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CALCULATIONS OF THE GROUND STATE ENERGY
AND EXCHANGE INTEGRALS OF CRYSTALLINE ³HE

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Chapter I. Introduction and Summary

In the past decade there has been considerable interest in Quantum Crystals.¹⁻¹⁰ From a theoretical point of view, the differences between a quantum crystal and its classical counterpart are mainly the large zero-point motion of the particles and the fact that quantum statistics are important for such a system. In the past, the only known quantum crystals were the solid phases of ^3He and ^4He , which can exist only under pressure at a low temperature. Recent interest has, however, shifted to nuclear matter and neutron stars, which were predicted to exist as quantum crystals by several authors.¹¹ In the case of helium, the quantum effect of the zero-point motion arises because of the weak attractive potential coupled with light mass. Furthermore, the kinetic energy of the system is comparable to its potential energy and that makes theoretical calculation quite complicated. The large zero-point motion (about 30% of the nearest neighbour distance in ^3He), also results in the failure of the traditional lattice dynamical theory¹⁰ which uses the small-oscillation expansion. Also, because of this large zero point motion, the strong, short-range repulsion between two atoms becomes important so that correlations between particles cannot be neglected. This results in the failure of variational calculations using only single-particle wave functions, e.g., the Hartree type of calculations.¹ A successful treatment, therefore, has to start with quantum mechanics, and should be able to account

for not only the elementary excitations of the solid, which is a property of all crystals, but also the effects due to the short-range repulsion. Progress has been made in using systematic perturbation theory,⁵ self-consistent phonon theory,⁷ t-matrix calculations,⁹ and cluster expansion calculations^{2,3,6} in computing the ground-state energy of solid helium and its various properties. Our purpose is to formulate a scheme, using the variational principle, with a trial wave function that has the required properties to adequately describe the quantum crystal problem, namely to be able to account for the short-range repulsion. Such a scheme offers several advantages: 1) Since it is a variational calculation, the wave function produced is optimal and we also obtain an upper bound to the ground state energy. 2) Since a variational calculation always produces a wave function of the system, the result of the calculation can be checked by Monte-Carlo methods which means that the approximations that we have made in the calculation can be checked. 3) If we have obtained a good ground state wave function, it can be used as a starting basis for perturbation calculation. Such a method has been devised by Feenberg and is called a correlated basis function type of calculation.¹²

In Chapter II we shall derive and discuss the variational equation using Jastrow-type wave functions, which are a product of pair wave functions, as our trial wave function. The logic in choosing such a trial wave function is that it is the simplest wave function that can describe the correlation effect, i.e.,

that the wave function should vanish if any two particles are close to one another. In our calculation, we shall ignore statistics since, for ^3He , the exchange energy is three orders of magnitude smaller than the ground state energy. We shall, therefore, treat statistics in an approximate way after the ground-state energy is computed by first ignoring statistics.¹³

The variational equation is non-linear. However, it can be simplified by writing it in terms of two, three and four particle distribution functions. In such a form, the relationship between the wave functions and the two-particle distribution function is provided by the BBGKY equation. We shall also discuss the various approximations used in simplifying the variational equations so as to obtain a form that is possible to solve numerically. This involves essentially truncating the equation so that the three and four particle distribution function can be approximated by some form of the two particle distribution function. In actual numerical calculations, we separate both the wave function and the two particle distribution function into two parts, one part involves the phonon (or localization) and the other part takes care of the short-range correlations. Such a separation is essential in the calculation since the variational equation probably does not have unique solutions as it can describe both the liquid phase and the solid phase as well. In doing the separation, we are looking for a solid phase solution. Furthermore, in our numerical calculation, we are always very careful so that the localization part of the

wave function is treated exactly and we only approximate on the short-range correlation part.

In Chapter III, we discuss the numerical approximations and simplifications and also present the results. It is obvious from the results that the approximations have destroyed the variational principle since at high density the calculated ground-state energy is lower than experimental values. We wish to emphasize, however, that the motivation is to demonstrate the feasibility of such a calculation and furthermore, we are able to obtain an approximate wave function for the system which can be used in a Monte-Carlo calculation for the ground-state energy and for the exchange integrals in solid ^3He .

In Chapter IV we describe a Monte Carlo calculation using the resulting wave function of Chapter III. The Monte Carlo calculation is indeed exact and produces an upper bound which compares favorably with all previous calculations. This indicates that the wave functions we obtained are really not too bad. We also describe a Monte Carlo-Variational calculation combination in which the short range correlation function is obtained from a differential equation (the approximate variational equation) and the Gaussian wave function is determined variationally from Monte Carlo.

Chapter V deals mainly with exchange in ^3He . The exchange interaction in solid ^3He , though small compared with the ground-state energy, has a pronounced effect on the low temperature thermodynamics and nuclear magnetic resonance properties of solid ^3He . It has received a considerable amount of theoretical and

experimental attention over the past ten years.¹⁴ In particular, it has been pointed out by McMahan¹³ that exchange is a many-body effect, and that in order to obtain a correct estimate of the exchange energy, one has to calculate the full many-body exchange integral. Secondly, Zane¹⁵ has suggested that the apparently anomalous results of Kirk and Adams¹⁶ might be explained by the presence of significant triple exchange. Our wave function, in particular, the one that is obtained by Monte Carlo calculation, offers an excellent means of testing these ideas. We, therefore, computed by Monte-Carlo methods, the nearest-neighbour, next-nearest neighbour and the only important triple exchange the 112-triple exchange integral. We find the value of J_1 to be roughly equal to the experimental value at the two densities that we performed our calculation. Further, we find the ratio of $|J_{112}|/|J_1|$ to range from $\sim 1/28$ at $R = 3.65 \text{ \AA}$ to $\sim 1/54$ at $R = 3.45 \text{ \AA}$, and that the value of $|J_2|$ and $|J_{112}|$ are about equal. An extrapolation of these points to the density at which the Kirk-Adams experiment was performed yields an estimate of the ratio $|J_{112}|/|J_1|$ of about $1/20$ which is in relatively good agreement with the value of $1/15$ estimated to be that which is necessary to account for the Kirk-Adams results in terms of triple exchange.

However, due to the many approximations involved in these calculations, it is not clear how accurately the exchange integrals are determined. An estimate¹⁷ of the Néel temperature of solid ^3He using the data of Adams et.al.¹⁸ and a nearest neighbor Heisenberg antiferromagnet model leads to a value of about 2.0 mK . Adams and

Nosanow¹⁹ have estimated the effect of triple exchange on the Néel temperature and found that it raises T_N to about 2.5 mK. Recently, Lee, Richardson and coworkers²⁰ at Cornell have made new measurements which indicate that the transition occurs at about 1.2 mK, which is in the wrong direction for triple exchange. Thus, although our calculations indicate that triple exchange is likely to be important in crystalline ^3He , the full understanding of the magnetic properties of crystalline ^3He must still be considered as an open problem.

Chapter II. Ground State Energy of Quantum Crystals.

For Quantum crystals, because of their large zero point energy, the motion of the particles will be strongly influenced by the large short-range repulsion in the interaction and must, therefore, be carefully treated in any calculation. For a variational calculation, one therefore has to build into the trial wave functions this correlation effect. The simplest type of wave function that can take care of this effect is the general Jastrow-type wave function, which is a product of factors, each of which depends on the relative coordinate of two particles and may be written

$$\psi(1 \dots N) = \prod_{i,j} g_{ij}(ij), \quad (2.1)$$

where the subscript indicates the lattice sites and i and j indicate the positions of particles i and j respectively. The best $\psi(1 \dots N)$ of the form (2.1) for a system is determined by the variational equation $\delta E / \delta g_{ij}(ij) = 0$, where E is the expectation value of the Hamiltonian using the wave function (2.1). For a crystal $g_{ij}(ij)$ would be different for different lattice site i and j . In particular, for a generalized harmonic oscillator problem,⁷ the exact wave function is in the form $\exp(-\frac{1}{2} \sum_{ij} \underline{u}_i \cdot \underline{G}_{ij} \cdot \underline{u}_j)$ where $\underline{u}_i = \underline{r}_i - \underline{R}_i$ and $\underline{G}$ is a $3N \times 3N$ matrix. The $\underline{u}_i$'s are related to the normal

coordinate by a unitary transformation. The same unitary transformation when applied to the matrix G_{ij} produces the phonon frequencies. Since this wave function is translational invariant, it can be expressed in the Jastrow form (2.1) where

$$g_{ij}(ij) = \exp\left(-\frac{1}{4} \underline{u}_{ij} \cdot \underline{G}_{ij} \cdot \underline{u}_{ij}\right) \quad \text{and} \quad \underline{u}_{ij} = \underline{u}_i - \underline{u}_j.$$

Therefore, the generalized Jastrow form is sufficiently general to take care of both the short-range correlations and the phonon collective modes of the crystal.

For solid ^3He , the effect of statistics on the energy is down by three orders of magnitude and may therefore be neglected in a first calculation. The Hamiltonian of the system is given by

$$H = -\frac{\hbar^2}{2m} \sum_i \nabla_i^2 + \sum_{i,j}' v(ij), \quad (2.2)$$

where the pair interaction is assumed to be symmetric; i.e. $v(ij) = v(ji)$ and is usually taken to depend only on $|\underline{r}_i - \underline{r}_j|$. In particular, the Lennard-Jones 6-12 potential is often taken for helium; i.e.,

$$v(ij) = 4 \epsilon \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^6 \right], \quad (2.3)$$

where $\epsilon = 10.22^\circ\text{K}$ and $\sigma = 2.556 \text{ \AA}^{0.1}$.

To obtain the best $g_{ij}(ij)$ one does a variation

$$\delta \langle \psi | H | \psi \rangle / \delta g_{ij}(ij) = 0$$

which produces an integro-differential equation for $g_{ij}(ij)$

$$\begin{aligned} & -\frac{\hbar^2}{2m} [(v_1^2 + v_2^2) g_n(12)] \int \psi_n^2(1 \dots n) d\tau_3 \dots d\tau_n - g_n(12) \frac{\hbar^2}{2m} (v_1^2 + v_2^2) \times \\ & \int \psi_n^2(1 \dots n) d\tau_3 \dots d\tau_n - \frac{\hbar^2}{2m} v_1 g_n(11) \cdot v_1 \int \psi_n^2(1 \dots n) d\tau_3 \dots d\tau_n - \\ & \frac{\hbar^2}{2m} v_2 g_n(11) \cdot v_2 \int \psi_n^2(1 \dots n) d\tau_3 \dots d\tau_n + v(12) g_n(12) \int \psi_n^2(1 \dots n) d\tau_3 \dots d\tau_n \\ & + g_n(12) \int \psi_n^2(1 \dots n) \sum_{\substack{i,j \\ i \neq n}}' \tilde{v}_{ij}(ij) d\tau_3 \dots d\tau_n - E g_n(12) = 0, \end{aligned} \quad (2.4)$$

where

$$\tilde{v}_{ij}(ij) = v(ij) + \frac{1}{2} (t_i + t_j) \ln g_{ij}(ij),$$

$$\psi_n(1 \dots n) = \psi(1 \dots n) / g_n(12),$$

$$t_i = -\frac{\hbar^2}{2m} v_i^2.$$

Equation (2.4) is perfectly general and can be applied to any N-body system.

In the form written, (2.4) is a non-linear integro-differential

equation. It can be simplified (at least in form) if we express it in terms of two-three- and four-particle distribution functions defined as

$$F_{1\dots n}(1\dots n) \equiv \int |\psi|^2 d\tilde{r}_{n+1} \dots d\tilde{r}_N \quad (2.5)$$

After some manipulation, equation (2.4) becomes

$$\begin{aligned} \frac{1}{4}(t_1 + t_2) F_{12}(12) + \tilde{v}_{12}(12) F_{12}(12) + \\ \sum_{3 \neq 1,2} \int F_{123}(123) [\tilde{v}_{13}(13) + \tilde{v}_{23}(23)] d\tilde{r}_3 + \\ \sum_{3,4 \neq 1,2} \int F_{1234}(1234) \tilde{v}_{34}(34) d\tilde{r}_3 d\tilde{r}_4 = E F_{12}(12) \end{aligned} \quad (2.6)$$

Equations similar to (2.4) and (2.6) have been derived by Lee and Broyles²¹ for liquid helium and by Leckner²² and Werthamer²³ for a general N-particle system.

It can be shown that equations (2.6) and (2.4) can be obtained by integrating $\psi H \psi = E |\psi|^2$ over all coordinates except 1 and 2. If instead we integrate over all coordinates except the first n, we can obtain an equation for $F_{1\dots n}(1\dots n)$ similar to (2.6), i.e.,

$$\begin{aligned}
& \frac{1}{4} (t_1 + \dots + t_n) F_{1\dots n}(1\dots n) + \sum_{i,j}^n \tilde{v}_{ij}(ij) F_{1\dots n}(1\dots n) + \\
& \sum_{j \neq 1\dots n}^N \int F_{1\dots nj}(1\dots nj) [\tilde{v}_{ij}(ij) + \dots + \tilde{v}_{nj}(nj)] d\mathbf{r}_j + \\
& \sum_{\substack{1 \leq j \\ i,j \neq 1\dots n}}^N \int F_{1\dots nj}(1\dots nj) \tilde{v}_{ij}(ij) d\mathbf{r}_i d\mathbf{r}_j = E F_{1\dots n}(1\dots n) \quad (2.7)
\end{aligned}$$

We see, therefore, that there is a whole chain of equations that relate all the $F_{1\dots n}(1\dots n)$ and to solve for them we have an infinite system of coupled equations which must be solved self-consistently. For practical reasons, we will truncate (2.6) by introducing approximations for $F_{123}(123)$ and $F_{1234}(1234)$.

A close examination of equation (2.6) indicates that the first three terms are of order one and the last two of order N . Since the energy E is a constant of order N , the term $\sum_{j \neq 1\dots n}^N \int F_{1234}(1234) \tilde{v}_{jk}(jk) d\mathbf{r}_3 d\mathbf{r}_4$ must also contain a large constant term which equals E , and in general another term of order one. This fact can be most easily seen by studying the harmonic problem,⁷ which can be solved exactly. When one evaluates the four-particle term in this case, one sees that it is equal to a constant $\times F_{12}(12)$ plus a general two particle harmonic term $\times F_{12}(12)$. Equation (2.6) can also be regarded as a natural extension of the single-particle Hartree equation to a two-particle case. For, if we write the single-particle Hartree equation in the form of distribution functions

similar to equation (2.6) we obtain a similar equation; i.e.,

$$\begin{aligned} \frac{1}{4} t_1 F_1(1) + \sum_{2 \neq 1}^N \int F_{12}(12) \tilde{v}_{12}(12) d\tilde{r}_2 + \\ \sum'_{\substack{23 \neq 1 \\ 2 < 3}}^N \int F_{123}(123) d\tilde{r}_2 d\tilde{r}_3 \tilde{v}_{23}(23) = E F_1(1) \end{aligned} \quad (2.8)$$

The first two terms are of order one and the last two of order N .

The nice feature of the single-particle Hartree equation is that in

this case $\sum'_{23 \neq 1, 2 < 3}^N \int F_{123}(123) \tilde{v}_{23}(23) d\tilde{r}_2 d\tilde{r}_3$ is exactly equal to a constant $\times F_1(1)$ so that we have exactly

$$\frac{1}{4} t_1 F_1(1) + \sum_{2 \neq 1}^N \int F_{12}(12) \tilde{v}_{12}(12) d\tilde{r}_2 = E_1 F_1(1). \quad (2.9)$$

By analogy, one wishes to consider the following approximation for the two-particle Hartree case:

$$\sum'_{\substack{34 \neq 12 \\ 34 \neq 12}}^N \int F_{1234}(1234) \tilde{v}_{34}(34) d\tilde{r}_3 d\tilde{r}_4 \approx \text{constant} \times F_{12}(12). \quad (2.10)$$

This approximation is analyzed in Appendix I. It is exact if a

single-particle wave function is assumed if the approximation $F_{1234}(1234) \simeq F_{12}(12) F_{34}(34)$ is made, if the superposition approximation is made for $F_{1234}(1234)$ in the case of liquid helium²¹ or if one approximates

$$F_{1234}(1234) \simeq \phi_{1234}(1234) G_{12}(12) G_{34}(34)$$

with

$$\phi_{1234}(1234) = \gamma e^{\frac{-A}{4}(u_{12}^2 + u_{13}^2 + u_{14}^2 + u_{23}^2 + u_{24}^2 + u_{34}^2)},$$

$$u_{12} = (x_1 - x_2) - (R_1 - R_2)$$

Therefore, the first major approximation that we make is the truncation of the four-particle term; i.e.,

$$\left\{ E - \sum_{34 \neq 12} \left[\frac{F_{1234}(1234)}{F_{12}(12)} \tilde{U}_{34}(34) \right] \right\} F_{12}(12) \simeq \epsilon_{12} F_{12}(12) \quad (2.11)$$

With (2.11), equation (2.7) is reduced to a two-body equation

$$\begin{aligned} \frac{1}{4} (t_1 + t_2) F_{12}(12) + \tilde{U}_{12}(12) F_{12}(12) + \sum_{34 \neq 12} \left\{ F_{123}(123) [\tilde{U}_{13}(13) + \tilde{U}_{23}(23)] \right\} \\ = \epsilon_{12} F_{12}(12) \end{aligned} \quad (2.12)$$

This equation is still quite general for an N-body problem with a two-body potential. Both the liquid phase and solid phase can be described by such an equation. In order to find a solution for the solid phase, let us define

$$F_{1,\dots,n}(1,\dots,n) \equiv \phi_{1,\dots,n}(1,\dots,n) G_{1,\dots,n}(1,\dots,n), \quad (2.13)$$

where in this factorization, it is intended that the $\phi_{1,\dots,n}(1,\dots,n)$ describe the localization and $G_{1,\dots,n}(1,\dots,n)$ the short-range correlations. If we examine equation (2.12), we see that localization will arise mainly from the three-particle terms which describe the effective potential well imposed on the two particles (1 and 2) by all the rest of the particles. for $F_{12}(12)$, we can therefore define localization as the part of the two particle distribution function that is produced by this effective potential well, i.e.,

$$\begin{aligned} \frac{1}{4}(t_1 + t_2) \phi_{12}(12) + \sum_{j \neq 12} \int \frac{F_{123}(123)}{F_{12}(12)} [\tilde{v}_{13}(13) + \tilde{v}_{23}(23)] d\mathbf{r}_3 \phi_{12}(12) \\ = \varepsilon_0 \phi_{12}(12) \end{aligned} \quad (2.14)$$

If this is so, then we can substitute (2.14) into (2.12) and obtain a differential equation for $G_{12}(12)$

$$\left\{ \frac{1}{4} (\epsilon_1 + \epsilon_2) + \tilde{v}_{12}(12) - \frac{\hbar^2}{4m} \sum_{i=1}^2 \ln \Phi_{12}(12) \cdot \sum_{i=1}^2 \ln G_{12}(12) - \right.$$

$$\left. \frac{\hbar^2}{4m} \sum_{i=1}^2 \ln \Phi_{12}(12) \cdot \sum_{i=2}^2 \ln G_{12}(12) \right\} G_{12}(12) = (\epsilon_{12} - \epsilon_0) G_{12}(12)$$

(2.15)

There are still two unknown functions we have to determine before we can solve for $F_{12}(12)$, i.e., $g_{ij}(ij)$ and $F_{123}(123)$. The general function $g_{ij}(ij)$ can be similarly factored into a localized part and a short-range correlation part

$$g_{ij}(ij) \equiv \varphi_{ij}(ij) f_{ij}(ij),$$

$$\psi = \phi F,$$

$$\phi = \prod_{i,j}^n \varphi_{ij}(ij),$$

$$F = \prod_{i,j}^n f_{ij}(ij). \quad (2.16)$$

Further we define

$$\begin{aligned} \Phi_{1 \dots n}(1 \dots n) &\equiv \int \Phi^2(1 \dots n) \, d\mathbf{r}_{n+1} \dots d\mathbf{r}_N, \\ &\equiv \int \prod_{i,j}^n \varphi_{ij}^2(ij) \, d\mathbf{r}_{n+1} \dots d\mathbf{r}_N. \end{aligned}$$

(2.17)

Eventually, it is our aim to take ϕ as a generalized Gaussian wave function $\exp(-\frac{1}{2} \sum_{ij} G_{ij} \cdot \underline{r}_i \cdot \underline{r}_j)$ or, in simplified cases, just the product of single particle gaussians. The relationship between $F_{12}(12)$ and $g_{ij}(ij)$ can be taken from equation (2.5). However, it is more convenient to use the BBGKY equation

$$\nabla_1 F_{12}(12) = \nabla_1 \int \psi^2 d\underline{r}_3 \dots d\underline{r}_N, \quad (2.18)$$

or after some simple manipulations:

$$\begin{aligned} \nabla_1 \ln F_{12}(12) &= 2 \nabla_1 \ln g_{12}(12) + \\ &+ 2 \sum_{3 \neq 1} \int \frac{F_{123}(123)}{F_{12}(12)} \nabla_1 \ln g_{13}(13) d\underline{r}_3. \end{aligned} \quad (2.19)$$

There are two reasons for the introduction of these definitions. First of all, we are looking for a solution which describes a solid so that the functions would therefore have to describe both the elementary excitations and the short-range repulsion. Secondly these definitions also will provide a scheme which is amenable to practical computation. In addition, these definitions enable us to treat the localized part of the distribution function (and the related wave function) more exactly. This is possible because for a generalized gaussian (or for single-particle gaussians), Eq. (2.17) can be solved exactly therefore ensuring that the ϕ

that relates ψ , $F_{12}(12)$, $F_{123}(123)$ and $F_{1234}(1234)$ is treated exactly. For $F_{123}(123)$ we will write

$$\begin{aligned}
 F_{123}(123) &\equiv \Phi_{123}(123) G_{123}(123) \\
 &\approx \Phi_{123}(123) G_{12}(12) G_{13}(13) G_{23}(23),
 \end{aligned}
 \tag{2.20}$$

where only the short-range correlation part is approximated by the superposition approximation.

Chapter III. Numerical Computation and Results.

Numerically, we have to solve equations (2.14), (2.15) and (2.19) self consistently. Without further simplifications, however, they are still too complicated to solve. As a first approach we will make enough approximations to make numerical calculation feasible. Then, in Chapter IV, we can use these results as a basis for Monte-Carlo type calculations.

Firstly, it is convenient to work with a ψ such that $\phi = \prod_i \phi_i(i)$. Then,

$$\begin{aligned}\phi_{1\dots n}(1\dots n) &= \int \prod_i^N \phi_i^2(i) d\mathbf{r}_{n+1} \dots d\mathbf{r}_N \\ &= \phi_1(r) \dots \phi_n(n)\end{aligned}\quad (3.1)$$

The BBGKY equation becomes

$$\begin{aligned}\nabla_1 \ln G_{12}(12) &= 2 \nabla_1 \ln f_2(12) + \\ &2 \sum_{3 \neq n} \int \phi_3^2(3) \phi_{13}^2(13) G_{23}(23) \nabla_1 \ln f_3(13) d\mathbf{r}_3\end{aligned}\quad (3.2)$$

This is a rather complicated integro-differential equation for $f_{12}(12)$. Because of severe computational difficulties we have been unable to solve (3.2) as it stands. A possible approach is suggested in Appendix II. For this reason we have studied the

problem by looking at the simplest approximation to (3.2); i.e., keeping only the leading term

$$\nabla_i \ln G_{12}(12) \simeq 2 \nabla_i \ln f_{12}(12), \quad (3.3)$$

which implies that $G_{12}(12) \simeq f_{12}^2(12)$. There are two justifications for this approximation. In the first place, we are only approximating the short-range correlation function, whereas the localized part which is the gaussian, needs no approximation. Secondly, in doing so, we have arrived at a differential equation for the short range correlation function which has been independently arrived at by a number of workers^{5,6,8} from very different approaches. Such a correlation function should thus be adequate for a first-order calculation.

The single-particle wavefunction $\varphi_i(i)$ can be taken as gaussian (in general, no such assumption is necessary and we do it for numerical convenience)

$$\varphi_i(i) = \left(\frac{A}{\pi}\right)^{3/2} e^{-\frac{A}{2}(r_i - R_i)^2}. \quad (3.4)$$

This modifies the effective potential $\tilde{v}_{ij}(ij)$ slightly: i.e.,

$$\sum_{i,j} \tilde{v}_{ij}(ij) = \sum_{i,j} \left[v(ij) + \frac{1}{2}(t_i + t_j) \ln f_{ij}(ij) \right] + \frac{3N_A^2 A}{4m}, \quad (3.5)$$

where the constant term comes from $\frac{1}{2} \sum_i t_i \ln \phi_i(i)$.

Therefore, for practical purposes, we can regard

$$\tilde{v}_{ij}(ij) = v(ij) + \frac{1}{2} (t_i + t_j) \ln f_{ij}(ij), \quad (3.6)$$

since the constant term which arises from the Gaussians will only modify the eigenvalue $\mathcal{E}_{12}$ and will leave the eigenfunction unchanged.

With (3.3) and (3.6), equation (2.15) for $G_{12}(12)$ can be written in terms of $f_{12}(12)$ and A ; i.e.,

$$\left\{ t_1 + t_2 + v(12) - \frac{\hbar^2 A}{2m} \frac{d}{dr_1} (r_1 - R_1)^2 \cdot \nabla_1 \ln f_{12}(12) - \right. \\ \left. \frac{\hbar^2 A}{2m} \frac{d}{dr_2} (r_2 - R_2)^2 \cdot \nabla_2 \ln f_{12}(12) \right\} f_{12}(12) = \mathcal{E}'_{12} f_{12}(12), \quad (3.7)$$

and for a spherically symmetric f -function

$$\left\{ -\frac{\hbar^2}{m} \frac{1}{r^2} \frac{d}{dr} r^2 \frac{d}{dr} + v(r) + \frac{\hbar^2 A}{m} (r - R_\alpha) \frac{d}{dr} \right\} f_\alpha(r) \\ = \mathcal{E}_\alpha f_\alpha(r), \quad (3.8)$$

where the subscript α denotes the neighbour number between the two lattice sites 1 and 2; for different neighbour pairs,

$f_d(r)$ would be different. Equation (3.8) is the same equation that other workers have obtained using different approaches.^{5,6,8} We therefore have confidence that such a short-range correlation function would give reasonable answers. The effective potential term $\tilde{\omega}(12)$ in equation (2.14) is:

$$\begin{aligned}\tilde{\omega}(12) &= \sum_{3 \neq 1,2} \int \frac{F_{123}(123)}{F_{12}(12)} [\tilde{v}_{13}(13) + \tilde{v}_{23}(23)] d\mathbf{r}_3 \\ &\approx \tilde{\omega}(1) + \tilde{\omega}(2),\end{aligned}\quad (3.9)$$

where we have approximated

$$\tilde{\omega}(1) \approx \sum_{3 \neq 1,2} \int \phi_1(3) G_{13}(13) \tilde{v}_{13}(13) d\mathbf{r}_3; \quad (3.10)$$

i.e., we approximate $G_{23}(23) \equiv 1$ in the expressions for $\tilde{\omega}(1)$ and $\tilde{\omega}(2)$. The validity of (3.10) still needs to be investigated. A similar approximation was discussed by Hetherington, Mullin and Nosanow in their cluster expansion calculation.³ They found that, in the three-body term, the integral $\langle f(\vec{r}_{12}) f(\vec{r}_{13}) f(\vec{r}_{23}) \rangle$ can be well approximated by $\langle f(\vec{r}_{12}) \rangle \langle f(\vec{r}_{13}) \rangle \langle f(\vec{r}_{23}) \rangle$. Here, $\langle \rangle$ denotes expectation value with respect to the gaussian

single particle wave functions, and $f(r)$ is the Nosanow correlation function. However, the integral $\langle \tilde{v}(1,2) f(\tilde{u}_2) f(\tilde{u}_3) f(\tilde{r}_1) \rangle$ cannot be satisfactorily approximated by replacing either $f(1-3)$ or $f(2-3)$ by unity. We believe that in making approximation (3.10) we are faced with the same types of difficulties. Moreover, the approximation would be worse at higher densities where the hard core effect is more important. Therefore, we expect (3.10) to break down at high density; whereas, it may be more satisfactory at lower densities.

Numerically, we symmetrize the effective potential $\tilde{\omega}(1)$ and $\tilde{\omega}(2)$ with respect to the coordinates $\underline{u}_1 = \underline{r}_1 - \underline{R}_1$ and $\underline{u}_2 = \underline{r}_2 - \underline{R}_2$ and the Gaussian A value is determined from the curvature at the minimum of the potential well $\tilde{\omega}(1)$ and $\tilde{\omega}(2)$. In principle, there is a different Gaussian A value for different pairs of neighbours. For simplicity, we have taken A from the potential well $\tilde{\omega}(1)$ when 2 is at infinity.

Equations (3.10) and (3.8) are then solved self-consistently. We found that a four neighbour approximation was adequate, i.e., we determined $f_\alpha(r)$ up to $\alpha = 4$, and took $f_\alpha(r) = f_4(r)$ for $\alpha \geq 4$.

The energy of the system is

$$E = \frac{\langle \psi | H | \psi \rangle}{\langle \psi | \psi \rangle}$$

$$= \frac{3\pi^2 A N}{4m} + \sum_{i,j}^N \frac{\int F_{ij}(ij) \tilde{v}_{ij}(ij) d\underline{u}_i d\underline{u}_j}{\int F_{ij}(ij) d\underline{u}_i d\underline{u}_j} \quad (3.11)$$

with $\tilde{v}_{ij}(ij)$ defined in (3.6) and $F_{ij}(ij)$ are the pair distribution functions for particles i and j .

We have done the calculations at two different densities: $R_1 = 3.45 \text{ \AA}$ and $R_1 = 3.65 \text{ \AA}$. The results are summarized in Tables I and Figures 1, 2, 3. Although there is qualitative agreement with experiment, the energy and its derivatives are not as good as those obtained previously. Still there is one striking feature; namely, that the gaussian A value obtained in this way is much smaller than that obtained by other workers. In particular, McMahan and Guyer have pointed out that the exchange integrals are particularly sensitive to the Gaussian A value.²⁵ Because this is such an important point, we have studied this question further by using these results in a Monte-Carlo calculation of the energy which is discussed in Chapter IV. Using the same correlation function, we find that the A value which minimizes the energy calculated via Monte-Carlo methods is about the same as the A value we found in this chapter. Thus, we may conclude that this approach is very useful in finding an appropriate trial wave function for use in a Monte-Carlo calculation.

Chapter IV. Monte Carlo Calculation of E_0 .

If the wave function of a system is known, then the Monte Carlo method is the best way to evaluate the pair distribution function or the ground state energy. Such a calculation is in principle exact and therefore for any wave function, by the variational theorem, would give an upper bound to the energy. In the past various workers²⁶ have performed Monte Carlo calculations using either the Nosanow wave function or the form $f(r) = \exp(-\frac{1}{2}(b/r)^5)$ with b as variational parameter and obtained good agreements with experiment. Since we have obtained approximate wave functions, a Monte Carlo calculation would give an upper bound to the energy for such wave functions. A comparison with our previous calculations of the ground-state energy and pair distribution function would provide further insight into our calculations.

At this point we would like to direct the reader's attention to a recent paper by McGee and Murphy²⁷ who have calculated the ground state energy and pair distribution function of liquid ^4He by using an $f(r)$ that is derived from the solution of the two-body zero energy S-wave reduced Schrodinger equation

$$-\frac{\hbar^2}{m} \frac{d^2 \omega}{dr^2} + V(r) \omega(r) = 0, \quad (4.1)$$

where $V(r)$ is the L-J potential. They have obtained excellent agreement with previous variational calculations and with experiment.

We believe our equation (2.5)

$$\left(-\frac{\hbar^2}{m} \nabla^2 + V(r) + \frac{\hbar^2 A}{m} (r - R_s) \frac{d}{dr}\right) f_s(r) = \epsilon'_s f_s(r) \quad (4.2)$$

for the solid short range correlation function, is the counterpart of equation (4.1) for the solid. The only difference is that we have an additional undetermined parameter A . Now, the wave function that we used is

$$\Phi = c \prod_i \varphi_i(i) \prod_{ij} f_{ij}(ij),$$

where

$$\varphi_i(i) = \exp \left[-\frac{A}{2} (i - R_s)^2 \right],$$

and $f_{ij}(ij)$ are the solutions to equation (4.2) for a given A value. There are two possible Monte Carlo approaches. In the first place, we can approach the problem as a Monte Carlo calculation with A as a variational parameter and $f_{ij}(ij)$ determined uniquely for each A value and relative lattice distance R_{ij} . Such an f is likely to be a good choice since, it is mainly determined by the strong short-range repulsive part of the interaction. The other approach is to use the Monte Carlo method as a check for our numerical results given in the previous chapter since for any A and f , the Monte Carlo will give an upper bound to the ground-state energy and a pair

distribution function as well.

The Monte Carlo integration technique has been fully described in the literature²⁸. Briefly, one would like to evaluate the integral

$$I = \int G(\underline{r}_1, \dots, \underline{r}_N) V(\underline{r}_1, \dots, \underline{r}_N) d\underline{r}_1 \dots d\underline{r}_N, \quad (4.3)$$

where the function G is now negative and normalized. One samples configurations of the system $R^{(i)} = (\underline{r}_1^{(i)}, \dots, \underline{r}_N^{(i)})$ according to the probability distribution $G(R^{(i)})$ and approximates the integral by

$$I \approx \frac{1}{n} \sum_{i=1}^n V(R^{(i)}) \quad (4.4)$$

where n is the number of such samples. In the limit of large n , (4.3) and (4.4) become equal. In fact one needs only to know G to within a multiplicative constant and the Monte Carlo integration will effectively use $G/\langle G \rangle$ as can be seen by setting $V = 1$ in equation (4.4).

The integral that we meant to evaluate by Monte Carlo technique is the ground state energy

$$E = \frac{3\hbar^2 AN}{4m} + \sum_{ij} \frac{\int \prod_i \phi_i^2 \prod_{ij} f_{ij}^2 \tilde{v}_{ij} d\underline{r}_1 \dots d\underline{r}_N}{\int \prod_i \phi_i^2 \prod_{ij} f_{ij}^2 d\underline{r}_1 \dots d\underline{r}_N} \quad (4.5)$$

Therefore, we take $G = \phi^2$ since ϕ^2 is non-negative and normalized, and

$$\tilde{v}_{ij}(ij) = v_{ij} + \frac{1}{2}(t_i + t_j) \ln f_{ij}(ij).$$

The Monte Carlo integration will also evaluate the pair distribution function. In fact, one first evaluates by Monte Carlo, all the various $F_{ij}(r_{ij})$ as a function of r_{ij} only, and the energy is simply obtained by integrating $F_{ij}(ij) \tilde{v}_{ij}(ij)$ over r_{ij} , i.e., the pair distribution function $G(r)$ is

$$G(r) = \frac{\sum_{j \neq i} F_{ij}(r_{ij})}{\int F_{ij}(r_{ij}) dr_{ij}}. \quad (4.6)$$

The Monte Carlo integration is performed for a system of 54 particles in bcc lattice, arranged in the shape of a cube and with periodic boundary conditions imposed. For simplicity, we only use a two shell approximation, i.e., only the nearest-neighbour and next-nearest neighbour f's are included. This is done because the other neighbours do not depend crucially on f. The Monte Carlo method is the biased random walk type described by McMillan.²⁹ We discard the first 12,000 configurations and average over the next 75,000 configurations.

The time taken on a CDC 6600 computer is about 15 minutes for each computation. We have computed the pair distribution function and ground state energy at two different densities ($R = 3.45 \text{ \AA}$ and $R = 3.65 \text{ \AA}$) using the numerical f and A of Chapter III. We have also done a variational calculation with A as a variational parameter and minimized ground-state energy with respect to A . In Figure 1 we see that the dependence of f on A and R is very weak. Especially in the region of importance ($0 - 4 \text{ \AA}$) the difference between the f of $R = 3.65 \text{ \AA}$, $A = 1.15$ and $R = 3.45 \text{ \AA}$, $A = 1.40$ is negligible. Therefore, in the variation of A , we can effectively keep f unchanged. The results are shown in Tables II, III and Figure 4 where we show the minimum E_0 as a function of A for $R = 3.45$ and $R = 3.65$, and we show the pair distribution function as compared with that calculated from Chapter III. We also show E_0 as a function of R , the nearest-neighbour distance and compare with experiment and other calculations. We note that the Monte Carlo result gives a better density dependence of the ground-state energy and the minimum A value is very close to that obtained from Chapter III. Furthermore, the Gaussian A value is consistently smaller than most other calculations. This, we believe, would give us a better agreement with experiment in the calculation of the exchange frequency in solid ^3He .

It is interesting to compare our Monte Carlo results with those of Hensen and Levesque²⁶ who use a correlation function that is equal to $\exp(-\frac{1}{2}(b/r)^n)$ with b and n as variational parameters. They have obtained a lower ground state energy than our calculation. However,

it is clear from Figure 2 that their calculation gives a poor density dependence at low density. We believe this poor density dependence is due mainly to a poor choice of correlation function. More recently, Hensen and Pollock³⁰ have done Monte Carlo calculations using correlated Gaussian wave functions and $\exp(-\frac{1}{2}(b/r)^5)$ as correlation function and found essentially no improvements over the previous calculation. Koehler has pointed out⁷ that a self-consistent phonon calculation would improve the ground state energy calculation by about 2°K over the calculation using single particle wave functions. This is substantiated by his calculation using the Nosanow correlation function and he obtained a 2°K lowering compared with the Nosanow single particle results. The discrepancy of the Hensen and Pollock result is again probably due to their correlation function which rises very slowly to unity as compared with the Nosanow correlation and our correlation function (see Figure 1). We believe that since our correlation function resembles the Nosanow function, the inclusion of correlated phonon wave function would improve our results. If that improvement is again roughly 2°K, we would obtain an excellent agreement with experiment. Another criticism of the Hansen and Levesque result is their unusually low Gaussian A value. Physically, this low A value is necessary to compensate for the large cut off by their correlation function. Furthermore, their wave function is unable to produce a reasonable exchange integral calculation for solid ³He.³¹ The

fact that the Hensen and Levesque wave function fails to produce a reasonable exchange integral is another indication that their wave function is not an adequate description of the true system. In conclusion, we believe we have found a reasonable approach to obtain a good ground state wave function for solid ^3He .

Chapter V. Monte Carlo Calculation of Exchange in ^3He .

One consequence of the large zero point motion in solid ^3He , is the existence of a relatively large exchange interaction.¹⁴ The resultant exchange energy is about three orders of magnitude larger than the nuclear spin magnetic dipole-dipole interaction energy, and about three orders of magnitude smaller than the ground state energy. This exchange interaction has a pronounced effect on the low temperature thermodynamics and nuclear magnetic resonance properties of solid ^3He . It has received a considerable amount of theoretical and experimental attention over the past ten years.¹⁴ Although the theoretical work has appeared reasonably successful in its agreement with experiment, several complications have become apparent recently. For one, it has been pointed out²⁵ that most of the calculation of the near neighbour pair exchange frequency obtain fortuitous agreement with experiment since they have neglected the many-body short range correlations which may reduce the result by as much as a factor of six. Secondly, Zane¹⁵ has suggested that the apparently anomalous measurements of Kirk and Adams¹⁶ who measured the excess pressure of solid ^3He in strong external magnetic field might be explained by the significant presence of triple exchange processes---which were neglected in previous theoretical calculations based purely on the near neighbour pair Heisenberg Hamiltonian. Zane¹⁵ and McMahan and Guyer²² have subsequently given treatments which include triple exchange. In summary, they have shown that the leading terms in the exchange Hamiltonian involve pair and triple

exchanges. The inclusion of triple exchange has the effect of modifying the Heisenberg Hamiltonian with near-neighbour and next-nearest-neighbour interaction such that the effect of triple exchange is to "renormalize" the two pair exchange parameters and causes the next-nearest-neighbour interaction to be ferromagnetic.

McMahan¹³ and more recently McMahan and Guyer²⁵ have formulated a theory of exchange which emphasizes that exchange in solid ^3He is a full many-body effect, and that any calculation of the exchange integral has to start with a full many-body wave function. For solid ^3He , the true Hamiltonian, which is spin independent, may be described by an effective Hamiltonian

$$H_{\text{eff}} = H_L + H_x, \quad (5.1)$$

where H_L is the Lattice Hamiltonian and describes phonon like excitation of the solid. It also forms a set of home-based functions $\{\phi_m\}$ for exchange calculations. And H_x is the exchange Hamiltonian. The true Hamiltonian H of the system is then diagonalized in the manifold $\{Q(\phi_m, \mathcal{E}_\alpha)\}$ where Q is the antisymmetrized and $\mathcal{E}_\alpha$ are a complete set of orthonormal spin functions for the N atoms. By expressing the antisymmetrizer in terms of permutation operators

$$Q = \frac{1}{N!} \sum_P (-1)^P P, \quad (5.2)$$

where $P = p^{(r)} p^{(\sigma)}$, r = coordinate variables,
 σ = spin variables. By using the approximate relation

$$\langle \phi_m | H | \phi_n \rangle = E_n \delta_{mn}, \quad (5.3)$$

and

$$\langle \phi_m | \phi_n \rangle = \delta_{mn}, \quad (5.4)$$

for the low lying states, the Hamiltonian can be replaced approximately by

$$\begin{aligned} H &= \sum_n |\phi_n\rangle E_n \langle \phi_n| + \sum_P' (-1)^P g_P P^{(\sigma)} \\ &= H_L + H_X \end{aligned} \quad (5.5)$$

The permutation P can be written in terms of pairs, triple, cyclic four particleetc. exchanges so that H_X can be expressed as

$$\begin{aligned} H_X &= -2 \sum_{i < j} g_{ij} \left(\frac{1}{4} + \vec{I}_i \cdot \vec{I}_j \right) + 2 \sum_{i < j < k} g_{ijk} \left(\frac{1}{4} + \vec{I}_i \cdot \vec{I}_j \right. \\ &\quad \left. + \vec{I}_i \cdot \vec{I}_k + \vec{I}_j \cdot \vec{I}_k \right) + \text{higher order terms} \end{aligned} \quad (5.6)$$

Further approximations lead to an expression for the exchange operator given by

$$\langle \phi_m | \mathcal{J}_P | \phi_n \rangle = \langle \phi_m | (H - \frac{1}{2}E_m - \frac{1}{2}E_n) \frac{1}{2}(P + P^{-1}) | \phi_n \rangle, \quad (5.7)$$

and the exchange frequency is

$$\mathcal{J}_P = \text{Tr}(\mathcal{S}_L \mathcal{J}_P). \quad (5.8)$$

In the lowest order approximation, if we retain only the two particle exchange terms $\mathcal{J}_{ij}$, H is reduced to the Heisenberg model. For low temperature, the temperature dependence of $\mathcal{J}_P$ is small and it is usually replaced by an exchange integral $\mathcal{J}_P$ which is the expectation value of $\mathcal{J}_P$ on phonon ground state; i.e.,

$$\begin{aligned} \mathcal{J}_P &= \langle \phi_0 | \mathcal{J}_P | \phi_0 \rangle \\ &= \langle \phi_0 | (H - E_0) \frac{1}{2}(P + P^{-1}) | \phi_0 \rangle, \end{aligned} \quad (5.9)$$

and

$$\mathcal{J}_P = \langle \phi_0 | (H - E_0) P | \phi_0 \rangle \quad (5.10)$$

for $\phi_0 = \text{real}$. It is obvious from the above expression that the net effect of the three particle exchange is to "renormalize" the pair exchanges. It turns out from actual numerical computation²⁵ that the only significant contribution to H_x comes from J_1 , J_2 and J_{112} , where J_1 is the near neighbour pair exchange, J_2 the next near neighbour pair exchange and J_{112} the triple exchange for a triangle of atoms containing two first neighbour sides and one second neighbour side. If we make the approximation

$$\begin{aligned} (H - E_0) \phi_0 &= 0 \quad \text{near side of } \Sigma, \\ (H - E_0) P\phi_0 &= 0 \quad \text{far side of } \Sigma, \end{aligned} \quad (5.11)$$

where Σ is the hypersurface in configuration space separating and midway between the regions where ϕ_0 and $P\phi_0$ are large, the exchange integral J_p can be expressed in terms of surface integral

$$J_p = \frac{-\hbar^2}{2m} \int_{\Sigma} d\vec{s} \cdot [\phi_0^* \nabla_{3N} P\phi_0 - (P\phi_0) \nabla_{3N} \phi_0^*], \quad (5.12)$$

where $d\vec{s}$ is the $3N-1$ dimensional surface element whose direction is perpendicular to Σ . ϕ_0 is the ground state phonon wave function and P the permutation operator, ∇_{3N} is the $3N$ -dimensional gradient. This is a many-body integral and can be evaluated by Monte Carlo method. In the actual numerical

computation ϕ_0 is taken to be the approximate ground state wave function

$$\begin{aligned}\phi_0 &= \phi F \\ \phi &= c \prod_i \exp \left\{ -\frac{A}{2} (x_i - R_i)^2 \right\} \\ F &= \prod_{i,j}^N f_{ij}(r_{ij})\end{aligned}\tag{5.4}$$

The $f_{ij}(r_{ij})$ are numerical functions taken from our previous calculation and which are different for different neighbours. In our calculation we take $f_{ij}(r_{ij})$ to be the first neighbour short range correlation function throughout. We estimated that in doing so, we might have introduced an error of about 25%. The numerical computation technique is a combination of cluster approximation and Monte Carlo technique. Successively larger cluster of particles are taken in the Monte Carlo calculation of J until the calculation converges. In most cases a cluster of 20 particles is sufficient to ensure convergence of the integrals. A comparison with the result of McMahan and Guyer show that the short range correlation, as compared to the Nosanow correlation function affects the results (for a given density and same Gaussian A value) of first-neighbour exchange by a factor of two. This shows clearly that short range correlation function is important in the calculation of exchange. We have also found that the results for J_1 is in good agreement with experiment. Furthermore, we find the ratio

$|J_{112}|/|J_1|$ to range from $\sim 1/28$ at $R = 3.65 \text{ \AA}$,
 $A = 1.0 \text{ \AA}$ to $\sim 1/54$ at $R = 3.45 \text{ \AA}$, $A = 1.4 \text{ \AA}$, and that the
 value of $|J_2|$ and $|J_{112}|$ are about equal. An extrapolation
 of these points to the density at which the Kirk-Adams experiment
 was performed ($R \approx 3.73 \text{ \AA}$) yields an estimate of this ratio of
 about $1/20$, which is in relatively good agreement with the value
 of $1/15$ estimated to be that which is necessary to account for
 the Kirk-Adams results in terms of triple exchange. We have
 clearly shown that the effect of triple exchange is significant.
 However, whether triple exchange can explain the full magnetic
 properties of crystalline ^3He is not clear. First of all, we have
 made numerous approximations in our calculations, and though the
 consistency of our results are encouraging, it is not clear in
 particular, how accurately the exchange integrals are determined.
 Secondly, the inclusion of triple exchange would modify the
 estimation of the Neel temperature of solid ^3He . An estimate¹⁷
 of the Neel temperature of solid ^3He using the data of Adams et.al.¹⁸
 and a nearest-neighbour Heisenberg antiferromagnet model leads to a
 value of about 2.0 mK . Adams and Mosanow¹⁹ have estimated the
 effect of triple exchange on the Neel temperature and found that
 it raises T_N to about 2.3 mK . More recently, Lee, Richardson and
 coworkers²⁰ at Cornell have made new measurements which indicate
 that the transition occurs at about 1.2 mK . It seems therefore,
 that triple exchange gives a wrong contribution to T_N . Thus,
 although our calculations indicate that triple exchange is likely
 to be important in crystalline ^3He , the full understanding of the

magnetic properties of crystalline ^3He must still be considered an open problem.

We have summarized our results in Figure 5 and in Tables IV and V.

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Appendix I. The Four-Particle Term.

The purpose of this appendix is to show that the four-particle term in the variational equation (2.6) reduces simply to a constant term times $F_{12}(12)$ if we make the appropriate approximations; i.e., we would like to investigate the validity of the approximation

$$\int F_{1234}(1234) \tilde{U}_{34}(34) d\vec{r}_3 d\vec{r}_4 \approx \text{constant} \times F_{12}(12). \quad (\text{I.1})$$

In the Hartree approximation (I.1) is certainly true, since the four-particle distribution function is simply a product of four single-particle distribution functions.

In general, we define

$$F_{1234}(1234) \equiv \phi_{1234}(1234) G_{1234}(1234), \quad (\text{I.2})$$

where ϕ_{1234} represents the localization; i.e.,

$$\phi_{1234}(1234) = \int \phi^2 d\vec{r}_5 \dots d\vec{r}_N, \quad (\text{I.3})$$

and

$$\phi_{12}(12) = \int \phi_{1234}(1234) d\vec{r}_3 d\vec{r}_4 \quad (\text{I.4})$$

Obviously in approximating the correlation term, we have to at least include a short-range correlations for the pair (3,4) so as to renormalize the hard core in $\tilde{v}_{34}(34)$.

Therefore, the simplest approximation for F_{1234} is

$$F_{1234}(1234) \simeq \phi_{1234}(1234) G_{12}(12) G_{34}(34). \quad (1.5)$$

With this approximation, it is again obvious that (I.1) is true in the Hartree approximation.

We will study next correlated Gaussians with a single A value. This is a most important case since such wave functions contain many correlations and it is not clear a priori how good (I.1) is for a correlated wave function. In this case

$$\phi_{12}(12) = \left(\frac{A}{2\pi}\right)^{3/2} e^{-\frac{A}{2} u_{12}^2}, \quad (1.6)$$

$$\phi_{1234}(1234) = \eta e^{-\frac{A}{4}(u_{12}^2 + u_{13}^2 + u_{14}^2 + u_{23}^2 + u_{24}^2 + u_{34}^2)} \quad (1.7)$$

where $u_{12} = r_{12} - R_{12} = u_1 - u_2$.

Then

$$\int F_{1234}(1234) \tilde{v}_{34}(34) d\vec{r}_3 d\vec{r}_4 \propto G_{12}(12) e^{-\frac{A}{2} u_{12}^2} \int e^{-\frac{A}{4}(u_{13}^2 + u_{14}^2 + u_{23}^2 + u_{24}^2 + u_{34}^2)} G_{34}(34) \tilde{v}_{34}(34) d\vec{r}_3 d\vec{r}_4 \quad (1.8)$$

Furthermore, in our calculation, we assume that $G_{34}(34)$ and $\tilde{V}_{34}(34)$ are both spherically symmetric and depend only on $|r_{34}|$. Let us make a change of variable for u_3 and u_4 ; i.e.,

$$u_3 = -u + \frac{1}{3}u_1 + \frac{1}{3}u_2 + \frac{1}{3}u_4, \quad (1.9)$$

$$u_4 = -v + \frac{1}{3}u_1 + \frac{1}{3}u_2, \quad (1.10)$$

i.e., the integrals over u_3 and u_4 becomes integrals over u and v . Then

$$u_{34} = u_3 - u_4 = -u + \frac{2}{3}v, \quad (1.11)$$

and thus

$$\begin{aligned} & \int F_{1234}(1234) \tilde{V}_{34}(34) du_3 du_4 \sim \\ & e^{-\frac{A}{2}u^2} G_n(12) \int e^{-\frac{3A}{2}v^2 - \frac{3A}{4}u^2} \tilde{G}_{34}(1-u+\frac{2}{3}v+R_{34}) \\ & \quad \cdot G_{34}(1u-\frac{2}{3}v+R_{34}) du dv \end{aligned}$$

$$\sim \text{constant} \cdot F_{12}(12). \quad (1.12)$$

In this approximation; therefore, (I.1) holds. Thus we believe that it is reasonable to conclude that (I.1) is a good first approximation to use in the variational equation.

Appendix II. The Solid BBGKY Equation.

In this section we will derive an expression for the solid BBGKY equation suitable for numerical computation.

The BBGKY equation is:

$$\begin{aligned} \nabla_1 \ln F_{12}(12) &= 2 \nabla_1 \ln g_{12}(12) + \\ &+ 2 \sum_{3 \neq 12} \int \frac{F_{123}(123)}{F_{12}(12)} \nabla_1 \ln g_{13}(13) d\mathbf{r}_3. \end{aligned} \quad (II.1)$$

And, according to the definitions of equations (2.15), (2.18), (2.20) and (3.1)

$$\begin{aligned} \nabla_1 \ln G_{12}(12) + \nabla_1 \ln \phi_{12}(12) &= 2 \nabla_1 \ln \phi_{12}(12) + 2 \nabla_1 \ln f_{12}(12) \\ &+ 2 \sum_{3 \neq 12} \int \frac{F_{123}(123)}{F_{12}(12)} \nabla_1 \ln \phi_{13}(13) d\mathbf{r}_3 \\ &+ 2 \sum_{3 \neq 12} \int \frac{F_{123}(123)}{F_{12}(12)} \nabla_1 \ln f_{13}(13) d\mathbf{r}_3. \end{aligned} \quad (II.2)$$

From the definition of $\phi_{12}(12)$, we have

$$\begin{aligned} \nabla_1 \ln \phi_{12}(12) &= 2 \nabla_1 \ln \phi_{12}(12) + \\ &+ 2 \sum_{3 \neq 12} \int \frac{\phi_{123}(123)}{\phi_{12}(12)} \nabla_1 \ln \phi_{13}(13) d\mathbf{r}_3. \end{aligned} \quad (II.3)$$

We can therefore approximate in (II.2)

$$\sum_1 \ln \phi_{12}(12) \approx 2 \sum_1 \ln \varphi_{12}(12) + 2 \sum_{3 \neq 1,2} \int \frac{F_{123}(123)}{F_{12}(12)} \sum_1 \ln \varphi_{13}(13) d\mathbf{r}_3. \quad (\text{II.4})$$

We see that the use of (II.4) introduces an error of the order

$$2 \sum_{3 \neq 1,2} \int \phi_{123}(123) \sum_1 \ln \varphi_{13}(13) [G_{13}(13) G_{23}(23) - 1] d\mathbf{r}_3,$$

but this is probably not serious because $G_{13}(13) G_{23}(23) \approx 1$ whenever $\phi_{123}(123)$ is large and $\phi_{123}(123) \approx 0$ otherwise. With this approximation (II.2) becomes

$$\sum_1 \ln G_{12}(12) \approx 2 \sum_1 \ln f_{12}(12) + 2 \sum_{3 \neq 1,2} \int \frac{F_{123}(123)}{F_{12}(12)} \sum_1 \ln f_{13}(13) d\mathbf{r}_3. \quad (\text{II.5})$$

In addition, if the localized part of the wave function is a product of single particle functions, (II.5) is exact. In particular, if the single particle functions are Gaussians, and if we assume that $G_{12}(12)$ and $f_{12}(12)$ depend only on 1-2, the relative coordinator, we can integrate (II.5) over the center of mass

coordinate and solve only for the relative part. The procedures and the resulting expression would be identical to the results that we will derive in the following pages. Therefore, the approximation (II.3) is probably a good one even for a general correlated Gaussian wave function. For numerical simplicity, we have also assumed that both $G_{12}(12)$ and $f_{12}(12)$ are spherically symmetric functions. To change (II.4) into a scalar equation, we multiply it by a unit vector along $\hat{r}_{12}$ and obtain

$$\begin{aligned} \phi_{12}(12) \frac{d}{dr_{12}} \ln G_{12}(12) &= 2 \phi_{12}(12) \frac{d}{dr_{12}} \ln f_{12}(12) + \\ &2 \sum_{3 \neq 12} \int \phi_{12}(12) G_{13}(13) G_{23}(23) \frac{d}{dr_{13}} \ln f_{13}(13) \hat{r}_{12} \cdot \hat{r}_{13} d\mathbf{r}_3 \end{aligned} \quad (II.6)$$

If we let $\eta'_{12}(12) = d/dr_{12} \ln f_{12}(12)$ and $\zeta'_{12}(12) = d/dr_{12} \ln G_{12}(12)$ and if we sphericalize (II.6) about r_{12} and rearrange terms, we find

$$\begin{aligned} \zeta'_{12}(r_{12}) &= 2 \eta'_{12}(r_{12}) + \\ &\frac{2}{\int \frac{d\mathbf{r}_n}{4\pi} \phi_{12}(12)} \sum_{3 \neq 12} \int \frac{d\mathbf{r}_n}{4\pi} \phi_{12}(12) G_{13}(13) G_{23}(23) \eta'_{13}(r_{13}) \hat{r}_{12} \cdot \hat{r}_{13} \end{aligned} \quad (II.7)$$

For correlated Gaussian wave functions with a single A value

$$\begin{aligned}\phi_{123}(123) &= \left(\frac{A^2}{3\pi}\right)^{3/2} e^{-\frac{A}{3}(u_1^2 + u_2^2 + u_{23}^2)} \\ \phi_{12}(12) &= \left(\frac{A}{2\pi}\right)^{3/2} e^{-\frac{A}{2}u_{12}^2}\end{aligned}\quad (11.8)$$

where $u_{12} = u_1 - u_2 = r_{12} - R_{12}$. Therefore, in the BBGKY equation, we have to do integrals of the form:

$$\begin{aligned}I(r_{12}) &= \int \frac{d\Omega_{12}}{4\pi} \int d\mathbf{r}_{13} \left(\frac{A^2}{3\pi}\right)^{3/2} e^{-\frac{A}{3}(u_1^2 + u_2^2 + u_{23}^2)} \\ &\quad G_{12}(r_{12}) \eta'_{12}(r_{12}) G_{23}(r_{23}) \hat{r}_{12} \cdot \hat{r}_{13}\end{aligned}\quad (11.9)$$

If we now substitute

$$\hat{r}_{12} \cdot \hat{r}_{13} = \frac{r_{12}^2 + r_{13}^2 - r_{23}^2}{2 r_{12} r_{13}}$$

and define

$$\begin{aligned}u &= r_{12} \quad , \quad v = r_{23} \quad , \quad w = r_{13} \quad , \\ a &= R_{12} \quad , \quad b = R_{23} \quad , \quad c = R_{13} \quad ,\end{aligned}$$

(II.9) becomes

$$\begin{aligned}
 I_{abc}(u) &= \frac{1}{4\pi} \left(\frac{A^2}{3u^2} \right)^{3/2} \int_0^\infty d\omega \, \omega^2 G_c(\omega) \eta_c'(\omega) \\
 &\quad \int d\Omega_u \int d\Omega_\omega G_b(v) \frac{u^2 + \omega^2 - v^2}{2u\omega} \exp \left\{ \right. \\
 &\quad \left. -\frac{A}{3} [u^2 + v^2 + \omega^2 + a^2 + b^2 + c^2 + \right. \\
 &\quad \left. 2u \cdot (b-a) + 2\omega \cdot (b-a)] \right\} .
 \end{aligned}
 \tag{II.10}$$

we can now make use of the transformations derived by Hetherington, Mullin and Nosanow³ to obtain

$$\begin{aligned}
 I_{abc}(u) &= \frac{1}{4\pi} \left(\frac{A^2}{3u^2} \right)^{3/2} e^{-\frac{A}{3}u^2} \int_0^\infty d\omega \, \omega^2 G_c(\omega) \eta_c'(\omega) \int_{|u-\omega|}^{u+\omega} dv \, \frac{v}{u\omega} \\
 &\quad G_b(v) \frac{u^2 + \omega^2 - v^2}{2u\omega} \exp \left\{ -\frac{A}{3}(v^2 + \omega^2 + a^2 + b^2 + c^2) \right\} \\
 &\quad I_1(P, Q)
 \end{aligned}
 \tag{II.11}$$

where

$$P = \left\{ \frac{2A}{3} \left[3(u^2 a^2 + v^2 b^2 + w^2 c^2) - \frac{1}{2} (u^2 + v^2 + w^2)(a^2 + b^2 + c^2) + 24 \Delta_{uvw} \Delta_{abc} \right]^{\frac{1}{2}} \right\},$$

$$Q = \left(\frac{2A}{3} \right)^2 48 \Delta_{uvw} \Delta_{abc} / P^2,$$

$$\Delta_{abc} = \frac{1}{4} \left[2(a^2 b^2 + b^2 c^2 + c^2 a^2) - (a^4 + b^4 + c^4) \right]^{\frac{1}{2}},$$

and

$$I_1(P, Q) = 8\pi^{\frac{3}{2}} \sum_{n=0}^{\infty} \Gamma(n + \frac{1}{2}) \left(\frac{-PQ}{2} \right)^n i_n(P) / (\Gamma(n+1))^2, \quad (II.12)$$

with $i_n(P) = \left(\frac{\pi}{2P} \right)^{\frac{1}{2}} I_{n+\frac{1}{2}}(P)$ the modified spherical Bessel function and $\Gamma(n+\frac{1}{2})$ the Gamma function. The expression (II.12) is useful for small PQ. For large PQ ($PQ > 15$ say), an asymptotic series is used:

$$I_1(P, Q) = \frac{8\pi}{(P^2 Q)^{\frac{1}{2}}} \sum_{n=0}^{\infty} \left(\Gamma(n + \frac{1}{2}) \right)^2 I_n(P) \left(\frac{2}{PQ} \right)^n / \Gamma(n+1) \quad (II.13)$$

As an example, let us work out the expression of a BBGKY equation with a two-shell approximation. For a two-shell approximation,

we assume that

$$G_b(r) \equiv 1$$

$$f_b(r) \equiv 1$$

for $b \geq 2$. That means, $\eta'_b(r) = 0$ for $b \geq 2$. The subscript 2 denotes the second neighbour.

Then (II.6) becomes

$$\zeta'_a(u) = 2 \eta'_a(u) + \frac{2}{\int \frac{d\lambda}{4\pi} \phi_a(u)} \sum_{\substack{b \leq 2 \\ c}}^{(\text{restricted})} I_{abc}(u) \quad (II.14)$$

For a bcc lattice, in the two-shell approximation, the set of coupled BBGKY equations is

$$\begin{aligned} \zeta'_i(u) = 2 \eta'_i(u) + \frac{2}{\int \frac{d\lambda}{4\pi} \phi_i(u)} \{ & 3 I_{i2i}(u) + 3 I_{i4i}(u) \\ & + I_{i5i}(u) + 3 I_{i12}(u) + 3 I_{i42}(u) \} \end{aligned} \quad (II.15)$$

$$\zeta_2'(u) = 2\eta_2'(u) + \frac{2}{\int \frac{d\Omega_u}{k\eta} \phi_2(u)} \left\{ k I_{211}(u) + k I_{241}(u) \right. \\ \left. + k I_{232}(u) + I_{262}(u) \right\}. \quad (11.16)$$

(11.13) and (11.14) can be solved self-consistently for $\eta(r)$ if coupled with equations (13) and (14) of Chapter II which produce the Gaussian A value and $\zeta(r)$.

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TABLE I

Calculated values of E_0 , ground-state energy and A for the nearest-neighbour distances $R = 3.45$ and $R = 3.65$.

R (Å)	A (Å ⁰⁻²)	E_0 (°K)
3.45	1.40	3.4
3.65	1.15	2.3

TABLE II

Monte-Carlo ground-state energy E_0 as a function of A for $R = 3.65$. The correlation function was the same for each value of A .

A (Å ⁰⁻²)	E_0 (°K)
1.15	4.42
1.05	3.70
1.00	3.54 (min.)
0.95	3.57
0.85	4.17

TABLE III

Monte-Carlo ground-state energy E_0 as a function of A for $R = 3.45$. The correlation function was the same for each value of A .

$A \text{ (}\overset{0-2}{\text{\AA}}\text{)}$	$E_0 (^{\circ}\text{K})$
1.30	7.27
1.40	6.80 (min.)
1.50	7.61

TABLE IV

Convergence of the n-body cluster integral for the first-neighbour exchange frequency (J_1) the second-neighbour exchange frequency (J_2) and the 112-triple exchange frequency (J_{112}) for the nearest-neighbour distance $R = 3.65$ Å and $A = 1.15$ Å. o-2

n	$J_1(10^{-5} \text{ } ^\circ\text{K})$	$J_2(10^{-6} \text{ } ^\circ\text{K})$	$J_{112}(10^{-6} \text{ } ^\circ\text{K})$
2	-48.4	-42.2	--
5	--	--	-7.0 ± 0.2
6	--	-2.40 ± 0.04	--
8	-6.1 ± 0.2	--	--
10	--	--	-2.45 ± 0.35
14	-10.3 ± 0.3	-2.03 ± 0.14	--
15	--	--	-2.5 ± 0.17
18	--	-2.4 ± 0.3	--
20	-10.6 ± 0.3	--	--

TABLE V

Typical values of J_1 , J_2 and J_{112} for nearest-neighbour distances of 3.45 Å and 3.65 Å.

	$J_1(10^{-5} \text{ } ^\circ\text{K})$	$J_2(10^{-6} \text{ } ^\circ\text{K})$	$J_{112}(10^{-6} \text{ } ^\circ\text{K})$
$R = 3.65$ $A = 1.15$	-10.6 ± 0.3	-2.4 ± 0.3	-2.5 ± 0.17
$R = 3.65$ $A = 1.0$	-40.0 ± 4	-14.3 ± 1.4	-14.0 ± 1.0
$R = 3.45$ $A = 1.40$	-1.9 ± 0.1	-0.37 ± 0.06	-0.35 ± 0.03

Figure Captions

Figure 1: The correlation function f compared with the Nosanow correlation function (N), the Guyer correlation function (G) and the Hensen-Levesque correlation function (HL). Here f_1 indicates the first-neighbour correlation function and f_2 the second-neighbour correlation for a first-neighbour distance $R = 3.65 \text{ \AA}$ and $A = 1.15 \text{ \AA}^{-2}$, whereas f_1' indicates the first-neighbour correlation function for $R = 3.45 \text{ \AA}$ and $A = 1.40 \text{ \AA}^{-2}$.

Figure 2: Calculated volume dependence of the ground-state energy as compared with the experimental values (E), the Nosanow values (N), the Guyer values (G), and the Hensen and Levesque values (HL). The two points are calculated from the variational equation. The curves (I) and (II) indicate the Monte-Carlo calculations; (I) used the same A values as the numerical results, whereas (II) indicates the Monte-Carlo minimum using the numerical correlation functions.

Figure 3: The pair distribution functions calculated from the differential equation (solid line) and the Monte-Carlo method (dashed line).

Figure 4: The sphericalized one-particle distribution function calculated by Monte Carlo methods ($R = 3.65 \text{ \AA}$, $A = 1.15 \text{ \AA}^{-2}$). The dashed line is the Hartree one-particle distribution function with $A = 1.15 \text{ \AA}^{-2}$.

Figure 5: The exchange integral J as a function of the nearest-neighbour distance R . The dashed lines labelled RHM and PA are experimental values of J_1 from the thermodynamic data of Panczyk and Adams (PA), and the NMR data of Richardson, Hunt and Meyer (RHM). The remaining curves labelled ES are theoretical results for J_1 , J_2 , J_{112} , obtained using the values of the Einstein parameter due to Ebner and Sung (ES) and the Nosanow correlation function. The points and solid lines are the results of the present calculation. For our calculation, $|J_2| \simeq |J_{112}|$.

Energy per Particle ($^{\circ}\text{K}$)