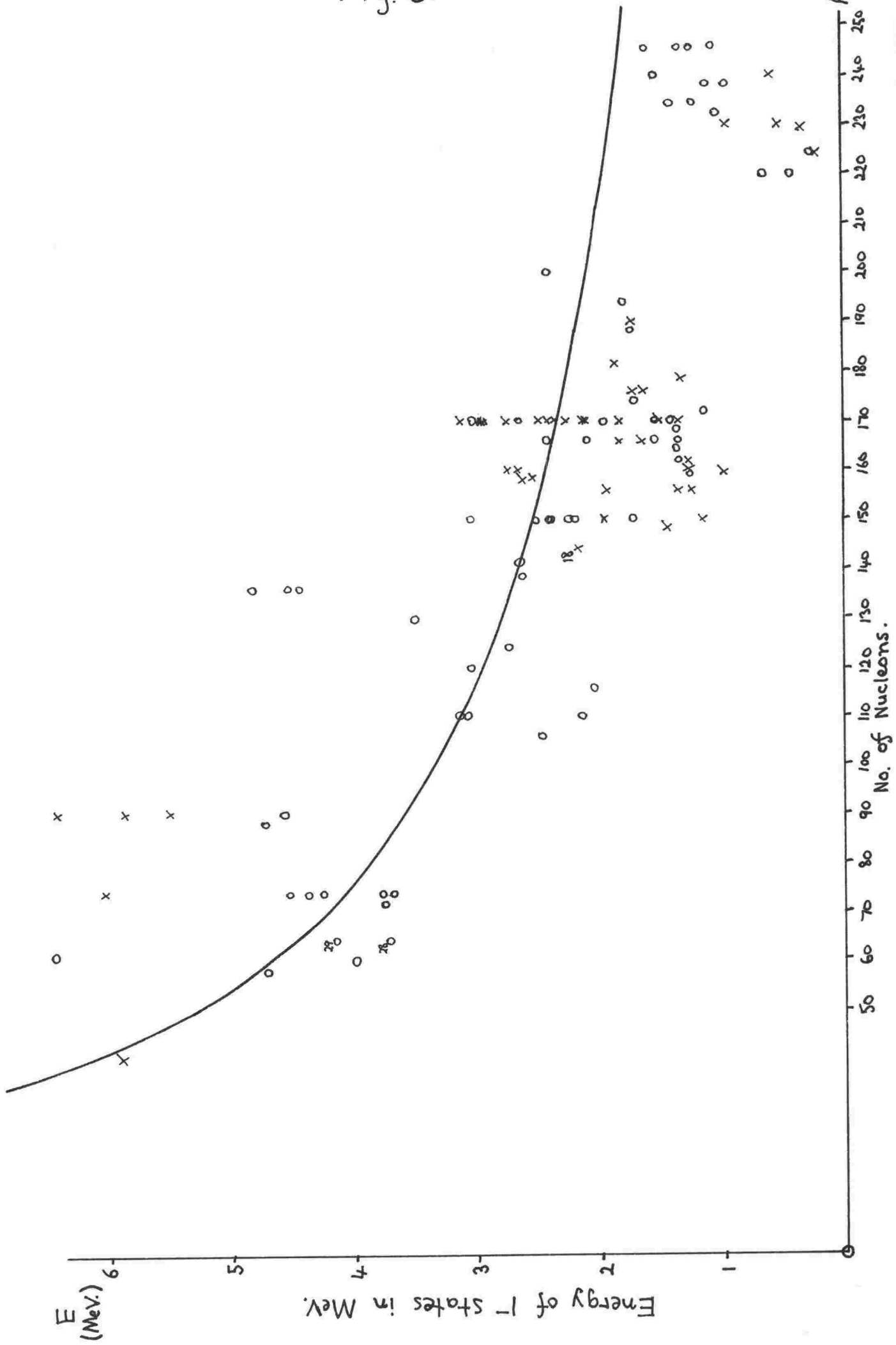

Fig. 2.

Fig. 3.

AII. 49.

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III. COMPRESSIONAL VIBRATIONAL STATES OF SPHERICAL NUCLEI.

*In this section trial wavefunctions for vibrational states of a quantum many-body system are considered and the energies of the states evaluated. The many-body system is taken to be composed of A spinless uncharged identical particles, as for example helium microdroplets, or to a reasonable approximation even-even nuclei where we can assume that the spins are paired up to give zero $\underline{S}$ along with neglecting the differences between proton and neutron masses and interactions. In evaluating the energies we will further assume that the ground state is spherically symmetric and also we will neglect all correlations between particles other than those described by the finiteness of the system (this is the weak correlation approximation discussed previously in sections AI and AII (i.e. [1])). *The weak correlation approximation is not as crude an approximation as might be expected, because it is only used to calculate the collective vibrational energy difference between the excited state and the ground state, so that no calculations are made of any part of the ground state energy. For our purpose of calculating only the collective excitation energy the weak correlation approximation should be fairly good for nuclei, as the hard core of the nucleons occupies less than a percent of the total volume (nucleon radius is approximately six times the hard core radius), but will only give a rough estimate for helium microdroplets where the hard core volume is about 20% of the total volume (helium radius is approximately 1.7 times the hard core radius). The spherical ground state and weak correlation assumptions mean that the calculated vibrational state energies are quantitatively valid only for the non deformed even-even nuclei, but the results could also be qualitatively extended to all even-even nuclei and helium microdroplets.

The trial wavefunctions considered are all Feynman type wavefunctions [2] of the same form as the rotational excitations

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* See the note on page AIII. 44.

considered in [1] , namely

$$\Psi = F\Phi \tag{1}$$

where F is a symmetric function of all the particles and Φ is the ground state wavefunction, or Φ could also be an excited state and then we would be creating a vibration of this excited state. (From now on Φ will be taken to be the ground state as we will not consider vibrational states built on excited states.) The Hamiltonian of the self-bound A particle system is assumed to be of the type

$$H = -\frac{\hbar^2}{2m} \sum_{k=1}^{A} \nabla_k^2 + V(r_1,...,r_A) \tag{2}$$

where V is completely symmetric with respect to permutations of the particle coordinates. Because H is a real linear operator the ground state Φ can be taken to be real. To evaluate the energy of the state (1) we use the identity proved in [1] , that for F a symmetric function of all the particles the expectation value of $H-E_o$ in the state Ψ can be written as

$$\frac{\int d1....dA \, \Psi^*(H-E_o)\Psi}{\int d1....dA \, |\Psi|^2} = \frac{A\hbar^2 \langle |\nabla_1 F|^2 \rangle}{2m \langle |F|^2 \rangle} \tag{3}$$

where $H\Phi = E_o\Phi$ and $\langle \rangle$ is the ground state expectation value defined by

$$\langle B \rangle = \frac{\int d1....dA \, B\,\Phi^2}{\int d1....dA \, \Phi^2} \tag{4}$$

In evaluating expectation values of the type (4) we further assume that Φ^2 is spherically symmetric. That is, we take the ground state to be described by a purely radial single particle number density (see AI(2))

$$n(r_1) = \frac{A \int d2....dA \, \Phi^2}{\int d1....dA \, \Phi^2} \tag{5}$$

where r_1 is the distance of particle one from the centre of mass.

The physical excitations of a self-bound many-body quantum system must be translationally invariant states. This is because we need to work in the rest frame of the system, in order that the angular momentum of the system be independent of the choice of origin. That is, we require $\underline{P}\Psi = 0$, but the centre of mass $\underline{R}$ and the total momentum $\underline{P}$ are conjugate variables so that by the uncertainty principle $\underline{R}$ is indeterminate when we work in the rest frame. Thus in order to be certain of the physical reality of any trial vibrational state we must use a wavefunction that is translationally invariant, so that it does not depend on the indeterminate variable $\underline{R}$. If translationally variant wavefunctions are used, for example those that are constructed on the assumption that the centre of mass is fixed at the origin, then they may give rise to spurious states that represent unphysical motions of the centre of mass rather than internal motions $[3, 4, 5]$. The problem of identifying spurious states has been shown to have a solution only in the special case of harmonic interactions $[3, 5]$. Note, that the use of a radial ground state density (5), is only a means of describing the density variation in the system and does not lead to spurious states as a translationally variant wavefunction might. (5) is inconsistent in the sense that the right hand side is translationally invariant because of Φ while the left hand side is a function of only one variable and so could only be translationally invariant if it was a constant. However providing (5) is not used to solve for the surface structure you can temporarily fix the centre of mass of Φ at the origin and then (5) will describe a radial density variation of a finite system from its centre of mass without leading to spurious results.

For the wavefunction $\Psi = F\Phi$ to represent a vibrational state built on the ground state Φ we require

$$\underline{L} F = 0 \tag{6}$$

where $\underline{L} = (L_x, L_y, L_z)$ is the angular momentum operator. (N.B. we have already ignored spin coordinates so that angular momentum means orbital angular momentum.) With condition (6) Ψ is an angular momentum eigenstate with the same eigenvalue as Φ , which is the definition of what I mean by a vibrational state. It might also be argued that Ψ should have the same parity (inversion symmetry) and permutation symmetry as Φ which is satisfied provided F has positive parity and Bose symmetry. (The results will only be quantitatively applied to non deformed even-even nuclei where the ground state Φ has zero angular momentum and positive parity; i.e. Φ is taken to be a 0^+ state.) Now because Ψ has the same angular momentum as Φ in order for Ψ to be a true excited state it must be made orthogonal to Φ . The condition for Ψ to be orthogonal to Φ is

$$\int \underline{d1} \dots \underline{dA} \, \Psi^* \Phi \;=\; 0 \;=\; \int \underline{d1} \dots \underline{dA} \, \Psi \Phi^* \tag{7}$$

Using (1) and (4) we can write (7) as

$$\langle F \rangle \;=\; 0 \tag{8}$$

Having imposed the condition (8) we can then use the variational principle to prove that (3) is the variational upper bound to the difference between the true excited state energy E and the ground state energy E_o . That is,

$$\Delta E \;\equiv\; E - E_o \;\leq\; \frac{A\hbar^2}{2m} \frac{\langle |\nabla F|^2 \rangle}{\langle |F|^2 \rangle} \tag{9}$$

From the above we conclude that for $\Psi = F\Phi$ to be a true vibrational state of Φ then F must be translationally invariant, must be an eigenstate of angular momentum zero, must have positive parity and Bose permutation symmetry, and must satisfy (8). The simplest form for F satisfying these conditions is

$$F \;=\; \sum_{i<j}^{A} \sum^{A} f(r_{ij}) \tag{10}$$

where $\Gamma_{ij} = |\underline{r}_i - \underline{r}_j|$ and the orthogonality condition (8) becomes

$$\langle f(\Gamma_{12}) \rangle = 0 \qquad (11)$$

That (10) is translationally invariant is obvious, while the positive parity and Bose symmetry properties of (10) follow from the identity $\Gamma_{ij} = \Gamma_{ji}$. To prove (6) we first calculate from (10)

$$\frac{\partial F}{\partial x_k^{(\alpha)}} = \sum_{j \neq k} \frac{x_{kj}^{(\alpha)}}{\Gamma_{kj}} f'(\Gamma_{kj}) \qquad (12)$$

where $x^{(\alpha)} = \{ x^{(1)}, x^{(2)}, x^{(3)} \} = \{ x, y, z \}$, $x_{kj}^{(\alpha)} = x_k^{(\alpha)} - x_j^{(\alpha)}$
and $f'(r) = df/dr$. Writing L_x, L_y, L_z as L_α where $\alpha = 1, 2, 3$
respectively, then

$$L_\alpha F = -i\hbar \sum_k \left(x_k^{(\alpha \oplus 1)} \frac{\partial F}{\partial x_k^{(\alpha \oplus 2)}} - x_k^{(\alpha \oplus 2)} \frac{\partial F}{\partial x_k^{(\alpha \oplus 1)}} \right) \qquad (13)$$

where $\alpha \oplus a$ means the $a\text{th}$ element to the right of $\alpha \in \{1, 2, 3\}$
in the sequence $1,2,3,1,2,3, \cdots$. Substituting (12) into (13) we
find

$$L_\alpha F = -i\hbar \sum_k \sum_{j \neq k} \left(x_k^{(\alpha \oplus 1)} \frac{x_{kj}^{(\alpha \oplus 2)}}{\Gamma_{kj}} f'(\Gamma_{kj}) - x_k^{(\alpha \oplus 2)} \frac{x_{kj}^{(\alpha \oplus 1)}}{\Gamma_{kj}} f'(\Gamma_{kj}) \right) \qquad (14)$$

Using the identity

$$\sum_k \sum_{j \neq k} A_k B_{kj} = \sum_{k<j} \sum A_{kj} B_{kj} \, , \, (B_{kj} = -B_{jk}) \qquad (15)$$

we see immediately from (14) that $\underline{L}F = 0$, that is

$$L_\alpha F = 0 \qquad \text{for all } \alpha \qquad (16)$$

(In fact, (16) follows automatically from (10) because any F of the
form (10) is rotationally invariant.) If, as for non deformed
even-even nuclei the ground state Φ is a 0^+ state, then $\Psi = F\Phi$
with F given by (10) and (11) is a 0^+ vibrational state. Using

(12) and $\underline{P}\Phi = 0$ we also have

$$\underline{P}\Psi = -i\hbar\Phi\left(\sum_k \frac{\partial F}{\partial x_k}, \sum_k \frac{\partial F}{\partial y_k}, \sum_k \frac{\partial F}{\partial z_k}\right) = 0 \qquad (17)$$

which confirms that we are working in the rest frame, as desired.

If we relax the translational invariance condition but keep all of the vibrational state requirements then the simplest form for F would be

$$F = \sum_{j=1}^{A} f(r_j) \qquad (18)$$

where $r_j = |\underline{r}_j|$ is the distance from the centre of mass which is now fixed at the origin, and the orthogonality condition (8) becomes

$$\langle f(r_i) \rangle = 0 \qquad (19)$$

(From (13) we see immediately that (18) satisfies (6) and the positive parity and Bose symmetry of (18) is obvious.) $\Psi = F\Phi$ with F given by (18) and (19) is a valid trial wavefunction for real vibrational states except when the broken translational invariance has lead to spurious unphysical states in the manner described previously. Unfortunately general methods of recognising spurious states are not known for non harmonic forces $[3, 5]$, so that (18) can only be used with the knowledge that it may sometimes represent non physical states.

THE PHYSICAL NATURE OF Ψ :

Classically we would expect the simplest and lowest energy compressional vibrational state of a spherical system to be the breathing mode oscillation, where the system periodically expands and contracts as a whole. Thus an important point to consider is whether the trial wavefunctions $\Psi = F\Phi$ with F given by (10) or (18) could describe a breathing mode state. Firstly let's define the excited

state single particle number density as (cf. (5))

$$n_1(\underline{r}_1) = \frac{A \int d2 \dots dA |\Psi|^2}{\int d1 \dots dA |\Psi|^2}$$

$$= \frac{A}{V} \frac{\langle |F|^2 \rangle_1 (\underline{r}_1)}{\langle |F|^2 \rangle} \tag{20}$$

where $\langle \ \rangle$ is defined by (4) and $\langle \ \rangle_1$ is defined by

$$\langle B \rangle_1 (\underline{r}_1) = \frac{V \int d2 \dots dA \, B \, \Phi^2}{\int d1 \dots dA \, \Phi^2} \tag{21}$$

We have assumed that Φ^2 is spherically symmetric so that our A

particle system is spherical and $n_1(\underline{r}_1)$ becomes $n_1(r_1)$; r_1 the

distance of particle one from the centre of mass. Let's now make the

approximation that the ground state number density $n(r_1)$ given by

(5) is the cutoff density (see AI(13)),

$$\begin{aligned} n(r_1) &= A/V \quad , \quad r_1 \le R \\ &= 0 \quad , \quad r_1 > R \end{aligned} \tag{22}$$

so that the ground state is a sphere of uniform density and volume

$V = \frac{4}{3}\pi R^3$. Note that from (5), (21) and (22) we have

$$\begin{aligned} \langle 1 \rangle_1 &= 1 \quad , \quad r_1 \le R \\ &= 0 \quad , \quad r_1 > R \end{aligned} \tag{23}$$

The question we now consider is how does the excited state number

density $n_1(r_1)$ vary with r_1 when $n(r_1)$ is given by (22).

We will first discuss the simpler translationally *variant*

vibrational state, namely $\Psi = F\Phi$ with F given by (18) and (19).

From (18) and (21) we find

$$\langle |F|^2 \rangle_1 (r_1) = \left\langle \sum_i^A \sum_j^A f^*(r_i) f(r_j) \right\rangle_1$$

$$= |f(r_1)|^2 \langle 1 \rangle_1 + (A-1) f^*(r_1) \langle f(r_2) \rangle_1 + (A-1) f(r_1) \langle f^*(r_2) \rangle_1$$

$$+ (A-1) \left\langle |f(r_2)|^2 \right\rangle_1 + (A-1)(A-2) \left\langle f^*(r_2) f(r_3) \right\rangle_1 \qquad (24)$$

Using (22) and the two and three particle density correlation

functions $n(1 \cdots S)$ defined by AI(2), it follows that for $h(r_j)$

a general function of just r_j , that

$$\left\langle h(r_2) \right\rangle_1 = \frac{V}{A(A-1)} \int \underline{d2} \, n(12) h(r_2) \qquad (25a)$$

and $\left\langle h(r_2) h(r_3) \right\rangle_1 = \frac{V}{A(A-1)(A-2)} \int \underline{d2} \, \underline{d3} \, n(123) h(r_2) h(r_3) \qquad (25b)$

If we now use the weak correlation approximation discussed previously,

then $n(12)$ becomes $(A-1)/A \, n(1) n(2)$ (see App AI(1)) and hence (25a)

becomes

$$\left\langle h(r_2) \right\rangle_1 = \frac{V}{A^2} \, n(1) \int \underline{d2} \, n(2) h(r_2) = \left\langle 1 \right\rangle_1 \left\langle h(r_1) \right\rangle \qquad (26a)$$

Similarly we can define a partial weak correlation limit by neglecting

correlations between (1,3) and (2,3) to give $n(123) = \frac{(A-2)}{A} n(12) n(3)$,

which leads to (25b) becoming

$$\left\langle h(r_2) h(r_3) \right\rangle_1 = \frac{V}{A^2(A-1)} \int \underline{d2} \, n(12) h(r_2) \int \underline{d3} \, n(3) h(r_3)$$

$$= \left\langle h(r_2) \right\rangle_1 \left\langle h(r_1) \right\rangle \qquad (26b)$$

In the weak correlation approximation the orthogonality condition (19)

becomes using (4) and (5)

$$\left\langle f(r_1) \right\rangle = \frac{1}{A} \int \underline{d1} \, n(1) f(r_1) = 0 \qquad (27)$$

Therefore from (25), (26) and (27) we see that $\left\langle f(r_2) \right\rangle_1$,

$\left\langle f^*(r_2) \right\rangle_1$, and $\left\langle f^*(r_2) f(r_3) \right\rangle_1$ are all zero and $\left\langle |f(r_2)|^2 \right\rangle =$

$\left\langle 1 \right\rangle_1 \left\langle |f(r_1)|^2 \right\rangle$.

Therefore in the weak correlation limit (24) becomes

$$\langle |F|^2 \rangle_1 (r_1) = \langle 1 \rangle_1 \left\{ |f(r_1)|^2 + (A-1) \langle |f(r_1)|^2 \rangle \right\} \tag{28}$$

where $\langle 1 \rangle_1$ is given by (23). Evaluating $\langle |F|^2 \rangle$ in a similar fashion to (24) and using the weak correlation approximation with the orthogonality condition (27), we find from (20), (22), (23) and (28) that

$$n_1(r_1) = n(r_1) \left[1 + \frac{|f(r_1)|^2 - \langle |f(r_1)|^2 \rangle}{A \langle |f(r_1)|^2 \rangle} \right] \tag{29}$$

where $n(r_1)$ is given by (22). Hence the excited state density $n_1(r_1)$ varies only to order $\frac{1}{A} n(r_1)$ from the constant ground state density $n(r_1)$ and this small variation is a function of $|f(r_1)|^2$. Furthermore if we substitute the cutoff density (22) into the orthogonality condition (27) we find,

$$\int_0^R dr_1 \, r_1^2 \, f(r_1) = 0 \tag{30}$$

so that requiring Ψ to be orthogonal to Φ implies that the real and imaginary parts of $f(r_1)$ must change sign at least once as r_1 varies from zero to R. Restricting the discussion to $f(r_1)$ real, which will be true for all the vibrational states of the type (18) that we will consider, we see that (29) and (30) imply that $n_1(r_1)$ must have at least one shallow minimum. Before discussing the physical signifi-cance of this we will consider the variation of $n_1(r_1)$ for the translationally invariant vibrational wavefunction.

In investigating the behaviour of $n_1(r_1)$ for the translationally invariant states (10) with (11) we will discuss only the case of F real, as all our specific trial wavefunctions will be real and any imaginary part could have been discussed in a similar way. From (10) and (21) we find

$$\langle |F|^2 \rangle_1 (\Gamma_i) = \langle \sum_{i<j}^A \sum_{k<\ell}^A f(\Gamma_{ij}) f(\Gamma_{k\ell}) \rangle_1$$

$$= (A-1)\langle f^2(\Gamma_{12}) \rangle_1 + \frac{(A-1)(A-2)}{2} \langle f^2(\Gamma_{23}) \rangle_1 + (A-1)(A-2)\langle f(\Gamma_{12}) f(\Gamma_{13}) \rangle_1$$

$$+ (A-1)(A-2)\langle f(\Gamma_{12}) f(\Gamma_{23}) \rangle_1 + (A-1)(A-2)^2 \langle f(\Gamma_{23}) f(\Gamma_{24}) \rangle_1$$

$$+ (A-1)(A-2)(A-3)\langle f(\Gamma_{12}) f(\Gamma_{34}) \rangle_1 + \frac{(A-1)(A-2)(A-3)(A-4)}{4} \langle f(\Gamma_{23}) f(\Gamma_{45}) \rangle_1$$

$$\tag{31}$$

Let's now introduce the two, three, four and five particle density correlation functions $n(1...5)$ defined by AI(2). The orthogonality condition (11) becomes

$$\langle f(\Gamma_{12}) \rangle = \frac{1}{A(A-1)} \int d\underline{1} . d\underline{2} \, n(12) f(\Gamma_{12}) = 0 \tag{32}$$

If we use the full weak correlation approximation then $n(1...5)$ splits into a product of $n(j)'s$, but for our purposes it will not always be necessary to neglect all the hard core interparticle correlations and so in order to make explicit the extent of the approximations needed we will continue as in deriving (26b), to neglect correlations only as necessary. The last two terms in (31) are zero if we use the partial weak correlation approximations, $n(12345) = \left[\frac{(A-3)(A-4)}{A(A-1)} \right] n(123) n(45)$

and $n(1234) = \left[\frac{(A-2)(A-3)}{A(A-1)} \right] n(12) n(34)$, because then

$$\langle f(\Gamma_{23}) f(\Gamma_{45}) \rangle_1 = \langle f(\Gamma_{23}) \rangle_1 \langle f(\Gamma_{45}) \rangle = 0 \tag{33}$$

and $\langle f(\Gamma_{12}) f(\Gamma_{34}) \rangle_1 = \langle f(\Gamma_{12}) \rangle_1 \langle f(\Gamma_{34}) \rangle = 0 \tag{34}$

where the last steps of (33) and (34) follow from the orthogonality condition (32). The approximation $n(1234) = \frac{(A-3)}{A} n(1) n(234)$ with (22) is enough to give

$$\langle f(\Gamma_{23}) f(\Gamma_{24}) \rangle_1 = \langle 1 \rangle_1 \langle f(\Gamma_{12}) f(\Gamma_{13}) \rangle \quad , \tag{35}$$

while the weak correlation approximation $n(12) = \frac{(A-1)}{A} n(1) n(2)$

and $n(123) = \frac{(A-1)(A-2)}{A^2} n(1) n(2) n(3)$ yields

$$\langle f(r_{12}) f(r_{13}) \rangle_1 = \langle f(r_{12}) \rangle_1^2 \tag{36}$$

and $$\langle f^2(r_{23}) \rangle_1 = \langle 1 \rangle_1 \langle f^2(r_{12}) \rangle \tag{37}$$

The last remaining term in (31), $\langle f(r_{12}) f(r_{23}) \rangle_1$, is slightly more complicated since using the approximation $n(123) =$ $\frac{(A-1)(A-2)}{A^2} n(1) n(2) n(3)$ only gives

$$\langle f(r_{12}) f(r_{23}) \rangle_1 = \langle 1 \rangle_1 \frac{1}{A} \int \underline{d2}\, n(2) f(r_{12}) \int \underline{d3}\, n(3) f(r_{23}) \tag{38}$$

So from (33) - (38) we find that the weak correlation approximation, or for some terms less than this, reduces (31) to

$$\langle |F|^2 \rangle_1 (r_1) = (A-1) \langle f^2(r_{12}) \rangle_1 + (A-1)(A-2) \Big\{ \langle f(r_{12}) \rangle_1^2 +$$

$$\langle f(r_{12}) f(r_{23}) \rangle_1 + \frac{\langle 1 \rangle_1}{2} \langle f^2(r_{12}) \rangle + (A-1)(A-2)^2 \langle 1 \rangle_1 \langle f(r_{12}) f(r_{13}) \rangle \Big\} \tag{39}$$

Evaluating $\langle |F|^2 \rangle$ with F given by (10) and (11) in a similar fashion to (31) and then using the partial weak correlation approximation $n(1234) = \left[\frac{(A-2)(A-3)}{A(A-1)} \right] n(12) n(34)$ along with the

orthogonality condition (32), we find that the four body term of $\langle |F|^2 \rangle$ is zero and so

$$\langle |F|^2 \rangle = \frac{A(A-1)}{2} \langle f^2(r_{12}) \rangle + A(A-1)(A-2) \langle f(r_{12}) f(r_{13}) \rangle \tag{40}$$

To calculate $n_1(r_1)$ from (20), (39) and (40) we can drop the first terms in (39) and (40) provided $\langle f(r_{12}) f(r_{13}) \rangle \gg O\left(\frac{1}{A} \langle f^2(r_{12}) \rangle \right)$ and assuming that A is larger than about 20. Then (20), (22), (23), (39) and (40) give

$$\eta_1(\vec{r}_1) \doteq n(\vec{r}_1) \left\{ 1 + \frac{\left[\langle f(\vec{r}_{12})\rangle_1^2 (\vec{r}_1) + \langle f(\vec{r}_{12}) f(\vec{r}_{23})\rangle_1 (\vec{r}_1) + \frac{\langle f^2(\vec{r}_{12})\rangle}{2} - 2\langle f(\vec{r}_{12}) f(\vec{r}_{13})\rangle \right]}{A \langle f(\vec{r}_{12}) f(\vec{r}_{13})\rangle} \right\}$$

(41)

Thus comparing (41) with (29) we see that provided $\langle f(\vec{r}_{12}) f(\vec{r}_{13})\rangle$

$\gg O\left(\frac{1}{A} \langle f^2(\vec{r}_{12})\rangle\right)$, then $\eta_1(\vec{r}_1)$ for the transla-

tionally invariant vibrational state (10) is very similar to the

density of the translationally variant state (18). Namely $\eta_1(\vec{r}_1)$

varies only to order $\frac{1}{A} n(\vec{r}_1)$ from the constant ground state

density $n(\vec{r}_1)$. Note from (20), (39) and (40), that if the

orthogonality condition happened to give $\langle f(\vec{r}_{12}) f(\vec{r}_{13})\rangle \lesssim O\left(\frac{1}{A} \langle f^2(\vec{r}_{12})\rangle\right)$

then the variable part of $\eta_1(\vec{r}_1)$ would be of the order of the

average value of $\eta_1(\vec{r}_1)$, so that these special states will have a

much higher energy than the type of state described by (41). Now

from (21) and (4) we have in the cutoff density approximation

$$\langle f(\vec{r}_{12})\rangle = \frac{4\pi}{V} \int_0^R d\vec{r}_1 \, r_1^2 \langle f(\vec{r}_{12})\rangle_1 (\vec{r}_1)$$

(42)

So using the orthogonality condition (11) we see that $\langle f(\vec{r}_{12})\rangle_1 (\vec{r}_1)$

must change sign at least once as $\vec{r}_1$ varies from zero to R. From

(38) and (32) we can also expect that $\langle f(\vec{r}_{12}) f(\vec{r}_{23})\rangle_1 (\vec{r}_1)$ will

have a minimum very near the values of $\vec{r}_1$ at which $\langle f(\vec{r}_{12})\rangle_1 (\vec{r}_1) = 0$

Therefore the orthogonality condition implies that $\eta_1(\vec{r}_1)$ (That is,

(41)) must pass through at least one shallow minimum as $\vec{r}_1$ varies

from zero to R.

So we have established that both the translationally invariant

state (10) and the translationally variant state (18) have densities

which pass through at least one shallow minimum. If F is oscillatory

and characterized by a wavenumber $\vec{k}$, then the orthogonality condition

will yield a discrete series of states labelled by increasing k .
We would expect the lowest energy state to have the smallest wave-
number (i.e. longest wavelength) and to have an excited state density
$n_1(r_1)$ with only one shallow minimum. For each wavenumber series
it is likely that the nth excited state will have a density with n
shallow minima. These shallow minima divide the excited system into
regions of mild compression and rarefaction, but the physical inter-
pretation of the vibration that produces these regions seems to have
two possibilities. One interpretation is that the vibrational motion
is small scale reflecting the shallowness of the density minima, and
the minima divide the system into regions moving out of phase with
one another. That is, the vibrational states are standing spherical
compressional waves with the lowest energy states having a wavelength
of the order of the size of the system. This interpretation therefore
excludes anything resembling the classical breathing mode oscillation
from being described by $\Psi = F \Phi$ with F of the form (10) or (18).
However this could be wrong if the excited state density $n_1(r_1)$
defined by (20) is too much of a quantum mechanical average. That is,
it may be that $n_1(r_1)$ averages out all oscillatory motion leaving
only the nett transfer of matter in the same way that occurred with
the quantum mechanical velocity field discussed in section BI.
In fact in appendix BI(2) $n_1(r_1)$ is calculated for a Feynman phonon
in bulk liquid helium,

$$\Psi = F \Phi \quad ; \quad F = \sum_j e^{i \underline{k} \cdot \underline{r}_j} \tag{43}$$

and in the same approximations used to derive (26) it was found that
$n_1(r_1) = n(r_1)$. Because it is known that as $k \to 0$ (43)
represents large scale periodic density variations we concluded in BI
that expressions of the type (20) are quantum mechanical averages of
all the motion, so that any oscillatory density variations are averaged

out. (See London [6] page 317 for a discussion on similar averaging problems in quantum mechanics.) As another example we note that the quantum mechanical velocity field BI(16) is zero for any state with a real wavefunction and all the vibrational states that we consider will be real. Thus the quantum velocity field has averaged out all the oscillatory motion and the zero result means only that there is no nett transfer of matter due to the vibrational motion, as we would expect for a pure vibration. So it is possible that the results (29) and (41) for the vibrational state density $\eta_i(r_i)$ have smoothed out all the vibrational motion and the shallow minima are just a small difference between the average density distribution of the excited state and the ground state, occurring because of the necessity that Ψ be orthogonal to Φ . If this is the correct interpretation of $\eta_i(r_i)$ then the vibrational motion may be a large scale density variation with the largest wavelength state closely resembling a breathing mode oscillation.

Another hint to the physical significance of our vibrational states is the close relation they have to the Feynman phonon-roton excitations of liquid helium, (43). In [2] Feynman describes the physical nature of the trial wavefunction $\sum_j f(r_j) \Phi$ and concludes that in the long wavelength region it represents a compressional density wave inside the fluid, (i.e. a sound wave). The only difference between this wavefunction and our finite system broken translational invariance state $\Psi = F\Phi$ with F given by (18), is that Feynman's state is characterized by a vector $\underline{k}$ and so acts in a specific direction, while (18) is purely radial. Hence a reasonable guess would be that in the long wavelength region $\Psi = F\Phi$ with F given by (18) should describe finite system radial analogues of Feynman's phonons. That is, it should describe radial compressional density waves. Now from the similarity of (29) and (41) it seems

likely that $\Psi = F \Phi$ with F given by (10) includes many of the translationally invariant forms of the states with F given by (18). This is closely related to the rotational states investigated in [1] where the 2^+ state was shown to be the translationally invariant form of the $L=2$ component of Feynman's wavefunction (43). So we expect that the vibrational states $\Psi = F \Phi$ with F given by (10) are translationally invariant relations of the states with F given by (18), which in turn appear to be finite system radial analogues of Feynman's compressional density wave excitations.

Let's now summarise on what the vibrational states $\Psi = F \Phi$ with F of the form (10) or (18) are likely to represent physically. Firstly they should describe radial, compressional, density oscillations directly analogous to Feynman's phonon excitations in liquid helium. The lowest energy states should have wavelengths of the order of the size of the system and there seems to be two possible interpretations of these large wavelength vibrations. One interpretation is that they are standing compressional density waves and could not include large scale inphase motion like a breathing mode oscillation, because the orthogonality condition insists that there be at least two regions of out of phase motion. The other interpretation suggested that the shallow density variations due to the orthogonality condition do not necessarily indicate out of phase motion, but may only be describing small density differences separating the excited state from the ground state after all the breathing mode like motion has been averaged out.

TRANSLATIONALLY INVARIANT VIBRATIONAL STATES.

We want to calculate the variational energy of the trial vibrational wavefunction $\Psi = F \Phi$ with F of the form (10) for a spherical, self-bound, many-body quantum system, described by a

Hamiltonian of the type (2). Provided the orthogonality condition (11) holds, then the variational upper bound to the vibrational energy E is given by (9). Inserting (10) into (9) we have (cf. [1] eq.(7))

$$\Delta E \equiv E - E_0 \leq \frac{\hbar^2}{m} \left\{ \frac{\langle f'^2(r_{12}) \rangle + (A-2) \langle \nabla_1 f(r_{12}) \cdot \nabla_1 f(r_{13}) \rangle}{\langle f^2(r_{12}) \rangle + 2(A-2) \langle f(r_{12}) f(r_{13}) \rangle + \frac{(A-2)(A-3)}{2} \langle f(r_{12}) f(r_{34}) \rangle} \right\}$$

(44)

where $H\Phi = E_0 \Phi$, $' = \frac{d}{dr_{12}}$ and $\langle \cdot \rangle$ is the ground state expectation value defined by (4). In order to evaluate (44) analytically it is necessary to make some approximations to the two, three and four particle density correlation functions, $n(1...S)$ (see AI(2)) that are needed to calculate the expectation values appearing in (44). Firstly we make the partial weak correlation approximation used to derive (34), namely

$$n(1234) = \left[\frac{(A-2)(A-3)}{A(A-1)} \right] n(12) n(34)$$

(45)

Note that (45) amounts to ignoring all pair correlations between particles (1,3) (1,4) (2,3) and (2,4) and all higher order correlations in the system, except those due to restricting the particles to move within the finite system. One type of error in this sort of approximation comes from ignoring the hard core interaction between particles, but as we have noted previously, the fact that the nucleon hard cores occupy less than a percent of the total volume, suggests that these approximations should be good for nuclei when calculating ΔE . *

The orthogonality condition (11) together with (45) immediately gives for the four body term of $\langle |F|^2 \rangle$,

$$\langle f(r_{12}) f(r_{34}) \rangle = \langle f(r_{12}) \rangle^2 = 0$$

(46)

We can prove that the three body terms of $\langle |\nabla_1 F|^2 \rangle$ and $\langle |F|^2 \rangle$ are positive definite by making the fairly mild approximation

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* See the note on page AIII.44.

$$n(123) = n(1)n(2)n(3) g(r_{12}) g(r_{13}) \tag{47}$$

where $g(r)$ is the pair correlation function. (cf. AI(6)). (47) amounts to keeping the pair correlations between particles (1,2) and (1,3) but ignores all other correlations other than those due to the finiteness of the system. Using (4), AI(2) and (47) it follows that

$$\left\langle \underline{\nabla}_1 f(r_{12}) \cdot \underline{\nabla}_1 f(r_{13}) \right\rangle = \mathcal{I}^{-1} \int \underline{d1}\, n(1)\, \underline{A}^2(1) \geq 0 \tag{48a}$$

and

$$\left\langle f(r_{12}) f(r_{13}) \right\rangle = \mathcal{I}^{-1} \int \underline{d1}\, n(1)\, B^2(1) \geq 0 \tag{48b}$$

where

$$\mathcal{I} = \int \underline{d1}\, \underline{d2}\, \underline{d3}\, n(123) \tag{48c}$$

$$\underline{A}(1) = \int \underline{d2}\, n(2)\, g(r_{12})\, \underline{\nabla}_1 f(r_{12}) \quad , \quad A^2 = \underline{A} \cdot \underline{A} \tag{48d}$$

$$B(1) = \int \underline{d2}\, n(2)\, g(r_{12})\, f(r_{12}) \tag{48e}$$

Therefore with only (45) and (47) we have proved that every term in (44) is positive definite, which is a result that from (9) seems likely to be completely general.

We now assume that the ground state is spherically symmetric, so that for example, $\left\langle x_{12}^2 \right\rangle = \left\langle y_{12}^2 \right\rangle = \left\langle z_{12}^2 \right\rangle$, $\left\langle x_{12} x_{13} \right\rangle = \left\langle y_{12} y_{13} \right\rangle$, etc. The two major approximations made to evaluate the ground state expectation values appearing in (44) will be the weak correlation approximation for $n(12)$ and $n(123)$ and the cutoff density approximation (22) for the ground state number density. We have already discussed the validity of the weak correlation approximation for evaluating the excited state collective energy and we concluded that it should be fairly

good for even-even nuclei. The cutoff density approximation should describe the ground states of non deformed even-even nuclei reasonably well when A is greater than about 20. Further discussions can be found in [1] where the same approximations are used to evaluate the energy of a related rotational excitation. Because of these approximations we will restrict the quantitative calculations to non deformed even-even nuclei, although the results could be qualitatively applied to other many-body quantum systems.

Let $h(r_{12})$ be a general function of r_{12}, then using spherical bipolar coordinates (see AI(7) and [1] eq's (32) and (35)) and the weak correlation approximation for $n(12)$ and $n(123)$, we have

$$\langle h(r_{12}) \rangle = \frac{\int_o^\infty dr_1\, r_1\, n(r_1) \int_o^\infty dr_2\, r_2\, n(r_2) \int_{|r_1-r_2|}^{r_1+r_2} dr_{12}\, r_{12}\, h(r_{12})}{2\left[\int_o^\infty dr_1\, r_1^2\, n(r_1)\right]^2} \tag{49a}$$

$$\langle h(r_{12}) h(r_{13}) \rangle = \frac{\int_o^\infty dr_1\, n(r_1) \int_o^\infty dr_2\, r_2\, n(r_2) \int_o^\infty dr_3\, r_3\, n(r_3) \int_{|r_1-r_2|}^{r_1+r_2} dr_{12}\, r_{12}\, h(r_{12}) \int_{|r_1-r_3|}^{r_1+r_3} dr_{13}\, r_{13}\, h(r_{13})}{4\left[\int_o^\infty dr_1\, r_1^2\, n(r_1)\right]^3} \tag{49b}$$

Next, define the variables x, y, t by

$$x = \frac{r_1}{R}, \quad y = \frac{r_2}{R}, \quad t = \frac{r_{12}}{R} \tag{50}$$

then using (50) it follows that the cutoff density approximation (22) reduces (49) to

$$\langle h(r_{12}) \rangle = \frac{9}{2} \int_o^1 dx\, x \int_o^1 dy\, y \int_{|x-y|}^{x+y} dt\, t\, h(Rt) \tag{51a}$$

$$\langle h(r_{12}) h(r_{13}) \rangle = \frac{27}{4} \int_o^1 dx \left[\int_o^1 dy\, y \int_{|x-y|}^{x+y} dt\, t\, h(Rt)\right]^2 \tag{51b}$$

The next step is to decide what specific forms for (10) we might expect to represent low energy states of the wavefunction $\Psi = F\Phi$ with F given by (10). Towards this end let's first discuss the translationally variant form (18), in the hope of generalising a specific choice to its translationally invariant form in a similar way to the rotational states investigated in [1] . Feynman has shown that for an infinite system the best wavefunction of the form $\sum_j f(r_j) \Phi$ satisfies the equation,([2] page 265.)

$$-\frac{\hbar^2}{2m} \nabla_1^2 f(r_1) = \Delta E \left\{ f(r_1) + \int d2 \, n(2) f(r_2) g(r_{12}) \right\} \qquad (52)$$

Using the weak correlation approximation (i.e. $g(r_{12})$ = a constant) and the orthogonality condition (cf. (27)), (52) becomes

$$\nabla_1^2 f(r_1) + \left(\frac{2m\Delta E}{\hbar^2} \right) f(r_1) = 0 \qquad (53)$$

Now (53) has more than just the plane wave solution (43). If we write

$$f(r_1) = h(r_1) Y_{LM}(\theta_1, \phi_1) \quad \text{then in the spherical coordinates}$$
$$(r, \theta, \phi) = (r_1, \theta_1, \phi_1) \quad , \text{ (53) is}$$

$$\nabla_r^2 h(r) + \left(k^2 - \frac{L(L+1)}{r^2} \right) h(r) = 0 \qquad (54)$$

where $k^2 = \left(\frac{2m\Delta E}{\hbar^2} \right)$ and $\nabla_r^2 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial}{\partial r} \right) = \frac{\partial^2}{\partial r^2} + \frac{2}{r} \frac{\partial}{\partial r}$.

From appendix AI(4) we see that the regular solution of (54) is the spherical Bessel $h(r) = j_L(kr)$. Therefore we have found that for a spherical system and neglecting surface effects, that the best state of angular momentum L of the type $\Psi = F\Phi$ with F given by

$$F = \sum_j Y_{LM}(\theta_j, \phi_j) h(r_j) \qquad (55a)$$

is, at least in the weak correlation approximation,

$$\Psi = \sum_j Y_{LM}(\theta_j, \phi_j) \, j_L(k r_j) \cdot \Phi \tag{55b}$$

Hence an obvious candidate for a low lying vibrational state of the form $\Psi = F\Phi$ is the $L = 0$ component of (55), namely

$$F = \sum_j j_o(k r_j) = \sum_j \frac{\sin k r_j}{k r_j} \tag{56}$$

Unfortunately the translationally invariant form of (56) is not apparent, but a wavefunction of the form (10) that might be closely related to (56) and hence should be investigated to test this, is obviously

$$F = \sum_{i<j}^{A} \sum^{A} j_o(k r_{ij}) \tag{57}$$

Another way to create a translationally invariant vibrational state of the form $\Psi = F\Phi$ is to superpose two phonon states (43) with opposite momenta, to produce an excitation of zero total momentum. Following Feynman ([2] page 268) we expect the wavefunction for this excitation to be $\Psi = F\Phi$ with

$$F = \sum_i \sum_{j \neq i} e^{i k \cdot (r_i - r_j)} = 2 \sum_{i<j} \sum \cos k \cdot (r_i - r_j) \tag{58}$$

If we choose the Z axis to be in the direction of k , then the non radial vibration (58) becomes

$$F = 2 \sum_{i<j} \sum \cos k Z_{ij} \tag{59}$$

This suggests that a low lying radial vibration of the type $\Psi = F\Phi$, with F of the form (10), is given by

$$F = \sum_{i<j}^{A} \sum^{A} \cos k r_{ij} \tag{60}$$

So we will now investigate the two translationally invariant wave-functions (57) and (60), to see if they include vibrational states whose energies are close enough to the ground state energy to be experimentally detected in non deformed even-even nuclei.

$$\Psi = \sum_{i<j}^{A} \sum_{j}^{A} J_0(k r_{ij}) \, \Phi \; .$$

(61)

The orthogonality condition (11) becomes using (51a)

$$\int_0^1 dx \, x \int_0^1 dy \, y \int_{|x-y|}^{x+y} dt \, t \, J_0(\alpha t) = 0$$

(62)

where $\alpha = kR$. Substituting $J_0(\alpha t) = \frac{\sin \alpha t}{\alpha t}$ into (62) and integrating over dt gives

$$\left[\int_0^1 dx \, x \sin \alpha x \right]^2 = 0$$

(63)

From appendix AI(4)(iii) we see that the orthogonality condition reduces to

$$J_1(\alpha) = 0 \qquad , \quad \alpha = kR$$

(64)

The first solution to (64) is $\alpha \doteq 4 \cdot 4934$. From (51b) we have

$$\left\langle J_0(k r_{12}) J_0(k r_{13}) \right\rangle = \frac{27}{4\alpha^2} \int_0^1 dx \left[\int_0^1 dy \, y \int_{|x-y|}^{x+y} dt \, \sin \alpha t \right]^2$$

$$= \frac{27}{\alpha^4} \int_0^1 dx \, x \sin^2 \alpha x \left[\int_0^1 dy \, y \sin \alpha y \right]^2$$

$$= \frac{27 J_1^2(\alpha)}{\alpha^4} \int_0^1 dx \, x \sin^2 \alpha x$$

which from (64)

$$= 0$$

(65)

Note that if we removed the cutoff density approximation (22) and kept $n(1)$ as a general radial density $n(r_i)$, then (65) would still hold because of the orthogonality condition, which becomes $\int_0^\infty dx \, x \, n(x) \sin \alpha x = 0$.

From appendix AI(4) $j_0'(k r_{12}) = -k \, j_1(k r_{12})$, so that (44), (46) and (65) give

$$\Delta E \leq \frac{\hbar^2}{m} \left\{ \frac{k^2 \langle j_1^2(k r_{12}) \rangle + (A-2) \langle \nabla_1 \, j_0(k r_{12}) \cdot \nabla_1 \, j_0(k r_{13}) \rangle}{\langle j_0^2(k r_{12}) \rangle} \right\} \qquad (66)$$

Now we have already shown with (48a) that

$\langle \nabla_1 \, j_0(k r_{12}) \cdot \nabla_1 \, j_0(k r_{13}) \rangle$ is non negative, so that the right hand side of (66) is at least

$$\frac{\hbar^2 k^2}{m} \frac{\langle j_1^2(k r_{12}) \rangle}{\langle j_0^2(k r_{12}) \rangle} \qquad (67)$$

and is probably of order A larger than this for small k. In appendix AIII(i) it is proved that $j_1(\alpha) = 0$ implies that $\langle j_1^2(k r_{12}) \rangle$ tends to $\langle j_0^2(k r_{12}) \rangle$ rapidly for large α , and that for the smallest value of α ($\alpha = 4.4934$) $\langle j_1^2(k r_{12}) \rangle \doteqdot 0.891 \langle j_0^2(k r_{12}) \rangle$.

Therefore the variational bound of the vibrational energy is at least of order

$$\frac{\hbar^2 k^2}{m} = \frac{\hbar^2 \alpha^2}{m R^2} \qquad (68)$$

For spherical nuclei we put $R = r_0 A^{1/3}$ with $r_0 = 1.2 \, fm.$, giving (cf. [1])

$$\frac{\hbar^2}{m R^2} = 28.77 \, A^{-2/3} \, Mev. \qquad (69)$$

From above we see that the lowest energy state of (61), namely

$\alpha \doteqdot 4.4934$, has a variational energy bound that is at least

$517 \, A^{-2/3} \, Mev$. Now it is very unlikely that for small α

$\langle \nabla_1 \, j_0(k r_{12}) \cdot \nabla_1 \, j_0(k r_{13}) \rangle \leq O\left(\frac{k^2}{A} \langle j_1^2(k r_{12}) \rangle \right)$ as well as (65), so

that the variational energy is almost certainly of order A higher than (68) for small α , and because even if $\langle \underline{\nabla}_1 j_0(kr_{12}) \cdot \underline{\nabla}_1 j_0(kr_{13}) \rangle$ was zero the energy would still be greater than $517 A^{-2/3}$ Mev , it is not worth the large effort to evaluate this remaining term. Note that in the previous work on $\eta_1(r_i)$ (see discussion after (41)) we found that if (65) held then there was a much larger difference between the ground state and excited state densities than usual for states of the form (10). So the conjecture that the energy of (61) is of order A higher than (68), is supported by the previous discussion of $\eta_1(r_i)$. Therefore we will end the investigation of (61), which was necessary because of the apparent relation between (56) and (57), and instead go on to consider other vibrational states including some that have energies significantly lower than $517 A^{-2/3}$ Mev , thereby definitely eliminating the wavefunction (61) from being important.

$$\Psi = \sum_{i<j}^{A}\sum^{A} \cos k r_{ij} \; \Phi \; : \tag{70}$$

The orthogonality condition for the wavefunction (70) is, using (11), (50), (51a) and defining $\alpha = kR$,

$$\int_0^1 dx\, x \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, t \cos\alpha t \;\; = 0 \tag{71}$$

This can be written as

$$0 \;\; = \frac{d}{d\alpha} \int_0^1 dx\, x \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, \sin\alpha t$$

$$= 2\frac{d}{d\alpha}\left(\frac{1}{\alpha} \int_0^1 dx\, x \sin\alpha x \int_0^1 dy\, y \sin\alpha y\right) \tag{72}$$

From appendix AI(4) (72) becomes

$$0 = \frac{d}{d\alpha}\left(\frac{j_1^2(\alpha)}{\alpha}\right) = \frac{j_1(\alpha)}{\alpha}\left[2\,j_0(\alpha) - \frac{5}{\alpha}\,j_1(\alpha)\right] \tag{73}$$

Therefore (70) has two distinct series of states, defined by

$$j_1(\alpha) = 0 \quad : \quad \alpha \doteq 4.4934,\ 7.7253,\ \cdots \tag{74a}$$

$$j_0(\alpha) = \frac{5}{2\alpha}j_1(\alpha) : \quad \alpha \doteq 1.5255,\ 5.8513,\ \cdots \tag{74b}$$

Inserting (70) into (44) with (46), we have

$$\Delta E \leq \frac{\hbar^2 k^2}{m}\left\{\frac{\langle \sin^2 k r_{12}\rangle + (A-2)\left\langle \frac{\sin k r_{12}}{r_{12}}\frac{\sin k r_{13}}{r_{13}}(\underline{r}_{12}\cdot\underline{r}_{13})\right\rangle}{\langle \cos^2 k r_{12}\rangle + 2(A-2)\langle \cos k r_{12}\cos k r_{13}\rangle}\right\} \tag{75}$$

In appendix AIII(2) we calculate, in the weak correlation and cutoff density approximations,

$$\langle \cos^2 k r_{12}\rangle = 1 - \langle \sin^2 k r_{12}\rangle = \frac{1}{2} + \frac{9\,j_1(2\alpha)}{2\alpha}\left\{j_0(2\alpha) - \frac{5\,j_1(2\alpha)}{4\alpha}\right\} \tag{76a}$$

and

$$\langle \cos k r_{12}\cos k r_{13}\rangle = 27\,j_1^2(\alpha)\int_0^1 dx\, x^4 j_1^2(\alpha x) + $$
$$27\left(j_0(\alpha)-\frac{2}{\alpha}j_1(\alpha)\right)^2\int_0^1 dx\, x^2 j_0^2(\alpha x) - 54\,j_1(\alpha)\left(j_0(\alpha)-\frac{2}{\alpha}j_1(\alpha)\right)\int_0^1 dx\, x^3 j_0(\alpha x)j_1(\alpha x) \tag{76b}$$

Let's now consider the states $\quad j_1(\alpha) = 0$

$$j_1(\alpha) = 0 \Rightarrow j_0(\alpha) = \cos\alpha,\ j_0(2\alpha) = j_0^2(\alpha),\ j_1(2\alpha) = \frac{\alpha}{2}j_0^2(\alpha) \tag{77}$$

From (76a), (76b) and (77) we get

$$\langle \cos^2 k r_{12} \rangle = \frac{1}{2} + \frac{27}{32} \, j_0^4(\alpha) \tag{78a}$$

$$\langle \sin^2 k r_{12} \rangle = \frac{1}{2} - \frac{27}{32} \, j_0^4(\alpha) \tag{78b}$$

and $\quad \langle \cos k r_{12} \cos k r_{13} \rangle = 27 \, j_0^2(\alpha) \int_0^1 dx \, x^2 \, j_0^2(\alpha x) \tag{79a}$

which from appendix AIII(2) ((i) is transformed using $1 = \cos^2 + \sin^2)$

and (77), reduces to

$$\langle \cos k r_{12} \cos k r_{13} \rangle = \frac{27}{2} \, j_0^4(\alpha) \tag{79b}$$

From Table I and appendix AIII(2) we find

$$\alpha = 1.5255 \; : \; \langle \cos k r_{12} \cos k r_{13} \rangle = 0.0248 ; \tag{80b}$$

$$\langle \cos^2 k r_{12} \rangle = 0.256 , \; \langle \sin^2 k r_{12} \rangle = 0.744$$

$$\alpha = 5.8513 \; : \; \langle \cos k r_{12} \cos k r_{13} \rangle = 0.0033 ; \tag{80c}$$

$$\langle \cos^2 k r_{12} \rangle = 0.502 , \; \langle \sin^2 k r_{12} \rangle = 0.498$$

The last remaining term to evaluate in (75) is the three body numerator term, which is of the form

$$\langle h(r_{12}) h(r_{13}) \, \underline{r_{12}} . \underline{r_{13}} \rangle = \langle h(r_{12}) h(r_{13}) (r_{12}^2 - r_2^2 + \underline{r_2} . \underline{r_3}) \rangle \tag{81}$$

The right hand side of (81) follows from the symmetry of particles 2 and 3 and the identity

$$r_{12}^2 = r_1^2 + r_2^2 - 2 \underline{r_1} . \underline{r_2} \tag{82}$$

Using the partial weak correlation approximation (47) we find, in the same way that (48) was derived,

$$\langle h(r_{12}) h(r_{13}) \underline{r_2} . \underline{r_3} \rangle = g^{-1} \int \underline{d_1} \, n(1) \, \underline{C}^2(1) \qquad \geq 0 \tag{83a}$$

where $\quad \underline{C}(1) = \int \underline{d_2} \, n(2) \, g(r_{12}) h(r_{12}) \underline{r_2} \quad , \quad C^2 = \underline{C} . \underline{C} \tag{83b}$

and $\mathcal{g}$ is defined by (48c). For a spherical system we can evaluate $\underline{C}(1)$ by rotating $\underline{d r_2}$ about the $\underline{r_1}$ axis for all values of r_{12} , so

$$\underline{C}(1) = \underline{r_1} \, \widetilde{C}(r_1) \tag{84}$$

where $\widetilde{C}(r_1)$ is some function of r_1 , yet to be found. If we now equate $\underline{r_1} . \underline{C}(1)$ for $\underline{C}(1)$ given by both (83b) and (84) we have

$$\widetilde{C}(r_1) = \frac{1}{r_1^2} \int \underline{d_2} \, n(2) \, g(r_{12}) h(r_{12}) (\underline{r_1} . \underline{r_2}) \tag{85}$$

From (84) $\underline{C}(1) \cdot \underline{C}(1) = r_1^2 \widetilde{C}(r_1)^2 = C^2(r_1)$ where, using (82)

$$C(r_1) = \frac{1}{2r_1} \int d2\, n(2)\, g(r_{12})\, h(r_{12})\, (r_1^2 + r_2^2 - r_{12}^2) \qquad (86)$$

If we now use the weak correlation and cutoff density approximations then (83a) and (86) give

$$\left\langle h(r_{12})\, h(r_{13})\, \underline{r_2} \cdot \underline{r_3} \right\rangle = \frac{27}{16} R^2 \int_0^1 dx\, \frac{F^2(x)}{x^2} \qquad (87a)$$

where $\qquad F(x) = x^2 L(x) + K(x) - P(x)$ $\qquad\qquad (87b)$

$$L(x) = \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, t\, h(Rt) \qquad (87c)$$

$$K(x) = \int_0^1 dy\, y^3 \int_{|x-y|}^{x+y} dt\, t\, h(Rt) \qquad (87d)$$

$$P(x) = \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, t^3\, h(Rt) \qquad (87e)$$

and X, y, t are defined by (50). Therefore from (81) and (87) the three body numerator term of (75) is

$$\left\langle \frac{\sin k r_{12}}{r_{12}} \frac{\sin k r_{13}}{r_{13}} \left(\underline{r_{12}} \cdot \underline{r_{13}}\right) \right\rangle = \frac{27}{16} R^2 \int_0^1 dx \left\{ x^2 L^2(x) + \right.$$
$$\left. \frac{1}{x^2} (P(x) - k(x)) \left[2x^2 L(x) + P(x) - k(x) \right] \right\} \qquad (88)$$

where $L(x), k(x), P(x)$ are given by (87c,d,e) with

$$h(Rt) = \frac{\sin \alpha t}{Rt} \quad ; \quad \alpha = kR.$$

For the states $J_1(\alpha) = 0$ (see (77)), we see from appendix AIII(2) that,

$$J_1(\alpha) = 0 \implies L(x) = 0, \; R(P(x) - k(x)) = 4x^2 J_1(\alpha x) J_0(\alpha) \qquad (89)$$

Therefore from (88), when $j_1(\alpha) = 0$

$$\left\langle \frac{\sin k \, r_{12}}{r_{12}} \frac{\sin k \, r_{13}}{r_{13}} \cdot \underline{r_{12}} \cdot \underline{r_{13}} \right\rangle = 27 j_0^2(\alpha) \int_0^1 dx \, x^2 \, j_1^2(\alpha x) \tag{90}$$

which from appendix AIII(2) and (76b) becomes

$$\left\langle \frac{\sin k \, r_{12}}{r_{12}} \frac{\sin k \, r_{13}}{r_{13}} \underline{r_{12}} \cdot \underline{r_{13}} \right\rangle = \frac{27}{2} j_0^4(\alpha) \tag{91}$$

For the states $j_0(\alpha) = \frac{5}{2\alpha} j_1(\alpha)$ (see Table I for the first two states), we have from appendix AIII(2),

$$R \, L(x) = 2 \times j_0(\alpha x) j_1(\alpha)$$

$$R(P(x) - k(x)) = 2 x^3 j_1(\alpha) \left(j_0(\alpha x) - \frac{1}{\alpha x} j_1(\alpha x) \right) \tag{92}$$

From (88) and (92) we have,

$$\left\langle \frac{\sin k \, r_{12}}{r_{12}} \frac{\sin k \, r_{13}}{r_{13}} \underline{r_{12}} \cdot \underline{r_{13}} \right\rangle = 27 j_1^2(\alpha) \left\{ \int_0^1 dx \, x^4 \, j_0^2(\alpha x) - \right. \tag{93a}$$

$$\left. \frac{1}{\alpha} \int_0^1 dx \, x^3 j_0(\alpha x) j_1(\alpha x) + \frac{1}{4\alpha^2} \int_0^1 dx \, x^2 j_1^2(\alpha x) \right\}$$

From Table I and appendix AIII(2) we find for the first two

$$j_0(\alpha) = \frac{5}{2\alpha} j_1(\alpha) \text{ states,}$$

$$\alpha = 1.5255 : \quad \left\langle \frac{\sin k \, r_{12}}{r_{12}} \frac{\sin k \, r_{13}}{r_{13}} \underline{r_{12}} \cdot \underline{r_{13}} \right\rangle = 0.3204 \tag{93b}$$

$$\alpha = 5.8513 : \quad \left\langle \frac{\sin k \, r_{12}}{r_{12}} \frac{\sin k \, r_{13}}{r_{13}} \underline{r_{12}} \cdot \underline{r_{13}} \right\rangle = 0.0039 \tag{93c}$$

We are now in a position to calculate the variational bound of the energy of the vibrational state (70), in the weak correlation and cutoff density approximations.

(a) For the series of states defined by $j_1(\alpha) = 0$, we have from (75), (78), (79) and (91)

$$\Delta E \leq \frac{\hbar^2 k^2}{m} \left\{ \frac{\left(\frac{1}{2} - \frac{27}{2} J_0^4(\alpha)\right) + (A-2)\frac{27}{2} J_0^4(\alpha)}{\left(\frac{1}{2} + \frac{27}{2} J_0^4(\alpha)\right) + 2(A-2)\frac{27}{2} J_0^4(\alpha)} \right\} \tag{94}$$

Now $J_0(\alpha) = \frac{\sin\alpha}{\alpha}$ so that $J_0^4(\alpha) \leq \frac{1}{\alpha^4}$. Therefore (94) varies from $\frac{\hbar^2 k^2}{2m}$ to $\frac{\hbar^2 k^2}{m}$ depending on the relative values of A and α . For the lowest state, namely $\alpha \doteq 4.4934$, we see that for $A \gtrsim 50$

$$\Delta E \lesssim \frac{\hbar^2 k^2}{2m} = \frac{\hbar^2 \alpha^2}{2m R^2} = 10.1 \frac{\hbar^2}{m R^2} \quad , \quad \alpha = 4.4934 \tag{95a}$$

For spherical nuclei we use (69) to obtain, for the lowest state of the series $J_1(\alpha) = 0$

$$\Delta E \lesssim 290\, A^{-2/3}\, \text{Mev} \quad , \quad A \gtrsim 50 \tag{95b}$$

As an example, for $A = 208 \left(\text{i.e. } Pb^{208}\right)$, the exact expression (94) gives $\Delta E \leq 8.5\, \text{Mev}$. So we see that this vibrational state is outside the well documented region of experimental nuclear physics, but is still experimentally accessible. (It has an energy of about half that of the giant resonance states [8] page 734.)

(b) For the excited states given by $J_0(\alpha) = \frac{5}{2\alpha} J_1(\alpha)$, we will calculate the energies of only the first two states, namely $\alpha \doteq 1.5255$, and $\alpha \doteq 5.8513$. From (75), (80) and (93) we have

$$\alpha \doteq 1.5255 \;, \quad \Delta E \leq \frac{\hbar^2 k^2}{m} \left\{ \frac{0.744 + (A-2).0.3204}{0.256 + 2(A-2).0.0248} \right\} \tag{96a}$$

$$\alpha \doteq 5.8513 \;, \quad \Delta E \leq \frac{\hbar^2 k^2}{m} \left\{ \frac{0.498 + (A-2).0.0039}{0.502 + 2(A-2).0.0033} \right\} \tag{96b}$$

From (80), (93) and appendix AIII(2) it is easily seen that for large α,

$\langle \cos k \vec{r}_{12} \cos k \vec{r}_{13} \rangle$ tends to $\langle \frac{\sin k \vec{r}_{12}}{\vec{r}_{12}} \frac{\sin k \vec{r}_{13}}{\vec{r}_{13}} \vec{r}_{12} \cdot \vec{r}_{13} \rangle$ and they both become of order $\left(\frac{1}{\alpha} \right)^4$. So the variational energy again varies between $\frac{\hbar^2 k^2}{2m}$ and $\frac{\hbar^2 k^2}{m}$ depending on the relative values of A and α , except for the lowest state (96a) which is around $6 \frac{\hbar^2 k^2}{m}$. When $A \gtrsim 50$ we can drop the first terms in the numerator and denominator of (96a), so that for spherical nuclei (i.e. using (69)) the energy of the lowest state in the series

$$j_0(\alpha) = \frac{5}{2\alpha} j_1(\alpha) \text{ is,}$$

$$\alpha \doteq 1 \cdot 5255 \quad : \quad \Delta E \lesssim \frac{15 \hbar^2}{mR^2} \doteq 430 A^{-2/3} \text{ Mev} \tag{97}$$

When A is in the region 100 to 200 we see from (96b) that the next state, namely $\alpha \doteq 5.8513$, has a variational energy of around,

$$\Delta E \sim \frac{3}{4} \frac{\hbar^2 \alpha^2}{mR^2} = 25.7 \frac{\hbar^2}{mR^2} \quad , \quad \alpha = 5.8513 \tag{98}$$

Therefore we have found, using the weak correlation and cutoff density approximations, that the vibrational state (70) has two series of states defined by $j_1(kR) = 0$ and $j_0(kR) = \frac{5}{2kR} j_1(kR)$, and depending on the relative values of A and kR the energies of both series vary between $\frac{\hbar^2 k^2}{2m}$ and $\frac{\hbar^2 k^2}{m}$, except for the lowest state of the second series which is anomolously higher than this. Within each series of states the larger k is (i.e. the smaller the wavelength) the higher the energy, which is what you would expect. The state with the lowest energy is the first $j_1(kR) = 0$ state,

$$kR \doteq 4 \cdot 4934 \quad \text{and for spherical nuclei its energy was}$$
$$\Delta E \lesssim 290 A^{-2/3} \text{ Mev} \quad \text{for } A \gtrsim 50.$$

To conclude the investigation into the state (70) we will now discuss the excited state density variations given by equation (41). That is, whenever $\langle \cos k \vec{r}_{12} \rangle_1 (\vec{r}_1)$ is zero the excited state density has a shallow minimum. Using the bipolar coordinates introduced in equation (49), it follows from (42), in the weak correlation and

cutoff density approximations, that

$$\langle h(r_{12}) \rangle_1 (r_1) = \frac{\int_0^R dr_2 \, r_2 \int_{|r_1 - r_2|}^{r_1 + r_2} dr_{12} \cdot r_{12} \, h(r_{12})}{\int_0^R dr_2 \, r_2 \int_{|r_1 - r_2|}^{r_1 + r_2} dr_{12} \cdot r_{12}} \tag{99}$$

Using the variables x, y, t defined by (50) and $\alpha = kR$, we have

$$\langle \cos\alpha t \rangle_1 (x) = \frac{3}{2x} \int_0^1 dy \, y \int_{|x-y|}^{x+y} dt \, t \cos\alpha t \tag{100a}$$

From appendix AIII(2) equation (d) this becomes

$$\langle \cos\alpha t \rangle_1 (x) = 3\left(j_0(\alpha) - \frac{2}{\alpha} j_1(\alpha) \right) j_0(\alpha x) - 3x \, j_1(\alpha x) \, j_1(\alpha) \tag{100b}$$

For the two series of states, we have from (100), as x varies from 0 to 1,

$$j_1(\alpha) = 0 \; ; \quad \langle \cos\alpha t \rangle_1 (x) = 3 j_0(\alpha) \, j_0(\alpha x) \tag{101a}$$

$$j_0(\alpha) = \frac{5}{2\alpha} j_1(\alpha) \; ; \quad \langle \cos\alpha t \rangle_1 (x) = \frac{3}{5} j_0(\alpha)\left(2\cos\alpha x - j_0(\alpha x) \right) \tag{101b}$$

In figure 1. the function $\langle \cos\alpha t \rangle_1 (x)$ is plotted for the first two states of each series (i.e. $j_1(\alpha) = 0 : \alpha \doteq 4.4934, \, 7.7253 ;$ $j_0(\alpha) = \frac{5}{2\alpha} j_1(\alpha) : \alpha \doteq 1.5255, \, 5.8513 .$)

We see that the lowest energy (longest wavelength) state of each series has only one shallow density minimum, while the second lowest states have two shallow minima. If the density minima are interpreted as dividing the system into regions of vibrational motion of alternating phase then the above leads us to conclude that the wavefunction (70) represents two series of spherical standing compressional waves, with the nth member of each series containing n radial nodes.

$\langle \cos \alpha t \rangle_1 (x)$

Fig. 1.

$\langle \cos \alpha t \rangle_1(x)$ is plotted for:

A $\alpha \doteq 4.4934$

B $\alpha \doteq 7.7253$

C $\alpha \doteq 1.5255$

D $\alpha \doteq 5.8513$
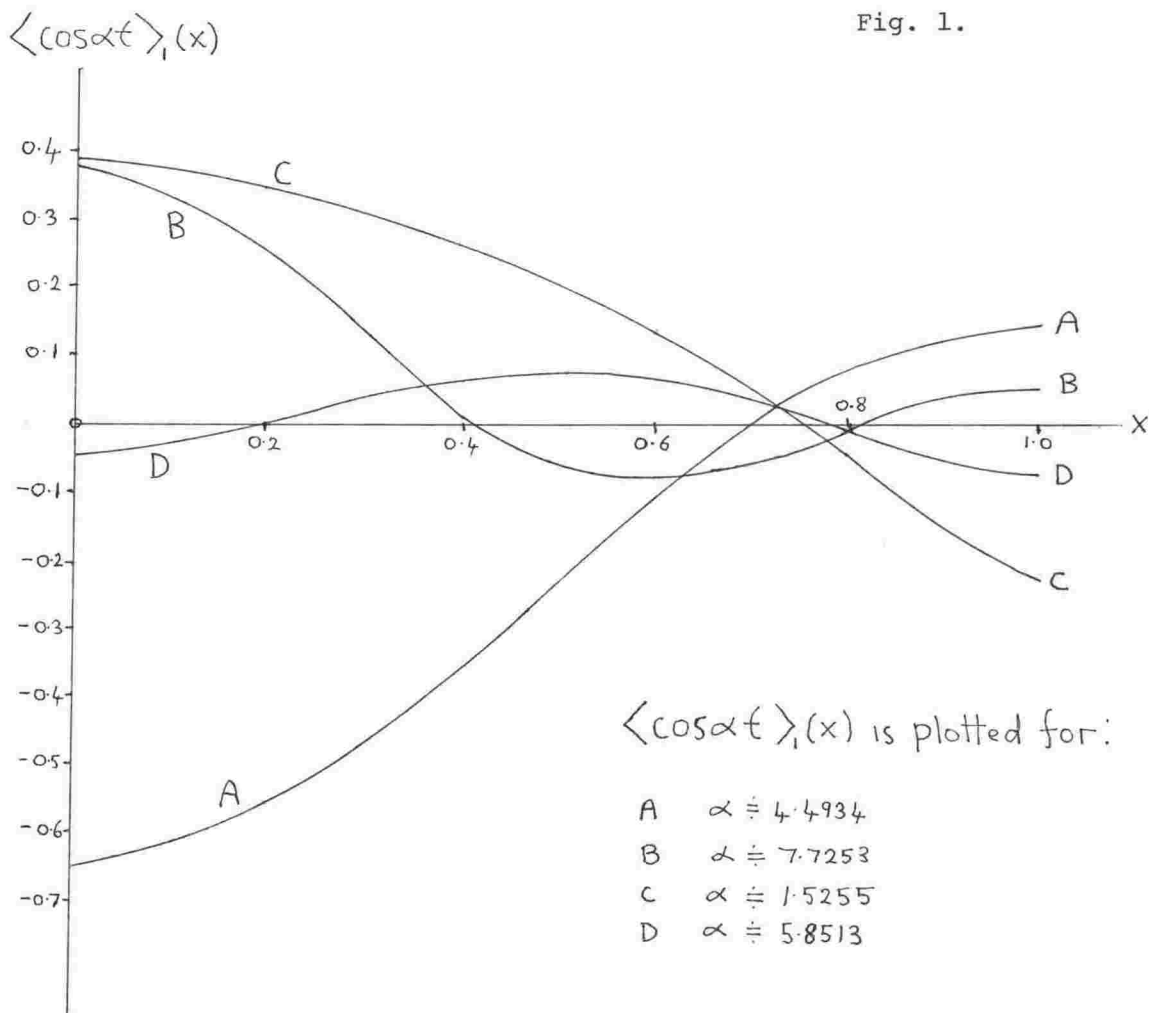
OTHER TRANSLATIONALLY INVARIANT STATES:

It is of importance to the interpretation of the physical nature of the vibrational states to consider for comparison, the two-phonon non radial vibrational (58), namely

$$\Psi = \sum_i^A \sum_{j \neq i}^A e^{i \underline{k} \cdot (\underline{r}_i - \underline{r}_j)} \, \Phi \; = 2 \sum_{i < j}^A \sum^A \cos \underline{k} \cdot (\underline{r}_i - \underline{r}_j) \, \Phi \qquad (102)$$

For this wavefunction the weak correlation approximation reduces the orthogonality condition (8) to

$$\langle e^{i \underline{k} \cdot \underline{r}_i} \rangle = 0 \qquad (103)$$

Because the system is spherical we can integrate over the angles and

then using the cutoff density approximation, we see that (103) becomes

$$\int_0^R dr_1 \, r_1^2 \, j_0(kr_1) = 0 \tag{104}$$

From appendix AI(4) and defining $\alpha = kR$, the orthogonality condition reduces to

$$j_1(\alpha) = 0 \tag{105}$$

Now, inserting (102) into (9), and using the partial weak correlation approximation (45) along with the orthogonality condition to make the four-body denominator term zero, we have

$$\Delta E \leq \frac{\hbar^2}{m} \left\{ \frac{\langle |\nabla_1 e^{i\underline{k}\cdot\underline{r}_{12}}|^2 \rangle + (A-2)\langle \nabla_1 e^{i\underline{k}\cdot\underline{r}_{12}} \cdot \nabla_1 e^{-i\underline{k}\cdot\underline{r}_{13}} \rangle}{\langle |e^{i\underline{k}\cdot\underline{r}_{12}}|^2 \rangle + 2(A-2)\langle e^{i\underline{k}\cdot\underline{r}_{12}} e^{-i\underline{k}\cdot\underline{r}_{13}} \rangle} \right\} \tag{106}$$

Using $\quad \nabla_1 e^{i\underline{k}\cdot\underline{r}_{12}} = e^{-i\underline{k}\cdot\underline{r}_2} \nabla_1 e^{i\underline{k}\cdot\underline{r}_1} = i\underline{k}\, e^{i\underline{k}\cdot\underline{r}_{12}} \quad$, we see that

$$\left\langle \nabla_1 e^{i\underline{k}\cdot\underline{r}_{12}} \cdot \nabla_1 e^{i\underline{k}\cdot\underline{r}_{13}} \right\rangle = k^2 \left\langle e^{i\underline{k}\cdot\underline{r}_{12}} e^{-i\underline{k}\cdot\underline{r}_{13}} \right\rangle = k^2 \left\langle e^{i\underline{k}\cdot\underline{r}_{32}} \right\rangle = 0 \tag{107}$$

where the last step in (107) is the orthogonality condition.

Therefore we find, with only the approximation (45),

$$\Delta E \leq \frac{\hbar^2 k^2}{m} = \frac{\hbar^2 \alpha^2}{mR^2} \quad , \quad j_1(\alpha) = 0 \tag{108}$$

(108) verifies Feynman's statement ([2] page 268), that (102) represents a superposition of two phonons with a total energy of twice that of a single phonon ($E = \frac{\hbar^2 k^2}{2m}$ in the weak correlation limit) within correction terms of order $1/\sqrt{}$, that would appear if we hadn't used the partial weak correlation approximation (45). From (105) and (108) we notice the close relation of this known state to our radial vibrations. Thus we have further evidence that for long wavelengths the vibrational states $\Psi = F\Phi$, with F given by (10) or (18) are finite system radial analogues of Feynman's compressional phonons.

To find more candidates for low energy translationally invariant vibrational states, we will try to find exact eigenstates of the type $\Psi = \sum_{i<j}\sum f(r_{ij})\,\Phi$, when Φ is the ground state of an A particle system with harmonic pair interactions, namely

$$\Phi = \exp\left\{-\beta \sum_{i<j}^{A}\sum^{A} r_{ij}^{2}\right\} \qquad , \qquad \beta \text{ a constant} \qquad (109)$$

Except for very small A (109) is a poor approximation to a real nuclear ground state, but its simplicity will allow us to find exact eigenstates of the form $\sum_{i<j}\sum f(r_{ij})\,\Phi$ which should be fairly good trial wavefunctions when Φ is left as the exact ground state. With the Hamiltonian H of the form (2) and $H\Phi = E_{o}\Phi$, we have when $\Psi = F\Phi$,

$$\left(H-E_{o}\right)\Psi = -\frac{\hbar^{2}}{2m}\sum_{k}\left\{\Phi \nabla_{k}^{2}F + 2\underline{\nabla}_{k}\Phi\cdot\underline{\nabla}_{k}F\right\} \qquad (110)$$

With F of the form (10) $\left(F = \sum_{i<j}\sum f(r_{ij})\right)$, we have from (12) and (109)

$$\nabla_{k}^{2}F = \sum_{k}\sum_{j\neq k}\left(f''(r_{kj}) + \frac{2f'(r_{kj})}{r_{kj}}\right) = 2\sum_{k<j}\sum\left(f''(r_{kj}) + 2\frac{f'(r_{kj})}{r_{kj}}\right) \quad (111)$$

and

$$\sum_{k}\underline{\nabla}_{k}\Phi\cdot\underline{\nabla}_{k}F = -2\beta\Phi\sum_{k}\sum_{j\neq k}\sum_{\ell\neq k}\frac{\underline{r}_{k\ell}\cdot\underline{r}_{kj}}{r_{kj}}f'(r_{kj}) \qquad (112)$$

$$= -2A\beta\Phi\sum_{k}\sum_{j\neq k}(\underline{r}_{k}-\underline{R})\cdot\frac{\underline{r}_{kj}}{r_{kj}}f'(r_{kj})$$

where $\underline{R} = (X,Y,Z) \qquad ; \qquad X = \frac{1}{A}\sum_{j}^{A}x_{j} \qquad$ etc.

Then using the identity (15) in (112) we have

$$\sum_{k}\underline{\nabla}_{k}\Phi\cdot\underline{\nabla}_{k}F = -2A\beta\Phi\sum_{k<j}\sum r_{kj}f'(r_{kj}) \qquad (113)$$

Therefore from (110), (111) and (113) we find

$$(H-E_0)\Psi = -\frac{\hbar^2 \Phi}{m} \sum_{k<j}^{A} \sum_{}^{A} \left\{ f''(r_{kj}) + f'(r_{kj}) \left(\frac{2}{r_{kj}} - 2A\beta r_{kj} \right) \right\} \quad (114)$$

From (114) we see that for Φ given by (109), $\Psi = \sum_{i<j} \sum f(r_{ij}) \Phi$ is an exact eigenstate (with $\Delta E = \hbar^2 q_i / m$), provided

$$f''(r_{12}) + f'(r_{12}) \left(\frac{2}{r_{12}} - 2A\beta r_{12} \right) + q_i f(r_{12}) = 0 \quad (115)$$

We can break the translational invariance of Ψ using the identity ([1] eq.(43))

$$\sum_{i<j}^{A} \sum_{}^{A} r_{ij}^2 = A \sum_{j}^{A} r_j^2 - A^2 R^2 \quad (116)$$

and putting $\underline{R} = (0,0,0)$. Then Φ becomes,

$$\Phi = \exp \left\{ -A\beta \sum_{j}^{A} r_j^2 \right\} \quad (117)$$

and for F of the form (18) $\left(F = \sum_{j}^{A} f(r_j) \right)$, (110) leads to

$$(H-E_0)\Psi = \frac{\hbar^2 q_v}{m} \Psi \qquad \text{provided}$$

$$f''(r) + f'(r) \left(\frac{2}{r} - 4A\beta r \right) + 2 q_v f(r) = 0 \quad (118)$$

The solutions to (115) and (118) are combinations of Hermite polynomials (cf. [9] page 111), with the simplest one being $f(r) = r^2 - C$; a constant. That is, for F of the form (10) and (18) respectively, exact eigenstates for a harmonic pair interaction ground state are :

$$F = \sum_{i<j}^{A} \sum_{}^{A} (r_{ij}^2 - C_i) \quad (119a); \qquad F = \sum_{j}^{A} (r_j^2 - C_v) \quad (119b)$$

Inserting (119) into (115) and (118) we find,

$$q_i = q_v = 4A\beta (=q) \; ; \; C_i = 6/q \;, \; C_v = 3/q \quad (120)$$

The orthogonality conditions (11) and (19) yield for (119)

$$C_i = \langle r_{12}^2 \rangle \quad ; \quad C_v = \langle r_1^2 \rangle \quad (121)$$

So from (120) and (121) we have $\langle \Gamma_{12}^2 \rangle = 2 \langle \Gamma_1^2 \rangle$, which is

a result that was verified in the weak correlation approximation in

[1] (eq.(32)), where it was shown that $g(\Gamma_{12}) = 1$ led to

$$\langle \Gamma_{12}^2 \rangle = 2 \overline{r^2} = \langle \Gamma_1^2 \rangle \quad \text{with}$$

$$\overline{f(r)} \equiv \frac{\int_0^\infty dr\, r^2 n(r) f(r)}{\int_0^\infty dr\, r^2 n(r)} \tag{122}$$

Now, using the identity (116) and putting $\underline{R} = (0,0,0)$ we can break

the translational invariance of (119a) to obtain,

$$\sum_{i<j}^A \sum^A (\Gamma_{ij}^2 - C_i) = A \sum_J^A \Gamma_J^2 - \frac{A(A-1)}{2} C_i \tag{123}$$

$$= A \sum_J^A \left(\Gamma_J^2 - \frac{C_i}{2} \right) + O(1)$$

So (119a) is the translationally invariant form of (119b), which

explains why (120) gave $q_i = q_v$ and $C_i = 2 C_v$

We now revert to leaving $\overline{\Phi}$ as the exact ground state and

consider the vibrational state

$$\Psi = \sum_{i<j}^A \sum^A \left(\Gamma_{ij}^2 - \langle \Gamma_{12}^2 \rangle \right) \overline{\Phi} . \tag{124}$$

The wavefunction (124) is just one orthogonal state. Inserting

(124) into (44) and using (46) we have

$$\Delta E \leq \frac{4\hbar^2}{m} \left\{ \frac{\langle \Gamma_{12}^2 \rangle + (A-2) \langle \underline{\Gamma_{12}} \cdot \underline{\Gamma_{13}} \rangle}{\langle (\Gamma_{12}^2 - \langle \Gamma_{12}^2 \rangle)^2 \rangle + 2(A-2) \langle (\Gamma_{12}^2 - \langle \Gamma_{12}^2 \rangle)(\Gamma_{13}^2 - \langle \Gamma_{12}^2 \rangle) \rangle} \right\} \tag{125}$$

For a spherical system we can use the identity

$$2 Z_{12} Z_{13} = Z_{12}^2 + Z_{13}^2 - Z_{23}^2 \tag{126}$$

to show that

$$\langle \underline{\Gamma_{12}} \cdot \underline{\Gamma_{13}} \rangle = 3 \langle Z_{12} Z_{13} \rangle = \frac{3}{2} \langle Z_{12}^2 \rangle = \frac{\langle \Gamma_{12}^2 \rangle}{2} \tag{127}$$

Using the weak correlation approximation we see from (49), that

$$\langle r_{12}^2 \rangle = 2 \overline{r^2} \ ; \ \langle r_{12}^4 \rangle = 2 \overline{r^4} + \frac{10}{3}(\overline{r^2})^2 \ ; \ \langle r_{12}^2 r_{13}^2 \rangle = \overline{r^4} + 3(\overline{r^2})^2 \tag{128}$$

where $\overline{f(r)}$ is defined by (122). In the cutoff density approximation,

$$\overline{r^n} = \frac{3}{n+3} R^n \tag{129}$$

For $A \gtrsim 50$ we can drop the first terms in the numerator and denominator of (125), so that, using (128) and (129)

$$\Delta E \lesssim 2 \frac{\hbar^2}{m} \frac{\overline{r^2}}{(\overline{r^4} - (\overline{r^2})^2)} = 17.5 \frac{\hbar^2}{m R^2} \tag{130}$$

For spherical nuclei (130) becomes, using (69),

$$\Delta E \lesssim 500 A^{-2/3} \ \text{Mev} \tag{131}$$

The rule that the smoother the wavefunction the lower the energy suggests with (119) that we should try the wavefunction

$$\underline{\Psi = \sum_{i<j}^A \sum^A (r_{ij} - \langle r_{12} \rangle) \Phi \ .} \tag{132}$$

The wavefunction (132) is just one orthogonal state and should have an energy smaller than (124), despite not being an exact eigenstate when Φ is the harmonic pair interaction ground state. Inserting (132) into (44) and using (46) gives

$$\Delta E \leq \frac{\hbar^2}{m} \left\{ \frac{1 + (A-2) \langle \frac{r_{12} \cdot r_{13}}{r_{12} \ r_{13}} \rangle}{\langle (r_{12} - \langle r_{12} \rangle)^2 \rangle + 2(A-2) \langle (r_{12} - \langle r_{12} \rangle)(r_{13} - \langle r_{12} \rangle) \rangle} \right\} \tag{133}$$

For $A \gtrsim 50$ (133) reduces to

$$\Delta E \lesssim \frac{\hbar^2 \left\langle \frac{\vec{r}_{12} \cdot \vec{r}_{13}}{r_{12} r_{13}} \right\rangle}{2m \left[\langle r_{12} r_{13} \rangle - \langle r_{12} \rangle^2 \right]} \tag{134}$$

In the weak correlation and cutoff density approximations, $\left\langle \frac{\vec{r}_{12} \cdot \vec{r}_{13}}{r_{12} r_{13}} \right\rangle$

is given by the right hand side of (88) where $L(x)$, $K(x)$ and

$P(x)$ are given by (87 c-e) with $h(R t) = 1/Rt$

That is,

$$RL(x) = \int_0^x dy\, y \int_{x-y}^{x+y} dt + \int_x^1 dy\, y \int_{y-x}^{x+y} dt = x - \frac{x^3}{3} \tag{135a}$$

$$RK(x) = \int_0^x dy\, y^3 \int_{x-y}^{x+y} dt + \int_x^1 dy\, y^3 \int_{y-x}^{x+y} dt = \frac{x}{2} - \frac{x^5}{10} \tag{135b}$$

$$RP(x) = \int_0^x dy\, y \int_{x-y}^{x+y} dt\, t^2 + \int_x^1 dy\, y \int_{y-x}^{x+y} dt\, t^2 = \frac{x}{2} + \frac{x^3}{3} - \frac{x^5}{30} \tag{135c}$$

From (88) and (135) we have

$$\left\langle \frac{\vec{r}_{12} \cdot \vec{r}_{13}}{r_{12} r_{13}} \right\rangle = \frac{27}{16} \int_0^1 dx \left\{ \frac{16}{9} x^4 - \frac{32}{45} x^6 + \frac{16}{225} x^8 \right\}$$

$$= \frac{232}{525} \doteq 0.4419 \tag{136}$$

Using (51) we also have

$$\langle r_{12} r_{13} \rangle = \frac{27}{4} R^4 \int_0^1 dx\, P^2(x)$$

$$= \frac{27}{4} R^2 \int_0^1 dx \left\{ \frac{x^2}{4} + \frac{x^4}{3} + \frac{7x^6}{90} - \frac{x^8}{45} + \frac{x^{10}}{900} \right\}$$

$$\doteq 1.0716 R^2 \tag{137}$$

and

$$\langle r_{12} \rangle = \frac{9}{2} R^2 \int_0^1 dx\, x\, P(x)$$

$$= \frac{9}{2} R \int_0^1 dx \left\{ \frac{x^2}{2} + \frac{x^4}{3} - \frac{x^6}{30} \right\}$$

$$= \frac{36}{35} R \qquad (138)$$

Therefore from (134) and (136-8), we find that for $A \gtrsim 50$ the variational energy of the state (132), in the weak correlation and cutoff density approximations, is

$$\Delta E \leq 16.2 \frac{\hbar^2}{m R^2} \qquad (139)$$

which for spherical nuclei becomes, using (69),

$$\Delta E \leq 466 A^{-2/3} \, Mev \qquad (140)$$

TRANSLATIONALLY *VARIANT* VIBRATIONAL STATES :

We now consider trial wavefunctions of the type $\Psi = F \Phi$, where Φ is a spherical ground state and F is of the form (18), namely $F = \sum_j^A f(r_j)$. Taking F to be real and enforcing the orthogonality condition (19), we find from (9) that the variational bound for the vibrational energy is

$$\Delta E = E - E_0 \leq \frac{\hbar^2}{2m} \left\{ \frac{\langle f'^2(r_1) \rangle}{\langle f^2(r_1) \rangle + (A-1) \langle f(r_1) f(r_2) \rangle} \right\} \qquad (141)$$

where $f'(r_1) = \frac{df(r_1)}{dr_1}$ and $\langle \ \rangle$ is the ground state expectation value (4). To evaluate (141) we will again use the weak correlation approximation, $n(12) = \frac{(A-1)}{A} n(1) n(2)$, so that together with the orthogonality condition (19) we have

$$\langle f(r_1) f(r_2) \rangle = \langle f(r_1) \rangle^2 = 0 \qquad (142)$$

That is, in the weak correlation approximation

$$\Delta E \leq \frac{\hbar^2}{2m} \frac{\langle f'^2(r_i) \rangle}{\langle f^2(r_i) \rangle} \tag{143}$$

where $$\langle f(r_i) \rangle = 0$$

Before evaluating (143) for specific choices of F let's summarise the many conclusions and approximations that we have previously discussed. Firstly, Φ is taken to be the spherical ground state of a self-bound many-body system described by a Hamiltonian of the type (2). So for nuclei we have neglected spin and any differences between the protons and neutrons, when the vibrational energy is calculated. The weak correlation approximation is also used in calculating the vibrational energy and this restricts the quantitative validity of our results to the case of non deformed even-even nuclei. $\Psi = \sum_j f(r_j) \Phi$ has the same parity, permutation symmetry and angular momentum as the ground state Φ. (e.g. For even-even nuclei Ψ and Φ are 0^+ states.) In evaluating expectation values we approximate the ground state density by the cutoff density

$$
\begin{aligned}
n(r) &= A/V \quad , \quad r \leq R \quad \left(V = \tfrac{4}{3}\pi R^3 \right) \\
&= 0 \quad , \quad r > R
\end{aligned}
\tag{144}
$$

Note, that defining $x = r_i/R$, and using (144) gives for $h(r_i)$ a function of r_i

$$\langle h(r_i) \rangle = 3 \int_0^1 dx \, x^2 \, h(Rx) \tag{145}$$

Finally, the fact that $F = \sum_j f(r_j)$ is not translationally invariant means that unless F can be shown to be closely related to a translationally invariant state, it is possible that $\Psi = F\Phi$ may be a spurious centre of mass motion state, that doesn't represent a real internal motion.

What are some likely low energy trial wavefunctions of the form $\sum_j f(r_j) \Phi$? Firstly we have already seen from (52-6) that when $f(r_j) = j_0(k r_j)$, that for small k, $F\Phi$ should represent a finite system spherical analogue of a Feynman phonon. By analogy with (70) we will also look at $\sum_j \cos k r_j \Phi$. Also, we have already found (119b) that $\sum_j (r_j^2 - \bar{r}^2) \Phi$ is an exact eigenstate of the Hamiltonian (2) when Φ is the harmonic pair interaction ground state. This is a very poor approximation to a nuclear ground state but the result (119b) does suggest that $f(r_j) = (r_j^2 - \bar{r}^2)$ will be a low lying vibrational state of the form (18). A wavefunction that is worth considering because it has less curvature than the wavefunction $\sum_j (r_j^2 - \bar{r}^2) \Phi$ and hence a lower energy, is $\sum_j (r_j - \bar{r}) \Phi$ In appendix AIII(3) we evaluate ΔE for the above trial wavefunctions and the results are summarised and compared with the translationally invariant wavefunctions in table II (next page).

TABLE II :

Trial Wavefunction	Orthogonality Condition. (if required) $\alpha = kR$	Variational bound to vibrational energy. ie. $\Delta E \leq$	ΔE for the lowest state of spherical nuclei. $\left(\frac{\hbar^2}{mR^2} = 28.77\, A^{-2/3}\,\text{Mev.}\right)$	Example: $A=208$ ie. Pb^{208} (Energy of the lowest state.)
$\sum\limits_{j}^{A} J_0(kr_j)\Phi$	$J_1(\alpha) = 0$	$\dfrac{\hbar^2 k^2}{2m} = \dfrac{\hbar^2 \alpha^2}{2mR^2}$	$290\, A^{-2/3}\,\text{Mev}$ $(\alpha = 4.4934)$	8.27 Mev
$\sum\limits_{j}^{A} \cos k r_j\, \Phi$	$J_0(\alpha) = \dfrac{2}{\alpha} J_1(\alpha)$	$\dfrac{\hbar^2 \alpha^2}{2mR^2}\left[\dfrac{1-3\left(J_0(2\alpha)-\frac{J_1(2\alpha)}{\alpha}\right)}{1+3\left(J_0(2\alpha)-\frac{J_1(2\alpha)}{\alpha}\right)}\right]$	$390\, A^{-2/3}\,\text{Mev}$ $(\alpha = 2.0816)$	11.1 Mev
$\sum\limits_{j}^{A} (r_j^2 - \bar{r}^2)\Phi$		$17.5\,\dfrac{\hbar^2}{mR^2}$	$500\, A^{-2/3}\,\text{Mev}$	14.3 Mev
$\sum\limits_{j}^{A} (r_j - \bar{r})\Phi$		$\dfrac{40}{3}\,\dfrac{\hbar^2}{mR^2}$	$384\, A^{-2/3}\,\text{Mev}$	10.9 Mev

Translationally Invariant Wavefunctions :

Trial Wavefunction	Orthogonality Condition	Variational bound to vibrational energy	ΔE for the lowest state of spherical nuclei	Example: $A=208$
$\sum\limits_{i<j}^{A}\sum^{A} J_0(k r_{ij})\Phi$	$J_1(\alpha) = 0$	At least (and probably of order A larger.) $\sim \dfrac{\hbar^2 \alpha^2}{mR^2}$	At least (and probably of order A larger.) $517\, A^{-2/3}\,\text{Mev}$ $(\alpha = 4.4934)$	
$\sum\limits_{i<j}^{A}\sum^{A} \cos k r_{ij}\,\Phi$	$J_1(\alpha) = 0$	$\dfrac{\hbar^2 \alpha^2}{2mR^2}$, $A \geq \dfrac{\alpha^4}{8}$	$290\, A^{-2/3}\,\text{Mev}$ $(\alpha = 4.4934\,;\, A \geq 50)$	8.5 Mev (exact expression (94))
	$J_0(\alpha) = \dfrac{5}{2\alpha} J_1(\alpha)$		$430\, A^{-2/3}\,\text{Mev}$ $(\alpha = 1.5255\,;\, A \geq 50)$	12.3 Mev
$\sum\limits_{i<j}^{A}\sum^{A} (r_{ij}^2 - \langle r_{12}^2 \rangle)\Phi$		$A \geq 50$ $17.5\,\dfrac{\hbar^2}{mR^2}$	$500\, A^{-2/3}\,\text{Mev}$	14.3 Mev
$\sum\limits_{i<j}^{A}\sum^{A} (r_{ij} - \langle r_{12} \rangle)\Phi$		$A \geq 50$ $16.2\,\dfrac{\hbar^2}{mR^2}$	$466\, A^{-2/3}\,\text{Mev}$	13.3 Mev

CONCLUDING REMARKS :

(1) We proved in (123) that $\sum_{i<j}\sum (r_{ij}^2 - \langle r_{12}^2 \rangle) \Phi$ is the translationally invariant form of $\sum_j (r_j^2 - \bar{r}^2) \Phi$. The results in Table II verify this.

(2) From Table II we see that the energy of $\sum_j (r_j - \bar{r}) \Phi$ is significantly lower than the energy of $\sum_{i<j}\sum (r_{ij} - \langle r_{12} \rangle) \Phi$ and so we cannot be sure that $\sum_j (r_j - \bar{r}) \Phi$ doesn't represent spurious motions occurring because it is not translationally invariant.

(3) From Table II we see that $\sum_j \cos k r_j \Phi$ has a very different orthogonality condition to $\sum_{i<j}\sum \cos k r_{ij} \Phi$ and so these wavefunctions are probably not closely related. Instead, from its energy the lowest state of $\sum_j \cos k r_j \Phi$ appears to be approximating the same motion as $\sum_j (r_j - \bar{r}) \Phi$, which unfortunately we cannot be certain isn't spurious.

(4) Also from Table II we see that $\sum_{i<j}\sum j_0(k r_{ij}) \Phi$ cannot be related to $\sum_j j_0(k r_j) \Phi$ because it has a much higher energy.

(5) The lowest states are, the first of the $j_1(\alpha) = 0$ series of $\sum_{i<j}\sum \cos k r_{ij} \Phi$ and the lowest state of $\sum_j j_0(k r_j) \Phi$. These two wavefunctions have the same orthogonality condition and for $A \gtrsim \alpha^4/8$ the same variational energy. So for low k they appear to be closely related.

The conclusion (5) is the most important one, because we found that for small k $\sum_j j_0(k r_j) \Phi$ is a radial analogue of a Feynman phonon. The fact that in the weak correlation approximation the energy was found to be $\dfrac{\hbar^2 k^2}{2m}$ further strengthens this. But, the real wavefunction of a self-bound many-body system must be translationally invariant, so the best wavefunction is

$$\Psi = \sum_{i<j}^A \sum^A \cos k r_{ij} \Phi \tag{70}$$

From conclusion (5) we can expect that for low k (70) represents a standing radial compressional density wave, analogous to Feynman's phonons in liquid helium. The lowest energy state of (70) is

$$kR \doteq 4.4934$$ and, like all the states we considered, is proportional to $A^{-2/3}$. For Pb^{208} the lowest state gave

$$\Delta E = 8.5 \, Mev$$, which places it in an energy range in between the low lying states and the giant resonance states.

Finally, as a speculative aside, we should remember that there could be a relationship between these $\hbar^2 k^2 / 2m$ phonon states and the breathing mode state. Namely, when investigating the excited state density we found that the orthogonality condition insisted that there be at least one shallow minimum in the density. This immediately led us to compressional phonon like states, but because of the large degree of cancellation that is always present in quantum mechanical averages like the density expression, it was not possible with the lowest state to rule out large scale in-phase motion similar to a breathing mode. This would be important if we could find an $\hbar^2 k^2 / 2m$ state such that $kR = \pi/2 \, (\lambda = 4R)$, since then

$$\frac{\hbar^2 k^2}{2m} = 35.5 \, A^{-2/3} \, Mev$$ (using (69)), which is a line that passes near to many of the low 0^+ states of even-even nuclei, including the new collective states discovered by Maher et.al. [10]. However, for a radial wavefunction of the form $\sum_j f(kr_j) \Phi$, the orthogonality condition would require $kR \gtrsim \pi \, (\lambda \lesssim 2R)$ so that we would need a wavefunction at least as complicated as

$$\sum_{i<j} \sum f(kr_{ij}) \Phi .$$

NOTE :

This Section was written (together with sections AI and BI) some time before the rest of this thesis, as part of a submission to the

1851 Science Research Scholarship Committee. Since this time I have

learnt more about the physical nature of the above vibrational states

and have come to doubt the validity of the weak correlation approximation,

but it did not seem sensible to change section AIII from the form in

which it was first written. Instead I have preferred to leave the new

results, which includes calculating the contributions to the $S(EO)_{class}$

sum rule from each of the vibrational states, to section AIV. In

particular, a more accurate description of the physical nature of these

states is given at the end of section AIV. Also, see note 5) of

section BII.

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APPENDIX AIII(1) : $\left\langle J_1^2(k\Gamma_{12})\right\rangle$ and $\left\langle J_0^2(k\Gamma_{12})\right\rangle$.

We wish to prove that in the cutoff density and weak correlation approximations the orthogonality condition $J_1(kR) = 0$ implies $\left\langle J_1^2(k\Gamma_{12})\right\rangle \approx \left\langle J_0^2(k\Gamma_{12})\right\rangle$. Firstly we rewrite (51a) using the result AI(19) which holds in the stated approximations, to obtain

$$\left\langle h(\Gamma_{12})\right\rangle = \frac{3}{R^3}\int_0^{2R}dr\,r^2\left[1-\frac{3}{2}\left(\frac{\Gamma}{2R}\right)+\frac{1}{2}\left(\frac{\Gamma}{2R}\right)^3\right]h(r)$$

Defining $y = kr$, $\alpha = kR$, we have

$$\left\langle J_1^2(k\Gamma_{12})\right\rangle = \frac{3}{16\alpha^3}\int_0^{2\alpha}dy\,y^2\left[16-12\frac{y}{\alpha}+\frac{y^3}{\alpha^3}\right]J_1^2(y)\tag{a}$$

$$\left\langle J_0^2(k\Gamma_{12})\right\rangle = \frac{3}{16\alpha^3}\int_0^{2\alpha}dy\,y^2\left[16-12\frac{y}{\alpha}+\frac{y^3}{\alpha^3}\right]J_0^2(y)\tag{b}$$

From now on we make constant use of the identities listed in appendix AI(4). Integrating by parts we find

$$\int_0^{2\alpha}dy\,(y^2J_1(y))J_1(y) = -4\alpha^2J_1(2\alpha)J_0(2\alpha) + \int_0^{2\alpha}dy\,y^2J_0^2(y)\tag{c}$$

$$\int_0^{2\alpha}dy\,(y^3J_1(y))J_1(y) = -8\alpha^3J_1(2\alpha)J_0(2\alpha)+\int_0^{2\alpha}dy\,y^2J_0(y)J_1(y) + \int_0^{2\alpha}dy\,y^3J_0^2(y)\tag{d}$$

$$\int_0^{2\alpha}dy\,(y^5J_1(y))J_1(y) = -32\alpha^5J_1(2\alpha)J_0(2\alpha)+3\int_0^{2\alpha}dy\,y^4J_0(y)J_1(y) + \int_0^{2\alpha}dy\,y^5J_0^2(y)\tag{e}$$

From (a) – (e) we have

$$\left\langle J_1^2(k\Gamma_{12})\right\rangle = \left\langle J_0^2(k\Gamma_{12})\right\rangle + \frac{3}{16\alpha^3}\left\{-\frac{12}{\alpha}\int_0^{2\alpha}dy\,y^2J_0(y)J_1(y) + \frac{3}{\alpha^3}\int_0^{2\alpha}dy\,y^4J_0(y)J_1(y)\right\}\tag{f}$$

Further integration by parts yields

$$\int_0^{2\alpha} dy (y^2 f_0(y)) f_1(y) = -4\alpha^2 f_0^2(2\alpha) + \int_0^{2\alpha} dy f_0(y) [2y f_0(y) - y^2 f_1(y)] \tag{g}$$

and

$$\int_0^{2\alpha} dy (y^2 f_0(y))(y^2 f_1(y)) = 16\alpha^4 f_1^2(2\alpha) - \int_0^{2\alpha} dy\, y^2 f_1(y)\, y^2 f_0(y) \tag{h}$$

Therefore from (g) and (h)

$$\int_0^{2\alpha} dy\, y^2 f_0(y) f_1(y) = -2\alpha^2 f_0^2(2\alpha) + \int_0^{2\alpha} dy\, y f_0^2(y) \tag{i}$$

and

$$\int_0^{2\alpha} dy\, y^4 f_0(y) f_1(y) = 8\alpha^4 f_1^2(2\alpha) \tag{j}$$

(f), (i) and (j) yield

$$\left\langle f_1^2(k r_{12}) \right\rangle = \left\langle f_0^2(k r_{12}) \right\rangle + \frac{q}{2\alpha^2} \left[f_0^2(2\alpha) + f_1^2(2\alpha) \right] - \frac{q}{4\alpha^4} \int_0^{2\alpha} dy\, y f_0^2(y) \tag{k}$$

When $\quad f_1(\alpha) = 0 \qquad$ (k) becomes (see (77))

$$\left\langle f_1^2(k r_{12}) \right\rangle = \left\langle f_0^2(k r_{12}) \right\rangle + \frac{q}{2\alpha^6} \sin^4\alpha \left(1 + \frac{\alpha^2}{4}\right) - \frac{q}{4\alpha^4} \int_0^{2\alpha} dy\, y f_0^2(y) \tag{l}$$

The last term left to calculate is,

$$\int_0^{2\alpha} dy\, y f_0^2(y) = \int_0^{2\alpha} dy\, \frac{\sin^2 y}{y} = \int_0^{2\alpha} dy\, \frac{(1-\cos 2y)}{2y}$$

$$= \frac{1}{2} \int_0^{4\alpha} dt\, \frac{(1-\cos t)}{t} \tag{m}$$

Now (m) can be expressed in terms of Euler's constant $(\gamma \doteqdot 0.577)$ and the cosine integral $\left(C_i(x) \right)$, $[7]$ page 231 :

$$\frac{1}{2} \int_0^{4\alpha} dt\, \frac{(1-\cos t)}{t} = \frac{1}{2} \left(\gamma + \ln 4\alpha - C_i(4\alpha) \right) \tag{n}$$

Therefore we have finally, when $\quad f_1(\alpha) = 0$

$$\left\langle f_1^2(k r_{12}) \right\rangle = \left\langle f_0^2(k r_{12}) \right\rangle + \frac{q}{2\alpha^6} \sin^4\alpha \left(1 + \frac{\alpha^2}{4}\right) - \frac{q}{8\alpha^4} \left(\gamma + \ln 4\alpha - C_i(4\alpha) \right) \tag{o}$$

From (b) we find, for $\alpha \gtrsim \pi$

$$\langle \jmath_0^2(k r_{12}) \rangle \approx \frac{9}{8\alpha^2} \tag{p}$$

(For $\alpha = 4.4934$, $\langle \jmath_0^2(k r_{12}) \rangle = \frac{8.878}{8\alpha^2} = 0.055$)

So from (o) and (p) we see that $\jmath_1(\alpha) = 0$ implies that $\langle \jmath_1^2(k r_{12}) \rangle$ rapidly tends to $\langle \jmath_0^2(k r_{12}) \rangle$ for large α. For the lowest value of α, namely $\alpha \doteqdot 4.4934$, we have from [7] page 243, $C_i(4\alpha) \doteqdot .044$ and taking $\gamma \doteqdot 0.577$, then (o) becomes

$$\langle \jmath_1^2(k r_{12}) \rangle \doteqdot \langle \jmath_0^2(k r_{12}) \rangle - 0.006 \tag{q}$$

Therefore from (p) and (q) when $\alpha = 4.4934$

$$\langle \jmath_1^2(k r_{12}) \rangle \doteqdot 0.891 \langle \jmath_0^2(k r_{12}) \rangle \tag{r}$$

———————

APPENDIX AIII(2) : Calculations towards the evaluation of (75).

In the following calculations we will make frequent use of the weak correlation limit and cutoff density approximation expressions (51a) and (51b) and the Bessel function properties listed in appendix AI(4). Firstly,

$$\langle \cos^2 k r_{12} \rangle = \frac{q}{2} \int_0^1 dx \, x \int_0^1 dy \, y \int_{|x-y|}^{x+y} dt \, t \cos^2 \alpha t$$

$$= \frac{1}{2} + \frac{q}{4} \int_0^1 dx \, x \int_0^1 dy \, y \int_{|x-y|}^{x+y} dt \, t \cos 2\alpha t$$

$$= \frac{1}{2} + \frac{q}{4} \frac{d}{d\beta} \left(\int_0^1 dx \, x \int_0^1 dy \, y \int_{|x-y|}^{x+y} dt \sin \beta t \right) \quad , \quad \beta = 2\alpha$$

$$= \frac{1}{2} + \frac{q}{2} \frac{d}{d\beta} \left(\frac{J_1^2(\beta)}{\beta} \right)$$

$$= \frac{1}{2} + \frac{q}{2\alpha} J_1(2\alpha) \left\{ J_0(2\alpha) - \frac{5}{4} \frac{J_1(2\alpha)}{\alpha} \right\} \tag{a}$$

Then using $\cos^2 + \sin^2 = 1$,

$$\langle \sin^2 k r_{12} \rangle = \frac{1}{2} - \frac{q}{2\alpha} J_1(2\alpha) \left\{ J_0(2\alpha) - \frac{5}{4} \frac{J_1(2\alpha)}{\alpha} \right\} \tag{b}$$

From (51b) we have

$$\langle \cos k r_{12} \cos k r_{13} \rangle = \frac{27}{4} \int_0^1 dx \, J^2(x) \tag{c}$$

Where

$$J(x) = \int_0^1 dy \, y \int_{|x-y|}^{x+y} dt \, t \cos \alpha t$$

$$= \frac{d}{d\alpha} \left(\int_0^1 dy \, y \int_{|x-y|}^{x+y} dt \sin \alpha t \right)$$

$$= \frac{d}{d\alpha} \left(2x J_0(\alpha x) J_1(\alpha) \right)$$

$$= 2x \left[J_0(\alpha x) J_1'(\alpha) - x J_1(\alpha x) J_1(\alpha) \right] \quad ; \quad \left(' = \frac{d}{d\alpha} \right) \tag{d}$$

(c) and (d) give

$$\langle \cos k\Gamma_{12} \cos k\Gamma_{13} \rangle = 27\, j_1^2(\alpha) \int_0^1 dx\, x^4 j_1^2(\alpha x) \;+$$

$$27\left(j_0(\alpha) - \frac{2}{\alpha} j_1(\alpha)\right)^2 \int_0^1 dx\, x^2 j_0^2(\alpha x) - 54\, j_1(\alpha)\left(j_0(\alpha) - \frac{2}{\alpha} j_1(\alpha)\right) \int_0^1 dx\, x^3 j_0(\alpha x) j_1(\alpha x) \tag{e}$$

Integrating by parts we find, putting $y = \alpha x$

$$\int_0^1 dx\, x^4 j_1^2(\alpha x) = \frac{1}{\alpha^5} \int_0^\alpha dy\, y^4 j_1^2(y)$$

$$= -\frac{j_0(\alpha) j_1(\alpha)}{\alpha} + \frac{1}{\alpha^5} \int_0^\alpha dy\, j_0(y)\left[4y^3 j_1(y) + 4\left(j_0(y) - \frac{2}{y} j_1(y)\right)\right]$$

$$= -\frac{j_0(\alpha) j_1(\alpha)}{\alpha} + \frac{2}{\alpha} \int_0^1 dx\, x^3 j_0(\alpha x) j_1(\alpha x) + \int_0^1 dx\, x^4 j_0^2(\alpha x) \tag{f}$$

and

$$\int_0^1 dx\, x^3 j_0(\alpha x) j_1(\alpha x) = \frac{1}{\alpha^4} \int_0^\alpha dy\, y^3 j_0(y) j_1(y)$$

$$= \frac{j_1^2(\alpha)}{\alpha} - \frac{1}{\alpha^4} \int_0^\alpha dy\, y^2 j_1(y)\left[j_1(y) + y\left(j_0(y) - \frac{2}{y} j_1(y)\right)\right]$$

$$= -\frac{j_1^2(\alpha)}{\alpha} + \frac{1}{\alpha} \int_0^1 dx\, x^2 j_1^2(\alpha x) - \int_0^1 dx\, x^3 j_0(\alpha x) j_1(\alpha x)$$

Therefore $$\int_0^1 dx\, x^3 j_0(\alpha x) j_1(\alpha x) = \frac{j_1^2(\alpha)}{2\alpha} + \frac{1}{2\alpha} \int_0^1 dx\, x^2 j_1^2(\alpha x) \tag{g}$$

Also

$$\int_0^1 dx\, x^2 j_1^2(\alpha x) = \frac{1}{\alpha^3} \int_0^\alpha dy\, y^2 j_1^2(y)$$

$$= -\frac{j_0(\alpha) j_1(\alpha)}{\alpha} + \frac{1}{\alpha^3} \int_0^\alpha dy\, j_0(y) y^2 j_0(y)$$

$$= -\frac{j_0(\alpha) j_1(\alpha)}{\alpha} + \int_0^1 dx\, x^2 j_0^2(\alpha x) \tag{h}$$

And

$$\int_0^1 dx\, x^2\, J_0^2(\alpha x) = \frac{1}{\alpha^2} \int_0^1 dx\, \sin^2 \alpha x$$

$$= \frac{1}{2\alpha^2} \int_0^1 dx\, (1 - \cos 2\alpha x)$$

$$= \frac{1}{2\alpha^2} \left(1 - J_0(2\alpha) \right) \tag{i}$$

Finally

$$\int_0^1 dx\, x^4\, J_0^2(\alpha x) = \frac{1}{2\alpha^2} \int_0^1 dx\, x^2(1 - \cos 2\alpha x)$$

$$= \frac{1}{6\alpha^2} + \frac{1}{2\alpha^2} \frac{d^2}{d\beta^2} \int_0^1 dx\, \cos \beta x \qquad , \beta = 2\alpha$$

$$= \frac{1}{6\alpha^2} + \frac{1}{2\alpha^2} \frac{d^2}{d\beta^2} J_0(\beta)$$

$$= \frac{1}{2\alpha^2} \left(\frac{1}{3} + \frac{J_1(2\alpha)}{\alpha} - J_0(2\alpha) \right) \tag{j}$$

We now go on to evaluate (87(c),(d),(e)) when $h(Rt) = k\, J_0(\alpha t)$

From (87c), $\quad RL(x) = \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, \sin \alpha t$

$$= 2x\, J_0(\alpha x)\, J_1(\alpha) \tag{k}$$

From (87d),

$$Rk(x) = \int_0^1 dy\, y^3 \int_{|x-y|}^{x+y} dt\, \sin \alpha t$$

$$= 2x\, J_0(\alpha x) \int_0^1 dy\, y^3 \sin \alpha y$$

$$= -2x\, J_0(\alpha x)\, \frac{d^2}{d\alpha^2} J_1(\alpha)$$

$$= 2x\, J_0(\alpha x) \left[J_1(\alpha) + \frac{2}{\alpha} J_0(\alpha) - \frac{6}{\alpha^2} J_1(\alpha) \right] \tag{l}$$

From (87e), $\quad RP(x) = \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, t^2 \sin \alpha t$

$$= -\frac{d^2}{d\alpha^2} L(x)$$

$$= -2x \, J_1(\alpha) \frac{d^2}{d\alpha^2} J_0(\alpha x) - 4x \frac{d}{d\alpha} J_1(\alpha) \frac{d}{d\alpha} J_0(\alpha x) + K(x)R$$

$$= 2x^3 J_1(\alpha)\left(J_0(\alpha x) - \frac{2}{\alpha x} J_1(\alpha x)\right) + 4x^2 J_1(\alpha x)\left(J_0(\alpha) - \frac{2}{\alpha} J_1(\alpha)\right) + RK(x) \quad \text{(m)}$$

For the first two states in the $J_0(\alpha) = \frac{5}{2\alpha} J_1(\alpha)$ series we calculate from the above, using table I,

	$\alpha = 1.5255$	$\alpha = 5.8513$
$\int_0^1 dx \, x^4 J_0^2(\alpha x) =$	0.1126	0.0057
$\int_0^1 dx \, x^3 J_0(\alpha x) J_1(\alpha x) =$	0.06445	0.0035
$\int_0^1 dx \, x^2 J_0^2(\alpha x) =$	0.2085	0.0156
$\int_0^1 dx \, x^4 J_1^2(\alpha x) =$	0.0256	0.0049
$\int_0^1 dx \, x^2 J_1^2(\alpha x) =$	0.03695	0.0135

APPENDIX AIII(3) : Translationally *Variant* States.

In the following calculations we make frequent use of (145) to evaluate (143); the approximations inherent in this are summarised in the main text. Also, we define $X = \Gamma_j/R$, $\alpha = kR$.

(1) $\Psi = \sum\limits_{j}^{A} j_0(k\Gamma_j) \, \bar{\Phi}$:

The orthogonality condition is

$$\int_0^1 dx \, x^2 \, j_0(\alpha x) = 0 \qquad\qquad\qquad \text{(i)}$$

$$\Rightarrow \quad j_1(\alpha) = 0 \qquad\qquad\qquad \text{(ii)}$$

Also, $\quad j_0'(k\Gamma_i) = -k \, j_1(k\Gamma_i) \qquad$ and

$$\langle j_1^2(k\Gamma_i) \rangle = 3 \int_0^1 dx \, x^2 \, j_1^2(\alpha x)$$

which after integrating by parts

$$= 3\left[-\frac{j_0(\alpha x)}{\alpha} x^2 j_1(\alpha x) \Big|_0^1 \right] + 3 \int_0^1 dx \, x^2 j_0^2(\alpha x)$$

using (ii) $\qquad\qquad = \langle j_0^2(k\Gamma_i) \rangle \qquad\qquad\qquad$ (iii)

Therefore from (143)

$$\Delta E \leq \frac{\hbar^2 k^2}{2m} = \frac{\hbar^2 \alpha^2}{2mR^2} \quad , \quad j_1(\alpha) = 0 \qquad\qquad \text{(iv)}$$

For non deformed even-even nuclei (iv) becomes, using (69),

$$\Delta E \leq 14.4\,\alpha^2 \, A^{-2/3} \, \text{Mev} \quad , \quad j_1(\alpha) = 0 \, (\text{i.e. } \alpha \doteq 4.4934; \cdots) \quad \text{(v)}$$

(2) $\Psi = \sum\limits_{j}^{A} \cos k\Gamma_j \, \bar{\Phi}$:

The orthogonality condition is

$$\int_0^1 dx \, x^2 \cos\alpha x = 0 \qquad\qquad\qquad \text{(vi)}$$

$$\Rightarrow \frac{d}{d\alpha} \int_0^1 dx \, x \sin\alpha x = 0$$

That is, $\dfrac{d}{d\alpha} f_1(\alpha) = 0$, which is $f_0(\alpha) = \dfrac{2}{\alpha} f_1(\alpha)$ (vii)

The lowest α to satisfy (vii) is $\alpha \doteq 2.0816$ (viii)

From (143) we have

$$\Delta E \leq \frac{\hbar^2 k^2}{2m} \frac{\langle \sin^2 k r_1 \rangle}{\langle \cos^2 k r_1 \rangle}$$ (ix)

and

$$\langle \sin^2 k r_1 \rangle = 1 - \langle \cos^2 k r_1 \rangle$$

$$= 3 \int_0^1 dx\, x^2 \sin^2 \alpha x$$

$$= \frac{1}{2} - \frac{3}{2} \int_0^1 dx\, x^2 \cos 2\alpha x$$

$$= \frac{1}{2} - \frac{3}{4} \frac{d}{d\alpha} f_1(2\alpha)$$

$$= \frac{1}{2} - \frac{3}{2} \left[f_0(2\alpha) - \frac{f_1(2\alpha)}{\alpha} \right]$$ (x)

So as α becomes large (i.e. $k \gg 1/R$) then $\langle \sin^2 k r_1 \rangle \approx$
$\langle \cos^2 k r_1 \rangle \approx 1/2$. Hence for large k,

$$\Delta E \leq \frac{\hbar^2 k^2}{2m}$$ (xi)

as for (1). For $\alpha = 2.0816$, $f_0(2\alpha) \doteq -0.2049$ and
$f_1(2\alpha) \doteq 0.0762$,giving

$$\Delta E \underset{(\alpha = 2.0816)}{\leq} 13.56 \frac{\hbar^2}{mR^2} , \qquad = 390 A^{-2/3} \text{Mev for spherical nuclei.}$$ (xii)

(3) $\Psi = \sum_j^A (r_j^2 - \overline{r^2})\, \Phi$:

This wavefunction is just one orthogonal state (i.e. $\langle r_1^2 \rangle = \overline{r^2}$)

From (143)

$$\Delta E \leq \frac{\hbar^2}{2m} \frac{4\overline{r^2}}{\langle (r_1^2 - \overline{r^2})^2 \rangle} = \frac{2\hbar^2}{m} \frac{\overline{r^2}}{(\overline{r^4} - (\overline{r^2})^2)}$$ (xiii)

Using (129), $\overline{r^4} = \dfrac{25}{21} (\overline{r^2})^2$, hence

$$\Delta E \leq \frac{21}{2} \frac{\hbar^2}{m \bar{r}^2} \qquad = 17.5 \frac{\hbar^2}{mR^2} \tag{xiv}$$

$$= 500 A^{-2/3} \text{Mev for spherical nuclei} \tag{xv}$$

(4) $\Psi = \sum_{j}^{A} (r_j - \bar{r}) \Phi$:

This wavefunction is also just one orthogonal state

(i.e. $\langle r_i \rangle = \bar{r}$)   . From (143)

$$\Delta E \leq \frac{\hbar^2}{2m} \frac{1}{\langle (r_i - \bar{r})^2 \rangle} \qquad = \frac{\hbar^2}{2m(\bar{r^2} - (\bar{r})^2)} \tag{xvi}$$

That is,

$$\Delta E \leq \frac{40}{3} \frac{\hbar^2}{mR^2} , \quad = 384 A^{-2/3} \text{Mev for spherical nuclei} \tag{xvii}$$

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IV   SUM RULES :

If the sum of all the transition probabilities multiplied by
some power of each transition energy can be calculated for transitions
from a given level, then it is called a sum rule. Since each term
in the sum is positive definite, a sum rule gives an upper bound for
the value of the transition probability times the transition energy
to the particular power, for every transition. Usually, sum rules
are restricted to transitions of a given multipolarity. The most
important class of sum rules are those for which the transition energy
factor in each term is to the power of one only. These sum rules are
sometimes called oscillator sums (see [1] pp 399-404). All the sum
rules considered in this section will be oscillator sums.

In reference [2] (pp 709-15), it is shown that the sum of all the
transition probabilities times the transition energy, for the
excitation of a nucleus from a given state $|i\rangle$ to all higher excited
levels $|f\rangle$, by the absorption of photons of multipolarity j, can
be written as

$$S_j^{(1)} \equiv \frac{8\pi^3}{\hbar c} \frac{(j+1)}{j[(2j+1)!!]^2} \frac{1}{(2J_i+1)} \sum_{M_i,f,m} (E_f - E_i)\left|\langle f|Q_{jm}|i\rangle\right|^2 \qquad (1)$$
$$(j\geq 1)$$

From now on the transitions are assumed to be of electric multipolarity
and I will only be using (1) when $|i\rangle$ is the ground state of an
even-even nucleus.   (Note, j = 0 is a special case because EO radiative
transitions are forbidden. The oscillator sum for j = 0 is discussed
separately, later on.) For $j \geq 2$ the electric multipole moments
can be approximated by (cf. AII (73))

$$Q_{jm} = e \sum_{\rho=1}^{Z} r_\rho^j Y_{jm}^*(\Sigma_\rho) \qquad (j\geq 2) \qquad (2)$$

The exact expressions must be translationally invariant, just as the true wavefunction for an internal excitation of a self-bound nucleus is translationally invariant (see the discussions near the beginning of sections AII and AIII). In particular, the centre of mass cannot be fixed at the origin because during a transition the nucleus must recoil with a momentum equal and opposite to that of the radiated photon. However, it can be shown that the recoil corrections for all multipole moments other than j = 1 (the electric dipole operator), are of order $1/A$ only (see [2] p 709, and [3]). For the dipole case we must use the translationally invariant form of the dipole moment : (cf. [4] eqn (1))

$$Q_{lm} = e \sum_{p=1}^{Z} \left[r_p Y_{lm}(r_p) - R Y_{lm}(R) \right] \tag{3}$$

$$= e \left\{ \frac{N}{A} \sum_{p=1}^{Z} r_p Y_{lm}(r_p) - \frac{Z}{A} \sum_{n=1}^{N} r_n Y_{lm}(r_n) \right\}$$

(R is the centre of mass coordinate and r_p and r_n are proton and neutron coordinates.)

The sum rules $S_j^{(l)}$ are evaluated by using the property of closure (see [2] p 711), i.e.

$$\sum_{f,m} (E_f - E_i) \left| \langle f | Q_{jm} | i \rangle \right|^2 = \frac{1}{2} \sum_m \langle i | \left[Q_{jm}^*, [H, Q_{jm}] \right] | i \rangle \tag{4}$$

where H is the Hamiltonian. When the potential energy commutes with Q_{jm}, the only contribution to (4) and hence $S_j^{(l)}$, comes from the kinetic energy. Using equations (1-4) the result for the kinetic energy term can be shown to be (see p 713 of [2]),

$$S_j^{(l)} = \pi^2 \frac{e^2}{\hbar c} \frac{(j+1)}{[(2j-1)!!]^2} \frac{\hbar^2}{m} Z \langle i | r_p^{2j-2} | i \rangle \left[1 + V_j \right] \tag{5a}$$
$$(j \geq 2)$$

$$S_1^{(l)} = 2\pi^2 \frac{e^2}{\hbar c} \frac{NZ}{A} \frac{\hbar^2}{m} \left(1 + V_1 \right) \tag{5b}$$

In these expressions the contribution from the potential energy has been written as a correction term V_j . It is very difficult to obtain an accurate estimate for V_j , but for the dipole case rough estimates and experimental results suggest that V_1 is about unity (see [2] pp 713-4, 736). It should also be pointed out that the above sum rules have treated the nucleons as elementary particles and so they exclude any contributions from meson production excitations.

I will now consider the contributions of particular excitations of even-even nuclei to the kinetic energy part of the $S_j^{(1)}$ sum rule. That is, the potential energy is assumed to commute with Q_{jm} , which implies the neglect of charge exchange and velocity dependent inter-actions, as was done in sections AII and AIII (see also [1] p 403 and [2] p 712). In this approximation $S_j^{(1)}$ is said to be a classical sum rule, i.e. $S_j^{(1)}$(class). Taking $|i\rangle$ to be the ground state we have

$$\langle i | \Gamma_\rho^{2j-2} | i \rangle = \langle \Gamma_\rho^{2j-2} \rangle \tag{6}$$

where $\langle \; \rangle$ is the ground state expectation value defined by AIII (4). So, when $|i\rangle$ is the ground state,

$$S_j^{(1)}(\text{class}) = \pi^2 \frac{Ze^2}{\hbar c} \frac{(j+1)}{[(2j-1)!!]^2} \frac{\hbar^2}{m} \langle \Gamma_\rho^{2j-2} \rangle \tag{7a}$$
$$(j \geq 2)$$

$$S_1^{(1)}(\text{class}) = 2\pi^2 \frac{e^2}{\hbar c} \frac{NZ}{A} \frac{\hbar^2}{m} \tag{7b}$$

Consider the contribution to $S_j^{(1)}$(class) from a single excited state with angular momentum quantum numbers (j,m), of the form

$$|f\rangle = F_{jm} |i\rangle \qquad (j \geq 1) \tag{8}$$

where $|i\rangle$ is the ground state of an even-even nucleus (i.e. $|i\rangle$ is a 0^+ state). From AII (4) we have, (neglecting the difference between

proton and neutron masses)

$$E_f - E_o = \frac{\hbar^2}{2m} \sum_{k=1}^{A} \frac{\langle |\nabla_k F_{jm}|^2 \rangle}{\langle |F_{jm}|^2 \rangle} \tag{9}$$

and (cf. AII (77))

$$|\langle f | Q_{jm} | i \rangle|^2 = \frac{\langle |F_{jm} Q_{jm}| \rangle^2}{\langle |F_{jm}|^2 \rangle} \tag{10}$$

Inserting (9) and (10) into (1), we see that the state (8) contributes to $S_j^{(1)}$(class), the amount

$$\frac{4\pi^3 (j+1)}{j[(2j+1)!!]^2} \frac{\hbar}{mc} \cdot \frac{\langle |F_{jm} Q_{jm}| \rangle^2}{\langle |F_{jm}|^2 \rangle^2} \cdot \sum_{k=1}^{A} \langle |\nabla_k F_{jm}|^2 \rangle \tag{11}$$

I will now discuss two examples of the type (8), for reference in section AII.

(i) j = 1 :

The particular case of j = 1 is important because the $S_1^{(1)}$ sum rule happens to be the total absorption cross section for dipole transitions (see [2] p 710), which is a direct experimental quantity. By far the largest contribution to the nuclear $S_1^{(1)}$ sum rule comes from the giant dipole resonance, which is known experimentally to exhaust about $S_1^{(1)}(class) \big/ S_1^{(1)}$ of the total sum rule (see [2] p 736). In reference [4] and part (b) of section AII, the giant dipole resonance state is investigated by using the isovector wavefunctions

$$\Psi_{1m} = F_{1m} = e^{-1} \alpha_{1m}^{-1} Q_{1m} \Phi \qquad (Q_{1m} \text{ defined by (3))} \tag{12}$$

where Φ is the ground state of an even-even nucleus and α_{1m} is the normalization factor for Y_{1m} (i.e. $\alpha_{11} = \alpha_{1-1} = \sqrt{\frac{3}{8\pi}}$, $\alpha_{10} = \sqrt{\frac{3}{4\pi}}$). For spherical nuclei (i.e. Φ spherically

symmetric) the three states are degenerate, but this degeneracy is broken in deformed nuclei (see AII part (b)). Inserting (12) into (11) we see that each m state, (m = -1,0,1), contributes to the $S_1^{(1)}$(class) sum rule, the amount

$$\frac{8}{9}\pi^2\frac{\hbar}{mc}\sum_{k=1}^{A}\langle|\nabla_k Q_{1m}|^2\rangle = \frac{2}{3}\pi^2\frac{e^2}{\hbar c}\frac{NZ}{A}\frac{\hbar^2}{m} \tag{13}$$

Comparing (13) with (7b) I note that the three giant dipole states Ψ_{1m} , together exhaust the entire $S_1^{(1)}$(class) sum rule. This proof could be regarded as unnecessary, since from (12) we see that Ψ_{1m} has complete overlap with the dipole operator Q_{1m} , and so it must exhaust the total m component of the $S_1^{(1)}$ sum rule (see [2] p 736). Generalizing this idea it follows that one way to derive (7) would be to consider the wavefunction $|f\rangle = Q_{jm}|i\rangle$, which must exhaust the entire **mth** component of the $S_j^{(1)}$ sum rule, because $|f\rangle$ has total overlap with the multiple moment Q_{jm} . The result (13) is a direct proof of this for the particular case when the potential energy is assumed to commute with Q_{1m} .

(ii) j = 2 :

In reference [5] and part (a) of section AII, a particular isoscalar 2^+ state of even-even nuclei was proposed :

$$\Psi_{2m} = F_{2m}\Phi = \alpha_{2m}^{-1}\sum_{i=1}^{A} r_i^2 Y_{2m}(\Gamma_i)\Phi \tag{14}$$

where α_{2m} is the normalization factor for Y_{2m} (see AII (74)). From (2) and (14), and ignoring any differences between protons and neutrons (the isoscalar wavefunction (14) has already assumed this since it treats protons and neutrons identically), we have

$$\langle|F_{2m}Q_{2m}|\rangle = e\alpha_{2m}\frac{Z}{A}\langle|F_{2m}|^2\rangle \tag{15}$$

Inserting (14) and (15) into (11) we see that each m state

(m = -2,-1,0,1,2) contributes to the $S_2^{(1)}$(class) sum rule, the

amount

$$\frac{2\pi^3}{75} \alpha_{2m}^2 \frac{e^2}{\hbar c} \frac{Z^2}{A} \frac{\hbar^2}{m} \left\langle |\nabla_1 F_{2m}|^2 \right\rangle \tag{16}$$

When Φ is spherically symmetric (i.e. $\left\langle x_1^2 \right\rangle = \left\langle y_1^2 \right\rangle = \left\langle z_1^2 \right\rangle$, etc.),

(16) reduces to

$$\frac{\pi^2}{15} \frac{e^2}{\hbar c} \frac{Z^2}{A} \frac{\hbar^2}{m} \left\langle r_1^2 \right\rangle \tag{17}$$

For spherical nuclei all five states are degenerate and from (7a) we

see that together they exhaust the factor Z/A of the $S_2^{(1)}$(class)

sum rule.

In reference [1] p 404, the oscillator sum rule governing EO

transitions to the ground state is defined as

$$S(EO) \equiv \sum_f (E_f - E_0) \left| \left\langle f \left| e \sum_{p=1}^{Z} r_p^2 \right| 0 \right\rangle \right|^2 . \tag{18}$$

Zero-zero radiative transitions are forbidden, but EO transitions can

be caused by Coulomb interaction between nuclear protons and atomic

shell electrons penetrating inside the nucleus. The probability of

such a transition from the ground state is proportional to

$$\left| \left\langle f \left| \sum_p r_p^2 \right| 0 \right\rangle \right|^2 \qquad \text{, (see [6]). We can use the}$$

translationally variant monopole operator because it is known that

recoil corrections for EO transitions are of order $1/A$ only (see [3]

p 486). It is shown in [1] p 404, that when velocity dependent and

charge exchange interactions are neglected, that the S(EO) sum rule

is given by

$$S(EO)_{class} = 2 Z e^2 \frac{\hbar^2}{m} \left\langle r_p^2 \right\rangle \tag{19a}$$

This result is easily proved by evaluating the term in (18) coming from the hypothetical wavefunction $|f\rangle = \sum_p r_p^2 |0\rangle$, which has total overlap with the monopole operator and hence must exhaust the entire S(EO) sum rule. In the cutoff density approximation (see AII (44-5)), (19a) becomes

$$S(EO)_{Class} = \frac{6}{5} Z e^2 \frac{\hbar^2}{m} . R^2 \qquad (19b)$$

(R is the radius of the ground state nucleus which is assumed to be spherical.) Consider the trial wavefunctions for isoscalar vibrational states of even-even nuclei, discussed in section AIII. They are all of the form

$$\Psi = F \Phi \qquad \text{(F is always real)} \qquad (20)$$

where Φ is the ground state of a non deformed even-even nucleus. Since the wavefunctions are isoscalar we can ignore the differences between protons and neutrons and so the contribution of Ψ to the $S(EO)_{class}$ sum rule is (cf. (10))

$$Z e^2 \Delta E_o \frac{\langle r_1^2 F \rangle^2}{\langle F^2 \rangle} \qquad (21)$$

where ΔE_o is given by AIII (3), i.e.

$$\Delta E_o = \frac{A \hbar^2}{2m} \frac{\langle (\nabla_1 F)^2 \rangle}{\langle F^2 \rangle} \qquad (22)$$

In appendices AIV (1) and AIV (2), the weak correlation approximation is used to calculate the value of (21) for the lowest states of all of the vibrational wavefunctions summarized in table II of section AIII (i.e. p AIII. 42.). The results are given in table I on the next page. I have also included the values of ΔE_o for A = 208 from table II of section AIII, together with a normalized ratio of (21) over ΔE_o for A = 208, which is proportional to the transition rate.

From table I we see that in the weak correlation approximation

TABLE I :   Properties of Vibrational Wavefunctions, Calculated in
the Weak Correlation Approximation.

Trial Wavefunction $\left(\alpha = kR ,\ R = 1\cdot2\,A^{1/3}\,fm\right)$	Column two Contributions to the $S(EO)_{class}$ sum rule, In units of $S(EO)_{class}$	Column three ΔE_o for A=208 (MeV)	$\left\{\begin{array}{l}\text{Column two}\\ \text{divided by}\\ \text{Column three}\end{array}\right\}\times 14\cdot71.$
$\sum\limits_{j}^{A} j_o(k r_j)\,\Phi$ $\left(j_1(\alpha)=0 \right)$	$\dfrac{10}{\alpha^2}\dfrac{Z}{A}\left\{\begin{array}{l}=0\cdot495\,\dfrac{Z}{A}\\ (\alpha=4\cdot4934)\end{array}\right.$	$8\cdot3$ $(\alpha=4\cdot4934)$	$0\cdot88\ \dfrac{Z}{A}$ $(\alpha=4\cdot4934)$
$\sum\limits_{j}^{A}\cos(k r_j)\,\Phi$ $(\alpha=2\cdot0816)$	$0\cdot766\,\dfrac{Z}{A}$	$11\cdot1$	$1\cdot015\,\dfrac{Z}{A}$
$\sum\limits_{j}^{A}(r_j^2-\bar{r^2})\,\Phi$	$\dfrac{Z}{A}$	$14\cdot3$	$1\cdot03\,\dfrac{Z}{A}$
$\sum\limits_{j}^{A}(r_j-\bar{r})\,\Phi$	$0\cdot741\,\dfrac{Z}{A}$	$10\cdot9$	$1\cdot00\,\dfrac{Z}{A}$
Translationally Invariant Wavefunctions :			
$\sum\limits_{i<j}^{A}\sum j_o(k r_{ij})\,\Phi$	0	very large	0
$\sum\limits_{i<j}^{A}\sum\cos k r_{ij}\cdot\Phi$ $\begin{array}{l}j_1(\alpha)=0\\ A\gtrsim\dfrac{\alpha^4}{8}\end{array}$	$\dfrac{10}{\alpha^2}\dfrac{Z}{A}\left\{\begin{array}{l}=0\cdot495\,\dfrac{Z}{A}\\ (\alpha=4\cdot4934,A\gtrsim50)\end{array}\right.$	$8\cdot3$ $(\alpha=4\cdot4934,A\gtrsim50)$	$0\cdot88\,\dfrac{Z}{A}$ $(\alpha=4\cdot4934,A\gtrsim50)$
$\begin{array}{l}j_o(\alpha)=\dfrac{5}{2\alpha}j_1(\alpha)\\ \alpha=1\cdot5255\\ A\gtrsim50\end{array}$	$0\cdot845\,\dfrac{Z}{A}$	$12\cdot3$	$1\cdot01\,\dfrac{Z}{A}$
$\sum\limits_{i<j}^{A}\sum(r_{ij}^2-\langle r_{12}^2\rangle)\,\Phi$ $(A\gtrsim50)$	$\dfrac{Z}{A}$	$14\cdot3$	$1\cdot03\,\dfrac{Z}{A}$
$\sum\limits_{i<j}^{A}\sum(r_{ij}-\langle r_{12}\rangle)\,\Phi$ $(A\gtrsim50)$	$0\cdot922\,\dfrac{Z}{A}$	$13\cdot3$	$1\cdot02\,\dfrac{Z}{A}$

that the nine trial wavefunctions separate into three different states.
The $\sum \sum J_o(k\, r_{ij})\, \Phi$ is a strange state with very high energy
and zero transition rate. Of the rest of the wavefunctions two appear
to describe a different excitation from the rest. Namely, $\sum J_o(k\, r_j)\, \Phi$
and the $J_1(\alpha) = 0$ series of $\sum \sum \cos k\, r_{ij}\, \Phi$ are very closely
related and have a smaller contribution to the sum rule than the other
states. The rest of the trial wavefunctions all have an energy within
the range $12 \cdot 6 \pm 1 \cdot 7$ MeV for A = 208, and the transition rate matrix
elements are remarkably insensitive to the differences in the wave-
functions.

However, in section AII I discovered that it is quite likely that
the results for the energy (and the transition rate matrix elements)
are qualitatively incorrect. That is, the weak correlation approximation
leads to $\Delta E_o \sim A^{-2/3}$, but the inclusion of short ranged correlations
and the requirement of number conservation may cancel the weak
correlation terms, leaving $\Delta E_o \sim A^{-1/3}$. This surprising
complication was discovered after writing section AIII, in the case
of the 2^+ and 1^- states discussed in section AII. The results for the
contributions to the $S(EO)_{class}$ sum rule should not be unduly
influenced by the weak correlation approximation because the
qualitative errors occur only in the $\langle r_1^2 F\rangle$ and $\langle F^2\rangle$ terms, so
that the errors will tend to cancel in the expression (21). In fact
for the particular state

$$F\, \Phi = \sum_j (r_j^2 - \overline{r^2})\, \Phi$$

there is no error at all, since the orthogonality condition

$$\langle F\rangle = 0 \quad \text{, implies}$$

$$\langle r_1^2 F\rangle = \tfrac{1}{A}\langle F^2\rangle$$

and so from (21) and (22) we see that the contribution to the
$S(EO)_{class}$ sum rule is exactly,

$$\frac{Z}{A} e^2 \frac{\hbar^2}{2m} \left\langle (\nabla_1 F)^2 \right\rangle = 2 \frac{Z}{A} e^2 \frac{\hbar^2}{m} \left\langle r_1^2 \right\rangle = \frac{Z}{A} S(E0)_{class} \quad (23)$$

I will now summarise the available evidence that indicates the physical nature of the vibrational states in table I. (I ignore the strange $\sum \sum j_o(k r_{ij}) \Phi$ wavefunction.) Firstly, in section BII it is argued that the translationally variant wavefunction

$$\sum_j f(r_j) \Phi$$

corresponds to a classical flow field given by a velocity potential f(r) (i.e. $\underline{v}(r) \sim \underline{\nabla} f(r)$). From this it follows that the lowest states of each translationally variant wavefunction should approximate a simple in and out vibration, i.e. a breathing mode. In particular, the $\sum (r_j^2 - \overline{r^2}) \Phi$ state should be a good approximation. From (23) it appears that $\sum (r_j^2 - \overline{r^2}) \Phi$ describes a giant monopole resonance, since it exhausts such a large part of the S(E0) sum rule, in direct analogy with the resonance states Ψ_{2M} and $\underline{\Psi}_{IM}$ of section AII. Using this together with the argument of section BII it follows that the giant monopole resonance and the breathing mode state are one in the same, which seems perfectly reasonable. In section AIII I showed that $\sum \sum (r_{ij}^2 - \langle r_{12}^2 \rangle) \Phi$ is the translationally invariant form of $\sum (r_j^2 - \overline{r^2}) \Phi$, (where the centre of mass is fixed at the origin), so the above results also apply to the translationally invariant wavefunction $\sum \sum (r_{ij}^2 - \langle r_{12}^2 \rangle) \Phi$. From appendix 6A-3 of reference [1] and using the argument from section BII, we see that the wavefunction $\sum j_o(k r_j) \Phi$ should describe the vibrational modes of a liquid drop. Thus the lowest state of $\sum j_o(k r_j) \Phi$ should also be the breathing mode. However, the orthogonality condition $j_1(\alpha) = 0$ is different from the liquid drop model boundary condition for a free surface oscillation,

namely $j_0(\alpha) = 0$. It is thus possible that the differences between the $\sum j_0(kr_j) \Phi$ and $\sum (r_j^2 - \overline{r^2}) \Phi$ wavefunctions in table I are physically correct. For example, the lowest state of $\sum j_0(kr_j) \Phi$ may correspond to a vibration with a zero velocity at the surface, whereas the breathing mode has maximum velocity at the surface. This interpretation could explain the physical difference between the lowest states of the two separate series belonging to the translationally invariant wavefunction $\sum \sum \cos kr_{ij} \Phi$: the $j_1(\alpha) = 0$ series corresponds to the $\sum j_0(kr_j) \Phi$ states (note from table I above and table II of section AIII, that for $A \gtrsim \frac{\alpha^4}{8}$, that the energies and the sum rule contributions, in the weak correlation approximation, are the same), while the lowest state of the $j_0(\alpha) = \frac{5}{2\alpha} j_1(\alpha)$ series approximates the giant monopole resonance. I note from reference [1] p 668, that the lowest vibrational state in the liquid drop model has an energy $\Delta E_0 \simeq 65 A^{-1/3} MeV$. So if $\sum (r_j^2 - \overline{r^2}) \Phi$ (and its translationally invariant form) is to represent the giant monopole resonance it follows that the weak correlational approximation result $\Delta E_0 \simeq 500 A^{-2/3} MeV$ is spurious. This possibility is strongly supported by the result 19c of section AII, which is very nearly satisfied in the cutoff density approximation and could easily be exactly satisfied for the true density profile (note, because of AII (19c) $\Delta E_2 \sim A^{-1/3}$ and not $A^{-2/3}$.) Since from equation (xiii) of appendix AIII (3), we see that if the result AII (19c) is substituted directly into the weak correlation expression for the vibrational energy of $\sum (r_j^2 - \overline{r^2}) \Phi$, it gives $\Delta E_0 = \infty$. Thus it appears that once again the weak correlation terms in the energy denominator are cancelled by contributions from the short ranged correlations, so that the $A^{-2/3}$ variation becomes an $A^{-1/3}$ variation. I hope to be able to prove the above conjecture in the not too distant future.

APPENDIX AIV(1) : Contributions to the S(EO)$_{class}$ Sum Rule from
Translationally *Variant* States.

We need to evaluate $\langle r_1^2 F \rangle$, where F is of the form

$$F = \sum_{j=1}^{A} f(r_j) \qquad\qquad (i)$$

From (i) we have

$$\langle r_1^2 F \rangle = \langle r_1^2 f(r_1) \rangle + (A-1)\langle r_1^2 f(r_2) \rangle$$

In the weak correlation approximation the last term becomes

$$\langle r_1^2 f(r_2) \rangle = \langle r_1^2 \rangle \langle f(r_2) \rangle \qquad\qquad ,$$

where I have used the orthogonality condition

$$\langle f(r) \rangle = 0 \qquad\qquad (ii)$$

Defining $x = r_1/R$, (R the radius of the spherical ground state),
and using the cutoff density approximation (see AIII (144)), the
result for $\langle r_1^2 F \rangle$ in the weak correlation limit can be written
as (cf. AIII (145))

$$\langle r_1^2 F \rangle = 3R^2 \int_0^1 dx\, x^4 f(Rx) \qquad\qquad (iii)$$

---

(1) $F = \sum_j^A j_0(k r_j)$: $\qquad\qquad (iv)$

In appendix AIII (3) the orthogonality condition (ii) is shown
to imply

$$j_1(\alpha) = 0 \qquad\qquad (\text{i.e. } \alpha \doteq 4.4934, \cdots)$$

where $\alpha \equiv kR$. Using (iii) we have

$$\langle r_1^2 F \rangle = \frac{3}{\alpha} R^2 \int_0^1 dx\, x^3 \sin\alpha x$$

$$= \frac{3}{\alpha} R^2 \frac{d^3}{d\alpha^3} \int_0^1 dx \cos\alpha x$$

$$= \frac{3}{\alpha} R^2 \frac{d^3}{d\alpha^3} j_0(\alpha)$$

$$= \frac{6}{\alpha^2} R^2 j_0(\alpha) \qquad\qquad , \ j_1(\alpha) = 0 \qquad \text{(v)}$$

From AIII (143) and appendix AIII(2) (i) we have, in the weak correlation approximation,

$$\langle F^2 \rangle = A \langle j_0^2(k\, r_i) \rangle$$

$$= 3A \int_0^1 dx\, x^2 j_0^2(\alpha x)$$

$$= \frac{3A}{2\alpha^2} (1 - j_0(2\alpha)) \qquad\qquad \text{(vi)}$$

Using (v), (vi) and the results AIII (77) for $j_1(\alpha) = 0$, I find

$$\frac{\langle r_i^2 F \rangle^2}{\langle F^2 \rangle} = \frac{24}{A} \frac{R^4}{\alpha^4} \qquad\qquad , \ j_1(\alpha) = 0$$

Then using the result calculated in appendix AIII(3) for ΔE_0 , it follows from (19b) and (21), that the contribution of (iv) to the $S(EO)_{class}$ sum rule is

$$12 \frac{Z}{A} e^2 \frac{\hbar^2}{m} \frac{R^2}{\alpha^2} = \left[\frac{10}{\alpha^2} \frac{Z}{A} \right] \cdot S(EO)_{class} \qquad , \ j_1(\alpha) = 0 \qquad \text{(vii)}$$

(2) $F = \sum_j^A \cos k\, r_j :$ (viii)

The orthogonality condition is (see app AIII(3))

$$\frac{d}{d\alpha} j_1(\alpha) = 0 \qquad , \text{i.e.} \ j_0(\alpha) = \frac{2}{\alpha} j_1(\alpha) \qquad , (\alpha \doteq 2.0816, \cdots)$$

Using (iii) we have

$$\langle r_i^2 F \rangle = 3R^2 \int_0^1 dx\, x^4 \cos\alpha x$$

$$= 3R^2 \frac{d^4}{d\alpha^4} \int_0^1 dx \cos\alpha x$$

$$= 3R^2 \frac{d^4}{d\alpha^4} j_0(\alpha)$$

$$= 6R^2 \frac{j_1(\alpha)}{\alpha^3}(4-\alpha^2) \qquad\qquad , \frac{d}{d\alpha} j_1(\alpha) = 0$$

$$\doteqdot -0.0966 \, R^2 \qquad\qquad , \alpha = 2.0816$$

$$\text{(ix)}$$

From appendix AIII(3) we have, in the weak correlation approximation,

$$\langle F^2 \rangle = A \langle \cos^2 k r_1 \rangle$$

$$= 3A \int_0^1 dx \, x^2 \cos^2 \alpha x$$

$$= A\left\{ \frac{1}{2} + \frac{3}{2}\left(j_0(2\alpha) - \frac{j_1(2\alpha)}{\alpha}\right)\right\}$$

$$\doteqdot 0.1377 \, A \qquad\qquad , \alpha = 2.0816 \qquad \text{(x)}$$

Using (ix) and (x), for the lowest state, I find

$$\frac{\langle r_1^2 F \rangle^2}{\langle F^2 \rangle} \doteqdot 0.0678 \frac{R^4}{A} \qquad\qquad , \alpha = 2.0816$$

Then using the result for ΔE_0 calculated in appendix AIII(3), it

follows from (19b) and (21), that the contribution of the lowest state

of (viii) to the $S(EO)_{class}$ sum rule is

$$0.9194 \, \frac{Z}{A} e^2 \frac{\hbar^2}{m} R^2 = 0.766 \frac{Z}{A} S(EO)_{class} \quad , \alpha = 2.0816$$

$$\text{(xi)}$$

$$(3) \quad F = \sum_j^A (r_j^2 - \overline{r^2}) : \qquad\qquad\qquad \text{(xii)}$$

Since $\langle F \rangle = 0$, it follows exactly that

$$\langle r_1^2 F \rangle = \frac{1}{A} \langle F^2 \rangle$$

So from (19a), (21) and (22) we have, without any approximations, that

the contribution from (xii) to the $S(EO)_{class}$ sum rule is (see also (23))

$$\frac{Z}{A} \, S(EO)_{class} \tag{xiii}$$

(4) $\quad F = \sum_{j}^{A} (\Gamma_j - \bar{F}) : \tag{xiv}$

In the weak correlation approximation we have

$$\langle \Gamma_1^2 F \rangle = \overline{\Gamma^3} - (\overline{\Gamma^2})\bar{F} = \frac{R^3}{20}$$

(using the cutoff density result $\overline{\Gamma^n} = \frac{3}{n+3} R^n$)

From appendix AIII(3), the weak correlation approximation also gives

$$\langle F^2 \rangle = A \langle (\Gamma_1 - \bar{F})^2 \rangle$$

$$= A \left[\overline{\Gamma^2} - (\bar{F})^2 \right]$$

$$= \frac{3}{80} A R^2$$

So

$$\frac{\langle \Gamma_1^2 F \rangle^2}{\langle F^2 \rangle} = \frac{R^4}{15A}$$

Using the result in appendix AIII(3) for ΔE_0 , it follows from (19b) and (21), that the contribution from (xiv) to the $S(EO)_{class}$ sum rule is

$$\frac{8}{9} \frac{Z}{A} e^2 \frac{\hbar^2}{m} R^2 = \left[\frac{20}{27} \frac{Z}{A} \right] S(EO)_{class} \tag{xv}$$

APPENDIX AIV(2) : Contributions to the $S(EO)_{class}$ Sum Rule from
Translationally Invariant States.

We need to evaluate $\langle r_1^2 F \rangle$ in the weak correlation

approximation, where F is of the form

$$F = \sum_{i<j}^{A}\sum^{A} f(r_{ij}^2)$$ (i)

That is, we wish to calculate expressions of the form

$$\langle r_1^2 F \rangle = (A-1)\langle r_1^2 f(r_{12})\rangle + \frac{(A-1)(A-2)}{2}\langle r_1^2 f(r_{23})\rangle$$

In the weak correlation approximation the last term is zero and so,

assuming a cutoff density, we obtain (see equations 49 and 51 of

section AIII)

$$\langle r_1^2 F \rangle = \frac{q}{2}(A-1)R^2 \int_0^1 dx\, x^3 \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, t\, f(Rt)$$ (ii)

(1) $$F = \sum_{i<j}^{A}\sum^{A} J_o(k r_{ij}) :$$ (iii)

From AIII (63), the orthogonality condition for a general radial

density is

$$\int_0^\infty dx\, x\, n(x)\sin\alpha x = 0 \qquad , \alpha = kR$$ (iv)

For a cutoff density (iv) reduces to

$$J_1(\alpha) = 0 \qquad , (\alpha = 4.4934,\dots)$$

Using (ii) we have

$$\langle r_1^2 F \rangle = \frac{q}{2}(A-1)R^2 \int_0^1 dx\, x^3 \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, \left(\frac{\sin\alpha t}{\alpha}\right)$$

$$= \frac{q}{\alpha^2}(A-1)R^2 \left[\int_0^1 dx\, x^3 \sin\alpha x\right]\left[\int_0^1 dy\, y\sin\alpha y\right]$$

$$= 0 \qquad , J_1(\alpha) = 0$$ (v)

Note from (iv), that (v) would still hold for a general radial

density $n(x)$. So in the weak correlation approximation the

contribution of (iii) to the S(EO)$_{\text{class}}$ sum rule is zero. In fact

from the general expression for $\langle \Gamma_i^2 F \rangle$ we see that the contribution

would still be zero if short ranged correlations between particles

2 & 3, but not 1 & 2 and 1 & 3, were included in the term $\langle \Gamma_i^2 f(\Gamma_{23}) \rangle$.

If all the correlations were included then the transition matrix

element for excitation to the state (iii) would probably no longer

be zero, but it should be at least an order of magnitude smaller

than for the rest of the states considered in this appendix.

(2) $$F = \sum_{i<j}^{A} \sum_{j}^{A} \cos k\Gamma_{ij} \; : \qquad\qquad\qquad\qquad\qquad\qquad \text{(vi)}$$

In section AIII it is shown, in the weak correlation approximation,
that this wavefunction describes two different sets of states, defined
by the orthogonality conditions

$$J_1(\alpha) = 0 \qquad\qquad\qquad (\alpha = 4.4934, \cdots)$$

$$J_0(\alpha) = \frac{5}{2\alpha} J_1(\alpha) \qquad\qquad (\alpha = 1.5255, \cdots)$$

Using (ii) we have

$$\langle \Gamma_i^2 F \rangle = \frac{9}{2}(A-1)R^2 \int_0^1 dx\, x^3 \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, t \cos\alpha t$$

$$= \frac{9}{2}(A-1)R^2 \frac{d}{d\alpha} \left\{ \int_0^1 dx\, x^3 \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt \sin\alpha t \right\}$$

$$= -9(A-1)R^2 \frac{d}{d\alpha} \left\{ \frac{J_1(\alpha)}{\alpha} \frac{d^2}{d\alpha^2} J_1(\alpha) \right\}$$

$$= 9(A-1)R^2 \left\{ 42 \frac{J_1^2(\alpha)}{\alpha^4} - 7 \frac{J_1^2(\alpha)}{\alpha^2} - 20 \frac{J_0(\alpha) J_1(\alpha)}{\alpha^3} \right.$$

$$\left. + 2 \frac{J_1(\alpha) J_0(\alpha)}{\alpha} + 2 \frac{J_0^2(\alpha)}{\alpha^2} \right\}$$

$$= \begin{cases} 18(A-1)R^2 \dfrac{J_0^2(\alpha)}{\alpha^2} & , \quad J_1(\alpha) = 0 \\[20pt] \dfrac{18}{25}(A-1)R^2 \dfrac{J_0^2(\alpha)}{\alpha^2}(9-4\alpha^2) & , \quad J_0(\alpha) = \dfrac{5}{2\alpha}J_1(\alpha) \end{cases}$$

In section AII it is shown, in the weak correlation approximation, that for $A \gtrsim 50$ (and $A \gtrsim \frac{\alpha^4}{8}$ for $J_1(\alpha) = 0$), that

$$\langle F^2 \rangle \doteq A(A-1)(A-2) \langle \cos k\,r_{12}\, \cos k\,r_{13} \rangle$$

$$\doteq A^3 \begin{cases} \dfrac{27}{2} J_0^2(\alpha) & , \quad J_1(\alpha) = 0 \\[14pt] 0.0248 & , \quad \alpha = 1.5255 \end{cases}$$

So

$$\frac{\langle r_1^2 F \rangle^2}{\langle F^2 \rangle} \simeq \begin{cases} \dfrac{24}{A}\dfrac{R^4}{\alpha^4} & , \quad J_1(\alpha) = 0 \quad , \quad A \gtrsim \dfrac{\alpha^4}{8} \\[18pt] 0.0676\dfrac{R^4}{A} & , \quad \alpha = 1.5255 \quad , \quad A \gtrsim 50 \end{cases}$$

Then using the results for ΔE_0, summarised in Table II of section AIII, it follows from (19b) and (21), that the contribution of (vi) to the $S(EO)_{class}$ sum rule is

$$\simeq \begin{cases} 12\dfrac{Z}{A}e^2\dfrac{\hbar^2}{m}\dfrac{R^2}{\alpha^2} = \left[\dfrac{10}{\alpha^2}\dfrac{Z}{A}\right]S(EO)_{class} & , \quad \begin{array}{l} J_1(\alpha) = 0 \\ A \gtrsim \dfrac{\alpha^4}{8} \end{array} \\[24pt] 1.014\dfrac{Z}{A}e^2\dfrac{\hbar^2}{m}R^2 = 0.845\dfrac{Z}{A}S(EO)_{class} & , \quad \begin{array}{l} \alpha = 1.5255 \\ A \gtrsim 50 \end{array} \end{cases}$$

$$\text{(vii)}$$

(3) $$F = \sum_{i<j}^{A}\sum^{A} \left(\Gamma_{ij}^{2} - \langle \Gamma_{12}^{2} \rangle \right) :$$ (viii)

In the weak correlation approximation we have

$$\langle \Gamma_{1}^{2} F \rangle = (A-1) \left\langle \Gamma_{1}^{2} \left(\Gamma_{12}^{2} - \langle \Gamma_{12}^{2} \rangle \right) \right\rangle$$

$$= (A-1) \left\{ \left\langle \Gamma_{1}^{2} \left(\Gamma_{1}^{2} + \Gamma_{2}^{2} - 2 \, \underline{\Gamma}_{1}.\underline{\Gamma}_{2} \right) \right\rangle - \overline{\Gamma^{2}} \langle \Gamma_{12}^{2} \rangle \right\}$$

$$= (A-1) \left[\overline{\Gamma^{4}} - \left(\overline{\Gamma^{2}} \right)^{2} \right]$$ (ix)

where I have used the weak correlation limit results;

$$\langle \Gamma_{12}^{2} \rangle = 2 \, \overline{\Gamma^{2}} \quad , \quad \langle \Gamma_{1}^{2} \left(\underline{\Gamma}_{1}.\underline{\Gamma}_{2} \right) \rangle = 0$$

It is easy to verify (ix) for a cutoff density by using (ii). From equation 130 of section AIII and neglecting terms of order $\frac{1}{A}$ smaller, we have

$$\langle \Gamma_{1}^{2} F \rangle = \frac{1}{A^{2}} \langle F^{2} \rangle$$

So from (21) and (22), it follows that the contribution of (viii) to the $S(EO)_{class}$ sum rule is

$$\frac{1}{2} \frac{Z}{A^{3}} e^{2} \frac{\hbar^{2}}{m} \left\langle \left(\nabla_{1} F \right)^{2} \right\rangle$$ (x)

From (19a) and equation (127) of section AIII, we see that (x) reduces to

$$\left[\frac{Z}{A} + O\left(\frac{1}{A} \right) \right] . \; S(EO)_{class} \qquad , \; A \gtrsim 50$$ (xi)

(4) $$F = \sum_{i<j}^{A}\sum^{A} \left(\Gamma_{ij} - \langle \Gamma_{12} \rangle \right) :$$ (xii)

In the weak correlation approximation we have

$$\langle \Gamma_{1}^{2} F \rangle = (A-1) \left\langle \Gamma_{1}^{2} \left(\Gamma_{12} - \langle \Gamma_{12} \rangle \right) \right\rangle$$

$$= (A-1) \left\{ \langle \Gamma_{1}^{2} \Gamma_{12} \rangle - \overline{\Gamma^{2}} \langle \Gamma_{12} \rangle \right\}$$ (xiii)

Using (ii) I find

$$\langle \Gamma_1^2 \Gamma_{12} \rangle = \frac{9}{2} R^3 \int_0^1 dx\, x^3 \int_0^1 dy\, y \int_{|x-y|}^{x+y} dt\, t^2$$

$$= \frac{9}{2} R^3 \int_0^1 dx\, x^3 \left(\frac{x}{2} + \frac{x^3}{3} - \frac{x^5}{30} \right) \qquad \text{, from AIII (135c)}$$

$$= \frac{68}{105} R^3 \qquad\qquad\qquad \text{(xiv)}$$

Substituting (xiv) into (xiii) and using the cutoff density result

for $\qquad \overline{\Gamma^2} \langle \Gamma_{12} \rangle \qquad\qquad$ (see AIII (138)), we obtain

$$\langle \Gamma_1^2 F \rangle = \frac{16}{525} (A-1) R^3$$

From section AIII we have, in the weak correlation approximation,

$$\langle F^2 \rangle = A^3 \left\{ \langle \Gamma_{12} \Gamma_{13} \rangle - \langle \Gamma_{12} \rangle^2 \right\} + O\left(\frac{1}{A}\right)$$

$$\doteq 0.0136\, A^3 R^2$$

So

$$\frac{\langle \Gamma_1^2 F \rangle^2}{\langle F^2 \rangle} \doteq 0.0683 \frac{R^4}{A}$$

Using the result AIII (139) for ΔE_o , it follows from (19b) and

(21) that the contribution of (xii) to the $S(EO)_{class}$ sum rule is

approximately

$$1.106 \frac{Z}{A} e^2 \frac{\hbar^2}{m} R^2 = 0.922 \frac{Z}{A} S(EO)_{class} \quad , A \gtrsim 50 \quad \text{(xv)}$$

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PART   B

FLOW   IN   QUANTUM   MECHANICS

ABSTRACT

I consider the problem of how to describe flow in quantum fluids.
In particular, we want to be able to identify the physical motion
represented by any given many-body wavefunction. Section I derives
a quantum mechanical velocity field for a many-body system, paying
special attention to the need for a quantum continuity equation. It
is found that when the wavefunction has the usual time dependence
$e^{-i\omega t}$, that the quantum velocity formula averages over all
oscillatory motion, so that much of the physical nature of the flow
field is lost. In section II a particular wavefunction is proposed
to represent the quantum excitation corresponding to any given
potential flow field. The results obtained by considering specific
examples are very encouraging.

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CONTENTS

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I.   CURRENT AND VELOCITY FIELDS IN NON-RELATIVISTIC MANY-BODY

QUANTUM MECHANICS.

The non-relativistic current density for a single particle quantum system described by a wavefunction $\psi(\underline{r},t)$ is derived from the one-particle Schrödinger equation

$$H\psi = i\hbar \frac{\partial \psi}{\partial t} \quad , \quad H = -\frac{\hbar^2}{2m}\nabla^2 + V(\underline{r}) \tag{1}$$

and the continuity equation embodying the conservation law for the flow of the system

$$\frac{\partial |\psi|^2}{\partial t} + \underline{\nabla}\cdot\underline{J} = 0 \quad , \qquad \underline{J} \text{ is the current density.} \tag{2}$$

By noting that $|\psi|^2 d\underline{r}$ is the probability of finding the particle in the volume element $d\underline{r}$, we see that $|\psi|^2$ is a single particle number density, so that (2) is directly analogous to the classical continuity equation for a fluid,

$$\frac{\partial \rho}{\partial t} + \underline{\nabla}\cdot(\rho\underline{v}) = 0 \tag{3}$$

We can therefore define a single particle velocity field by

$$\underline{v} = \frac{\underline{J}}{|\psi|^2} \tag{4}$$

From (1) and its complex conjugate, (note that H is a real, linear operator)

$$\frac{\partial |\psi|^2}{\partial t} = \psi^* \frac{\partial \psi}{\partial t} + \psi \frac{\partial \psi^*}{\partial t}$$

$$= \frac{1}{i\hbar}\left(\psi^* H\psi - \psi H\psi^*\right)$$

$$= \frac{i\hbar}{2m}\left(\psi^* \nabla^2 \psi - \psi \nabla^2 \psi^*\right)$$

$$= \frac{i\hbar}{2m} \underline{\nabla}\cdot\left(\psi^* \underline{\nabla}\psi - \psi \underline{\nabla}\psi^*\right)$$

$$= -\frac{\hbar}{m} \underline{\nabla}\cdot I_m(\psi^* \underline{\nabla}\psi) \tag{5}$$

From (2) and (5) the single particle current density is

$$\underline{J} = \frac{\hbar}{m} \text{Im} (\psi^* \underline{\nabla} \psi) \tag{6}$$

and then from (4) the velocity field of the particle is given by

$$\underline{V}(\underline{r}) = \frac{\hbar}{m} \text{Im} \left(\frac{\underline{\nabla} \psi}{\psi} \right) \tag{7}$$

I will now extend the above theory to a many-body system, paying special attention to a suitable generalisation of the continuity equation (2).

Consider a non relativistic system of N distinguishable particles, with wavefunction $\Psi(\underline{r}_1, \ldots, \underline{r}_N, t)$, and Hamiltonian

$$H = -\frac{\hbar^2}{2} \sum_{j=1}^{N} \frac{1}{m_j} \nabla_j^2 + V(\underline{r}_1, \ldots, \underline{r}_N) \tag{8}$$

Then from Schrodinger's equation $H\Psi = i\hbar \frac{\partial \Psi}{\partial t}$, and following the same steps used to derive (5)

$$\frac{\partial |\Psi|^2}{\partial t} + \hbar \sum_{j=1}^{N} \frac{1}{m_j} \underline{\nabla}_j \cdot \text{Im}(\Psi^* \underline{\nabla}_j \Psi) = 0 \tag{9}$$

[Aside: If we defined a current density for particle j by

$$\underline{j}_j(\underline{r}_j, t) = \frac{\hbar \text{Im}}{m_j} (\Psi^* \underline{\nabla}_j \Psi) \quad , \text{ then (9) becomes}$$

$$\frac{\partial |\Psi|^2}{\partial t} + \sum_j \underline{\nabla}_j \cdot \underline{j}_j = 0 \quad , \text{ which is } \underline{\text{not}} \text{ of the}$$

form (2) with $\underline{J} = \sum_j \underline{j}_j$. Hence we reject this guess

for $\underline{j}_j$.]

We define the number density for particle one as, (compare with AI(2))

$$n_1(1) = \frac{\int d\underline{r}_2 \ldots d\underline{r}_N |\Psi|^2}{\int d\underline{r}_1 \ldots d\underline{r}_N |\Psi|^2} \tag{10}$$

Similarly and using (6), we define the current density for particle one as,

$$\underline{j}_1(1) = \frac{\hbar}{m_1} \frac{\int d\underline{r}_2 \ldots d\underline{r}_N \text{Im}(\Psi^* \underline{\nabla}_1 \Psi)}{\int d\underline{r}_1 \ldots d\underline{r}_N |\Psi|^2} \tag{11}$$

In appendix BI(1) I prove that particle one satisfies the conservation law

$$\frac{\partial n_1(1)}{\partial t} + \underline{\nabla}_1 \cdot \underline{j}_1(1) = 0 \tag{12}$$

Hence by analogy with (3) we find that the velocity field for particle one is given by

$$\underline{v}_1(1) = \underline{j}_1(1) \Big/ n_1(1) = \frac{\hbar}{m_1} \frac{\int d\underline{r}_2 \ldots d\underline{r}_N \, \mathrm{Im}(\Psi^* \underline{\nabla}_1 \Psi)}{\int d\underline{r}_2 \ldots d\underline{r}_N |\Psi|^2} \tag{13}$$

From now on we will restrict ourselves to a system of N indistinguishable particles each of mass m . Then the number density and probability current density for the entire system are,

$$n(\underline{r},t) = N \, n_1(\underline{r},t) \quad , \quad \underline{J}(\underline{r},t) = N \, \underline{j}_1(\underline{r},t) \tag{14}$$

and the conservation law for the flow within the system is given by the continuity equation

$$\frac{\partial n(\underline{r},t)}{\partial t} + \underline{\nabla} \cdot \underline{J}(\underline{r},t) = 0 \tag{15}$$

Therefore the velocity field is the same for each particle, namely,

$$\underline{v}(1) = \frac{\hbar}{m} \frac{\int d\underline{r}_2 \ldots d\underline{r}_N \, \mathrm{Im}(\Psi^* \underline{\nabla}_1 \Psi)}{\int d\underline{r}_2 \ldots d\underline{r}_N |\Psi|^2} \tag{16}$$

After writing this section it subsequently came to my notice that Feenberg has previously published the single particle conservation condition (12), using it to discuss phonons in liquid helium ([1] , Chapter 4). Several authors have previously stated the result (10), (11) and (14), without derivation or reference to a continuity equation. (See London page 147 [2] , and Feynman page 292 [3]). We should also note that other authors have used a slightly different approach by defining a many-body velocity field in operator language. (Landau [4] , but note that London [5] , believes Landau's velocity operator to be invalid, and Feynman page 294 [3]).

THE PHYSICAL NATURE OF $\underline{\mathcal{V}}(1)$:

From (16) we see that if Ψ is an energy eigenstate (i.e.

$$\Psi = \Psi_0 \, e^{-i\omega t}$$ where Ψ_0 is not a function of time), then

the velocity is independent of time and is zero if Ψ_0 is pure real

or imaginary. Hence it appears that $\underline{\mathcal{V}}(1)$ is a quantum mechanical average

velocity that averages out all motions that do not contribute to the

nett transport of matter. This conclusion is verified by the specific

examples considered below.

Let us consider a very simple wavefunction first put forward to

describe mass motion of superfluid helium, (see Feynman [6] page 269,

and [3] page 335.),

$$\Psi = \prod_{j=1}^{N} \exp\left(i \, S(\underline{r}_j)\right) \Phi \tag{17}$$

where Φ is the ground state wavefunction and $S(\underline{r})$ is some function of

position. Letting the ground state $\Phi = \Phi_0 \, e^{-i\omega t}$ we can always

take Φ_0 to be real because H is a real linear operator, So,

$$\text{Im}\left(\Psi^* \underline{\nabla}_i \Psi\right) = \text{Re}\left(\underline{\nabla}_i S(\underline{r}_i)\right). |\Psi|^2 \tag{18}$$

Therefore, from (18) and (16) we find that the velocity field for the

state (17) is

$$\underline{\mathcal{V}}(\underline{r}) = \frac{\hbar}{m} \, \text{Re}\left(\underline{\nabla} S(\underline{r})\right) \tag{19}$$

Equation (19) plays a central role in superfluid physics ([7]), but

because it was first obtained only heuristically by Feynman ([6] , page 269)

many people did not realise that it could be rigorously derived in the

manner above. We see from (19) that the wavefunction (17) describes only

irrotational flow, $\underline{\nabla} \times \underline{\mathcal{V}}(\underline{r}) = 0$. Two well known examples of flows

accurately represented by wavefunctions of the type (17) are,

(i) For the entire fluid in bodily motion

$$\Psi = \exp\left(i \, \underline{K}.\underline{R}\right) \Phi \quad , \quad \underline{R} = \frac{1}{N} \sum_{j=1}^{N} \underline{r}_j \tag{20}$$

and from (19) $\quad \underline{V}(\underline{\Gamma}) = \dfrac{\hbar \underline{k}}{Nm}$ $\qquad$ as expected.

(ii) For vortex motion about a line singularity

$$\Psi = \prod_j f(\rho_j) \exp(i \ell \phi_j) \; \Phi \tag{21}$$

where f is real and the phase $S(\underline{\Gamma})$ is an integral multiple of the azimuthal angle of the jth particle. (See [8] page 86). Using (19) we have, as expected, an azimuthal velocity

$$V_\phi = \dfrac{\hbar \ell}{m \rho} \tag{22}$$

where ρ is the distance from the line singularity.

An important question that we will now consider is whether we can make up the wavefunction corresponding to any classical flow field represented by a real velocity potential $\phi(\underline{\Gamma})$ (That is, $\underline{V}(\underline{\Gamma}) = \underline{\nabla} \phi(\underline{\Gamma})$) by writing the wavefunction in the form (17) with

$$S(\underline{\Gamma}) = \dfrac{m}{\hbar} \phi(\underline{\Gamma}) \tag{23}$$

Firstly we have $\quad |\Psi|^2 = |\Phi|^2 = \Phi_o^2 \quad$, so from (9), (18) and (23) with $S(\underline{\Gamma})$ real, we find

$$\sum_j \left\{ \Phi_o \, \nabla_j^2 \phi(\underline{\Gamma}_j) + 2 \, \underline{\nabla}_j \phi(\underline{\Gamma}_j) . \, \underline{\nabla}_j \Phi_o \right\} = 0 \tag{24}$$

Therefore the flow field is incompressible ($\nabla^2 \phi = 0$) if

$\sum_j \underline{\nabla}_j \phi(\underline{\Gamma}_j) . \underline{\nabla}_j \Phi_o = 0 \qquad$. Note, both (20) and (21) are incompressible flow fields and satisfy (24) because Φ_o is in the rest frame, that is $\sum_j \underline{\nabla}_j \Phi_o = 0 \qquad$. Now it was shown in [11] , that for Hamiltonians of the type (8) with $m_j = m$ for all j and the potential V independent of momenta, that the expectation value of the energy of the state $\Psi = F \Phi$ is given by (when F is symmetric)

$$\Delta E \equiv E - E_o = \dfrac{N \hbar^2}{2m} \dfrac{\langle |\nabla_1 F|^2 \rangle}{\langle |F|^2 \rangle} \tag{25a}$$

where $\quad H \Phi = E_o \Phi \quad$ and

$$\langle B \rangle = \dfrac{\int d\underline{\Gamma}_1 \ldots d\underline{\Gamma}_N \, B \, \Phi_o^2}{\int d\underline{\Gamma}_1 \ldots d\underline{\Gamma}_N \, \Phi_o^2} \tag{25b}$$

Inserting (23) into (17), we have

$$|\nabla_1 F|^2 = (\underline{\nabla}_1 S(\underline{r}))^2 = \frac{m^2}{\hbar^2}(\underline{\nabla}_1 \phi(\underline{r}_1))^2 \tag{26}$$

and $|F|^2 = 1$, as S is real.

Therefore using (25), (26) and (19)

$$\Delta E = \frac{Nm}{2} \frac{\int d\underline{r}_1 \ldots d\underline{r}_N (\underline{\nabla}_1 \phi(\underline{r}_1))^2 \underline{\Phi}_0^2}{\int d\underline{r}_1 \ldots d\underline{r}_N \Phi_0^2}$$

$$= \frac{1}{2} \int d\underline{r}\, \rho(\underline{r})\, \underline{v}^2(\underline{r}) \tag{27}$$

where $\rho(\underline{r}) = m.n(\underline{r})$ is the mass density, and $\underline{v}$ the fluid velocity.
Thus we see that the substitution (23) just leads to the quantum
variational energy becoming the kinetic energy of a classical liquid
undergoing potential flow with fluid velocity $\underline{v}(\underline{r}) = \underline{\nabla}\phi(\underline{r})$.
([9] page 8.) Unfortunately this promising result is misleading
because the quantum velocity field (19) is an average velocity and as
we shall see below cannot represent a classical fluid velocity unless
the entire fluid is moving as a body. In fact there are probably only
two exact quantum states of the type (17), namely the ones given by
(20) and (21). ([8] page 76.) This is because for a wavefunction to
be an exact excited quantum state it must be orthogonal to the ground
state and must have a suitable time dependence. Now let's look at the
time dependence of a state given by

$$\Psi = \exp(i S)\Phi_0 \quad, \quad S = \sum_{j=1}^{N} s(\underline{r}_j, t) \tag{28}$$

If we wish $S(\underline{r}, t)$ to have a time dependence like $\cos\omega t$, as would often be
the case if we used (23), then Ψ would have a time dependence of the
form $\exp(i\cos\omega t)$, instead of the usual form $e^{-i\omega t}$. Thus
for an S given by (23) to have a suitable time dependence necessary to
describe back and forth flow, the wavefunction (28) doesn't have the
required time dependence. There is also a problem with satisfying the

orthogonality condition for the state (28). The orthogonality

condition $\int d\Gamma_1 \ldots d\Gamma_N \Psi \Phi_0 = 0$ leads to, for (28),

$$\left\langle \cos\left(\sum_j S(\Gamma_j)\right) + i\sin\left(\sum_j S(\Gamma_j)\right) \right\rangle = 0, \quad \langle\ \rangle \text{ defined by (25b)} \qquad (29)$$

For the special case of uniform motion, (20), we can see that (29) does

hold by transforming to the normal coordinates $\underline{\xi}_\lambda$, defined by, [10]

$$\underline{\xi}_\lambda = \sqrt{\frac{\lambda}{\lambda+1}}\left(\underline{\Gamma}_{\lambda+1} - \frac{1}{\lambda}\sum_{i=1}^{\lambda}\underline{\Gamma}_i\right), \ (\lambda = 1,2,\cdots,N-1) \ ; \quad \underline{\xi}_N = \frac{1}{\sqrt{N}}\sum_{i=1}^{N}\underline{\Gamma}_i \qquad (30)$$

Then $\langle \exp(i\underline{K}.\underline{R}) \rangle$ contains

$$\int d\underline{\xi}_N \exp\left(i\underline{K}.\underline{\xi}_N/\sqrt{N}\right) = \frac{(2\pi)^3}{\sqrt{N}}\delta(\underline{K}) \ , \ (=0 \ |\underline{K}| \neq 0)$$

However in general (29) seems an almost impossible condition to satisfy

with S given by (23). So the wavefunction (28) is almost certainly an

energy eigenstate for only two cases : uniform motion (20), and vortex

motion (21), both of which are states where the entire flow is nett flow,

unlike a phonon or any other type of oscillatory motion. An interesting

question is, what would (28) represent if we chose $S(\underline{\Gamma},t)$ so that

$\frac{\hbar}{m} \text{Re}(\underline{\nabla}S)$ describes an oscillatory motion? From above we

know that such a wavefunction could not be a single excited state, but

it seems plausible that it would represent a large amplitude disturbance

characterised by the velocity potential $\frac{\hbar}{m}S$. A large-scale motion

such as this is not an eigenstate of the Hamiltonian, but rather consists

of a multitude of excitations with different eigenvalues.

To get more insight into what the velocity field (16) represents

physically, we will next consider the phonon wavefunction,

$$\Psi = \sum_{j=1}^{N} e^{i\underline{k}.\underline{\Gamma}_j} \Phi \qquad (31)$$

For small $|\underline{k}|$ (31) represents a longitudinal compressional sound wave

within the fluid. (See Feynman, [6]). Now the time independent part of the velocity potential for a sound wave in a classical fluid satisfies the wave equation, ([9] pages 492,6)

$$(\nabla^2 + k^2)\,\phi_o(\underline{r}) = 0 \quad , \quad \phi = \phi_o e^{-i\omega t} \tag{32}$$

where $k = \omega/c$ is the same wavenumber appearing in (31). The plane wave solution of (32) is

$$\phi(\underline{r},t) = A\,e^{i(\underline{k}\cdot\underline{r} - \omega t)} \tag{33}$$

Thus (31) is actually the wavefunction

$$\Psi = \sum_{j}^{N} \phi(\underline{r}_j,t)\,\overline{\Phi}_o \tag{34}$$

where ϕ is the classical velocity potential for the fluid motion corresponding to the quantum excitation (31). From (33) we have for the time independent part of the classical velocity field,

$$\underline{v}(\underline{r}) = \text{Re}\,\underline{\nabla}\phi_o(\underline{r}) = -A\,\underline{k}\,\sin\underline{k}\cdot\underline{r} \tag{35}$$

which as we expected, represents a longitudinal density oscillation within the fluid. However, in appendix BI(2) we show that our quantum velocity field applied to (31), gives at least approximately,

$$\underline{v}(1) = \frac{\hbar\underline{k}}{Nm} \tag{36}$$

Comparing (35) with (36) we see that our quantum velocity formula (16) has averaged out all the internal fluid oscillation described by (35) and only describes a small nett flow of matter of the order of $1/N$ times the total motion. Thus (16) is only the velocity field of the nett flow of matter in the system. Another example is the real vibrational wavefunction discussed in AIII. In this case (16) gives

$\underline{v} = 0$, as it does for any real wavefunction, which we now know only means that the nett flow of matter is zero, as we would expect for a pure vibration.

The inability of the quantum velocity formula to describe oscillatory motion is due to the problem of time dependence. In particular, in setting up the velocity field we derived a continuity equation (12) that holds for any given time dependence of the excited state number density and current density (provided $H\Psi = i\hbar \frac{\partial \Psi}{\partial t}$), but from the definitions of $n_i(1)$ and $\underline{j}_i(1)$ (eqns (10) and (11)) we see that they are both independent of time whenever Ψ is an energy eigenstate. This follows because when Ψ is a solution of Schrodinger's equation $H\Psi = i\hbar \frac{\partial \Psi}{\partial t}$, we can write $\Psi = \Psi_0 e^{-i\omega t}$, so that $|\Psi|^2$ is independent of time. However, assuming the velocity formula remains at least approximately correct for non energy eigenstates we can write down the wavefunction for a large amplitude time dependent disturbance described by a velocity potential $\phi(\underline{r},t)$ (i.e. $\underline{v}(\underline{r},t) = Re(\underline{\nabla}\phi(\underline{r},t))$) :

$$\Psi = \prod_j e^{i\frac{m}{\hbar}\phi(\underline{r},t)} \Phi_0 \qquad (37)$$

This is important since the part of (37) that is linear in the amplitude of ϕ is the sum wavefunction (34), which has the time dependence of ϕ $(\sim e^{-i\omega t})$ and is easy to make orthogonal to Φ_0 . In the following section BII, this idea is investigated with a view to understanding what the wavefunction is for an excitation corresponding to a given classical flow field.

APPENDIX BI(1) :   The Continuity Equation.

Firstly we have from (9) that,

$$\frac{\partial}{\partial t} \int \underline{dr_1} \ldots \underline{dr_N} |\Psi|^2 = \int \underline{dr_1} \ldots \underline{dr_N} \frac{\partial |\Psi|^2}{\partial t}$$

$$= -\hbar \sum_{j=1}^{N} \frac{1}{m_j} \int \underline{dr_1} \ldots \underline{dr_N} \underline{\nabla}_j \cdot Im(\Psi^* \underline{\nabla}_j \Psi)$$

$$= -\hbar \sum_{j=1}^{N} \frac{1}{m_j} \int \underline{dr_1} \ldots \underline{dr_{j-1}} \underline{dr_{j+1}} \ldots \underline{dr_N} \int \underline{dr_j} \underline{\nabla}_j Im(\Psi^* \underline{\nabla}_j \Psi)$$

$$= 0$$

because, (using periodic boundary conditions, or taking Ψ to be zero for $r_j = \pm \infty$) ,

$$\int \underline{dr_j} \underline{\nabla}_j \cdot Im(\Psi^* \underline{\nabla}_j \Psi) = 0 \qquad\qquad (a)$$

Therefore from (10) we have,

$$\frac{\partial n_1(1)}{\partial t} = \frac{\int \underline{dr_2} \ldots \underline{dr_N} \frac{\partial |\Psi|^2}{\partial t}}{\int \underline{dr_1} \ldots \underline{dr_N} |\Psi|^2}$$

$$= -\frac{\hbar}{g} \sum_{j=1}^{N} \frac{1}{m_j} \int \underline{dr_2} \ldots \underline{dr_N} \underline{\nabla}_j \cdot Im(\Psi^* \underline{\nabla}_j \Psi) \quad , \text{ from (9)}$$

(where $g = \int \underline{dr_1} \ldots \underline{dr_N} |\Psi|^2$)

from (11) $\qquad = -\underline{\nabla}_1 \cdot \underline{j}_1(1) - \frac{\hbar}{g} \sum_{j=2}^{N} \frac{1}{m_j} \int \underline{dr_2} \ldots \underline{dr_N} \underline{\nabla}_j \cdot Im(\Psi^* \underline{\nabla}_j \Psi)$

using (a) $\qquad = -\underline{\nabla}_1 \cdot \underline{j}_1(1)$

Thus, $\qquad \dfrac{\partial n_1(1)}{\partial t} + \underline{\nabla}_1 \cdot \underline{j}_1(1) = 0$

APPENDIX BI(2) : The Quantum Velocity Field of a Phonon.

Define $\langle \ \rangle$ by (25b) and $\langle \ \rangle_1$ by

$$\langle B \rangle_1 = V \frac{\int d\underline{r}_2 \ldots d\underline{r}_N B \Phi_0^2}{\int d\underline{r}_1 \ldots d\underline{r}_N \Phi_0^2}$$

For $\Psi = F \Phi_0$, Φ_0 real, we have

$$\text{Im}(\Psi^* \underline{\nabla}_1 \Psi) = \Phi_0^2 \text{Im}(F^* \underline{\nabla}_1 F)$$

Therefore, (16) becomes for $\Psi = F \Phi_0$

$$\underline{v}(1) = \frac{\hbar}{m} \frac{\langle \text{Im}(F^* \underline{\nabla}_1 F) \rangle_1}{\langle |F|^2 \rangle_1} \tag{a}$$

We wish to apply (a) to (31), namely

$$F = \sum_{j=1}^{N} e^{i\underline{k} \cdot \underline{r}_j} \quad , \text{ (time is suppressed as it cancels in (a))} \tag{b}$$

Taking the ground state density $n(1)$, (see AI(2)) to be the constant N/V , we have, $\langle 1 \rangle_1 = 1$

Therefore, $\langle |F|^2 \rangle_1 = N + 2(N-1)\langle \cos \underline{k} \cdot \underline{r}_{12} \rangle_1 + (N-1)(N-2)\langle \cos \underline{k} \cdot \underline{r}_{23} \rangle_1$ (c)

$$\langle \cos \underline{k} \cdot \underline{r}_{12} \rangle_1 = \frac{V}{N(N-1)} \int d\underline{r}_2 \cos \underline{k} \cdot \underline{r}_{12} \, n(12) \quad , \quad n(12) \text{ defined by AI(2)}.$$

Let us use the approximation for $n(12)$ (AI(6)),

$$n(12) = n(1) \, n(2) \, g(r_{12}) = \frac{N^2}{V^2} g(r_{12}) \tag{d}$$

Therefore, $\langle \cos \underline{k} \, \underline{r}_{12} \rangle_1 = \frac{N}{(N-1)} \frac{1}{V} \int d\underline{r}_2 \cos \underline{k} \cdot \underline{r}_{12} \, g(r_{12})$

$$= \frac{N}{(N-1)} \frac{1}{V} \int d\underline{r} \cos \underline{k} \cdot \underline{r} \, g(r) \tag{e}$$

Let's also define, $S(k) = \frac{\langle |F|^2 \rangle}{N}$, F given by (b)

Therefore, $S(k) = 1 + \frac{1}{N} \langle \sum_{i}^{N} \sum_{j \neq i}^{N} \cos \underline{k} \cdot \underline{r}_{ij} \rangle$

$$= 1 + (N-1) \langle \cos \underline{k} \cdot \underline{r}_{12} \rangle$$

$$= 1 + \frac{1}{N} \int d\underline{r}_1 \, d\underline{r}_2 \cos \underline{k} \cdot \underline{r}_{12} \, n(12)$$

$$= 1 + \frac{N}{V} \int d\underline{r} \cos \underline{k} \cdot \underline{r} \, g(r) \qquad \text{, using (d)} \qquad \text{(f)}$$

Thus from (e) $\quad \langle \cos \underline{k} \cdot \underline{r}_{12} \rangle_1 = \frac{1}{(N-1)} (S(k) - 1)$ (g)

To evaluate the last term of (c) we use the approximation,

$$\langle \cos \underline{k} \cdot \underline{r}_{23} \rangle_1 = \langle \cos \underline{k} \cdot \underline{r}_{12} \rangle$$

$$= \frac{1}{(N-1)} (S(k) - 1) \qquad \text{(h)}$$

((h) is rigorously true if we can write $n(123) = \frac{N^3}{V^3} g(r_{23}) \frac{(N-2)}{N}$,

(compare with AI(2), AI(6)) when evaluating $\langle \cos \underline{k} \cdot \underline{r}_{23} \rangle_1$. This

is like defining a weak correlation limit, (see appendix AI(1))

$$g(r_{12}) \, g(r_{13}) = \frac{(N-2)}{N} = 1 - \frac{2}{N} \qquad)$$

From (g), (h) and (c) we get,

$$\langle |F|^2 \rangle_1 = N \, S(k) \qquad \text{(i)}$$

From (b) we have $\quad F^* \underline{\nabla}_1 F = i \underline{k} \sum_{j=1}^{N} e^{i \underline{k} \cdot \underline{r}_{1j}}$

Therefore, $\quad \langle \text{Im}(F^* \underline{\nabla}_1 F) \rangle_1 = \underline{k} \{ \langle 1 \rangle_1 + (N-1) \langle \cos \underline{k} \cdot \underline{r}_{12} \rangle_1 \}$

$$= \underline{k} \, S(k) \qquad \text{(j)}$$

Finally from (i), (j) and (a) we obtain

$$\underline{v}(1) = \frac{\hbar \underline{k}}{N m} \qquad \text{(k)}$$

We should note that (k) is a surprising result in that it is not a

function of $\underline{r}_1$. In fact the step (h) is equivalent to taking the

excited state number density $n_1(1)$, (10), to be the constant $\frac{1}{V}$.

Thus for an interacting fluid the approximation (h) may have resulted

in small correction terms dependent on $\underline{r}_1$ being left out of (k) .

(Any such terms would need to satisfy the continuity equation (12).)

However, we can see that (k) is the velocity corresponding to the nett momentum transferred by the excitation (31), by calculating the momentum eigenvalue. With $\Psi = F \Phi$, we put the ground state $\overline{\Phi}$ in the zero momentum frame, namely, $\underline{P} \Phi = 0$, where

$$\underline{P} = -i\hbar \sum_{\ell=1}^{N} \nabla_\ell .$$

Firstly, let's calculate $\underline{P} \Psi$ for a uniform translational motion

$$\Psi' = \prod_j e^{i \underline{k} \cdot \underline{r}_j} \Phi$$, (compare with (20))

$$\underline{P} \Psi' = -i\hbar \sum_\ell i \underline{k} \, \Psi' = N\hbar\underline{k} \, \Psi' .$$

This is bodily flow with velocity $\dfrac{N\hbar\underline{k}}{Nm} = \dfrac{\hbar\underline{k}}{m}$

For the phonon wavefunction (31), $\Psi = \sum_j e^{i \underline{k} \cdot \underline{r}_j} \Phi$

$$\underline{P} \Psi = -i\hbar \sum_\ell i \underline{k} \, e^{i \underline{k} \cdot \underline{r}_\ell} \Phi = \hbar\underline{k} \, \Psi \qquad (1)$$

Therefore the phonon has a total momentum eigenvalue $O\left(\frac{1}{N}\right)$ times that of a comparable bodily flow. We can see from (1) that the total nett momentum transported by a phonon excitation (31), corresponds to a velocity $\underline{v} = \hbar\underline{k} / Nm$. That is, a nett mass flow of the order of $\frac{1}{N}$ times the total motion.

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II. WAVEFUNCTIONS FOR QUANTUM STATES CORRESPONDING TO CLASSICAL FLOW FIELDS.

In Section BI simple product and simple sum wavefunctions (BI (17), (28) and (34)) were discussed, while investigating the nature of the quantum velocity field $\underline{v}$, BI (16). So let us begin this section by summarising what we have learnt about these wavefunctions.

a) $\prod_j \exp(i\, S(\underline{r}_j))\, \Phi \quad : \left(\Phi = \Phi_o e^{-i\omega t} \right)$ (1)

(i) $\underline{v}(\underline{r}) = \frac{\hbar}{m}\, Re(\underline{\nabla}\, S(\underline{r}))$. Note $\underline{\nabla} \times \underline{v} = 0$.

(ii) With $S(\underline{r})$ real it follows from BI (24) that $\nabla^2 \underline{v} \doteq 0$ since $\underline{\nabla}_j \Phi_o$ changes sign rapidly over a microscopic variation of $\underline{r}_j$, while $\underline{\nabla}_j \phi(\underline{r}_j)$ will always be slowly varying on the microscopic scale for low energy states.

(iii) With $S(\underline{r})$ real $E - E_o = \frac{m}{2} \int \underline{dr}\, n(\underline{r})\, \underline{v}^2(r)$, where E is the expectation value of the excited energy and $H\Phi = E_o \Phi$.

(iv) If we let $S(\underline{r})$ be complex then (ii) and (iii) no longer hold. The effect of a complex $S(\underline{r})$ is to create a permanent deformation in the density, e.g. the factor $f(\rho_j)$ in BI (21) produces the hole around the vortex line.

(v) Because the wavefunction (1) has the time dependence $e^{-i\omega t}$, $\underline{v}$ is independent of time and so gives only the nett flow of matter after averaging out any oscillatory motion.

(vi) It is very hard to make this wavefunction orthogonal to Φ (see BI (29)).

(vii) It is likely that the only exact eigenstates of this type are uniform motion and vortex motion, i.e. BI (20-21).

From the above we can conclude that this product wavefunction

describes permanent mass flow and not oscillatory density variations. As an aside it is worth showing that the particular example

$$\Psi = \prod_j e^{i\underline{k}.\underline{r}_j} \Phi \tag{2}$$

is the wave-mechanical method of applying a Galilean transformation to Φ : $\underline{v}' = \underline{v} + \underline{u}$ where $\underline{u} = \dfrac{\hbar \underline{k}}{m}$. This follows since

$$|\Psi|^2 = |\Phi|^2 , \quad \underline{\nabla_1} \Psi = i\underline{k}\,\Psi + \prod_j e^{i\underline{k}.\underline{r}_j} \underline{\nabla_1}\Phi$$

and so using BI (16),

$$\underline{v}' = \frac{\hbar}{m} \frac{\int d\underline{r}_2 \ldots d\underline{r}_N\, \text{Im}(\Psi^* \underline{\nabla} \Psi)}{\int d\underline{r}_1 \ldots d\underline{r}_N\, |\Psi|^2}$$

$$= \frac{\hbar \underline{k}}{m} + \frac{\int d\underline{r}_2 \ldots d\underline{r}_N\, \text{Im}(\Phi^* \underline{\nabla_1} \Phi)}{\int d\underline{r}_1 \ldots d\underline{r}_N\, |\Phi|^2} \tag{3}$$

$$= \underline{u} + \underline{v}$$

b) $\displaystyle\sum_j \phi(\underline{r}_j, t)\, \Phi_o$: $\tag{4}$

    (i) has the time dependence of ϕ.

    (ii) is easy to make orthogonal to Φ_o . (In the notation of BI (29) we require only $\langle \phi \rangle = 0$.)

    (iii) The wavefunction (4) describes oscillatory density variations, but the quantum velocity field will average it all out to give only the nett flow.

    (iv) By analogy with phonons (i.e. BI (31)) it is plausible to speculate that to a first approximation the classical (dynamic) flow field corresponding to the state (4) is $\underline{v}_c \sim \text{Re}(\underline{\nabla}\phi)$.

Note (iv) is important since if it was true in general, (4) would be the single excitation corresponding to the classical potential flow

field of small amplitude, $\underline{v_c}(\underline{r}, t) = Re(\underline{\nabla}\phi(\underline{r}, t))$, with $\phi(\underline{r}, t) = \phi_0(\underline{r})e^{-i\omega t}$. This idea is strongly supported by the argument due to Chang and Cohen [1] , discussed below.

c) $\prod_j \exp(i S(\underline{r}, t)) \Phi_0 :$ (5)

(i) Except for the special case $S(\underline{r}, t) = S(\underline{r}) - \omega t$ (i.e. (1)), (5) is not a solution of Schrodinger's equation and cannot be made orthogonal to Φ_0 .

(ii) Assuming that BI (16) is still correct we have $\underline{v}(\underline{r}, t) = \frac{\hbar}{m} Re(\underline{\nabla} S(\underline{r}, t))$. Note $\underline{\nabla} \times \underline{v} = 0$

(iii) From BI (25) I calculate E, the expectation value of the energy in the state (5), to be

$$E - E_0 = \frac{m}{2} \int d\underline{r}\, \eta_1(\underline{r})\, \underline{\tilde{v}}^* \cdot \underline{\tilde{v}} \quad , \quad H\Phi = E_0\Phi \quad (6)$$

where $\underline{\tilde{v}} = \frac{\hbar}{m}(\underline{\nabla} S)$ and $\eta_1(\underline{r})$ is the density in the excited state (5). (Compare this with BI (27).)

(iv) From (iii) and by analogy with the wavefunction (1) it seems plausible that (5) represents a large-amplitude disturbance with velocity field $\underline{v}$.

Chang and Cohen [1] assume the conclusion c)(iv), that the wavefunction (5) represents large-scale hydrodynamic motion with velocity potential S, and then use this to show that the wavefunction (4) is the single excitation corresponding to the same velocity field (i.e. conclusion b(iv)). Let us write the velocity potential as $\alpha\phi$ where α is proportional to the amplitude and absorb the constant $\frac{\hbar}{m}$ into α when appropriate, then Chang and Cohen's argument is to expand the wavefunction (5) :

$$\exp\left(i\alpha\sum_j \phi(\underline{r}_j, t)\right)\Phi_0 = \left[1 + i\alpha\sum_j \phi - \alpha^2\left(\sum_j \phi\right)^2 - i\alpha^3\left(\sum_j \phi\right)^3 + \cdots\right]\Phi_0 \quad (7)$$

They identify this expansion as a superposition of the ground state Φ_0 with the state of a single excitation, plus the state of two excitations, and so on. So the state of a single excitation with velocity potential $\emptyset$ is the term linear in α :

$$\sum_j \phi(\underline{r}_j, t)\, \Phi_0 \tag{4}$$

Because a quantum mechanical wavefunction is normalised the amplitude has dropped out of (4), unlike the wavefunction (7). So (4) describes motion in the small amplitude limit. If (4) has momentum eigenvalue $\underline{k}$ then (according to Chang and Cohen) the term in (7) proportional to α^n belongs to momentum eigenvalue $n\underline{k}$, and represents a state composed of n excitations. The argument due to Feynman referred to on page AIII. 20., that the state of two phonon excitations of momentum $\hbar\underline{k}$ is

$$\left(\sum_j e^{i\underline{k}\cdot\underline{r}_j}\right)^2 \Phi$$ and has momentum eigenvalue $\simeq 2\hbar\underline{k}$,

agrees with Chang and Cohen's identification. Now, the wavefunction (4) is very important since it is a single excitation and in the limit of small amplitude (long wavelength) it is likely to be an exact energy eigenstate. But before investigating examples of the type (4) I will first consider the large amplitude wavefunction (7) since Chang and Cohen's argument is based on the assumption that it represents a flow field with velocity potential $\alpha\phi$.

Large-amplitude flow wavefunctions :

The problem to consider is : because the wavefunction $\prod_j e^{i\alpha\phi_j} \Phi_0$ is not an exact solution of Schrodinger's equation (except for centre of mass motion and vortex motion), the proof of the continuity equation BI (15) and hence the derivation of the velocity field

$\underline{v} \sim Re(\nabla\phi)$ no longer follows. That is, can the large amplitude wavefunction approximate this flow field despite the fact that it cannot be made orthogonal to the ground state, nor has the

correct time dependence, and is not quantised (i.e. contains arbitrary α) ? Writing the wavefunction (7) in the form

$$\Psi = e^{iX}\, \Phi_{o} \qquad , \qquad X = A + iB$$

we have

$$\text{Im}\left(\Psi^{*}\nabla_{j}\Psi\right) = \text{Re}\left(\nabla_{j}X\right)|\Psi|^{2}, \quad |\Psi|^{2} = e^{-2B}\Phi_{o}^{2} \ .$$

Therefore equation BI (9), which is the condition that the derivation of the continuity equation BI (15) and the velocity field BI (16) holds, is

$$\frac{\partial|\Psi|^{2}}{\partial t} + \sum_{j}\nabla_{j}.\left(\underline{v}_{j}|\Psi|^{2}\right) = 0 \tag{8}$$

where

$$\underline{v}_{j} \equiv \frac{\hbar}{m}\,\text{Re}\left(\nabla_{j}X\right) \tag{9}$$

The velocity field will be correctly described by (9) provided (8) is satisfied for the chosen X . Equation (8) reduces to

$$e^{-2B}\Phi_{o}^{2}\left\{-2\frac{\partial B}{\partial t} + \frac{\hbar}{m}\sum_{j}\nabla_{j}^{2}A\right\} + \frac{\hbar}{m}\sum_{j}\nabla_{j}A.\nabla_{j}\left(e^{-2B}\Phi_{o}^{2}\right) \tag{10}$$

Now since $\nabla_{j}\Phi_{o}$ changes $\overset{\text{sign}}{\wedge}$ rapidly over a microscopic variation of $\underline{r}_{j}$ and $\nabla_{j}A$ is slowly varying for low energy (long wavelength) states, we have almost exactly that

$$\sum_{j}\nabla_{j}A.\nabla_{j}\Phi_{o} = 0 \tag{11}$$

From (11), (10) becomes

$$-2\frac{\partial B}{\partial t} + \frac{\hbar}{m}\sum_{j}\nabla_{j}^{2}A - \frac{2\hbar}{m}\sum_{j}\nabla_{j}A.\nabla_{j}B = 0 \tag{12}$$

For wavefunctions of the form (7) we have

$$X = A + iB = \sum_{j}\left[a(\underline{r}_{j},t) + i\,b(\underline{r}_{j},t)\right]$$

and (12) reduces to

$$-2\dot{b} + \frac{\hbar}{m}\nabla^{2}a - 2\frac{\hbar}{m}\nabla a.\nabla b = 0 \qquad \left(\dot{} \equiv \frac{\partial}{\partial t}\right) \tag{13}$$

When X is real $b = 0$, and (13) is just

$$\nabla^{2}a = 0$$

With this condition we have from (9) that $\nabla \cdot \underline{v} = 0$, so a real velocity potential can only describe incompressible flow. From BI (27) we see that the extra energy associated with the incompressible flow is

$$\Delta E = \frac{\hbar^2}{2m} \int \underline{dr}\, n(\underline{r})\, \nabla a \cdot \nabla a \qquad (14)$$

Taking variations with respect to a gives the condition for a minimum in the energy as $\nabla \cdot (\nabla a) = 0$, that is $\nabla^2 a = 0$. So in this case the approximation (11) is exact.

States of compressible flow will be described by the wavefunction (7) if we allow χ to be complex, that is $b \neq 0$. However (13) will no longer be satisfied exactly and so the expression (9) for the velocity field is at the best approximate. But if we make the same assumption that is usually made for classical compressible flows, that the amplitude remains small enough to ignore all terms not linear in amplitude, then (13) becomes

$$-2\dot{b} + \frac{\hbar}{m}\nabla^2 a \simeq 0 \qquad (15)$$

This condition should be compared with the classical continuity equation for compressible flow, in the same approximation ([2] page 476)

$$\delta\dot{\rho} + \nabla \cdot \underline{v} = 0 \qquad , \left(\rho = \rho_0(1+\delta\rho)\right)$$

If the continuity equation (15) holds then from BI we can identify $\underline{v} = \frac{\hbar}{m}\nabla a$ and the wavefunction $\prod_i \exp(i[a_i + i b_i])\Phi_0$ will approximate a state of large-scale flow with velocity field $\underline{v}$, which is the assumption that Chang and Cohen's argument requires.

To investigate the solutions of (15) let us consider two examples, one of incompressible flow and the other compressible flow.

Large amplitude surface wave :

Take the ground state to be a semi-infinite liquid with a free

surface in the xy plane, and let the density approach zero as $z \rightarrow +\infty$. Then the velocity potential for a classical capillary wave propagating in the x-direction can be taken to be ([2] page 457, [3] page 238))

$$\alpha\phi = \alpha e^{kz} \cos(kx - \omega t) \tag{16}$$

where $\omega^2 = \frac{\sigma}{n_0 m} k^3$; σ is the surface tension and n_0 the bulk number density. So the large emplitude surface wave state should be

$$\Psi_{sw} = \prod_j \exp\left(i\alpha\, e^{kz_j} \cos(kx_j - \omega t)\right) \Phi_0 \tag{17}$$

Since we have taken ϕ to be real the continuity equation (15) is just $\nabla^2 \phi = 0$, which is satisfied by (16). That is, the flow is incompressible. From (14) and taking the ground state number density to be $n(z) = n_0 \; z < 0 \; , = 0 \; z > 0$, it is easy to show that

$$\Delta E = A\alpha^2 \frac{\hbar^2 k}{4m} n_0 \qquad , \text{A the surface area.}$$

Then a simple classical calculation of the increase in surface area (ΔA) to lowest order in α yields the classical dispersion relation

$$\omega^2 = \frac{\sigma}{n_0 m} k^3 \qquad , \sigma = \frac{\Delta E}{\Delta A} \quad . \text{ That is, because (17)}$$

represents incompressible flow it leads to the classical flow energy (14), which ensures that Ψ_{sw} is consistent with the assumption that it represents a large amplitude surface wave. It is obviously a large amplitude disturbance since $\Delta E \sim \alpha^2$.

Large amplitude sound wave :

Take the ground state to be an infinite liquid of density n_0. Now, a classical sound wave travelling in the $\hat{k}$ direction could be represented by a velocity potential ([2] page 496, [3] page 248)

$$\alpha\phi = \alpha \cos(\underline{k}.\underline{r} - \omega t) \qquad , \tag{18a}$$

or $\qquad \alpha\phi = \alpha\, e^{i(\underline{k}.\underline{r} - \omega t)} \tag{18b}$

From (15) we see that ϕ must be taken to be complex or it could not represent compressional flow (i.e. (18a) cannot satisfy the quantum continuity equation (15)). So the large amplitude sound wave state would have to be

$$\Psi = \prod_j \exp\left(i\alpha\, e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}\right)\Phi_0 \tag{19}$$

Inserting (18b) into (15), (i.e. $a+ib=\alpha\phi$), leads to $2\omega - \dfrac{\hbar k^2}{m} = 0$. So the continuity equation gives the large amplitude dispersion relation as $\omega = \dfrac{\hbar k^2}{2m}$. This is different to the small amplitude classical relation $\omega = ck$, c the constant velocity of sound. Thus (19) is a solution of (15) but does not give the expected dispersion relation. From (6), the flow energy of the state Ψ is

$$\begin{aligned}
\Delta E &= \frac{\hbar^2}{2m}\alpha^2 \int d\mathbf{r}\, n_i(\mathbf{r})\,|\nabla\phi|^2 \\
&= \alpha^2 \frac{\hbar^2 k^2}{2m} \int d\mathbf{r}\, n_i(\mathbf{r}) \\
&\simeq N\alpha^2 \frac{\hbar^2 k^2}{2m} \\
&= N\alpha^2 \hbar\omega
\end{aligned} \tag{20}$$

This large amplitude energy should be compared with the energy of a single sound wave excitation,

$$\Delta E = \hbar\omega = \hbar ck \tag{21}$$

So the assumption of Chang and Cohen's, that the wavefunction $\prod_j \exp(i\alpha\phi(j))\Phi_0$ represents large scale motion with velocity field $\mathbf{v}\sim \mathrm{Re}(\nabla\phi)$, seems to be approximately correct but there are some problems. For example, ϕ must be real to describe incompressible flow but when using the single excitation wavefunction $\sum_j \phi(j)\Phi_0$ we require the complex form $\phi\sim e^{-i\omega t}$, to get the necessary

time dependence. The compressible flow has a complex ϕ but the energy (6) is no longer the exact classical flow energy. In fact it seems that only kinetic energy is present, which would be half the energy of a classical sound wave. Also the large amplitude sound wave had a strange dispersion relation, but this could be expected since classically $\omega \sim k$ only in the small amplitude limit.

Single excitations :

Chang and Cohen's argument (and BI) led to the conclusion that the wavefunction

$$\sum_j \phi(r_j, t) \, \Phi_o \tag{4}$$

is a single quantum excitation corresponding to a <u>classical</u> potential flow field $\underline{v} \sim Re(\underline{\nabla}\phi)$. In the remainder of this section I will consider further evidence for this important idea with emphasis on specific examples.

1) Firstly, the wavefunction (4) has the required properties to be an energy eigenstate. That is, it has the correct time dependence by taking $\phi \sim e^{-i\omega t}$, and is easy to make orthogonal to the ground state (i.e. $\langle \phi \rangle = 0$ where $\langle \, \rangle$ is defined by BI (25b)). Also, because the classical amplitude has dropped out of (4), it will be best in the small amplitude limit.

2) It may also be possible to derive the nature of (4) using the operator approach of collective coordinates. In CII the Hamiltonian in the continuum approximation, for states which represent small longitudinal deviations from uniform density, is shown to be

$$H_{ph} \simeq \frac{1}{2} \sum_k m_k \left\{ |\dot{n}_k|^2 + \omega_k^2 |n_k|^2 \right\} \tag{22}$$

where $m_k = m/N k^2$, $\omega_k = ck$, and the collective

coordinate η_k is the Fourier transform of the number density operator.

Now H_{ph} is a harmonic oscillator Hamiltonian and so the first excited

state wavefunction is the ground state multiplied by η_k . The Hamiltonian

(22) describes quantised sound waves or phonons and in this case

$$
\begin{aligned}
\eta_k &= \int \underline{dr}\, \phi(\underline{r}, t)\, n(\underline{r}) \\
&= \int \underline{dr}\, \phi(\underline{r}, t) \sum_j \delta(\underline{r}_j - \underline{r}) \\
&= \sum_j \phi(\underline{r}_j, t) \\
&= \sum_j e^{i(\underline{k}\cdot\underline{r}_j - \omega_k t)}
\end{aligned}
\tag{23}
$$

So the first excited state $\eta_k \Phi_0$ is the Feynman phonon wavefunction

BI (31), which is of the form (4). If we generalise the above and

assume that any small amplitude deviation from the ground state will

be described by a Hamiltonian of the form (22), in terms of a

collective coordinate η_k for the particular flow field, then by analogy

with the harmonic oscillator we know that a good variational wavefunction

for the lowest excitation with this flow field may be obtained by

multiplying the ground state by the collective coordinate : (cf. [4]

page 23)

$$
\Psi = \eta_k \Phi_0
\tag{24}
$$

That is, if η_k is the collective coordinate for the small amplitude

flow field described by velocity potential ϕ, then (24) is a good wave-

function for the single excitation corresponding to this flow field.

We see that (24) and (4) are identical if (23) is true in general,

i.e. provided

$$
\eta_k = \int \underline{dr}\, \phi(\underline{r}, t)\, n(\underline{r}) = \sum_j \phi(\underline{r}_j, t)
\tag{25}
$$

This equation is true for phonons and according to Chang and Cohen

([1] eqn (26)) also holds for quantised surface waves. I have not

seen a general statement of (25) but it may be a consequence of the fact, proved by Kronig and Thellung $\begin{bmatrix} 5 \end{bmatrix}$, that η and $\emptyset$ are canonically conjugate variables (η is the generalised momentum corresponding to the variable $\emptyset$).

3) Phonons (quantised sound waves) :

I have previously noted ((18b) and BI) that the velocity potential for a small amplitude classical sound wave is

$$\phi \sim e^{i(\mathbf{k}.\mathbf{r} - \omega t)}$$ where $\omega = ck$ (c the velocity of sound). So, assuming (4) is correct, the wavefunction for a single sound wave excitation (i.e. phonon) is:

$$\Psi_{ph} = \sum_j e^{i\mathbf{k}.\mathbf{r}_j} \, \Phi \tag{26}$$

This wavefunction is also obtained from the normal mode Hamiltonian which by analogy with a harmonic oscillator led to (24), as described in 2) above. Feynman $\begin{bmatrix} 6 \end{bmatrix}$ first proposed the wavefunction (26) for excitations in a bose liquid (liquid He^4), giving extensive physical arguments to conclude that for small k, Ψ_{ph} represents a longitudinal compressional sound wave with energy $\hbar\omega = \hbar c k$. Feynman also proved that Ψ_{ph} was the optimum wavefunction of the form $\sum_j f(\mathbf{r}_j) \, \Phi$. Applying BI (25a) we have from the variational principle (Ψ_{ph} is orthogonal to Φ for $k \neq 0$ since $\langle e^{i\mathbf{k}.\mathbf{r}} \rangle \sim \delta(\mathbf{k})$),

$$\Delta E = E - E_0 \leq \frac{\hbar^2 k^2}{2m \, S(k)} \tag{27}$$

where $H\Phi = E_0 \Phi$ and the structure factor S(k) is

$$S(k) = \frac{1}{N} \langle | \sum_j e^{i\mathbf{k}.\mathbf{r}_j} |^2 \rangle \tag{28}$$

($\langle \rangle$ is the ground state expectation value defined by BI (25b)). Defining the pair correlation function $g(r_{12})$ from the pair density (see AI (6))

$$n(\underline{r}_1,\underline{r}_2) = n_0^2 \, g(r_{12})$$

(28) becomes

$$S(k) = 1 + \frac{1}{N} \int d\underline{r}_1 \, d\underline{r}_2 \, n(\underline{r}_1,\underline{r}_2) \, e^{i\underline{k}\cdot(\underline{r}_1-\underline{r}_2)}$$

$$= 1 + n_0 \int d\underline{r} \, e^{-i\underline{k}\cdot\underline{r}} \, (g(r)-1) \quad ,$$

(29)

where the orthogonality condition has been used to replace g by (g-1) in the last line of (29). Inverting (29) we have

$$g(r)-1 = \frac{1}{(2\pi)^3 n_0} \int d\underline{r} \, e^{i\underline{k}\cdot\underline{r}} \, (S(k)-1)$$

$$= \frac{1}{2\pi^2 n_0 r} \int_0^\infty dk \, k \sin kr \, (S(k)-1)$$

(30)

Now we expect a long wavelength phonon to have the classical energy $\Delta E = \hbar c k$, which from (27) implies $S(k) \to \hbar k / 2mc$, as $k \to 0$. Integrating by parts the right hand side of (30) we see that this condition holds provided the long range part of (g-1) is

$$(g(r)-1) \longrightarrow -\frac{1}{2\pi^2 n_0} \frac{\hbar}{mc} \frac{1}{r^4}$$

(31)

The above self-consistent results are well accepted properties of the bulk superfluid phase of a Bose liquid (liquid He^4), but as pointed out by Chang and Cohen ([1] p 1064) the result (31) has not been derived from first principles because $\Delta E \to \hbar c k$ is really an assumption. (Feenberg has a proof of (31) using normal modes and sum rules, that might not have this hidden assumption, [7] , but I am not sure.)

In Appendix BI (2) I proved that $\underline{P} \, \Psi_{ph} = \hbar \underline{k} \, \Psi_{ph}$. That is, a phonon is a momentum eigenstate with an eigenvalue appropriate to the motion of a single particle rather than bodily flow. This agreed with the calculation of the quantum velocity field;

$\underline{v}_{qu} = \hbar\underline{k}/Nm$ (N the number of particles). So the quantum excitation includes a small nett transfer of matter of order of $\frac{1}{N}$ times the total motion. This is very interesting since it is known that the classical sound wave also has a second order nett transfer of matter. Landau and Lifshitz ([3] p 252) calculate the total momentum of a long wavelength classical sound wave to be $\hat{n}\,E/c$, where $\hat{\underline{n}}E$ is the energy of the sound wave directed along the line of propagation $\hat{\underline{n}}$. Substituting the assumed quantum energy $\hat{\underline{n}}E = \hbar c\underline{k}$, gives the proved result $\hbar\underline{k}$. This helps to confirm the universally accepted assumption, that in the limit $k\rightarrow o$ the wavefunction (26) is an exact energy eigenstate with an energy $\hbar\omega$ corresponding to the classical dispersion relation $\omega = ck$.

4) Surface excitations :

The velocity potential for a classical capillary wave (restoring force is σ , the surface tension), can be taken to be $\phi \sim e^{kz} e^{i(\underline{k}\cdot\underline{r} - \omega t)}$, where $\underline{k}$ is parallel to the surface and the liquid occupies the volume $z \leq 0$. The classical dispersion relation is $\omega^2 = \left(\sigma/n_o m\right) k^3$ where n_o is the bulk number density. Assuming that (4) is the correct wavefunction for a single quantum surface excitation we have

$$\psi_{sw} = \sum_j e^{kz_j} e^{i(\underline{k}\cdot\underline{r}_j - \omega t)} \Phi_o \tag{32}$$

For the large amplitude surface wave (17) it was necessary to use the real part of ϕ because the flow field is incompressible, but the complex form of ϕ is required in (32) in order for ψ_{sw} to have the correct time dependence $e^{-i\omega t}$. On the basis of the previously described argument that led to (4), Chang and Cohen [1] proposed the wavefunction (32) for the surface excitations of liquid helium four. In a paper supplied with this thesis [8], the wavefunction (32) is

used to analytically rederive the numerical results of Chang and Cohen. The purpose of [1] and [8] is to calculate the excitation energy of a single surface mode.

The analytic calculation of [8] is analogous to the derivation of the phonon excitation energy, described in 3) above. The surface mode excitation energy is found to be (cf. (27))

$$\Delta E \leq \frac{\hbar^2 k^2}{2m\, S^*(k)} \tag{33}$$

where S^* is the surface structure factor (cf. (28))

$$S^*(k) = \frac{\left\langle \left| \sum_j e^{kz_j} e^{i\mathbf{k}\cdot\mathbf{r}_j} \right|^2 \right\rangle}{2N \left\langle e^{2kz_1} \right\rangle} \tag{34}$$

Defining a generalised pair correlation function by

$$n(\mathbf{r}_1,\mathbf{r}_2) = n(z_1)\, n(z_2)\, g(r_{12}, z_1, z_2) \quad,$$

it is shown in [8] , that (34) becomes

$$S^*(k) = \frac{1}{2}\left\{ 1 + \frac{2\pi \int_{-\infty}^{\infty} dz_1\, n(z_1) e^{kz_1} \int_{-\infty}^{\infty} dz_2\, n(z_2) e^{kz_2} \int_0^{\infty} d\rho\, \rho\, J_0(k\rho) \left[g(r_{12}, z_1, z_2) - 1\right]}{\int_{-\infty}^{\infty} dz\, n(z)\, e^{2kz}} \right\} \tag{35}$$

To evaluate (35) it was necessary to make the same approximation that Chang and Cohen made : that the bulk properties held right up to the surface, i.e. the contribution to the ground state from the zero-point motion of the surface modes is neglected. Now in the phonon case the zero-point motion of the phonon modes can be shown, with various approximations and assuming the classical dispersion relation, to lead to equation (31) for the long ranged part of [g−1] . (This calculation is given in section CII.) But it was this variation of (g-1) that was required to reproduce the classical dispersion relation in the limit $k \to 0$. Hence it is not surprising that [1] and [8] failed to reproduce the classical energy in the long wavelength limit :

$$\hbar\omega \to \hbar\sqrt{\frac{\sigma}{n_0 m}}\, k^{3/2} \quad, \quad S^* \to \frac{\hbar}{2}\sqrt{\frac{n_0}{\sigma m}}\, k^{1/2} \tag{36}$$

In fact the approximations made in [1] and [8] gave a result that was much closer to $\omega \sim k$ than $\omega \sim k^{3/2}$. However the results for intermediate k were good and it was concluded in both papers that if the correct ground state was used (i.e. if the zero-point motion of the surface modes was included), then the classical result $\omega \sim k^{3/2}$ would be obtained in the limit $k \to 0$ using the wavefunction (32). The work required to prove this is summarised at the end of reference [8].

It is worth pointing out that the wavefunction (32) could also be used to calculate the energy of a surface excitation of Fermi liquid (liquid He^3). This is because any wavefunction of the form $F\Phi$ where F is a symmetric function of the coordinates has the permutation symmetry of the ground state Φ, and so the wavefunction (4) can be applied to either bose or Fermi systems by choosing the appropriate ground state.

From (36) we see that if the calculations of [8] could be improved so that in the long wavelength limit the classical dispersion relation was obtained, then a value for the surface tension would be found. The results of [8] are restricted to temperature T = 0, where the free energy is the total energy and the expectation values are not complicated thermal averages. So the system of most importance is superfluid He^4, where the surface tension at T = 0 is a well known experimental quantity (see Atkins and Narahara [9]). Assuming that the dispersion relation derived at T = 0 continues to hold at higher temperatures, it is also possible to calculate the temperature dependence of the surface tension (note, σ is the free energy per unit area of surface). The free energy excited by the surface modes at temperature T is given by the usual statistical mechanical expression for non interacting bosons with zero chemical potential (e.g. photons, phonons, surface modes) :

$$\Delta F \ (\equiv F - F_0) \ = \ -T \sum_{\underline{\ell}} \log \mathcal{Z}_\ell \tag{37}$$

where $\mathcal{Z}_\ell = \sum_{n=0}^{\infty} \exp(-n\epsilon_\ell/T) = \dfrac{1}{1-e^{-\epsilon_\ell/T}}$

and $F_0 = \dfrac{1}{2}\sum_{\underline{\ell}} \epsilon_\ell$ is the energy at T = 0. If ϵ_ℓ is a function

of a wavenumber $\underline{k}$ that is parallel to a surface of area $A = L^2$

$\left(e.g. \ \epsilon_\ell = \hbar c k_\ell \ ; \ \epsilon_\ell = \hbar \sqrt{\dfrac{\sigma}{n_0 m}} \, k_\ell^{3/2} \right)$, then using

the periodic boundary conditions $k_x = (2\pi/L)\ell_x, \ k_y = (2\pi/L)\ell_y$,

we have

$$\sum_{\underline{\ell}} = \sum_{\ell_x}\sum_{\ell_y} = \dfrac{A}{(2\pi)^2}\int_{-\infty}^{\infty}dk_x\int_{-\infty}^{\infty}dk_y = \dfrac{A}{2\pi}\int_0^\infty dk\, k \tag{38}$$

From (37) and (38) we obtain

$$\sigma(T) - \sigma_0 = \dfrac{T}{2\pi}\int_0^\infty dk\, k \log\left[1-e^{-\epsilon(k)/T}\right] \tag{39}$$

If we now take $\epsilon(k)$ to be of the form

$$\hbar\omega(k) = \hbar a k^b \qquad (a, b \ \text{constants}; \ a, b > 0)$$

then integrating (39) by parts gives

$$\sigma(T) = \sigma_0 - \dfrac{\hbar ab}{4\pi}\int_0^\infty dk\, k^{b+1}\dfrac{e^{-\epsilon(k)/T}}{1-e^{-\epsilon(k)/T}} \tag{40}$$

$$= \sigma_0 - \dfrac{\hbar a}{4\pi}\left(\dfrac{T}{\hbar a}\right)^{\frac{b+2}{b}}\sum_{n=1}^{\infty}\int_0^\infty dx\, x^{2/b}\, e^{-nx}$$

So if the dispersion relation is $\omega \sim k^b$ then the temperature

dependence of σ is $\left(\sigma(T)-\sigma_0\right) \sim T^{1+2/b}$. For liquid

helium four and assuming the classical dispersion relation

$\omega = \sqrt{\dfrac{\sigma}{n_0 m}} \, k^{3/2}$, Atkins [10] calculates

$$\sigma = \sigma_0 - 6.9\times10^{-3}\, T^{7/3} \quad erg\,cm^{-2} \ (T \text{ in } {}^\circ K) \tag{41}$$

In [1] and [8] the approximations resulted in a linear dispersion

relation for intermediate k. The numerical result of [1] is

$$\omega \simeq \dfrac{C_{ph}}{2}k \simeq 120 k \quad s^{-1} \ (k \text{ in } m^{-1})$$

Substituting this in (40) yields

$$\sigma = \sigma_o - 3.2 \times 10^{-3} T^3 \quad erg\, cm^{-2} \quad (T \text{ in } {}^\circ K) \qquad (42)$$

These values should be compared with the experimental result of
Atkins and Narahara [9] , for the temperature dependence of the
surface tension of liquid helium four near T = 0. The best fit to
their data was

$$\sigma = 0.373 - 8.1 \times 10^{-3} T^{5/2} \quad erg\, cm^{-2} \quad (T \text{ in } {}^\circ K) \qquad (43)$$

Ignoring the minor differences in the temperature exponent we see from
(41), (42) and (43) that the classical dispersion relation accounts
for 85% of the experimental temperature variation, while the result of
[1] accounts for only 40%. If the temperature dependence of (43) was
exact then from (40) we have that the quantum dispersion relation is

$$\omega \sim k^{4/3} \qquad \text{(assuming that it is the same over the entire}$$

temperature range), instead of $\omega \sim k^{3/2}$. It is also worth

pointing out that the above calculations have assumed that the temperature
dependent part of the surface free energy arises entirely from the
surface excitations. Chang and Cohen ([1] p 1064) state that there
is a small contribution to $\sigma - \sigma_o$ due to the effect of the free
surface on the bulk excitations, and they give a generalisation of
equation (39) to take account of this. There would also be a
contribution from any temperature dependent density variation but
this is negligible for superfluid He^4 , [10] .

5) Vibrational wavefunctions :

In AIII, translationally invariant wavefunctions of the type
$\sum_{i<j} \sum f(r_{ij}) \Phi$ and related translationally variant wavefunctions
of the form $\sum_j f(r_j) \Phi$, are proposed as compressional vibrational
states of spherical nuclei. The $\sum_j f(r_j) \Phi$ wavefunction is of the form (4)

and so should describe a vibration with a velocity potential
$\phi \sim f(r)$, where r is the distance from the centre of mass.
So the "classical" velocity field is purely radial, which agrees
with the result proved in AIII that $\sum_j f(r_j) \Phi$ has zero angular
momentum. The quantum velocity field BI (16) is zero because the
wavefunctions chosen were all real, but this means only that there
is no nett transfer of matter.

An important vibrational state of the form (4) was found to
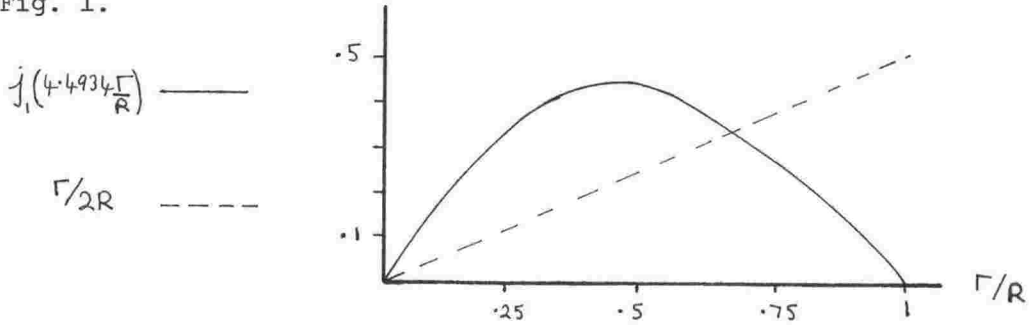be (see AIII and AIV)

$$\sum_j j_0(k r_j) \Phi \qquad , \qquad j_1(kR) = 0 \qquad (44)$$

This wavefunction had the lowest energy in the weak correlation limit
and was closely related to a translationally invariant state, proving
that it is not a spurious state. It was concluded by analogy with
the wavefunction (26) that (44) is a spherical compressional wave,
i.e. a radial phonon. (Note, (44) is the zero angular momentum
projection of (26).) The orthogonality condition $j_1(kR) = 0$
suggested that the lowest state $(kR = 4.4934)$ was not the breathing
mode oscillation, but instead had two regions of motion out of phase
with one another. On page AIII. 44. we speculated that if a wave-
function existed with a similar energy to the state (44) but had a
wavelength such that $kR \simeq \pi/2$ instead of 4.5, then it would
be the breathing mode state.

If we accept the arguments of this section then the "classical"
velocity field of the lowest state is

$$\upsilon(r,t) = \upsilon(r) e^{-i\omega t} , \quad \upsilon(r) \sim j_0'(kr) = -k j_1(4.4934 \, r/R) \qquad (45)$$

Fig. 1.

$$j_1\left(4.4934\frac{r}{R}\right)$$ ————

$$r/2R$$ — — — —

From Fig. 1. we see that (45)'s interpretation of the wavefunction (44) is that there is no out of phase motion in the system, but it is still not the breathing mode oscillation since in this state the surface should have the largest velocity. From Lamb ([2] p 506) we note that the velocity field (45) is that of the longest wavelength sound vibration enclosed in a spherical envelope with a fixed rigid boundary.

In AIV we found that apart from one odd state of very high energy, that all the other wavefunctions proposed in AIII formed a group of related states, higher in energy than (44) and its translationally invariant relative. From the lifetimes it appeared that this group all represented the same physical motion. One of these states was

$$\sum_j (r_j^2 - \overline{r^2})\, \Phi \tag{46}$$

which is the same as $\sum_{i<j}\sum (r_{ij}^2 - \langle r_{12}^2\rangle)\,\Phi$, $(M=0)$. If we interpret the velocity potential as $\phi(r) \sim r^2 - \overline{r^2}$ then the flow field of (46) is $v(r) \sim 2r$. (The dotted line in Fig. 1. is $r/2R$.) So this state would be a good candidate for the breathing mode state if the argument about the nature of (4) was correct. This conclusion pours cold water on the speculation at the end of section AIII where I guessed that some of the low lying 0^+ states of even-even nuclei could be the breathing mode state.

6) Dipole rotational wavefunctions :

In AII I discussed the 1^- giant dipole nuclear wavefunction

$$\Psi_{10} = \sum_{\rho}^{Z} \sum_{n}^{N} z_{\rho n} \Phi = \left[N \sum_{\rho}^{Z} z_{\rho} - Z \sum_{n}^{N} z_{n} \right] \Phi \tag{47}$$

(Z is the number of protons, N the number of neutrons)

Now, Ψ_{10} is a sum of two wavefunctions of the form (4), so it should

describe a flow field with a proton velocity potential $\phi_{\rho} \sim N z_{\rho}$

and a neutron velocity potential $\phi_{n} \sim - Z z_{n}$. This interpretation

implies that Ψ_{10} represents dipole motion in which the protons and

neutrons oscillate out of phase and parallel to the z axis. This is

the same conclusion about the physical nature of Ψ_{10} that was

arrived at by other means in AII.

7) 2$^+$ rotational wavefunctions :

In section AII and [11] a particular type of many-body rotational

excitation was proposed for nuclei :

$$\Psi_{2M} = \sum_{i<j}^{A} \sum_{}^{A} r_{ij}^{2} Y_{LM}(\Sigma_{ij}) f(r_{ij}) \Phi \tag{48}$$

where A is the number of nucleons. With $f = 1$ it was noticed that by fixing

the centre of mass at the origin that (48) reduces to the wavefunction

$$\Psi_{2M}' = \sum_{j}^{A} r_{j}^{2} Y_{LM}(\Sigma_{j}) \Phi \tag{49}$$

which is of the form (4) with velocity potential $\phi \sim r^{2} Y_{LM}(\Sigma)$.

In [11] the wavefunction Ψ_{20}' was generalised to

$$\sum_{j} (3z_{j}^{2} - r_{j}^{2}) f(r_{j}) \Phi \tag{50}$$

and it was proved that in the weak correlation approximation and

assuming Φ to be spherically symmetric, that the optimum wavefunction

of the form (50) is given by $f(r_{j}) = j_{2}(kr_{j}) r_{j}^{-2}$, i.e.

$$\sum_{j} Y_{20}(\Sigma_{j}) j_{2}(kr_{j}) \Phi \tag{51}$$

On the basis that (51) is the L = 2 component of Feynman's phonon

wavefunction (26), we concluded in [11] that both (51) and (49) were

finite system compressional phonon excitations carrying angular

momentum L = 2. This conclusion is supported by analogy with (4)

for the wavefunction (51), since the velocity potential for a classical

sound wave satisfies the equation ($\begin{bmatrix} 2 \end{bmatrix}$ p 496)

$$(\nabla^2 + k^2)\, \phi \; = \; 0 \quad , \tag{52}$$

which has the non singular spherical solutions

$$\phi \sim \mathcal{J}_L(kr)\, Y_{LM}(\Sigma) \tag{53}$$

However, for the wavefunction (49) (i.e. f=1), the analogy with (4)

leads to a different physical interpretation, since the velocity

potential $r^L\, Y_{LM}(\Sigma)$ satisfies the incompressible wave equation

$\nabla^2 \phi = 0$ and represents surface oscillations instead of sound waves.

$\{$ aside: It is not the purpose of the rest of this section to discuss

the properties of Ψ_{2M} and Ψ'_{2M}, which are extensively described

in AII and $\begin{bmatrix} 11 \end{bmatrix}$, but rather to investigate the finite system classical

flow field with velocity potential $\sim r^2\, Y_{2M}(\Sigma)$. The results of this

section are used in AII to help understand the physical nature of the

nuclear wavefunctions (48) and (49). $\}$

In the small amplitude region the oscillation of a spherical drop

of incompressible fluid under the action of gravitational or capillary

forces, is known to be described by the velocity potential (see $\begin{bmatrix} 2 \end{bmatrix}$

pp 450, 474 and $\begin{bmatrix} 3 \end{bmatrix}$ p 239)

$$\phi_{LM} \sim r^L\, Y_{LM}(\Sigma)\, e^{-i\omega t} \tag{54}$$

The oscillations described by (54) cause the surface to be deformed

from its spherical equilibrium state, with a surface radius vector

given by

$$R + \eta\, Y_{LM}(\Sigma) \tag{55}$$

where R is the radius of the undisturbed sphere and η is the amplitude.

The classical velocity field $\underline{V}_{LM} = Re\,\underline{\nabla}\phi_{LM}$ can represent standing oscillations or travelling surface waves, depending on the values of L and M. Now these small amplitude oscillations are in the nature of vibrational modes but for certain values of L and M, ϕ_{LM} also represents rotational modes with a non spherical shape and large amplitude vibrations about non spherical equilibrium. That is, some of the small amplitude vibrational degrees of freedom given by (54) are really rotational degrees of freedom and large amplitude vibrational degrees of freedom. Using group theory it should be possible to identify all these different degrees of freedom but I will restrict my discussion to the case L = 2, which has relevance to the rotational excitations of the deformed even-even nuclei (see AII).

We wish to investigate solutions for irrotational incompressible motion of a liquid contained in an ellipsoidal envelope (or the irrotational rotation of a self-bound ellipsoid). Let the ellipsoidal boundary be given by

$$\frac{x'^2}{a^2} + \frac{y'^2}{b^2} + \frac{z'^2}{c^2} = 1 \tag{56}$$

where (x', y', z') are body fixed coordinates, and consider a rotation about the z axis. The velocity potential is that solution of $\nabla^2\phi = 0$ for which the rotating ellipsoid remains a fixed shape, i.e. the boundary has a velocity whose normal component $\underline{V}.\underline{n}$ is equal to the normal component of the flow at the boundary $\underline{\nabla}\phi.\underline{n}$. In the following calculation the coordinate system is the rotating frame and so the coordinates are time dependent. If the system is rotating about the z axis with constant angular velocity ω, then in the body fixed frame the boundary condition is constant :

$$\underline{V}.\underline{n} = \underline{\nabla}\phi.\underline{n} \tag{57}$$

where $\underline{V} = \underline{\omega} \times \underline{r}' = (-\omega y', \omega x', 0)$. From coordinate geometry we know that if $\underline{P}$ is a point on a surface defined by

$f(x, y, z) = 1$, then $\nabla f(P)$ is normal to every curve on the surface passing through P . So from (56) we have

$$\underline{n} \sim \left(\frac{x'}{a^2} , \frac{y'}{b^2} , \frac{z'}{c^2} \right) \qquad ,$$

and hence the boundary condition (57) is

$$-\omega \frac{y'x'}{a^2} + \omega \frac{x'y'}{b^2} = \frac{\partial \phi}{\partial x'} \frac{x'}{a^2} + \frac{\partial \phi}{\partial y'} \frac{y'}{b^2} + \frac{\partial \phi}{\partial z'} \frac{z'}{c^2} \qquad (58)$$

The solution to (58) (which is also a solution of $\nabla^2 \phi = 0$) is

$$\phi = \omega \left(\frac{a^2 - b^2}{a^2 + b^2} \right) x' y' \qquad (59)$$

When the body fixed frame is at an angle Θ to the lab frame we have

$$\begin{aligned}
x' &= x \cos\Theta + y \sin\Theta \\
y' &= -x \sin\Theta + y \cos\Theta \\
z' &= z
\end{aligned} \qquad (60)$$

So transforming (59) into lab coordinates we have

$$\phi = \omega \left(\frac{a^2 - b^2}{a^2 + b^2} \right) \left\{ \sin2\Theta . \frac{(y^2 - x^2)}{2} + \cos2\Theta . xy \right\} \qquad (61)$$

We can identify this as the real part of ϕ_{22} given by (54) (note, $r^2 Y_{22} \sim (x+iy)^2$)

$$\text{Re} \, \phi_{22} \sim \cos\omega t . (x^2 - y^2) + \sin\omega t . 2xy \qquad (62)$$

provided we have

$$\omega t = 2\Theta + \frac{\pi}{2} \qquad (63)$$

For rigid body rotation $\omega = \dot{\Theta}$ instead of $\omega = 2\dot{\Theta}$ as given by (63). This difference is because after the boundary has rotated 180° the surface wave containing the motion has undergone one complete cycle. The real part of ϕ_{2-2} also reduces to (61), provided (note, $r^2 Y_{2-2} \sim (x-iy)^2$)

$$\omega t = -2\Theta - \frac{\pi}{2} \qquad ,$$

which is just a rotation in the opposite manner to ϕ_{22} . So the

(2,2) and the (2,-2) small amplitude oscillations of a sphere (54),
describe rotational motion of an ellipsoid. In lab coordinates this
motion can be regarded equally as either a finite amplitude
travelling wave on the surface of a sphere, or the rotation of an
ellipsoid of fixed shape.

To calculate the rotational energy, consider the case of an
ellipsoid of revolution symmetric about the x axis (eqn (56) with
b=c) and rotating about the z axis. Writing $n(\underline{r}')$ for the number
density in the body fixed frame ($m\int d\underline{r}'n(\underline{r}') = Nm = M$, the total
fluid mass), we can show by changing to cylindrical coordinates,

$$\frac{1}{N}\int d\underline{r}'n(\underline{r}')x'^2 = \frac{a^2}{5} \quad , \quad \frac{1}{N}\int d\underline{r}'n(\underline{r}')y'^2 = \frac{b^2}{5} \tag{64}$$

The rotational energy is a constant of motion and so it can be evaluated
when the body fixed frame coincides with the lab frame (this is easily
checked by using (60)) :

$$E = \frac{m}{2}\int d\underline{r}'n(\underline{r}')\left(\nabla\phi(\underline{r}')\right)^2$$

$$= \frac{m}{2}\omega^2\left(\frac{a^2-b^2}{a^2+b^2}\right)^2\int d\underline{r}'\,n(\underline{r}')(x'^2+y'^2) \tag{65}$$

$$= \frac{1}{2}\mathcal{I}_{irrot}\,\omega^2$$

where $\mathcal{I}_{irrot}$ is the moment of inertia for irrotational flow :

$$\mathcal{I}_{irrot} = \frac{1}{5}\frac{(a^2-b^2)^2}{a^2+b^2}M \tag{66}$$

For comparison with (66), the rigid body moment of inertia is

$$\mathcal{I}_{rig} = m\int d\underline{r}'n(\underline{r}')(x'^2+y'^2) = \frac{(a^2+b^2)}{5}M \tag{67}$$

The angular momentum of the rotating ellipsoid can also be evaluated :

$$L = m\int d\underline{r}'n(\underline{r}')\left[x'\frac{\partial\phi}{\partial y'} - y'\frac{\partial\phi}{\partial x'}\right] \tag{68}$$

$$= m\omega\left(\frac{a^2-b^2}{a^2+b^2}\right)\int d\underline{r}' n(\underline{r}')(x'^2-y'^2)$$

$$= \mathcal{I}_{irrot}\,\omega$$

From (64) and (68) it follows that

$$E = \frac{L^2}{2\mathcal{I}_{irrot}} = \frac{1}{2}\mathcal{I}_{irrot}\,\omega^2 \tag{69}$$

These results are the same for rigid body rotation except that $\mathcal{I}_{irrot}$ is replaced by the rigid body value (67). So the irrotational rotational energy is smaller than the rigid body rotational energy by the factor

$$\frac{\mathcal{I}_{irrot}}{\mathcal{I}_{rig}} = \left(\frac{a^2-b^2}{a^2+b^2}\right)^2 \tag{70}$$

Lamb ([2] p 147) also gives the solution for a vibrating, stationary, ellipsoidal mass whose changing form is always ellipsoidal. If in (56) the lengths of the axes are changing at the rates $\dot{a},\dot{b},\dot{c}$, Lamb finds the velocity potential

$$\phi = -\frac{1}{2}\left(\frac{\dot{a}}{a}x^2 + \frac{\dot{b}}{b}y^2 + \frac{\dot{c}}{c}z^2\right) \tag{71}$$

where $\nabla^2\phi = 0$ implies $\dot{a}/a + \dot{b}/b + \dot{c}/c = 0$.

Now, the real part of the velocity potential for the $(L,M) = (2,0)$ surface oscillation of the type (54) is (note, $r^2 Y_{20} \sim 2z^2-x^2-y^2$)

$$Re\,\phi_{20} \sim (2z^2-x^2-y^2)\cos\omega t , \tag{72}$$

which can be identified as (71) provided

$$\frac{\dot{c}}{c} = -2\frac{\dot{a}}{a} = -2\frac{\dot{b}}{b} \sim \cos\omega t ,$$

This is satisfied, for example, by

$$c = c_0 e^{-2\alpha\sin\omega t}, \quad a = a_0 e^{\alpha\sin\omega t}, \quad b = b_0 e^{\alpha\sin\omega t}$$

So the (2,0) small amplitude oscillation of a sphere can also describe

large emplitude vibrational motion of an ellipsoid.

The remaining members of ϕ_{2M} ; ϕ_{21} and ϕ_{2-1} , appear to represent only small amplitude travelling waves on the surface of a spherical system. From $r^2 Y_{21} \sim z(x+iy)$ we have

$$Re\,\phi_{21} \sim \cos\omega t.\,zx + \sin\omega t.\,zy \qquad (73)$$

It is probable that certain members of ϕ_{LM} for $L > 2$ also represent rotational motion of a deformed shape, or large amplitude vibrations of deformed shapes. These deformed shapes will have a surface described by Y_{LM} (see (55)) and so will be more complicated than the L = 2 ellipsoid shapes, but their identification may be possible using group theory. (Gustafson [12] , has considered approximate deviations from an ellipsoidal equilibrium and finds that for fixed volume rotations, E/Q_o remains virtually constant, where Q_o is the quadrupole moment.)

To sum up - we have found that of the five small amplitude surface oscillations of a sphere, described by velocity potential ϕ_{2M} $(M = -2,-1, 0, 1, 2)$ two of them ϕ_{22} , ϕ_{2-2} , represent the rotation of an ellipsoid of fixed shape (which is also a finite amplitude travelling wave on a sphere), and another one, ϕ_{20} , can represent the vibration of an ellipsoid (which is a standing wave). The question of whether the nuclear wavefunctions (49) describe such motions is investigated in AII, with some success.

The discussions numbered 1) to 7) above have all been, at least in part, evidence supporting the idea that the quantum wavefunction of the form $\sum_{j} f(r_j) \Phi$ represents a single excitation corresponding

to the classical potential flow field with velocity potential

$$\phi(\underline{r}) \sim f(\underline{r})$$. For the future I note that another excited

mode, with experimental results for liquid helium four, that may

also be described by this type of wavefunction, is a small amplitude

oscillation of a vortex line (see [13] p 204).

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PART   C

PHYSICS   OF   SURFACES

ABSTRACT

I investigate properties of surfaces. Section I presents a
theoretical description of the tension, energy and thickness of a
classical liquid-vapour interface. In section II the classical
results are extended to describe the surface of a quantum system,
namely superfluid helium four. Problems occur for the quantum
system if the correlations arising from the zero-point-motion
of the phonon modes are included in the ground state wavefunction.
Finally, in section III I discuss generalized virial theorems
that give the change in the free energy of a system undergoing
an infinitesimal deformation. For example, a particular
deformation gives the expression used in II, for the surface
tension of a plane quantum surface.

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CONTENTS

I. LIQUID-VAPOUR INTERFACE OF A CLASSICAL FLUID :

A liquid-vapour transition zone has different properties to the bulk phases arising from the variation of density through the zone. In particular it is possible to associate with the transition zone a surface energy and a surface tension. The presence of a positive surface energy is easy to see by comparing an atom in the bulk liquid with one at the surface of the liquid, since there must be a positive addition to the energy of a surface atom due to the absence of the negative potential energy of its missing neighbours. The presence of a surface tension is not so simply understood, but it is easy to detect experimentally, for example if one pulls a wire hoop from out of a liquid surface an obvious tension (pull) will be experienced.

From Newton's law it can be shown ([1] p 291), for a liquid-vapour system in hydrostatic equilibrium, that the pressure normal to the surface, P_N , is constant and the only pressure variation allowed is that of the pressure tangential to the surface, P_T . In fact P_T decreases enormously in the interface region changing into a tension. This is shown schematically in Fig. 1., where the magnitude of the drop in pressure at the surface, $-P_S$, is of the order of $10^3 . P_0$ for an average example, like water away from its critical point. ([1]).

Fig. 1.
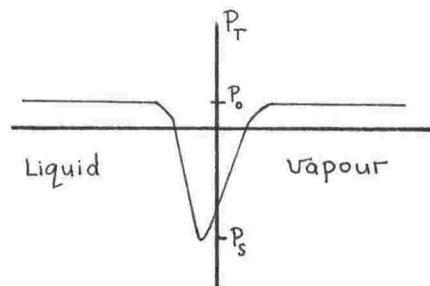
(The variation of tangential pressure through the interface, from [1] p 293)

So if we take any line in the surface of unit length, there acts a force σ with which the liquid on one side pulls the liquid on the other side. This is one of two ways in which surface tension, σ, is defined and is called the mechanical definition of σ. Note that σ has the units of force per unit length ($dyne\ cm^{-1}$) or energy per unit area ($erg\ cm^{-2}$).

Let us now consider a flat liquid-vapour interface situated in the xy plane, with the liquid occupying the volume $z \leq 0$. It should be noted that physically, in order to get a plane surface, a gravitational field is required, however it is known experimentally that the earth's gravitational pressure gradients can be neglected apart from temperatures extremely close to the critical point ([2] p 421). So it can be assumed that the only effect of gravity is to cause the surface to be plane. For this geometry the mechanical definition of surface tension, as the stress across a strip of unit width normal to the interface, can be written as

$$\sigma = \int_{-\infty}^{\infty} \left(P_N - P_T(z) \right) dz \tag{1}$$

The integrand of the right hand side of (1) is non zero only in the interface (see Fig. 1.), which apart from temperatures near the critical point has a width of the order of $\overset{\circ}{A}$.

For a curved surface in hydrostatic equilibrium there is always an excess pressure on the concave side, given by Laplace's equation (see [3] p 231),

$$\Delta P \ (\equiv P_1 - P_2) \ = \ \sigma \left(\frac{1}{R_1} + \frac{1}{R_2} \right) \tag{2}$$

where R_1 and R_2 are the principle radii of curvature at the given point on the surface (R_k is positive if drawn into medium 1). For a sphere of radius R we see that the pressure inside is greater by

$$2\sigma / R \tag{3}$$

The above can be summarised as -surface tension causes any interface to appear stretched so that it always wishes to contract. From this it follows that in the absence of a gravitational field the shape of a self-bound liquid mass would be spherical, since this shape has the least surface area.

The second definition of surface tension is the <u>thermodynamic definition</u> : σ is the isothermal change in the Helmholtz free energy during the formation (at constant volume) of a unit area of surface (i.e. σ is the free energy per unit surface area). To see the equivalence of the two definitions for σ consider a deformation of a surface such that the change in volume is zero, $\Delta V = 0$, while the area increases from A to A + dA. Then from the mechanical definition of σ, the work done during the deformation is

$$dw = \sigma \, dA \tag{4}$$

Writing the total thermodynamic energy as E, the free energy as F, and the entropy as S, we know from thermodynamics that,

$$
\begin{aligned}
dF &= d(E - TS) \\
&= dE - T\,ds - S\,dT \\
&= -S\,dT + dW \\
&= -S\,dT + \sigma \, dA
\end{aligned}
\tag{5}
$$

So from (5) we have the thermodynamic definition

$$\sigma = \left(\frac{\partial F}{\partial A}\right)_{T,V} \tag{6}$$

From (6) we can write

$$F = F_v V + \sigma A \tag{7}$$

so that σ can be regarded as a surface free energy. We can also define a surface entropy

$$S_s A = -\left(\frac{\partial F_s}{\partial T}\right)_A$$

$$= -\frac{d\sigma}{dT} \cdot A \tag{8}$$

Therefore, introducing ϵ as the surface part of the total energy E per unit surface area, we have using E = F + TS,

$$\epsilon = \sigma - T\frac{d\sigma}{dT} \tag{9}$$

That is, the total surface energy per unit area ϵ , and the surface free energy per unit area σ , are related via the thermodynamic relation (9). At this point it should be noted that when we define surface variables like S_s, ϵ and F_s , we are also defining the position of the surface. To see this consider a plane interface and write the number density profile n(z) as

$$n(z) = \begin{cases} n_\ell + (n_\ell - n_v)\delta f(z) & z < D \\ n_v + (n_\ell - n_v)\delta f(z) & z > D \end{cases} \tag{10}$$

With (10) we have located the surface at z = D, but in doing so we have placed a constraint on $\delta f(z)$. This is because a plane surface quantity Q_s per unit area, is defined as the difference between the total value of Q for the system defined by (10) and $Q_\ell + Q_v$ divided by the surface area, where Q_ℓ and Q_v are the bulk values of Q when taking the system to be two distinct bulk phases. So there must be the same number of particles in the system defined by (10) as in the system defined by $n(z) = \{ n_\ell \ z < D \ , \ n_v \ z > D \}$. This implies

$$\int_{-\infty}^{D} dz(n(z) - n_\ell) + \int_{D}^{\infty} dz(n(z) - n_v) = 0 \qquad ,$$

which, from (10), can be written as

$$\int_{-\infty}^{\infty} dz \ \delta f(z) = 0 \tag{11}$$

The surface location defined by (10) and (11) is called the Gibb's dividing surface of vanishing superficial density of matter, or

sometimes just the equimolecular dividing surface. For a plane interface it is only for this particular definition of the surface (which is also the obvious one), that surface quantities are uniquely defined, so that for example σ can be identified as the surface free energy. If (11) is not satisfied then instead of (9) we have for a plane interface, $\epsilon = \sigma + T S_s + \Gamma \mu$, where μ is the chemical potential and Γ is the surface excess matter, which is non zero unless (11) holds (see reference 20 of [4]).

In the rest of this section I discuss the results of a paper supplied with this thesis [4] , which presents a theoretical description of the tension, energy, and thickness of the surface of a simple classical fluid. The phrase, simple classical fluid, means a single component non-polar fluid that is accurately described by central pairwise forces (e.g. the rare-gas liquids, Ar, Kr, Xe, Ne, as well as liquid oxygen and nitrogen, but not water). That is, the potential energy is assumed to be of the form

$$ U = \sum_{i<j}\sum u(r_{ij}) \qquad , \quad r_{ij} = |\underline{r}_i - \underline{r}_j| \qquad (12) $$

Reference [4] considers a two phase classical system of N atoms inside a box, with a flat liquid-vapour interface parallel to the xy plane. The system was kept finite to ensure mathematical rigor and so contributions from the discontinuities at the walls had to be identified and discarded. Now, an exact treatment of the properties of the interface would require two steps : (1) calculate the unique density profile determined by the experimentally derived potential U , and (2) with this density profile calculate σ and ϵ . In [4] only the second step is carried out and specific results are obtained from general expressions by choosing particular forms for the density profiles. So the surface structure is approximated by simple physical density profiles and not explicitly calculated.

This procedure does not cause serious difficulties for classical systems, but it should be noted for reference in section CII, where the surface properties of a quantum fluid are investigated by extending the classical theory. An important property of classical fluids is that the kinetic energy is isotropic. For an N particle system the kinetic energy is $\frac{3}{2}NT$, i.e. $\frac{3}{2}T$ per particle irrespective of whether the particle is in the bulk or at the surface. Hence the surface properties of a classical fluid are determined by the potential energy alone.

The surface energy theory of [4] is new, while microscopic formulations of surface tension theory go back at least forty years. The first general treatment was due to Kirkwood and Buff ([4] reference 1.) who used the mechanical definition of surface tension to obtain an expression for σ in terms of the pair correlation function. The same expression was later obtained from the thermo-dynamic definition by Buff ([4] reference 2) and MacLellan ([4] reference 3). For the sake of completeness [4] includes a simple derivation of Kirkwood and Buff's formula, before going on to evaluate σ and ϵ for a simple exponential density profile and for a general density in the limit of large surface thickness. These results are then used to obtain the surface thickness and to discuss relations between exponents of critical power laws. There is another theory for σ which leads to a general expression in terms of the direct correlation function. In the final section of [4] this expression is evaluated in the same way as was done for the Kirkwood and Buff formula. The two theories should be equivalent, at least when U is of the form (12), and this is proved for the special case of the low density limit. A full summary of these results is given at the beginning of [4]. For the rest of this section the discussion is in the manner of appendices to reference [4] and is restricted to

new or extended results.

1) Isothermal Deformations :

In section 2 of [4] a general expression is derived for the change (to lowest order) in the total thermodynamic energy of a classical system undergoing a small isothermal deformation :

$$\Delta E = \langle \Delta U \rangle + \frac{1}{T}\{\langle U \rangle \langle \Delta U \rangle - \langle U \Delta U \rangle\} \qquad (13)$$

where $\langle\ \rangle$ is the classical expectation value,

$$\langle A \rangle \equiv \frac{\int \underline{d1}\dots \underline{dN}\, A\, e^{-U/T}}{\int \underline{d1}\dots \underline{dN}\, e^{-U/T}} \qquad (14)$$

There are no kinetic energy terms in (13) because total classical kinetic energy $\frac{3}{2}NT$, is constant in isothermal deformations. Hence $\Delta E = \Delta\langle U \rangle$. It was also shown in [4] that the change in the free energy is, to lowest order in the deformation, given by the first term on the right hand side of (13) :

$$\Delta F = \langle \Delta U \rangle \qquad (15)$$

So from (13), (15) and the thermodynamic relation E = F + TS, we have

$$\Delta S = \frac{1}{T^2}\{\langle U \rangle \langle \Delta U \rangle - \langle U \Delta U \rangle\} \qquad (16)$$

The expression (15) is an example of the generalised virial theorems discussed in CIII. In [4] a particular constant volume deformation was used, together with (12) and (15), to derive the Kirkwood and Buff expression for σ from the thermodynamic definition of surface tension. Now, with U of the form (12), the expression (16) for the change in entropy involves four particle correlations through the term $\langle U \Delta U \rangle$. For this reason the surface energy was not calculated from (13), but instead was found by extracting that part of $\langle U \rangle$ (which involves only two particle correlations when U

is given by (12)) that is proportional to the surface area.

I will now show that (13), (15) and (16) are the classical limit of a general statistical mechanical result. The total energy of a statistical mechanical system at temperature T is of the form

$$E = \sum_{\ell} \epsilon_\ell P_\ell \quad , \quad P_\ell = \frac{e^{-\epsilon_\ell/T}}{\sum_{\ell} e^{-\epsilon_\ell/T}} \tag{17}$$

If we now differentiate E with respect to some parameter λ, at constant temperature (i.e. perform an infinitesimal isothermal deformation), we have

$$\left(\frac{dE}{d\lambda}\right)_T = \sum_{\ell}\left(\frac{d\epsilon_\ell}{d\lambda}\right)_T P_\ell + \sum_{\ell} \epsilon_\ell \left(\frac{dP_\ell}{d\lambda}\right)_T$$

$$= \sum_{\ell}\left\{\left(\frac{d\epsilon_\ell}{d\lambda}\right)_T P_\ell + \frac{1}{T}\left[\epsilon_\ell P_\ell\left(\sum_{\ell}\left(\frac{d\epsilon_\ell}{d\lambda}\right)_T P_\ell\right) - \epsilon_\ell\left(\frac{d\epsilon_\ell}{d\lambda}\right)_T P_\ell\right]\right\}$$

$$= \left[\!\!\left[\left(\frac{dE}{d\lambda}\right)_T\right]\!\!\right] + \frac{1}{T}\left\{E\left[\!\!\left[\left(\frac{dE}{d\lambda}\right)_T\right]\!\!\right] - \left[\!\!\left[\widetilde{E\left(\frac{dE}{d\lambda}\right)_T}\right]\!\!\right]\right\} \tag{18}$$

where $[\![\ ]\!]$ denotes the statistical mechanical average and I have introduced a special notation for the last term in (18),

$$\sum_{\ell}\epsilon_\ell\left(\frac{d\epsilon_\ell}{d\lambda}\right)_T P_\ell \equiv \left[\!\!\left[\widetilde{E\left(\frac{dE}{d\lambda}\right)_T}\right]\!\!\right]$$

, to highlight the connection with its classical value $\left\langle U\left(\frac{dU}{d\lambda}\right)_T\right\rangle$. We see that (18) is the generalised result corresponding to (13).

By differentiating the statistical mechanical expression for the free energy,

$$F = -T\log\left(\sum_{\ell} e^{-\epsilon_\ell/T}\right) \quad , \tag{19}$$

it follows that

$$\left(\frac{dF}{d\lambda}\right)_T = \left[\!\!\left[\left(\frac{dE}{d\lambda}\right)_T\right]\!\!\right] \tag{20}$$

This is the generalised form of the classical result (15).

By comparing (18) and (20) with the relation E = F + TS, we can find a general expression for the change in entropy (compare with (16)) :

$$T^2\left(\frac{dS}{d\lambda}\right)_T = E\left[\!\left[\left(\frac{dE}{d\lambda}\right)_T\right]\!\right] - \left[\!\left[\widetilde{E\left(\frac{dE}{d\lambda}\right)_T}\right]\!\right] \tag{21}$$

This expression can also be proved directly by differentiating the result (see [5] p 118)

$$S = -\sum_\ell P_\ell \log P_\ell \tag{22}$$

and using the condition $\sum_\ell P_\ell = 1$.

2) Fermi function density profiles :

In section 4 of [4] the general expressions for σ and ϵ , in the plane surface approximation $n_2 = n(z_1)n(z_2)\, g(r_{12}, \bar{n})$, were evaluated for a simple exponential variation of the density. It is the purpose of this note to point out that in a paper (supplied with this thesis) on the tension and energy of the surface of superfluid helium four ([6] and CII), the expressions for σ and ϵ are also given for the Fermi function density profile (surface is at z = o) :

$$n(z) = n_v + \frac{(n_\ell - n_v)}{e^{z/\delta} + 1} \tag{23}$$

or

$$\delta f(z) = \frac{sgn(z)}{e^{|z|/\delta} + 1} \tag{24}$$

Note that since $\delta f(z)$ is odd about z = o the surface condition (11) is satisfied for all δ . The classical result for ϵ with this density profile, is just equation (27) of [6] with n_0^2 replaced by $(n_\ell - n_v)^2$ and putting $\emptyset = 0$ (i.e. the kinetic energy term is zero for the classical case). The corresponding classical result for σ is equation (46) of [6] , putting $\emptyset = 0$ and replacing n_0^2 by $(n_\ell - n_v)^2$.

For temperatures at which the density is slowly varying on the microscopic scale, it was found in [4] from a Taylor expansion of the density, that σ and ϵ acquire the limiting forms

$$\sigma_\infty = A(\eta_\ell - \eta_v)^2 / \lambda \qquad\qquad (\,[4]\ \text{eqn 84}) \qquad (25)$$

$$\epsilon_\infty = B(\eta_\ell - \eta_v)^2 \lambda \qquad , \qquad (\,[4]\ \text{eqn 90}) \qquad (26)$$

where A and B are independent of λ (λ is a measure of the surface width). Substituting the Fermi function density (24) into the expressions for A and B given in [4], we find (cf. [6] eqns 29 & 47)

$$\frac{A}{\lambda} = \frac{1}{\delta}\frac{\pi}{45}\int_0^\infty dr\, r^5 g(r, \bar{n})\frac{du(r)}{dr} \qquad (27)$$

$$B\lambda = -2\pi\delta\int_0^\infty dr\, r^2 g(r, \bar{n})u(r) \qquad (28)$$

3) The physical significance of ϵ_∞:

It was noted in [4] that σ_∞ and ϵ_∞ are good approximations for temperatures all the way from the critical point to perhaps even the triple point. To be precise, the exponential density expressions for σ_∞ and ϵ_∞ were very good at $\lambda = d$ (d the hard core diameter, λ defined by eqn (71) of [4]), and the triple point value of λ was in the region $\lambda \simeq 0.6d$. In [6] a simple physical interpretation of ϵ_∞ was found, which I will now outline.

It is shown in section 3 of [6] that for both the exponential density profile and the Fermi function density profile, that the limiting surface energy $A\epsilon_\infty$ can be written as

$$A\epsilon_\infty \simeq -\frac{1}{2}\Delta N\left(\frac{E}{V}\right) \qquad (29)$$

where ΔN is the number of atoms in the interface and E/V is the bulk energy density (for the classical case E is the energy of a bulk phase

at density $\bar{\eta}$). From (29) the physical meaning of ϵ_{∞} is obvious :
the positive surface energy ϵ_{∞} , is due to the loss of approximately
one half of the binding energy per atom for every atom in the surface
layer. For a quantum surface this explanation cannot be complete
because of the contributions to ϵ from the kinetic energy and from
the zero-point motion of the surface modes, but for a classical
liquid the loss of binding energy from atoms at the surface is the
entire physical reason for the presence of a surface energy. So it
is not surprising that ϵ_{∞} is a good approximation for a classical
interface even near the triple point.

4) The surface thickness of simple classical fluids :

In section 8 of [4] the exponential density expression for ϵ_{∞}
together with experimental data on triple point bulk energies, was
used to calculate the surface thickness of the rare-gas liquids Ar,
Kr and Xe, at the triple point. Because ϵ_{∞} was a good approximation
for the total expression ϵ , it was simply assumed that the triple
point energy was $\epsilon_{t} \simeq \epsilon_{\infty}$. However for the corresponding quantum
calculation given in [6] , the relatively worse approximation of
ϵ_{∞} to ϵ resulted in the development of a more accurate method.
So the classical calculations of section 8 of [4] could be improved
in the following manner.

Let us assume, as in [4] , that at the triple point,
$$g(r, \bar{\eta}) = g(r, \eta_{\ell}) \qquad \text{and } \eta_{v} \simeq 0 \text{ so that } (\eta_{\ell} - \eta_{v})^2 \simeq \eta_{\ell}^2 \quad .$$
Then from equation 108 of [4] we have

$$\lambda_{t} = -\frac{4}{3} \frac{1}{\left(\frac{\langle u \rangle_{\ell}}{V_{\ell}}\right)_{t}} \cdot \epsilon_{t} \left(\frac{\epsilon_{\infty}}{\epsilon}\right)_{t} \tag{30}$$

where $\dfrac{\langle u \rangle_{\ell}}{V_{\ell}}$ is the potential energy per unit volume of the bulk

liquid and λ_t is the triple point value of the exponential density surface width parameter. In [4] the surface widths of Ar, Kr and Xe were calculated from (30) by setting $(\epsilon_\infty / \epsilon)_t = 1$. The average result was $\lambda_t / d = 0.63$ with a maximum deviation of less than 4 per cent (note, the rare-gas liquids can all be described by the same type of pair potential differing only in the strength and the core diameter d, and since the strengths scale with the triple point temperatures, the surface widths should scale with d). From Fig. 4 of [4] we see that the ratio $(\epsilon_\infty / \epsilon)_t$ is less than unity so that a more accurate calculation will slightly lower the value of λ_t . To estimate this correction we could use the low density pair correlation function

$$g = e^{-u/T}$$, to calculate the values of $(\epsilon_\infty)_t$ and ϵ_t for (λ / d)

from zero to about 1 (see Fig. 3 of [6]). Despite the approximate g it could be expected that the ratio $(\epsilon_\infty / \epsilon)_t$ would be quite accurate. Then to get the accurate value of λ_t one would plot the calculated ratio of $(\epsilon_\infty / \epsilon)_t$, together with its value found from (30) using experimental data for $\left(\langle u \rangle_e / V_e \right)_t$. The intercept of the straight line found from (30) with the curve $(\epsilon_\infty / \epsilon)_t$ is the required value of λ_t (see Fig. 4 of [6]). Since $(\epsilon_\infty / \epsilon)_t$ is slightly less than unity it is most likely from the results of [4] that the corrected value of λ_t for the rare-gas liquids would lie in the range

$$0.5 \lesssim (\lambda_t / d) \lesssim 0.6$$. The calculation outlined above will be carried out and published in the near future.

Another aspect of the quantum calculation in [6] that is worth using in the classical case, is the introduction of a precise definition of surface width. In [4] the surface width was just taken to be about $\left| (n_e - n_v) / n'(0) \right|$ which is 2λ for the exponential density profile. In [6] it was found that the best (most consistent) definition of surface width is the 10 - 90 thickness t , which is distance in which $n(z) - n_v$ rises from 10% of

$(n_\ell - n_v)$ to 90% of $(n_\ell - n_v)$. This gives for the exponential and Fermi function profiles respectively,

$$t_e = 3.22 \lambda , \quad t_F = 4.39 \delta \tag{31}$$

These widths are the same when $\lambda / \delta = 1.365$, which compares well with result obtained by equating the expressions for ϵ_∞ for the two profiles, namely $\lambda / \delta = 4/3$. So taking $\lambda \simeq 0.6 d$ from $[4]$, we have that the 10 - 90 surface width for the rare-gas liquids at the triple point is

$$t_t \simeq 1.9 d \tag{32}$$

i.e. The surface width is about two core diameters.

In section 7 of $[4]$ we concluded that the critical power laws should hold over the same temperature range as the limiting forms σ_∞ and ϵ_∞ were good approximations, which seemed to be all the way from the critical point to the triple point. In particular the power law

$$\lambda = \lambda_t \tau^{-\nu} , \quad \tau = \frac{T_c - T}{T_c - T_t} \tag{33}$$

should hold from $\tau = 0$ to $\tau = 1$. In reference 8 of $[4]$ the expression (33) was found to hold far from the critical region with $\nu = 0.65$, while in reference 20 of $[4]$ the experimental results for the critical region yielded $\nu = 0.64 \pm .03$. So the calculation (32) together with (33), gives (at least approximately) values for the surface thickness of the rare-gas liquids at any temperature between the triple and critical points.

5) σ and ϵ for curved surfaces :

The calculations of $[4]$ refer to a plane interface, or to a curved surface in the limit of large radius of curvature. A question

that is important for the study of small droplets is, does a curved interface have different expressions for σ and ϵ ? In section AI I found that there was no curvature correction term for the surface energy (at least for an exponential density profile) when the equimolecular dividing surface (i.e. the spherical equivalent of (11)) is chosen to be the location of the interface. That is, for this definition of the surface the expression for ϵ was the same for both a droplet and a plane interface.

However, the surface tension of small droplets has not been defined with respect to the equimolecular dividing surface, but instead a dividing surface called the "surface of tension" is introduced (see reference 20 of [4] and [7]). For spherical surfaces this surface location is smaller than the equimolecular surface radius and is introduced so that the work of formation of a spherical droplet is given by $W = 4\pi R^2 \sigma$ where R is the radius of the "surface of tension". (See [7] page 349). This should be compared with (4) and the plane surface discussion following equation (9). Denoting σ_p as the surface tension of a plane interface it is found that the surface tension of a droplet with respect to the "surface of tension", is of the form (see reference 20 of [4]),

$$\sigma = \sigma_p + \left(\frac{c}{s}\right)_p (c_1 + c_2) + \cdots \tag{34}$$

where $c_1 + c_2$ is the mean curvature and $\left(c/s\right)_p$ is an additional thermodynamic parameter related to the asymmetry of the density profile about the "surface of tension" radius. To a first approximation (34) is, [7]

$$\frac{\sigma}{\sigma_p} \simeq 1 - \frac{2z_0}{R} \quad , \tag{35}$$

where z_0 is the difference between the two dividing surfaces. It is important to note that the density profile should be "centred" on the equimolecular dividing surface, or the number of particles in the

in the system will change as the surface width parameter is varied

(see AI).

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II. THE SURFACE OF A QUANTUM FLUID.

For a classical system with a surface the presence of a positive

surface energy is due entirely to the fact, that because at atom at

the surface has missing neighbours it experiences less negative

potential energy. However, for a quantum system there are two other

factors that influence the surface energy, [1] : (i) a surface atom

is in a region of lower density than a bulk atom, which means a

smoother variation of the wavefunction and so the kinetic energy is

reduced, i.e. there is a negative kinetic energy contribution to the

surface energy of a quantum fluid. (ii) There is a positive addition

to the surface energy arising from the quantum surface excitations

(see also the discussion following equation BII (37)).

The two best known examples of a quantum liquid are, liquid He^3

(which is a Fermi liquid for temperatures above $\approx 10^{-3} k$) and super-

fluid liquid He^4 (which is a bose liquid). Liquid helium is remarkable

in that it remains a liquid for all temperatures down to absolute zero,

provided the pressure is less than about 25 atmospheres. Below the

λ point ($2.17 K$) liquid He^4 transforms into the spectacular

superfluid phase, while liquid He^3 undergoes a similar transition at

very low temperatures in the region of $10^{-3} K$. Most of this section

applies specifically to the bose quantum liquid, superfluid He^4 .

A quantum fluid at absolute zero has some simplifying properties that

a classical liquid cannot have. For example it is a single phase

(i.e. $n_v = 0$), and the surface tension (σ) and the surface

energy per unit area (ϵ), are identical (see CI (9)). Also a system

at T = 0 is in its ground state, which makes He^4 the simplest quantum

liquid to understand theoretically. Note that even though a liquid

at T = 0 is in its ground state there is still a contribution to the

surface energy arising from surface excitations, namely the zero-point

energy of the surface modes, [1] . For temperatures slightly greater than absolute zero it is found in the case of superfluid He^4 , that the majority of the temperature variation of σ comes from the surface modes (see discussion following BII (37)).

In a paper that is reprinted at the end of this thesis [2] , the classical calculations of CI (i.e. [3]) are extended to describe the tension energy and thickness of the surface of liquid He^4. The results of [2] are based on the concept that the wavefunction

$$\prod_{i<j}^{N} f(r_{ij})$$

(1)

describes the ground state of a droplet of liquid He^4 . Arguments supporting this idea are given in section one of [2] . This wavefunction is known to give a good description of the ground state of bulk liquid (i.e. an infinite system) and variational forms for f(r) have been calculated by minimising the total energy. The expressions for σ and ϵ are evaluated by choosing particular f(r) determined from the infinite system. So the calculations of [2] are an attempt to find σ and ϵ directly from the ground state of a droplet and are not variational calculations, since f is determined by minimising the total energy, which is dominated by the bulk energy for macroscopic droplets. Another way of stating this concept is - a finite number of liquid He^4 atoms at T = 0 will form a droplet with a surface energy that is just a consequence of the finiteness of the system, and hence ϵ is determined by the ground state alone. In previous calculations of the surface energy of liquid He^4 the ground state has been approximated by

$$\prod_{i<j} f(r_{ij}) \prod_{k} s(r_k)$$

(2)

instead of (1), where $s(r)$ is treated as a variational parameter. Thus the results of [2] are a zeroth order approach, but are important for this fact alone, since the simplest problem highlights the important

physics. A subtle point that is worth mentioning for reference later on, is that a self-bound droplet is translationally invariant and while the simplest wavefunction (1) is translationally invariant, the variational wavefunction (2) is not. However, by introducing a particular singlet density $n(r)$, the calculations of [2] have effectively broken the translational invariance. An exact calculation of the surface energy of a droplet would first require the determination of the unique density profile from the exact ground state (note that this density profile must be translationally invariant). Most of the expressions for σ and ϵ that are derived in [2] , apply to a flat surface in direct analogy to [3] . The assumption is made that the wavefunction (1) also describes a droplet in the limit of its radius tending to infinity.

If we write the assumed ground state wavefunction (1) in the form

$$\Phi = \prod_{i<j}^{N} \exp \tfrac{1}{2} \phi(r_{ij}) = \exp\left\{\frac{1}{2} \sum_{i<j}^{N} \sum^{N} \phi(r_{ij})\right\} \quad , \quad (3)$$

then the probability density Φ^2 is seen to have a direct analogy with the classical probability density for N atoms interacting via pairwise central forces, namely

$$\exp\left\{-\frac{1}{T} \sum_{i<j}^{N} \sum^{N} u(r_{ij})\right\} \quad .$$

Also, the potential energy of liquid He^4 is well described by a classical two body potential

$$U = \sum_{i<j} \sum u(r_{ij}) \quad .$$

Because of this analogy the quantum expressions for σ and ϵ of reference [2] , follow from the classical results of section CI, i.e. [3] . In order to obtain analytic results it was necessary to make an approximation for the pair density, analogous to equation 34 of [3] , namely

$$n(r_1, r_2) = n(r_1) n(r_2) g(r_{12}) \qquad (4)$$

The assumption that the pair correlation function g is a function of only Γ_{12} , is not true at the surface. However without this approximation analytic results would be virtually impossible. The derivation of the expression for ϵ (the surface part of the expectation value of the Hamiltonian, $\langle H \rangle$), is also given in section AI. The surface tension σ is evaluated from an expression derived in section CIII, which was first given by Toda (1955) and Brout and Nauenberg (1958), (see references in [2]). The difference between the quantum and classical expressions for σ and ϵ is the addition of an extra kinetic energy term. The results of [3] can be used because the kinetic energy term is of the same form as the potential energy term, with integrands involving $\emptyset(r)$ instead of U(r). The two main effects of the kinetic energy term are : (i) it is a negative contribution to σ and ϵ , (ii) it makes the slowly varying density limiting forms, σ_{∞} and ϵ_{∞} , less accurate than the classical results. Because of (ii) the surface thickness was calculated by a more accurate extension of the method used in [3] , as described in note 4 of section CI.

To illustrate the nature of general formulae derived in [2] , the expressions for σ and ϵ at T = O were evaluated by choosing particular forms for the wavefunction i.e. $\emptyset(r)$, the density profile $n(z)$, and by using the simplest pair correlation function, g(r) = exp$\emptyset$(r). As in [3] , the exponential density profile was used for numerical results, although expressions were also given for the Fermi function profile. Numerical results are very sensitive to the choice of pair correlation function, as explained in section 2 of [2] . The choice $g = e^{\phi}$, which is exact in the low density limit but poor for liquid He^{4}, happened to give good results because of cancelling errors. The function $\emptyset$ was chosen from variational calculations for the infinite system (i.e. without a surface), with the possibility of

including a non variational factor arising out of the zero-point-motion
of the bulk phonon modes.

(a) Excluding the phonon factor : The optimum ϕ among the class
$-a\left(\frac{b}{r}\right)^n$, tried by McMillan (1965), (referenced in [2]), is

$$\phi(r) = -2\left(\frac{d}{r}\right)^5 \qquad (5)$$

where d is the hard core diameter of the Lennard-Jones potential.
Note that $\phi(r)$ decays rapidly at large   . With this choice for the
wavefunction very good results were obtained : The expressions for
σ and ϵ were calculated as a function of λ/d , for the exponential
density profile. The surface thickness was found by the method outlined
in note 4 of section CI, yielding a 10 - 90 thickness of $3 \cdot 9 Å$.
This method is more accurate than the values for σ and ϵ since
the only variable that has to be calculated is the ratio ϵ_∞/ϵ ,
which should be relatively insensitive to the approximate nature of
the pair correlation function. From equation 9 of section CI we know
that $\sigma = \epsilon$ at T = 0. From the curves of σ and ϵ versus λ/d
this yielded a 10 - 90 thickness of $3 \cdot 4 Å$, in good agreement with
the accurate method.

(b) Including the phonon factor : In section 5 of [2] the
consequences of including the zero-point-motion of the phonon modes
is discussed. Reatto and Chester [4] , show that these modes imply
an additional term to (5), (i.e. an extra factor in the wavefunction),
that for large r tends to

$$\phi_P = -\frac{b^2}{r^2} \qquad , \qquad b = \left(\frac{mc}{\pi^2 n_0 \hbar}\right)^{1/2} \qquad (6)$$
$$\simeq 2 \cdot 6 Å \text{ for liquid } He^4$$

So at large r $(r \gtrsim 10 Å)$, $\phi \to \phi_P$, which unlike (5) decays
slowly with r . However, the inclusion of ϕ_P led to a $-R^2 \log R$
term in the energy of a droplet of radius R, implying a logarithmic

divergence of σ and ϵ . Reatto and Chester were also able to extend the result (6) to non zero temperatures,

$$\phi(r,T) \rightarrow \phi_p(r,T) = -\frac{b^2}{r^2} \cdot \frac{\pi T r/\hbar c}{\sinh(\pi T r/\hbar c)} \qquad (7)$$

At $T > 0$ ϕ_p caused by log T divergence of σ and ϵ . In both cases, for large enough R or small enough T, the surface energy becomes negative, which is spurious since a droplet or a flat surface would disintegrate if the surface energy was negative. But even worse than this, the phonon term ϕ_p led to strange results for the bulk part of the liquid. Firstly at T = 0, the long range part of the phonon factor raised the bulk energy which means that from a variational viewpoint it should be left out (note that a short ranged contribution could change this). At $T > 0$ the phonon factor implied the existence of a completely unstable temperature region, during part of which ϵ was positive, where the total specific heat would be negative. i.e. The phonon factor led to a negative bulk specific heat.

The above problems implied two alternative conclusions :
(i) the phonon factor ϕ_p is spurious, or (ii) the wavefunction $\prod_{i<j} f(r_{ij})$ is too simple to describe a droplet. These two possibilities are investigated in section 5 of [2] , but it was not possible to decide which one is correct. The rest of this section is devoted to filling in the detail about these two alternatives that is not given in the paper.

1) The zero-point-motion of the phonon modes :

Since the phonon factor caused so many problems, not only with the surface properties, but also in the bulk region, it is worth investigating whether or not it really exists. To begin with I will give a full derivation of the results that lead to equation (6),

for ϕ_p . The derivation holds for any system whose Hamiltonian can be written in the form

$$H = H' + H_{ph} \qquad , \qquad (8)$$

where H_{ph} is the harmonic contribution from the long wavelength phonon modes (phonons, i.e. quantised sound waves, are discussed in note 3 of section BII). So the basic assumption is that the long wavelength phonon modes propogate independently of one another and any other modes. This assumption is supported by strong evidence in the case of liquid He^4 at low temperatures (see [4] p 90), but it may also be valid for many other systems both bose and Fermi. From (8) it follows that the ground state wavefunction is of the form

$$\Phi = \Phi' \Phi_{ph} \qquad , \qquad (9)$$

where Φ' and Φ_{ph} are the ground states of H' and H_{ph} respectively. The approximate wavefunction that gave the good results in reference [2] , i.e. Φ given by (3) and (5), assumed $\Phi_{ph} = 1$, (i.e. $H_{ph} = 0$). To derive H_{ph} we note that because it is the contribution from long wavelength modes we can ignore the structure on the atomic level. That is, we can apply a continuum approximation to the fluid. (See [5] p 56, and reference 5 of BII.) In the continuum approximation we have

$$H_{ph} = \int d\underline{r} \left\{ \tfrac{1}{2} \rho \underline{v}^2 + \epsilon(\rho) \right\} \qquad (10)$$

where ρ is the local mass density, $\underline{v}$ the local velocity, and $\epsilon(\rho)$ the potential energy density. For longitudinal density oscillations (i.e. sound waves) the potential energy is a function of ρ alone. Assuming that the external pressure of the system is zero we have $\left(\frac{\partial \epsilon}{\partial \rho} \right)_0 = 0$, where the subscript zero refers to the mean value. Then in the small amplitude limit, i.e. considering only those states with a slight deviation from the mean density ρ_0 , we can approximate (10) by

$$H_{ph} \simeq \frac{1}{2} \int \underline{dr} \left\{ \rho_0 \underline{v}^2 + (\rho - \rho_0)^2 \left(\frac{\partial^2 \epsilon}{\partial \rho^2} \right)_0 \right\} \tag{11}$$

From the expression for the speed of sound c, $\quad c^2 = \left(\frac{\partial P}{\partial \rho} \right)_0$,

where P is the total pressure $-\left(\frac{\partial E}{\partial V} \right)_S$, it follows, using $(\partial \epsilon / \partial \rho)_0 = 0$,

that (see also [6] p 250 and note that the derivatives are at constant

entropy, since a sound wave is adiabatic)

$$c^2 = \rho_0 \left(\frac{\partial^2 \epsilon}{\partial \rho^2} \right)_0$$

Therefore we can write equation (11) as

$$H_{ph} \simeq \frac{m}{2} \int \underline{dr} \left\{ n_0 \underline{v}^2 + \frac{c^2}{n_0} (n - n_0)^2 \right\} \tag{12}$$

where $\rho_0 = n_0 m$, m the atomic mass. The expression (12) is a

classical Hamiltonian describing small amplitude, long wavelength

sound waves. So we need to quantise the Hamiltonian, i.e. to go from

classical sound waves to phonon modes. Consider a system of N atoms

in a volume V, then to quantise the motion one introduces the

collective (normal) coordinate $n_{\underline{k}}$, defined as the Fourier component

of the number density $n(\underline{r})$:

$$n_{\underline{k}} = \int \underline{dr} \, e^{i\underline{k} \cdot \underline{r}} \, n(\underline{r}) \quad , \quad n(\underline{r}) = \frac{1}{(2\pi)^3} \int \underline{dk} \, e^{-i\underline{k} \cdot \underline{r}} \, n_{\underline{k}} \tag{13}$$

From the quantum mechanical density expression, $n(\underline{r}) = \sum_j \delta(\underline{r} - \underline{r}_j)$,

the left hand side of (13) becomes

$$n_{\underline{k}} = \sum_{j=1}^{N} e^{i\underline{k} \cdot \underline{r}_j} \tag{14}$$

and using periodic boundary conditions to change the integral over $\underline{k}$

to a sum over the discrete levels, the right hand side of (13) is

$$n(\underline{r}) = \frac{1}{V} \sum_{\underline{k}} e^{-i\underline{k} \cdot \underline{r}} \, n_{\underline{k}} \tag{15}$$

Writing $k = |\underline{k}|$, we have from (14) and (15) that

$$n_0 = \frac{1}{V} \int \underline{dr} \, n(\underline{r})$$

$$= \frac{1}{V} \, n_{k=0} \qquad ,$$

where the second line follows because for discrete $\underline{k}$,

$$\frac{1}{V} \int \underline{dr}\, e^{i\underline{k}\cdot\underline{r}} = \delta_{k,o} = \begin{cases} 1 & k=o \\ 0 & k \neq o \end{cases} \tag{16}$$

Therefore we see that

$$\int \underline{dr}\,(n-n_o)^2 = \frac{1}{V^2} \int \underline{dr} \sum_{\substack{\underline{k}\\(k>0)}} \sum_{\substack{\underline{k}'\\(k'>0)}} e^{-i(\underline{k}+\underline{k}')\cdot\underline{r}}\, n_{\underline{k}}\, n_{\underline{k}'}$$

$$= \frac{1}{V} \sum_{\substack{\underline{k}\\(k>0)}} n_{\underline{k}}\, n_{-\underline{k}} \tag{17}$$

To evaluate the first term of (12), we note that the classical sound wave has a velocity potential $(\underline{v}=\underline{\nabla}\phi)$, $\phi \sim e^{i\underline{k}\cdot\underline{r}}$, (see BII) and so we can introduce the collective coordinate $\phi_{\underline{k}}$, in the same manner as $n_{\underline{k}}$, i.e.

$$\phi(\underline{r}) = \frac{1}{V} \sum_{\underline{k}} e^{-i\underline{k}\cdot\underline{r}} \phi_{\underline{k}} \quad , \quad \phi_{\underline{k}} = \int \underline{dr}\, e^{i\underline{k}\cdot\underline{r}} \phi(\underline{r}) \tag{18}$$

Thus

$$\underline{v}(\underline{r}) = \underline{\nabla}\,\phi(\underline{r})$$

$$= \frac{1}{V} \sum_{\underline{k}} e^{-i\underline{k}\cdot\underline{r}}(-i\underline{k}\,\phi_{\underline{k}}) \tag{19}$$

and, using (16)

$$\int \underline{dr}\,\underline{v}^2 = \frac{1}{V^2} \int \underline{dr} \sum_{\underline{k}} \sum_{\underline{k}'} e^{-i(\underline{k}+\underline{k}')\cdot\underline{r}}(-\underline{k}\cdot\underline{k}')\,\phi_{\underline{k}}\,\phi_{\underline{k}'}$$

$$= \frac{1}{V} \sum_{\underline{k}} k^2\,\phi_{\underline{k}}\,\phi_{-\underline{k}} \tag{20}$$

To get $\phi_{\underline{k}}$ in terms of $n_{\underline{k}}$, we use the continuity equation for small amplitude disturbances,

$$\dot{n} + n_o \nabla^2\phi = 0 \qquad , \qquad (\cdot \equiv \partial/\partial t) \tag{21}$$

By integrating both sides of (21) over $\underline{dr}\, e^{-i\underline{k}\cdot\underline{r}}$, we can rewrite the continuity equation as

$$\dot{n}_{\underline{k}} - k^2 n_o \phi_{\underline{k}} = 0 \tag{22}$$

From (20) and (22) it follows that

$$\int \underline{dr}\, \underline{v}^2 = \frac{1}{V} \sum_{\substack{\underline{k} \\ (k>0)}} \frac{\dot{n}_{\underline{k}} \dot{n}_{-\underline{k}}}{k^2 n_o^2} \tag{23}$$

So the normal mode Hamiltonian for phonons has been shown to be,

from (12), (17) and (23)

$$H_{ph} = \frac{m}{2V} \sum_{\substack{\underline{k} \\ (k>0)}} \left\{ \frac{\dot{n}_{\underline{k}} \dot{n}_{-\underline{k}}}{k^2 n_o^2} + \frac{c^2}{n_o} \cdot n_{\underline{k}} n_{-\underline{k}} \right\}$$

$$= \frac{1}{2} \sum_{\substack{\underline{k} \\ (k>0)}} m_{\underline{k}} \left\{ \dot{n}_{\underline{k}} \dot{n}_{\underline{k}}^* + \omega_{\underline{k}}^2 n_{\underline{k}} n_{\underline{k}}^* \right\} \quad , \tag{24}$$

where I have put $m_{\underline{k}} = m/Nk^2$, and used the assumed classical

dispersion relation $\omega_{\underline{k}} = ck$.

$\left\{ \right.$ aside: A subtle point that has been suppressed is the question of

the time dependence of $n_{\underline{k}}$. If we use the second line of (24) we

can take the complex form $n_{\underline{k}} \sim e^{-i\omega_{\underline{k}} t}$ and then H_{ph} is independent

of time. In the classical case it is necessary to take

$\phi \sim Re\; e^{i(\underline{k} \cdot \underline{r} - \omega t)}$. Since $\dot{n}_{\underline{k}} = -i\omega_{\underline{k}} n_{\underline{k}}$,

we see that the two terms of (24) are equal, which corresponds to the

well known classical result that the potential energy density and the

kinetic energy density of a sound wave are idential, ([6] p 250). $\left. \right\}$

The Hamiltonian (24) is a set of harmonic oscillators, so we can

immediately write down the ground state wavefunction of H_{ph} , as

$$\Phi_{ph} = exp\left[-\sum_{\substack{\underline{k} \\ (k>0)}} \frac{m_{\underline{k}} \omega_{\underline{k}}}{2\hbar} n_{\underline{k}} n_{\underline{k}}^* \, G(k, k_c) \right] \tag{25}$$

where $G(k, k_c)$ is introduced to cutoff the sum over $\underline{k}$ for large

values of k, at some wavevector k_c (the limit $k_c \to \infty$ can be taken

later). { aside: The one-dimensional harmonic oscillator Hamiltonian,

$$H = -\frac{\hbar^2}{2m}\frac{d^2}{dx^2} + \frac{m\omega^2 x^2}{2} \quad , \text{ (i.e. } \tfrac{1}{2}m[v^2 + \omega^2 x^2] \quad) ,$$

has the ground state $\psi = e^{-\alpha x^2}$, where $\alpha = m\omega/2\hbar$,

i.e. $H\psi = (\hbar^2 \alpha/m)\psi = \frac{\hbar\omega}{2}\psi$.}

From (25) and (14) we obtain

$$\Phi_{ph} = \exp\left[-\frac{1}{2}\sum_{j<\ell}^{N}\sum_{\substack{\underline{k}\\(k>0)}} \frac{2m_k\omega_k}{\hbar} G(k,k_c).\, e^{i\underline{k}\cdot\underline{r}_{j\ell}}\right] \tag{26}$$

Reatto and Chester [4], make the approximation that by letting the

volume tend to infinity, the discrete sum over $\underline{k}$ can be written as

an integral from zero to infinity. Then taking $G(k,k_c) = e^{-k/k_c}$ and

writing

$$\Phi_{ph} = \prod_{i<j}^{N} \exp\tfrac{1}{2}\phi_p(r_{ij}) \quad , \tag{27}$$

they calculate

$$\phi_p(r) = -\frac{2}{\hbar}\sum_{\underline{k}} m_k\omega_k\, e^{-k/k_c}\, e^{i\underline{k}\cdot\underline{r}}$$

$$\simeq -\left(\frac{2mc}{N\hbar}\right)\frac{V}{(2\pi)^3}\int d\underline{k}\,\frac{e^{-k/k_c}}{k}\, e^{i\underline{k}\cdot\underline{r}}$$

$$= -\frac{mc}{\pi^2 n_0 \hbar}\frac{1}{r}\int_0^\infty dk\, e^{-k/k_c}\sin kr$$

$$= -\frac{mc}{\pi^2 n_0 \hbar}\frac{1}{\left(r^2 + \frac{1}{k_c^2}\right)} \tag{28}$$

For large r we have

$$\phi_p \longrightarrow -\frac{b^2}{r^2} \quad , \quad \left\{ b = \left(\frac{mc}{\pi^2 n_0 \hbar}\right)^{1/2} \simeq 2\cdot 6\,\text{Å for} \atop \text{liquid He}^4. \right. \tag{29}$$

So we have derived equation (6), for the contribution to the ground

state wavefunction of liquid He^4 from the correlations due to the

zero-point-motion of the long wavelength phonon modes. Provided

equation (8) holds (and hence (9)), any system which has long wave-

length phonon excitations should have a factor in the ground state

of the form (27) and (29). Furthermore, it appears that the above

results are part of a general quantum principle : that the ground state

of a system is composed (at least partly) of the zero-point-motion

contributions of all its excited states (see also note 4 of BII).

In note 3 of section BII, I introduced the structure factor S(k)

and the pair correlation function g(r) for phonon excitations, in the

infinite system at T = O :

$$S(k) = 1 + n_o \int \underline{dr} \, e^{-i\underline{k}\cdot\underline{r}} \left[g(r) - 1 \right] \qquad (30)$$

$$g(r) - 1 = \frac{1}{2\pi^2 n_o r} \int_0^\infty dk \, k \sin kr \left[S(k) - 1 \right] \qquad (31)$$

The expectation value of the energy of a single phonon was,

$$E(k) = \frac{\hbar^2 k^2}{2m \, S(k)} \qquad , \qquad (32)$$

and so the assumption that $E(k) \rightarrow \hbar c k$ in the limit of small k

(i.e. assuming the classical dispersion relation to hold in the long

wavelength limit), implied

$$S(k) \longrightarrow \frac{\hbar k}{2mc} \qquad (33)$$

Integrating the right hand side of (31) by parts and using (33), gives

an expression for the long range part of g(r),

$$\left[g(r) - 1 \right] \rightarrow - \frac{1}{2\pi^2 n_o} \cdot \frac{\hbar}{mc} \cdot \frac{1}{r^4} \qquad (34)$$

That is, the presence of phonon modes implies that the long range part

of the ground state pair correlation function is given by (34). When

the ground state wavefunction is of the form $\prod_{i<j} \exp \frac{1}{2} \phi(r_{ij})$ there are approximate formulae giving $\phi(r)$ as a functional in $g(r)$ and $S(k)$. To the lowest order of density it follows from the definition of g that, (cf. eqn 33 of $[3]$)

$$g(r) \longrightarrow \exp \phi(r) \quad , \text{ i.e. } \log g = \phi \tag{35}$$

If we write the exact expression for uniform liquid He^4 as

$$\log g(r) = \phi(r) + \gamma(r) \tag{36}$$

then γ is given approximately by the hypernetted chain (HNC) and Percus-Yevick (PY) formulae (see eqns 69, 70 of $[2]$),

$$\gamma(r) = \begin{cases} P(r) & (HNC) \\ \log[1 + P(r)] & (PY) \end{cases} \tag{37}$$

where

$$P(r) = \frac{1}{2\pi^2 n_0 r} \int_0^\infty dk \, k \sin kr \, \frac{[S(k)-1]^2}{S(k)} \tag{38}$$

Inserting (33) into (38) and integrating by parts yields

$$P(r) = \frac{mc}{\pi^2 n_0 \hbar} \frac{1}{r^2} - \frac{1}{2\pi^2 n_0} \cdot \frac{\hbar}{mc} \cdot \frac{1}{r^4} + \cdots \tag{39}$$

From (36), (37) and (39) we see that both the HNC and PY approximations yield the result (34) for the long range part of $g(r)$, provided

$$\phi(r) \longrightarrow -\left(\frac{mc}{\pi^2 n_0 \hbar}\right) \frac{1}{r^2} = \phi_p \tag{29}$$

Since the proof of (34) relies only on the obvious assumption that the energy of a phonon tends to $\hbar ck$ in the limit of small k, it is much less likely to be incorrect than the proof of (29) for the long range part of ϕ. Thus unless the HNC or PY approximations are incorrect, the zero-point-motion correlations of the phonon modes almost certainly imply that $\prod_{i<j} \exp \frac{1}{2} \phi_p(r_{ij})$ is a factor of the ground state of the infinite system.

It is worth speculating on the possibility that the HNC and PY approximations are incorrect, at least in as much as the first term of (39) may be spurious. Both formulae are derived from an incomplete summation of diagrams in a perturbation analysis. Thus it is possible that the partial summations may be invalid and that if the series was summed completely, that the first term of (39) would disappear. This could be analogous to suggesting that either the approximation (8) or (11) for H_{ph} is invalid and that if the anharmonic terms, or the harmonic terms of higher order in the density variation were introduced then a different result to equation (29) would be found. Note that if in equation (24) for H_{ph} we replaced ω_k by $\hbar \dfrac{k^2}{2mS(k)}$ instead of ck, then (28) would become (for $G(k,k_c) = 1$),

$$\phi(r) = -\frac{1}{2\pi^2 n_0 r} \int_0^\infty dk\, k\left(\sin kr \middle/ S(k)\right)$$

This is very similar to the HNC and PY result (38) for $P(r)$. In a paper by Bogoliubov and Zubarev [7] , the ground state of a system of interacting bosons is calculated as a series of terms of increasing order in accuracy. The zeroth approximation yields a ground state that includes the factor Φ_{ph} given by equation (26) with $G(k,k_c) = 1$ (see eqns 3.12 and 4.6 of [7] and substitute $E(k) = \hbar c k$, $m = N k^2 m_k$, $\omega_k = ck$). This is another derivation of the result (29), for ϕ_p to the zeroth order of accuracy. What is required is a proof that the sum of the contributions from all the higher order terms does not cancel the zeroth order term (29).

The above derivation of ϕ_p was restricted to T = 0, i.e. (6). The $T > 0$ result (7), was derived by Reatto and Chester from an evaluation of the density matrix ([4] p 91). The diagonal part of the phonon factor in the density matrix was shown to have the form of a product wavefunction $\prod_{i<j} \exp\frac{1}{2} \phi_p(r_{ij}, T)$ with $\phi_p(r, T)$ given by (7). The non diagonal part represents phonon-phonon interactions

which are assumed to be negligible, i.e. the temperature must be low enough so that the phonons can be regarded as non-interacting. I will now give another proof of Reatto and Chester's result, using the HNC/PY formulae (36) - (38) and the finite temperature structure factor. In the small k region the structure factor for liquid He^4, for $T > 0$, is, ([8] p 1203)

$$S(k,T) = \frac{\hbar k}{2mc} \coth\left(\frac{\hbar c k}{2T}\right) \tag{40}$$

Since $\coth z \to 1/z$ as $z \to 0$ we have

$$S(0,T) = \frac{T}{mc^2} = n_0 T \chi_s \tag{41}$$

where χ_s is the adiabatic compressibility. It can be shown that as $T \to 0$, $\chi_s \to \chi_T$, the isothermal compressibility, so that (41) reduces to the result of equation (d) of appendix AI (1). Also, since $\coth z \to 1$ as $z \to \infty$, we recover (33) in the limit T = 0 :

$$S(k,0) = \frac{\hbar k}{2mc} \tag{33}$$

Because $\phi_p(r,T)$ is exponentially damped for $r \gtrsim \hbar c/\pi T$ (= $5.76\,\text{Å}$ at $1K$, for liquid He^4), see equation (7), we cannot use the same method to evaluate $P(r,T)$ that led to (39). This is because integrating by parts will only lead to terms decaying slower than $r^{-\infty}$, i.e. if we insert (40) into (38) and integrate by parts we will find every term to be zero. So we need to integrate (38) explicitly with S(k) replaced by $S(k,T)$. Now in the small k limit

$$k\frac{[S(k,T)-1]^2}{S(k,T)} \longrightarrow \frac{k}{S(k,T)} = \frac{2mc}{\hbar}\tanh\gamma k \;,\; \left\{ \gamma = \frac{\hbar c}{2T} \right. \tag{42}$$

To get the asymptotic form of $P(r,T)$ (i.e. large r), we can use (42) in (38), since for large r the presence of $\sin kr$ in the integrand means that only the small k part will contribute. Inserting (42) into (38) we have

$$P(r,T) \rightarrow \left(\frac{mc}{\pi^2 n_o \hbar}\right) \frac{1}{r} \int_0^\infty dk \sin kr \tanh \gamma k \qquad , \quad \gamma = \frac{\hbar c}{2T} \quad (43)$$

We can evaluate the integral as follows :

$$I \equiv \int_0^\infty dk \sin kr \tanh \gamma k$$

$$= \frac{1}{2} \mathfrak{I}_m \left\{ \int_{-\infty}^\infty dk \, e^{ikr} \tanh \gamma k \right\}$$
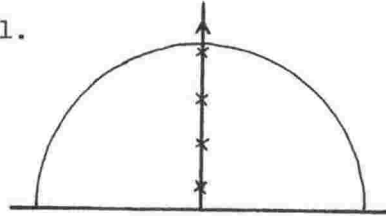
$$\equiv \frac{1}{2} \mathfrak{I}_m \{J\} \qquad (44)$$

To calculate J we integrate over the contour of Fig. 1.

which has simple poles

at $\cosh \gamma k = 0$, i.e.

$$\gamma k = (n + \frac{1}{2})\pi i \, , \quad n = 0,1,2,\cdots$$

The residue at $(n + \frac{1}{2})\pi i$ is

$$\frac{1}{\gamma} e^{-\frac{\pi r}{2\gamma}} e^{-n\frac{\pi r}{\gamma}}$$

Fig. 1.

Therefore

$$J = \frac{2\pi i}{\gamma} e^{-\frac{\pi r}{2\gamma}} \sum_{n=0}^\infty e^{-\frac{n\pi r}{\gamma}}$$

$$= \frac{\pi i}{\gamma} \frac{1}{\sinh\left(\frac{\pi r}{2\gamma}\right)} \qquad (45)$$

From (43) - (45), we have for the long range part of $P(r,T)$

$$P(r,T) \rightarrow \left(\frac{mc}{\pi^2 n_o \hbar}\right) \frac{1}{r^2} \cdot \frac{\pi r T/\hbar c}{\sinh\left(\pi r T/\hbar c\right)} \qquad (46)$$

Equations (36), (37) and (46), give the result (7) for $\phi_p(r,T)$.

In section 5 of $[2]$, the inclusion of $\phi_p(r,T)$ led to a negative

bulk specific heat for a droplet with the ground state wavefunction

$$\prod_{i<j} \exp \frac{1}{2} \phi(r_{ij})$$. For the uniform (infinite) system the

above results have shown that unless the HNC or PY equations are incorrect, the factor $\prod_{i<j} \exp \frac{1}{2} \phi_p(r_{ij})$ is almost certainly a real part of the ground state of liquid He^4.

One possible solution to the problems of section 5 of $\left[2\right]$, could be that for a finite droplet ϕ_p changes enough to remove the spurious results. I will investigate this possibility for a finite system at T = O, where the inclusion of the infinite system ϕ_p led to negative divergences in σ and ϵ. One thing that is immediate is that $\phi_p(r)$ will no longer be infinite ranged since the system is finite. Also, because a finite system has a discrete set of states (k values), the replacement of $\sum_k$ by $\int dk$ in the derivation of ϕ_p is at best an approximation (cf. eqn (28)). From (26) we have, (putting $G(k,k_c) = 1$)

$$\Phi_{ph} = \prod_{i<j}^{N} \exp \frac{1}{2} \phi_p(r_{ij}) \tag{47}$$

where

$$\phi_p(r) = -\left(\frac{2mc}{N\hbar}\right) \sum_{\substack{k \\ (k>0)}} \frac{e^{ik \cdot r}}{k} \quad , \quad k = |k| \tag{48}$$

For a finite system k has discrete values and so the exact solution requires an explicit evaluation of the sum. Another important point is that the derivation of (48) has assumed cubic symmetry and not spherical symmetry. This is because the normal coordinate η_k was taken to be $\sum_j^N e^{ik \cdot r_j}$, which applies to cubic symmetry. Taking the system to be a cube of length L and using periodic boundary conditions, the discrete values of $k = (k_x, k_y, k_z)$ are given by

$$k_x, k_y, k_z \quad \in \left\{ \frac{2n\pi}{L} \quad , \quad n = 0, \pm 1, \pm 2, \cdots \right\} \tag{49}$$

So for a finite cube we need to evaluate

$$\phi_p(r) = -\left(\frac{2mc}{N\hbar}\right) \sum_{n_x=-\infty}^{\infty} \sum_{\substack{n_y \\ =-\infty}}^{\infty} \sum_{\substack{n_z \\ =-\infty}}^{\infty} \frac{e^{i\frac{2\pi}{L}r n_x} e^{i\frac{2\pi}{L}r n_y} e^{i\frac{2\pi}{L}r n_z}}{\left(\frac{2\pi}{L}\right)\sqrt{n_x^2+n_y^2+n_z^2}}$$

$$\underset{\substack{(excluding \\ n_x=n_y=n_z=0)}}{} \tag{50}$$

I have been unable to calculate the triple sum involved in (50), but it is possible to give a plausible guess as to what form it should take. Firstly, at T = 0 the phonon correlations will extend over the entire system, by analogy with the infinite ranged correlations present in the infinite system. Also, the result of (50) for ϕ_p cannot be completely spherically symmetric because of the walls. However, it is most likely that deep inside the system the expression (50) will tend to the infinite system result (29).

As an aside it is worth pointing out that for the one dimensional (1-D) system ϕ_p can be found exactly, although of course it is completely different to the three dimensional result. In 1-D you cannot evaluate (48) by taking the infinite system limit because of the k^{-1} singularity, so the sum over k must be evaluated explicitly. If L is the length of the 1-D system, we have from (48), using periodic boundary conditions,

$$\phi_p(r) = -\left(\frac{2mc}{N\hbar}\right)\cdot\frac{L}{2\pi}\sum_{\substack{n=-\infty \\ (\neq 0)}}^{+\infty}\frac{e^{i\frac{2\pi}{L}rn}}{|n|}$$

$$= -\left(\frac{2mc}{\pi n_o\hbar}\right)\sum_{n=1}^{\infty}\frac{\cos\left(\frac{2\pi r}{L}n\right)}{n}$$

$$= \left(\frac{2mc}{\pi n_o\hbar}\right)\log\left[2\sin\frac{\pi r}{L}\right] \qquad\qquad \text{, (see [8])} \quad (51)$$

This result is given by Reatto and Chester ([4] p 96). They note that $\prod_{i<j}\exp\tfrac{1}{2}\phi_p(r_{ij})$ where ϕ_p is given by (51), is the exact ground state of a 1-D system of point impenetrable bosons. For this particular system $\left(2mc/\pi n_o\hbar\right) = 2$.

We have already seen that the long range part of $\phi_p(r)$ is determined by the small k values. So the major difference between a finite system ϕ_p and the infinite system ϕ_p should be accounted for

by replacing the $\sum\limits_{k}$ by $\int \underline{dk}$, but integrating up from the minimum k value instead of zero. For comparison with section 5 of $\boxed{2}$ I will consider a droplet of radius R, writing the minimum value of k as k_{min} (by analogy with (49) we could guess $k_{min} = 2\pi/R$). Then instead of (28) we now have

$$
\begin{aligned}
\phi_p(r) &\simeq -\left(\frac{mc}{\pi^2 n_0 \hbar}\right) \frac{1}{r} \int_{k_{min}}^{\infty} dk\, e^{-k/k_c} \sin kr \\
&= -\left(\frac{mc}{\pi^2 n_0 \hbar}\right) \frac{1}{r} \mathcal{I}_m\left\{ \int_{k_{min}}^{\infty} dk\, e^{k(ir - 1/k_c)} \right\} \\
&= -\left(\frac{mc}{\pi^2 n_0 \hbar}\right) \frac{e^{-k_{min}/k_c}}{(r^2 + 1/k_c^2)} \left[\cos k_{min} r + \frac{\sin k_{min} r}{k_c r} \right]
\end{aligned}
$$
(52)

Taking $k_c \to \infty$ and choosing $= 2\pi/R$, we see that the long range part of ϕ_p is

$$
\phi_p(r) \longrightarrow -\frac{b^2}{r^2} \cos\left(\frac{2\pi r}{R}\right) \qquad , \qquad b^2 = \left(\frac{mc}{\pi^2 n_0 \hbar}\right)
$$
(53)

This result should be compared with (29). For $r/R \ll 1$ (53) reduces to the infinite system value (29). The conclusion is that the only difference between a finite system and an infinite system, for the contribution to the ground state wavefunction from the zero-point-motion of the phonon modes, occurs for interparticle separations of the order of the size of the system. To get the corresponding result to equation 55 of $\boxed{2}$, for the direct contribution to the energy arising out of the zero-point-motion correlations, we require

$$
\Delta E = \frac{2}{3}(\hbar c n_0) R^3 \int_{2a}^{2R} dr \left[1 - \frac{3}{2}\left(\frac{r}{2R}\right) + \frac{1}{2}\left(\frac{r}{2R}\right)^3 \right] \frac{d}{dr}\left(r^2 \frac{d}{dr}\left(\frac{\cos k_{min} r}{r^2} \right) \right)
$$
(54)

Since k_{min} is about $\frac{2\pi}{R}$, it follows for $R \gg a\, (\simeq 10\,\text{Å})$, that

$$\Delta E \simeq \frac{2}{3} (\hbar c n_0) R^2 \left\{ \frac{R}{a} - \frac{3}{2} \int_{2a}^{2R} dr \frac{\cos k_{min} r}{r} + C + O\left(\frac{1}{R}\right) \right\}$$

(55)

where C is a constant of order unity depending on the value of k_{min} .
From (55) we see that the "surface energy" at T = O still contains the
divergent term, since

$$\epsilon = -\frac{\hbar c n_0}{4\pi} \left\{ \int_{2a}^{2R} dr \frac{\cos k_{min} r}{r} + \frac{2}{3} C \right\}$$

(56)

and as $R \to \infty$ the "surface energy" diverges to minus infinity as
before, namely

$$\epsilon \to -\frac{\hbar c n_0}{4\pi} \log\left(\frac{R}{a}\right)$$

(57)

So the more accurate result (53) for ϕ_ρ , does not solve the problem
of the divergence of σ and ϵ at T = O.

To sum up - the problems caused by the inclusion of the infinite
system ϕ_ρ when calculating the surface properties of a droplet, will
almost certainly not disappear if the exact finite system ϕ_ρ was used.
The derivation of ϕ_ρ (eqns (6) and (7)), did not seem to contain any
hidden errors or wrong assumptions. However, if the HNC or PY formulae
were incorrect then this might change. For example, if the calculation
was done to all orders it might just be that the zeroth term, ϕ_ρ , is
cancelled by part of the sum of all the higher order terms. The most
serious problem caused by the inclusion of the long range phonon factor,
in section 5 of [2] , is that it gives a negative contribution to the
bulk specific heat that would dominate at low temperatures ($T \le 10^{-2} k$
for a droplet of millimetre size). This problem must also be present
for the infinite system when the wavefunction is taken to be of the
form $\prod_{i<j} \exp \frac{1}{2} \phi(r_{ij})$. From section 5 of [2] we see that the

inclusion of the term (7) in the ground state wavefunction implies a negative T^3 dependence of the specific heat that dominates the normal positive T^3 term due to thermal excitation of phonons, for $T \lesssim 1K$. This result must be spurious, since a region of negative specific heat is unstable and could never be obtained in nature. It should be pointed out however, that the proof of this negative specific heat relies on an approximation that may be invalid. Namely, the contribution to the bulk energy per atom from the long range correlations of the phonon modes at $T > 0$, is assumed to be

$$ E/N = -\frac{\pi n_0 \hbar^2}{2m} \int_a^\infty dr\, r^2 \nabla^2 \phi_\rho \qquad , \quad (a \simeq 10 \text{\AA}) \qquad (58) $$

where ϕ_ρ is given by (7). That is, the $T > 0$ expression for ϕ_ρ is inserted into the T = 0 expectation value of the energy. To be completely correct the statistical mechanical expectation value should be used (see BII (17)). However there is a good physical reason for believing that this approximation is not important for calculating the contribution from the zero-point-motion of the phonons, as distinct from the normal statistical term due to the thermal excitation of phonons. The argument is given in section 5 of [2] and is as follows : the term $h(r) = -\frac{\hbar^2}{4m} \nabla^2 \phi_\rho$ tends to $\frac{\hbar^2 b^2}{2m} \frac{1}{r^4}$ at T = 0 and hence can be regarded as an effective repulsive r^{-4} inter- action. From (7) we see that at $T > 0$ h(r) is exponentially damped, i.e. it is a repulsive r^{-4} interaction which is screened thermally. Because the higher the temperature the greater the screening, the energy due to the long range phonon correlations decreases with temperature, i.e. the specific heat is negative. This argument also suggests a possible flaw in the reasoning of section 5 of [2] . Namely, a short ranged contribution to $\phi_\rho(r,T)$ would include an effective attractive interaction (eg. (28) for $r \lesssim 1/k_c$). Thus it is possible that the temperature variation due to the short range part

of ϕ_ρ might cancel the negative specific heat coming from the long range part. However, it is very difficult to see how the short ranged correlations could remove the divergencies in σ and ϵ (in this connection it might pay to be beware of the approximation (4) for the pair density).

If the phonon factor $\prod_{i<j} \exp \frac{1}{2}\phi_\rho(r_{ij})$ is a true part of the exact ground state of liquid He^4 (or perhaps any system), then it would imply that the wavefunction $\prod_{i<j} f(r_{ij})$ is too simple to describe the ground state of a droplet. This may also apply to the infinite system at $T > 0$, as discussed above. This brings us to the question of whether a better ground state wavefunction will remove the problems associated with the phonon factor.

2) Variational Wavefunctions :

It was shown in section 5 of $\begin{bmatrix} 2 \end{bmatrix}$ that by using the variational wavefunction

$$\Psi = \prod_{i<j} \exp \frac{1}{2}\phi(r_{ij}) \prod_{k} \exp \frac{1}{2} t(r_k) \qquad , \qquad (59)$$

that the problems associated with the phonon factor are either removed, or are shifted to a term involving only the density profile. The function $t(r)$ can either be optimised variationally, or it can be eliminated by means of the Yvon equation

$$\underline{\nabla}_1 n(r_1) = n(r_1) \underline{\nabla}_1 t(r_1) + \int \underline{dr}_2 \, n(r_1, r_2) \, \underline{\nabla}_1 \phi(r_{12}) \qquad , \quad (60)$$

in which case the density profile is optimised. The inclusion of an extra variational parameter $t(r)$, will lower the total energy and so will probably give a better description of the surface, but it is hard to see how it could remove divergences in σ and ϵ . Chang and Cohen $\begin{bmatrix} 10 \end{bmatrix}$, who first suggested the method based on (59) and (60),

state that a long range behaviour of $\emptyset(r)$ (e.g. $\phi_\rho(r)$) necessitates a long range variation of $t(\underline{r})$ into the bulk, to avoid unphysical behaviour of the density. The expectation value of the Hamiltonian (ϵ is the surface part of $\langle H \rangle$) for the wavefunction (59), is

$$\langle H \rangle = \frac{1}{2} \int d\underline{r}_1 \, d\underline{r}_2 \, n(\underline{r}_1, \underline{r}_2) \left\{ u(r_{12}) - \frac{\hbar^2}{4m} \nabla_1^2 \emptyset(r_{12}) \right\}$$
$$- \frac{\hbar^2}{8m} \int d\underline{r} \, n(\underline{r}) \, \nabla^2 t(\underline{r}) \tag{61}$$

It is worth investigating two other attempts to calculate ϵ , based on wavefunctions of the form (59), that were not as successful as Chang and Cohen's, to see where they went wrong :

(1) Bowley [11] , used the wavefunction (59) but did not use (60). He assumed that $n(\underline{r}) = n_0 \exp t(\underline{r})$, which is plausible considering the definition of the number density,

$$n(\underline{r}_1) = N \frac{\int d\underline{r}_2 \ldots d\underline{r}_N |\Psi|^2}{\int d\underline{r}_1 \ldots d\underline{r}_N |\Psi|^2} \tag{62}$$

This assumption reduces the last term in (61) to

$$\frac{\hbar^2}{8m} \int d\underline{r} \, n(\underline{r}) \left(\nabla \log n(\underline{r}) \right)^2 \tag{63}$$

which turns out to be a small virtually irrelevant term, for physical density profiles of liquid He^4. So Bowley's assumption results in the same answer as obtained in [2] for the $\prod_{i<j} \exp \frac{1}{2} \emptyset(r_{ij})$ wavefunction, apart from the small positive definite term (63). Hence the inclusion of ϕ_ρ leads to a divergence of ϵ , as before. The parametrisation $n(\underline{r}) = n_0 \exp t(\underline{r})$ is bad because it is equivalent to ignoring the second term of (60), and it is shown in section 5 of [2] that if the exact YBon equation is used then not only is (63) obtained, but also another term arises which cancels the divergent part of the first term of $\langle H \rangle$. In fact, if the HNC and PY equations are used, all the terms involving ϕ_ρ cancel in the

expression for ϵ .

(2) Lekner [12] , attempted to calculate ϵ for a flat surface in the xy plane, by using the wavefunction

$$\Psi = \prod_k S(z_k) \Phi \quad , \tag{64}$$

where Φ was taken to be the exact ground state of the infinite system, instead of approximating it by $\prod_{i<j} \exp \frac{1}{2}\phi(r_{ij})$ as in (59). Lekner assumed that the surface energy was given by $\Delta E = E - E_o$, where

$$H\Phi = E_o \Phi \qquad \text{and E is the expectation value of the Hamiltonian}$$

in the state Ψ . He calculates

$$\Delta E = \frac{\hbar^2}{2m} \int dr\, n(z) \left[s'(z) / s(z) \right]^2 \tag{65}$$

Substituting $S = e^{\frac{1}{2}t}$ and integrating (65) by parts we see that ΔE is just the last term in (61). The attempt to treat $S(z)$ as a variational parameter failed because ΔE is minimised by taking $S(z) \to 1$, since (65) is positive definite. By comparing (61) and (65) we see that the problem with Lekner's method is that the surface energy is contained within the two body part of the wavefunction, i.e. $\prod_{i<j} \exp \frac{1}{2} \phi(r_{ij})$, which has been left out of (65), (i.e. it is part of E_o). We cannot use the result (65) to prove that the optimum wavefunction of the form (59) is given by $e^{\frac{1}{2}t} = 1$, since by taking Φ to be the exact ground state (which means that E_o must be the minimum energy), Φ will still be better described by (59) than by $\prod_{i<j} \exp \frac{1}{2} \phi(r_{ij})$.

From equation (68) of [2] we see that the wavefunction (59) will also remove the $\nabla^2\phi_\rho$ term in the infinite system expression for $\langle H \rangle$. That is, we can use (60) to eliminate $t(r)$ in favour of $n(r)$ and then put $n(r) \to n_o$ so that the surface contribution (63) becomes zero. From (60) this implies that for the infinite system

$$\nabla_1 t(r_1) = -4\pi n_o \int_0^\infty dr_2\, r_2^2\, \nabla_1 \phi(r_{12}) \quad , \tag{66}$$

which has the solution

$$t(\underline{r}_1) \ = \ -4\pi n_o \int_0^\infty dr_2 \, r_2^2 \, \phi(r_{12}) \ + \ const. \tag{67}$$

Since the right hand side of (67) is translationally invariant the only physical solution would be $t(\underline{r}_1)$ = constant, however by putting $\phi(r_{12}) = \phi_\rho \sim 1/r_{12}^2$, we see that the right hand side of (67) is divergent. [aside: Infinity is a constant but I do not regard this as satisfactory.] This throws doubt on the method of Chang and Cohen because if (60) does not hold for their chosen density profile when ∅ includes ∅p, then their argument breaks down, and is only transforming the spurious results of ∅p from a divergence in $\in$ to an inability to satisfy the Yvon equation.

This brings us to the conclusion of [2] , that until the consequences of ∅p on the density profile are explicitly calculated we cannot be sure whether the wavefunction $\prod_{i<j} exp\frac{1}{2} \phi(r_{ij})$ is too simple, or whether it is the ∅p factor that is incorrect. The density profile resulting from a given wavefunction is usually calculated from the Yvon equation (60), (see for example [13]), so we can put it another way and conclude that we must first prove that the Yvon equation makes sense if ∅ varies as $1/r_{12}^2$. This difficult problem will be attempted in the future, but I will make some short comments about it, that are also applicable to the classical surface structure problem, $\phi \longleftrightarrow \frac{-u}{T}$. Firstly, we note that a self bound droplet is translationally invariant and so we require $n(\bar{r})$ where $\bar{r}$ is relative to the centre of mass. However, the definition (62) for $n(\underline{r}_1)$, from which the Yvon equation is derived by differentiation, is not satisfactory in this regard. In fact, if we take $|\Psi|^2$ to be translationally invariant then the right hand side of (62) is translationally invariant and hence the only solution to (62) will be $n(\underline{r})$ a constant. So what is required is a translationally invariant

density definition, capable of describing a droplet. Also, in order
to calculate the exact density profile, the zero-point-motion
correlations of the surface modes would have to be included in the
ground state (see below). We note that Chang and Cohen's wavefunction
(59) is not translationally invariant, but this may be necessary once
the translationally variant density $n(\underline{r})$ is introduced, via the
approximation

$$n(\underline{r}_1, \underline{r}_2) = n(\underline{r}_1) n(\underline{r}_2) g(\underline{r}_{12}) \tag{4}$$

Bowley, whose method of calculating ϵ is virtually equivalent to
$[2]$, had a different explanation of why the phonon factor caused a
divergence in ϵ . He suggested that the correlations arising out of
the zero-point-motion of the surface modes cancelled the phonon mode
correlations deep inside the liquid, and so removed the divergent $\emptyset p$
term. In note 4 of section BII I pointed out that the presence of
surface modes implies a factor $\prod_{i<j} \exp \frac{1}{2} \phi_s (\underline{r}_i, \underline{r}_j)$ in the ground
state, in direct analogy with the phonon modes leading to the factor
$\prod_{i<j} \exp \frac{1}{2} \phi_p (\underline{r}_{ij})$. Bowley's argument was that because the
surface mode energy $\epsilon_s \sim \hbar k^{3/2}$ was lower than the phonon energy
$\epsilon_p \sim \hbar k$ for small k, that the long range part of $\emptyset_s$ was even more
slowly varying than $\emptyset p$. Because of this he suggested that the phonon
factor was cancelled. However, this idea is almost certainly incorrect
for two reasons : (1) The surface wave term $\phi_s(\underline{r}_i, \underline{r}_j)$ will contain the
symmetry of the surface and will vary the slowest parallel to the
surface, while $\phi_p (\underline{r}_{ij})$ is a bulk function. Hence even if $\emptyset_s$
cancelled $\emptyset p$ at the surface it could not do so deep inside the fluid.
(2) Since $\emptyset_s$ is even more slowly varying than $\emptyset p$ it should give an
even worse divergence of ϵ , using the methods of $[2]$ and Bowley.

It is worth considering just how bad an approximation it is to
neglect the surface mode factor in the ground state when calculating ϵ ,

either by the methods of sections one to four of $\begin{bmatrix} 2 \end{bmatrix}$, or by the
variational method of Chang and Cohen. At the very beginning of this
section I noted that there were three physical contributions to the
surface energy of a quantum fluid : (i) a positive potential energy
term, (ii) a negative contribution from the kinetic energy and (iii) a
positive addition arising out of the zero-point-motion of the surface
modes. Atkins $\begin{bmatrix} 1 \end{bmatrix}$, calculates that the total zero point energy of
the surface modes is of the order of the total surface energy, so the
question is, why did $\begin{bmatrix} 2 \end{bmatrix}$ and Chang and Cohen get good results by
neglecting the zero-point motion factor in the ground state? The
answer must be that the main physical effect of the presence of
surface modes is on the density profile. So, by choosing specific
forms for the density profile and varying the surface width parameter,
as in $\begin{bmatrix} 2 \end{bmatrix}$ and Chang and Cohen, most of the effects of the surface
modes are taken into account. However, it would be necessary to
include the surface mode factor if the ground state wavefunction was
used to calculate the density profile, as would be necessary for an
exact determination of ϵ .

To conclude - the question of whether the phonon term ϕ_ρ is
incorrect, or whether the wavefunction $\prod_{i<j} f(r_{ij})$ is too simple,
will probably have to wait until the density profiles that are implied
by the wavefunctions (3) and (59) can be calculated. If ϕ_ρ is incorrect
then the only possibility that I have been able to suggest, is that the
HNC and PY formulae may be wrong and that a complete summation of all
terms will result in a cancellation of the zeroth order term, $\emptyset p$.
Finally, the fact that a self-bound droplet is translationally invariant
needs to be kept in mind, especially if the translationally invariant
wavefunction $\prod_{i<j} f(r_{ij})$ turns out to be too simple to describe a droplet
of liquid He^4 .

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III. GENERALIZED VIRIAL THEOREMS.

In this section I discuss generalized virial theorems that give the change in the free energy of a system during an infinitesimal isothermal deformation. A deformation will affect the kinetic energy and the potential energy separately so that these virial theorems relate expressions involving the momentum of the particles to expressions involving the interactions between the particles. I have already discussed an example of a generalized virial theorem in section CI, (i.e. [1]), where a particular isothermal deformation of a classical system was considered, in order to derive a formula for the surface tension of a classical liquid-vapour interface. In this section I will investigate the general virial theorem for any first order deformation and consider a particular example of a second order deformation.

Consider first, generalized virial theorems for quantum systems. To deform a quantum system we scale the wavefunction to fit the new shape. One of the first persons to derive a generalized quantum virial theorem by scaling was Toda (1955), [2] , who even introduced the idea of a quantum mechanical stress tensor. Another extensive discussion on the scaling problem and the virial theorem is given by Löwdin (1959), [3] . Much of the results of this section appear to have been derived by Toda (although he restricted his discussion to pair interactions), but I prefer to work from a recent paper by McLellan (1974), [4] , who derives a general expression for the change in energy during any first order infinitesimal deformation. I will now prove McLellan's result by a slightly different procedure that I found easier to understand, before going on to investigate its many uses.

Consider an N particle quantum system of volume V and assume it to be an eigenstate of a Hamiltonian H = K + U, i.e. we are discussing a single excited state or an entire system at temperature T = O. The wavefunction Ψ and the energy E are given by

$$H\Psi = E\Psi \tag{1a}$$

$$\int_V d\tau\, \Psi^*\Psi = 1 \qquad , \quad d\tau = \prod_{j=1}^{N} d\mathbf{r}_j \tag{1b}$$

$$E = \int_V d\tau\, \Psi^* H \Psi \tag{1c}$$

To deform the system from V to V' we scale the wavefunction by a change of variables :

$$\Psi(\mathbf{r}_1,\ldots,\mathbf{r}_N) \longrightarrow \Psi(\mathbf{r}_1',\ldots,\mathbf{r}_N') \equiv \Psi' \tag{2}$$

where $\mathbf{r}'$ varies over the same limits as $\mathbf{r}$ used to, i.e. $\mathbf{r}'$ varies over V and $\mathbf{r}$ now varies over V'. A general infinitesimal deformation can be written as a matrix, $\underline{\Lambda} = (\underline{1} + \underline{\lambda})$, where $\underline{1}$ is the identity and $\underline{\lambda}$ is an infinitesimal matrix. In the notation of (2) we have

$$\mathbf{r} \equiv \begin{pmatrix} x \\ y \\ z \end{pmatrix} = \begin{pmatrix} 1+\lambda_{11} & \lambda_{12} & \lambda_{13} \\ \lambda_{21} & 1+\lambda_{22} & \lambda_{23} \\ \lambda_{31} & \lambda_{32} & 1+\lambda_{33} \end{pmatrix} \begin{pmatrix} x' \\ y' \\ z' \end{pmatrix} \equiv \underline{\Lambda}\, \mathbf{r}' \tag{3}$$

Note that when $\lambda_{ij} > 0$ the deformation is stretching with respect to this particular element of $\underline{\lambda}$. From (1c) and (2) the energy of the deformed system is

$$E' = \int_{V'} d\tau\, \Psi^{*\prime} H \Psi' \qquad , \tag{4a}$$

with the normalization constraint

$$\int_{V'} d\tau\, \Psi^{*\prime}\Psi' = 1 \tag{4b}$$

(i.e. the total number of particles is a constant.)

Letting H' stand for H written with dashed coordinates, we can rewrite

(4) as

$$E' = \int_{V'} d\tau \, \Psi'^* \left[H' + (H - H') \right] \Psi'$$

$$= E + \int_{V'} d\tau \, \Psi'^* (H - H') \Psi \tag{5}$$

where the second line follows exactly from the first, since

$$H' \Psi' = E \Psi' \tag{6}$$

That is, a change of coordinates does not alter the energy eigenvalue.

I will now introduce a more convenient notation :

$$\underline{X} = (x_1, x_2, x_3) \equiv (x, y, z) \tag{7}$$

and

$$\underline{X}^{(\mu)} \equiv \underline{r}_\mu$$

The summation convention for Greek indices will apply whenever a sum is not written explicitly. We wish to calculate the partial derivative of E with respect to an element λ_{ab} of $\underline{\lambda}$. I choose to do this by considering the particular deformation $\lambda_{ij} = 0 , (i,j) \neq (a,b)$, i.e.

$$X_\alpha = X'_\alpha \qquad , \alpha \neq a$$
$$X_a = X'_a + \lambda_{ab} X'_b \tag{8}$$

The change in the potential energy is found by expanding U in a Taylor series about U' :

$$U(\underline{x}^{(1)}, \ldots, \underline{x}^{(N)}) = U(\underline{x}^{(1)'}, \ldots \underline{x}^{(N)'}) + \sum_{\mu=1}^{N} (\underline{x}^{(\mu)} - \underline{x}^{(\mu)'}) \cdot (\underline{\nabla}^{(\mu)} U)' + \cdots$$

$$= U' + \lambda_{ab} \sum_{\mu=1}^{N} x_b^{(\mu)'} \left(\frac{\partial U}{\partial X_a} \right)' + 0(\lambda_{ab}^2) \tag{9}$$

The total kinetic energy is given by

$$K = -\frac{\hbar^2}{2m} \sum_{\mu=1}^{N} \sum_{\alpha=1}^{3} \frac{\partial^2}{\partial x_\alpha^{(\mu)2}} = \frac{1}{2m} \sum_\mu \sum_\alpha p_\alpha^{(\mu)2} \tag{10}$$

To write $\dfrac{\partial^2}{\partial x_\alpha^2}$ in terms of the dashed coordinates apply the chain rule

$$\frac{\partial}{\partial x_\alpha} = \left(\frac{\partial x_1'}{\partial x_\alpha}\right)_{x_2', x_3'} \cdot \frac{\partial}{\partial x_1'} + \left(\frac{\partial x_2'}{\partial x_\alpha}\right)_{x_1', x_3'} \cdot \frac{\partial}{\partial x_2'} + \left(\frac{\partial x_3'}{\partial x_\alpha}\right)_{x_1', x_2'} \cdot \frac{\partial}{\partial x_3'} \quad (11)$$

and use (8),

(1) $a = b$:
$$\frac{\partial^2}{\partial x_\alpha^2} = \begin{cases} \dfrac{\partial^2}{\partial x_\alpha'^2} & , \quad \alpha \neq a \\[2ex] (1 + \lambda_{aa}^{-2})\dfrac{\partial^2}{\partial x_a'^2} & , \quad \alpha = a \end{cases} \qquad (12a)$$

(2) $a \neq b$:
$$\frac{\partial^2}{\partial x_\alpha^2} = \begin{cases} \dfrac{\partial^2}{\partial x_\alpha'^2} & , \quad \alpha \neq b \\[2ex] \dfrac{\partial^2}{\partial x_b'^2} - 2\lambda_{ab}\dfrac{\partial^2}{\partial x_a' \partial x_b'} + \lambda_{ab}^2 \dfrac{\partial^2}{\partial x_a'^2} & , \quad \alpha = b \end{cases} \qquad (12b)$$

In both cases we have

$$\sum_{\alpha=1}^{3} \frac{\partial^2}{\partial x_\alpha^2} = \sum_{\alpha=1}^{3} \frac{\partial^2}{\partial x_\alpha'^2} - 2\lambda_{ab}\frac{\partial^2}{\partial x_a' \partial x_b'} + O(\lambda_{ab}^2)$$

$$(13)$$

Therefore (10) and (13) give

$$K = K' - \frac{\lambda_{ab}}{m}\sum_{\mu=1}^{N} p_a^{(\mu)'} p_b^{(\mu)'} + O(\lambda_{ab}^2) \qquad (14)$$

Using the summation convention, we see from (9) and (14) that

$$H - H' = \lambda_{ab}\left\{ -\frac{1}{m} p_a^{(\mu)'} p_b^{(\mu)'} + x_b^{(\mu)'}\left(\frac{\partial U}{\partial x_a^{(\mu)}}\right)' \right\} + O(\lambda_{ab}^2) \qquad (15)$$

Inserting (15) into (5) and using

$$\int_{V'} d\tau \, \Psi^* A' \Psi' = \int_V d\tau \, \Psi^* A \Psi \equiv \langle A \rangle \qquad (16)$$

(aside: (16) is only exact if $d\tau = d\tau'$, but it will always be correct to the first order in λ_{ab}), we find

$$E' - E = \lambda_{ab}\left\{ -\frac{1}{m}\langle p_a^{(\mu)} p_b^{(\mu)} \rangle + \langle x_a^{(\mu)} \frac{\partial U}{\partial x_b^{(\mu)}} \rangle \right\} + O(\lambda_{ab}^2)$$

$$(17)$$

So we have proved McLellan's result :

$$\left(\frac{\partial E}{\partial \lambda_{ab}}\right)_{\lim \lambda_{ab} \to 0} = -\frac{1}{m}\left\langle p_a^{(\mu)} p_b^{(\mu)}\right\rangle + \left\langle x_a^{(\mu)} \frac{\partial U}{\partial x_b^{(\mu)}}\right\rangle \tag{18}$$

Equation (18) applies to a single energy eigenstate, but we can obtain the corresponding result for a system at $T > 0$ by taking a statistical mechanical average. The total energy E is given by (cf. CI eqns (17) - (20))

$$E = \sum_{\ell} \epsilon_{\ell} P_{\ell} \quad , \quad P_{\ell} = \frac{e^{-\epsilon_{\ell}/T}}{\sum_{\ell} e^{-\epsilon_{\ell}/T}}$$
$$\equiv [\![E]\!] \tag{19}$$

where $[\![\ ]\!]$ indicates the statistical mechanical average. The total free energy is

$$F = -T \log \sum_{\ell} e^{-\epsilon_{\ell}/T} \tag{20}$$

From (20) we have

$$\left(\frac{\partial F}{\partial \lambda_{ab}}\right)_T = \frac{\sum_{\ell} \left(\frac{\partial \epsilon_{\ell}}{\partial \lambda_{ab}}\right)_T e^{-\epsilon_{\ell}/T}}{\sum_{\ell} e^{-\epsilon_{\ell}/T}} = \left[\!\left[\left(\frac{\partial E}{\partial \lambda_{ab}}\right)_T\right]\!\right] \tag{21}$$

So the quantum mechanical virial theorem for $T > 0$ is

$$\left(\frac{\partial F}{\partial \lambda_{ab}}\right)_T\bigg|_{\lim \lambda_{ab} \to 0} = -\frac{1}{m}\left[\!\left[\left\langle p_a^{(\mu)} p_b^{(\mu)}\right\rangle\right]\!\right] + \left[\!\left[\left\langle x_a^{(\mu)} \frac{\partial U}{\partial x_b^{(\mu)}}\right\rangle\right]\!\right] \tag{22}$$

At T = 0, (22) reduces to (18). For an infinite or self-bound system the free energy will be a minimum, so that in these cases the left hand side of (22) is zero. However, this will not be true for finite systems at $T > 0$, since they must be contained within boundary walls, and the energy of the system inside the walls is not in general a minimum with respect to a deformation of the walls. If the system is finite a typical deformation will usually change both the volume (V) and the surface area (A). Now in CI we saw that the free energy of a finite system can be divided into a volume free energy and a surface

free energy, $F(V,A) = F_V + F_A$. There is even a surface free energy (i.e. surface tension) for an interacting system enclosed by rigid walls, i.e. the Fowler term (see [1] and references 3 and 15 of [1]), which is equivalent to assuming a completely sharp surface for a self-bound system. So using the chain rule, the left hand side of (22) becomes, (see also [5] eqn (10))

$$\left(\frac{\partial F}{\partial \lambda_{ab}}\right)_T = \left(\frac{\partial F}{\partial V}\right)_{A,T} \frac{\partial V}{\partial \lambda_{ab}} + \left(\frac{\partial F}{\partial A}\right)_{V,T} \frac{\partial A}{\partial \lambda_{ab}} \tag{23}$$

By differentiating the formula F = E - TS, (S the entropy), one obtains

$$dF = -SdT + PdV + \sigma dA \tag{24}$$

where P is the pressure and σ the surface tension. Using (23) and (24) we see that the generalized quantum virial theorem for a finite system can be written as

$$-P\left(\frac{\partial V}{\partial \lambda_{ab}}\right)_{\lim \lambda_{ab} \to 0} + \sigma\left(\frac{\partial A}{\partial \lambda_{ab}}\right)_{\lim \lambda_{ab} \to 0} = -\frac{1}{m}\left[\!\left[\left\langle p_a^{(\mu)} p_b^{(\mu)}\right\rangle\right]\!\right]$$
$$+ \left[\!\left[\left\langle x_b^{(\mu)} \frac{\partial U}{\partial x_a^{(\mu)}}\right\rangle\right]\!\right] . \tag{25}$$

Examples of the use of (25) :

From (25) we can find the change in the free energy during any given isothermal infinitesimal deformation, provided the change in energy is first order in the deformation.

A) Isotropic Deformations.

Consider a simple isotropic expansion of a finite system :

$$\lambda_{ii} = \delta , \quad \lambda_{ij} = 0 \quad i \neq j \quad , \text{ i.e.}$$

$$\underline{\underline{\Lambda}} = \begin{pmatrix} 1+\delta & 0 & 0 \\ 0 & 1+\delta & 0 \\ 0 & 0 & 1+\delta \end{pmatrix} \tag{26}$$

We can write $V \sim L^3$ and $A \sim L^2$, where L is some characteristic length. Then applying the deformation (26) we have $V \rightarrow V(1+\delta)^3$ and $A \rightarrow A(1+\delta)^2$, so that

$$\left(\frac{\partial V}{\partial \delta}\right)_{\lim \delta \to 0} = 3V \quad , \quad \left(\frac{\partial A}{\partial \delta}\right)_{\lim \delta \to 0} = 2A \tag{27}$$

Therefore, applying (25) three times and using (27), we find

$$-3PV + 2\sigma A = -2 [\![\langle K \rangle]\!] + [\![\langle C \rangle]\!] \tag{28a}$$

where

$$C \equiv \underline{x}^{(\mu)} \underline{\nabla}^{(\mu)} U = x_\alpha^{(\mu)} \frac{\partial U}{\partial x_\alpha^{(\mu)}} \tag{28b}$$

1) A quantum droplet at T = 0 :

Consider a self-bound quantum droplet of radius R at T = 0, e.g. liquid He^4 . Because the temperature is at absolute zero the pressure outside the drop is zero (i.e. there is no vapour), hence from Laplace's equation (see eqns 2 and 3 of section CI) we have

$$P = \frac{2\sigma}{R} \quad , \quad \text{i.e. } 3PV = 2\sigma A \tag{29}$$

From (28) and (29) we must have

$$-2\langle K \rangle + \langle C \rangle = 0 \tag{30}$$

for a self-bound quantum system at T = 0. Equation (30) means that $\left(\frac{\partial E}{\partial \delta}\right)_{\lim \delta \to 0} = 0$, which is just a consequence of the fact that a quantum droplet at T = 0 is in its ground state and so its energy is a minimum with respect to any deformation. Note that for this case the surface term $2\sigma A$ has the same value as the volume term $3PV$ and cannot be neglected as one might at first expect.

2) Finite systems at $T > 0$:

At $T > 0$ there arises an immediate complication due to the fact that for a finite system to be in equilibrium it must be confined in a container. This is because a self-bound liquid would now be surrounded by a vapour and to maintain a finite system in equilibrium one requires fixed boundaries. To avoid the extra problems associated with two phases (i.e. one must identify the separate contributions to P and σ), we can consider either a fluid above its critical point or a gas, inside a container. Equation (30) will no longer hold since the walls of the container exert an external pressure on the system. That is, the walls are an external influence and the system is not in an energy minimum with respect to changes in position of the walls. For an interacting system of particles the surface term $2\sigma A$ is still present, since the surface tension due to a wall is equivalent to the surface tension of a completely sharp surface. From equation 40 of reference [1] , the surface tension of a classical (i.e. coming from U only) fluid of density $\bar{n}$, with pair interactions, due to the presence of a wall, is given by Fowler's formula

$$\sigma_F = \frac{\pi}{8} \bar{n}^2 \int_0^\infty dr \, r^4 g(r, \bar{n}) \frac{du(r)}{dr} \tag{31}$$

where $U = \sum_{i<j}\sum u(r_{ij})$ and the pair density has been approximated by $\bar{n}^2 g(r, \bar{n})$.

The classical form of the generalized virial theorem can also be derived by a deformation method, but I will first give McLellan's proof since a problem arises with his method in trying to identify the $2\sigma A$ term. For a classical system we have

$$\frac{1}{m} p_a^{(\mu)} p_b^{(\mu)} = \dot{x}_a^{(\mu)} p_b^{(\mu)} \qquad (\dot{} \equiv d/dt) \tag{32}$$

$$= \frac{d}{dt}\left(x_a^{(\mu)} p_b^{(\mu)} \right) - x_a^{(\mu)} \dot{p}_b^{(\mu)}$$

Now the classical time average of a quantity $\frac{df}{dt}$ is

$$\overline{\frac{df}{dt}} \equiv \lim_{\tau \to \infty} \frac{1}{\tau} \int_0^{\tau} dt \, \frac{df}{dt} \tag{33}$$

$$= \lim_{\tau \to \infty} \left[\frac{f(\tau) - f(0)}{f(\tau)} \right]$$

So provided the system is finite (i.e. contained) and $f(t)$ is non divergent, we find

$$\overline{df/dt} = 0 \tag{34}$$

From (32) and (34) we obtain

$$-\frac{1}{m} \overline{p_a^{(\mu)} p_b^{(\mu)}} = - \overline{x_a^{(\mu)} \dot{p}_b^{(\mu)}} \tag{35}$$

$$= - \overline{x_a^{(\mu)} \frac{\partial U}{\partial x_b^{(\mu)}}} \tag{35}$$

where U is the total potential energy, including the wall potential. For $a \neq b$ the left hand side of (35) is zero since the kinetic energy is isotropic in classical fluids. By assuming the ergothic hypothesis (i.e. ensemble averages and time averages are equivalent), we can replace the time average by the usual statistical average

$$[\![A]\!] \equiv \frac{\int dx^{(1)} \ldots dx^{(N)} A e^{-U/T}}{\int dx^{(1)} \ldots dx^{(N)} e^{-U/T}} \tag{36}$$

Considering the isotropic virial theorem, i.e. a = b, we have proved

$$0 = -2 [\![K]\!] + [\![x_a^{(\mu)} \frac{\partial U}{\partial x_a^{(\mu)}}]\!] \tag{37}$$

Note, for a classical system in three dimensions

$$[\![K]\!] = \frac{3}{2} NT \tag{38}$$

Comparing (37) with the quantum virial theorem (28) it appears at first sight that the terms $-3PV$ and $2\sigma A$ are missing in (37). However, proper consideration of the wall potential should yield

these terms. It is easy to show that the volume term contributed by
the walls is $-3PV$, since the pressure P is exerted on the system
by the walls, and we can write

$$\left[\!\left[x_\alpha^{(\mu)} \frac{\partial U}{\partial x_\alpha^{(\mu)}} \right]\!\right]_{Vol/walls} = -P \int \underline{ds} \cdot \underline{x} \quad , \quad (\underline{ds} \text{ is over wall surface})$$

$$= -P \int \underline{dx} \; \underline{\nabla} \cdot x \quad , \quad (\text{by Gauss's theorem})$$

$$= -3PV \tag{39}$$

Unfortunately I cannot see how the surface term $2\sigma A$, which must be
present for an interacting classical system (i.e. Fowler's formula),
can be obtained from the above proof. However, it does appear in
another method of proving the classical virial theorem, by deforming
the system in a manner similar to the quantum derivation. To show
this I will consider only the isotropic deformation (26). Then
following exactly the same process as equations 3 - 8 of reference [1]
which is based on the classical expression for $e^{-F/T}$, one obtains

$$\Delta F = -3NT\delta + [\![\Delta U]\!] \tag{40}$$

where ΔU is defined by

$$U(\underline{x}^{(1)}_{(1+\delta)}, \ldots, \underline{x}^{(N)}_{(1+\delta)}) = U(\underline{x}^{(1)}, \ldots \underline{x}^{(N)}) + \Delta U \tag{41}$$

The first term of (40) occurs because the Jacobian of the change of
variables is not unity. For the deformation (26) we have

$$\Delta U = \delta \left(x_\alpha^{(\mu)} \frac{\partial U}{\partial x_\alpha^{(\mu)}} \right) + O(\delta^2) \tag{42}$$

From (38), (40) and (42) it follows that

$$\Delta F = \delta \left\{ -2[\![K]\!] + [\![C]\!] \right\} + O(\delta^2) \tag{43}$$

Now the left hand side of (43) includes a volume term and a surface
term in the same way as (23), i.e. from the chain rule

$$\Delta F = \left(\frac{\Delta F}{\Delta V}\right)_A \Delta V + \left(\frac{\Delta F}{\Delta A}\right)_V \Delta A$$

$$= (-3PV + 2\sigma A)\delta + O(\delta^2)$$

Therefore we have shown that

$$-3PV + 2\sigma A = -2[\![K]\!] + [\![C]\!] \tag{44}$$

From a comparison of (28) and (44) we see, as should be expected, that the only difference between the classical and quantum virial theorems is the different averages.

Another reason why the surface term on the left hand side of (44) should not be left out is that for an interacting classical system, $[\![C]\!]$ will have a surface term as well as a volume term, in the same manner as was calculated for $[\![U]\!]$ in reference [1]. To see if the surface terms on both sides of (44) could cancel, consider a classical fluid of density $\bar{n} = N/V$, confined to a cubic box of length L, with the particles interacting via central pairwise forces, i.e.

$$U = \sum_{i<j}^{N}\sum^{N} u(r_{ij}) \tag{45}$$

Then from equation 15 of reference [1] and switching to the $\underline{r}_j$ notation, we have

$$[\![C]\!] \equiv \left[\!\!\left[x_\alpha^{(\mu)} \frac{\partial U}{\partial x_\alpha^{(\mu)}} \right]\!\!\right] = \frac{1}{2} N(N-1) \left[\!\!\left[r_{12} \frac{du(r_{12})}{dr_{12}} \right]\!\!\right] \tag{46}$$

$$= \frac{1}{2} \int d\underline{r}_1 d\underline{r}_2 \, n(\underline{r}_1,\underline{r}_2) \, r_{12} \frac{du(r_{12})}{dr_{12}}$$

where the pair density is defined by

$$n(\underline{r}_1,\underline{r}_2) = N(N-1) \frac{\int d\underline{r}_3 \dots d\underline{r}_N \, e^{-U/T}}{\int d\underline{r}_1 \dots d\underline{r}_N \, e^{-U/T}} \tag{47}$$

Define a generalized pair correlation function $g(\Gamma_1, \Gamma_2)$ by

$$n(\Gamma_1, \Gamma_2) = n(\Gamma_1) \, n(\Gamma_2) \, g(\Gamma_1, \Gamma_2)$$

$$= \bar{n}^2 \, g(\Gamma_1, \Gamma_2) \qquad \text{, (inside the cube)}$$

(48)

Then if we use the same approximation for the pair correlation function as in [1], namely

$$g(\Gamma_1, \Gamma_2) = g(r_{12}, \bar{n}) \qquad ,$$

(49)

which is incorrect near the walls but enables us to obtain analytic results, we have from section three of [1]

$$[\![c]\!] = [\![c]\!]_V + [\![c]\!]_A$$

(50a)

$$[\![c]\!]_A = -A \frac{\pi}{2} \bar{n}^2 \int_0^\infty dr \, r^4 \, g(r, \bar{n}) \frac{du(r)}{dr}$$

(50b)

$$[\![c]\!]_V = V \cdot 2\pi \, \bar{n}^2 \int_0^\infty dr \, r^3 \, g(r, \bar{n}) \frac{du(r)}{dr}$$

(50c)

The last equation is derived from (46), (48) and (49), by ignoring the walls and changing $\int d\Gamma_1 \, d\Gamma_2 \longrightarrow V \int d\Gamma_{12}$. The upper limits in (50b) and (50c) can be extended to infinity because of the short-ranged nature of du/dr. Unfortunately the result (50b) is rather strange, since from (31) we see that $[\![c]\!]_A = -4\sigma_F A$ and not $2\sigma_F A$. That is, $[\![c]\!]$ definitely contains a surface term but it does not appear to cancel with $2\sigma_F A$. From [1] I note that $2\sigma_F A$ would follow from a slightly different expression than (46), namely

$$\frac{A}{2} \int d\Gamma_1 \, d\Gamma_2 \, n(\Gamma_1, \Gamma_2) \frac{(r_{12}^2 - 3z_{12}^2)}{r_{12}} \frac{du(r_{12})}{dr_{12}}$$

(51)

From the results of section CII we see that this lack of cancellation is also present for the potential energy contributions to the quantum virial theorem for the case of liquid He^4 in a tank. For an

interacting quantum system there will also be a surface term from

$[[\langle K \rangle]]$, just as there is a kinetic energy contribution to the

surface tension of a quantum fluid. By ignoring all the surface terms

one obtains from (44) and (50c), the usual formula

$$P = \bar{n}T - \frac{2}{3}\pi\bar{n}^2 \int_0^\infty dr\, r^3 g(r,\bar{n}) \frac{du(r)}{dr} \qquad (52)$$

3) A finite system of non-interacting particles :

For non interacting particles we have C = 0, so that (52) becomes

$$PV = \frac{2}{3}\bar{E} \qquad (53a)$$

where $\bar{E}$ is the total average energy. From equation (28) we see that

(53) also holds for a non-interacting quantum system. For the classical

case the surface tension term is zero since σ is due entirely to

interactions, however it is not necessary to assume that σ is zero

for the quantum case. This is because we can identify σA as a surface

free energy F_s , so that (53) for the quantum system could be

written as

$$PV = \frac{2}{3}\bar{E} = \frac{2}{3}(F_v + F_s) \qquad (53b)$$

where $\bar{E} = [[\langle K \rangle]] + \sigma A$. For non-interacting quantum

systems it may be that σ is always zero, but there is a possibility

that the presence of Fermi statistics could result in an anistropic

kinetic energy near the walls, and thus produce a surface free energy.

4) Deformations with respect to other parameters :

The generalized virial theorem (18) or (22) can be applied to

deformations of any parameter. For a discussion on the first

derivative of the energy with respect to an arbitrary parameter see

reference [3]. I will include an example (also in [3]) to illustrate

the idea. Consider a diatomic molecule with an internuclear

distance R. Take the potential energy to be the electrostatic

potential $u(r_{12}) \sim 1/r_{12}$, i.e. $r_{12} \frac{du}{dr_{12}} = -u(r_{12})$.

If we now apply the isotropic deformation (26), then the parameter R

changes from R to $R(1+\delta)$ and the virial theorem (18) becomes

$$\frac{dE}{d\delta}\bigg|_{\lim \delta \to 0} = -2\langle K \rangle - \langle u \rangle \qquad (54)$$

The left hand side is easily evaluated since

$$\frac{dE}{d\delta} = \frac{dE}{dR}\frac{dR}{d\delta} = R\frac{dE}{dR} \qquad (55)$$

From (54) and (55) we obtain

$$2\langle K \rangle + \langle u \rangle + R\frac{dE}{dR} = 0 \qquad (56)$$

If the diatomic molecule is in its equilibrium position (i.e. uncon-

strained), or if R is infinity, then $dE/dR = 0$, so that we

recover the usual virial theorem for self-bound or infinite electro-

static systems, namely $2\langle K \rangle + \langle u \rangle = 0$.

However if the nuclei are fixed away from their equilibrium positions

then the correct virial theorem is (56).

B)   Anisotropic Deformations.

From (25) we see that by considering a deformation in which the

change in volume is zero we can calculate the surface tension σ .

1)   The surface tension of a plane quantum surface :

Following the method of Toda [2] , let us apply the deformation

given by $\Lambda_{11} = 1+\delta, \Lambda_{22} = 1, \Lambda_{33} = (1+\delta)^{-1}$ and $\Lambda_{ij} = 0, i \neq j$.

That is,

$$\underline{\Lambda} = \begin{pmatrix} 1+\delta & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & (1+\delta)^{-1} \end{pmatrix} \qquad (57)$$

If we take a flat surface in the xy plane then the deformation (57)

stretches the x direction and compresses the z direction :

$$V \longrightarrow V(1+\delta)(1+\delta)^{-1} = V$$
$$A \longrightarrow A(1+\delta)$$

(58)

The volume of both the liquid and the vapour remain constant so that all volume terms are zero. The change in the liquid surface area is δA . Note that the surface area of some of the walls will change so that to evaluate the expression for σ one needs to identify and discard surface terms arising from the change in density at the walls (see [1]). From (58) we have

$$\frac{\partial F}{\partial \delta} = \frac{\partial F}{\partial A} \frac{\partial A}{\partial \delta} = \sigma A$$

(59)

and from (57) it follows that

$$\frac{\partial F}{\partial \delta} = \frac{\partial F}{\partial \lambda_{11}} \frac{\partial \lambda_{11}}{\partial \delta} + \frac{\partial F}{\partial \lambda_{33}} \frac{\partial \lambda_{33}}{\partial \delta}$$
$$= \frac{\partial F}{\partial \lambda_{11}} - \frac{\partial F}{\partial \lambda_{33}} + O(\delta)$$

(60)

So from the generalized virial theorem (22) we find

$$\sigma = \frac{1}{A}\left\{ -2\left[\!\left[\langle k_x - k_z \rangle \right]\!\right] + \left[\!\left[\left\langle x_\alpha^{(\mu)} \frac{\partial U}{\partial x_\alpha^{(\mu)}} - z_\alpha^{(\mu)} \frac{\partial U}{\partial z_\alpha^{(\mu)}} \right\rangle \right]\!\right] \right\}$$

(61)

This is the Toda (see also Brout and Nauenberg, referenced in [6]) expression for the surface tension of a plane quantum surface, extended to a general potential U. The potential term on the right hand side of (61) is the classical result, which was proved in [1] by applying the same deformation to a classical system. For pair inter-actions, i.e. $U = \sum_i \sum_j u(r_{ij})$, we have from (61), (using eqn 15 of [1] and changing the notation from $\underline{x}^{(\mu)}$ to $\underline{r}_\mu$)

$$\sigma = \frac{1}{A}\left\{ -\frac{N\hbar^2}{m}\left[\!\left[\left\langle \frac{\partial^2}{\partial z_i^2} - \frac{\partial^2}{\partial x_i^2} \right\rangle \right]\!\right] + \frac{N(N-1)}{4}\left[\!\left[\left\langle \frac{(r_{12}^2 - 3z_{12}^2)}{r_{12}} \frac{du(r_{12})}{dr_{12}} \right\rangle \right]\!\right] \right\}$$

(62)

This formula was used, for the case of liquid He^4 at T = 0, in CII (i.e. section four of [6]).

2) The surface energy of a quantum droplet at T = 0 :

Consider a self-bound droplet of a quantum liquid at absolute zero (e.g. liquid He^4). The ground state will be spherical since this minimises the surface area which in turn minimises the surface energy. At T = 0 the free energy and the total energy are the same so that the surface tension σ is equal to the surface energy per unit area. We can deform the sphere into a prolate ellipsoid, symmetric about the Z axis, by applying the deformation $\lambda_{ij} = 0 \quad i \neq j$,

$$\lambda_{11} = \lambda_{22} = \left(e^{-\delta/2} - 1 \right) \quad , \quad \lambda_{33} = \left(e^{\delta} - 1 \right) \qquad \text{i.e.}$$

$$\underline{\Delta} = \begin{pmatrix} e^{-\delta/2} & 0 & 0 \\ 0 & e^{-\delta/2} & 0 \\ 0 & 0 & e^{\delta} \end{pmatrix} \tag{63}$$

In the (x,y,z) notation of (3) this is

$$x = x' e^{-\delta/2} \quad , \quad y = y' e^{-\delta/2} \quad , \quad z = z' e^{\delta} \tag{64}$$

{aside: In reference [7] p 452, it is stated that the lines of motion for a second order surface oscillation of a liquid globe (i.e. sphere into ellipsoid) are given by $z \, \varpi^2$ = constant, where ϖ is the distance from the axis of symmetry, the z axis. Taking the point (x', y', z') in the undeformed sphere and applying the scale change (2) with (64) we see that $z \left(x^2 + y^2 \right) = z' \left(x'^2 + y'^2 \right)$.

This is a proof of my statement that (63) changes a sphere into an ellipsoid of revolution about the z axis. } To calculate the change in the surface area during the deformation we need the surface area of an ellipsoid of revolution whose boundary is defined by

$$\frac{x^2}{a^2} + \frac{y^2}{a^2} + \frac{z^2}{b^2} = 1 \qquad , \tag{65a}$$

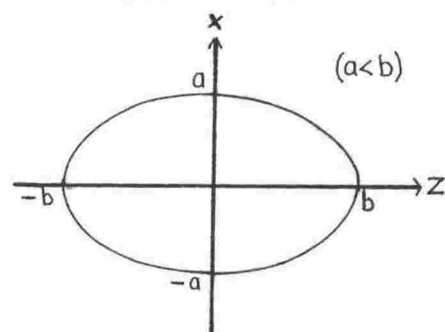$$a = R e^{-\delta/2} \quad , \quad b = R e^{\delta} \qquad , \tag{65b}$$

where R is the radius of the initial sphere. Note that

$$V = \frac{4}{3}\pi a^2 b = \frac{4}{3}\pi R^3$$

, so that the change in the volume is identically zero. By rotating the ellipse of Fig. 1. about the z axis, we can calculate (somewhat laboriously)

Fig. 1.

$(a < b)$

$$A = 2 \int_{z=0}^{z=b} ds\, 2\pi |x| \quad , \quad ds^2 = dx^2 + dz^2$$

$$= 4\pi \frac{a}{b} \int_0^b dz\, \sqrt{b^2 - \epsilon^2 z^2} \quad , \quad \epsilon^2 = 1 - \frac{a^2}{b^2} > 0$$

$$= 2\pi a^2 + 2\pi ab\, \frac{\sin^{-1}\epsilon}{\epsilon}$$

$$= 2\pi a^2 + 2\pi ab \left\{ 1 + \frac{\epsilon^2}{6} + \frac{3}{40}\epsilon^4 + \cdots \right\} \tag{66}$$

From (65b) we have

$$\epsilon^2 = 3\delta - \frac{9}{2}\delta^2 + O(\delta^3) \quad , \quad \epsilon^4 = 9\delta^2 + O(\delta^3) \tag{67}$$

and hence (66) is

$$A = 2\pi a^2 + 2\pi ab \left\{ 1 + \frac{\delta}{2} - \frac{3}{40}\delta^2 + O(\delta^3) \right\}$$

$$= 4\pi R^2 \left\{ 1 + \frac{2}{5}\delta^2 + O(\delta^3) \right\} \tag{68}$$

So to the lowest order in δ , the change in the surface area during the deformation is

$$\Delta A = A_0 \frac{2}{5}\delta^2 \quad , \quad A_0 = 4\pi R^2 \tag{69}$$

We see that the change in area is second order in the deformation parameter, unlike the plane surface deformation in part B 1) above. I shall now show that the change in energy is also second order in

δ . If we use the first order generalized virial theorem (18),

with $\underline{\Lambda}$ given by (63), then

$$\left(\frac{\partial E}{\partial \delta}\right)_{\lim \delta \to 0} = \left(\frac{\partial E}{\partial \lambda_{11}}\frac{\partial \lambda_{11}}{\partial \delta} + \frac{\partial E}{\partial \lambda_{22}}\frac{\partial \lambda_{22}}{\partial \delta} + \frac{\partial E}{\partial \lambda_{33}}\frac{\partial \lambda_{33}}{\partial \delta}\right)_{\lim \delta \to 0}$$

$$= \left(-\frac{e^{-\delta/2}}{2}\left[\frac{\partial E}{\partial \lambda_{11}} + \frac{\partial E}{\partial \lambda_{22}}\right] + e^{\delta}\frac{\partial E}{\partial \lambda_{33}}\right)_{\lim \delta \to 0}$$

$$= \left\langle K_x + K_y - 2K_z\right\rangle + \left\langle \frac{x^{(\mu)}}{2}\frac{\partial U}{\partial x^{(\mu)}} + \frac{y^{(\mu)}}{2}\frac{\partial U}{\partial y^{(\mu)}} - z^{(\mu)}\frac{\partial U}{\partial z^{(\mu)}}\right\rangle$$

$$= 0 \tag{70}$$

The last line follows, because the expectation value is in the ground

state which is spherically symmetric. This result is the same as was

found in part A 1) for isotropic deformations of a self-bound system,

and is a consequence of the fact that an isolated system in

equilibrium occupies an energy minimum with respect to any

deformation.

So the first order generalized virial theorem does not enable us

to find the surface energy of a self-bound droplet since σ is deter-

mined by second order terms. That is, instead of (23) we have (note

that the change in volume is identically zero)

$$\left(\frac{\partial^2 E}{\partial \delta^2}\right)_0 = \frac{\partial E}{\partial A}\left(\frac{\partial^2 A}{\partial \delta^2}\right)_0 \tag{71}$$

where the subscript zero refers to the non deformed limit, i.e.

$\lim \delta \to 0$. From (69) and (71) we find

$$\sigma = \frac{\delta^2}{2\Delta A}\left(\frac{\partial^2 E}{\partial \delta^2}\right)_0 \tag{72}$$

Another way of obtaining this result is to expand the energy of the

deformed system in a Taylor series about the ground state energy E_0 :

$$E = E_0 + \delta\left(\frac{\partial E}{\partial \delta}\right)_0 + \frac{1}{2}\delta^2\left(\frac{\partial^2 E}{\partial \delta^2}\right)_0 + \cdots \tag{73}$$

We have already seen that $\left(\frac{\partial E}{\partial \delta}\right)_0 = 0$, unlike for a flat surface, so that the change in the energy divided by the change in area yields equation (72). Inserting (69) into (72) gives

$$\sigma = \frac{5}{16\pi R^2}\left(\frac{\partial^2 E}{\partial \delta^2}\right)_0 \tag{74}$$

Note that since E_0 is a minimum with respect to the deformation we must have $\left(\partial^2 E / \partial \delta^2\right)_0 \geq 0$, i.e. $\sigma \geq 0$.

Instead of calculating the general second order virial theorem for $\left(\partial^2 E / \partial \lambda_{ab}^2\right)$, I will just consider the particular case of the deformation (64) and calculate the change in energy directly from equation (5) :

$$\Delta E \ (\equiv E - E_0) = \int_{V'} d\tau \ \Psi'^*(H-H')\Psi' \ , \tag{5}$$

where from (64) we see that $d\tau' = d\tau$ so that equation (16) holds exactly :

$$\int_{V'} d\tau \ \Psi'^* A'\Psi' = \int_V d\tau \ \Psi^* A\Psi \equiv \langle A \rangle \tag{16}$$

($\langle \ \rangle$ is the spherically symmetric ground state expectation value.) Using (64) we can write the kinetic energy operator as

$$K = -\frac{\hbar^2}{2m}\sum_{j=1}^{N}\left(\frac{\partial^2}{\partial x_j^2} + \frac{\partial^2}{\partial y_j^2} + \frac{\partial^2}{\partial z_j^2}\right)$$

$$= -\frac{\hbar^2}{2m}\sum_j\left\{e^\delta\left(\frac{\partial^2}{\partial x_j'^2} + \frac{\partial^2}{\partial y_j'^2}\right) + e^{-2\delta}\frac{\partial^2}{\partial z_j'^2}\right\} \tag{75}$$

Therefore from (5), (16) and (75) we have

$$\int_{V'} d\tau \ \Psi'^*(K-K')\Psi = \langle K_x + K_y\rangle(e^\delta - 1) + \langle K_z\rangle(e^{-2\delta} - 1)$$

$$= \delta\langle K_x + K_y - 2K_z\rangle + \delta^2\langle \tfrac{K_x}{2} + \tfrac{K_y}{2} + 2K_z\rangle + 0(\delta^3)$$

$$= \delta^2\langle K \rangle + 0(\delta^3) \tag{76}$$

where the last line follows from the spherical symmetry of the ground state, i.e. $\langle K_x \rangle = \langle K_y \rangle = \langle K_z \rangle$. By expanding the potential $U(x_1, \ldots, z_N)$ about $U(x_1', \ldots, z_N')$ we obtain

$$U = U' + \sum_{j=1}^{N} (\underline{r}_j - \underline{r}_j') \cdot (\nabla U)' + \frac{1}{2} \sum_{j=1}^{N} \sum_{\alpha=1}^{3} \sum_{\beta=1}^{3} (x_\alpha^{(j)} - x_\alpha^{(j)})(x_\beta^{(j)} - x_\beta^{(j)}) \left(\frac{\partial^2 U}{\partial x_\alpha^{(j)} \partial x_\beta^{(j)}} \right)$$

$$+ \cdots$$

$$= U' + \sum_{j=1}^{N} \left\{ (e^{-\delta/2} - 1) \left[x_j' \left(\frac{\partial U}{\partial x_j} \right)' + y_j' \left(\frac{\partial U}{\partial y_j} \right)' \right] + (e^{\delta} - 1) z_j' \left(\frac{\partial U}{\partial z_j} \right)' \right.$$

$$+ \frac{1}{2} (e^{-\delta/2} - 1)^2 \left[x_j'^2 \left(\frac{\partial^2 U}{\partial x_j^2} \right)' + y_j'^2 \left(\frac{\partial^2 U}{\partial y_j^2} \right)' \right] + (e^{\delta} - 1)^2 z_j'^2 \left(\frac{\partial^2 U}{\partial z_j^2} \right)' \quad (77)$$

$$\left. + (e^{-\delta/2} - 1)(e^{\delta} - 1) \left[x_j' z_j' \left(\frac{\partial^2 U}{\partial x_j \partial z_j} \right)' + y_j' z_j' \left(\frac{\partial^2 U}{\partial y_j \partial z_j} \right)' \right] + (e^{-\delta/2} - 1)^2 x_j' y_j' \left(\frac{\partial^2 U}{\partial x_j \partial y_j} \right)' \right\}$$

So

$$\int_{V'} d\tau \, \Psi'^* (U - U') \Psi' = N \left\{ (e^{-\delta/2} - 1) \left\langle x_1 \frac{\partial U}{\partial x_1} + y_1 \frac{\partial U}{\partial y_1} \right\rangle + (e^{\delta} - 1) \left\langle z_1 \frac{\partial U}{\partial z_1} \right\rangle \right.$$

$$+ \frac{(e^{-\delta/2} - 1)^2}{2} \left\langle x_1^2 \frac{\partial^2 U}{\partial x_1^2} + y_1^2 \frac{\partial^2 U}{\partial y_1^2} \right\rangle + (e^{\delta} - 1)^2 \left\langle z_1^2 \frac{\partial^2 U}{\partial z_1^2} \right\rangle$$

$$\left. + (e^{-\delta/2} - 1)(e^{\delta} - 1) \left\langle x_1 z_1 \frac{\partial^2 U}{\partial x_1 \partial z_1} + y_1 z_1 \frac{\partial^2 U}{\partial y_1 \partial z_1} \right\rangle + (e^{-\delta/2} - 1)^2 \left\langle x_1 y_1 \frac{\partial^2 U}{\partial x_1 \partial y_1} \right\rangle \right\}$$

$$= N \frac{3}{4} \delta^2 \left\langle x_1 \frac{\partial U}{\partial x_1} + x_1^2 \frac{\partial^2 U}{\partial x_1^2} - x_1 y_1 \frac{\partial^2 U}{\partial x_1 \partial y_1} \right\rangle + O(\delta^3) \quad (78)$$

The last line in (78) follows from the spherical symmetry of the ground state. From (5), (76), (78) and (74) we have

$$\sigma = \frac{5}{8\pi R^2} \left\{ \langle K \rangle + \frac{3}{4} \sum_{j=1}^{N} \left\langle x_j \frac{\partial U}{\partial x_j} + x_j^2 \frac{\partial^2 U}{\partial x_j^2} - x_j y_j \frac{\partial^2 U}{\partial x_j \partial y_j} \right\rangle \right\} \quad (79)$$

This result is rather strange since the right hand side of (79) contains volume terms (i.e. proportional to R^3/R^2), while σ should be independent of the radius R. So for the expression (79) to make sense the volume terms must cancel, which leads to a condition for the infinite system :

$$\left\langle K \right\rangle_{inf} + \frac{3}{4} \sum_j \left\langle x_j \frac{\partial U}{\partial x_j} + x_j^2 \frac{\partial^2 U}{\partial x_j^2} - x_j y_j \frac{\partial^2 U}{\partial x_j \partial y_j} \right\rangle_{inf} = 0 \qquad (80)$$

where $\left\langle \ \right\rangle_{inf}$ is the expectation value over the infinite system (i.e. pure bulk terms). By using the first order virial theorem

$$2 \left\langle K \right\rangle_{inf} = 3 \left\langle \sum_j x_j \frac{\partial U}{\partial x_j} \right\rangle_{inf} \qquad , \qquad (81)$$

we can write the condition (8) purely in terms of the potential, namely

$$\left\langle 3x_1 \frac{\partial U}{\partial x_1} + x_1^2 \frac{\partial^2 U}{\partial x_1^2} - x_1 y_1 \frac{\partial^2 U}{\partial x_1 \partial y_1} \right\rangle_{inf} = 0 \qquad (82)$$

This condition on U must hold in order for the expression (79) for σ to be independent of the size of the droplet.

For the case of liquid He^4 at T = 0, it is usual to assume a pair potential, i.e.

$$U = \sum_i^N \sum_{i<j}^N u(r_{ij}) \qquad (45)$$

For this potential it would be more useful to rewrite the expression for σ in terms of the translationally invariant coordinates $r_{ij} = | \underline{r}_i - \underline{r}_j |$. To do this I found it necessary to expand $U(r_{12})$ about $U(r_{12}')$, instead of using (77). Applying the deformation (64) I calculate

$$r_{12} = r_{12}' \left\{ 1 + \frac{\delta}{2} \left(\frac{3z_{12}'^2}{r_{12}'^2} - 1 \right) + \frac{\delta^2}{8} \left(1 + 12 \frac{z_{12}'^2}{r_{12}'^2} - 9 \frac{z_{12}'^4}{r_{12}'^4} \right) + O(\delta^3) \right\} \quad (83)$$

and hence

$$U(r_{12}) = U(r_{12}') + (r_{12} - r_{12}') \left(\frac{du(r_{12})}{dr_{12}} \right)' + \frac{1}{2} (r_{12} - r_{12}')^2 \left(\frac{d^2 u(r_{12})}{dr_{12}^2} \right)' + \cdots$$

$$= u(r_{12}') + \frac{\delta}{2}\left(\frac{3z_{12}'^2}{r_{12}'^2} - 1\right)r_{12}'\left(\frac{du(12)}{dr_{12}}\right)' + \frac{\delta^2}{8}\left\{\left(1 + 12\frac{z_{12}'^2}{r_{12}'^2} - 9\frac{z_{12}'^4}{r_{12}'^4}\right)\times\right.$$

$$r_{12}'\left(\frac{du(12)}{dr_{12}}\right)' + \left(1 - 6\frac{z_{12}'^2}{r_{12}'^2} + 9\frac{z_{12}'^4}{r_{12}'^4}\right)r_{12}'^2\left(\frac{d^2u(12)}{dr_{12}^2}\right)'\bigg\}$$

$$+ O(\delta^3) \tag{84}$$

From the spherical symmetry of the ground state it follows that

$$\left\langle z_{12}^2 f(r_{12})\right\rangle = \frac{1}{3}\left\langle r_{12}^2 f(r_{12})\right\rangle, \quad \left\langle z_{12}^4 f(r_{12})\right\rangle = \frac{1}{5}\left\langle r_{12}^4 f(r_{12})\right\rangle \tag{85}$$

Then using (16), (84) and (85), I find

$$\int_{V'} d\tau \,\Psi'^*(u(12) - u(12'))\Psi' = \frac{\delta^2}{8}\left\langle\frac{16}{5}r_{12}\frac{du(12)}{dr_{12}} + \frac{4}{5}r_{12}^2\frac{d^2u(12)}{dr_{12}^2}\right\rangle + O(\delta^3) \tag{86}$$

Therefore the corresponding expression to (79) is

$$\sigma = \frac{5}{8\pi R^2}\left\{\langle K\rangle + \frac{1}{10}\sum_{i<j}^{N}\sum^{N}\left\langle 4r_{ij}\frac{du(ij)}{dr_{ij}} + r_{ij}^2\frac{d^2u(ij)}{dr_{ij}^2}\right\rangle\right\} \tag{87}$$

The infinite system first order virial theorem for the potential (45)

is, (see eqn (46))

$$\langle K\rangle_{inf} = \frac{1}{2}\sum_{i<j}\sum\left\langle r_{ij}\frac{du(ij)}{dr_{ij}}\right\rangle_{inf} \tag{88}$$

Therefore in order for σ to be independent of the radius R, it must

follow that

$$\left\langle 9r_{12}\frac{du(12)}{dr_{12}} + r_{12}^2\frac{d^2u(12)}{dr_{12}^2}\right\rangle_{inf} = O \tag{89}$$

Consider the Lennard-Jones potential used in [6] :

$$u(12) \sim \left(\frac{d}{r_{12}}\right)^{12} - \left(\frac{d}{r_{12}}\right)^6 \tag{90}$$

An attempt to satisfy (89) with (90) fails since the left hand side of

(89) becomes proportional to

$$\left\langle 4\left(\frac{d}{r_{12}}\right)^{12} + \left(\frac{d}{r_{12}}\right)^{6} \right\rangle_{inf} \qquad ,$$

which is greater than zero. It may be that the pair potential is too simple to accurately describe a droplet. Remember that (90) must first satisfy (88) before it can be used in (89). The pair potential also gave a strange result for the surface part of $[\![c]\!]$, i.e. equation (50b).

To sum up - A second order generalized virial theorem is required to find the surface tension of a self-bound droplet. The resulting expression implies an extra condition on the potential in the infinite system, or σ would be proportional to the size of the droplet. However, the condition is not readily satisfied, so that the validity of this theory for σ remains in doubt.

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A Collective Rotational State
of Spherical Nuclei

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Abstract

We consider a particular many-body rotational excitation Ψ of a spherical self-bound system of particles, of the form studied by Lekner (1974). This angular momentum eigenstate is translationally invariant and thus is not a spurious state. The energy of Ψ is found from first principles to be substantially larger than that of the first 2^+ excited states of even–even nuclei, with the exception of ^{208}Pb. The quadrupole moment is negative, the g-factor is approximately Z/A and the lifetime is shorter than the single-particle (Weisskopf) value by a factor of the order of A/Z^2. It is suggested that these states are the finite system rotational analogues of Feynman's phonons and rotons.

1. Introduction

Let $\Phi(r_1, ..., r_A)$ be a translationally and rotationally invariant ground or vibrational state of a self-bound system of A particles, with $H\Phi = E_0\,\Phi$. It has been shown (Lekner 1974; hereinafter referred to as Paper I) that $\Psi = F\Phi$, where

$$F = \tfrac{1}{2} \sum_{i=1}^{A} \sum_{j=1}^{A} (x_{ij}+\mathrm{i}y_{ij})^L f(r_{ij}) \qquad (L \text{ even}) \tag{1}$$

is an eigenstate of L^2 and L_z with eigenvalues $L(L+1)\hbar^2$ and $L\hbar$. The wavefunctions Ψ have the same permutation and inversion symmetry as Φ, and they are translationally invariant. These properties hold irrespective of the masses or permutation symmetries of the constituent particles, but it is clear that, since equation (1) treats each pair of particles in the same way, Ψ is best suited to describe rotational states of systems composed of particles with nearly identical masses and pair interactions, e.g. nuclei and helium microdroplets. We note in passing that Karl and Obryk (1968) and Castilho Alcarás and Leal Ferreira (1971) have found only one symmetric $L = 2$ eigenstate for the three-body system, namely the state Ψ with $f = 1$. The translational invariance of the Ψ considered here guarantees that it is not a spurious state (Elliott and Skyrme 1955; Lipkin 1958; Aviles 1968), i.e. we can be sure that Ψ describes a genuine internal motion of the self-bound system.

In Paper I it was further shown that for harmonic pairwise interactions between A identical Bose particles, that is,

$$H = -(\hbar^2/2m) \sum_{i=1}^{A} \nabla_i^2 + V(r_1, ..., r_A) \tag{2a}$$

with

$$V(r_1, ..., r_A) = \sum_{i<j}\sum v(1 + r_{ij}^2/a^2), \tag{2b}$$

the state Ψ with $f = 1$ is an exact energy eigenstate, with

$$E_L = E_0 + L(2\hbar^2 vA/ma^2)^{\frac{1}{4}} \tag{3}$$

(this equation corrects an error of a factor of $\sqrt{2}$ in Paper I (eqn 30), arising from the same erroneous factor in I(28)). The purpose of the present paper is to carry the analysis of Paper I further, by evaluating the expectation values of the energy, quadrupole moment, magnetic dipole moment and lifetime of the state with $L = 2$ and $f = 1$. These are then compared with experimental values for nuclei with spherical ground states.

2. Expectation Value of Energy

We will assume here and in the remainder of this paper that Φ^2 has complete permutation symmetry, e.g. we neglect the differences between proton and neutron masses and interactions. We also assume that the spins are paired up to give zero S, because $L = 0$ as well as $J = 0$ in the ground state Φ. Thus we are discussing the nondeformed even–even nuclei. These assumptions also imply that, for example, $\langle x_{12}^2 \rangle = \langle y_{12}^2 \rangle = \langle z_{12}^2 \rangle$ where the expectation value is defined in equation (5) below.

Since Ψ is an eigenstate of angular momentum with $L = 2$, it is orthogonal to Φ and thus the expectation value of the Hamiltonian in the state $\Psi = F\Phi$ gives a variational bound

$$
\begin{aligned}
E - E_0 &\leqslant \int d1 \dots dA \; \Psi^*(H - E_0)\Psi \Big/ \int d1 \dots dA \; |\Psi|^2 \\
&= \frac{A\hbar^2}{2m} \int d1 \dots dA \; |\nabla_1 F|^2 \Phi^2 \Big/ \int d1 \dots dA \; |F|^2 \Phi^2 \\
&= \frac{A\hbar^2}{2m} \frac{\langle |\nabla_1 F|^2 \rangle}{\langle |F|^2 \rangle},
\end{aligned}
\tag{4}
$$

where

$$\langle B \rangle = \int d1 \dots dA \; B\Phi^2 \Big/ \int d1 \dots dA \; \Phi^2 \tag{5}$$

denotes a ground-state expectation value. The second step in obtaining the expression (4) comes from Paper I(25), and is valid for any Hamiltonian of the form (2a) with an interaction $V(r_1, \dots, r_A)$ which is completely symmetric and independent of spins and momenta.

We take $f = 1$ in the wavefunction (1), since this gives exact energy eigenstates for harmonic interactions, and also because this is mathematically the simplest and most tractable. A further reason for taking $f = 1$ is given in Section 6. We find

$$\langle |\nabla_1 F|^2 \rangle = 4A(A-1)\langle x_{12}^2 + y_{12}^2 \rangle = \tfrac{8}{3} A(A-1)\langle r_{12}^2 \rangle \tag{6}$$

and

$$
\begin{aligned}
\langle |F|^2 \rangle = &\; \tfrac{1}{2} A(A-1)\langle (x_{12}^2 + y_{12}^2)^2 \rangle \\
&+ A(A-1)(A-2)\langle (x_{12}^2 - y_{12}^2)(x_{13}^2 - y_{13}^2) + 4x_{12}y_{12}x_{13}y_{13} \rangle \\
&+ \tfrac{1}{4} A(A-1)(A-2)(A-3)\langle (x_{12}^2 - y_{12}^2)(x_{34}^2 - y_{34}^2) + 4x_{12}y_{12}x_{34}y_{34} \rangle
\end{aligned}
\tag{7}
$$

(this equation corrects two counting errors in Paper I(39), namely a factor 2 in the three-body term and a factor $\tfrac{1}{2}$ in the four-body term).

In the remainder of this section, the expression for $\langle |F|^2 \rangle$ and hence also that for $E_2 - E_0$ is reduced to expectation values involving only r_{ij} terms. We first note that the energy cannot depend on the azimuthal quantum number. Thus

$$\Psi_{22} = \tfrac{1}{2} \sum_i \sum_j (x_{ij} + iy_{ij})^2 \Phi, \qquad \Psi_{20} = \tfrac{1}{2} \sum_i \sum_j (3z_{ij}^2 - r_{ij}^2) \Phi \tag{8,9}$$

must have the same value of $\langle |\nabla_1 F|^2 \rangle / \langle |F|^2 \rangle$. For Ψ_{20} we find

$$\langle |\nabla_1 F|^2 \rangle = 4A(A-1)\langle r_{12}^2 \rangle \tag{10}$$

and

$$\begin{aligned}
\langle |F|^2 \rangle = {}& \tfrac{1}{2} A(A-1)\langle (3z_{12}^2 - r_{12}^2)^2 \rangle + A(A-1)(A-2)\langle (3z_{12}^2 - r_{12}^2)(3z_{13}^2 - r_{13}^2) \rangle \\
& + \tfrac{1}{4} A(A-1)(A-2)(A-3)\langle (3z_{12}^2 - r_{12}^2)(3z_{34}^2 - r_{34}^2) \rangle .
\end{aligned} \tag{11}$$

Thus equations (6), (7), (10) and (11) give the equality

$$\begin{aligned}
\langle (3z_{12}^2 & - r_{12}^2)^2 + 2(A-2)(3z_{12}^2 - r_{12}^2)(3z_{13}^2 - r_{13}^2) + \tfrac{1}{2}(A-2)(A-3)(3z_{12}^2 - r_{12}^2)(3z_{34}^2 - r_{34}^2) \rangle \\
& = \tfrac{3}{2}\langle (x_{12}^2 + y_{12}^2)^2 + 2(A-2)\{(x_{12}^2 - y_{12}^2)(x_{13}^2 - y_{13}^2) + 4x_{12}\,y_{12}\,x_{13}\,y_{13}\} \\
& \quad + \tfrac{1}{2}(A-2)(A-3)\{(x_{12}^2 - y_{12}^2)(x_{34}^2 - y_{34}^2) + 4x_{12}\,y_{12}\,x_{34}\,y_{34}\} \rangle .
\end{aligned} \tag{12}$$

We will now prove the equality of the two- and three-body terms in equation (12), and thus show the equality of the four-body terms.

In the two-body terms, use of the facts that

$$\langle z_{12}^2 r_{12}^2 \rangle = \tfrac{1}{3}\langle (x_{12}^2 + y_{12}^2 + z_{12}^2) r_{12}^2 \rangle = \tfrac{1}{3}\langle r_{12}^4 \rangle \tag{13}$$

and

$$\langle z_{12}^4 \rangle = \tfrac{1}{5}\langle r_{12}^4 \rangle \tag{14}$$

(obtained by angular integration) demonstrates equality. The value of the two-body term is

$$\langle (3z_{12}^2 - r_{12}^2)^2 \rangle = \tfrac{4}{5}\langle r_{12}^4 \rangle . \tag{15}$$

In the three-body term, we use in addition the identity

$$2z_{12} z_{13} = z_{12}^2 + z_{13}^2 - z_{23}^2 \tag{16}$$

to show that

$$\langle z_{12}^2 z_{13}^2 \rangle = \tfrac{1}{10}\langle r_{12}^4 \rangle . \tag{17}$$

Thus

$$\langle (3z_{12}^2 - r_{12}^2)(3z_{13}^2 - r_{13}^2) \rangle = \tfrac{9}{10}\langle r_{12}^4 \rangle - \langle r_{12}^2 r_{13}^2 \rangle . \tag{18}$$

In the same way the three-body expectation value on the right-hand side of equation (12) is given by (omitting the factor $2(A-2)$)

$$\tfrac{3}{2}\langle 2x_{12}^2 x_{13}^2 + 3x_{12}^2\,y_{12}^2 - 4x_{12}^2\,y_{13}^2 \rangle . \tag{19}$$

The first term we know from equation (17). The second term we find from $\langle r_{12}^4 \rangle = \langle (x_{12}^2 + y_{12}^2 + z_{12}^2)^2 \rangle$ and equation (14) to be

$$\langle x_{12}^2\,y_{12}^2 \rangle = \tfrac{1}{15}\langle r_{12}^4 \rangle . \tag{20}$$

The last term we obtain by expanding $\langle r_{12}^2 r_{13}^2 \rangle$:

$$\langle x_{12}^2 y_{13}^2 \rangle = \tfrac{1}{6}\langle r_{12}^2 r_{13}^2 \rangle - \tfrac{1}{20}\langle r_{12}^4 \rangle. \tag{21}$$

These identities reduce the expression (19) to the right-hand side of equation (18), so that we have demonstrated the equality of the three-body terms. We may thus put

$$\langle (3z_{12}^2 - r_{12}^2)(3z_{34}^2 - r_{34}^2) \rangle = \tfrac{3}{2}\langle (x_{12}^2 - y_{12}^2)(x_{34}^2 - y_{34}^2) + 4x_{12}\, y_{12}\, x_{34}\, y_{34} \rangle. \tag{22}$$

By use of the identity

$$2x_{12}\, x_{34} = x_{14}^2 + x_{23}^2 - x_{13}^2 - x_{24}^2, \tag{23}$$

the right-hand side of equation (22) reduces to

$$3\langle x_{12}^2 x_{34}^2 + x_{12}^2 y_{34}^2 + 2x_{14}^2 y_{14}^2 - 4x_{14}^2 y_{13}^2 \rangle. \tag{24}$$

The last two terms of this expression we know from equations (20) and (21). We can find a relationship between the first two by expanding $r_{12}^2 r_{34}^2$:

$$\langle r_{12}^2 r_{34}^2 \rangle = \langle (x_{12}^2 + y_{12}^2 + z_{12}^2)(x_{34}^2 + y_{34}^2 + z_{34}^2) \rangle = \langle 3x_{12}^2 x_{34}^2 + 6x_{12}^2 y_{34}^2 \rangle. \tag{25}$$

Now the left-hand side of equation (22) is equal to

$$\langle 9z_{12}^2 z_{34}^2 - 6z_{12}^2 r_{34}^2 + r_{12}^2 r_{34}^2 \rangle = \langle 9z_{12}^2 z_{34}^2 - r_{12}^2 r_{34}^2 \rangle. \tag{26}$$

We thus have, equating (26) to the expression (24),

$$\langle 9x_{12}^2 x_{34}^2 - r_{12}^2 r_{34}^2 \rangle = 3\langle x_{12}^2 x_{34}^2 + x_{12}^2 y_{34}^2 + \tfrac{1}{3} r_{12}^4 - \tfrac{2}{3} r_{12}^2 r_{13}^2 \rangle. \tag{27}$$

We can now evaluate the x and y terms using equations (25) and (27):

$$\langle x_{12}^2 x_{34}^2 \rangle = \tfrac{1}{15}\langle 2r_{12}^4 - 4r_{12}^2 r_{13}^2 + 3r_{12}^2 r_{34}^2 \rangle, \tag{28}$$

$$\langle x_{12}^2 y_{34}^2 \rangle = \tfrac{1}{15}\langle -r_{12}^4 + 2r_{12}^2 r_{13}^2 + r_{12}^2 r_{34}^2 \rangle. \tag{29}$$

The four-body term in equation (12) is thus

$$\langle (3z_{12}^2 - r_{12}^2)(3z_{34}^2 - r_{34}^2) \rangle = \tfrac{2}{5}\langle 3r_{12}^4 - 6r_{12}^2 r_{13}^2 + 2r_{12}^2 r_{34}^2 \rangle, \tag{30}$$

and the variational bound for the excitation energy of the state Ψ is

$$\frac{\hbar^2}{m}\, \frac{2A\langle r_{12}^2 \rangle}{\langle \tfrac{2}{5} r_{12}^4 + (A-2)(\tfrac{9}{10} r_{12}^4 - r_{12}^2 r_{13}^2) + \tfrac{1}{4}(A-2)(A-3)\tfrac{2}{5}(3r_{12}^4 - 6r_{12}^2 r_{13}^2 + 2r_{12}^2 r_{34}^2) \rangle}. \tag{31}$$

No approximations have been made to this stage; the result (31) is a rigorous expectation value of the energy in the state Ψ. To evaluate this exactly, however, we would need to know the two-, three- and four-particle correlation functions of the system. In the next section we evaluate (31) in the simplest physically meaningful approximation, namely that in which the particles are correlated simply by coexisting in a finite system.

3. Weak Correlation Approximation

As a first approximation we assume that the A particles are correlated by virtue of the finiteness of the self-bound system, i.e. we assume the system to be characterized solely by a number density $n(r)$ (measured relative to the centre of mass of the system). The use of this 'weak correlation' approximation is supported to some extent by the fact that in nuclei the hard core of the nucleons occupies only about 1% of the total volume (Irvine 1972; de Shalit and Feshbach 1974) so that pair correlations due to nucleon–nucleon interactions can be expected not to be dominant in the evaluation of the expectation values in the result (31). It turns out that the weak correlation approximation is sufficient to make the three-body term

$$\langle (3z_{12}^2 - r_{12}^2)(3z_{13}^2 - r_{13}^2) \rangle$$

nonzero (and in fact positive–definite in this approximation), whereas in a completely uncorrelated (infinite) system it would be zero.

When the system is characterized solely by a single-particle density $n(r)$, depending only on the radial distance from the centre of mass, the expectation values needed for the evaluation of the energy bound (31) can be found by working in spherical bipolar coordinates (Hill 1956). We have

$$\langle r_{12}^2 \rangle = \frac{\displaystyle\int_0^\infty dr_1\, r_1\, n(r_1) \int_0^\infty dr_2\, r_2\, n(r_2) \int_{|r_1-r_2|}^{r_1+r_2} dr_{12}\, r_{12}^3}{\displaystyle\int_0^\infty dr_1\, r_1\, n(r_1) \int_0^\infty dr_2\, r_2\, n(r_2) \int_{|r_1-r_2|}^{r_1+r_2} dr_{12}\, r_{12}}$$

$$= \frac{\displaystyle\int_0^\infty dr_1\, r_1^2\, n(r_1) \int_0^\infty dr_2\, r_2^2\, n(r_2)\, (r_1^2 + r_2^2)}{\left(\displaystyle\int_0^\infty dr\, r^2\, n(r)\right)^2} = 2[\![r^2]\!], \tag{32}$$

where

$$[\![f(r)]\!] \equiv \int_0^\infty dr\, r^2\, n(r)\, f(r) \Big/ \int_0^\infty dr\, r^2\, n(r). \tag{33}$$

Similarly

$$\langle r_{12}^4 \rangle = 2[\![r^4]\!] + \tfrac{10}{3}[\![r^2]\!]^2. \tag{34}$$

The three-particle correlations are a little more complicated: $\langle r_{12}^2\, r_{13}^2 \rangle$ is given by

$$\frac{\displaystyle\int_0^\infty dr_1\, n(r_1) \int_0^\infty dr_2\, r_2\, n(r_2) \int_{|r_1-r_2|}^{r_1+r_2} dr_{12}\, r_{12}^3 \int_0^\infty dr_3\, r_3\, n(r_3) \int_{|r_1-r_3|}^{r_1+r_3} dr_{13}\, r_{13}^3}{\displaystyle\int_0^\infty dr_1\, n(r_1) \int_0^\infty dr_2\, r_2\, n(r_2) \int_{|r_1-r_2|}^{r_1+r_2} dr_{12}\, r_{12} \int_0^\infty dr_3\, r_3\, n(r_3) \int_{|r_1-r_3|}^{r_1+r_3} dr_{13}\, r_{13}}$$

$$= \frac{\displaystyle\int_0^\infty dr_1\, r_1^2\, n(r_1) \int_0^\infty dr_2\, r_2^2\, n(r_2) \int_0^\infty dr_3\, r_3^2\, n(r_3)\, (r_1^2 + r_2^2)(r_1^2 + r_3^2)}{\left(\displaystyle\int_0^\infty dr\, r^2\, n(r)\right)^3}$$

$$= [\![r^4]\!] + 3[\![r^2]\!]^2. \tag{35}$$

Thus the three-body term (18) is positive–definite:

$$\langle \tfrac{9}{10} r_{12}^4 - r_{12}^2 r_{13}^2 \rangle = \tfrac{4}{5} [\![r^4]\!] . \tag{36}$$

In the weak correlation approximation, the four-particle term $\langle r_{12}^2 r_{34}^2 \rangle$ factorizes as

$$\langle r_{12}^2 r_{34}^2 \rangle = \langle r_{12}^2 \rangle^2 = 4 [\![r^2]\!]^2 . \tag{37}$$

It follows that the total four-body term in the expectation value of the energy is zero:

$$\langle 3r_{12}^4 - 6r_{12}^2 r_{13}^2 + 2r_{12}^2 r_{34}^2 \rangle = 6[\![r^4]\!] + 10[\![r^2]\!]^2 - 6[\![r^4]\!] - 18[\![r^2]\!]^2 + 8[\![r^2]\!]^2 = 0 . \tag{38}$$

These results are true for arbitrary radial variation of the density.

In the weak correlation approximation, the energy of the $L = 2$ state thus reduces to

$$\Delta E_2 \equiv E_2 - E_0 \leqslant \frac{\hbar^2}{m} \frac{A [\![r^2]\!]}{\tfrac{1}{3}[\![r^2]\!]^2 + (A-1)\tfrac{1}{5}[\![r^4]\!]} . \tag{39}$$

When $A \gtrsim 20$, we can write

$$\Delta E_2 \lesssim (5\hbar^2/m)[\![r^2]\!]/[\![r^4]\!] . \tag{40}$$

If we further assume that the system has a uniform density up to a sharp cutoff at radius R, we have

$$[\![r^2]\!] = \tfrac{3}{5} R^2 , \qquad [\![r^4]\!] = \tfrac{3}{7} R^4 , \tag{41}$$

so that

$$\Delta E_2 \lesssim 7\hbar^2/mR^2 . \tag{42}$$

In the next section we see how these results may be obtained much more simply by breaking the translational invariance of the wavefunction.

4. Wavefunctions with Broken Translational Invariance

In Paper I it was pointed out that (i) the orbital angular momentum of a system of particles is independent of the choice of origin if and only if the system has zero total momentum (i.e. its wavefunction is translationally invariant) and (ii) because of the uncertainty principle it is impossible in quantum mechanics to fix the centre of mass of a system (at the origin, for example) when the system has zero total momentum. Thus the only rigorous way to treat the problem of rotational excitations of a self-bound system which is not fixed in space by external forces is to deal exclusively with translationally invariant wavefunctions. We have done so here (up to this point) thus ensuring that the wavefunction considered does correspond to an actual internal excitation, and not to a spurious state.

Having set up the excitation in a translationally invariant way, however, we are free to break the translational invariance of that wavefunction without risk of spurious states; that is, we are sure from its origins that the translationally *variant* wavefunction represents an internal excitation. We find, in a simple calculation, that breaking the translational invariance leads to an error in the energy of order A^{-1}. We are also able to readily obtain the quadrupole moment and the lifetime.

Consider Ψ_{22} given by equation (8). Since

$$\tfrac{1}{2} \sum_{i=1}^{A} \sum_{j=1}^{A} x_{ij}^2 = A \sum_{i=1}^{A} x_i^2 - A^2 X^2 \tag{43}$$

and

$$\sum_{i=1}^{A} \sum_{j=1}^{A} x_{ij} y_{ij} = 2A \sum_{i=1}^{A} x_i y_i - 2A^2 XY, \tag{44}$$

where

$$X = A^{-1} \sum_{i=1}^{A} x_i \quad \text{etc.,} \qquad \mathbf{R} = (X, Y, Z),$$

we have

$$\tfrac{1}{2} \sum_i \sum_j (x_{ij} + iy_{ij})^2 = A \sum_j (x_j + iy_j)^2 - A^2 (X + iY)^2. \tag{45}$$

Similarly

$$\tfrac{1}{2} \sum_i \sum_j (3z_{ij}^2 - r_{ij}^2) = A \sum_j (3z_j^2 - r_j^2) - A^2 (3Z^2 - R^2). \tag{46}$$

Thus when we break the translational invariance by fixing the centre of mass at the origin of the coordinate system $X = 0$, $Y = 0$, $Z = 0$, the wavefunctions Ψ_{22} and Ψ_{20} (equations 8, 9) become

$$\Psi'_{22} = \sum_j (x_j + iy_j)^2 \Phi \equiv F_{22} \Phi, \qquad \Psi'_{20} = \sum_j (3z_j^2 - r_j^2) \Phi \equiv F_{20} \Phi \tag{47, 48}$$

(we have dropped the factor A for simplicity). These states are angular momentum eigenstates as before, with energy ΔE_2 given by (4) above. For the Ψ'_{20} state we have

$$\langle (\nabla_1 F)^2 \rangle = 8 \langle r_1^2 \rangle \tag{49}$$

and

$$\langle F^2 \rangle = A \langle (3z_1^2 - r_1^2)^2 \rangle + A(A-1) \langle (3z_1^2 - r_1^2)(3z_2^2 - r_2^2) \rangle. \tag{50}$$

In the weak correlation approximation (and for spherical Φ) the second term is zero, so we have

$$\langle F^2 \rangle = A \tfrac{4}{5} \langle r_1^4 \rangle. \tag{51}$$

Thus the weak correlation approximation gives

$$\Delta E_2 \leqslant (5\hbar^2/m) \langle r_1^2 \rangle / \langle r_1^4 \rangle. \tag{52}$$

Since these expectation values are taken with the centre of mass fixed at the origin, we have $\langle r^n \rangle = [\![r^n]\!]$ as defined by equation (33), so that the result (52) is the same as (40), and differs from (39) by terms of order A^{-1}. We have thus shown that, in the weak correlation approximation, breaking translational invariance leads to an error of order A^{-1} only, as could be expected.

It is also easy to calculate the quadrupole moment Q in the same approximation. We have

$$Q = e \int d1 \dots dA \sum_{j=1}^{Z} (3z_j^2 - r_j^2) |\Psi'_{22}|^2 \bigg/ \int d1 \dots dA \, |\Psi'_{22}|^2$$

$$= Ze \langle (3z_1^2 - r_1^2)(x_1^2 + y_1^2)^2 \rangle / A \langle (x_1^2 + y_1^2)^2 \rangle$$

$$= -\tfrac{4}{7} Ze A^{-1} \langle r_1^6 \rangle / \langle r_1^4 \rangle, \tag{53}$$

so that the excited state is oblate (pancake-shaped), with a small negative deformation parameter, proportional to A^{-1}. For uniform density up to a cutoff radius R, equation (53) becomes

$$Q = -\tfrac{4}{9} Z e A^{-1} R^2 . \tag{54}$$

We can also estimate the magnetic dipole moment on the assumption that the spin contribution (for even–even nuclei) is negligible. The orbital contribution is, irrespective of translational invariance,

$$\boldsymbol{\mu} = (e/2mc) \sum_{j=1}^{Z} \langle l_j \rangle \approx (e/2mc) Z A^{-1} \langle L \rangle ,$$

so that

$$\mu = (e\hbar/mc) Z A^{-1} , \qquad g = Z A^{-1}$$

in the Ψ_{22} state.

The lifetime of the state is readily obtained from the transition rate (Blatt and Weisskopf 1952, p. 595)

$$T_E(L, M) = \frac{8\pi(L+1)}{L\{(2L+1)!!\}^2} \frac{k^{2L+1}}{\hbar} |\langle f | Q_{LM} | i \rangle|^2 .$$

We have

$$Q_{20} = \tfrac{1}{4}(5/\pi)^{\frac{1}{2}} e \sum_{i=1}^{Z} (3z_i^2 - r_i^2) ,$$

with

$$| i \rangle = N_i^{-1} \sum_j (3z_j^2 - r_j^2) \Phi , \qquad | f \rangle = N_f^{-1} \Phi ,$$

where N_i and N_f are normalization factors. In the weak correlation approximation we find

$$T_E(2, 0) = (\tfrac{1}{75} Z^2 e^2 k^5 / A\hbar) \langle r_1^4 \rangle . \tag{55}$$

From equations (55) and (41), the linewidth Γ of the 2^+ state is

$$\Gamma \approx 1 \cdot 2 (Z^2/A) \Gamma_{\mathrm{W}} , \tag{56}$$

where Γ_{W} is the Weisskopf width (de Shalit and Feshbach 1974, p. 702). Thus the width is large relative to the single-particle value, as befits a collective state.

5. Effect of Interactions on Correlations

We saw in Section 3 that the four-particle term given by equation (30) is zero in the weak correlation approximation, i.e. when the particles are correlated solely because of the finiteness of the self-bound system. The validity of this 'zeroth' approximation needs to be examined in more detail, since the four-particle term in the exact expression (31) for the energy is multiplied by the factor $\tfrac{1}{4}(A-3)$ relative to the three-particle term.

Let the pair interactions be characterized by a range a. If one particle is placed randomly in the system (of radius R) the probability of placing a second particle in the range of interaction with the first is of order $(a/R)^3$. This leads us to expect that, for example,

$$\langle r_{12}^n \rangle - \langle r_{12}^n \rangle_0 \sim \langle r_{12}^n \rangle_0 (a/R)^3 , \tag{57}$$

where $\langle r_{12}^n \rangle_0$ is the expectation value calculated in the weak correlation approximation. A precise formulation in terms of the pair correlation function $g(r)$ gives (c.f. equation 32)

$$\langle r_{12}^n \rangle = \frac{\displaystyle\int_0^\infty dr_1\, r_1\, n(r_1) \int_0^\infty dr_2\, r_2\, n(r_2) \int_{|r_1-r_2|}^{r_1+r_2} dr_{12}\, r_{12}\, g(r_{12})\, r_{12}^n}{\displaystyle\int_0^\infty dr_1\, r_1\, n(r_1) \int_0^\infty dr_2\, r_2\, n(r_2) \int_{|r_1-r_2|}^{r_1+r_2} dr_{12}\, r_{12}\, g(r_{12})}; \tag{58}$$

$\langle r_{12}^n \rangle_0$ is the same expression with $g = 1$. Thus we see that, writing $g = 1+(g-1)$ and using equation (32),

$$\langle r_{12}^2 \rangle = \frac{2\displaystyle\int_0^\infty dr_1\, r_1^2\, n(r_1) \int_0^\infty dr_2\, r_2^2\, n(r_2)(r_1^2+r_2^2) + K_2}{2\left(\displaystyle\int_0^\infty dr\, r^2\, n(r)\right)^2 + K_0}, \tag{59}$$

where

$$K_n \equiv \int_0^\infty dr\, r\, n(r) \int_0^\infty ds\, s\, n(s)\, G_n(r,s), \tag{60}$$

with

$$G_n(r,s) \equiv \int_{|r-s|}^{r+s} dt\, t\{g(t)-1\}\, t^n. \tag{61}$$

We can evaluate G_n explicitly for a simple model where $g = 0$ for $r < a$ and $g = 1$ for $r > a$ (a correlation hole arising out of hard core repulsions):

$$G_n(r,s) = 0, \qquad\qquad\qquad\qquad\qquad |r-s| > a; \tag{62a}$$

$$= -(n+2)^{-1}\{(r+s)^{n+2} - |r-s|^{n+2}\}, \qquad |r-s| < a,\ r+s < a; \tag{62b}$$

$$= -(n+2)^{-1}\{a^{n+2} - |r-s|^{n+2}\}, \qquad |r-s| < a,\ r+s > a. \tag{62c}$$

If we further assume that the density $n(r)$ is a constant n_0 up to $r = R$ and zero for $r > R$, we find

$$K_n = n_0^2\{-\tfrac{2}{3}(n+3)^{-1}R^3 a^{n+3} + \tfrac{1}{2}(n+4)^{-1}R^2 a^{n+4} + O(a^{n+6})\}. \tag{63}$$

Substitution into equation (59) gives

$$\langle r_{12}^2 \rangle = \tfrac{6}{5} R^2\{1+(a/R)^3 + HO(a/R)\}, \qquad \text{while} \qquad \langle r_{12}^2 \rangle_0 = \tfrac{6}{5} R^2, \tag{64}$$

where $HO(a/R)$ indicates terms of higher order in a/R. Thus we have justified the approximation (57) in detail for a specific case.

We now see that, for this simple model, the total four-body term appearing in the result (31) is of the order of the three-body term multiplied by $A(a/R)^3$. For nuclei and helium droplets we have $R = r_0 A^{\frac{1}{3}}$, where r_0 is approximately independent of A. It is thus plausible that the neglected four-body term is smaller than the three-body term by the factor $(a/r_0)^3$. For nuclei the core size is of order $0\cdot4$ fm, with $r_0 \approx 1\cdot2$ fm $\approx 3a$, while for liquid helium $a = 2\cdot56$ Å and $r_0 \approx 0\cdot85\,a$. Thus we expect the above approximations to give reasonable results for nuclei but only a rough estimate for helium microdroplets.

6. Comparison with Experiment

The weak correlation approximation gives the formula (39) for the energy ΔE_2 of the 2^+ state. For large A, and on the assumption of a fairly sharp surface, this result simplifies to $\Delta E_2 \lesssim 7\hbar^2/mR^2$ (equation 42). If we put $R = r_0 A^{\frac{1}{3}}$ with $r_0 = 1 \cdot 2$ fm (Irvine 1972), we have for nuclei

$$\Delta E_2 \lesssim 200 A^{-\frac{2}{3}} \text{ MeV}. \tag{65}$$

This is a large excitation energy for nuclei. For $A = 208$ we have $\Delta E_2 \lesssim 5 \cdot 7$ MeV, while the first 2^+ state of ^{208}Pb is at $4 \cdot 085$ MeV. For all other even–even nuclei, the first 2^+ state is considerably below the bound (65), although the $A^{-\frac{2}{3}}$ trend is roughly followed by spherical nuclei.

There are (at least) three possible explanations of the above discrepancy:

(i) correlations may make the four-body term significant,

(ii) the trial wavefunction we have used needs to be improved, or

(iii) the first 2^+ excited states of most spherical even–even nuclei (except perhaps ^{208}Pb) are not collective rotations of the type described by our wavefunction.

Since four-particle correlations are difficult to discuss rigorously, we have given a heuristic discussion of (i) in the previous section. We can test the explanation (ii) as follows. Consider, instead of Ψ'_{20}, the wavefunction

$$\sum_j (3z_j^2 - r_j^2) f(r_j) \Phi. \tag{66}$$

This wavefunction is also an eigenstate of angular momentum with $L = 2$. We can calculate the expectation value of the energy in this state in the weak correlation approximation, as we did before with $f = 1$. We find

$$\Delta E_2 \leqslant \frac{\hbar^2}{m} \frac{5\langle r^2 f^2 \rangle + 2\langle r^3 f f' \rangle + \frac{1}{2}\langle r^4 f'^2 \rangle}{\langle r^4 f^2 \rangle}, \tag{67}$$

where f' denotes df/dr and the expectation values are calculated in the spherical state Φ as before, with r the distance from the centre of mass. We now optimize with respect to f. A short variational calculation gives the following differential equation to be satisfied by the best f:

$$f'' + (6r^{-1} + n'n^{-1})f' + (q^2 + 2r^{-1}n'n^{-1})f = 0. \tag{68}$$

Here n is again the number density, and $\hbar^2 q^2/2m = \Delta E_2$. For n constant, the regular solution is $f = r^{-2} j_2(qr)$ and so $f = $ const. is a good approximation while qr is small, i.e. r/R small from the result (42). Thus our trial wavefunction with $f = 1$ is good inside the nucleus.

We can turn the problem around and ask: what density $n(r)$ has $f = 1$ as the optimum solution? From equation (68) we find

$$n'n^{-1} = -\frac{1}{2}rq^2,$$

so that

$$n(r) = n(0)\exp(-\tfrac{1}{4}q^2 r^2) \approx n(0)\exp(-7r^2/2R^2). \tag{69}$$

This describes a typical nuclear density variation fairly well, and gives $\langle r^2 \rangle = 3R^2/7$, whereas a sharp boundary has $\langle r^2 \rangle = 3R^2/5$. We thus see that, on both counts, $f = 1$ gives a suitable trial wavefunction, and it is unlikely that the considerable extra mathematical complexity of a general $f(r)$ is warranted.

We conclude then that the collective rotational states studied here are unlikely to be the lowest 2^+ excitations of even–even spherical nuclei (except perhaps for ^{208}Pb, where the energy is of the right order; from equation (56), the width comes to about $38\Gamma_w$ whereas experiment gives approximately $8\Gamma_w$ (Lewis 1971, p. 266)). This conclusion has been reached from first principles.

7. Physical Significance of Proposed States

It was conjectured in Paper I that these new states represented surface oscillations, on the basis of the similarity between the effective moment of inertia,

$$I_2 \geqslant \tfrac{3}{4} Am\langle (x_{12}^2 - y_{12}^2)(x_{34}^2 - y_{34}^2) \rangle / \langle x_{12}^2 + y_{12}^2 \rangle, \tag{70}$$

and the irrotational moment of inertia of an ellipsoid of constant density deformed along the x axis, rotating about the z axis (Gustafson 1955; Katz 1962), namely

$$I_2 = Am\langle x^2 - y^2 \rangle^2 / \langle x^2 + y^2 \rangle \tag{71}$$

(in equation (71) the expectation values are to be taken in the rotating state). However, the similarity of equation (70) to (71) is misleading, for two reasons: firstly because (70) is zero in the weak correlation approximation, and secondly because the considerations of the next paragraph point to a different physical interpretation.

We have seen that on breaking translational invariance of the Ψ_{20} state we get the wavefunction

$$\Psi'_{20} = \sum_j (3z_j^2 - r_j^2)\Phi.$$

This wavefunction has the same form that Feynman (1954, 1972) proposed for excitations in liquid helium, namely $\Psi = F\Phi$ with $F = \sum_j f(r_j)$. The analogy is closer than this similar form however. Feynman showed that, in the bulk, the optimum f is a plane wave $\exp(i\mathbf{k}.\mathbf{r})$. Now the plane wave can be expanded in angular momentum eigenfunctions as

$$\exp(ikr\cos\theta) = \sum_{L=0}^{\infty} (2L+1)i^L j_L(kr) P_L(\cos\theta)$$

and we thus see that the $L = 2$ component of Feynman's wavefunction is

$$\sum_j j_2(kr_j) P_2(\cos\theta_j)\Phi, \tag{72}$$

which we showed to be the optimum wavefunction of the type (66). For small k, the form (72) is just our Ψ'_{20}. Thus the proposed states are finite-system, angular momentum projections of Feynman's excitations.

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THE GIANT DIPOLE RESONANCE

ABSTRACT

A microscopic calculation of the energy of the giant dipole resonance is carried out using the wavefunction proposed by deShalit and Feshbach. The result for the variational bound of the excitation energy (obtained without any adjustable parameters) is

$$E - E_o \lesssim 100\, A^{-1/3}\ MeV$$; which compares well with the experimental large A limit of about $80\, A^{-1/3}\ MeV$. The expectation value of the energy is shown to be determined by the differences in the mean square distances between neutron-neutron, proton-proton and neutron-proton pairs. The nucleon pair correlations are approximated by the ideal Fermi gas correlation functions; Coulomb and other contributions to the pair correlation functions have been neglected. It is shown, however, that the qualitative variation of the energy ($E - E_o \sim A^{-1/3}$) is independent of the nature of the short ranged correlations.

1. Introduction.

DeShalit and Feshbach [1] have written down an approximate wave-function $\Psi_{1M} = D_{1M} \Phi$ for the giant dipole resonance state. Here D is the translationally invariant dipole operator

$$
\begin{aligned}
D &= e \sum_p (r_p - R_{cm}) \\
&= \frac{e}{A} \left[N \sum_p r_p - Z \sum_n r_n \right] \\
&= \frac{e}{A} \sum_p \sum_n r_{pn}
\end{aligned}
\tag{1}
$$

and $\Phi(1...A)$ is the many-body ground state.

The deShalit and Feshbach wavefunction has some points in common with the wavefunctions we introduced [2,3] to describe collective isoscalar rotational modes, namely

$$
\Psi_{LM} = \left\{ \sum_i^A \sum_j^A r_{ij}^L \, Y_{LM}(r_{ij}) \, f(r_{ij}) \right\} \Phi
\qquad \text{(L even)} \tag{2}
$$

These wavefunctions are translationally invariant and have the same permutation symmetry and parity as the ground state Φ. In ref. [2] it is shown that if Φ has L = S = O then Ψ_{LM} is an angular momentum eigenstate $|L, M\rangle$, and further, that when Φ is the ground state of a system of bose particles interating via harmonic pairwise interactions :

$$
V(1...A) = v \sum_{i<j} \sum \left(\frac{r_{ij}}{a} \right)^2 \qquad + \text{constant} \qquad , \tag{3}
$$

then (2) with f = 1 is also an exact energy eigenstate, with eigenvalue

$$
E_L = E_o + L \left(\frac{2\hbar^2 v A}{m a^2} \right)^{1/2} \tag{4}
$$

(an erroneous factor of $\sqrt{2}$ in ref. [2] has been removed). Since (2) makes no distinction between protons and neutrons, Ψ_{LM} describes isoscalar excitations.

The isovector analogues of the wavefunctions (2) are obtained by simply distinguishing between protons and neutrons :

$$\Psi_{LM} = \left\{ \sum_p^Z \sum_n^N r_{pn}^L \, Y_{LM}(\hat{r}_{pn}) \, f(r_{pn}) \right\} \Phi \tag{5}$$

For these wavefunctions L can be any integer unlike for the isoscalar wavefunctions Ψ_{LM}, which are zero when L is odd. Note that the odd L states have opposite parity to the even L states. It is straightforward to show that if Φ has L = S = 0 then Ψ_{LM} is an angular momentum eigenstate $|L,M\rangle$, and further, that for a hypothetical mixture of two kinds of bosons interacting via the harmonic pair potential (3), that Ψ_{LM} with f = 1 is an energy eigenstate with eigenvalue again given by (4).

In this paper we shall investigate only the simplest of the isovector wavefunctions, namely the deShalit and Feshbach state, Ψ_{1M} with f = 1. In order to guarantee that Ψ_{1M} is a rotational eigenstate $|1,M\rangle$ as indicated by the subscripts, we assume that Φ is spherically symmetric and that it has (at least approximately) L = 0 and S = 0 as well as J = 0. So we are restricting Φ to be the ground state of a non deformed even-even nucleus. Since for spherical systems the energy cannot depend on the azimuthal quantum number, the three Ψ_{1M} states are degenerate. We shall evaluate the energy by considering the particular state M = 0 :

$$\Psi_{10} = \sum_p^Z \sum_n^N z_{pn} \, \Phi \tag{6}$$

{aside: For deformed nuclei the Ψ_{1M} states are no longer degenerate and the dipole resonance is split into two or three components depending on whether the nucleus has axial symmetry or not. The splitting between the modes is, to leading order, proportional to the deformation, which means a change of less than 30% for most nuclei [ref. 4) p.490] .}

It is easy to see that Ψ_{10} has the properties of a nuclear

dipole vibration. In particular, the probability $|\psi_{10}|^2$ is largest

when the neutron and proton centres-of-mass are separated, which

implies that the neutron and proton fluids oscillate out of phase,

as was originally proposed by Goldhaber and Teller [5]. Also,

because ψ_{10} has total overlap with the dipole operator it must

exhaust the entire El sum rule, [ref. [1] p.736]. This property

implies that ψ_{10} is an approximate nuclear wavefunction (it would

be almost exact if there were no velocity dependent or charge exchange

nuclear forces), since the giant dipole resonance in medium to large

nuclei is known experimentally to exhaust about one half of the El

sum rule and is even less for the light nuclei. However, since we

neglect charge exchange and velocity dependent interactions in order

to evaluate the energy, the wavefunction (6) is the appropriate

wavefunction to use. Another approximation pointed out by deShalit

and Feshbach is that the real dipole resonance is particle unstable

whereas ψ_{10} is a bound state.

2. A General expression for the energy.

Consider $\Psi = F\Phi$, where Φ is the exact ground state of the nucleus, i.e.

$$H\Phi = E_0 \Phi \qquad , \qquad (7)$$

and

$$F = \sum_{p}^{Z} \sum_{n}^{N} z_{pn} \qquad (8)$$

Then assuming that the potential energy commutes with F (i.e. neglecting charge exchange and velocity dependent interactions), the excitation energy is given by $\left[\text{cf. refs.}^{[2,3]} \right]$

$$\Delta E \equiv E - E_0 \leq \frac{\hbar^2}{2m} \sum_{j=1}^{A} \frac{\langle (\nabla_j F)^2 \rangle}{\langle F^2 \rangle} \qquad (9)$$

The angular brackets denote an expectation value taken in the ground state :

$$\langle B \rangle = \frac{\int d1 \ldots dA \, B \, \Phi^2}{\int d1 \ldots dA \, \Phi^2} \qquad , \qquad (10)$$

where the integral sign includes a sum over spin variables. The expression (9) ignores the difference between the proton and neutron masses, but not the differences in the interactions.

In the numerator of (9) we have

$$(\nabla_j F)^2 = \begin{cases} N^2 & \text{when j denotes a proton} \\ Z^2 & \text{when j denotes a neutron} \end{cases}$$

so that

$$\sum_j \langle (\nabla_j F)^2 \rangle = NZA \qquad (11)$$

In the denominator of (9), we have

$$\langle F^2 \rangle = NZ \left\langle z_{pn}^2 + (N-1) z_{pn} z_{pn'} + (Z-1) z_{pn} z_{p'n} \right.$$
$$\left. + (N-1)(Z-1) z_{pn} z_{p'n'} \right\rangle \qquad (12)$$

Using the identities

$$2 z_{pn} z_{pn'} = z_{pn}^2 + z_{pn'}^2 - z_{nn'}^2$$

$$2 z_{pn} z_{p'n} = z_{pn}^2 + z_{p'n}^2 - z_{pp'}^2$$

$$2 z_{pn} z_{p'n'} = z_{pn'}^2 + z_{p'n}^2 - z_{pp'}^2 - z_{nn'}^2$$

we find

$$\langle F^2 \rangle = \frac{1}{2} N Z \Big\{ Z \langle z_{nn'}^2 \rangle + N \langle z_{pp'}^2 \rangle \tag{13}$$
$$- N Z \langle z_{nn'}^2 + z_{pp'}^2 - 2 z_{pn}^2 \rangle \Big\}$$

By restricting Φ to be the ground state of a non deformed
even-even nucleus we can assume that the ground state neutron and
proton distributions are separately spherically symmetric, so that
each $\langle z_{ij}^2 \rangle$ can be replaced by $\frac{1}{3} \langle r_{ij}^2 \rangle$. The expression (9)
then gives

$$\Delta E \leq \frac{3\hbar^2/m}{\frac{Z}{A} \langle r_{nn'}^2 \rangle + \frac{N}{A} \langle r_{pp'}^2 \rangle - \frac{NZ}{A} \langle r_{nn'}^2 + r_{pp'}^2 - 2 r_{pn}^2 \rangle} \tag{14}$$

3. An approximation that must be avoided.

The evaluation of the expression (14) would be very simple if, having first obtained the correct permutation symmetry with the wavefunction (6), we could use the approximation in which the protons and neutrons are regarded as indistinguishable. In particular, the last term in the denominator would be zero :

$$\left\langle \Gamma_{nn'}^2 + \Gamma_{pp'}^2 - 2\Gamma_{pn}^2 \right\rangle = -2\left\langle \underline{\Gamma}_n.\underline{\Gamma}_{n'} + \underline{\Gamma}_p.\underline{\Gamma}_{p'} - 2\underline{\Gamma}_p.\underline{\Gamma}_n \right\rangle \qquad (15)$$
$$= 0$$

where the last line follows when the correlations between each of the three types of pairs of particles are assumed to be identical. For example, using the weak correlation approximation introduced in ref. [3], where all the correlations in the nucleus are neglected apart from those arising out of the finiteness of the system, we have

$$\left\langle \underline{\Gamma}_i.\underline{\Gamma}_j \right\rangle_0 = \left\langle \underline{\Gamma}_i \right\rangle_0^2 \qquad i \neq j$$
$$= 0 \qquad ,$$

assuming the ground state centre of masses to be at the origin (i.e. $\sum \underline{\Gamma}_i = \underline{0}$), so that in this case the result (15) holds for any radial densities $n_n(r)$ and $n_p(r)$ of the neutron and proton fluids. However, in the approximation (15), the energy would be

$$\Delta E \lesssim \frac{3\hbar^2}{2m\langle r^2\rangle} \simeq \frac{5}{2}\frac{\hbar^2}{mR^2} \qquad (16)$$

(taking a step function density with the same radius R for both the neutrons and protons). With $R = 1.2\,A^{1/3}\,fm$ (16) gives $\Delta E \lesssim 72\,A^{-2/3}\,MeV$, which is lower than the experimental giant dipole energy ($80\,A^{-1/3}\,MeV$) by a factor of about $A^{-1/3}$. Thus the approximation (15) leads to a completely unsatisfactory result. The physical reason for this will

be shown to be, that requiring number conservation to hold for the neutrons and protons separately, implies a significant difference between the expectation value $\langle \Gamma_{pn}^2 \rangle$ and the expectation values $\langle \Gamma_{nn'}^2 \rangle$ and $\langle \Gamma_{pp'}^2 \rangle$.

4. Effect of distinguishing between neutrons and protons.

Physically we expect $\langle \Gamma_{nn'}^2 + \Gamma_{pp'}^2 - 2\Gamma_{p\kappa}^2 \rangle$ to be greater than zero because of (i) the correlation hole between like fermions, and (ii) the Coulomb repulsion between protons. To obtain a reasonable approximation for the value of ΔE, we shall include the effect of (i) by using the spin average of the pair correlation functions g_+ and g_- for like fermions [chapter 2, eqns 5.10 and 5.13 of ref. ')] :

$$g = \frac{3}{4} g_- + \frac{1}{4} g_+$$

which gives

$$g(\Gamma_{12}) = 1 - \frac{9}{2} \left[\frac{j_1(k_F \Gamma_{12})}{k_F \Gamma_{12}} \right]^2 \tag{17}$$

where j_1 is the spherical Bessel function of order one, and k_F is the Fermi wavenumber. The functions $g_\pm$ have been derived for bulk systems, where $k_F^3 = 3\pi^2.$(number density). We shall not attempt here to calculate the exact Fermi correlations in nuclei. Further, we shall assume a uniform density of both the neutron and proton fluids up to a common cutoff radius R (the generalization to separate cutoff radii is simple but any corrections involved are smaller than the effect of assuming a step function density in the first place). So

$$(k_F R)^3 = \begin{cases} \frac{9\pi}{4} N & \text{for neutrons} \\ \frac{9\pi}{4} Z & \text{for protons} \end{cases} \tag{18}$$

Denoting the mean square separation of like fermions by $\langle \Gamma_{12}^2 \rangle$, we use (16) to obtain [cf. eqn. 58 of ref.^{3})]

$$\langle \Gamma_{12}^2 \rangle = \frac{\int_0^R d\Gamma_1 \Gamma_1 \int_0^R d\Gamma_2 \Gamma_2 \int_{|\Gamma_1-\Gamma_2|}^{\Gamma_1+\Gamma_2} d\Gamma \, \Gamma \, g(\Gamma) \Gamma^2}{\int_0^R d\Gamma_1 \Gamma_1 \int_0^R d\Gamma_2 \Gamma_2 \int_{|\Gamma_1-\Gamma_2|}^{\Gamma_1+\Gamma_2} d\Gamma \, \Gamma \, g(\Gamma)} \tag{19}$$

where R is defined by (18). Since we are only including Fermi pair

correlations the value of $\langle \Gamma_{pn}^2 \rangle$ is given by (19) with g = 1, i.e.

$$\langle \Gamma_{pn}^2 \rangle = \langle \Gamma_{pn}^2 \rangle_0 = \frac{6}{5} R^2 \tag{20}$$

The triple integrals in (19) can be reduced to a single one

[see eqns. 9, 11, and 13 of ref. [6])] :

$$\langle \Gamma_{12}^2 \rangle = \frac{\int_0^{2R} dr\, r^4 g(r)\, \gamma(r/2R)}{\int_0^{2R} dr\, r^2 g(r)\, \gamma(r/2R)} \tag{21}$$

where $\gamma(x) = 1 - \frac{3}{2} x + \frac{1}{2} x^3$.

On substituting from (17) and defining $\alpha = 2 k_F R$, we have

$$\langle \Gamma_{12}^2 \rangle = \frac{6}{5} R^2 \left\{ \frac{1 - \frac{360}{\alpha^2} \int_0^1 dy\, y^2 j_1^2(\alpha y)\, \gamma(y)}{1 - \frac{108}{\alpha^2} \int_0^1 dy\, j_1^2(\alpha y)\, \gamma(y)} \right\} \tag{22}$$

Now α is a large parameter (e.g. α_n = 17.8 for N = 100), and so when

calculating the energy it is sufficient to include terms to order α^{-4}

only. This is straightforward but lengthy, so we will give just two

key integrals and the final result. The integrals [7,8])

are

$$\int_0^\infty dx\, j_1^2(x) = \frac{\pi}{6}$$

and

$$\int_0^\infty dx\, x\, j_1^2(x) = \frac{1}{2} J_1^2(1)$$

We calculate

$$\langle \Gamma_{12}^2 \rangle = \frac{6}{5} R^2 \left\{ 1 + 18\pi \alpha^{-3} - \left[108 + 81 J_1^2(1) \right] \alpha^{-4} + O(\alpha^{-5}) \right\}$$

Thus the neutron-neutron and proton-proton mean square separations

are given by, using (18)

$$\langle \Gamma_{nn'}^2 \rangle = \frac{6}{5} R^2 \left\{ 1 + \frac{1}{N} - 0.57\, N^{-4/3} + O(N^{-5/3}) \right\} \tag{23}$$

$$\langle \Gamma_{pp'}^2 \rangle = \frac{6}{5} R^2 \left\{ 1 + \frac{1}{Z} - 0.57\, Z^{-4/3} + O(Z^{-5/3}) \right\}$$

From (20) and (23) we have

$$\left\langle \Gamma_{nn'}^2 + \Gamma_{pp'}^2 - 2\Gamma_{pn}^2 \right\rangle \simeq \frac{6}{5} R^2 \left\{ \frac{A}{NZ} - 0.57 \left(N^{-1/3} + Z^{-1/3} \right) \right\} \quad (24)$$

and so the leading terms in the denominator of the energy (eqn. (14))

cancel exactly. We are left with

$$\Delta E \lesssim \frac{\frac{5\hbar^2}{2mR^2}}{0.57 \frac{NZ}{A} \left(N^{-1/3} + Z^{-1/3} \right)} \quad (25)$$

$$\simeq 100 A^{-1/3} \text{ MeV}.$$

where we have used $R = 1.2 A^{1/3}$ fm. This result is roughly 20%

higher than the experimental value for the medium to large nuclei,

of about $80 A^{-1/3}$ MeV [9].

At first sight it might seem remarkable that such a good result

(especially when compared with (16)) is obtained by including only

those pair correlations due to Fermi statistics, and even then we

used the infinite system pair correlation function. In particular,

the expression (24) is crucial, since it is because the term $\frac{6}{5} R^2 \frac{A}{NZ}$

cancels the dominant part of $\frac{1}{A} \left[Z\langle \Gamma_{nn'}^2 \rangle + N\langle \Gamma_{pp'}^2 \rangle \right]$

in the denominator of (14), that the variation of ΔE with nucleon

number changes from $A^{-2/3}$ to $A^{-1/3}$. This cancellation follows

from the results (20) and (23) for $\langle \Gamma_{pn}^2 \rangle$, $\langle \Gamma_{nn'}^2 \rangle$ and $\langle \Gamma_{pp'}^2 \rangle$.

We will now show that regardless of the nature of the short ranged

correlations, that the requirement of number conservation will lead

to expressions of the form (20) and (23) and hence the result

$\Delta E \sim A^{-1/3}$. First of all, what is the requirement of

number conservation for two-particle expectation values like (19)?

By insisting that the neutrons and protons be separately conserved

(i.e. neglecting charge exchange processes), we have three different

pair densities

$$n_2(\underline{r}_n, \underline{r}_{n'}) \ , \ n_2(\underline{r}_p, \underline{r}_{p'}) \ , \ n_2(\underline{r}_p, \underline{r}_n) \quad (26)$$

where $n_2(\underline{r}_1,\underline{r}_2)\,d\underline{r}_1\,d\underline{r}_2$ is defined as the probability of finding particle one in a volume $d\underline{r}_1$ about $\underline{r}_1$ together with particle two in a volume $d\underline{r}_2$ about $\underline{r}_2$. That is,

$$\int d\underline{r}_n\, d\underline{r}_{n'}\, n_2(\underline{r}_n,\underline{r}_{n'}) = N(N-1) \tag{27a}$$

$$\int d\underline{r}_p\, d\underline{r}_{p'}\, n_2(\underline{r}_p,\underline{r}_{p'}) = Z(Z-1) \tag{27b}$$

$$\int d\underline{r}_p\, d\underline{r}_n\, n_2(\underline{r}_p,\underline{r}_n) = NZ \tag{27c}$$

Defining generalized pair correlation functions by

$$n_2(\underline{r}_1,\underline{r}_2) = n(\underline{r}_1)\,n(\underline{r}_2)\,g(\underline{r}_1,\underline{r}_2) \tag{28}$$

where $n(r)$ is the singlet density (i.e. $\int d\underline{r}_n\, n(\underline{r}_n) = N$, $\int d\underline{r}_p\, n(\underline{r}_p) = Z$), we can write the exact expression for $\langle r_{12}^2 \rangle$ as (cf. (19))

$$\langle r_{12}^2 \rangle = \frac{\int d\underline{r}_1\, n(\underline{r}_1)\int d\underline{r}_2\, n(\underline{r}_2)[1+(g(\underline{r}_1,\underline{r}_2)-1)]\,r_{12}^2}{\int d\underline{r}_1\, n(\underline{r}_1)\int d\underline{r}_2\, n(\underline{r}_2)[1+(g(\underline{r}_1,\underline{r}_2)-1)]} \tag{29}$$

Because r_{12}^2 is maximum for largest separations, where $(g-1)$ tends to zero, the correction terms coming from $(g-1)$ in the numerator are of higher order than the terms coming from $(g-1)$ in the denominator. This was proved above for the particular pair correlation function (17), where the difference between the correction terms is of order $A^{-1/3}$. [See also ref.[3]), where the simple model $g = 0$ for $r < a$ and $g = 1$ for $r > a$, led to a difference between the two correction terms of order $A^{-2/3}$.] So the first two terms of an expansion of (29), arising from the exact short ranged correlations, are given by

$$\langle r_{12}^2 \rangle = \langle r_{12}^2 \rangle_0\left\{1 - \frac{\int d\underline{r}_1\, n(\underline{r}_1)\int d\underline{r}_2\, n(\underline{r}_2)[g(\underline{r}_1,\underline{r}_2)-1]}{\int d\underline{r}_1\, n(\underline{r}_1)\int d\underline{r}_2\, n(\underline{r}_2)} +...\right\} \tag{30}$$

Using the requirement of number conservation, expressed by (27) and (28), we have

$$\langle \Gamma_{nn'}^2 \rangle = \langle \Gamma_{nn'}^2 \rangle_0 \left\{ 1 + \frac{1}{N} + O(N^{-4/3}) \right\}$$

$$\langle \Gamma_{pp'}^2 \rangle = \langle \Gamma_{pp'}^2 \rangle_0 \left\{ 1 + \frac{1}{Z} + O(Z^{-4/3}) \right\} \tag{31}$$

$$\langle \Gamma_{pn}^2 \rangle = \langle \Gamma_{pn}^2 \rangle_0 \left\{ 1 + O(A^{-4/3}) \right\}$$

So (20) with (23) is a particular example of the general result (31), which does not depend on the nature of the short ranged correlations, but rather is a manifestation of number conservation. That is, the expression (14) for the variational energy is rigorously proportional to $1/R$ and not $1/R^2$ as it appears to be from looking at it.

14.

5. Summary and conclusions.

We have shown that the deShalit and Feshbach wavefunction gives a good value for the giant dipole resonance energy. This calculation was carried out from first principles, i.e. with no adjustable parameters, but we have neglected velocity dependent and charge exchange forces. By including only those pair correlations due to Fermi statistics we obtained $\Delta E \lesssim 100 A^{-1/3} MeV$, which is in good agreement with experiment. Justification for assuming that the correlations due to Fermi statistics play a dominant role in the microscopic description of isovector modes, like the giant dipole state, comes from the liquid drop (or hydrodynamic) model $\left[\text{see ref.}^{4)} \text{ p.670, and ref. }^{10)} \right]$. In this very successful phenomenological description of the giant dipole oscillation, the potential energy is taken to be the restoring force between the neutron and proton fluids, which is determined by the symmetry energy in the Weizsäcker mass formula. On page 128 of ref. $^{)}$ it is shown that Fermi statistics, i.e. the Pauli principle, accounts for about one half of the observed symmetry energy. However, it is possible that the good agreement with experiment of our calculation based on the approximate pair correlation function (17), is partly due to the correct normalization involved in (19). That is, (17) does not satisfy exactly the number conservation requirement for a finite nucleus, but by defining the expectation values as in (19) we have imposed the condition $\langle 1 \rangle = 1$, which may remove a significant amount of the error involved in (17).

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Theory of surface excitations of liquid helium four

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ABSTRACT

The wavefunction proposed by Chang and Cohen
for surface excitations of a quantum fluid is used
to calculate the excitation energy $\epsilon(k)$. A general
relation $\epsilon(k) \leq \hbar^2 k^2 / 2m\, S^*(k)$ is obtained, where
S^* (the surface structure factor) is determined in
terms of the density profile and the pair correlation
function. If one assumes that the liquid has
bulk properties right up to the surface, the theory
leads rigorously to $\epsilon(k) = \pi \hbar c k / \left\{ \log\left(\frac{2}{ka}\right) + \text{constant} \right\}$
in the long wavelength limit. This expression is in
qualitative agreement (for intermediate values of k)
with the semiclassical theory of Edwards, Eckardt and
Gasparini, and with the numerical work of Chang and
Cohen. We discuss the proposal that the inclusion
in the ground state of correlations due to the zero
point motion of the surface modes would lead to the
classical dispersion relation $\epsilon \sim k^{3/2}$.

1. INTRODUCTION

Chang and Cohen (1975) have proposed a many-body wavefunction for the surface excitations of liquid He^4 , namely

$$\Psi = \sum_j e^{i\underline{k}.\underline{r}_j} f(z_j) \Phi \qquad (1)$$

where Φ is the ground state wavefunction of the liquid with a free surface in the xy plane, and $\underline{k}$ is parallel to the surface. Chang and Cohen deduce the form of f in the small k limit by the following argument: Feynman (1954, 1972) showed that large-scale motion of a quantum fluid is described by the wavefunction

$$\exp\left\{ i \sum_j s(\underline{r}_j) \right\} \Phi \qquad (2)$$

where the velocity of the motion is

$$\underline{v}(\underline{r}) = \frac{\hbar}{m} Re \, \underline{\nabla} s(\underline{r}) \qquad (3)$$

Thus $(\hbar/m) s$ is the velocity potential of the motion. The classical velocity potential for a surface wave is $\eta \exp(i\underline{k}.\underline{r} + kz)$, where η is proportional to the amplitude. Thus a finite amplitude surface wave would be described by the wavefunction

$$\exp\left\{ i\eta \sum_j \exp(i\underline{k}.\underline{r}_j + kz_j) \right\} \Phi \qquad (4)$$

The wavefunction of a single surface excitation is identified by Chang and Cohen as the term in (4) linear in η , namely

$$\Psi = \sum_j \exp(i\underline{k}.\underline{r}_j + kz_j) \Phi \qquad (5)$$

which has the form (1) with $f(z) = e^{kz}$. Ψ is orthogonal to Φ because it has a different eigenvalue of momentum parallel to the surface, so (from the variational principle) the energy of a surface excitation is given by

$$\epsilon(k) \leq \frac{\int d1 \ldots dN \, \Psi^*(H-E_o)\Psi}{\int d1 \ldots dN |\Psi|^2} \qquad (6)$$

Just as Feynman's wavefunction for phonons is expected to give exactly the energy $\hbar c k$ in the long wavelength limit, so we may expect the small k limit of (6) to be $\hbar w(k)$, where the frequency is given by the classical dispersion relation for capillary waves,

$$w(k) = \left(\frac{\sigma}{nm}\right)^{1/2} k^{3/2}, \qquad (7)$$

where σ is the surface tension, n the number density and m the mass of a helium atom.

Chang and Cohen use the wavefunction (1) with $f(z) = e^{qz}$, and also with $f(z)$ determined by an integro-differential equation derived from the variation principle, the two methods being in numerical agreement. They find that the optinum value of q (treated as a variational parameter) differs somewhat from k, though for $k \leq 0.6 \, \overset{o}{A}^{-1}$ the effect on the excitation energy of replacing q by k is negligible.

We shall obtain a general reduction of the excitation energy to the form $\hbar^2 k^2 / 2m S^*(k)$ for the wavefunction (1), where the surface structure factor S^* is determined as a three-fold integral dependent on the pair correlation function and the density. We then specialize to the

wavefunction (5), and derive an expression for $\epsilon(k)$ on the assumption that the liquid has bulk properties right up to the surface. This leads (without any numerical work) to a dispersion relation which is approximately linear, except at very small k where $\epsilon \sim k/\log k^{-1}$. Chang and Cohen expect the classical dispersion relation $\epsilon \sim k^{3/2}$ to hold when the correlations due to the zero-point motion of the surface excitations are included in the ground state Φ. This proposition is discussed in the last section.

2. AN EXACT EXPRESSION FOR THE EXCITATION ENERGY

We shall evaluate the expectation value of the excitation energy in the state Ψ given by (1), later specializing to (5). It is convenient to write $\Psi = F \Phi$ with

$$F = \sum_{j=1}^{N} e^{i\underline{k}\cdot\underline{r}_j} f(z_j) \tag{8}$$

From (6) we have, using $H\Phi = E_0 \Phi$

$$\epsilon(k) \leq \frac{\hbar^2}{2m} \frac{N\langle |\nabla_1 F|^2\rangle}{\langle |F|^2\rangle} \tag{9}$$

where the expectation values are taken in the ground state:

$$\langle A \rangle = \frac{\int d1\ldots dN\, A\, \Phi^2}{\int d1\ldots dN\, \Phi^2} \tag{10}$$

To keep the analogy with bulk phonons for which $F = \sum_j e^{i\underline{k}\cdot\underline{r}_j}$ and

$$\epsilon(k) \leq \frac{\hbar^2 k^2}{2m S(k)} \tag{11}$$

where $S(k)$ is the bulk structure factor, we write (9) in the form

$$\epsilon(k) \leq \frac{\hbar^2 k^2}{2m S^*(k)} \tag{12}$$

S^* can be called the surface structure factor. From (8) we find

$$N\langle |\nabla_1 F|^2\rangle = \int d1\, n(1)\left[k^2 f^2(z) + \left(df/dz\right)^2\right] \tag{13}$$

where

$$n(1) = \frac{N\int d2\ldots dN\, \Phi^2}{\int d1\ldots dN\, \Phi^2} \tag{14}$$

is the number density, and

$$\langle |F|^2\rangle = \int d1 n(1) f^2(z_1) + \int d1 d2\, n(1,2) \exp(i\underline{k}\cdot\underline{r}_{12}) f(z_1) f(z_2) \quad (15)$$

where

$$n(1,2) = \frac{N(N-1)\int d3\ldots dN\, \Phi^2}{\int d1\ldots dN\, \Phi^2} \quad (16)$$

is the pair density. Thus the surface structure factor is given by

$$S^*(k) = \frac{k^2\langle |F|^2\rangle}{N\langle |\nabla_1 F|^2\rangle}$$

$$\simeq \frac{\int d1 n(1) f^2(z_1) + \int d1 d2\, n(1,2) e^{i\underline{k}\cdot\underline{r}_{12}} f(z_1) f(z_2)}{\int d1 n(1)\left[f^2(z_1) + k^{-2}(df/dz_1)^2\right]} \quad (17)$$

For a fluid with a surface in the xy plane, we can write (except near the walls of the container)

$$n(1,2) = n(z_1) n(z_2) g(r_{12}, z_1, z_2) \quad (18)$$

where g is a generalized pair correlation function. We take the side walls of the container to be at x=0 and L and y=0 and L, and the liquid surface to be at or near z=0. For Ψ to represent a surface wave, $f(z)$ must go to zero deep inside the liquid, so we can extend the z integrations to $-\infty$. Because of the number density factors, the z integrations can also be extended to $+\infty$ provided that when $f(z)$ is taken to grow exponentially as e^{kz}, then k must be less than

$$k_c = \left(-\frac{2m E_0/N}{\hbar^2}\right)^{1/2} \simeq 1.09\, \text{Å}^{-1} \quad (19)$$

since the density goes to zero asymptotically as

$\exp[-2k_c Z]$ (Saam, 1971). Thus we have using (18),

$$S^*(k) = \frac{\int_{-\infty}^{\infty} dz_1 \, n(z_1) f^2(z_1) + \int_{-\infty}^{\infty} dz_1 \, n(z_1) f(z_1) \int_{-\infty}^{\infty} dz_2 \, n(z_2) f(z_2) \, I(z_1, z_2)}{\int_{-\infty}^{\infty} dz \, n(z) \left[f^2(z) + k^{-2} \left(\frac{df}{dz} \right)^2 \right]} \qquad (20)$$

where

$$I(z_1, z_2) = L^{-2} \int_0^L dx_1 \int_0^L dy_1 \int_0^L dx_2 \int_0^L dy_2 \, e^{i \underline{k} \cdot \underline{r}_{12}} \, g(r_{12}, z_1, z_2) \qquad (21)$$

Since $\underline{k}$ is in the xy plane, and

$$\int_0^L dx_1 \int_0^L dx_2 \, \sin k x_{12} \, h(|x_{12}|) = 0 \quad, \qquad (22)$$

we can replace $\exp(i \underline{k} \cdot \underline{r}_{12})$ by $\cos \underline{k} \cdot \underline{r}_{12}$ in (21).

Further, the orthogonality of the excited state Ψ
to the ground state Φ implies $\langle F \rangle = 0$ and thus

$$\int_0^L dx_1 \int_0^L dy_1 \, e^{i \underline{k} \cdot \underline{r}_1} = 0 \qquad (23)$$

(which is satisfied for example by periodic boundary con-
ditions for which $k_x, k_y = \pm 2\pi/L, \pm 4\pi/L, \dots$). This
allows us to replace g by $g-1$ in (21). A further
reduction follows $\overset{from}{\wedge}$ the result (Lekner 1971)

$$\int_0^L dx_1 \int_0^L dx_2 \, h(|x_{12}|) = 2L \int_0^L dx \left(1 - \frac{x}{L} \right) h(x) \qquad (24)$$

Incorporating these three steps in (21) leads to

$$I(z_1, z_2) = 4 \int_0^L dx \left(1 - \frac{x}{L} \right) \int_0^L dy \left(1 - \frac{y}{L} \right) \cos \underline{k} \cdot \underline{\rho} \left[g(r_{12}, z_1, z_2) - 1 \right] \qquad (25)$$

where $\rho^2 = x^2 + y^2 = r_{12}^2 - z_{12}^2$. Provided $g-1$ goes to
zero faster than r_{12}^{-2} as $r_{12} \to \infty$, we can replace L by ∞
in the upper limits and omit the x/L and y/L terms.
Further, since g and $\cos \underline{k} \cdot \underline{\rho}$ are even in x and y ,

we can write

$$I(z_1, z_2) = \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy \, \cos \underline{k} \cdot \underline{\rho} \left[g(r, z_1, z_2) - 1 \right]$$

$$= \int_0^{\infty} d\rho \, \rho \left[g(r, z_1, z_2) - 1 \right] \int_0^{2\pi} d\phi \, \cos \left[k\rho \cos(\phi - \theta) \right] \quad (26)$$

$$= 2\pi \int_0^{\infty} d\rho \, \rho \, J_0(k\rho) \left[g(r, z_1, z_2) - 1 \right]$$

$$= 2\pi \int_{|z_{12}|}^{\infty} dr \, r \, J_0(k\sqrt{r^2 - z_{12}^2}) \left[g(r, z_1, z_2) - 1 \right]$$

We have thus shown that the excitation energy is given

by $E(k) \leq \hbar^2 k^2 / 2m \, S^*(k)$,

where

$$S^*(k) = \frac{\int_{-\infty}^{\infty} dz \, n(z) f^2(z) + 2\pi \int_{-\infty}^{\infty} dz_1 \, n(z_1) f(z_1) \int_{-\infty}^{\infty} dz_2 \, n(z_2) f(z_2) \int_0^{\infty} d\rho \, \rho \, J_0(k\rho) \left[g(r, z_1, z_2) - 1 \right]}{\int_{-\infty}^{\infty} dz \, n(z) \left[f^2(z) + k^{-2} (df/dz)^2 \right]}$$

(27)

The excitation energy is therefore real and independent

of the surface area (or indeed of all macroscopic dimensions

of the system). The above expression is an improvement

of that obtained by Chang and Cohen (eqn. 9) in two

ways: a four-fold integral has been reduced to a three-

fold integral, and g has been replaced by $g-1$.

In the remainder of this paper we shall take

$f = e^{kz}$, which (as discussed in the introduction) has

direct theoretical connection with classical surface waves.

For this f , the excitation energy is given by (12) with

$2S^* = 1 + \alpha$, where

$$\alpha(k) = \frac{2\pi \int_{-\infty}^{\infty} dz_1 \, n(z_1) e^{kz_1} \int_{-\infty}^{\infty} dz_2 \, n(z_2) e^{kz_2} \int_0^{\infty} d\rho \, \rho \, J_0(k\rho) \left[g(r, z_1, z_2) - 1 \right]}{\int_{-\infty}^{\infty} dz \, n(z) e^{2kz}}$$

(28)

In this case the energy is manifestly independent of

the z coordinate of the surface.

3. AN APPROXIMATE REDUCTION

To proceed further we shall approximate the density and pair correlation function as follows:

$$n(z) = \begin{cases} n_0 & z < 0 \\ 0 & z > 0 \end{cases} \qquad (29)$$

$$g(r, z_1, z_2) = g_0(r) , \qquad (30)$$

where n_0 and g_0 are the density and the pair correlation function in the bulk liquid. These approximations together amount to ignoring the structure of the liquid-vacuum interface, or (equivalently) to assuming that knowledge of the bulk properties is sufficient to determine the dispersion relation for surface excitations. We shall show rigorously that unsatisfactory results come from these assumptions.

Using the result

$$\int_0^\infty dz_1\, e^{-kz_1} \int_0^\infty dz_2\, e^{-kz_2}\, h(|z_{12}|) \qquad (31)$$

$$= k^{-1} \int_0^\infty dz\, e^{-kz}\, h(z)$$

and the approximations (29) and (30), we find

$$\alpha(k) = 4\pi n_0 \int_0^\infty dz\, e^{-kz} \int_0^\infty d\rho\, \rho\, J_0(k\rho)\left[g_0(r) - 1\right]$$

$$= 4\pi n_0 \int_0^\infty dz\, e^{-kz} \int_z^\infty dr\, r\, J_0(k\sqrt{r^2 - z^2})\left[g_0(r) - 1\right] \qquad (32)$$

$$= 4\pi n_0 \int_0^\infty dr\, r\left[g_0(r) - 1\right] \int_0^r dz\, e^{-kz}\, J_0(k\sqrt{r^2 z^2})$$

In the appendix we show that

$$\int_0^y dx\, e^{-x}\, J_0(\sqrt{y^2 - x^2}) = y\left\{1 - \int_0^y dx\, x^{-1}\, J_1(x)\right\} , \qquad (33)$$

which enables us to rewrite (32) as

$$\alpha(k) = 4\pi n_o \int_0^\infty dr\, r^2 \left[g_o(r) - 1\right] \left\{1 - \int_0^{kr} dx\, x^{-1}\, J_1(x)\right\}$$

$$= -1 - 4\pi n_o \int_0^\infty dr\, r^2 \left[g_o(r) - 1\right] \int_0^{kr} dx\, x^{-1}\, J_1(x) \;,$$ (34)

where we have used the result that the bulk structure

factor

$$S(k) = 1 + 4\pi n_o \int_0^\infty dr\, r^2 \left[g_o(r) - 1\right] \frac{\sin kr}{kr}$$

goes to zero as $k \to 0^+$. The excitation energy

has thus been reduced to

$$E(k) \lesssim \frac{\hbar^2 k^2}{m} \Big/ 4\pi n_o \int_0^\infty dr\, r^2 \left[1 - g_o(r)\right] \int_0^{kr} dx\, x^{-1}\, J_1(x)$$ (35)

4. THE LONG WAVELENGTH LIMIT

The asymptotic form of the bulk pair correlation function is known to be (see for example Feenberg 1970, p. 689)

$$g_0 \longrightarrow 1 - \frac{\hbar}{2\pi^2 n_0 mc} r^{-4} \qquad (36)$$

Let us assume that this is attained for $r > a$, where a would typically be several atomic diameters (say $a = 10\overset{\circ}{A}$). Then we can rewrite the double integral in (35) as the sum of a short-range and a long-range contribution:

$$\int_0^\infty dr\, r^2 \left[1 - g_0(r)\right] \int_0^{kr} dx\, x^{-1} J_1(x)$$

$$= \int_0^a dr\, r^2 \left[1 - g_0(r)\right] \int_0^{kr} dx\, x^{-1} J_1(x) \qquad (37)$$
$$+ \hbar/2\pi^2 n_0 mc \int_a^\infty dr\, r^{-2} \int_0^{kr} dx\, x^{-1} J_1(x)$$

The last double integral is equal to

$$\frac{1}{a} \int_0^{ka} dx\, x^{-1} J_1(x) + k \int_{ka}^\infty dx\, x^{-2} J_1(x) ,$$

of which the second term varies as $-k\log ka$ for small ka. Using $J_1 = x(J_0 - J_1')$, we find

$$\int_0^y dx\, x^{-1} J_1(x) + y \int_y^\infty dx\, x^{-2} J_1(x) = \int_0^y dx\, J_0(x) - \frac{1}{2} J_1(y) + \frac{y}{2} \int_y^\infty dx\, x^{-1} J_0(x)$$

The precise form of the logarithmic variation is given by (Abramowitz and Stegun 1964, p. 481)

$$\int_y^\infty dx\, x^{-1} J_0(x) = \log \frac{2}{y} - \gamma + \left(1 - 2 y^{-1} J_1(y)\right)$$

where $\gamma = \cdot577...$ is Euler's constant. Thus the limiting form of (37) is

$$\frac{\hbar k}{4\pi^2 n_0 mc} \left\{ \log\left(\frac{2}{ka}\right) + \frac{3}{2} - \gamma \right\} + \frac{1}{2} k \int_0^a dr\, r^3 \left[1 - g_0(r)\right] + O(ka)^2 ,$$

and the long wavelength form of the excitation energy becomes (from (35)):

$$\epsilon(k) \lesssim \hbar c k \Big/ \frac{1}{\pi} \left\{ \log\left(\frac{2}{ka}\right) + \frac{3}{2} - \gamma \right\} + \frac{2\pi n_0 mc}{\hbar} \int_0^a dr\, r^3 \left[1 - g_0(r)\right] \qquad (38)$$

Our assumptions (29) and (30) thus lead to the dispersion relation $\omega(k) \simeq \pi c k / \log\left(\frac{2}{ka}\right)$ at very small ka. However, for moderate k, (38) predicts an almost linear, phonon-like, dispersion relation. To estimate the relative size of the terms in (38), one can take $a \simeq d$ (the hard core diameter) and $g_0 = 0$ for $r < d$. Then the denominator becomes

$$\frac{1}{\pi}\left\{ \log\left(\frac{2}{kd}\right) + \frac{3}{2} - \gamma \right\} + \frac{\pi}{2} \frac{n_0 mc}{\hbar} d^4$$

With $d = 2.6 \overset{\circ}{A}$ and k measured in $\overset{\circ}{A}^{-1}$, these terms have the numerical magnitude

$$\frac{1}{\pi}\left\{ \log k^{-1} + 0.66 \right\} + 2.36$$

and thus the logarithmic term becomes dominant only when $k \lesssim 10^{-4} \overset{\circ}{A}^{-1}$. If we ignore the long-range contributions altogether, the excitation energy becomes $\epsilon \simeq \hbar c k / 2.4$, which is in qualitative agreement with Chang and Cohen, whose numerical results for $0.2 \overset{\circ}{A}^{-1} \le k \le 1 \overset{\circ}{A}^{-1}$ are closely approximated by $\hbar ck/2.0$, and with the semiclassical results of Edwards, Eckardt and Gasparini (1974) in the region $0.5 \overset{\circ}{A}^{-1} \le k \le 1.5 \overset{\circ}{A}^{-1}$.

5. DISCUSSION

We have seen that if the bulk properties of liquid helium are assumed to hold right up to the surface, the wavefunction (5) gives an excitation energy which is not

$$E(k) = \hbar \left(\frac{\sigma}{n_0 m} \right)^{1/2} k^{3/2} \tag{39}$$

in the long-wavelength limit, but lies closer to a phonon-like dispersion relation. Chang and Cohen assert that the correct limiting behaviour (39) will be obtained when one includes in the ground state the correlations due to the zero-point motion of the surface excitations. Their assertion is made on the basis of the analogy with bulk excitations, where the long-range behaviour of $g_0(r)-1$ is determined by the existence of phonon excitations and the Feynman relation (11), which implies that $S(k) \rightarrow \frac{\hbar k}{2mc}$ as $k \rightarrow 0$. In the surface case, (39) and (12) similarly imply

$$S^*(k) \longrightarrow \frac{\hbar}{2} \left(\frac{n_0}{\sigma m} \right)^{1/2} k^{1/2} \tag{40}$$

One can obtain this result by using our approximations (29) and (30) and postulating the asymptotic behaviour of the pair correlation function to be

$$g(r) \longrightarrow 1 - \frac{3}{8 \, \Gamma^2(1/4)} \left(\frac{\hbar^2}{\sigma n_0 m} \right)^{1/2} r^{-7/2} \tag{41}$$

instead of (36). We are not suggesting that (41) is correct, but it gives a clue as to what the exponent in the asymptotic value of $g(r, z_1, z_2) - 1$ might be

when the effect of surface excitations is included
in the ground state. In fact (41) must be too simple,
since we know that near the surface g is a function
of z_1 and z_2 as well as of r_{12}. Also, the expression
for the zero-point motion correlations in the wavefunction
written down by Chang and Cohen (their eqns (26) and
(27)) is a function of the variables $\rho_{ij} = \sqrt{r_{ij}^2 - z_{ij}^2}$
and $z_i + z_j$.

In the case of bulk excitations it was possible to
go directly from $S(k) \rightarrow \hbar k / 2mc$ to the asymptotic form
of $g - 1$. The surface case is more complicated: (40)
cannot be inverted to obtain the asymptotic form of
$g(r, z_1, z_2) - 1$ without knowledge of the density profile,
since (28) involves $n(z)$ as well as g. The complete
analysis would thus have to run as follows - (i) obtain
the contribution of the zero-point motion of the surface
excitations to the ground state wavefunction.
(ii) evaluate $n(z)$ and $g(r, z_1, z_2)$ for this wavefunction.
(iii) evaluate $S^*(k)$ from (28) and check that it has
the correct limiting form (40). This programme has
been started by Chang and Cohen (step (i)), but will
require a lot of analytical work to complete. In
particular, the statistical mechanics involved in
step (ii) is formidable.

APPENDIX

We first obtained the result (33) (which appears
to be new) in the form

$$\int_0^y dx\, e^{-x} J_0\left(\sqrt{y^2-x^2}\right) = y \int_y^\infty dx\, x^{-1} J_1(x) \qquad \text{(A1)}$$

by expanding the integrand on the left and summing the double series. In the shorter
proof which follows we take the Laplace transform
of both sides, following the suggestion of Dr Graeme
Wake. The right side becomes

$$\int_0^\infty dy\, e^{-py} y \int_y^\infty dx\, x^{-1} J_1(x)$$

$$= -\frac{d}{dp} p^{-1} \int_0^\infty dx\, x^{-1}\left(1-e^{-px}\right) J_1(x) \qquad \text{(A2)}$$

$$= \frac{1}{\sqrt{p^2+1}\left(1+\sqrt{p^2+1}\right)}$$

where we have used the result (Watson 1944, p. 386)

$$\int_0^\infty dx\, x^{-1} e^{-px} J_1(x) = \sqrt{p^2+1} - p$$

On the left side of (A1) we use the Bessel formula
(Watson, p. 21)

$$J_0\left(\sqrt{y^2-x^2}\right) = \frac{1}{2\pi} \int_{-\pi}^{\pi} d\phi\, e^{x\cos\phi + iy\sin\phi}$$

to obtain

$$\frac{1}{2\pi} \int_{-\pi}^{\pi} d\phi\, e^{iy\sin\phi} \frac{1-e^{-y(1-\cos\phi)}}{1-\cos\phi} \qquad \text{(A3)}$$

The Laplace transform of (A3) is

$$\frac{1}{2\pi} \int_{-\pi}^{\pi} d\phi\, \frac{1}{(p-i\sin\phi)(p-i\sin\phi+1-\cos\phi)} \qquad \text{(A4)}$$

On changing to the variable $z = e^{i\phi}$ this transforms
to the contour integral (around the unit circle)

$$\frac{1}{\pi i} \oint dz \; \frac{1}{(z^2 - 2pz - 1)(z - p - 1)} \qquad \text{(A5)}$$

Of the three poles, two lie outside the unit circle
and one inside for $p > 0$. Evaluating the residue
we find (A5) reduces to (A2).

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SURFACE TENSION AND ENERGY OF A CLASSICAL LIQUID-VAPOUR

INTERFACE

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ABSTRACT

Rigorous general expressions for the surface tension σ
and the surface energy per unit area ϵ are derived in
the form of three-fold integrals.   In the approximation
$$n_2(\vec{r}_1,\vec{r}_2) = n(z_1)n(z_2)g(r_{12},\bar{n})$$ we obtain the
following results: (i) Both σ and ϵ are proportional to
$(n_\ell - n_v)^2$. (ii) The expressions for σ and ϵ are formally
reduced to a single integral, with integrands determined
in terms of the density profile $n(z)$. (iii) Explicit
expressions are given for an exponential density variation.
(iv) In the limit of a density variation which is slow on
the scale of the molecular diameter, we derive the
general expressions $\sigma = A(n_\ell - n_v)^2/\lambda$, $\epsilon = B(n_\ell - n_v)^2\lambda$
from the microscopic theory (λ is a measure of the surface
thickness).   The same forms for σ and ϵ follow from
(iii), with explicit expressions for A and B.   These forms
for σ and ϵ are shown to be very good approximations
even well away from the critical point.   It is argued
that the critical power laws have the same range of validity.
(v) The critical exponents of A and B are determined, that
for A agreeing with the result of Fisk & Widom.   (vi) The
surface thickness is determined for Ar, Kr and Xe
near their triple points, using our theory for ϵ and

experimental data on the bulk energy of the liquids.
The results are in excellent agreement with other
estimates.

Similar results are obtained with the direct correlation
function expression for the surface tension: the general
expression is reduced to a three-fold integral, and
results analogous to (i) through (iv) are obtained in the
approximation $C(\underline{r}_1,\underline{r}_2) = C(r_{12},\bar{n})$. The equivalence of
the $C(\underline{r}_1,\underline{r}_2)$ and $g(\underline{r}_1,\underline{r}_2)$ formulations for σ is proved
in the low density limit.

1.   SUMMARY OF RESULTS

By using the thermodynamic definition $\sigma = \left(\partial F / \partial A\right)_{V,T}$ of surface tension, and by extracting that part of the total energy which is proportional to the surface area, we obtain, for a liquid-vapour interface lying in the xy plane, and assuming the total potential energy has the form $U(\vec{r}_1, \dots \vec{r}_N) = \sum_{i<j}^{N}\sum u(r_{ij})$,

$$'\sigma' = \frac{\pi}{2}\int_0^L dz_1 \int_0^L dz_2 \int_{|z_{12}|}^{\infty} dr\, n_2(r, z_1, z_2) \frac{du}{dr}\left(r^2 - 3z_{12}^2\right) \tag{25}$$

and

$$'\varepsilon' = \pi \int_0^L dz_1 \int_0^L dz_2 \int_{|z_{12}|}^{\infty} dr\, n_2(r, z_1, z_2)\, r\, u(r) \tag{29}$$

(The reason for the quotation marks is explained in the text). In the approximation

$$n_2(r, z_1, z_2) = n(z_1) n(z_2) g(r, \bar{n})$$

we derive from (25) and (29) the expressions

$$\sigma = \frac{\pi}{2}(n_\ell - n_v)^2 \int_0^{\infty} dr\, g(r, \bar{n})\frac{du}{dr}\left[\frac{1}{4}r^4 + p_1(r) + p_2(r)\right] \tag{50}$$

and

$$\varepsilon = \pi(n_\ell - n_v)^2 \int_0^{\infty} dr\, g(r, \bar{n})\, r\, u(r)\left[-\frac{1}{2}r^2 + q_1(r) + q_2(r)\right] \tag{69}$$

where the functions p_i and q_i are determined in terms of the density n(z).   For an exponential density variation (characteristic length λ), these formulae give (with $F = g\frac{du}{dr}$ and h = rgu)

$$\sigma = \frac{\pi}{2}(n_\ell - n_v)^2 \int_0^{\infty} dr\, F(r)\left\{\frac{1}{4}r^4 + \lambda^4\left[18 - 2\frac{r^2}{\lambda^2} - e^{-r/\lambda}\left(18 + 18\frac{r}{\lambda} + 7\frac{r^2}{\lambda^2} + \frac{r^3}{\lambda^3}\right)\right]\right\} \tag{77}$$

and

$$\varepsilon = -\frac{\pi}{2}(n_\ell - n_v)^2 \int_0^{\infty} dr\, h(r)\left\{r^2 + \lambda^2\left[4 - e^{-r/\lambda}\left(4 + \frac{r}{\lambda}\right)\right]\right\} \tag{78}$$

When λ is large these formulae simplify to

$$\sigma = \frac{\pi}{30}(n_\lambda - n_v)^2 \frac{1}{\lambda} \int_0^\infty dr\, F(r)\, r^5 \tag{80}$$

$$\epsilon = -\frac{3\pi}{2}(n_\lambda - n_v)^2 \lambda \int_0^\infty dr\, h(r)\, r \tag{81}$$

A comparison of integrands shows that (80) and (81) are good approximations to (77) and (78) even when λ equals the core diameter. An alternative treatment for slowly varying density, based on a Taylor expansion of the density, gives

$$\sigma = A\,(n_\lambda - n_v)^2 / \lambda \tag{84}$$

and

$$\epsilon = B\,(n_\lambda - n_v)^2 \lambda \tag{90}$$

where the coefficients A and B reduce to those of (80) and (81) for an exponential density variation.

In the limit $\lambda \to 0$ the expressions for σ and ϵ are shown to be thermodynamically consistent (i.e. to satisfy $\epsilon = \sigma - T\frac{d\sigma}{dT}$) for a dilute gas - wall interface. The thermodynamic relation between σ and ϵ leads to an equation for their critical exponents μ, μ'; namely

$$\mu' = \mu - 1 \tag{97}$$

This result is also derived by an extension of an argument due to Widom. The critical exponents α', β', ν and γ of A, B, λ^{-1} and χ_T^{-1} are shown to satisfy

$$\alpha' = \gamma - 2\nu \tag{103} \quad ; \quad \beta' = \gamma - 1 \tag{104}$$

The surface thickness ($\simeq 2\lambda$) of Ar, Kr and Xe is found from (81) and experimental data to be slightly larger than the molecular diameter near the triple point, in excellent agreement with the Egelstaff and Widom estimate.

Finally, similar results are obtained using the direct correlation function theory for the surface tension. The general expression corresponding to (25) is

$$\sigma_c = \frac{\pi}{2} T \int_{-\infty}^{\infty} dz_1 \frac{dn(z_1)}{dz_1} \int_{-\infty}^{\infty} dz_2 \frac{dn(z_2)}{dz_2} \int_{|z_{12}|}^{\infty} dr\, r\, C(r, z_1, z_2)(r^2 - z_{12}^2) \quad (110)$$

When the direct correlation function is approximated by $C(r_{12}, \bar{n})$,

$$\sigma_c = \frac{\pi}{2} T \int_0^{\infty} dr\, r\, c(r, \bar{n}) \int_{-\infty}^{\infty} dz_1 \frac{dn(z_1)}{dz_1} \int_{z_1-r}^{z_1+r} dz_2 \frac{dn(z_2)}{dz_2}(r^2 - z_{12}^2) \quad (113)$$

For an exponential density variation,

$$\sigma_c = \frac{\pi}{2} T (n_\ell - n_v)^2 \int_0^{\infty} dr\, r\, c(r, \bar{n}) \left\{ r^2 - 4\lambda^2 + \lambda^2 e^{-r/\lambda}\left(4 + 4\frac{r}{\lambda} + \frac{r^2}{\lambda^2}\right) \right\} \quad (115)$$

which becomes

$$\sigma_c = \frac{\pi}{6} T (n_\ell - n_v)^2 \frac{1}{\lambda} \int_0^{\infty} dr\, r^4 c(r, \bar{n}) \quad (116)$$

in the large λ limit. Corresponding to (84) we show that, for slowly varying density, $\sigma_c \doteq A_c (n_\ell - n_v)^2 / \lambda$. The pair correlation and the direct correlation theories are proved to be equivalent to the lowest order in density, but the question of their equivalence in general is left open.

2. GENERAL EXPRESSIONS FOR SURFACE TENSION AND SURFACE ENERGY.

Basically, there are two definitions of surface tension. One is the mechanical definition of σ as the stress transmitted across a strip of unit width normal to the interface, and the other is the thermodynamic definition as the isothermal change in the Helmholtz free energy during the formation (at constant volume) of a unit area of surface. The Kirkwood and Buff [1] general treatment used the mechanical definition, and their result was shown to be obtainable from the thermodynamic definition by Buff [2] and MacLellan [3]. These treatments arrive at a general expression for σ as a four-fold integral. We will give a simple derivation using the thermodynamic definition of σ and based on the deformation method of Harasima [4] and Toda [5]. This method is mathematically equivalent to that used by Buff and MacLellan, but is physically more transparent. We reduce the general expression for σ to a three-fold integral, and in addition obtain a general three-fold integral expression for the surface energy ε. (For some interesting alternative formulations of surface tension theory, see [6, 7, 8])

Fig. 1.

From now on we consider a two phase classical system of N atoms inside the box of figure 1. In the calculation of σ and ε we assume that the only effect of gravity is to create a flat liquid-vapour interface.

(Gravitational pressure gradients can be neglected *experimentally* for all temperatures such that $|T_c - T|/T_c$ is greater than about 10^{-5}; see Kadanoff et al [9] , p.421)

To change the area of the liquid-vapour interface without changing the total volume or the volume of either phase, we expand the box from L_1 to $L_1(1+\xi)$ in the x-direction, and contract it from L_3 to $L_3(1+\xi)^{-1}$ in the z-direction, leaving L_2 unchanged. The deformation is performed isothermally. The increase in the interface area is

$$\Delta A = L_1 L_2 (1 + \xi) - L_1 L_2 = \xi L_1 L_2 \qquad (1)$$

From the thermodynamic definition of surface tension, σ is contained within

$$"\sigma" = \lim_{\Delta A \to 0} \frac{\Delta F}{\Delta A} \quad , \qquad (2)$$

where ΔF is the increase in the Helmholtz free energy. The reason for the inverted commas around σ is that ΔF also contains terms due to the change in the liquid-wall and vapour-wall interfacial areas, and thus gives more than just the surface free energy of the liquid-vapour interface. We will continue to use this notation throughout the paper, finally doing away with the inverted commas when we have identified and discarded all the parts of ΔF which are not true interface terms. This procedure is required by the mathematical rigour which this subject demands (specifically by the need to keep the system under consideration

finite so that interchange of order of integration can be done rigorously).

The free energy of the original system is given by ([10], Ch.2, problem 33)

$$\exp\left(-\frac{F}{T}\right) = \frac{1}{N!}\left(\frac{mT}{2\pi\hbar^2}\right)^{\frac{3N}{2}} \int_0^{L_1}dx_1 \dots \int_0^{L_3}dz_N \exp\left(-\frac{U(x_1 \dots z_N)}{T}\right) \qquad (3)$$

where U is total potential energy, and the temperature T is given the dimension of energy. The free energy of the deformed system is given by

$$\exp\left(-\frac{F'}{T}\right) = \frac{1}{N!}\left(\frac{mT}{2\pi\hbar^2}\right)^{\frac{3N}{2}} \int_0^{L_1(1+\xi)}dx_1 \dots \int_0^{L_3(1+\xi)^{-1}}dz_N \exp\left(-\frac{U(x_1 \dots z_N)}{T}\right) \qquad (4)$$

If we introduce the scaled coordinates

$$x' = \frac{x}{1+\xi} \quad , \quad y' = y \quad , \quad z' = z(1+\xi) \qquad (5)$$

into (4), then integrations in the new coordinates have the same limits as in (3). The Jacobian of this change of variables is unity, so

$$\exp\left(-\frac{F'}{T}\right) = \frac{1}{N!}\left(\frac{mT}{2\pi\hbar^2}\right)^{\frac{3N}{2}} \int_0^{L_1}dx_1' \dots \int_0^{L_3}dz_N' \exp\left\{-\frac{U(x_1'(1+\xi) \dots z_N'(1+\xi)^{-1})}{T}\right\} \qquad (6)$$

If we now set $F' = F + \Delta F$ and

$$U(x_1(1+\xi) \dots z_N(1+\xi)^{-1}) = U(x_1 \dots z_N) + \Delta U \quad , \qquad (7)$$

then to lowest order

$$\Delta F = \frac{\int_0^{L_1}dx_1 \dots \int_0^{L_3}dz_N \, \Delta U \exp\left(-\frac{U}{T}\right)}{\int_0^{L_1}dx_1 \dots \int_0^{L_3}dz_N \exp\left(-\frac{U}{T}\right)} = \langle \Delta U \rangle \qquad (8)$$

(The last equality defines expectation value brackets). Note that the change in the free energy produced by this isothermal, constant volume deformation has the appearance of a change in the total energy of the system (the kinetic energy

is isotropic in classical fluids, and is constant in isothermal deformations). This is deceptive, however: the total thermodynamic energy E is (for monatomic systems),

$$E = \frac{3}{2}NT + \frac{\int_0^{L_4}dx_1 \dots \int_0^{L_3}dz_N\, U \exp(-U/T)}{\int_0^{L_4}dx_1 \dots \int_0^{L_3}dz_N\, \exp(-U/T)} \tag{9}$$

$$= \frac{3}{2}NT + \langle U \rangle$$

By the above arguments we find, for the same isothermal deformation,

$$\Delta E = \langle \Delta U \rangle + \frac{1}{T}\{\langle U \rangle\langle \Delta U \rangle - \langle U \Delta U \rangle\} \tag{10}$$

so that

$$\Delta E = \Delta\langle U \rangle \neq \langle \Delta U \rangle \tag{11}$$

The second term in (10) gives the entropy change caused by the deformation, since

$$E = F + TS, \quad \Delta E = \Delta F + T\Delta S \qquad \text{(isothermal change)} \tag{12}$$

Returning to the expression (8) for ΔF, we see from (7) that

$$\Delta U = \xi \sum_{j=1}^{N}\left(x_j\frac{\partial U}{\partial x_j} - z_j\frac{\partial U}{\partial z_j}\right) + O(\xi^2) \tag{13}$$

so that

$$\langle \Delta U \rangle = N\xi\left\langle x_1\frac{\partial U}{\partial x_1} - z_1\frac{\partial U}{\partial z_1}\right\rangle + O(\xi^2) \tag{14}$$

We now specialize to central pairwise forces, that is

$$U = \sum_{i<j}^{N}\sum^{N} u(r_{ij}), \quad r_{ij} = |\underline{r}_i - \underline{r}_j|$$

Then

$$\left\langle x_1\frac{\partial U}{\partial x_1}\right\rangle = (N-1)\left\langle x_1\frac{\partial u(r_{12})}{\partial x_1}\right\rangle$$

$$= (N-1)\left\langle \frac{x_1 x_{12}}{r_{12}}\frac{du(r_{12})}{dr_{12}}\right\rangle \tag{15}$$

$$= \frac{1}{2}(N-1)\left\langle \frac{x_{12}^2}{r_{12}}\frac{du(r_{12})}{dr_{12}}\right\rangle$$

where the last line follows from the equality of $\left\langle x_1 \frac{\partial u}{\partial x_1} \right\rangle$ and $\left\langle x_2 \frac{\partial u}{\partial x_2} \right\rangle$. We thus have

$$\langle \Delta u \rangle = \zeta \frac{1}{2} N(N-1) \left\langle \frac{x_{12}^2 - z_{12}^2}{r_{12}} \frac{du(r_{12})}{dr_{12}} \right\rangle + O(\zeta^2)$$

(16)

which we can write as

$$\langle \Delta u \rangle = \zeta \frac{1}{4} N(N-1) \left\langle \frac{r_{12}^2 - 3z_{12}^2}{r_{12}} \frac{du(r_{12})}{dr_{12}} \right\rangle + O(\zeta^2)$$

(17)

because of the physical equivalence of the x and y directions. In terms of the pair density n_2 defined by

$$n_2(\underline{r}_1, \underline{r}_2) = \frac{N(N-1) \int d\underline{r}_3 \ldots \int d\underline{r}_N \exp(-U/_T)}{\int d\underline{r}_1 \ldots \ldots \int d\underline{r}_N \exp(-U/_T)}$$

(18)

the surface tension becomes

$$"\sigma" = \lim_{\zeta \to 0} \frac{\langle \Delta u \rangle}{\zeta L_1 L_2}$$

$$= \frac{1}{4 L_1 L_2} \int d\underline{r}_1 \int d\underline{r}_2 \, n_2(\underline{r}_1, \underline{r}_2) \frac{r_{12}^2 - 3z_{12}^2}{r_{12}} \frac{du(r_{12})}{dr_{12}}$$

(19)

(This is easily transformed into the general expression of Kirkwood and Buff [1]). We can simplify this result by making use of the symmetry of the system, which implies that everywhere except near the walls of the box n_2 is function of only three variables r_{12}, z_1 and z_2 :

$$n_2(\underline{r}_1, \underline{r}_2) = n_2(r_{12}, z_1, z_2)$$

(20)

Then making use of the identity [11]

$$\int_0^L dx_1 \int_0^L dx_2 \, f(|x_{12}|) = 2L \int_0^L dx \left(1 - \frac{x}{L}\right) f(x) \quad ,$$

(21)

we can perform two integrations immediately :

$$"\sigma" = \int_0^{L_3} dz_1 \int_0^{L_3} dz_2 \int_0^{L_1} dx_{12} \left(1 - \frac{x_{12}}{L_1}\right) \int_0^{L_2} dy_{12} \left(1 - \frac{y_{12}}{L_2}\right) n_2(r_{12}, z_1, z_2) \frac{r_{12}^2 - 3z_{12}^2}{r_{12}} \frac{du(r_{12})}{dr_{12}}$$

(22)

The terms X_{12}/L_1 and Y_{12}/L_2 in the integrand can be identified as contributing only to the free energy of the liquid-and vapour-side wall interfaces. To see this, consider a fluid above its critical temperature: the pair density becomes $n_2 = n^2 g(r)$ where n is the density of the single phase filling the box. The change in free energy caused by the deformation of this single-phase system is (c.f. (22))

$$\Delta F = 2n^2 \xi \, L_1 L_2 L_3 \int_0^{L_1} dx_{12} \left(1 - \frac{x_{12}}{L_1}\right) \int_0^{L_2} dy_{12} \left(1 - \frac{y_{12}}{L_2}\right) \int_0^{L_3} dz_{12} \left(1 - \frac{z_{12}}{L_3}\right) g(r) \frac{r^2 - 3z_{12}^2}{r} \frac{du}{dr} \tag{23}$$

where we have dropped the suffix 12 on r . The term in X_{12}/L_1 is thus proportional to $\xi L_2 L_3$ (there are also ξL_2, ξL_3 and ξ terms which we can neglect) and therefore corresponds to a change in the yz-wall area. Thus we can discard the X_{12}/L_1 and Y_{12}/L_2 terms, and (22) reduces to

$$'\sigma' = \int_0^L dz_1 \int_0^L dz_2 \int_0^\infty dx_{12} \int_0^\infty dy_{12} \, n_2(r, z_1, z_2) \frac{r^2 - 3z_{12}^2}{r} \frac{du}{dr} \quad , \tag{24}$$

where we have dropped the suffix 3 on L_3 , and extended the range of the X_{12} and Y_{12} integrations to infinity. The latter is valid on the assumption that the force factor du/dr limits contributions to the integral to microscopic values of r(and therefore of X_{12} and Y_{12}).

We can now perform one more integration by changing from X_{12}, Y_{12} to the cylindrical coordinates ρ and ϕ :

$$X_{12} = \rho \cos\phi \,, \quad Y_{12} = \rho \sin\phi \,, \quad dx_{12} dy_{12} = \rho \, d\rho \, d\phi$$

Then (24) becomes

$$'\sigma' = \frac{\pi}{2} \int_0^L dz_1 \int_0^L dz_2 \int_0^\infty d\rho \, \rho \, n_2(r, z_1, z_2) \frac{r^2 - 3z_{12}^2}{r} \frac{du}{dr}$$

$$= \frac{\pi}{2} \int_0^L dz_1 \int_0^L dz_2 \int_{|z_{12}|}^\infty dr \, n_2(r, z_1, z_2)(r^2 - 3z_{12}^2) \frac{du}{dr} \tag{25}$$

and we have the surface tension reduced to a three-fold
integral.   The reason for keeping the quotation marks around
σ is that (25) still contains "floor" and "ceiling" terms.
These will be identified and discarded in Section 3.

We now consider the surface energy, i.e. the surface
part of the total thermodynamic energy   E   defined in eq.(9).
For pairwise central interactions, (9) reduces to the well-
known expression

$$E = \frac{3}{2}NT + \frac{1}{2}\int d\underline{r}_1 \int d\underline{r}_2\, n_2(\underline{r}_1, \underline{r}_2)\, u(r_{12}) \tag{26}$$

For a classical system, the kinetic energy density is constant,
i.e. there is no kinetic contribution to the surface energy
(this is not so for a quantum system).   Thus the surface
energy per unit area $\in$ is $(L_1 L_2)^{-1}$ times the surface part
of the potential energy

$$\langle u \rangle = \frac{1}{2}\int d\underline{r}_1 \int d\underline{r}_2\, n_2(\underline{r}_1, \underline{r}_2)\, u(r_{12}) \tag{27}$$

Instead of deforming the box to extract the surface part (i.e.
evaluating (10) explicitly), we prefer instead to extract the
term proportional to $L_1 L_2$ directly from (27).   When the
liquid-vapour interface lies in the xy plane $n_2 = n_2(r_{12}, z_1, z_2)$
and

$$\langle u \rangle = 2L_1 L_2 \int_0^{L_1} dx_{12}\left(1-\frac{x_{12}}{L_1}\right)\int_0^{L_2} dy_{12}\left(1-\frac{y_{12}}{L_2}\right)\int_0^{L_3} dz_1 \int_0^{L_3} dz_2\, n_2(r, z_1, z_2)\, u(r) \tag{28}$$

As noted for the free energy, the $\frac{x_{12}}{L_1}$ and $\frac{y_{12}}{L_2}$ terms do
not lead to liquid-vapour interface energies, so

$$'\in' = 2\int_0^L dz_1 \int_0^L dz_2 \int_0^\infty dx_{12}\int_0^\infty dy_{12}\, n_2(r, z_1, z_2)\, u(r)$$

$$= \pi \int_0^L dz_1 \int_0^L dz_2 \int_{|z_{12}|}^{\infty} dr \, n_2(r, z_1, z_2) \, r u(r) \tag{29}$$

The quotation marks are again necessary because (29) includes floor and ceiling surface terms (as for 'σ') and also, unlike the expression for 'σ' , volume terms for the two phases.

on the assumption $g(\underline{r}_1, \underline{r}_2) = g(r_{12})$, but do not deal with g(r) directly. Rather, they generate configurations (with probability exp(-^{U}/T)) consistent with a given density profile. Of course, approximations like (32) can be avoided altogether if the numerical work is based on the exact expressions (25) and (29).

We now proceed to evaluate 'σ' and 'ϵ' using

$$n_2(r_{12}, z_1, z_2) = n(z_1) n(z_2) g(r_{12}, \bar{n}) \tag{34}$$

From (25) and (34) we have

$$'\sigma' = \frac{\pi}{2} \int_0^L dz_1 n(z_1) \int_0^L dz_2 n(z_2) \int_{|z_{12}|}^\infty dr \, F(r) (r^2 - 3z_{12}^2) \tag{35}$$

where

$$F(r) = g(r, \bar{n}) \frac{du}{dr} \tag{36}$$

is (apart from its sign) the average force between two particles in the fluid separated by distance r. We now write

$$n(z) = n_v + (n_\ell - n_v) f(z) \tag{37}$$

where $f(z)$ tends to unity deep in the liquid and to zero far in the gas.

Then
$$'\sigma' = \frac{\pi}{2} n_v^2 \int_0^L dz_1 \int_0^L dz_2 \int_{|z_{12}|}^\infty dr \, F(r)(r^2 - 3z_{12}^2)$$

$$+ \frac{\pi}{2} n_v (n_\ell - n_v) \int_0^L dz_1 \int_0^L dz_2 [f(z_1) + f(z_2)] \int_{|z_{12}|}^\infty dr \, F(r)(r^2 - 3z_{12}^2) \tag{38}$$

$$+ \frac{\pi}{2} (n_\ell - n_v)^2 \int_0^L dz_1 f(z_1) \int_0^L dz_2 f(z_2) \int_{|z_{12}|}^\infty dr \, F(r)(r^2 - 3z_{12}^2)$$

Using (21) we see that the first term equals

$$\pi n_v^2 L \int_0^\infty dz \left(1 - \frac{z}{L}\right) \int_z^\infty dr \, F(r)(r^2 - 3z^2) \tag{39}$$

$$= \frac{\pi}{4} n_v^2 \int_0^\infty dr \, r^4 F(r)$$

We note for comparison the Fowler formula [15] which was derived using the approximations $n_V = 0$, $g(\underline{r}_1, \underline{r}_2) = g_\ell(r)$, namely

$$\sigma_F = \frac{\pi}{8} n_\ell^2 \int_0^\infty dr \, F_\ell(r) \, r^4 \tag{40}$$

Thus (39) represents the surface free energy of the interfaces between a vapour phase and the top and bottom of the box. We therefore discard the first term in (38), since it does not arise from the presence of a liquid-vapour interface.

Turing to the second term in (38), we note that it is proportional to

$$\int_0^L dz_1 \, f(z_1) \int_0^L dz_2 \int_{|z_{12}|}^\infty dr \, F(r)(r^2 - 3z_{12}^2)$$

$$= \int_0^L dz_1 \, f(z_1) \int_{z_1}^\infty dr \, F(r) \, z_1 (r^2 - z_1^2) \tag{41}$$

(We have used the fact that F(r) goes to zero in a microscopic distance, plus the presence of $f(z_1)$, to extend the range of z_2 to infinity in carrying out the integration). Because of F, the contributions to (41) come only from small z_1 , i.e. from near the bottom of the tank. Thus we discard the second term in (38), and are left with

$$"\sigma" = \frac{\pi}{2} (n_\ell - n_v)^2 \int_0^L dz_1 \, f(z_1) \int_0^L dz_2 \, f(z_2) \int_{|z_{12}|}^\infty dr \, F(r)(r^2 - 3z_{12}^2) \tag{42}$$

We have kept the quotation marks around σ because there is still one spurious term to be discarded. This can be seen as follows: we write

$$f(z) = f_D(z) + \delta f(z) \tag{43}$$

Fig. 2.

where $f_D(z)$ is a step function,

$$f_D(z) = \begin{cases} 1 & z \leq D \\ 0 & z > D \end{cases} \tag{44}$$

and the range of $\delta f(z)$ is the surface thickness. The contribution to (42) from the term $f_D(z_1) f_D(z_2)$ is of the same form as the first term in (38), with L replaced by D, and thus equals

$$\frac{\pi}{4} (n_\ell - n_v)^2 \int_0^\infty dr \, F(r) \, r^4$$

As with (39) this has combined two equal terms, one from the liquid-vapour interface ($z_1 \simeq D \simeq z_2$) and the other from the bottom of the tank (z_1 and z_2 both near zero). All other contributions to (42) are true interface terms since they involve δf's. Thus we have, finally without quotation marks

$$\sigma = \frac{\pi}{8}(n_\ell - n_v)^2 \int_0^\infty dr \, F(r) \, r^4$$
$$+ \pi(n_\ell - n_v)^2 \int_0^L dz_1 \, \delta f(z_1) \int_0^D dz_2 \int_{|z_{12}|}^\infty dr \, F(r) (r^2 - 3z_{12}^2)$$
$$+ \frac{\pi}{2}(n_\ell - n_v)^2 \int_0^L dz_1 \, \delta f(z_1) \int_0^L dz_2 \, \delta f(z_2) \int_{|z_{12}|}^\infty dr \, F(r) (r^2 - 3z_{12}^2) \tag{45}$$

When n_v is negligible the first term reduces to the Fowler expression (40).

For the second term we find

$$\int_o^D dz_2 \int_{|z_{12}|}^\infty dr\, F(r)(r^2 - 3z_{12}^2)$$

$$= -\int_{|z_1 - D|}^\infty dr\, F(r)(z_1 - D)[r^2 - (z_1 - D)^2]$$

(46)

It is now convenient to change to a height coordinate which has the surface at the origin: we put

$$\zeta = z - D \quad \text{and} \quad \delta f(z) = \widetilde{\delta f}(\zeta)$$

(47)

Using (46), (47) and the fact that D and L − D are macroscopic lengths, we obtain for the second term in (45)

$$-\pi(n_\ell - n_v)^2 \int_{-\infty}^\infty d\zeta\, \widetilde{\delta f}(\zeta)\, \zeta \int_{|\zeta|}^\infty dr\, F(r)(r^2 - \zeta^2)$$

(48)

$$= -\pi(n_\ell - n_v)^2 \int_o^\infty dr\, F(r) \int_{-r}^r d\zeta\, \widetilde{\delta f}(\zeta)\, \zeta(r^2 - \zeta^2)$$

In the last term of (45) we have

$$\int_{-\infty}^\infty d\zeta_1\, \widetilde{\delta f}(\zeta_1) \int_{-\infty}^\infty d\zeta_2\, \widetilde{\delta f}(\zeta_2) \int_{|\zeta_{12}|}^\infty dr\, F(r)(r^2 - 3\zeta_{12}^2)$$

$$= \int_{-\infty}^\infty d\zeta_1\, \widetilde{\delta f}(\zeta_1) \int_o^\infty dr\, F(r) \int_{\zeta_1 - r}^{\zeta_1 + r} d\zeta_2\, \widetilde{\delta f}(\zeta_2)(r^2 - 3\zeta_{12}^2)$$

(49)

$$= \int_o^\infty dr\, F(r) \int_{-\infty}^\infty d\zeta_1\, \widetilde{\delta f}(\zeta_1) \int_{\zeta_1 - r}^{\zeta_1 + r} d\zeta_2\, \widetilde{\delta f}(\zeta_2)(r^2 - 3\zeta_{12}^2)$$

Combining these results gives

$$\sigma = \frac{\pi}{2}(n_\ell - n_v)^2 \int_o^\infty dr\, F(r) \left[\frac{1}{4} r^4 + p_1(r) + p_2(r)\right]$$

(50)

where

$$p_1(r) = -2 \int_{-r}^r d\zeta\, \widetilde{\delta f}(\zeta)\, \zeta(r^2 - \zeta^2)$$

(51)

$$p_2(r) = \int_{-\infty}^\infty d\zeta_1\, \widetilde{\delta f}(\zeta_1) \int_{\zeta_1 - r}^{\zeta_1 + r} d\zeta_2\, \widetilde{\delta f}(\zeta_2)(r^2 - 3\zeta_{12}^2)$$

Note that we have proved, in the approximation $n_2(r, z_1, z_2) = n(z_1)\, n(z_2)\, g(r_{12}, \bar{n})$, that the surface tension is proportional to $(n_\ell - n_v)^2$. This result shows a desired symmetry between the liquid and vapour phases. MacLellan [3] has previously suggested a similar extension to the Fowler formula, namely

$$\sigma = \frac{\pi}{8} \int_0^\infty dr\, \frac{du}{dr}\, r^4 \left[n_\ell\, g_\ell(r)^{1/2} - n_v\, g_v(r)^{1/2} \right]^2 \ ,$$

which gives the first term of (50) on the assumption that

$$g(r, n_\ell) = g(r, n_v) = g(r, \bar{n}).$$

We now turn to the energy per unit area of the interface, or briefly, the surface energy. From the general expression for $'\epsilon'$, equation (29), and using the approximation $n_2 = n(z_1)\, n(z_2)\, g(r_{12}, \bar{n})$, we have

$$'\epsilon' = \pi \int_0^L dz_1\, n(z_1) \int_0^L dz_2\, n(z_2) \int_{|z_{12}|}^\infty dr\, h(r) \tag{52}$$

where

$$h(r) = g(r, \bar{n})\, r\, u(r) \tag{53}$$

Like $F(r)$, $h(r)$ goes to zero rapidly over a microscopic range. We write, as before

$$n(z) = n_v + (n_\ell - n_v) \left[f_0(z) + \delta f(z) \right]$$

Then

$$'\epsilon' = '\epsilon_0' + \epsilon_1 + \epsilon_2 \tag{54}$$

where $'\epsilon_0'$, ϵ_1 and ϵ_2 come respectively from the $n(z_1)\, n(z_2)$ terms

$$\left[n_v + (n_\ell - n_v) f_0(z_1) \right] \left[n_v + (n_\ell - n_v) f_0(z_2) \right] ,$$

$$(n_\ell - n_v)\left\{ \left[n_v + (n_\ell - n_v) f_0(z_1) \right] \delta f(z_2) + \delta f(z_1) \left[n_v + (n_\ell - n_v) f_0(z_2) \right] \right\} , \tag{55}$$

and $\quad (n_\ell - n_v)^2 \, \delta f(z_1) \, \delta f(z_2)$

Thus

$$'\epsilon_o' = \pi \left\{ n_\ell^2 \int_0^D dz_1 \int_0^D dz_2 + 2 n_\ell n_v \int_0^D dz_1 \int_D^L dz_2 + n_v^2 \int_D^L dz_1 \int_D^L dz_2 \right\} \int_{|z_{12}|}^\infty dr \, h(r) \quad (56)$$

Consider the first term of $'\epsilon_o'$. It is equal to (using (21) again)

$$\pi n_\ell^2 \, 2D \int_0^D dz \left(1 - \frac{z}{D} \right) \int_z^\infty dr \, h(r) \quad (57)$$

$$= D. \, 2\pi n_\ell^2 \int_0^\infty dr \, h(r) \, r \; - \; \pi n_\ell^2 \int_0^\infty dr \, h(r) \, r^2$$

The first part of (57) is not a surface energy of any kind, but rather the bulk energy of the liquid phase divided by $L_1 L_2$. The second part is twice the amount obtained in Fowler's approximation (Kirkwood and Buff [1], eqn. (31))

$$\epsilon_F = -\frac{\pi}{2} n_\ell^2 \int_0^\infty dr \, h(r) \, r^2$$

because it contains two contributions - one from the change in density at the bottom of the box, and an equal contribution coming from the liquid-vapour interface. (This is in exact correspondence to the discussion of the similar surface tension contributions). Thus the first and last terms of (56) give a contribution to ϵ_o adding to

$$-\frac{\pi}{2} (n_\ell^2 + n_v^2) \int_0^\infty dr \, h(r) \, r^2 \quad (58)$$

The middle term of (56) is entirely a true liquid-vapour interface term, and equals

$$\pi n_\ell n_v \int_0^\infty dr \, h(r) \, r^2 \quad (59)$$

Thus the liquid-vapour interface part of $'\epsilon_o'$ is

$$\epsilon_o = -\frac{\pi}{2} (n_\ell - n_v)^2 \int_0^\infty dr \, h(r) \, r^2 \quad (60)$$

Turning to ϵ_1 and ϵ_2 , we note that the presence of the δf's
in the second and third part of (55) guarantees that only
liquid-vapour interface terms are present.   We have

$$\epsilon_1 = 2\pi(n_\ell - n_v)\left\{ n_\ell \int_0^D dz_1 + n_v \int_D^L dz_1 \right\} \int_0^L dz_2\, \delta f(z_2) \int_{|z_{12}|}^\infty \frac{dr}{}\, h(r) \quad (61)$$

$$\equiv 2\pi(n_\ell - n_v)\left\{ n_\ell J_\ell + n_v J_v \right\}$$

The integrals J_ℓ and J_v can be reduced as follows.   We first
change the order of the z_1 and z_2 integrations, then introduce
the ζ-coordinates defined in (47), then interchange the order of
the ζ_1 and r integrations, carry out the ζ_1 integration,
and finally interchange the order of the r and ζ_2 integrations.
We find

$$J_\ell = \int_0^\infty dr\, h(r) \left\{ -\int_{-r}^r d\zeta\, \widetilde{\delta f}(\zeta)\, \zeta + r\left[\int_{-r}^r d\zeta\, \widetilde{\delta f}(\zeta) + 2\int_{-\infty}^{-r} d\zeta\, \widetilde{\delta f}(\zeta) \right] \right\} \quad (62)$$

and

$$J_v = \int_0^\infty dr\, h(r) \left\{ +\int_{-r}^r d\zeta\, \widetilde{\delta f}(\zeta)\, \zeta + r\left[\int_{-r}^r d\zeta\, \widetilde{\delta f}(\zeta) + 2\int_r^\infty d\zeta\, \widetilde{\delta f}(\zeta) \right] \right\} \quad (63)$$

So far we have not specified the location of the surface.
Ono and Kondo [16] pp139-142 (see also the discussion following
eq.(1) in Fisk and Widom [17])point out that when the surface($z=D$)
is chosen to be the Gibbs' dividing surface of vanishing super-
ficial density (sometimes called the equimolecular dividing surface),
then the surface tension becomes the superficial density of the
Helmholtz   free energy and there is a thermodynamic relation between
σ and ϵ analogous to that between F and E.   The location of the
equimolecular dividing surface is determined by

$$\int_0^D dz\, [n(z) - n_\ell] + \int_D^L dz\, [n(z) - n_v] = 0 \quad (64a)$$

or equivalently

$$\int_{-\infty}^{\infty} d\zeta \, \widetilde{\delta f}(\zeta) = 0 \qquad (64b)$$

This choice for D leads to considerable simplification in ε. For, using (64b) we find

$$J_\ell + J_v = 0 \qquad (65)$$

and ε_1 simplifies to

$$\varepsilon_1 = 2\pi(n_\ell - n_v)^2 \int_0^\infty dr \, h(r) \left\{ -\int_{-r}^{r} d\zeta \, \widetilde{\delta f}(\zeta) \zeta + r \left[\int_{-\infty}^{-r} d\zeta \, \widetilde{\delta f}(\zeta) - \int_{r}^{\infty} d\zeta \, \widetilde{\delta f}(\zeta) \right] \right\} \quad (66)$$

We come finally to ε_2, given by

$$\varepsilon_2 = \pi(n_\ell - n_v)^2 \int_{-\infty}^{\infty} d\zeta_1 \, \widetilde{\delta f}(\zeta_1) \int_{-\infty}^{\infty} d\zeta_2 \, \widetilde{\delta f}(\zeta_2) \int_{|\zeta_{12}|}^{\infty} dr \, h(r) \qquad (67)$$

By the same steps as used in the reduction of the last term of (45), this can be written as

$$\varepsilon_2 = \pi(n_\ell - n_v)^2 \int_0^\infty dr \, h(r) \int_{-\infty}^{\infty} d\zeta_1 \, \widetilde{\delta f}(\zeta_1) \int_{\zeta_1 - r}^{\zeta_1 + r} d\zeta_2 \, \widetilde{\delta f}(\zeta_2) \qquad (68)$$

Thus, provided (64) holds,

$$\varepsilon = \pi(n_\ell - n_v)^2 \int_0^\infty dr \, h(r) \left[-\tfrac{1}{2} r^2 + q_1(r) + q_2(r) \right] \qquad (69)$$

where

$$q_1(r) = -2 \int_{-r}^{r} d\zeta \, \widetilde{\delta f}(\zeta) \zeta - 2r \left[\int_{r}^{\infty} d\zeta \, \widetilde{\delta f}(\zeta) - \int_{-\infty}^{-r} d\zeta \, \widetilde{\delta f}(\zeta) \right] \qquad (70)$$

$$q_2(r) = \int_{-\infty}^{\infty} d\zeta_1 \, \widetilde{\delta f}(\zeta_1) \int_{\zeta_1 - r}^{\zeta_1 + r} d\zeta_2 \, \widetilde{\delta f}(\zeta_2)$$

We note that the choice (64) for the dividing surface has made ε proportional to $(n_\ell - n_v)^2$, in parallel to the dependence of σ. This will prove to be important in the discussion of critical exponents in Section 7.

4. EXPONENTIAL DENSITY VARIATION

In the previous section we derived, in the approximation $n_2 = n(z_1)n(z_2)g(r,\bar{n})$, expressions for σ and ε that are valid for any variation of the density. We now give analytic results for the surface tension and the surface energy for a simple exponential variation of the density, namely

$$
n(\zeta) = \begin{cases} n_\ell - \frac{1}{2}(n_\ell - n_v) e^{\zeta/\lambda} & , \quad \zeta < 0 \\[2ex] n_v + \frac{1}{2}(n_\ell - n_v) e^{-\zeta/\lambda} & , \quad \zeta > 0 \end{cases}
\tag{71}
$$

where λ is a measure of the surface width. The density and its derivative are continuous at $\zeta = 0$, where

$$
n(0) = \frac{1}{2}(n_\ell + n_v) \quad , \quad n'(0) = -\frac{1}{2}(n_\ell - n_v)/\lambda
\tag{72}
$$

The function $\widetilde{\delta f}$ corresponding to this choice of density is

$$
\widetilde{\delta f}(\zeta) = \frac{1}{2} \operatorname{sgn} \zeta \exp(-|\zeta|/\lambda) \quad ,
\tag{73}
$$

and, being odd, satisfies (64) automatically.

That this choice is correct at large $|\zeta|$ (at least near the critical point) has been shown by Fisk and Widom [17] from a generalized van der Waals theory. The exponential density variation has also been used by Berry, Durrans and Evans [12], and Freeman and McDonald [14].

The evaluation of p_1 and q_1 from (51) and (70) is straightforward. We find

$$p_1(r) = \lambda^4 \left[12 - 2\left(\frac{r}{\lambda}\right)^2 - e^{-r/\lambda}\left(12 + 12\frac{r}{\lambda} + 4\frac{r^2}{\lambda^2}\right) \right]$$

$$(74)$$

$$q_1(r) = -2\lambda^2\left(1 - e^{-r/\lambda}\right)$$

Both p_1 and q_1 are zero at r=o, and are otherwise negative and monotonically decreasing. The evaluation of p_2 and q_2 is made easier by noting that

$$\frac{dq_2}{dr} = \int_{-\infty}^{\infty} d\zeta \, \widehat{\delta f}(\zeta) \left[\widehat{\delta f}(\zeta + r) + \widehat{\delta f}(\zeta - r) \right]$$

$$= 2 \int_{-\infty}^{\infty} d\zeta \, \widehat{\delta f}(\zeta) \, \widehat{\delta f}(\zeta + r) \qquad (\text{for } \widehat{\delta f} \text{ odd})$$

$$= \frac{1}{2} \lambda \, e^{-r/\lambda} \left(1 - r/\lambda\right)$$

Thus
$$q_2(r) = \frac{1}{2}\lambda r \, e^{-r/\lambda} \, .$$

$$(75)$$

Similarly
$$p_2(r) = r^2 q_2(r) - 3 I(r)$$

where
$$I(r) = \int_{-\infty}^{\infty} d\zeta \, \widehat{\delta f}(\zeta) \int_{-r}^{r} dz \, z^2 \, \widehat{\delta f}(\zeta + z) \, ,$$

so that
$$\frac{dI}{dr} = r^2 \frac{dq_2}{dr}$$

thus
$$p_2(r) = \lambda^4 \left[6 - e^{-r/\lambda}\left(6 + 6\frac{r}{\lambda} + 3\frac{r^2}{\lambda^2} + \frac{r^3}{\lambda^3}\right) \right]$$

$$(76)$$

Both p_2 and q_2 are zero at r=o and are otherwise positive. While p_2 is monotonically increasing, q_2 has a maximum at r=λ. The sums $p_1 + p_2$ and $q_1 + q_2$ are both monotonically decreasing.

Using these results we find, for this one-parameter density,

$$\sigma = \frac{\pi}{2}(n_\ell - n_v)^2 \int_0^\infty dr \, F(r) \left\{ \frac{1}{4} r^4 + \lambda^4 \left[18 - 2\frac{r^2}{\lambda^2} - e^{-r/\lambda}\left(18 + 18\frac{r}{\lambda} + 7\frac{r^2}{\lambda^2} + \frac{r^3}{\lambda^3}\right) \right] \right\}$$

$$(77)$$

and

$$\epsilon = -\frac{\pi}{2}(n_\ell - n_v)^2 \int_0^\infty dr \, h(r) \left\{ r^2 + \lambda^2 \left[4 - e^{-r/\lambda}\left(4 + \frac{r}{\lambda}\right) \right] \right\}$$

$$(78)$$

The result for ε is new; the expression for σ differs from that obtained by Berry, Durrans and Evans [12] only in the replacement of n_ℓ^2 by $(n_\ell - n_v)^2$ (This difference, of course, proves to be very important near the critical point).

We consider the two limiting forms of σ and ε. The limit $\lambda \to 0$ (step function density) has $\delta f \equiv 0$, so from either (50) and (69) or from (77) and (78) we have

$$\sigma_0 = \frac{\pi}{8}(n_\ell - n_v)^2 \int_0^\infty dr\, F(r)\, r^4 \quad , \quad \varepsilon_0 = -\frac{\pi}{2}(n_\ell - n_v)^2 \int_0^\infty dr\, h(r)\, r^2 \qquad (79)$$

Neither of these expressions could be expected to be accurate, even near the triple point, since the thickness of the interface (approximately given by $|\Delta n / n'(0)| = 2\lambda$) is unlikely to be much smaller than molecular size. Surprisingly, Freeman and McDonald [14] found numerical values of σ_0 quite close to the triple point experimental values for Argon and Neon (see also Berry et al [12]). The reason for this is accidental, as can be seen from Fig. 3. There we have plotted as the full and dotted lines the integrand of the expression (77) for σ, taking

$$u(r) = 4v\left[\left(\frac{d}{r}\right)^{12} - \left(\frac{d}{r}\right)^6\right] \quad \text{and} \quad g = e^{-u/T}. \text{ The curves}$$

are for $\lambda = d$ (d is the core diameter, which is plausibly about half the interface thickness 2λ near the triple point), and for $\lambda = 0$. We see that there is a large degree of concellation between positive and negative parts of the integrand in both cases, and that (for a more accurate g) σ_0 happens to give a good value only because of this cancellation, since the integrand is quite different numerically from that with $\lambda = d$.

Fig. 3.

On the other hand, ε_o gives poor values for the surface energy [14]. The reason for this can be seen in Fig. 4, where we have plotted the integrand of the expression (78) for ε for $\lambda = d$ and $\lambda = o$. In this case there is almost no cancellation between the positive and negative parts of the integrand, so that ε_o is far from the experimental value, even at the triple point.

$$\text{Fig. 4.}$$

We can further see from Figs. 3 and 4 that $\sigma_o > \sigma$ and $\varepsilon_o < \varepsilon$, as found by Freeman and McDonald [14].

We now look at the limit where λ is large compared to the molecular size. This limit is of course attained as the critical point is approached, but expressions for large λ prove to be surprisingly useful even well away from the critical point. Since F and h rapidly become negligible as r/d becomes large, the values of r of importance in the integrand are then small compared with λ. Thus we can expand the integrands in σ and ε as a series in r/λ. The leading terms for large λ are

$$\sigma = \frac{\pi}{30} (n_\ell - n_v)^2 \frac{1}{\lambda} \int_0^\infty dr \, F(r) r^5 \tag{80}$$

and

$$\varepsilon = -\frac{3\pi}{2} (n_\ell - n_v)^2 \lambda \int_0^\infty dr \, h(r) r \tag{81}$$

Thus the surface tension and surface energy are proportional to λ^{-1} and λ for large λ. We will obtain these results in another way in the next section.

It is interesting to compare the integrands of the expressions for σ and ε in the large λ limit with those for $\lambda \to o$ and the full expressions given by (77) and (78). The large λ limit, evaluated at $\lambda = d$, is shown as the dashed curve in Figs 3 and 4. The agreement between the large λ limits and the full expressions is surprisingly good. In section 8 we will use this to extract the surface thickness from experimental data near the triple point.

5. SLOWLY VARYING DENSITY.

It is possible to obtain general expressions for σ and ε when the density variation is slow on the scale of the molecular diameter. The results given below are generalizations of the large λ limit of the exponential density variation. Consider the surface tension first, starting from (35):

$$
\begin{aligned}
\sigma &= \frac{\pi}{2} \int_0^L dz_1\, n(z_1) \int_0^L dz_2\, n(z_2) \int_{|z_{12}|}^\infty dr\, F(r)(r^2 - 3z_{12}^2) \\
&= \frac{\pi}{2} \int_0^L dz_1\, n(z_1) \int_{-z_1}^{L-z_1} dz\, n(z_1 + z) \int_{|z|}^\infty dr\, F(r)(r^2 - 3z^2) \qquad (82) \\
&= \frac{\pi}{2} \int_0^L dz_1\, n(z_1) \int_{-z_1}^{L-z_1} dz\, \left\{ n(z_1) + z\, n'(z_1) + \tfrac{1}{2} z^2 n''(z_1) + \cdots \right\} \int_{|z|}^\infty dr\, F(r)(r^2 - 3z^2)
\end{aligned}
$$

Taking the z integrals in turn, we see that because

$$
\int_{-\infty}^\infty dz \int_{|z|}^\infty dr\, F(r)(r^2 - 3z^2) = \int_0^\infty dr\, F(r) \int_{-r}^r dz\,(r^2 - 3z^2) = 0 ,
$$

the zeroth term in the Taylor expansion contributes only if z_1 is near 0 or L, and thus is not a liquid-vapour interface term. The second term similarly contains no interface contribution, since

$$
\int_{-\infty}^\infty dz\, z \int_{|z|}^\infty dr\, F(r)(r^2 - 3z^2) = 0
$$

Thus we are left with

$$
\sigma = \frac{\pi}{4} \int_0^L dz_1\, n(z_1) n''(z_1) \int_{-\infty}^\infty dz\, z^2 \int_{|z|}^\infty dr\, F(r)(r^2 - 3z^2)
$$

We discard contributions from the 0 and L boundaries in the z_1 integral, and perform the z integral to obtain

$$
\sigma = \frac{2\pi}{15} \int_0^\infty dr\, F(r)\, r^5 \int_{-\infty}^\infty d\zeta\, [n'(\zeta)]^2 \qquad (83)
$$

Thus we see that the microscopic theory leads to

$$\sigma = a \int_{-\infty}^{\infty} d\zeta \, [n'(\zeta)]^2 = \frac{A(n_\ell - n_v)^2}{\lambda} \qquad (84)$$

in the limit of slow density variation. This has the same
form as that obtained from the van der Waals theory by Cahn and
Hilliard [18] and Fisk and Widom [17], and also from a fluct-
uation theory by Triezenberg and Zwanzig [7]. According to
Lovett, De Haven, Vieceli and Buff [8], the same form was
obtained long ago by Ornstein and Zernike [19] with the
coefficient a given by

$$a = \frac{2\pi}{3} T \int_{0}^{\infty} dr \, c(r) \, r^4,$$

where $c(r)$ is the direct correlation function. This result is
readily seen to be identical to (83) in the low-density limit

$$g = e^{-u/T} \quad , \quad c = e^{-u/T} - 1 \qquad (85)$$

We now turn to the surface energy, which to our knowledge
has only been considered for a step-function density variation
[1].   Starting with (52) and proceeding as for σ,
we find

$$'\epsilon' = \pi \int_0^L dz_1 \, n(z_1) \int_0^L dz_2 \, n(z_2) \int_{|z_{12}|}^{\infty} dr \, h(r) \qquad (86)$$

$$= \pi \int_0^L dz_1 \, n(z_1) \int_{-z_1}^{L-z_1} dz \left\{ n(z_1) + z n'(z_1) + \frac{1}{2} z^2 n''(z_1) \right\} \int_{|z|}^{\infty} dr \, h(r)$$
$$+ \dots$$

Liquid-vapour interface contributions come only from $z_1 \simeq D \simeq z_2$,
and since D and L-D are macroscopic distances, the integral over
z may be taken from $-\infty$ to $+\infty$ when extracting the interface
contributions. The first derivative gives a zero contribution,

as before. The second derivative contributes to ε by the amount

$$-\frac{\pi}{3} \int_0^\infty dr\, h(r) r^3 \int_{-\infty}^\infty d\zeta\, [n'(\zeta)]^2 \tag{87}$$

which like σ is proportional to $(n_\ell - n_v)^2 / \lambda$. The dominant contribution for large λ is however not (87) but the liquid-vapour interface part of the first term in (86), i.e.

$$2\pi \int_0^L dz_1\, n^2(z_1) \int_0^\infty dr\, h(r) r \tag{88}$$

To extract the interface part of $\int_0^L dz\, n^2(z)$, we write the density as

$$n(z) = n_D(z) + (n_\ell - n_v)\, \delta f(z)$$

where
$$n_D(z) = \begin{cases} n_\ell & , \quad z < D \\ n_v & , \quad z > D \end{cases}$$

The constant parts n_ℓ and n_v of $n(z)$ give bulk terms, while the mixed term $2 n_D(z)(n_\ell - n_v) \delta f(z)$ contributes (to $\int_0^L dz\, n^2(z)$) by the amount

$$2(n_\ell - n_v)\left\{ n_\ell \int_{-\infty}^0 d\zeta\, \widetilde{\delta f}(\zeta) + n_v \int_0^\infty d\zeta\, \widetilde{\delta f}(\zeta) \right\}$$

On using the convention (64) for the location of the dividing surface, this becomes

$$-2(n_\ell - n_v)^2 \int_0^\infty d\zeta\, \widetilde{\delta f}(\zeta)$$

The last term in $\int_0^L dz\, n^2(z)$ is

$$(n_\ell - n_v)^2 \int_{-\infty}^\infty d\zeta\, [\widetilde{\delta f}(\zeta)]^2 ,$$

so that the total interface contribution to (88) is

$$2\pi(n_\ell - n_v)^2 \int_0^\infty dr\, h(r) r \left\{ -2 \int_0^\infty d\zeta\, \widetilde{\delta f}(\zeta) + \int_{-\infty}^\infty d\zeta\, [\widetilde{\delta f}(\zeta)]^2 \right\} \tag{89}$$

When the surface is characterized by a single length λ, both of the ζ integrals are proportional to λ; thus the dominant part of ε for a slowly varying density is

$$\varepsilon = B(n_\ell - n_v)^2 \lambda \tag{90}$$

This form is in agreement with the large λ limit obtained for the exponential density variation, equation (81), and the coefficient

$$B = -\frac{3\pi}{2} \int_0^\infty dr \, h(r) \, r \tag{91}$$

of (81) can be verified from the more general expression (89) with the substitution of (73) for $\widetilde{\delta f}$.

6. THERMODYNAMIC CONSISTENCY

The thermodynamics of surfaces is discussed in general terms by Ono and Kondo[16]. They show that when the dividing surface is chosen according to (64) (the equimolecular dividing surface), the surface tension σ, surface energy ε and surface entropy s are related by

$$\sigma = \varepsilon - Ts \quad , \quad s = -\frac{d\sigma}{dT} \quad , \tag{92}$$

in direct analogy to the relations in the bulk, namely

$$F = E - TS \quad , \quad S = -\left(\frac{\partial F}{\partial T}\right)_V$$

The two relations (92) imply

$$\varepsilon = \sigma - T\frac{d\sigma}{dT} = -T^2 \frac{d}{dT}\left(\frac{\sigma}{T}\right) \tag{93}$$

In principle this equation together with the expressions for σ and ε puts a constraint on the density variation. For example, it could be used to obtain the thermodynamically consistent λ by comparison of (77) and (78). Freeman and McDonald [14] were aware of this possibility, but did not have an expression for ε available. Analytically this programme is too complicated except in limiting cases, which we will now examine.

Consider the $\lambda \to 0$ (step function) limit first. As explained in Section 4, this limit does not give a satisfactory surface energy for the liquid-vapour interface. However, we can apply our theory to the interface between an infinitely steep wall and a dilute gas, for which we can use the low-density approximation (85) for g. The theory gives (cf(79))

$$\sigma_o = \frac{\pi}{8} n_v^2 \int_o^\infty dr\, r^4 e^{-u/T} \frac{du}{dr}$$

$$\epsilon_o = -\frac{\pi}{2} n_v^2 \int_o^\infty dr\, r^3 e^{-u/T} u \tag{94}$$

The expression for σ_o gives, on integration by parts,

$$\frac{\sigma_o}{T} = \frac{\pi}{2} n_v^2 \int_o^\infty dr\, r^3 \left(e^{-u/T} - 1\right)$$

so that the relation $-T^2 \frac{d}{dT}\left(\frac{\sigma_o}{T}\right) = \epsilon_o$ is exactly

satisfied for a temperature change at constant volume

(and thus constant density).

The consequence of thermodynamic consistency in the

large λ limit will be discussed in the next section.

7. CRITICAL EXPONENTS

As the critical point is approached, many physical quantities diverge or go to zero as T_c-T to some exponent. If we write

$$t = \frac{T_c-T}{T_c}$$

then, in the notation of Widom [20]

$$\sigma \sim t^{\mu}, \quad \lambda \sim t^{-\gamma}, \quad n_{\ell}-n_v \sim t^{\beta}, \quad C_v \sim t^{-\alpha}, \quad \chi_T \sim t^{-\gamma} \qquad (95)$$

We will also define exponents for ϵ and the coefficients A and B in the relations (84) and (90):

$$\epsilon \sim t^{\mu'}, \quad A \sim t^{\alpha'}, \quad B \sim t^{\beta'} \qquad (96)$$

The exponent μ' can be evaluated immediately from the thermodynamic relation (93):

$$\mu' = \mu - 1 \qquad (97)$$

This relation may also be obtained by a simple extension of the physical argument of Widom ([20], p.80), which shows that σ/λ is the singular part of the free energy density, and thus $\mu + \nu$ is linked to the specific heat exponent α by ([20],eq 3)

$$\mu + \nu = 2 - \alpha \qquad (98)$$

The same reasoning shows that ϵ/λ is that part of the bulk energy density which carries the specific heat singularity, so that

$$\mu' + \nu = 1 - \alpha \qquad (99)$$

Combining (98) and (99) we regain (97).

Turning to the exponents α' and β', we see from (84) and (90) that these are given by

$$\mu = \alpha' + 2\beta + \gamma \tag{100}$$

and

$$\mu' = \beta' + 2\beta - \gamma \tag{101}$$

Now if we assume the truth of the relations (3), (5) and (7) in Widom's review [20], we have (in three-dimensions) the equalities

$$\mu + \gamma = 2 - \alpha = 3\nu = \gamma + 2\beta \tag{102}$$

Using these in (100) we find for the exponent of A

$$\alpha' = \gamma - 2\nu \tag{103}$$

which is the result obtained by Fisk and Widom [17]. For β' we have, from (97), (101) and (102)

$$\beta' = \gamma - 1 \tag{104}$$

With the approximate numerical values [20]

$$\gamma \simeq 1.25 \;,\; \nu \simeq 0.64$$

we have

$$\alpha' \simeq -0.03 \;,\; \beta' \simeq 0.25$$

In our microscopic theory, the coefficients A and B are proportional to

$$\int_0^\infty dr\, r^5 g(r, \bar{n}) \frac{du}{dr} \quad \text{and} \quad \int_0^\infty dr\, r^2 g(r, \bar{n}) u(r)$$

and it is hard to see how these could be singular when u(r) is a short-ranged interaction potential. However, Rice and Chang [21] have pointed out that a similar difficulty exists with the bulk potential energy, which (above T_c) has the form of our B, and yet is singular, varying as $t^{1-\alpha}$. This leads to the question: why is β' not equal to $1-\alpha$? A possible answer is that B is proportional to the potential energy of an unstable phase (for $T < T_c$, $\bar{n}$ is in the unphysical region of the n, T plane).

Another possibility is that our microscopic expressions for the A and B coefficients are not complete, and that something slipped through the net when we made the approximation

$$n_2(\underline{r_1}, \underline{r_2}) = n(z_1)\, n(z_2)\, g(r_{12}, \bar{n}).$$

¶ We conclude this section with some observations concerning the range of validity of the power laws (95). In Section 4 we saw that the large λ limit integrands for σ and ε gave good agreement with the exact integrands for the expontential surface, even down to $\lambda = d$. This comparison was with $g = e^{-u/T}$, which is not adequate near the triple point; however, the qualitative features of the integrands and the accuracy of the large λ expressions are not expected to be different when a more accurate pair correlation function is used. Note that this increases the range of validity of the van der Waals theories (e.g. Fisk and Widom [17]) set up to operate in the critical region , which give the same form for σ as our large λ expression. It is known experimentally that $\sigma \sim t^{\mu}$ (see for example Zollweg et al. [22], Smith et al. [23]) and $(n_\ell - n_v) \sim t^{\beta}$ (Stansfield [24]), hold over a large temperature range and a recent theory of surface tension by Lovett et al. [8] has predicted that $\lambda \sim t^{-.65}$ also holds far from the critical point. We expect the power laws to hold over the same temperature range in which the large λ expressions

$$\sigma = A\,(n_\ell - n_v)^2 / \lambda \qquad , \quad \varepsilon = B\,(n_\ell - n_v)^2 \lambda \qquad (84),(90)$$

are a good approximation, which appears to be all the way to the triple point.

In terms of

$$\tau = \frac{T_c - T}{T_c - T_t} \qquad\qquad (105)$$

this would imply that the relations

$$\sigma = \sigma_t\, \tau^{\mu}, \quad \varepsilon = \varepsilon_t\, \tau^{\mu-1} \quad \text{and} \quad \lambda = \lambda_t\, \tau^{-\nu} \qquad\qquad (106)$$

were valid all the way from $\tau = 0$ to $\tau = 1$. We will show in Section 8 that the results of the large λ limit do indeed give satisfactory values for λ near the triple point.

8. THE SURFACE THICKNESS

¶ We have shown that the large λ limits are a good approximation when λ is of order of the core diameter, and that the coefficient B in the large λ expression for ϵ is proportional to the bulk potential energy of a fluid at density $\bar{n}$. These facts enable us to determine the surface thickness at the triple point. We proceed as follows: σ, $n_\ell - n_v$ and the potential energy per molecule in the liquid

$$\frac{\langle u \rangle}{N} = 2\pi n_\ell \int_0^\infty dr \; r^2 \, g(r, n_\ell) \, u(r) \; .$$

are all known from experiment, and ϵ can be deduced from the temperature variation of σ. From $\langle u \rangle$ we can obtain the value of our coefficient B (given by (91)) at the triple point, on the assumption that $\bar{n} \simeq n_\ell$:

$$n_\ell B = -\frac{3}{4} \frac{\langle u \rangle}{N} \tag{107}$$

Thus, from (90),

$$\lambda_t = \frac{\epsilon_t}{B_t (\Delta n)_t^2} \simeq \frac{\epsilon_t}{B_t \, n_{\ell t}^2} \tag{108}$$

i.e. λ can be found directly from experimental data.
Table 1 gives the relevant parameters for Ar, Kr and Xe.

TABLE 1.

We find an average value of λ/d = 0.63, with a maximum deviation of less than 4%.

This result is highly satisfactory when we remember that the surface thickness is approximately 2λ, i.e. the surface thickness near the triple point comes out to be a bit larger than the core diameter d.   It is also in accord with Egelstaff and Widom [25] , who point out that the fundamental length $\chi_T \sigma$ is approximately equal to 0.07λ at the triple point. Then using a relation proposed by Mayer from the theory of the hard sphere fluid [26] and approximating the hard core diameter by d, they find that $\chi_T \sigma \simeq 0.05d$. Thus Egelstaff and Widom have $\lambda/d \simeq 0.7$ near the triple point, in excellent agreement with our results above. Finally, Lovett, DeHaven, Vieceli and Buff [8] find similar values for surface thickness near the triple point by comparing their direct correlation function theory with experimental data for Argon, although the value they extract from Toxvaerd [6] in their table I looks the best agreement with our results.

9.   THE DIRECT CORRELATION FUNCTION THEORY FOR σ.

Alternative statistical mechanical expressions for the surface tension have been given by Yvon [29], Buff and Lovett [30], Triezenberg and Zwanzig [7] and most recently by Lovett, De Haven, Vieceli and Buff [8].   In these formulations the interatomic force does not appear explicitly, and correlations appear through the direct correlation function $c(r_1, r_2)$ rather than through the pair correlation function $g(r_1, r_2)$. We shall use the techniques developed in this paper to reduce the direct correlation function expression, and to obtain results which parallel those we obtained with the pair correlation function expression.   Finally, we show that the two theories

are equivalent in the lowest order in density.

Our starting point will be equation (11) of Lovett et al. [8]:

$$\sigma_c = \frac{1}{4} T \int_{-\infty}^{\infty} dz_1 \, \frac{dn(z_1)}{dz_1} \int_{-\infty}^{\infty} dz_2 \, \frac{dn(z_2)}{dz_2} \int_{-\infty}^{\infty} dx_{12} \int_{-\infty}^{\infty} dy_{12} \, C(r_{12}, z_1, z_2)(x_{12}^2 + y_{12}^2) , \quad (109)$$

which gives the surface tension of a plane liquid-vapour interface in the neighbourhood of $z = 0$. This expression may be reduced to a three-fold integral by the use of cylindrical polars (cf eqn. (25)):

$$\sigma_c = \frac{\pi}{2} T \int_{-\infty}^{\infty} dz_1 \, \frac{dn(z_1)}{dz_1} \int_{-\infty}^{\infty} dz_2 \, \frac{dn(z_2)}{dz_2} \int_{0}^{\infty} d\rho \, \rho \, C(\sqrt{\rho^2 + z_{12}^2}, z_1, z_2) \rho^2$$

$$= \frac{\pi}{2} T \int_{-\infty}^{\infty} dz_1 \, \frac{dn(z_1)}{dz_1} \int_{-\infty}^{\infty} dz_2 \, \frac{dn(z_2)}{dz_2} \int_{|z_{12}|}^{\infty} dr \, r \, C(r, z_1, z_2)(r^2 - z_{12}^2) \qquad (110)$$

Equation (110) may be applied immediately to a step function density at $z = 0$ (e.g. at the boundary of a single phase of density n_v and an ideal wall). We find

$$\sigma_0 = \frac{\pi}{2} T \, n_v^2 \int_0^{\infty} dr \, r^3 \, C(r, 0, 0) \qquad (111)$$

In the dilute gas limit, this reduces to the result obtained in Section 6.

We now give, with abbreviated discussion, results obtained with the approximation analogous to (32), namely

$$c(r_{12}, z_1, z_2) \simeq c(r_{12}, \bar{n}) \qquad (112)$$

Interchanging the order of integration as in (49), we have from (110)

$$\sigma_c = \frac{\pi}{2} T \int_0^{\infty} dr \, r \, c(r, \bar{n}) \int_{-\infty}^{\infty} dz_1 \, \frac{dn(z_1)}{dz_1} \int_{z_1 - r}^{z_1 + r} dz_2 \, \frac{dn(z_2)}{dz_2} (r^2 - z_{12}^2) \qquad (113)$$

For the exponential density variation (71),

$$\frac{dn}{dz} = -\frac{1}{2\lambda}(n_\ell - n_v) \exp(-|z|/\lambda) \qquad (114)$$

and we find (c.f. Section 4)

$$\sigma_c = \frac{\pi}{2} T (n_\ell - n_v)^2 \int_0^{\infty} dr \, r \, c(r, \bar{n}) \left\{ r^2 - 4\lambda^2 + \lambda^2 e^{-r/\lambda} \left(4 + 4 \frac{r}{\lambda} + \frac{r^2}{\lambda^2} \right) \right\} \qquad (115)$$

The $\lambda \to 0$ limit checks with eqn.(111). If we put $g = e^{-u/T}$ in (77) and integrate by parts, we get (115) with $c = e^{-u/T} - 1$.
In the large λ limit we find

$$\sigma_c = \frac{\pi}{6} T (n_\ell - n_r)^2 \frac{1}{\lambda} \int_0^\infty dr \, r^4 \, c(r, \bar{n}) \tag{116}$$

which is in agreement with (80) to the lowest order in density.
The integrands in the full expression (115) for σ_c, and in the limiting expressions for $\lambda \to 0$ and $\lambda \to \infty$ are compared in Fig.5 for $c = e^{-u/T} - 1$ at $\lambda = d$. We see that the large λ expression for σ_c is a very good approximation even at $\lambda = d$, while σ_0 is much larger than σ_c. This is in agreement with Fig.3 and the remark following eqn. (115).

$$\text{FIG. 5}$$

Next we apply the gradient method of Section 5.
Integrating eqn.(113) by parts we obtain

$$\sigma_c = \pi T \int_0^\infty dr \, r \, c(r, \bar{n}) \int_{-\infty}^\infty dz_1 \, n'(z_1) \int_{z_1 - r}^{z_1 + r} dz_2 \, z_{21} \, n(z_2) \tag{117}$$

The last integral may be written as

$$\int_{-r}^r d\zeta \, \zeta \, n(z_1 + \zeta)$$

$$= \int_{-r}^r d\zeta \, \zeta \, \left\{ n(z_1) + \zeta n'(z_1) + \tfrac{1}{2} \zeta^2 n''(z_1) + \dots \right\}$$

$$= 0 + \tfrac{2}{3} r^3 n'(z_1) + 0 + \dots$$

so that

$$\sigma_c \simeq \frac{2\pi}{3} T \int_0^\infty dr \, r^4 \, c(r, \bar{n}) \int_{-\infty}^\infty dz \, [n'(z)]^2 \tag{118}$$

This result was stated by Lovett et al. (eqn.(12) and (13)) and credited to Ornstein and Zernike, and is the counterpart of eqn.(83). The equivalence of (116) and (118) for the

particular case of the exponential density variation is easily verified.

We turn finally to the question of the equivalence or non-equivalence of the $g(\underline{r}_1, r_2)$ and $c(\underline{r}_1, \underline{r}_2)$ theories. Lovett et al. refer to the two theories as "complementary", and do not discuss their equivalence. But both theories are claimed to be general (except that the pair correlation function theory is usually restricted to pairwise interactions). Thus they are either rigorously equivalent, or at least one is approximate. Consider the general expression (110), transcribed to the geometry of Fig.1. Integrate by parts twice, neglecting the contributions which do not come from the liquid-vapour interface. This gives

$$'\sigma_c' = \frac{\pi}{2} T \int_0^L dz_1\, n(z_1) \int_0^L dz_2\, n(z_2) \frac{\partial^2}{\partial z_1 \partial z_2} \int_0^\infty dr\; r\; c(r, z_1, z_2)(r^2 - z_{12}^2) \qquad (119)$$

Comparison with

$$'\sigma_g' = \frac{\pi}{2} \int_0^L dz_1\, n(z_1) \int_0^L dz_2\, n(z_2) \int_{|z_{12}|}^\infty dr\; g(r, z_1, z_2) \frac{du}{dr}(r^2 - 3z_{12}^2) \qquad (25a)$$

shows that the two theories are equivalent for central pairwise interactions if

$$T \frac{\partial^2}{\partial z_1 \partial z_2} \int_{|z_{12}|}^\infty dr\; r\; c(r, z_1, z_2)(r^2 - z_{12}^2) = \int_{|z_{12}|}^\infty dr\; g(r, z_1, z_2) \frac{du}{dr}(r^2 - 3z_{12}^2) \qquad (120)$$

A general proof of equivalence is thus seen to require knowledge of relationships between c, g and u for inhomogeneous systems. It is possible to show, however, that σ_g and σ_c are identical to the lowest order in density. Using the identity

$$\frac{\partial^2}{\partial z_1 \partial z_2} f(|z_{12}|) = -2\delta(z_{12}) f'(|z_{12}|) - f''(|z_{12}|) \qquad (121)$$

we find that the left side of (120) reduces to

$$2T \left\{ \int_{|z_{12}|}^\infty dr\; r\; c(r, \bar{n}) - z_{12}^2\, c(|z_{12}|, \bar{n}) \right\}$$

when $c(\underline{r}_1, \underline{r}_2)$ is approximated as in (112).

The right side of (120) is easily shown to be equal to this expression when we take $g = e^{-u/T}$ and $c = e^{-u/T} - 1$.

TABLE 1.

	ARGON		KRYPTON		XENON	
ϵ (erg/cm^2)	35.0	[14, 27]	40.1	[27]	50.1	[14]
ρ (g/cm^3)	1.410	[28]	2.451	[28]	3.076	[28]
M (amu)	39.95		83.8		131.3	
n_ℓ (Å^{-3})	2.126×10^{-2}		1.762×10^{-2}		1.411×10^{-2}	
$\langle u \rangle / N$ (erg/atom)	-0.984×10^{-13}	[27]	-1.365×10^{-13}	[27]	-1.911×10^{-13}	[27]
$n_\ell B$ (erg/atom)	0.738×10^{-13}		1.023×10^{-13}		1.433×10^{-13}	
λ (Å)	2.231		2.225		2.478	
d (Å)	3.405	[14]	3.634	[14]	3.954	[14]
λ / d	0.655		0.612		0.627	

CAPTION: Evaluation of the surface thickness for Argon, Krypton and Xenon at their triple points.

FIGURE CAPTIONS

FIG.1.　The liquid-vapour system

FIG.2.　The function $f(z)$, where $n(z) = n_v + (n_\ell - n_v) f(z)$.

FIG.3.　Integrands of the exponential density expressions
for σ when $\lambda = d$ with $u(r) = 4v \left[\left(\frac{d}{r} \right)^{12} - \left(\frac{d}{r} \right)^{6} \right]$,
$g = e^{-u/T}$. The value of $\frac{4v}{T}$ was taken to be
5.69, which is the average of the rare gas triple point
values [28]. Full curve: general expression (77).
Dashed curve: large λ expression (80). Dotted
curve: $\lambda = 0$ limit (79). The area under a curve
times $\frac{\pi}{2} (n_\ell - n_v)^2 d^4 v$ gives the surface tension.

FIG.4.　Integrands of the exponential density expressions
for ϵ, when $\lambda = d$ with u, g and $\frac{v}{T}$ as in Fig.3.
Full curve: general expression (78). Dashed
curve: large λ expression (81). Dotted curve:
$\lambda = 0$ limit (79). The area under a curve times
$\frac{\pi}{2} (n_\ell - n_v)^2 d^4 v$ gives the surface energy.

FIG.5.　Integrands of the exponential density expressions
for σ_c, when $\lambda = d$; with $c = e^{-u/T} - 1$; u and T as
in Figs. 3 and 4. Full curve: general
expression (115). Dashed curve: large λ expression
(116). Dotted curve: $\lambda = 0$ limit. The area under
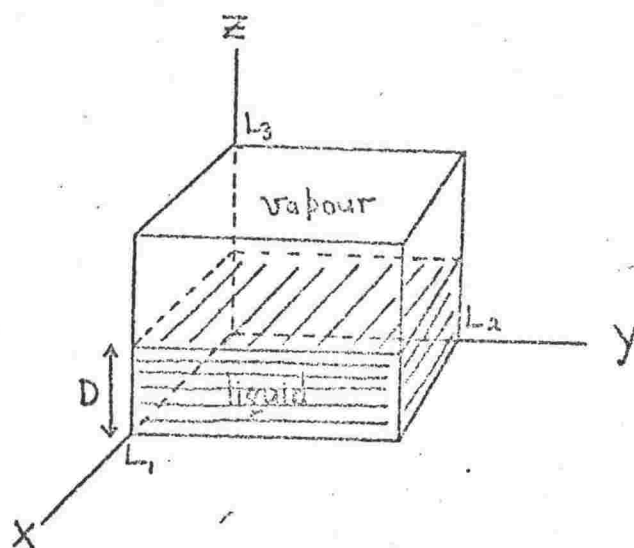a curve times $\frac{\pi}{2} (n_\ell - n_v)^2 d^4 T$ gives the surface
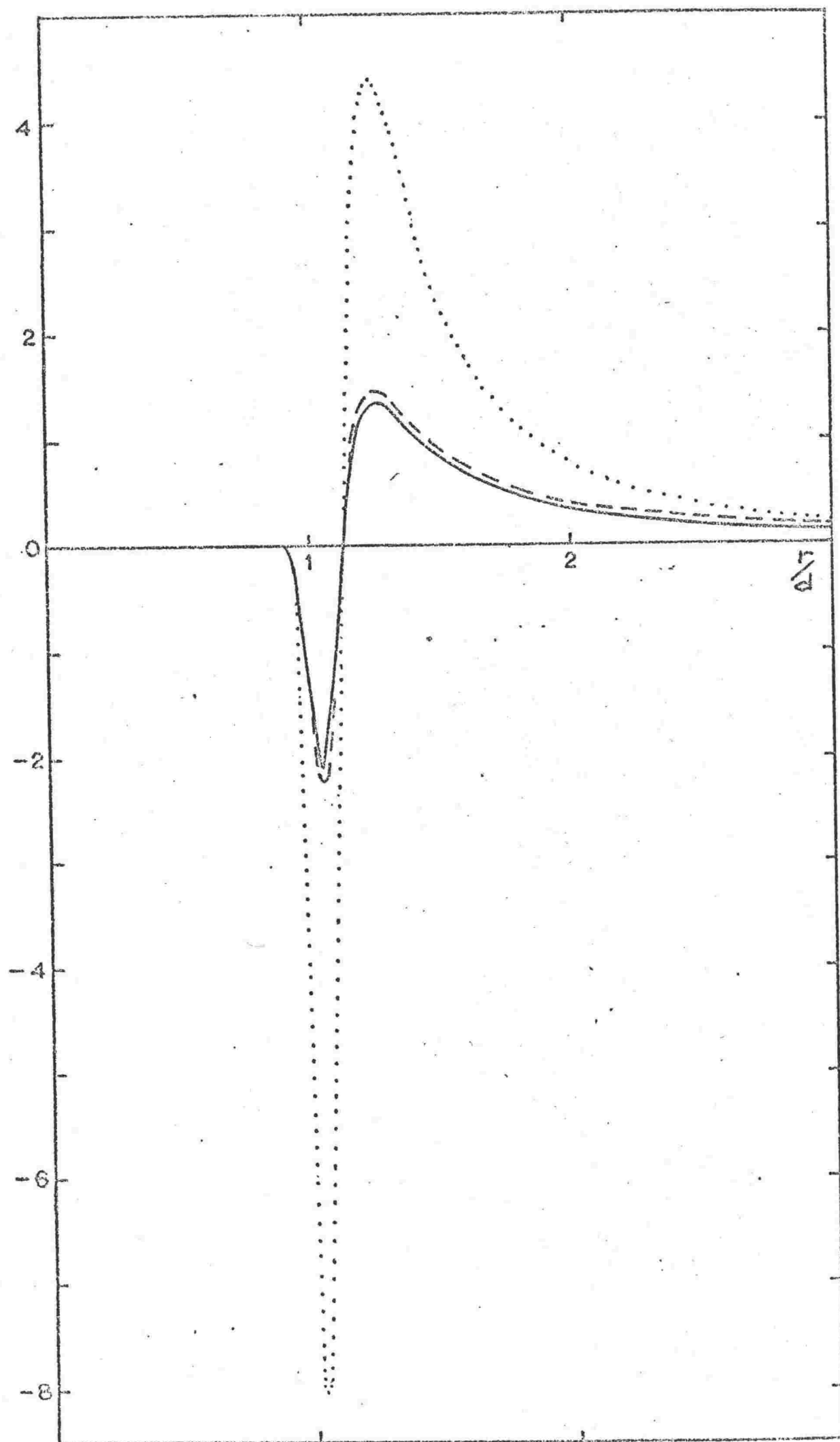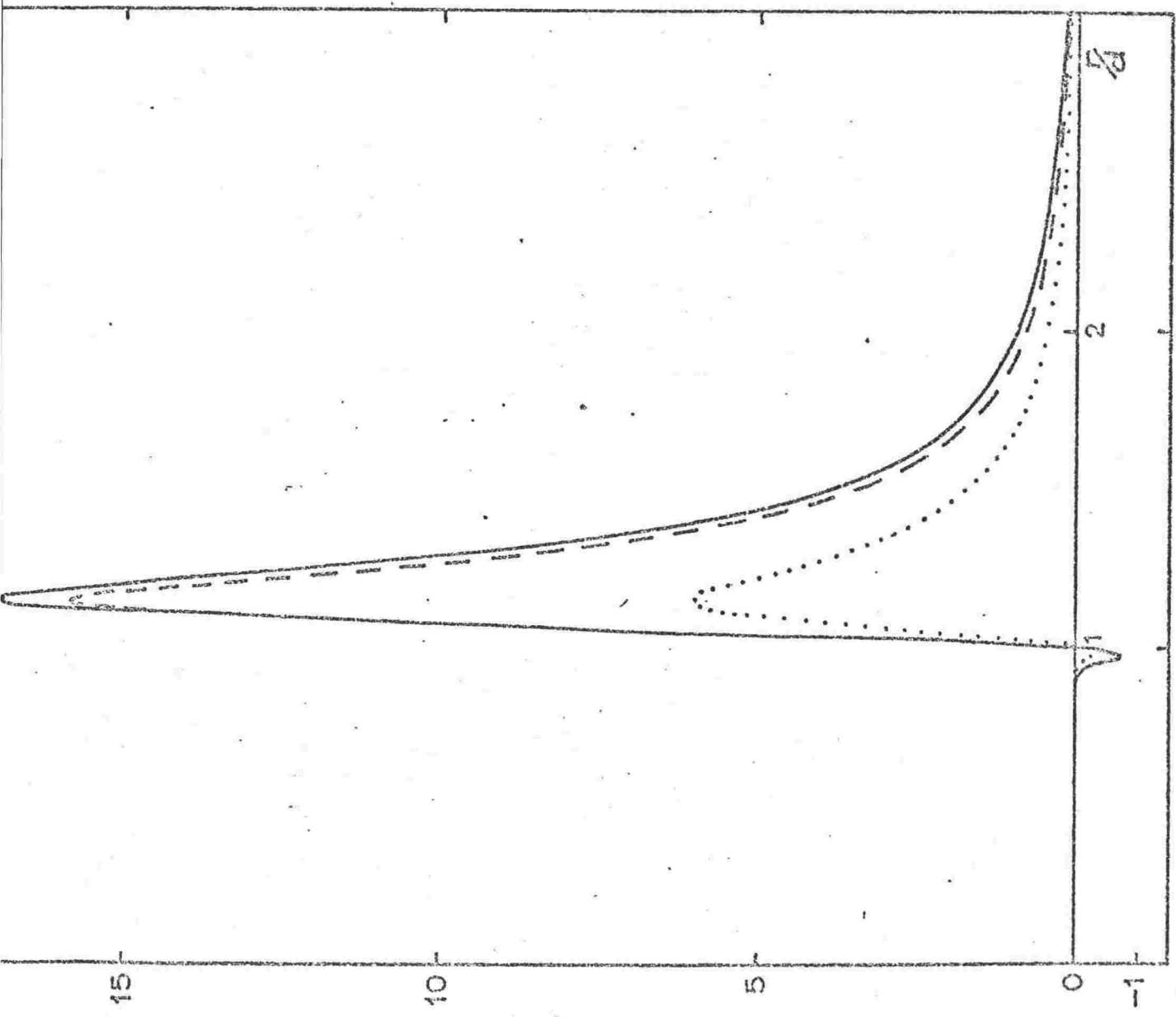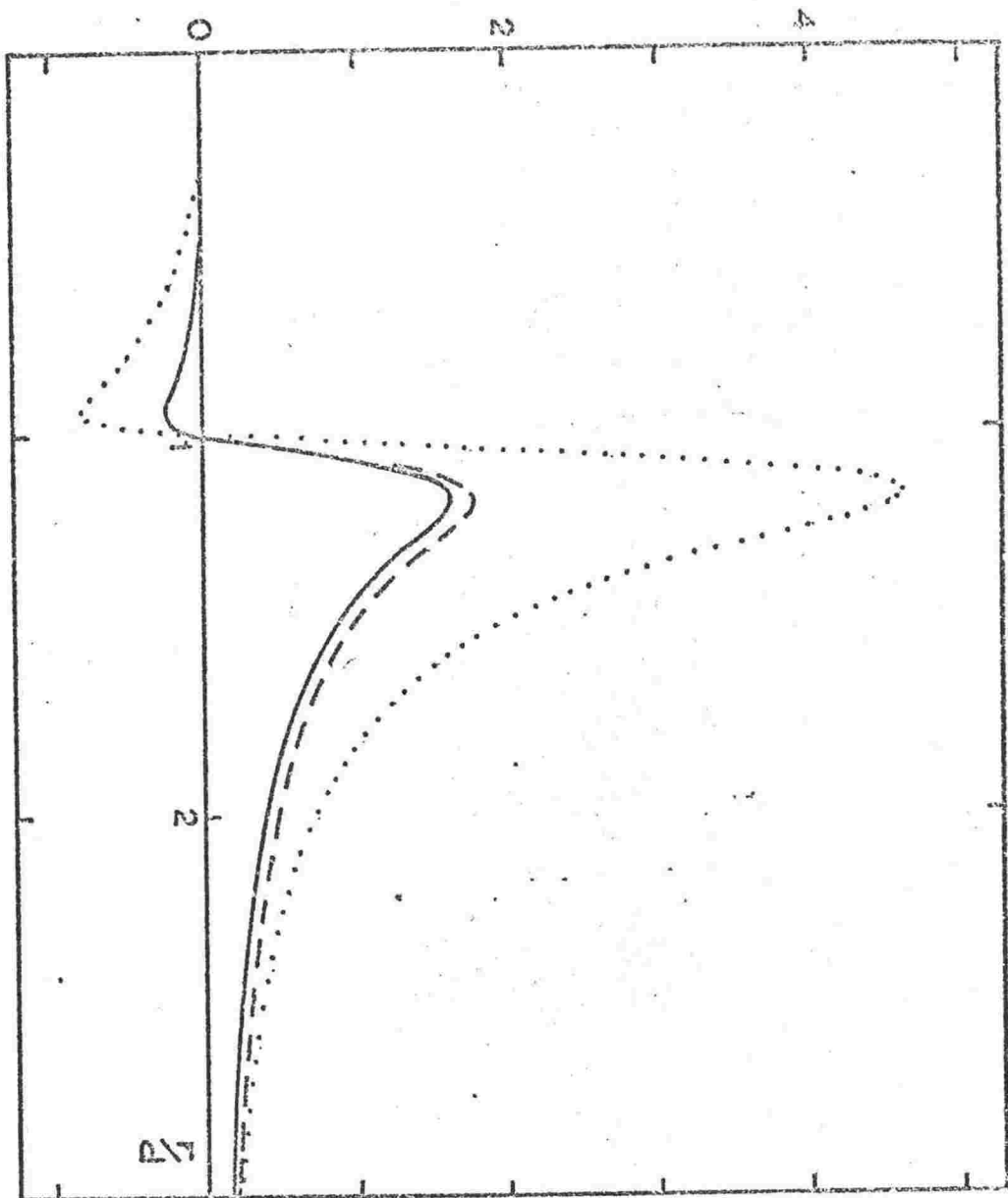tension.

Fig. 1
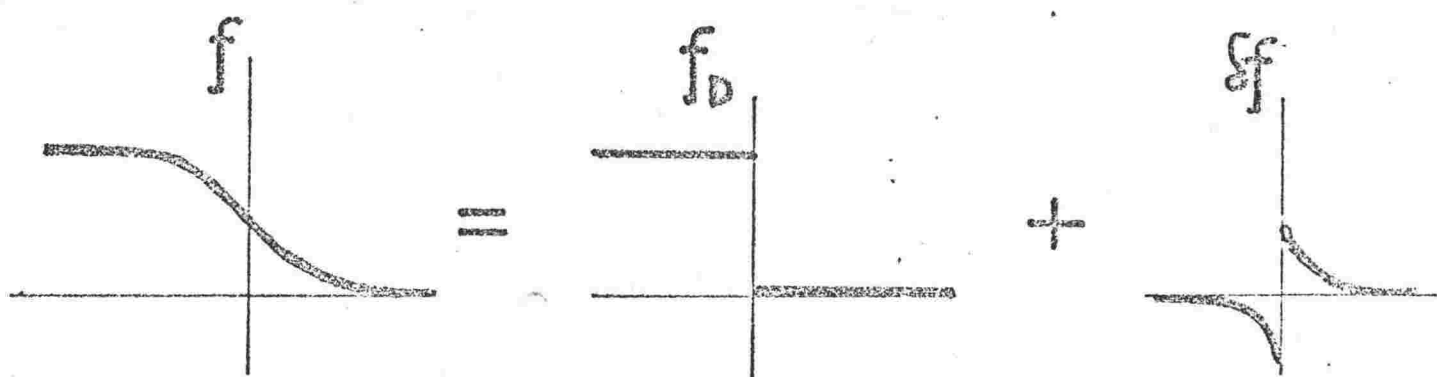
Fig 2.

Fig. 3

Fig. 4

Fig. 5

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THE SURFACE OF LIQUID He^4, BASED ON THE IDEA THAT

$\prod_{i<j} f(r_{ij})$ DESCRIBES A DROPLET

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ABSTRACT

We argue that the wavefunction $\prod f(r_{ij})$ describes the ground state of a droplet of liquid helium four. With this wavefunction expressions for the surface energy ε and the surface tension σ of liquid He^4 at T=0 are derived. Choosing particular f(r) and density profile, and the simplest pair correlation function, we plot the variation of ε and σ with surface thickness t. For slow variation of density at the surface, ε becomes proportional to t. The surface thickness is found to be about $4\overset{0}{A}$.

The inclusion of phonon zero-point motion correlations in the wavefunction leads (at T=0) to a $-R^2 \log R$ term in the energy of a droplet of radius R, implying a logarithmic divergence in both ε and σ. At T>0 the phonon correlations give a log T dependence of ε and σ and a negative bulk specific heat. Suggestions as to the reason for these problems are explored, but no definite conclusions are reached.

1. INTRODUCTION

The ground state of a drop of liquid helium is usually approximated by a Bijl-Jastrow product over pairs times a Hartree factor, namely

$$\prod_{i<j}^{N} f(r_{ij}) \quad \prod_{\ell}^{N} s(r_{\ell})$$

The function $s(r_{\ell})$ is used to control the density variation at the surface, and is determined variationally. (Bowley 1970; Shih and Woo 1973; Chang and Cohen 1973; Liu, Kalos and Chester 1975).

In this paper we put forward the view that the function

$$\Psi(1...N) = \prod_{i<j}^{N} f(r_{ij}) \tag{1}$$

already describes a droplet, and thus necessarily has a surface. There are three arguments to support this: First we note that Ψ is both translationally and rotationally invariant, so that it could describe only spherically symmetric self-bound systems. Second, we can argue by analogy with a classical fluid consisting of N atoms interacting with pairwise central forces. This has a probability density proportional to

$$\exp\{ -\frac{1}{T} \sum_{i<j}\sum u(r_{ij})\}$$

and thus has the same form as Ψ^2, the quantum probability density. For temperatures and pressures lying between the triple and the critical points, this probability density will describe a drop in equilibrium with its vapour. (We note in passing that this classical analogy implies that the function $f(r)$

should not be monotonically increasing (as is usually assumed)
if it is to describe a self-bound system, since a classical
system does not have condensation unless the pair potential
$u(r)$ contains an attractive part. A similar view has been
expressed recently by De Michelis and Reatto (1974)). The
third argument in favour of our assertion that $\prod f(r_{ij})$ describes
a droplet is provided cumulatively by the results of this paper,
where we calculate the surface energy and the surface tension
of the Bijl-Jastrow product (1). The results, both analytically
and numerically, are entirely satisfactory provided we leave
out the r^{-2} correlations, which Reatto and Chester (1967) assert must
exist in the bulk pair function f for all Bose systems that
have phonon excitations.

Our calculation of the surface energy and tension
with the wavefunction Ψ of equation (1) can be regarded as the
zeroth order approach to the problem. It is not a variational
calculation because the pair function f is determined by mini-
mizing the total energy, which is dominated by the bulk energy
when the radius of the drop is macroscopic. That is, we take
whatever function minimizes the bulk energy, and calculate the
consequent surface energy and tension. There is no doubt that
the inclusion of a variational factor such as s(r) will lower
the total energy and hence probably give a better description
of the surface. We believe, however, that our zeroth order
calculation is simpler   and yet both analytically
and numerically not very far from reality.

In Section 2 we calculate the surface energy of

a spherical drop at T=0 directly from the expectation
value of the Hamiltonian in the state Ψ. By letting the
radius of the drop tend to infinity we discuss the
simpler case of a plane surface with gravity and wall
forces absent. Extensive use is made of results previously
derived for monatomic classical liquids (Lekner and
Henderson (1977), referred to below as LH).

In Section 3 our results for ε (surface energy per
unit area) together with experimental values for ε and
the binding energy per atom in the bulk are used to calculate
the surface thickness of liquid He^4. We find a 10-90
thickness of about 3.9Å, in good agreement with recent
estimates.

In Section 4 we use the Toda (1955) and Brout and
Nauenberg (1958) expression for the surface tension σ of
a quantum fluid to obtain σ for the wavefunction Ψ. Curves
of σ and ε versus λ (a length characterizing the density
variation at the surface) are computed, using the simplest
approximation for the pair correlation function g(r).
Since $\sigma=\varepsilon$ at absolute zero, the curves should intercept
at the value of λ which corresponds to the actual density
profile. Despite the crude g(r) this method yields a surface
thickness in good agreement with Section 3 (about 3.4Å).

Finally in Section 5 we find that the inclusion of
Reatto and Chester's phonon factor leads to unphysical
results, namely divergences in ε and σ and a negative
bulk specific heat. We conclude that one of the following
is incorrect: the phonon factor, or our assumption that the
wavefunction Ψ is enough to describe a surface.

2. SURFACE ENERGY OF $\Pi f(r_{ij})$

The expectation value of the Hamiltonian

$$H = -\frac{\hbar^2}{2m} \sum_{i=1}^{N} \nabla_i^2 + \sum_{i<j}^{N}\sum^{N} u(r_{ij}) \tag{2}$$

when the wavefunction is of the form (1) may be written

as (see for example Lekner (1972))

$$\langle H \rangle = \frac{1}{2} \int d\underline{r}_1 d\underline{r}_2 \, n(\underline{r}_1,\underline{r}_2)\left[u(r_{12}) - \frac{\hbar^2}{4m}\left(\phi''(r_{12}) + \frac{2}{r}\phi'(r_{12})\right)\right] \tag{3}$$

where the two-particle density correlation function

is defined by

$$n(\underline{r}_1,\underline{r}_2) = N(N-1)\frac{\int d\underline{r}_3 \ldots d\underline{r}_N \, \Psi^2}{\int d\underline{r}_1 \ldots d\underline{r}_N \, \Psi^2} \tag{4}$$

and where we have written

$$f(r) = \exp\left[\tfrac{1}{2}\phi(r)\right] \tag{5}$$

To extract the surface part of (3), we make the approximation

$$n(\underline{r}_1,\underline{r}_2) = n(r_1)\, n(r_2)\, g(r_{12}) \tag{6}$$

where $n(r_1)$ is the single-particle density measured relative
to the centre-of-mass, and $g(r_{12})$ is the pair correlation
function.   Equation (6) really has two approximations in it:
the first is that we have broken the translational invariance
of the system described by Ψ of equation (1), and have by
some means localized the centre-of-mass of the droplet at
the origin.   The second approximation is that we have assumed
that the pair correlation function is a function of r_{12} only,
which can only be true inside the liquid.   This second
approximation has been discussed by LH($\S 3$) in the classical
context; for our purposes the disadvantage of its numerical
inaccuracy is outweighed by the simplicity of the analytic

results which it makes possible.

With the approximation (6) then, the expectation value
of the energy becomes

$$\langle H \rangle = \tfrac{1}{2} \int \underline{dr_1}\, \underline{dr_2}\; n(r_1)\, n(r_2)\, h(r_{12}) \tag{7}$$

where

$$h(r) = g(r)\left\{ u(r) - \frac{\hbar^2}{4m} \left[\phi''(r) + \frac{2}{r}\phi'(r) \right] \right\} \tag{8}$$

In bipolar coordinates (Hill 1956, p203), equation (7) reads

$$\langle H \rangle = (2\pi)^2 \int_0^\infty dr_1\, r_1\, n(r_1) \int_0^\infty dr_2\, r_2\, n(r_2) \int_{|r_1-r_2|}^{r_1+r_2} dr\, r\, h(r) \tag{9}$$

By successive interchange of the order of integration, the
energy may be written as

$$\langle H \rangle = 2\pi \int_0^\infty dr\, r^2\, h(r)\, Q(r) \tag{10}$$

where

$$Q(r) = \frac{2\pi}{r} \int_0^\infty dr_1\, r_1\, n(r_1) \int_{|r-r_1|}^{r+r_1} dr_2\, r_2\, n(r_2) \tag{11}$$

The function Q is proportional to the volume overlap of two
identical spherical density distributions, whose centers are
separated by distance r. $Q(r)$ may be evaluated analytically
for simple density variations. The simplest of these
is the step-function

$$n(r_1) = \begin{cases} n_0 & r_1 < R \\ 0 & r_1 > R \end{cases} \tag{12}$$

For the step-function density, Q becomes n_0^2 times the
volume common to two spheres of radius R, separated by distance
r. This function is well known (Hill 1956, p210):

$$Q_0(r) = \begin{cases} \frac{4}{3}\pi R^3 n_0^2 \left[1 - \frac{3}{2}\left(\frac{r}{2R}\right) + \frac{1}{2}\left(\frac{r}{2R}\right)^3\right] & (r < 2R) \\ 0 & (r > 2R) \end{cases} \tag{13}$$

We have thus shown that the expectation value of the energy of a droplet with a step-function density (radius R) is

$$E(R) = \left(\frac{4}{3}\pi R^3\right) 2\pi n_0^2 \int_0^{2R} dr\, r^2 h(r) \qquad \text{(volume term)}$$

$$\tag{14}$$

$$- (4\pi R^2)\frac{\pi}{2} n_0^2 \int_0^{2R} dr\, r^3 h(r) \qquad \text{(surface term)}$$

$$+ \frac{\pi^2}{6} n_0^2 \int_0^{2R} dr\, r^5 h(r) \qquad \text{(constant term)}$$

When h(r) is short-ranged the upper limit may be replaced by infinity in each case, and we regain the usual volume energy

$$E/N = 2\pi n_0 \int_0^\infty dr\, r^2 h(r) \tag{15}$$

plus a step-function contribution to the energy per unit area of the surface,

$$\epsilon_0 = -\frac{\pi}{2} n_0^2 \int_0^\infty dr\, r^3 h(r) \tag{16}$$

When h(r) varies as r^{-4} due to the zero point vibration of the phonons, the separation into volume, surface and constant terms is lost. We shall return to this problem in Section 5.

Having shown how we can extract a surface energy
from Ψ for spherical symmetry, we turn to the simpler case
of a plane geometry which is the $R \to \infty$ limit of the
spherical case. For the plane geometry we can use our
work on the surface energy of a classical liquid-vapour
interface (LH §3), since there is mathematically a one-to-
one correspondence between a classical system with probability
distribution

$$\exp\left\{-\frac{1}{T}\sum_{i<j}^{N}\sum u(r_{ij})\right\}$$

and our quantum system with probability distribution given
by

$$\Psi^2 = \exp\left\{\sum_{i<j}^{N}\sum \phi(r_{ij})\right\}$$

From (3), with the plane surface approximation

$$n(r_1, r_2) = n(z_1)n(z_2)g(r_{12}) \tag{17}$$

we have for the surface energy per unit area, (c.f. LH eqns (69),(70))

$$\epsilon = \pi n_0^2 \int_0^\infty dr\, r\, h(r)\left[-\tfrac{1}{2}r^2 + q_1(r) + q_2(r)\right] \tag{18}$$

with

$$q_1(r) = -2\int_{-r}^{r} dz\, z\, \delta f(z) - 2r\left[\int_r^\infty dz\, \delta f(z) - \int_{-\infty}^{-r} dz\, \delta f(z)\right] \tag{19}$$

$$q_2(r) = \int_{-\infty}^{\infty} dz_1\, \delta f(z_1)\int_{z_1-r}^{z_1+r} dz_2\, \delta f(z_2)$$

The function δf is defined by

$$\frac{n(z)}{n_0} = f(z) = f_0(z) + \delta f(z) \tag{20}$$

where f_0 is a step-function (see Fig.1).

$$\text{Fig. I.}$$

The location of the surface is not arbitrary, since the derivation of (18) and (19) depends on the property

$$\int_{-\infty}^{\infty} dz \, \delta f(z) = 0 \tag{21}$$

which defines the (Gibbs) dividing surface.   In the above formulae the dividing surface is at $z = 0$.

The first term of (18) has already been derived (eqn.(16)); the spherical counterparts of the other terms can also be derived and can be shown to be the same $_\wedge$ in the **as (19)** limit of large R, as   expected.

For an exponential density variation

$$n(z) = \begin{cases} n_0(1 - \tfrac{1}{2} e^{z/\lambda}) & z < 0 \\ \tfrac{1}{2} n_0 e^{-z/\lambda} & z > 0 \end{cases} \tag{22}$$

or $$\delta f(z) = \tfrac{1}{2} \operatorname{sgn} z \, \exp(-|z|/\lambda) \tag{23}$$

the surface energy becomes (LH §4)

$$\epsilon = -\frac{\pi}{2} n_0^2 \int_0^{\infty} dr \, r \, h(r) \left\{ r^2 + \lambda^2 \left[4 - e^{-r/\lambda}(4 + r/\lambda) \right] \right\} \tag{24}$$

For a Fermi function density variation,

$$n(z) = \frac{n_0}{e^{z/\delta}+1} \tag{25}$$

or

$$\delta f(z) = \frac{\text{sgn}(z)}{e^{|z|/\delta}+1} \tag{26}$$

it is straightforward to show (compare with Bowley's (1970) result):

$$\epsilon = -\frac{\pi}{2}n_0^2 \int_0^\infty dr\, r\, h(r)\left\{r^2 + 4\delta^2 \int_0^{r/\delta} dx\, \frac{x}{e^x-1}\right\} \tag{27}$$

As the surface thickness (proportional to λ and δ in the two chosen densities) goes to zero, we regain the step-function result, eq.(16) from both (24) and (27). As the surface thickness becomes large compared to the range of $h(r)$ we have the limiting forms

$$\epsilon \longrightarrow \epsilon_\infty = -\frac{3\pi}{2}n_0^2 \lambda \int_0^\infty dr\, r^2 h(r) \tag{28}$$

and

$$\epsilon \longrightarrow \epsilon_\infty = -2\pi n_0^2 \delta \int_0^\infty dr\, r^2 h(r) \tag{29}$$

The fact that in the limit of large surface thickness the surface energy becomes proportional to the thickness of the liquid-vacuum interface can be shown more generally by a Taylor expansion method $\left(\text{LH } \S 5\right)$. One finds that, neglecting second order and higher derivatives of the density, the surface energy is given by

$$\epsilon \simeq 2\pi n_0^2 \int_0^\infty dr\, r^2 h(r)\left\{\int_{-\infty}^\infty dz\, [\delta f(z)]^2 - 2\int_0^\infty dz\, \delta f(z)\right\} \tag{30}$$

When the density variation is characterized by a single length (such as λ or δ) both of the integrals in the curly brackets are proportional to this length; in particular on inserting (23) or (26) into (30) we regain (28) or (29).

In Fig. 2 we have plotted the integrands of the expressions (16), (24) and (28) for the surface energy. The full expression (24) and the slowly varying density limit (28) are both plotted for $\lambda=d$, where d is the hard core diameter of the Lennard-Jones potential

$$u(r) = 4\sigma[(\frac{d}{r})^{12} - (\frac{d}{r})^6]$$

$\longleftarrow$ The function ϕ was taken to be $-2(d/r)^5$ (this is close to the optimum among forms of the type $-a(b/r)^n$ tried by McMillan, 1965). The pair correlation function was approximated by e^ϕ.

Fig. 2 .

In Fig. 3 the values of the surface energy obtained for the exponential density variation are shown as a function of λ/d.

We see that ε is greater than ε_∞, and increases monotonically with λ. The fact that ε does not show even a local minimum at some physical value of λ/d need not surprise us, since we are not doing a variational calculation. However, we should issue a word of caution that the choice of g has a large effect on the curves in Fig. 3. From Fig. 2 it can be seen that the magnitude of g in the region $r \lesssim d$ affects the negative part of the integrand, while the first maximum, which is present in a realistic g, affects the positive part. It so happens that the crude approximation

$$g = e^{-2(d/r)^5}$$

contains cancelling errors that lead to a very reasonable result. If a more realistic pair correlation function is used in order to include the effect of a maximum, then equal care must be exercised in the $r \lesssim d$ region.

Fig. 3

3. THE SURFACE THICKNESS

The problem of assigning one length as the thickness of the surface has no unique solution when one is comparing different density profiles. The length we have found most useful for numerical comparisons is the 10-90 thickness, which is the distance in which the density rises from 10% of its bulk value to 90% of its bulk value. If we call this thickness t, then for the exponential and Fermi function profiles we have

$$t = 2\lambda \log 5 \simeq 3.22\lambda$$

and

$$t = 2\delta \log 9 \simeq 4.39\delta$$

In terms of this definition we see that the exponential and Fermi profiles give the same thickness when

$$\lambda/\delta = \log 9/\log 5 \simeq 1.365$$

Consider the slowly varying density limiting expressions (28) and (29) for the surface energy. From (14) we note that these can be written as

$$\varepsilon_\infty = -\tfrac{3}{4}\lambda E/V \tag{31}$$

and

$$\varepsilon_\infty = -\delta E/V \tag{32}$$

where E/V is the bulk energy per unit volume. [Note that these expressions are the same when $\lambda = (4/3)\delta$, which compares well with the 10-90 equivalence condition $\lambda = 1.365\delta$]. We are now in a position to evaluate the surface thickness from our $\varepsilon(\lambda)$ curve and experimental data: rewrite (31) as

$$\lambda = -\frac{4}{3} \left\{ \frac{\varepsilon}{E/V} \right\} \frac{\varepsilon_\infty}{\varepsilon} \tag{33}$$

For the quantities in the curly brackets we can substitute the experimental values $\varepsilon = 0.373 \, erg/cm^2$ (Atkins and Narahara 1965) and $E/N = -.99 \times 10^{-15} \, erg/atom$, $N/V = n_0 = 2.2 \times 10^{22} \, atoms/cm^3$; this gives $\lambda \simeq 2.3 \, (\varepsilon_\infty/\varepsilon) \, \text{Å}$. With $d = 2.6 \, \text{Å}$ we then find

$$\frac{\varepsilon_\infty}{\varepsilon} \simeq 1.14 \frac{\lambda}{d} \tag{34}$$

In Fig. 4 we plot this line, together with the ratio $\varepsilon_\infty/\varepsilon$ obtained from results of Fig. 3.

Fig. 4.

The line (34) and the curve $\varepsilon_\infty/\varepsilon$ intercept at $\lambda/d \simeq 0.46$, which gives a 10-90 thickness

$$t = 3.9 \overset{0}{A}$$

This estimate has the advantage that nature evaluates two very difficult integrals for us (ε and E/N), and all we have had to find is the ratio $\varepsilon_\infty/\varepsilon$, which we expect to be insensitive to the errors in our choice of the pair correlation function g and the pair function f. The agreement with recent estimates, based on a comparatively enormous amount of numerical work, is good: Chang and Cohen (1973) obtained $\lambda \simeq 1.4 \overset{0}{A}$ and $\delta \simeq 1.0 \overset{0}{A}$, giving $t \simeq 4.6 \overset{0}{A}$ and $4.4 \overset{0}{A}$ respectively, while Liu, Kalos and Chester (1975) calculate $t \simeq 5 \overset{0}{A}$.

We conclude this section by pointing out the physical meaning of the slowly varying density limits, eqns.(31) and (32). Consider eqn.(31) first and let A be the area of the surface. Then according to (31) the surface energy is

$$\Delta E = A\varepsilon \simeq -\frac{3}{4} \lambda A \frac{E}{V}$$

The 10-90 thickness of the surface is 3.22 λ, so that the volume ΔV of the interface is approximately 3.22 λA. The number of atoms in the interface is $\Delta N \simeq \frac{1}{2} n_o \Delta V$, so that

$$\Delta E \simeq -\frac{3}{4}(\frac{2}{3.22}) \Delta N \frac{E}{N} \simeq -\frac{1}{2} \Delta N \frac{E}{N}$$

The same approximate relation follows from the Fermi function limiting form (32). The physical meaning of (31) and (32) is thus clear: the positive surface

energy corresponds to the loss of approximately one half the binding energy per atom for every atom in the surface layer.

4.   SURFACE TENSION OF $\Pi f(r_{ij})$

Toda (1955) and Brout and Nauenberg (1958) have derived general expressions for the surface tension of a quantum liquid.   Their result is the sum of a potential energy contribution σ_p and a kinetic energy contribution σ_k :

$$\sigma = \sigma_p + \sigma_k \tag{35}$$

where, at T = 0

$$\sigma_p = \frac{1}{4 L_x L_y} \int d\underline{r_1} \int d\underline{r_2}\, n(\underline{r_1}, \underline{r_2})\, \frac{r_{12}^2 - 3 z_{12}^2}{r_{12}}\, \frac{du(r_{12})}{dr_{12}} \tag{36}$$

and

$$\sigma_k = \frac{1}{L_x L_y} \left(-\frac{N \hbar^2}{m} \right) \left\langle \frac{\partial^2}{\partial z_i^2} - \frac{\partial^2}{\partial x_i^2} \right\rangle \tag{37}$$

The above formulae have been written for a plane interface in the x-y plane, of area $L_x\, L_y$.   The potential energy contribution is the classical expression evaluated in

LH$\quad$, and the kinetic energy term may be put into the same form as the potential energy term when the wavefunction is a product of pair functions.   We have, with

$$\Psi = e^{\Phi/2}, \quad \Phi = \sum_{i < j} \phi(r_{ij}),$$

$$\left\langle \frac{\partial^2}{\partial z_i^2} \right\rangle = \int d\underline{r_1} \dots d\underline{r_N}\, \Psi \frac{\partial^2}{\partial z_i^2} \Psi$$

$$= \int d\underline{r_1} \dots d\underline{r_N}\, \Psi^2 \left\{ \frac{1}{4} \left(\frac{\partial \Phi}{\partial z_1} \right)^2 + \frac{1}{2} \frac{\partial^2 \Phi}{\partial z_i^2} \right\}$$

Integrating the RHS of the first equality by parts we also have

$$\left\langle \frac{\partial^2}{\partial z_1^2} \right\rangle = -\int d\underline{r}_1 \ldots d\underline{r}_N \left(\frac{\partial \Psi}{\partial z_1}\right)^2 = -\frac{1}{4}\int d\underline{r}_1 \ldots d\underline{r}_N \Psi^2 \left(\frac{\partial \Phi}{\partial z_1}\right)^2$$

Therefore

$$\left\langle \frac{\partial^2}{\partial z_1^2} \right\rangle = \frac{1}{4}\int d\underline{r}_1 \ldots d\underline{r}_N \Psi^2 \frac{\partial^2 \Phi}{\partial z_1^2}$$

$$= \frac{N-1}{4}\int d\underline{r}_1 \ldots d\underline{r}_N \frac{1}{r_{12}}\left\{ \frac{d\phi(r_{12})}{dr_{12}} + z_{12}^2 \frac{d}{dr_{12}}\left[\frac{1}{r_{12}}\frac{d\phi(r_{12})}{dr_{12}}\right]\right\} \Psi^2 \quad (38)$$

Thus

$$\sigma_k = \frac{1}{4L_xL_y}\left(-\frac{\hbar^2}{m}\right)N(N-1)\int d\underline{r}_1 \ldots d\underline{r}_N \frac{z_{12}^2 - x_{12}^2}{r_{12}}\frac{d}{dr_{12}}\left[\frac{1}{r_{12}}\frac{d\phi(r_{12})}{dr_{12}}\right]\Psi^2$$

$$= \frac{1}{4L_xL_y}\left(-\frac{\hbar^2}{2m}\right)\int d\underline{r}_1 \int d\underline{r}_2\, n(\underline{r}_1,\underline{r}_2)\frac{3z_{12}^2 - r_{12}^2}{r_{12}}\frac{d}{dr_{12}}\left[\frac{1}{r_{12}}\frac{d\phi(r_{12})}{dr_{12}}\right] \quad (39)$$

For a plane surface we can write

$$n(\underline{r}_1,\underline{r}_2) = n(z_1)n(z_2)g(r_{12},z_1,z_2) \quad (40)$$

and then the surface tension reduces to (cf. LH , Section 2)

$$\sigma = \frac{\pi}{2}\int_{-\infty}^{\infty}dz_1\, n(z_1)\int_{-\infty}^{\infty}dz_2\, n(z_2)\int_{|z_{12}|}^{\infty}dr\, g(r,z_1,z_2)[r^2-3z_{12}^2]\left[u' + \frac{\hbar^2}{2m}\left(\frac{\phi'}{r}\right)'\right] \quad (41)$$

If $g(r,z_1,z_2)$ is approximated by $g(r)$, we get (cf. eqn.(50) of LH)

$$\sigma = \frac{\pi}{2}n_0^2\int_0^{\infty}dr\, g(r)\left[u' + \frac{\hbar^2}{2m}\left(\frac{\phi'}{r}\right)'\right]\left\{\frac{1}{4}r^4 + p_1(r) + p_2(r)\right\} \quad (42)$$

where

$$P_1(r) = -2 \int_{-r}^{r} dz \, \delta f(z) \, z \, (r^2 - z^2)$$

$$P_2(r) = \int_{-\infty}^{\infty} dz_1 \, \delta f(z_1) \int_{z_1-r}^{z_1+r} dz_2 \, \delta f(z_2) (r^2 - 3z_{12}^2) \tag{43}$$

For the exponential density profile (23) we have from

LH, eqn.(77)

$$\sigma = \frac{\pi}{2} n_0^2 \int_0^{\infty} dr \, g(r) \left[u' + \frac{\hbar^2}{2m} \left(\frac{\phi'}{r} \right)' \right] \left\{ \frac{r^4}{4} + \lambda^4 \left[18 - 2\frac{r^2}{\lambda^2} - \bar{e}^{r/\lambda} \left(18 + 18\frac{r}{\lambda} + 7\frac{r^2}{\lambda^2} + \frac{r^3}{\lambda^3} \right) \right] \right\} \tag{44}$$

which has the large λ limit (cf. LH, eqn.(80)

$$\sigma \rightarrow \sigma_{\infty} = \frac{\pi}{30} n_0^2 \frac{1}{\lambda} \int_0^{\infty} dr \, g(r) \left[u' + \frac{\hbar^2}{2m} \left(\frac{\phi'}{r} \right)' \right] r^5 \tag{45}$$

For the Fermi function density profile (26) we use the

methods of LH (Section 4) to evaluate p_1 and p_2.

We find

$$\sigma = \frac{\pi}{2} n_0^2 \int_0^{\infty} dr \, g(r) \left[u' + \frac{\hbar^2}{2m} \left(\frac{\phi'}{r} \right)' \right] \left\{ \frac{r^4}{4} + 2\delta^4 \int_0^{r/\delta} dx \frac{x}{e^x - 1} \left(3x^2 - \frac{r^2}{\delta^2} \right) \right\} \tag{46}$$

The large δ limit is

$$\sigma \rightarrow \sigma_{\infty} = \frac{\pi}{45} n_0^2 \frac{1}{\delta} \int_0^{\infty} dr \, g(r) \left[u' + \frac{\hbar^2}{2m} \left(\frac{\phi'}{r} \right)' \right] r^5 \tag{47}$$

Note that the exponential and Fermi profiles give the

same σ_{∞} if $\lambda = \frac{3}{2} \delta$, whereas the surface energies were

equal in this limit when $\lambda = \frac{4}{3} \delta$.

The limiting results obtained above are special cases

of the general formula for slowly varying density (c.f.
LH eqn.(83))

$$\sigma = \frac{2\pi}{15} \int_0^\infty drg(r) \left[u' + \frac{\hbar^2}{2m} \left(\frac{\phi'}{r} \right)' \right] r^5 \int_{-\infty}^\infty dz \left(\frac{dn}{dz} \right)^2, \quad (48)$$

which is obtained by a Taylor expansion of the density,
neglecting fourth order and higher derivatives of the
density. As in the case of surface energy the odd deriva-
tive terms in the expansion give zero contribution so that
correction terms are two orders higher in the gradient.
In Fig. 5. we have plotted, in the manner of Fig. 2.,
the integrand of the step function limit for the surface
tension

$$\sigma_0 = \frac{\pi}{8} n_0^2 \int_0^\infty drg(r) \left[u' + \frac{\hbar^2}{2m} \left(\frac{\phi'}{r} \right)' \right] r^4 \qquad (49)$$

along with integrands of the exponential density expres-
sion (44) and its large λ limit (45), at $\lambda = d$.

$$\text{Fig. 5.}$$

The values of surface tension as a function of
λ/d, obtained with the zeroth approximation pair correlation
function, $g = e^\phi$, are shown in Fig. 3. The curves for ε
and σ intersect at $\lambda = .41d$ and since $\varepsilon = \sigma$ at absolute
zero, provided the location of the surface is defined
by (21) (see Section 3 of LH), we have another estimate

of the surface thickness: $t = 3.22\lambda \simeq 3.4\overset{0}{\text{A}}$. This value
is likely to be less accurate than our previous estimate
of Section 3, since it is based on a first principles
calculation with a crude g and no direct experimental
input. Nevertheless, the intercept energy 0.43 erg/cm^2
is remarkably close to the experimental value of 0.37 erg/cm^2,
implying that the severe approximations that we made to
obtain the curves of Fig. 3 contain cancelling errors
(see discussion at the end of Section 2).

Sections 2 to 4 support our initial assertion that
$\prod_{i<j} f(r_{ij})$ describes a droplet, but we should remember
that these results are based on the assumption that $\phi(r)$
decays rapidly at large r.

5. PROBLEMS WITH $\prod f(r_{ij})$ WHEN ZERO POINT MOTION OF PHONONS IS INCLUDED

In the previous sections we found that the wavefunction

$$\prod_{i<j} e^{\frac{1}{2}\phi(r_{ij})} \tag{50}$$

gave good results for the surface energy and thickness of liquid helium provided $\phi(r)$ tends to zero rapidly as $r \to \infty$. In particular the McMillan form

$$\phi(r) = -2\left(\frac{d}{r}\right)^5 \tag{51}$$

led to reasonable results.

However, Reatto and Chester (1967) have shown that the existence of phonon excitations in liquid helium implies that a factor of the ground state wavefunction is of the form (50), with

$$\phi(r) \to \phi_p = -\frac{b^2}{r^2} \tag{52}$$

where

$$b = \frac{1}{\pi}\left(\frac{mc}{n_0\hbar}\right)^{\frac{1}{2}} \simeq 2.6\overset{0}{A} \tag{53}$$

The term ϕ_p leads to a logarithmic divergence (to minus infinity) of both our surface energy and surface tension expressions, (18) and (42). This divergence has appeared in previous formulations for the surface energy of liquid helium and varying attempts have been made at understanding it (see Bowley (1970) and Chang and Cohen (1973), both of which are discussed below). Note also that the divergence invalidates the derivations of the large surface width expressions ε_∞ and σ_∞. To see clearly the

consequences of the phonon factor let us return to the droplet of
Section 2 and consider contributions to the total energy coming from
particle separations greater than 2a, where a is $\left(\text{say}\right)$ 5Å; then we
can take $\phi = \phi_\rho$, g = 1, and $u = 0$ to obtain the asymptotic
value

$$h(r) \longrightarrow \frac{\hbar^2 b^2}{2m} \frac{1}{r^4} \tag{54}$$

which can be regarded as an effective repulsive $1/r^4$ interaction.
Inserting (54) into (14) we find that the direct contribution of the
infinite-range correlations arising out of the zero point motion
of the phonon modes is

$$\Delta E = \frac{4}{3}\left(\hbar c n_0\right) R^3 \int_{2a}^{2R} dr\, r^{-2}\left(1 - \frac{3}{2}\left(\frac{r}{2R}\right) + \frac{1}{2}\left(\frac{r}{2R}\right)^3\right)$$

$$= \frac{2}{3}\left(\hbar c n_0\right) R^2 \left\{\frac{R}{a} - \frac{3}{2}\log\left(\frac{R}{a}\right) - \frac{3}{4} - \frac{1}{4}\frac{a^2}{R^2}\right\} \tag{55}$$

For a flat surface of area L^2 (54) similarly leads to a
positive volume term and a $-L^2\log\left(L/a\right)$ term. So at T = 0 the long
range part of the phonon correlations <u>raise</u> the bulk energy (i.e.
from a variational point of view they should be left out to lower
the total energy), and produce a negative $R^2\log\left(R/a\right)$ term, as well
as a negative R^2 term. The bulk energy is raised by $\hbar c / 2\pi a$
($\simeq 0.58 K$ when a = 5Å) per atom, which is small in comparison
with the experimental bulk energy, - 7.16 K per atom. On the other
hand, the "surface" energy is lowered by $\frac{\hbar c n_0}{4\pi}\left(\log\frac{R}{a} + \frac{1}{2}\right)$,
which is approximately 0.63 $erg\, cm^{-2}$ when R = .5 mm and a = 5 Å .
Thus for R large enough we have that at T = 0 the "surface" energy
would be negative, which seems impossible to accept.

It is known that at finite temperatures the long-range correlations due to the zero point motion are expoentially damped, so one might hope that the above problems in the ground state would disappear at finite temperature. However we find that this is not so. Reatto and Chester (1967) have shown that the diagonal part of the density matrix has the form of a product wavefunction, with the long-range part due to phonon zero point motion. The probability$_\wedge$ density is thus of the same form as before, with

$$\phi_p(r, T) \rightarrow -\frac{b^2}{r^2} \frac{\pi Tr/\hbar c}{\sinh(\pi Tr/\hbar c)} \qquad (56)$$

and so the kinetic energy term

$$- (\hbar^2/4m)\nabla^2 \phi_p$$

in the total energy again has the form of a repulsive interaction. The screening length $\hbar c/\pi T$ is approximately 5.8Å at 1K. Inserting (56) into (14) and considering contributions from particle separations greater than 2a, we find that the finite temperature phonon zero-point motion contribution to the bulk energy is (when $2\pi TR/\hbar c \gg 1$)

$$\Delta E_V = \tfrac{2}{3}(\hbar c \eta_o)\frac{R^3}{a} F\left(\frac{2\pi Ta}{\hbar c}\right) \qquad (57)$$

where $\quad F(x) = \tfrac{1}{2}\frac{x}{\sinh x}\left(1 + \frac{x\cosh x}{\sinh x}\right) \qquad (58)$

The low temperature expansion gives a negative contribution
to the specific heat proportional to T^3, since

$$F(x) = 1 - \frac{7}{360} x^4 + O(x^6) \qquad (59)$$

In fact the slope of F is always negative, so the screened
zero point motion contributes a negative specific heat,
with a maximum magnitude at $T \simeq .45\ \hbar c/a$. This result
is in accord with our statement above that the correlations
due to the zero point motion of the phonon modes are
equivalent to an effective repulsive r^{-4} interaction
which is screened thermally, because the higher the
temperature the better the screening and so the energy
due to the zero point motion of the phonons decreases
with T. For a of order of the atomic diameter or greater,
this negative T^3 specific heat is an order of magnitude
larger than the normal T^3 term due to thermal excitation
of phonons, so the total bulk specific heat is negative
up to $T \sim \hbar c/a \sim 1K$. This indicates an instability in the
system, since if the energy decreases with temperature
in any region, a spontaneous positive temperature fluc-
tuation will decrease the total energy and thus release
heat, further increasing the temperature, which will
in turn decrease the energy, and so on. Similarly, a neg-
atively temperature fluctuation will induce a continual
decrease in temperature.

The surface energy contribution of the zero point
motion of the phonon modes is readily obtained from (56)

and (14). We find

$$\Delta\epsilon = - \frac{\hbar c n_o}{8\pi} \left\{ \frac{x}{\sinh x} + 2F(x) - 2\log\tanh\frac{x}{2} \right\} \quad (60)$$

where $x = 2\pi Ta/\hbar c$. The derivation of this expression is valid only when the total surface energy is greater than zero, since a spherical shape is assumed in (14). The low temperature ($2\pi T \ll \hbar c/a$) expansion is

$$\Delta\epsilon = - \frac{\hbar c n_o}{4\pi} \left\{ \log\left(\frac{\hbar c}{\pi Ta}\right) + \tfrac{3}{2} + O\left(\frac{\pi Ta}{\hbar c}\right)^2 \right\} \quad (61)$$

The above expressions are again limited to the region $2\pi T R/\hbar c \gg 1$. From (61) we see that the surface contribution to the specific heat is positive, varies as T^{-1}, and will become larger than the previously noted negative T^3 bulk contribution when $T \lesssim \frac{\hbar c}{3a}(\frac{a}{R})^{\frac{1}{4}}$. This temperature is of the order of 10^{-2}K for a droplet of millimetre size.

The consequences of the long-range correlations arising out of the zero point motion of the phonons thus appear to be

(i) a negative bulk specific heat

(ii) a term in the energy varying as

$- R^2 \log(R/a)$ at T=0

(iii) a logarithmic temperature dependence of the surface energy at low temperatures, with measurable temperature variation at about 10^{-1}K and zero ϵ at about 10^{-3}K.

Bowley (1970), who first met one of the difficulties associated with the long-range phonon correlations (he found a divergence to minus infinity in the surface energy at T=0 when these correlations were included) has postulated that the inclusion of the zero point motion of the surface modes would remove this problem. The correlations due to surface mode zero point motion have yet to be evaluated explicitly, though Chang and Cohen (1975) have written down an expression for their contribution to the ground state wavefunction. We feel that it is unlikely that the surface wave zero point motion can remove a problem which arises from the existence of bulk modes, since we expect the correlations due to the surface modes to be of a different kind, and additional to the phonon modes, rather than cancelling them in a substantial part of the interior.

Another approach to these difficulties is that of Chang and Cohen (1973), who use the wavefunction

$$\Psi = \prod_{i<j} \exp \tfrac{1}{2}\phi(r_{ij}) \prod_{k} \exp \tfrac{1}{2}t(\underset{\sim}{r}_k) \tag{62}$$

where the function $t(\underset{\sim}{r})$ is optimized variationally, as in Bowley's work. However, instead of parametrizing t, as Bowley did, they eliminate t in favour of the density n, and parametrize n. We note in passing that Bowley's assumption that $n=n_o e^t$ leads to the same expression for ε as our work based on $\prod e^{\tfrac{1}{2}\phi}$, plus a small term

$$- (\hbar^2/8m) \int d\underset{\sim}{r} \, n(r) \, \nabla^2 t(r).$$

We will generalize Chang and Cohen's result for the energy and give a similar treatment for the surface tension. The expectation value of the Hamiltonian (2) taken with the trial function (62) is readily shown to be (c.f.(3))

$$\langle H \rangle = \tfrac{1}{2} \int d\underline{r}_1 \, d\underline{r}_2 \; n(\underline{r}_1, \underline{r}_2) \left\{ u(r_{12}) - \tfrac{\hbar^2}{4m} \nabla_1^2 \phi(r_{12}) \right\} - \tfrac{\hbar^2}{8m} \int d\underline{r}_1 \, n(\underline{r}_1) \nabla_1^2 t(\underline{r}_1) \tag{63}$$

The Yvon equation

$$\nabla_1 n(\underline{r}_1) = n(\underline{r}_1) \nabla_1 t(\underline{r}_1) + \int d\underline{r}_2 \; n(\underline{r}_1, \underline{r}_2) \, \nabla_1 \phi(r_{12}) \tag{64}$$

obtained by differentiating the defining relation for the density,

$$n(\underline{r}_1) = N \int d\underline{r}_2 \ldots d\underline{r}_N \, \Psi^2 \Big/ \int d\underline{r}_1 \ldots d\underline{r}_N \, \Psi^2 \tag{65}$$

can now be used to rewrite the last term in (63) in terms of the density n and the pair function ϕ. Then defining

$$n(\underline{r}_1, \underline{r}_2) = n(\underline{r}_1) n(\underline{r}_2) \, g(\underline{r}_1, \underline{r}_2) \tag{66}$$

and

$$g(\underline{r}_1, \underline{r}_2) = \exp\{\phi(r_{12}) + \gamma(\underline{r}_1, \underline{r}_2)\} \;, \tag{67}$$

the expectation value of the total energy becomes

$$\langle H \rangle = \tfrac{1}{2} \int d\underline{r}_1 \, d\underline{r}_2 \; n(\underline{r}_1, \underline{r}_2) \left\{ u(r_{12}) + \tfrac{\hbar^2}{4m} [(\nabla_1 \phi(r_{12}))^2 + \nabla_1 \phi(r_{12}) \cdot \nabla_1 \gamma(\underline{r}_1, \underline{r}_2)] \right\} + \tfrac{\hbar^2}{8m} \int d\underline{r}_1 \, n(\underline{r}_1) \, (\nabla_1 \log n(\underline{r}_1))^2 \tag{68}$$

In a uniform fluid, the HNC and PY equations for the pair correlation function may be used to evaluate γ (see for example Feenberg 1970, page 695)

$$\gamma(\underline{r}_1, \underline{r}_2) = \begin{cases} P(r_{12}) & \text{(HNC)} \\ \log[1+P(r_{12})] & \text{(PY)} \end{cases} \qquad (69)$$

where

$$P(r) = \frac{1}{(2\pi)^3 n_0} \int d\underline{k} \ e^{-i\underline{k}\cdot\underline{r}} \frac{[S(k)-1]^2}{S(k)}$$

$$= \frac{1}{2\pi^2 n_0 r} \int_0^\infty dk \ k \ \sin kr \ \frac{[S(k)-1]^2}{S(k)} \qquad , \qquad (70)$$

and

$$S(k) = 1 + n_0 \int d\underline{r} \ e^{i\underline{k}\cdot\underline{r}} [g(r)-1] \qquad (71)$$

is the structure factor. The large r variation of $P(r)$ (and hence of γ) is determined by the small k variation of $S(k)$. If we accept the correctness of the usual form, $S(k) \to \hbar k/2mc$ (Feynman (1954), Feenberg (1970)), then integrating the right hand side of (70) by parts and substituting for $S(k)$, gives for the asymptotic form of $P(r)$,

$$P(r) \to \frac{mc}{\pi^2 \hbar n_0} \frac{1}{r^2} - \frac{1}{2\hbar^2 n_0 mc} \frac{\hbar}{r^4} \frac{1}{r^4} + \dots \qquad (72)$$

Thus we have that

$$\gamma(r) \to P(r) = \frac{b^2}{r^2} + 0(r^{-4}) = -\phi_p(r) + 0(r^{-4}) \qquad (73)$$

i.e. using the HNC or PY equations the contributions of the zero-point phonon   correlations cancell in the first term of (68), at least deep inside the liquid. (We should however note that the equivalent classical fluid ($u = -T\phi$) corresponding to the product wavefunction (1) when $\phi \to -b^2/r^2$ has energy per particle and pressure both proportional to the size of the system, and thus the validity of the HNC and PY equations needs to be re-examined for this very singular case .) In fact provided γ goes to zero no

slower than r^{-1}, (68) will not diverge.

A similar result follows for the surface tension. We find, for the wavefunction (62), that the surface tension is given by (42), the expression derived for the wavefunction (1), plus the term

$$\Delta\sigma = \frac{1}{L_x L_y} \left(-\frac{\hbar^2}{4m}\right) \int d\underline{r}_1 \; t''(z_1) \; n(z_1) \tag{74}$$

(in this case we find it simpler to work with a plane surface). When we again eliminate t by using the Yvon equation, we find

$$\Delta\sigma = \frac{\hbar^2}{4m} \int dz \; n(z) \left(\frac{d}{dz} \log n(z)\right)^2$$
$$+\frac{\hbar^2}{4m} \frac{1}{L_x L_y} \quad \text{X surface part of} \int d\underline{r}_1 d\underline{r}_2 \, n(\underline{r}_1, \underline{r}_2)\Big\{\nabla_1^2\phi(r_{12})$$
$$+\big(\phi'(r_{12})\big)^2 + \nabla_1\phi(r_{12})\cdot\nabla_1\gamma(\underline{r}_1,\underline{r}_2)\Big\} \tag{75}$$

The singular part of the second term in (75) has the form (c.f.(3)&(18))

$$-\frac{\pi}{4} \; \frac{n_0^2 \hbar^2}{m} \; \int_0^\infty dr \; r^3 g(r) (\phi_p'' + \frac{2}{r} \phi_p') \tag{76}$$

while the singular part of (42) is

$$\frac{\pi}{16} \; \frac{n_0^2 \hbar^2}{m} \; \int_0^\infty dr \; r^4 \; g(r) \left(\frac{\phi_p''}{r}\right)' \tag{77}$$

and when ϕ_p varies as r^{-2}, these cancell.

Thus it appears that by using the wavefunction (62) instead of (1) the divergence in the surface tension and the surface energy has either been removed, or has been shifted to the density gradient term

$$\int d\underline{r} \; n(\underline{r}) \; (\nabla \log n(\underline{r}))^2 \; . \tag{78}$$

Chang and Cohen (1973) argue that when a physically reasonable density is chosen, this term will not be divergent. However,

it is not clear whether the long-range correlation
ϕ_p in the ground state gives such a physically reasonable
density.

To sum up - we have shown that good results for the
energy, tension and thickness of the surface of liquid
helium   are obtained with the wavefunction $\prod f(r_{ij})$
provided r^{-2} correlations due to phonon zero-point motion
are left out. When the phonon correlations are included
serious problems arise that may only be overcome by using
the method of Chang and Cohen and the wavefunction $\prod f(r_{ij}) \prod s(r_i)$.
The question of whether it is the wavefunction $\prod f(r_{ij})$ or the
phonon correlation factor that is at fault remains unanswered
however, since the exact density profiles that follow
from the suggested wavefunctions are not yet known.

We are grateful to Professor Alan Portis for several
stimulating discussions on the problems arising out of
the zero-point motion of the phonons.

FIGURE CAPTIONS

Fig 1.   The functions f and δf defining the density
            profile.

Fig 2.   Integrands of the exponential density expressions
            for ϵ and ϵ_∞ at $\lambda = d$, together with the integrand
            for the limiting expression ϵ_o. We took d=2.6Å
            and v=10.3K. The areas under the curves times
            $2\pi n_o^2 d^4 v \left(= 1.431 \; K/Å^2 \text{ or } 1.975 \, erg/cm^2 \right)$
            give the surface energies.

Fig 3.   Surface energy and surface tension for the
            exponential density profile. (ϵ and ϵ_∞ from
            eqns. (24) and (28), σ and σ_∞ from eqns.
            (44) and (45)).

Fig 4.   Determination of the surface thickness from
            eqn. (34) and the results of Fig. 3.

Fig 5.   The surface tension integrands, plotted in the
            same way as the energy integrands of Fig 2. The
            areas under the curves times $2\pi n_o^2 d^4 v$
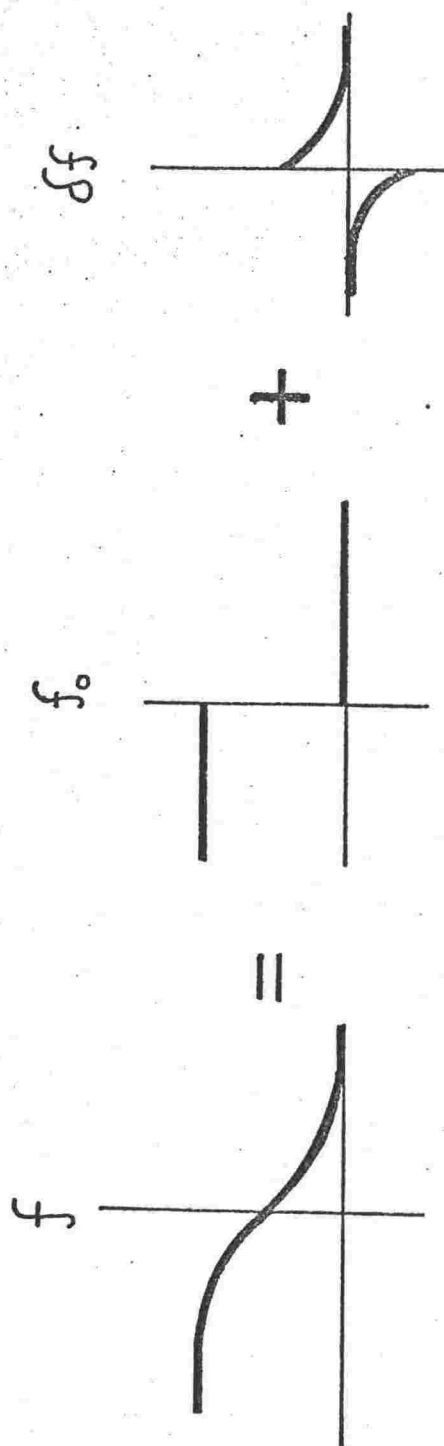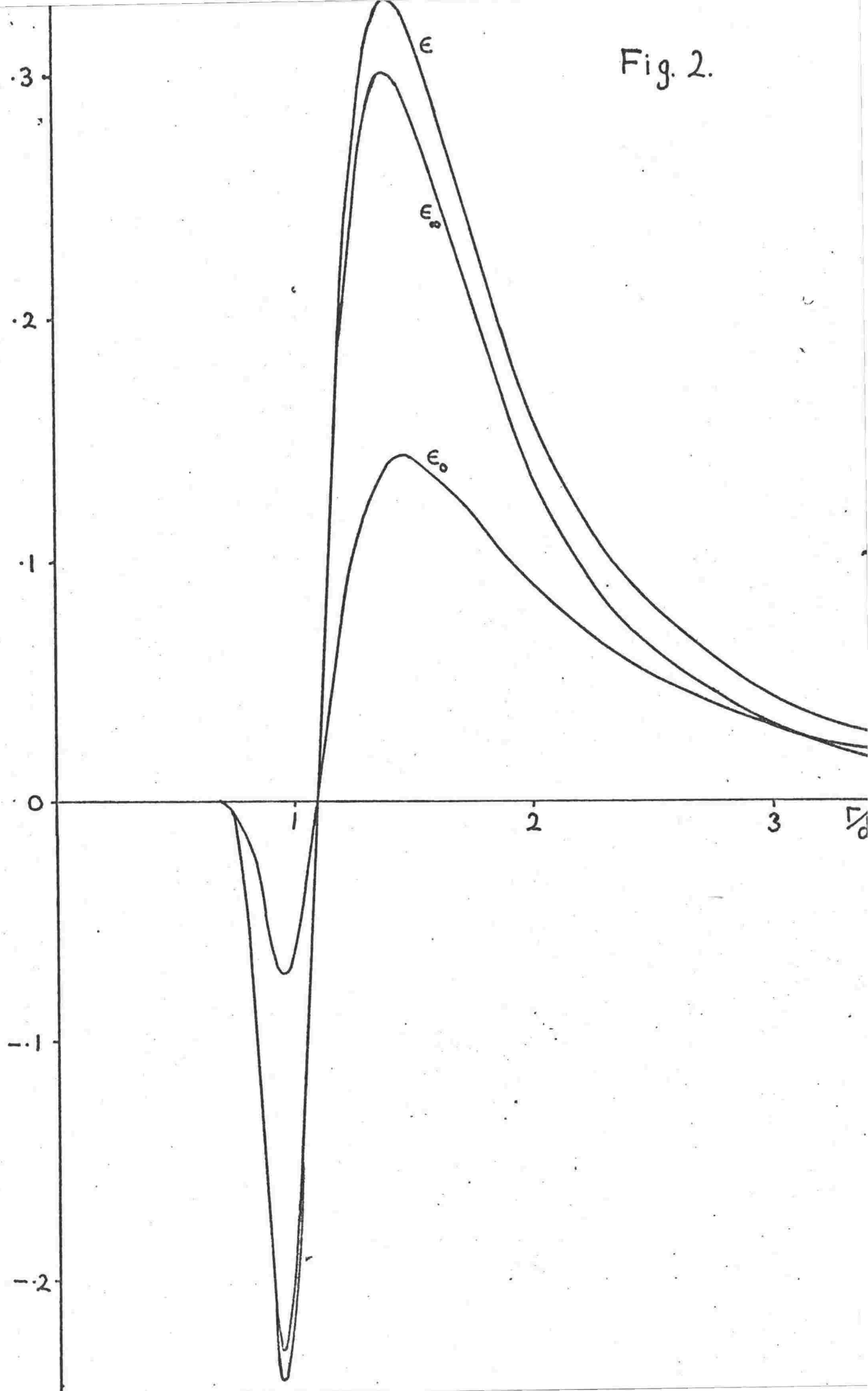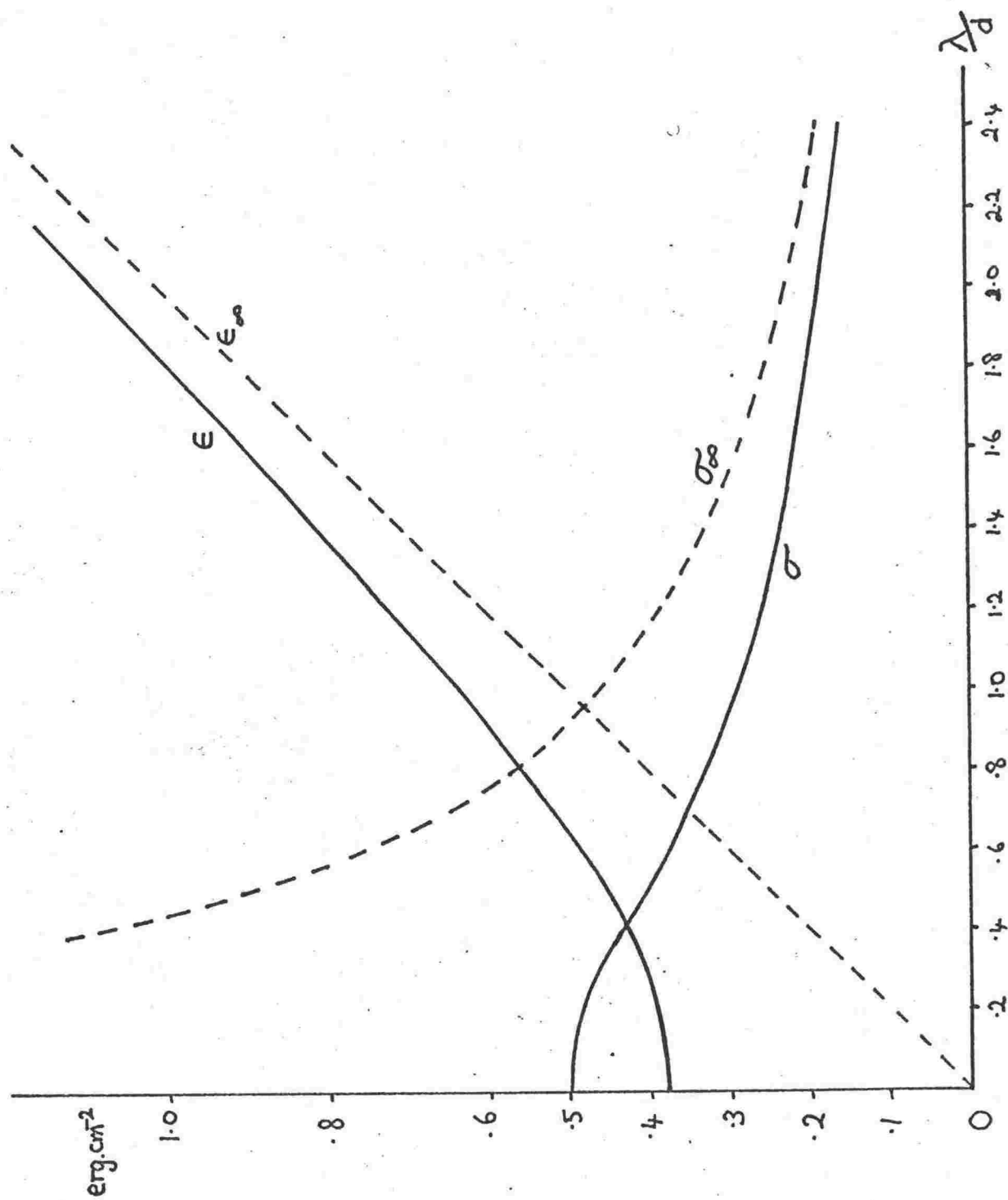   give the surface tensions.
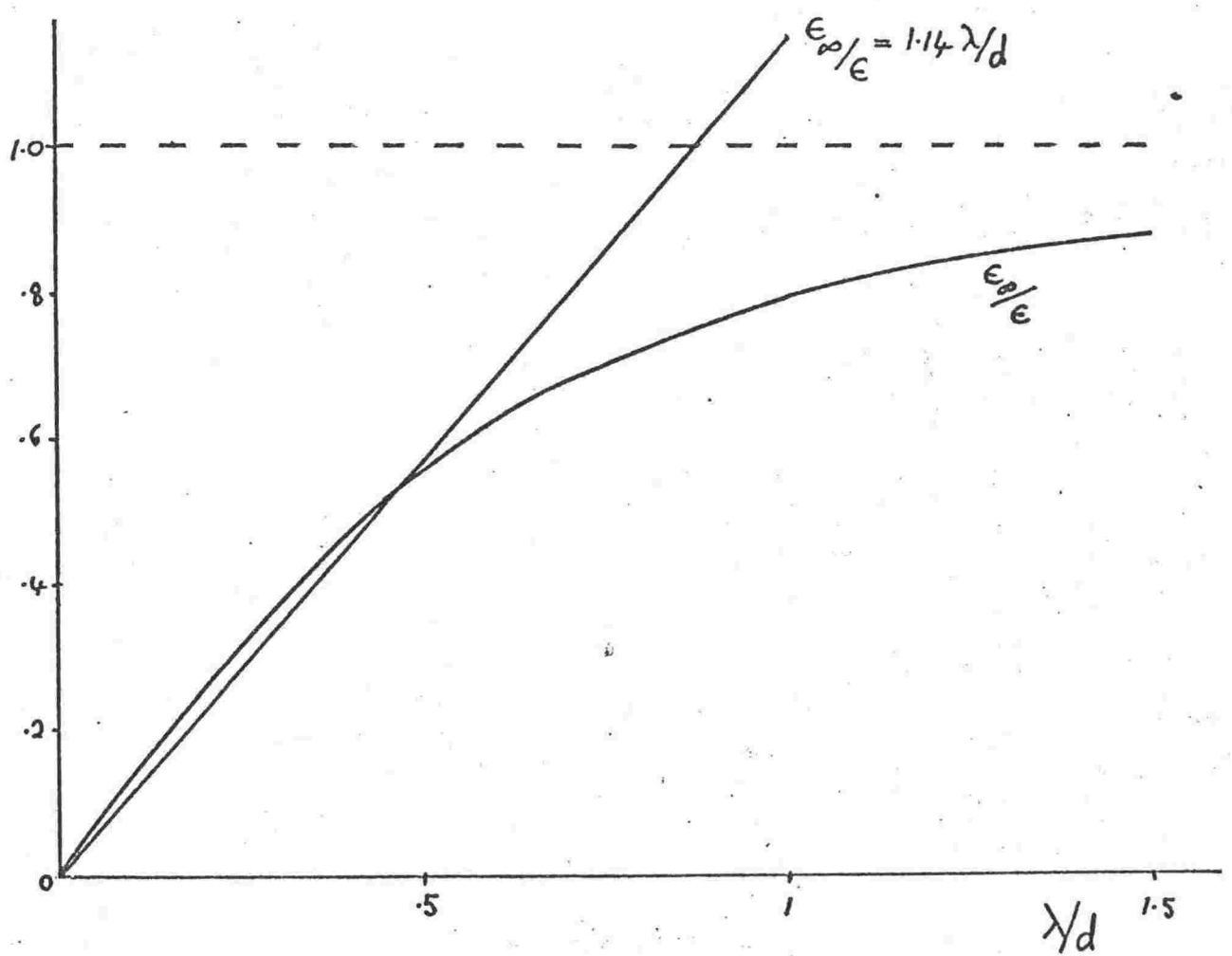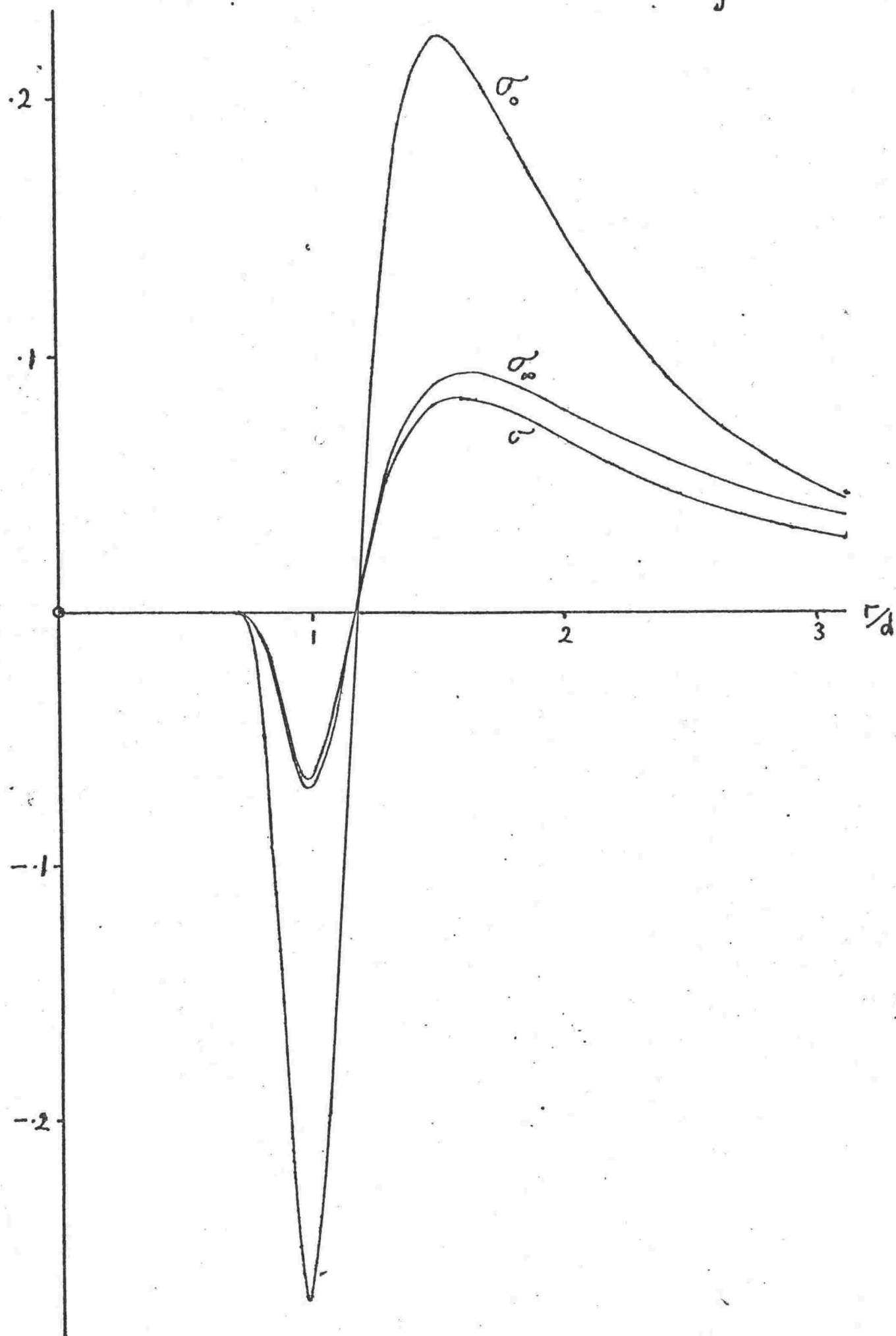
Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4.

Fig. 5.

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