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Tunneling and Exchange in Quantum Solids*

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Pairs of ^{3}He atoms in solid bcc ^{3}He have spatially symmetric or antisymmetric wave functions which correspond to energies ϵ^+ and ϵ^-, $\epsilon^\pm = \epsilon \pm \frac{1}{2}\Delta\epsilon$. The energy difference $\epsilon^+ - \epsilon^- = \Delta\epsilon$ is due to a tunneling process and an interaction process. The effect of these two processes can be simulated by adding to the Hamiltonian of the solid an exchange Hamiltonian,

$$\mathcal{H}_x = -2\Delta\epsilon \sum_{ij}' \vec{\sigma}_i \cdot \vec{\sigma}_j = -2(\Delta\epsilon_T + \Delta\epsilon_x) \sum_{ij}' \vec{\sigma}_i \cdot \vec{\sigma}_j ,$$

where $\Delta\epsilon$ is a sum of $\Delta\epsilon_T$ (due to the tunneling process) and $\Delta\epsilon_x$ (due to the interaction process). A theory of the magnitude and sign of $\Delta\epsilon_x$ and $\Delta\epsilon_T$ is given. We find $\Delta\epsilon_T < 0$ and $\epsilon_x > 0$. Using quite general arguments, we show that $|\Delta\epsilon_T| \gtrsim 2|\Delta\epsilon_x|$. The exchange in bcc solid ^{3}He is antiferromagnetic. Evaluation of the formulas for $\Delta\epsilon_T$ and $\Delta\epsilon_x$ using the ground-state wave function of Guyer and Sarkissian leads to $\Delta\epsilon = J$, in good agreement with experiment.

I. INTRODUCTION

A quantum solid is one in which the root-mean-square deviation of a particle from its lattice site, u_{rms}, is a large fraction of the near-neighbor distance Δ; $u_{\text{rms}}/\Delta > \frac{1}{5}$; the zero-point motion of the atoms is large. As a consequence of this large zero-point motion it is possible for atoms on neighboring lattice sites to change places. For a pair of ^{3}He atoms (fermions) on neighboring lattice sites R_1 and R_2, we write a wave function of the form

$$\psi_{R_1R_2}(\vec{x}_1\sigma_1, \vec{x}_2\sigma_2) = \Phi_{R_1R_2}(\vec{x}_1\vec{x}_2)X(\sigma_1\sigma_2)$$
$$= \varphi_{R_1}(\vec{x}_1)\varphi_{R_2}(\vec{x}_2)X(\sigma_1\sigma_2) , \quad (1)$$

where $\varphi_{R_1}(\vec{x}_1)$ and $\varphi_{R_2}(\vec{x}_2)$ are single-particle wave functions localized in the vicinity of lattice sites R_1 and R_2, and $X(\sigma_1\sigma_2)$ is the spin wave function for the pair.[1] If Eq. (1) is to be a good first approximation to the wave function of the pair, the overlap between

$$\varphi_{R_1}(\vec{x}_1) \quad \text{and} \quad \varphi_{R_2}(\vec{x}_1)$$

must be small. Although the zero-point motion of the atoms is large, it is not so large that we do not have

$$\int d\vec{x}_1\, \varphi_{R_1}(\vec{x}_1)\varphi_{R_2}(\vec{x}_1) \ll \int d\vec{x}_1\, \varphi_{R_1}(\vec{x}_1)\varphi_{R_1}(\vec{x}_1) . \quad (2)$$

The state

$$\psi_{R_1R_2}(\vec{x}_1\sigma_1, \vec{x}_2\sigma_2)$$

and the state

$$\psi_{R_1R_2}(\vec{x}_2\sigma_2, \vec{x}_1\sigma_1) ,$$

in which particle 1 is at lattice site R_2 and particle 2 is at lattice site R_1, are degenerate with energy eigenvalue E. But if there is any overlap between

$$\varphi_{R_1}(\vec{x}_1) \quad \text{and} \quad \varphi_{R_2}(\vec{x}_1) ,$$

we must admit the possibility of exchange, i.e., particle 1 can be at lattice site R_2 as well as R_1, and vice versa. We must write properly symmetrized pair wave functions (for a pair of fermions we need wave functions which are antisymmetric under exchange $\vec{x}_1 \leftrightarrow \vec{x}_2$, $\sigma_1 \leftrightarrow \sigma_2$). When the spins of the particles are parallel,

$$X(\sigma_1\sigma_2) = \delta_{\sigma_1\uparrow}\delta_{\sigma_2\uparrow} = X^- ,$$

we use $\Phi^-_{R_1R_2}(\vec{x}_1\vec{x}_2)$,

a space antisymmetric wave function for which

$$\Phi^-_{R_1R_2}(\vec{x}_1\vec{x}_2) = -\Phi^-_{R_1R_2}(\vec{x}_2\vec{x}_1) ; \quad (3)$$

then

$$\psi^-_{R_1R_2}(\vec{x}_1\sigma_1, \vec{x}_2\sigma_2) = \Phi^-_{R_1R_2}(\vec{x}_1\vec{x}_2)X^-(\sigma_1\sigma_2)$$

$$= -\psi^-_{R_1R_2}(\vec{x}_2\sigma_2, \vec{x}_1\sigma_1) \ .$$

When the spins of the particles are antiparallel

$$X^+(\sigma_1\sigma_2) = (\delta_{\sigma_1\uparrow}\delta_{\sigma_2\downarrow} - \delta_{\sigma_1\downarrow}\delta_{\sigma_2\uparrow})/\sqrt{2} \ ,$$

we use $\Phi^+_{R_1R_2}(\vec{x}_1\vec{x}_2)$,

a space symmetric wave function for which

$$\Phi^+_{R_1R_2}(\vec{x}_1\vec{x}_2) = \Phi^+_{R_1R_2}(\vec{x}_2\vec{x}_1) \ ; \qquad (4)$$

then

$$\psi^+_{R_1R_2}(\vec{x}_1\sigma_1, \vec{x}_2\sigma_2) = \Phi^+_{R_1R_2}(\vec{x}_1\vec{x}_2)X^+(\sigma_1\sigma_2)$$

$$= -\psi^+_{R_1R_2}(\vec{x}_2\sigma_2, \vec{x}_1\sigma_1) \ .$$

These space symmetric and antisymmetric wave functions have energy eigenvalues E^+ and E^-, respectively, and lift the degeneracy which existed for the unsymmetrized wave functions. Let us assume $E^\pm = E_0 \pm \Delta E$. The effect of admitting the possibility of exchange can be simulated in the Hamiltonian describing the pair by adding to the bare Hamiltonian $\mathcal{H}$, the spin-dependent term

$$-4\Delta E\vec{\sigma}_1\cdot\vec{\sigma}_2 \ . \qquad (5)$$

Since the overlap of near-neighbor pair wave functions is largest, we should add a term like Eq. (5) for all near-neighbor pairs in the solid,

$$\mathcal{H}_x = -4\sum_i \sum_{j\neq(i)} \Delta E_{ij}\vec{\sigma}_i\cdot\vec{\sigma}_j \ . \qquad (6)$$

The properties of the solid which follow because of the Hamiltonian $\mathcal{H}_x$ are referred to as exchange properties.[2] They depend on the sign and magnitude of ΔE. The exchange properties are many and they have been the subject of a long and extensive experimental exploration.[3]

The purpose of this paper is to discuss the theory of exchange in quantum solids. We will not deal with exchange phenomena, rather we will describe a theory of the magnitude and sign of ΔE.

The early theoretical work on exchange in quantum solids, that of Bernades and Primakoff[4] and Sanders,[5] is primarily of historical interest. More recently, Nosanow and co-workers[6-9] and Thouless[10] have developed theories of exchange. Nosanow and co-workers have developed a theory as an integral part of their quantum solids program. As such, there is a body of computational results which follows from the quantification of their theory. (i) When the properly symmetrized wave functions are taken to be a superposition of the unsymmetrized wave functions, Nosanow and co-workers find antiferromagnetism, i.e., $\Delta E < 0$, of about the right order of magnitude.[11] (ii) ΔE is very sensitive to the details of the short-range correlation function (both in magnitude and sign); ΔE is also very sensitive to the details of the single-particle wave functions.

The theory of exchange due to Thouless is a theory of tunneling.[2] It produces an intuitively appealing and simple result. (a) $\Delta E < 0$. The ground state of the system is manifestly antiferromagnetic. (b) This result follows because ψ^+ has one less node than ψ^- and hence $E^+ < E^-$; it depends in no way upon the details of the wave function describing the pair. The theory due to Thouless does not include the short-range correlations which are an integral part of the Nosanow theory. The Thouless theory can be modified to include short-range correlations with no qualitative change in conclusions (a) and (b) above.

There is a fundamental difference between the result of Thouless and those of Nosanow *et al.* Thouless finds manifest antiferromagnetism; Nosanow and co-workers find a magnetism which is extremely sensitive to details which are totally irrelevant to the Thouless theory. The computational procedures used by Thouless and Nosanow and co-workers are sufficiently different as to preclude easy comparison of these theories.

In Sec. II, we briefly review the theory of quantum solids which was recently developed by Guyer[12] and Guyer and Sarkissian.[13] We apply this theory in Sec. III to the calculation of ΔE as an illustration of the sort of computational procedure which is employed in tunneling calculations. The calculation is a generalization of the Thouless tunneling theory; it includes short-range correlations. We obtain manifest antiferromagnetism independent of the details of the pair wave function. The important point is that the sign of ΔE depends in no way upon the nature of the bare interaction between a pair of particles. In Sec. IV, we look more carefully at the pair problem which leads to ΔE in order to learn where the effects of the interaction are. We find that in order for the bare interaction to have an effect on the sign of ΔE the short-range correlation function must depend upon the symmetry of the pair wave function. We are then led to view the exchange process as a combination of two processes; (i) a tunneling process, and (ii) an interaction process. We find that the tunneling process drives the system toward an antiferromagnetic ground state, whereas the interaction process can lead to ferromagnetism or antiferromagnetism depending on the nature of the bare interaction. We exhibit formulas for the contribution of the tunneling process and the interaction process to ΔE. In Sec. V, we evaluate these formulas. We find that the tunneling process causes an energy shift (toward antiferromagnetism) about 2.5 times the energy shift of the interaction process. The exchange interaction is due to a tunneling process; the ground state of the system is antiferromagnetic. We discuss the physics of the tunneling process in detail. Our concluding remarks are in Sec. VI. In the Appendices, we include various analytic details which are needed to support the arguments in the body of the paper.

II. THEORY OF QUANTUM SOLIDS (REVIEW)

We begin this section by briefly reviewing the theory of the ground state of quantum solids due to Guyer[12] and Guyer and Sarkissian.[13]

The Hamiltonian which describes a solid ^{3}He crystal is

$$\mathcal{H}=\sum_{i=1}^{N} T(i)+\frac{1}{2}\sum_{i,j}{}' v(ij) \;, \tag{7}$$

where $T(i)=\vec{p}_i^{\,2}/2m$ and $v(ij)$ is the Lennard-Jones interaction between particles i and j;

$$v(ij)=v(r_{ij})=4\epsilon[(\sigma/r_{ij})^{12}-(\sigma/r_{ij})^{6}] \;, \tag{8}$$

$\epsilon=10.2$ °K and $\sigma=2.556$ Å. To do a formal perturbation theory of a system which is characterized by localization, it is useful to add to $\mathcal{H}$ an effective interaction between pairs of particles which will lead to a convenient localized first approximation. We write

$$\mathcal{H}=\sum_{i=1}^{N} T(i)+\frac{1}{2}\sum_{ij}{}' w_{ij}(ij) + \frac{1}{2}\sum_{ij}{}'[v(ij)-w_{ij}(ij)] \;, \tag{9}$$

where

$$w_{ij}(ij)=w_{R_iR_j}(\vec{x}_i\vec{x}_j)$$

is an effective interaction between $\vec{x}_i$ localized near lattice site R_i and $\vec{x}_j$ localized near lattice site R_j.[14] $\mathcal{H}$ is separated in the form $\mathcal{H}=\mathcal{H}_0+V$, where

$$\mathcal{H}_0=\sum_i T(i)+\frac{1}{2}\sum_{ij}{}' w_{ij}(ij) \;, \tag{10a}$$

and $$V=\frac{1}{2}\sum_{ij}{}' \tilde{v}(ij)=\frac{1}{2}\sum_{ij}{}' [v(ij)-w_{ij}(ij)] \;. \tag{10b}$$

We assume that the problem $\mathcal{H}_0\Phi_n(1\cdots N)=E_n\Phi_n(1\cdots N)$ is solvable and leads to a ground state which is characterized by the localization of particles in the vicinity of a lattice site. The complete set of states generated by $\mathcal{H}_0$ is to be used to do perturbation theory on V. To be able to be more explicit in the developments which follow, we will take $w_{ij}(ij)$ in such a form that the $\Phi_n(1\cdots N)$ are products of single-particle states. For example, we may choose

$$w_{ij}(ij)=w_{R_iR_j}(0)+\tfrac{1}{2}w''_{R_iR_j}(0):\; \vec{u}_{ij}\vec{u}_{ij}+\cdots, \tag{11}$$

where $\vec{u}_i=\vec{x}_i-\vec{R}_i,\ \vec{u}_{ij}=\vec{u}_i-\vec{u}_j$, such that

$$U_{R_i}(\vec{x}_i)=U_i(i)=\sum_{j\neq(i)}\int d\vec{x}_j\,|\varphi_j(j)|^2 w_{ij}(ij) \tag{12}$$

is well defined. Then the Hamiltonian $\mathcal{H}_0$ can be regarded as a sum of single-particle Hamiltonians;

$$\mathcal{H}_0=\sum_i[T(i)+\tfrac{1}{2}U_i(i)] \;, \tag{13}$$

and $\Phi_n(1\cdots N)$ is a product of single-particle wave functions, $\Phi_n(1\cdots N)=\Pi_i\,\varphi_i^{\alpha_i}(i)$, where

$$\mathcal{H}_0(1\cdots N)\Phi_n(1\cdots N)=E_n\Phi_n(1\cdots N) \tag{14}$$

and the $\varphi_i^{\alpha_i}(i)=\varphi_{R_i}^{\alpha_i}(\vec{x}_i)$

are solutions of [15]

$$[T(i)+U_i(i)]\varphi_i^{\alpha_i}(i) = h_i(i)\varphi_i^{\alpha_i}(i)=\epsilon_{\alpha_i}\varphi_i^{\alpha_i}(i). \tag{15}$$

In writing a product for $\Phi_n(1\cdots N)$ we will have a useful set of wave functions if

$$\langle\Phi_n(1\cdots N)|\Phi_{n'}(1\cdots N)\rangle=\delta_{nn'} \;, \tag{16}$$

where $n=\{\alpha_1\cdots\alpha_N\}$ denotes the excitation level of the particle at each lattice site. Equation (16) is satisfied if we have

$$\langle\varphi_i^{\alpha}(i)|\varphi_j^{\beta}(i)\rangle=\delta_{\alpha\beta}\delta_{R_iR_j} \;. \tag{17}$$

Of course, $\langle\varphi_i^{\alpha}(i)|\varphi_i^{\beta}(i)\rangle=\delta_{\alpha\beta}$ follows from Eq. (15). For a solid in which the particles are well localized in the vicinity of their lattice site, e.g., neon, argon, etc., we have

$$\langle\varphi_i^{\alpha}(i)|\varphi_j^{\beta}(i)\rangle\approx\delta_{\alpha\beta}\delta_{R_iR_j} \tag{18}$$

to good approximation for the low-lying states. For a quantum solid in which the single-particle wave functions are reasonably extended in space, we must be more careful. For example, in solid helium we have

$$\langle\varphi_i^{0}(i)|\varphi_j^{0}(i)\rangle/\langle\varphi_i^{0}(i)|\varphi_i^{0}(i)\rangle\lesssim e^{-\frac{1}{2}\alpha^2\Delta^2}=10^{-2}\ ,$$

where $\alpha^2\approx1-2$, and Δ is the distance between near-neighbor lattice sites R_i and R_j.[16] We may avoid the difficulty associated with the nonorthogonality of the single-particle states generated by $\mathcal{H}_0$ by several schemes.

(i) Put each particle in its own quantum field

$$\psi_{R_i}(\vec{x}_i)$$

described by the complete set of states at its lattice site only.[17] If particle 1 which is usually localized near R_1 is to be localized near R_2, it does so by existing in a proper superposition of

$$\varphi_{R_1}^{\alpha}(\vec{x}_1)\ .$$

Certainly this is not a convenient scheme for describing a system in which the particles change places. But it is useful for doing a Hartree or similar unsymmetrized calculation. In this case the question of nonorthogonality does not enter.[18]

(ii) Suppose we know that particles 1 and 2 can both be in the vicinity of lattice sites R_1 and R_2 while particles $3\cdots N$ stay in the vicinity of their respective lattice sites. Then, we may write

$$\Phi_n(12;3\cdots N)=\varphi_{R_1R_2}^{\alpha}(12)\prod_{i=3}^{N}\varphi_i^{\alpha_i}(i), \tag{19}$$

where $\varphi_{12}^{\alpha}(12)$ is a pair wave function which satisfies the equation

$$\begin{aligned}h_{12}(12)\varphi_{12}^{\alpha}(12)&=[T(1)+T(2)+U_{12}(1)+U_{12}(2)]\varphi_{12}^{\alpha}(12)\\&=\epsilon_{\alpha}(12)\varphi_{12}^{\alpha}(12)\ .\end{aligned} \tag{20}$$

The single-particle potentials in Eq. (20) tend to localize a particle in the vicinity of two lattice sites R_1 and R_2 (see Fig. 1). In this case we have adulterated the specification of $\mathcal{H}_0$; it is now a sum of single-particle Hamiltonians for all but the pair of particles 1 and 2. We have

$$\mathcal{H}_0(12;3\cdots N)=h_{12}(12)+\sum_{i=3}^{N}h_i(i)\ , \tag{21}$$

and

$$\begin{aligned}\mathcal{H}_0(12;3\cdots N)\Phi_n(12;3\cdots N)\\=E_n(12)\Phi_n(12;3\cdots N)\ .\end{aligned} \tag{22}$$

This complete set of states $\{\Phi_n(12;3\cdots N)\}$ will be particularly convenient for describing the exchange process between particles on lattice sites R_1 and R_2.

(iii) We may employ in place of the single-particle states generated by Eq. (15) the Wannier single-particle wave functions[19] $\{\theta_i^{\alpha}(i)\}$ for which

$$\langle\theta_i^{\alpha}(i)|\theta_j^{\beta}(i)\rangle=\delta_{\alpha\beta}\delta_{R_iR_j}\ . \tag{23}$$

This set of functions has the orthogonality property which is called for to permit second quantization of $\mathcal{H}$ and the use of the full range of field theoretic computational procedures. The Wannier states provide an immense computational advantage at the expense of wave-function simplicity. In what follows we will always use the formulation of perturbation theory within

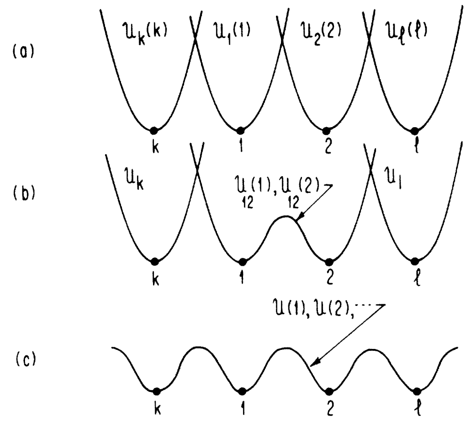

FIG. 1. Single-particle potentials I: (a) Each particle stays in the vicinity of its lattice site – no exchange. (b) Particles 1 and 2 can exchange. They see a common double well due to the other particles in the solid. (c) A periodic single-particle potential is seen by all of the particles in the solid.

the context of one of these three orthogonalization schemes.

Let us calculate the ground-state energy of the system described by the Hamiltonian $\mathcal{H}$, Eq. (7), using the perturbation theory appropriate to orthogonalization scheme 1. We will not consider the exchange process. The expectation value of $\mathcal{H}$ is given by the Rayleigh-Schrödinger perturbation series in V. For the energy shift, we have

$$\Delta E = \langle \mathcal{H}\rangle - E_0 = \langle V\rangle + \langle VGV\rangle + \langle VGVGV\rangle + \cdots - \langle V\rangle\langle VGGV\rangle + \cdots , \quad (24)$$

where $G = (E_0 - \mathcal{H}_0)^{-1}(1 - |\Phi_0\rangle\langle\Phi_0|)$, (25)

and $\langle\cdots\rangle = \langle\Phi_0(1\cdots N)|\cdots|\Phi_0(1\cdots N)\rangle$. (26)

Progress is made in obtaining ΔE using Eq. (24) by making various approximations to the full perturbation expansion.

We proceed by making a sequence of approximations to ΔE in which a pair of particles, a triple of particles, etc., are treated exactly. Here we look in detail at the pair approximation.

In each term of the perturbation expansion we follow a pair of particles through the term letting them interact among themselves only. We write

$$\langle V\rangle = \frac{1}{2}\sum_{ij}{}' \langle \bar v(ij)\rangle ,$$

$$\langle VGV\rangle = \frac{1}{2}\sum_{ij}{}' \langle \bar v(ij) G \bar v(ij)\rangle , \quad (27)$$

$$\langle VGVGV\rangle = \frac{1}{2}\sum_{ij}{}' \langle \bar v(ij) G \bar v(ij) G \bar v(ij)\rangle , \quad \text{etc.} \quad (28)$$

With this approximation to each term we have

$$\Delta E = \frac{1}{2}\sum_{ij}{}' [\langle \bar v(ij)\rangle + \langle \bar v(ij) G \bar v(ij)\rangle + \cdots - \langle \bar v(ij)\rangle\langle \bar v(ij) GG \bar v(ij)\rangle + \cdots] . \quad (29)$$

This energy shift is the sum of the energy shifts of the auxiliary problems

$$\mathcal{H}_{0,ij}\Phi_{0,ij}(1\cdots N) = [\mathcal{H}_0 + \bar v(ij)]\Phi_{0,ij}(1\cdots N) = E_{ij}\Phi_{0,ij}(1\cdots N) . \quad (30)$$

The Rayleigh-Schrödinger perturbation expansion for the energy shift in Eq. (30) is

$$E_{ij} - E_0 = \langle \bar v(ij)\rangle + \langle \bar v(ij) G \bar v(ij)\rangle + \langle \bar v(ij) G \bar v(ij) G \bar v(ij)\rangle + \cdots - \langle \bar v(ij)\rangle\langle \bar v(ij) GG \bar v(ij)\rangle + \cdots .$$

Thus, we write

$$\Delta E = \frac{1}{2}\sum_{ij}{}' (E_{ij} - E_0) . \quad (31)$$

It is important to recognize that Eq. (30) is a relatively simple equation of motion for the wave function of the pair of particles i and j. The perturbation $\bar v(ij)$ affects particles i and j only; we write[20]

$$\Phi_{0,12}(1\cdots N) = \psi_{12}(12)\prod_{i=3}^{N}\varphi_i^0(i)$$

and

$$[h_1(1) + h_2(2) + \bar v(12)]\psi_{12}(12) = \epsilon_{12}\psi_{12}(12) , \quad (32)$$

where $E_{12} = E_0 + \epsilon_{12} - 2\epsilon_0$. This equation describes a pair of particles each in the single-particle potential of the lattice medium and interacting with one another through $v(12)$. In Eq. (32) the single-particle potential on particle 1, in the vicinity of lattice site R_1, is not $U_{R_1}(\vec{x}_1)$ from Eq. (12); it is

$$\bar U_{R_1}(\vec{x}_1) = U_1(1) - \int d\vec{x}_2 |\varphi_2(2)|^2 w_{12}(12) , \quad (33)$$

a slightly skewed single-particle potential due to the treatment of particle 2 exactly (see Fig. 2).

We write the pair wave function which solves Eq. (32) in the form

$$\psi_{12}(12) = \varphi_1^0(1)\varphi_2^0(2)g_{12}(12) . \quad (34)$$

Then, the energy shift for the pair, $\epsilon_{12} - 2\epsilon_0$, can be written in terms of $g_{12}(12)$, viz.,

$$\epsilon_{12} - 2\epsilon_0 = \frac{\langle\Phi_0(1\cdots N)|\,\bar v(12)\,|\,\Phi_{0,12}(1\cdots N)\rangle}{\langle\Phi_0(1\cdots N)|\,\Phi_{0,12}(1\cdots N)\rangle} \quad (35)$$

$$\text{or} \quad \epsilon_{12} - 2\epsilon_0 = \frac{\langle \bar v(12) g_{12}(12)\rangle}{\langle g_{12}(12)\rangle} .$$

Using Eq. (35) in Eq. (31) we have

$$\Delta E = \frac{1}{2}\sum_{ij}{}' \left(\frac{\langle v(ij) g_{ij}(ij)\rangle}{\langle g_{ij}(ij)\rangle} - \frac{\langle w_{ij}(ij) g_{ij}(ij)\rangle}{\langle g_{ij}(ij)\rangle} \right) . \quad (36)$$

We expect a functional form for $w_{12}(12)$ which

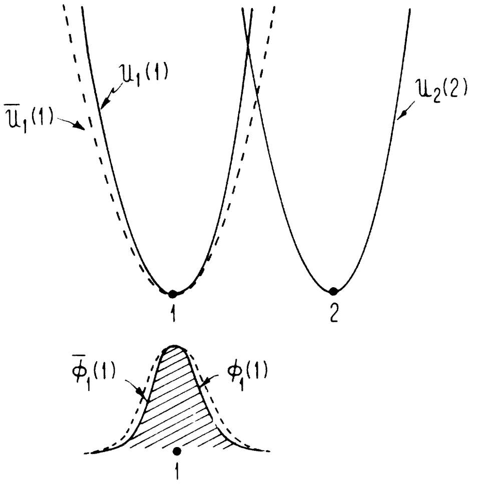

FIG. 2. Single-particle potentials II: Particle 1 near lattice site $\vec{R}_1$ sees the single-particle potential $U_1(1)$ which is due to all of the other particles in the solid. Particle 2 sees a similar potential centered at $\vec{R}_2$. Particle 2 contributes to $U_1(1)$. When the contribution to $U_1(1)$ of particle 2 is subtracted, particle 1 sees $\bar{U}_1(1)$.

leads to $U_1(1)$ of Eq. (12) in the form $U_1(1) \approx U_1(\vec{x}_1 - \vec{R}_1)$.

Thus, $\int d\vec{x}_j \,|\varphi_j(j)|^2 w_{1j}(1j) = w_{1j}(\vec{u}_1)$

depends upon $\vec{u}_1 = \vec{x}_1 - \vec{R}_1$, and $\langle w_{1j}(1j) g_{1j}(1j)\rangle / \langle g_{1j}(1j)\rangle$ is not sensitive to the weighting factor $g_{1j}(1j)$.

Thus,
$$\frac{\langle w_{ij}(ij) g_{ij}(ij)\rangle}{\langle g_{ij}(ij)\rangle} \approx \frac{\langle w_{ij}(ij)\rangle}{\langle 1\rangle} \tag{37}$$

is a good approximation.[21] Since E_0 is given by

$$E_0 = \sum_i \langle T(i)\rangle + \frac{1}{2} \sum_{ij}{}' \langle w_{ij}(ij)\rangle \quad , \tag{38}$$

we can use Eqs. (36) and (37) to write E in the form

$$E = \sum_i \langle T(i)\rangle + \frac{1}{2} \sum_{ij}{}' \frac{\langle v(ij) g_{ij}(ij)\rangle}{\langle g_{ij}(ij)\rangle} \quad .$$

We define the t matrix by

$$t_{12}(12) = v(12) g_{12}(12) / \langle g_{12}(12)\rangle \quad , \tag{39}$$

and, in terms of it, the energy of the system is

$$E = \sum_i \langle T(i)\rangle + \frac{1}{2} \sum_{ij}{}' \langle t_{ij}(ij)\rangle \quad . \tag{40}$$

The t matrix acts like an effective interaction between the pair of atoms i and j, localized near lattice sites R_i and R_j, respectively. It is found by solving Eq. (32) in the form given by Eq. (34). Equations (14), (32), and (31) constitute the pair approximation to $E - E_0$.

So far in the pair approximation we have an unknown, since we have not specified the effective interaction $w_{12}(12)$. It may be chosen in several ways. (a) Choose $w_{12}(12)$ so that the pair energy

$$\sum_{ij}{}' (\epsilon_{ij} - 2\epsilon_0)$$

vanishes identically. (b) Choose $w_{12}(12)$ so that E in the pair approximation is stationary with respect to variation of the parameters in $w_{12}(12)$. We briefly discuss these two alternatives:

(a) The choice

$$\sum_{ij}{}' (\epsilon_{ij} - 2\epsilon_0) = 0$$

implies, Eq. (36),

$$\sum_i \sum_{j \neq (i)} \langle t_{ij}(ij)\rangle = \sum_i \sum_{j \neq (i)} \frac{\langle w_{ij}(ij) g_{ij}(ij)\rangle}{\langle g_{ij}(ij)\rangle}$$

$$\approx \sum_i \sum_{j \neq (i)} \langle w_{ij}(ij)\rangle \quad ,$$

using the approximation of Eq. (37). From the definition of the single-particle potential in Eq. (12), we have

$$\sum_i \Bigl(\sum_{j \neq (i)} \langle t_{ij}(ij)\rangle - U_i(i) \Bigr) = 0 \quad . \tag{41}$$

Thus, the condition

$$\sum_{ij}{}' (\epsilon_{ij} - 2\epsilon_0) = 0$$

is satisfied when the expectation value of the energy of particle i in the single-particle potential $U_i(i)$ is the same as the expectation value of the energy of i interacting via the t matrix with the other particles in the lattice. For a particular choice of the functional form for $U_i(i)$, Eq. (41) may be further reduced to a computationally tractable form.[22]

(b) To apply method (b) a parametrized choice of the single-particle potential must be made. Let us choose

$$U_i(i) = U_0 + \tfrac{1}{2}k u_i^2 \quad . \tag{42}$$

Then for a given choice of k we have E, a functional of k, $E[k]$. We may vary $E[k]$ with respect to k to attempt to find a stationary (possibly minimum) value of E. Since the basic equations we are employing are much like those of Koehler[23] and Gillis, Werthamer, and Koehler[24] we expect to achieve a result similar to theirs. We find $\delta E[k]/\delta k$ implies[25]

$$1 = \frac{4m}{3\hbar^2} \sum_{j \neq (i)} [\langle u_i^2 t_{ij}(ij)\rangle - \langle t_{ij}(ij)\rangle \times \langle u_i^2 g_{ij}(ij)\rangle / \langle g_{ij}(ij)\rangle] \quad . \tag{43}$$

Finally, the pair approximation requires the solution to Eqs. (14), (32), and (31), subject to the constraint of Eq. (41) or Eq. (43).

The basic idea of the sequence of approximations (which starts with the pair approximation) is to treat larger and larger clusters of particles exactly while treating the interaction of the cluster with its medium in some approximate way. In the lowest-order approximation, $\mathcal{H}_0$ or E_0, all particles are coupled to one another by springs. In the pair approximation, the springs are removed for one pair at a time and replaced by the true Lennard-Jones interaction. In the triple approximation, a triple of particles interact among themselves via the Lennard-Jones interaction; they interact with all other particles in the system by springs (see Fig. 3).

III. TUNNELING

Let us begin the discussion of exchange by looking in some detail at the solution to the ground-state problem in the pair approximation of Sec. II. In this approximation the energy of the system is given by

$$E = E_0 + \frac{1}{2} \sum_{ij}{}' (E_{ij} - E_0) \quad , \tag{31}$$

where E_0 and E_{ij} are the energy eigenvalues of the auxiliary problems

$$\mathcal{H}_0(1\cdots N)\Phi_0(1\cdots N) = E_0\Phi_0(1\cdots N) \quad , \tag{14$'$}$$

and $$\mathcal{H}_{0,ij}\Phi_{0,ij}(1\cdots N) = E_{ij}\Phi_{0,ij}(1\cdots N) \quad . \tag{30$'$}$$

For a choice of $w_{ij}(ij)$ which leads to single-particle potentials at each lattice site [e.g., the choice in Eq. (12)] we have

$$E_0 = N[\epsilon_0 - \tfrac{1}{2}\langle U_i(i)\rangle] \quad ,$$

where $$[T(i) + U_i(i)]\varphi_i^0(i) = \epsilon_0 \varphi_i^0(i) \tag{44}$$

and $$\Phi_0(1\cdots N) = \prod_{i=1}^{N} \varphi_i^0(i) \quad . \tag{45}$$

The solution to Eq. (30) reduces to Eq. (32). Using the definition of $\bar{U}_i(i)$ in Eq. (33) we have

$$[T(i) + T(j) + \bar{U}_i(i) + \bar{U}_j(j) + v(ij)]\psi_{ij}(ij) = \epsilon_{ij}\psi_{ij}(ij) \quad . \tag{46}$$

Equation (46) is the equation of motion for a pair of particles, each localized in the vicinity of a lattice site due to the solid medium, interacting with one another through $v(ij)$.[26] When we write $\psi_{ij}(ij)$ in the product form of Eq. (34), Eq. (46) amounts to an equation of motion for $g_{ij}(ij)$. We have

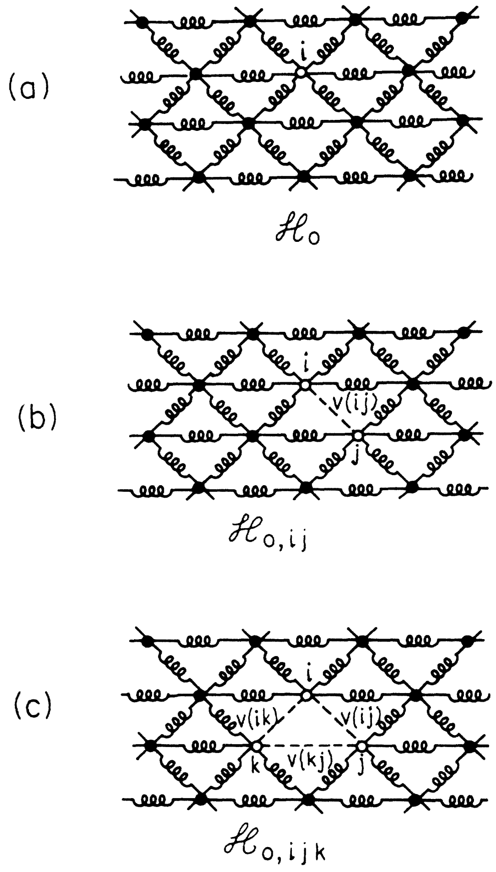

FIG. 3. Springs: (a) In the lowest approximation all of the particles are coupled together by springs. (b) For a pair of particles (ij) the spring $w_{ij}(ij)$ is replaced by the Lennard-Jones potential $v(ij)$. (c) For a triplet of particles (ijk) the springs $w_{ij}(ij)$, $w_{jk}(jk)$, and $w_{ki}(ki)$ are replaced by the Lennard-Jones potentials $v(ij)$, $v(jk)$, and $v(ki)$.

$$\Phi_{ij}(ij)[T(i)+T(j)+v(ij)$$

$$+m^{-1}\vec{\mathrm{p}}(ij)\ln\Phi_{ij}(ij)\cdot\vec{\mathrm{p}}(ij)\ln g_{ij}(ij)]$$

$$\times g_{ij}(ij)=(\epsilon_{ij}-2\epsilon_0)g_{ij}(ij), \tag{47}$$

where $\Phi_{ij}(ij)=\varphi_i(i)\varphi_j(j)$, and $\vec{\mathrm{p}}(ij)$ is a 6-component momentum vector $\vec{\mathrm{p}}(ij)=[p(i)_x, p(i)_y, p(i)_z, p(j)_x, p(j)_y, p(j)_z]$. For computational convenience let us assume that $\varphi_i(i)$ is a Gaussian;

$$\varphi_i(i)=A\exp[-\tfrac{1}{2}\alpha^2(\vec{\mathrm{x}}_i-\vec{\mathrm{R}}_i)^2] \ .$$

Then,

$$\vec{\mathrm{p}}(ij)\Phi_{ij}(ij)\cdot\vec{\mathrm{p}}(ij)=-a^2[\vec{\mathrm{u}}_i\cdot\vec{\mathrm{p}}(i)+\vec{\mathrm{u}}_j\cdot\vec{\mathrm{p}}(j)],$$

and we have

$$\vec{\mathrm{p}}(ij)\ln\Phi_{ij}(ij)\cdot\vec{\mathrm{p}}(ij)\ln g_{ij}(ij)$$

$$=-\tfrac{1}{2}\alpha^2(\vec{\mathrm{r}}-\vec{\Delta})\cdot\vec{\mathrm{p}}(r)g_{ij}(ij)$$

$$-2\alpha^2(\vec{\mathrm{R}}-\vec{\mathrm{d}})\cdot\vec{\mathrm{P}}(R)g_{ij}(ij) \ ,$$

where $\vec{\Delta}=\vec{\mathrm{R}}_i-\vec{\mathrm{R}}_j$, $\vec{\mathrm{d}}=\tfrac{1}{2}(\vec{\mathrm{R}}_i+\vec{\mathrm{R}}_j)$,

and $\vec{\mathrm{P}}(R)$ and $\vec{\mathrm{p}}(r)$ are the center of mass and relative coordinate momenta. Thus, Eq. (47) for $g_{ij}(ij)$ takes the form

$$[T(R)+T(r)+v(r)-\frac{i\hbar\alpha^2}{2m}(\vec{\mathrm{r}}-\vec{\Delta})\cdot\vec{\mathrm{p}}(r)\ (2i\hbar\alpha^2/m)$$

$$\times(\vec{\mathrm{R}}-\vec{\mathrm{d}})\cdot\vec{\mathrm{P}}(R)]\,g_{ij}(ij)=(\epsilon_{ij}-2\epsilon_0)g_{ij}(ij) \ .$$

The equation of motion for g separates into the center of mass and relative coordinate parts. The important thing which $g_{ij}(ij)$ is supposed to do is to describe the relative motion of i and j due to $v(r)$. We write $g_{ij}(ij)=g(\vec{\mathrm{r}};\vec{\Delta})G(\vec{\mathrm{R}};\vec{\mathrm{d}})$ and set $G(\vec{\mathrm{R}};\vec{\mathrm{d}})$ equal to 1. The equation of motion for $g(\vec{\mathrm{r}};\vec{\Delta})$ is

$$[T(r)+v(r)+(\hbar^2\alpha^2/2m)(\vec{\mathrm{r}}-\vec{\Delta})\cdot\nabla_r]g(\vec{\mathrm{r}};\vec{\Delta})$$

$$=\Delta\epsilon(\vec{\Delta})g(\vec{\mathrm{r}};\vec{\Delta}) \ . \tag{48}$$

This equation of motion is almost the equation of relative motion for two free particles interacting through $v(r)$. However, $g(\vec{\mathrm{r}};\vec{\Delta})$ is coupled through the third term, $(\hbar^2\alpha^2/2m)(\vec{\mathrm{r}}-\vec{\Delta})\cdot\nabla_r$, to the relative motion due to $\Phi_{ij}(ij)$. We will refer to the third term as the coupling term. It arises because of the product form of $\psi_{ij}(ij)$ and because the localization in $\Phi_{ij}(ij)$ constrains the relative motion of i and j,

$$\Phi_{ij}(ij)=\varphi(\vec{\mathrm{r}};\vec{\Delta})\varphi(\vec{\mathrm{R}},\vec{\mathrm{d}}) \ ,$$

$$\varphi(\vec{\mathrm{r}};\vec{\Delta})=\exp[-\tfrac{1}{4}\alpha^2(\vec{\mathrm{r}}-\vec{\Delta})^2] \ .$$

The coupling term is important in determining the asymptotic form of $g(\vec{\mathrm{r}};\vec{\Delta})$.

The coupling term depends upon the angle between $\vec{\mathrm{r}}$ and $\vec{\Delta}$. We argue that because of the localization of i and j near R_i and R_j, respectively, the important relative coordinate vectors are along the line $\vec{\mathrm{r}}\parallel\vec{\Delta}$ (see Fig. 4). Thus, we argue that (a) $g(r)$ may be taken to depend upon $|\vec{\mathrm{r}}|=r$ only and (b) the equation of motion for $g(r)$ should be solved one dimensionally along the line $\vec{\mathrm{r}}\parallel\vec{\Delta}$. In the work of Guyer and Sarkissian,[13] these two assumptions have been checked. They lead to a good first approximation. Corrections due to making them are small, nonetheless these corrections have been included in the detailed computational use of Eq. (48). Using assumptions (a) and (b) we have

$$\left(-\frac{\hbar^2}{m}\frac{d^2}{dr^2}+v(r)+\frac{\hbar^2\alpha^2}{2m}(r-\Delta)\frac{d}{dr}\right)g(r;\Delta)$$

$$=\Delta\epsilon(\Delta)g(r;\Delta) \ , \tag{49}$$

where $\Delta\epsilon(\Delta)=\epsilon_{ij}-2\epsilon_0$. As $r\to 0$ we have the WKB solution for the pair of particles in the hard core of $v(r)$,

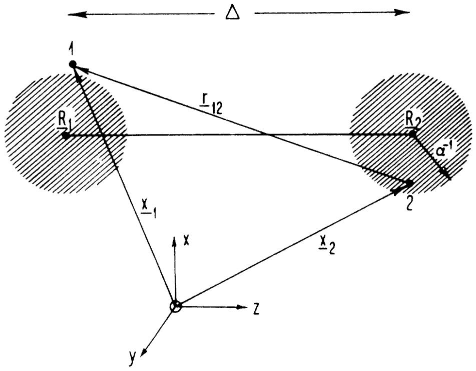

FIG. 4. Relative motion: The shaded area around $\vec{\mathrm{R}}_1$ is the region of space where particle 1 spends most of its time. Likewise, the shaded region around $\vec{\mathrm{R}}_2$ is the region of space in which particle 2 is found. The vector $\vec{\mathrm{r}}_{12}$ is most probably parallel to Δ. The relative motion of particles 1 and 2 is approximately one dimensional because each is localized in the vicinity of a lattice site.

$$g(r;\Delta)\sim\exp[-\beta(\sigma/r)^5],$$

$$\beta=\tfrac{2}{5}(\sigma/\hbar)(m\epsilon)^{1/2}\approx 1\quad .$$

As $r\rightarrow+\infty$ the coupling term determines the asymptotic form;

$$g(r;\Delta)\sim r^{\delta},\qquad \delta=3\Delta\epsilon(\Delta)/\epsilon_0\quad ,$$

where $\epsilon_0=3\hbar^2\alpha^2/2m$ and $\Delta\epsilon(\Delta)\approx v(\Delta)<0$; $\delta\rightarrow 0^-$ as $\Delta\rightarrow+\infty$. Thus $g(r;\Delta)$ has the form shown in Fig. 5. The details of $g(r;\Delta)$ for a particular equilibrium spacing Δ depends weakly upon Δ for small r and large r. We will often refer to $g(r,\Delta)$ or $g_{ij}(ij)$ as the correlation function. We do this because it is that part of $\psi_{ij}(ij)$ which measures the short-range correlation in the motion of i and j. The true pair correlation function can be related to the set of $g_{ij}(ij)$ by a cluster-expansion calculation.

Further on in this paper it will be necessary to understand the physics which determines the structure of $g_{ij}(ij)$. Our purpose above has been to lay the ground work for this understanding.

Within the context of the pair approximation we have been examining let us consider the exchange process. Particle 1 can be in the vicinity of lattice site R_2 as well as lattice site R_1 and the same for particle 2. We must make a fundamental change in our description of the pair of particles. The Hamiltonian in Eq. (46) must be replaced by

$$T(1)+T(2)+U_{12}(1)+U_{12}(2)+v(12)\quad ,$$

where $U_{12}(1)$ is a single-particle potential for particle 1 in the vicinity of R_1 and R_2 due to the lattice medium. We expect $U_{12}(1)\approx\bar{U}_1(1)$, for $\vec{x}_1\approx\vec{R}_1$; $U_{12}(1)\approx\bar{U}_2(1)$, for $\vec{x}_1\approx\vec{R}_2$ (see Fig. 6). The ground-state pair wave function for i and j must

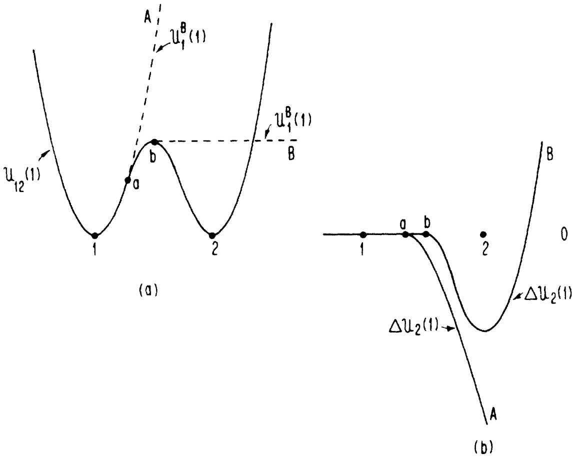

FIG. 6. Double well I: Particles 1 and 2, which are in the vicinity of $\vec{R}_1$ and $\vec{R}_2$, see a double well in this region of space due to the lattice medium. Two choices of the biased potentials $U_1^B(1)$ (A and B) are shown on (a) as dashed lines. These two biased potentials give rise to 2 potential differences $\Delta U_2(1)$ illustrated in (b). The biased potentials localize a particle near one of the lattice sites. The potential differences $\Delta U_2(1)$ are negative in the region of space near the other lattice site.

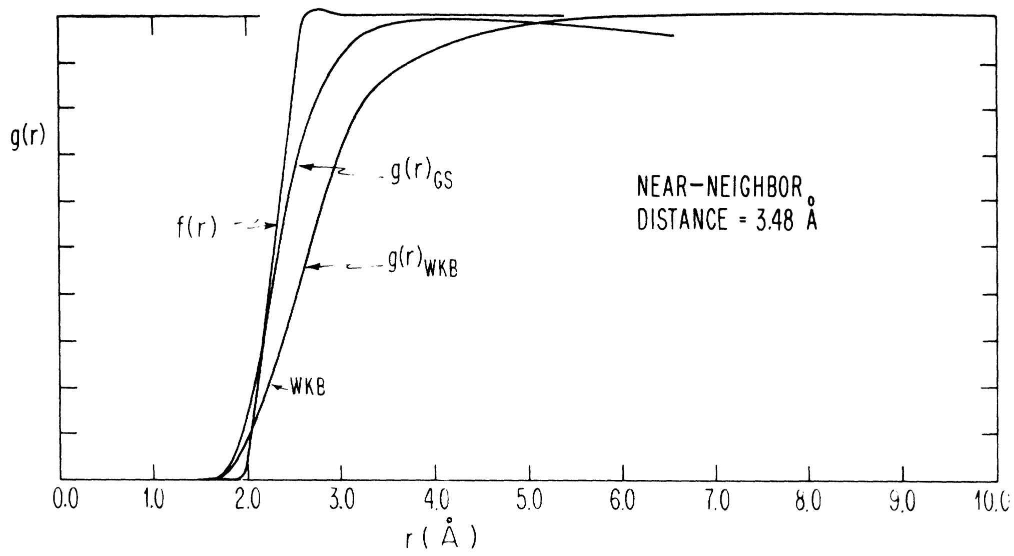

FIG. 5. Correlation functions: The functions $f(r)$, $g(r)_{\rm gs}$, and $g(r)_{\rm WKB}$ are the correlation functions of Nosanow (Ref. 6), Guyer and Sarkissian (Refs. 12 and 13), and the WKB correlation function. The common characteristic of all of these functions is that they go rapidly to zero for $r<\sigma$, i.e., as the particles penetrate one another's hard core. The numerical calculations of $\Delta\epsilon_T$ and $\Delta\epsilon_x$ discussed in Sec. V were done with these three correlation functions.

solve the equation

$$[T(1)+T(2)+U_{12}(1)+U_{12}(2) + v(12)]\psi_{12}^{\pm}(12)=\epsilon_{12}^{\pm}\psi_{12}^{\pm}(12) \quad , \qquad (50)$$

where the ± indicate that there are two nearly degenerate solutions to Eq. (50) which correspond to having the spins of the 2 particles parallel or antiparallel. We have $\psi_{12}^{\pm}(12)=\pm\psi_{12}^{\pm}(21)$. The Hamiltonian given by Eq. (50) is of the kind which is used in orthogonalization scheme 2. Equation (50) can be derived using Rayleigh-Schrödinger perturbation theory in scheme 2 in the same way it was used in Sec. II to obtain Eq. (29) in scheme 1. It is plausible to expect to construct an approximation to $\psi_{12}^{\pm}(12)$ in the form

$$\psi_{12}^{\pm}(12)=[\psi_{12}(12)\pm\psi_{12}(21)]/\sqrt{2} \quad , \qquad (51)$$

where $\psi_{12}(12)$ solves Eq. (46). These wave functions are spatially symmetric and antisymmetric, respectively, and belong to the spin states

$$\left(\delta_{\sigma_1\downarrow}\delta_{\sigma_2\uparrow}-\delta_{\sigma_1\uparrow}\delta_{\sigma_2\downarrow}\right)\leftrightarrow\psi^{+};$$

$$\left\}\begin{matrix}\delta_{\sigma_1\uparrow}\delta_{\sigma_2\uparrow}\\ \delta_{\sigma_1\downarrow}\delta_{\sigma_2\downarrow}\\ \delta_{\sigma_1\uparrow}\delta_{\sigma_2\downarrow}+\delta_{\sigma_1\downarrow}\delta_{\sigma_2\uparrow}\end{matrix}\right\}\leftrightarrow\psi^{-} \quad .$$

Let us attempt to find $\epsilon^{+}-\epsilon^{-}$. We can find an expression for the energy difference $\epsilon^{+}-\epsilon^{-}$ by using the following generalization of a standard procedure. The wave functions $\psi_{12}(12)$ and $\psi_{12}(21)$ which we add together in the superposition approximation in Eq. (51) are not exact solutions to Eq. (50), since, in $\psi_{12}(12)$, we constrain 1 and 2 to remain in the R_1 and R_2 sides of the double well, respectively. But $\psi_{12}(12)$ exactly satisfies the Schrödinger equation

$$h_{12}^{B}(12)\psi_{12}(12)=[T(1)+T(2)+U^{B}(1) + U^{B}(2)+v(12)]\psi_{12}(12)=\epsilon_{12}\psi_{12}(12) \quad , \qquad (52)$$

where $U_1^{B}(1)$ and $U_2^{B}(2)$ are biased single-particle potentials (see Fig. 6). $U_1^{B}(1)$ is so constructed as to reproduce exactly $U_{12}(1)$ in the region of space near R_1. It differs from $U_{12}(1)$ only in the vicinity of R_2. We have

$$[T(i)+U_i^{B}(i)]\varphi_i(i)=\epsilon_0\varphi_i(i) \quad . \qquad (53)$$

Now $h_{12}^{B}(12)\psi_{12}(12)=\epsilon_{12}\psi_{12}(12)$

and $h_{12}^{B}(21)\psi_{12}(21)=\epsilon_{12}\psi_{12}(21)$,

where $h_{12}^{B}(12)=T(1)+T(2)+U_1^{B}(1)+U_2^{B}(2)+v(12)$. Consider finding $\epsilon_{12}^{\pm}$ from Eq. (50),

$$h(12)\psi_{12}^{\pm}(12)=[T(1)+T(2)+U_{12}(1) + U_{12}(2)+v(12)]\psi_{12}^{\pm}(12)=\epsilon_{12}^{\pm}\psi_{12}^{\pm}(12).$$

As an approximation to $\psi_{12}^{\pm}(12)$ use

$$\psi_{12}^{\pm}(12)=[\varphi_1(1)\varphi_2(2)\pm\varphi_1(2)\varphi_2(1)](\sqrt{2})^{-1}g(r_{12}) = (1\pm X)(\sqrt{2})^{-1} \quad . \qquad (54)$$

We use the schematic notation 1 for $\varphi_1(1)\varphi_2(2)g(r_{12})$ and X for $\varphi_1(2)\varphi_2(1)g(r_{12})$. From Eq. (50) we have

$$\epsilon_{12}^{\pm}=\frac{\langle 1\pm X|h(12)|1\pm X\rangle}{\langle 1\pm X|1\pm X\rangle} =\frac{\langle 1|h(12)|1\rangle\pm\langle X|h(12)|1\rangle}{\langle 1|1\rangle\pm\langle X|1\rangle} \quad .$$

We may write $h(12)=h_{12}^{B}(12)+\Delta U_2(1)+\Delta U_1(2)$, where $\Delta U_2(1)=U_{12}(1)-U_1^{B}(1)$ [see Fig. 6(b)]. Then we have

$$\epsilon_{12}^{\pm}=\epsilon_{12}+2\frac{\langle 1|\Delta U_2(1)|1\rangle}{\langle 1|1\rangle}\pm2\frac{\langle X|\Delta U_2(1)|\rangle}{\langle 1|1\rangle} \quad . \qquad (55)$$

The first term is the pair energy before symmetrization, the second term is a small energy shift which is the same[27] for ϵ_{12}^{+} and ϵ_{12}^{-}, and the third term has the opposite sign for ϵ_{12}^{+} and ϵ_{12}^{-}. The third term is negative, since $\Delta U_2(1)$ is always negative. Thus, even before scrutinizing this term in detail we know that $\epsilon_{12}^{+}<\epsilon_{12}^{-}$. It is easy to understand the meaning of this result. The quantity $\langle X|\Delta U_2(1)|1\rangle$ is the reduction in potential energy which particle 1 of a symmetric pair sees in the vicinity of lattice site R_2 because there is a lower potential energy in the vicinity of this lattice site when we have $U_{12}(1)$ in place of $U_1^{B}(1)$.

The above calculations are tunneling calculations. The energy difference $\epsilon_{12}^{+}-\epsilon_{12}^{-}=\Delta\epsilon_T$ is twice ω_T, a tunneling frequency. In Sec. V, we discuss in detail the meaning of this result, estimate ω_T, and suggest a simple model for computing the tunneling frequency.

The magnitude of $\Delta\epsilon_T$ given by Eq. (55) depends on the bare interaction between i and j through $g(r_{ij})$. However, the sign of $\Delta\epsilon_T$ depends in no way upon $g(r_{ij})$ and, therefore, in no way on $v(r_{ij})$. This result is not a consequence of the model of the pair problem that is implied by Eq. (50) but rather follows from the assumptions we have made about the form of the pair wave function in treating this equation, i.e., in writing $\psi_{12}^{\pm}(12)$ in the form given by Eq. (54).

To learn the effect of the interaction process on the energetics which determine the magnetism of the system we look at two simple model systems which illustrate useful limiting cases.

Case 1. A pair of noninteracting particles in a double well. Consider a pair of noninteracting particles in the double well of Fig. 6(a). In the limit where the barrier is large enough that the particles are to first approximation localized on one or the other side of the well, the ground state is degenerate.[28] This degenerate ground state for a single particle is split by the finite probability of tunneling through the barrier. We have

$$\psi^{\pm}(1) = \varphi_1(1) \pm \varphi_2(1)$$

with the corresponding energies $\epsilon^{\pm}$ which are approximately

$$\epsilon^{\pm} = \epsilon_0 \mp t^2/\epsilon_0 \ ,$$

where t is a tunneling matrix element. The ground state for a pair of noninteracting fermions in the double well has wave function

$$\psi^{+}(12) = \psi^{+}(1)\psi^{+}(2)$$

and energy $2\epsilon^{+}$. If the particles are fermions, the ground state is antiferromagnetic. The first excited state is ferromagnetic, with energy

$$\epsilon = \epsilon^{+} + \epsilon^{-} = 2\epsilon_0 \ .$$

Quite generally a pair of noninteracting fermions in a double well have an antiferromagnetic ground state. It is tunneling through the barrier in the double well which yields this result.

Case 2. A pair of free fermions having a strong repulsive short-ranged interaction. Consider a pair of Fermi particles in a box with dimensions large compared to the fundamental length in the interaction between them. For the pair we can construct two free-particle states

$$\psi^{\pm}_{\vec{k}\vec{k}'} = (e^{i\vec{k}\cdot\vec{x}_1} e^{i\vec{k}'\cdot\vec{x}_2} \pm e^{i\vec{k}\cdot\vec{x}_2} e^{i\vec{k}'\cdot\vec{x}_1})$$

corresponding to wave vectors $\vec{k}$, $\vec{k}'$. Using the center of mass $\vec{R} = \frac{1}{2}(\vec{x}_1 + \vec{x}_2)$ and relative coordinate $\vec{r} = \vec{x}_1 - \vec{x}_2$ we can write

$$\psi^{\pm}_{\vec{k}\vec{k}'} = \psi^{\pm}_{\vec{K}\vec{\kappa}} = 2e^{i\vec{K}\cdot\vec{R}} \begin{pmatrix} \cos\vec{\kappa}\cdot\vec{r} \\ i\sin\vec{\kappa}\cdot\vec{r} \end{pmatrix} ,$$

where $\vec{K} = \frac{1}{2}(\vec{k} + \vec{k}')$ and $\vec{\kappa} = \vec{k} - \vec{k}'$. The expectation value of $v(r)$ for these two states is

$$V^{\pm} = \int d\vec{r}\, v(r) |\psi^{\pm}_{\vec{K}\vec{\kappa}}|^2 / \int d\vec{r}\, |\psi^{\pm}_{\vec{K}\vec{\kappa}}|^2 \ .$$

In the limit $\kappa\sigma \gg 1$ [σ is the range of $v(r)$], we have

$$V^{\pm} \approx V_0 \begin{pmatrix} 1 \\ \kappa^2\sigma^2 \end{pmatrix},$$

where $v(r)$ is approximated by $v(r) = V_0\theta(\sigma - r)$. Thus for $V_0 > 0$, the space antisymmetric or ferromagnetic state is the ground state for the pair. For $V_0 < 0$, the space symmetric or antiferromagnetic state is the ground state of the pair. The reason for this dependence of the magnetism on the sign of the interaction is quite simple. When the interaction is strongly repulsive, the pair chooses the space antisymmetric (spin parallel) wave function which is small at $r \to 0$ in order to minimize the potential energy. The space symmetric wave function (spin antiparallel) is large at $r \to 0$ and the opposite argument holds.

The purpose of looking at Case 1 and Case 2 is to expose the elementary physical mechanisms operating between a pair of Fermi particles which lead to magnetism, i.e., which lead to an energetic difference between a parallel spin pair and an antiparallel spin pair.

We have: (a) Tunneling. Because more than one single-particle well is accessible to a particle by tunneling, the energy of the state (↑↓) is lowered relative to the energy of the state (↑↑). (b) Interaction. Depending on the detailed nature of $v(r)$, a pair of particles may choose either the state (↑↓) or the state (↑↑) as the lower-energy state. For a positive hard-core interaction, the state (↑↑) has lower energy than the state (↑↓).

We take particular note of the fact that (Case 2) the interaction is capable of changing the sign of the magnetism. Why is there no evidence in Eq. (35) for the dependence of the sign of $\Delta\epsilon_T$ on the nature of $v(r)$? This is because the calculation, as we have carried it out above, has not correctly treated the interaction process. In Sec. IV, we will attempt to show how we must modify the above approach to include it.

IV. INTERACTION

Let us consider the solution to Eq. (50) when we write the pair wave functions in the form

$$\psi_{12}^{\pm}(12) = \Phi_{12}^{\pm}(12) g_{12}(12)^{\pm} = [\varphi_1(1)\varphi_2(2) \pm \varphi_1(2)\varphi_2(1)] (\sqrt{2})^{-1} g_{12}(12)^{\pm} \tag{56}$$

The motivation for writing $g_{12}^{\pm}(12)$ in place of $g_{12}(12)$ is that $g_{12}(12)$ is the manifestation of the influence of $v(r)$ on the relative motion of 1 and 2. This relative motion is coupled to the relative motion in $\Phi_{12}^{\pm}(12)$. If there is no feature of the relative motion of 1 and 2 due to $v(r_{12})$ which depends upon the symmetry of the pair wave function, it is hard to see how $v(r_{12})$ can have an influence of the sign of $\epsilon_{12}^{\pm}-\epsilon_{12}^{-}$. Thus, we argue that such influence as there is must appear through the coupling of $g_{12}(12)$ to $\Phi_{12}^{\pm}(12)$ and manifest itself as $g_{12}^{\pm}(12)$. This argument requires that we make a distinction between the present situation and that dealt with in Case 2 above. The motion which is important for determining the interplay of symmetry and $v(r)$ is the relative motion of the pair of particles. In Case 2, we symmetrized the plane waves $e^{i\vec{k}\cdot\vec{x}_1}$ and $e^{i\vec{k}'\cdot\vec{x}_2}$ to obtain two relative motion wave functions $\psi_{\vec{k}\vec{k}'}^{\pm}$ and calculated the expectation value of $v(r)$ using these wave functions. We can expect this procedure to work adequately so long as the relative motion of 1 and 2 is principally determined by $\psi_{\vec{k}\vec{k}'}^{\pm}$; i.e., so long as $v(r)$ is sufficiently weak that the relative motion of 1 and 2 embodied in $\psi_{\vec{k}\vec{k}'}^{\pm}$ is not seriously distorted by $v(r)$. In the case of two interacting ^{3}He atoms the Lennard-Jones potential dramatically distorts the relative motion of a pair of particles. Of course, it does this precisely in the region of space $(r_{12}\approx\sigma)$ where the interaction between 1 and 2 is most important. Thus, we cannot adequately treat the interaction process for a pair of strongly interacting particles by assuming that the symmetrization of the wave function need involve only the localized part.

Let us substitute Eq. (56) into Eq. (50) and find the resulting equation of motion for $g_{12}^{\pm}(12)$. After several straightforward manipulations we obtain

$$\begin{aligned}\Phi_{12}(12)[H(12)+m^{-1}\vec{p}(12)\ln\Phi_{12}(12)\cdot\vec{p}(12)\ln g_{12}^{\pm}(12)]g_{12}^{\pm}(12)\pm\Phi_{12}(21)[H(12)\\ +m^{-1}\vec{p}(12)\ln\Phi_{12}(21)\cdot\vec{p}(12)\ln g_{12}^{\pm}(12)]g_{12}^{\pm}(12)\\ =(\epsilon_{12}^{\pm}-2\epsilon_0)[\Phi_{12}(12)\pm\Phi_{12}(21)]\,g_{12}^{\pm}(12)\\ +[\Delta U_2(1)+\Delta U_1(2)]\,\Phi_{12}(12)g_{12}^{\pm}(12)\pm[\Delta U_1(1)+\Delta U_2(2)]\,\Phi_{12}(21)g_{12}^{\pm}(12)\quad,\end{aligned}\tag{57}$$

where $H(12)=T(1)+T(2)+v(12)$ and we have used Eq. (52). On the right-hand side of this equation we have two terms which are the potential energy manifestation of the tunneling process. We replace these terms by their expectation value.

Since $\langle 1\pm X|\,\Delta U_2(1)+\Delta U_1(2)\,|1\rangle=\pm\langle 1\pm X|\,\Delta U_1(1)+\Delta U_2(2)\,|X\rangle$,

we make the replacement

$$[\Delta U_2(1)+\Delta U_1(2)]\,|1\rangle\pm[\Delta U_1(1)+\Delta U_2(2)]\,|X\rangle=(\epsilon_D\pm\tfrac{1}{2}\Delta\epsilon_T)(1\pm X)\ ,$$

where $\epsilon_D=2\langle 1|\,\Delta U_2(1)\,|1\rangle/\langle 1|1\rangle$

and $\Delta\epsilon_T=4\langle X|\,\Delta U_2(1)\,|1\rangle/\langle 1|1\rangle$.

Equation (57) now has the simple form

$$\begin{aligned}\Phi_{12}(12)[H(12)+m^{-1}\vec{p}(12)\ln\Phi_{12}(12)\cdot\vec{p}(12)\ln g_{12}^{\pm}(12)]g_{12}^{\pm}(12)\pm\Phi_{12}(21)[H(12)\\ +m^{-1}\vec{p}(12)\ln\Phi_{12}(21)\cdot\vec{p}(12)\ln g_{12}^{\pm}(12)]\,g_{12}^{\pm}(12)=(\epsilon_{12}^{\pm}+\epsilon_D\pm\tfrac{1}{2}\Delta\epsilon_T-2\epsilon_0)[\Phi_{12}(12)\pm\Phi_{12}(21)]g_{12}^{\pm}(12)\ .\end{aligned}\tag{58}$$

There are two terms on the left-hand side of this equation. The first of these, which is multiplied by $\Phi_{12}(12)$, dominates the second when particle 1 is near lattice site R_1 and particle 2 is near lattice site R_2. It is convenient when dealing with this equation to look at its behavior when the interacting particles are in various regions of real space and of relative coordinate space. By relative coordinate space we mean the space of $\vec{r}_{12}=\vec{x}_1-\vec{x}_2$ (see Fig. 7). When particle 1 is near lattice site R_1 and particle 2 is near lattice site R_2, we have $\vec{r}_{12}$ parallel to $\vec{R}_1-\vec{R}_2$ and in the direct region of r_{12} space. When particle 1 is near lattice site R_2 and particle 2 is near lattice site R_1, we have $\vec{r}_{12}$ parallel to $\vec{R}_2-\vec{R}_1$ and in the exchange region of r_{12} space. To see what Eq. (58) looks like in the direct region of space we multiply from the left by $\Phi_{12}(12)^{-1}$ and obtain

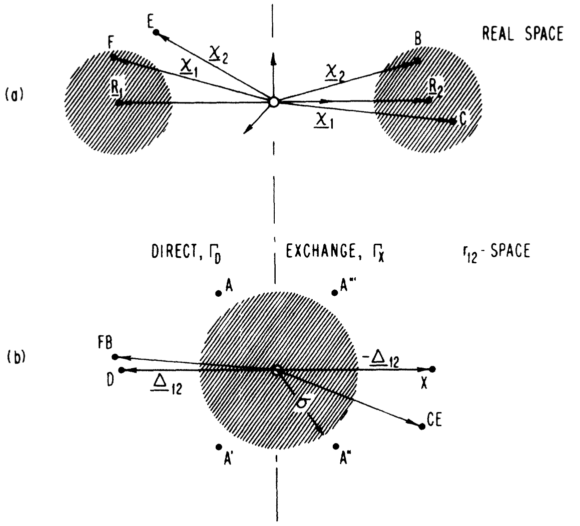

FIG. 7. Real space and $\vec{r}_{12}$ space: The vector $\vec{r}_{12}$ is in the direct part of $\vec{r}_{12}$ space when particle 1 is near $\vec{R}_1$ and particle 2 is near $\vec{R}_2$, e.g., $\vec{x}_1$ at F and $\vec{x}_2$ at B, $\vec{r}_{12}$ at FB. For the exchanged case (particle 1 near $\vec{R}_2$ and particle 2 near $\vec{R}_1$) $\vec{r}_{12}$ is in the exchange space e.g., $\vec{x}_1$ at C and $\vec{x}_2$ at E, $\vec{r}_{12}$ at CE. The equation for $g(r)$ is solved along the line DO. The important region of $\vec{r}_{12}$ space is on the border of the shaded region along the line DO where $r_{12} \approx \sigma$.

$$(1 \pm t_D)[H(12) + m^{-1}\vec{p}(12)\ln\Phi_{12}(12)\cdot\vec{p}(12)\ln g_{12}^{\pm}(12)]\, g_{12}^{\pm}(12)$$

$$\pm t_D\left(\frac{\vec{p}(12)}{m}\ln t_D\cdot\vec{p}(12)\ln g_{12}^{\pm}(12)\right) g_{12}^{\pm}(12) = (1 \pm t_D)E^{\pm} g_{12}^{\pm}(12) \ , \tag{59}$$

where $E^{\pm} = \epsilon_{12}^{\pm} + \epsilon_D \pm \frac{1}{2}\Delta\epsilon_T - 2\epsilon_0$, and $t_D = \Phi_{12}(12)^{-1}\Phi_{12}(21)$

is given by $t_D = \exp(-\alpha\vec{\Delta}_{12}\cdot\vec{r}_{12})$

in the Gaussian approximation to the single-particle wave functions. In the direct space, $\vec{\Delta}_{12}\cdot\vec{r}_{12} \geqslant 0$. When the pair of particles are at the distance of closest approach,

$$|\vec{r}_{12}| \approx \sigma, \quad t_D \approx e^{-\alpha^2\Delta\sigma} \approx e^{-13} \ .$$

On Fig. 7, we have shaded a region near the origin of approximate radius σ. A pair of particles have a hard time getting inside the shaded region due to $v(r_{12})$; $g_{12}(r_{12})$ vanishes rapidly as r_{12} becomes less than σ.

The behavior of $g_{12}(12)^{\pm}$ in the direct region of space (for example, along the line between D and O, DO) is principally determined by the first term in Eq. (59). In fact, if the second term in Eq. (59), proportional to t_D, is ignored we have $g_{12}^{+}(12) = g_{12}^{-}(12)$. This is the approximation which leads to tunneling only. The second term in Eq. (59) is the overlap term; it is a measure of the effect of the overlap of $\Phi_{12}(12)$ and $\Phi_{12}(21)$ on the relative motion of the particles in the direct space. As remarked above, t_D is a very small quantity. However, so is the energy shift due to tunneling. We must consider the effect the overlap term has on the equation of motion for $g_{12}^{\pm}(12)$ along DO and on the energy eigenvalue $E^{\pm}$. We can show that the equation of motion for $g_{12}^{\pm}(12)$ in the direct space is exactly the same as the equation of motion in the exchange space, i.e., $g_{12}^{\pm}(12)$ is the same function at points A, A', A'', and A''' in r_{12} space. Thus, we need look in detail only at the equation in direct space to understand its behavior everywhere.

We want to estimate the energy shift due to the t_D term which couples $g_{12}^{\pm}(12)$ to the overlap of $\Phi_{12}(12)$ and $\Phi_{12}(21)$. Let us calculate the expectation value of the coupling term. We have

$$\pm t_D\left(\frac{\vec{p}(12)}{m}\ln t_D\cdot\vec{p}(12)\ln g_{12}^{\pm}(12)\right) g_{12}^{\pm}(12) \approx \pm t_D \frac{\hbar^2\alpha^2}{2m}\frac{\vec{\Delta}\cdot\vec{r}_{12}}{r_{12}}\frac{g'(r_{12})}{g(r_{12})} \ .$$

In the direct region of space the term contributes an energy which we can estimate by multiplying it by $\Phi_{12}(12)^2$ and integrating over $x_{12} < 0$. Since $\Phi_{12}(12)^2 t_D = \Phi_{12}(12)\Phi_{12}(21)$, we obtain

$$\pm(\hbar^2\alpha^2/m)\tfrac{1}{4}\Delta\int_{\Gamma_D} d\vec{r}\, g'(r)g(r)e^{-\alpha^2 r^2/2} \ ,$$

where a factor of $\frac{1}{2}$ is the average value of $\vec{\Delta}\cdot\vec{r}/r$ over the direct space and $\int_{\Gamma_D} d\vec{r}\cdots$ is the integral over the direct space Γ_D.

We can show that the expectation value of the overlap term in exchange space is the same as that above except that we must integrate over Γ_X, the exchange space. Thus we have $E^{\pm} = \epsilon_{12} + \epsilon_D - 2\epsilon_0 \pm \frac{1}{2}\Delta\epsilon_T \pm \frac{1}{2}\Delta\epsilon_X$, where

$$\Delta\epsilon_X = +(\hbar^2\alpha^2/m)\tfrac{1}{2}\Delta \int d\vec{r}\, g'(r) g(r) e^{-\alpha^2 r^2/2} \quad . \tag{60}$$

We can easily determine the sign of $\Delta\epsilon_X$ by noting that $g(r)$ is given by something like $g(r) = \exp[-\gamma(r)]$, $g'(r) \approx -\gamma'(r)g(r)$, $\gamma(r) \approx (\sigma/r)^n$ and $\gamma'(r) = -(n/\sigma)(\sigma/r)\gamma(r)$; $\Delta\epsilon_X$ is positive. Thus the interaction process energy difference, $\Delta\epsilon_X$, is positive. As with the case of the tunneling process energy difference this sign is a consequence of quite general arguments. We may easily understand this positive energy shift. Due to the overlap of $\Phi_{12}(12)$ and $\Phi_{12}(21)$, there is more wave function in the vicinity of $r_{12} \approx \sigma$ for $\Phi_{12}(12) + \Phi_{12}(21)$ than there is for $\Phi_{12}(12) - \Phi_{12}(21)$. This extra wave function leads to positive interaction energy. Of course, this is the same argument as we made above in Case 2. But now a much more sophisticated mechanism has come into play. This extra wave function changes the relative motion of 1 and 2 through its coupling to $g_{12}(12)$.

The effect of the t_D term on the equation of motion for $g_{12}^{\pm}(12)$ is small. We will not bother to compute the corrections to the relative coordinate wave function due to this term. We only need the estimate of the energy shift due to it.

We may write the energy of a pair of particles in a double well and interacting with one another through $v(r_{12})$ in the form

$$E^{\pm} = \epsilon_{12} + \epsilon_D - 2\epsilon_0 \pm \tfrac{1}{2}[\Delta\epsilon_T + \Delta\epsilon_X] \quad ,$$

where $\Delta\epsilon_T$ and $\Delta\epsilon_X$ are given by

$$\Delta\epsilon_T = 4\langle X|\Delta U_2(1)|1\rangle = 4\int d\vec{r}\,\Delta U_2(1) g(r)^2 e^{-\alpha^2 r^2/2} \tag{61}$$

and $$\Delta\epsilon_X = (\hbar^2\alpha^2/m)\tfrac{1}{2}\Delta \int d\vec{r}\, g'(r) g(r) e^{-\alpha^2 r^2/2} \quad . \tag{62}$$

The choice of magnetic ground state for the system will depend upon the balance of $\Delta\epsilon_T$ and $\Delta\epsilon_X$. We note that although both $\Delta\epsilon_T$ and $\Delta\epsilon_X$ depend upon the overlap integral, they are sensitive to two quite different features of the system. $\Delta\epsilon_T$ depends upon the average of the lattice medium potential over the overlap trajectory,[29] whereas $\Delta\epsilon_X$ depends upon the behavior of $v(r)$ over the overlap trajectory.

As we would expect as $v(r) \to 0$, $g(r) \to 1$, and $g'(r) \to 0$. In this limit $\Delta\epsilon_X$ vanishes as it should; $\Delta\epsilon_T$ remains finite and proportional to the uncorrelated overlap integral.

V. TUNNELING, INTERACTION ?

In this section, we will discuss the evaluation of the two integrals obtained above which give the magnitude of $\Delta\epsilon_T$ and $\Delta\epsilon_X$. The physics of the tunneling process and interaction process boils down to these two integrals; we will spend some time learning what they mean. Let us begin with a description of the physical content of the tunneling integral.

A. Tunneling Integral

This integral is given by Eq. (61),

$$\Delta\epsilon_T = 4\frac{\langle X|\Delta U_2(1)|1\rangle}{\langle 1|1\rangle} \approx 4\langle\Delta U_2(1)\rangle_{\text{ot}} \frac{\langle X|1\rangle}{\langle 1|1\rangle} , \tag{63}$$

where we can write the second step because $\Delta U_2(1)$ is a very mild function of $\vec{x}_1$ and $\vec{x}_2$; $\langle\Delta U_2(1)\rangle_{\text{ot}}$ means average of $\Delta U_2(1)$ over the important region of space for overlap, the overlap trajectory (ot) (see below). A discussion and justification of this procedure is given in Appendix B.

We have $\Delta\epsilon_T = 4\langle\Delta U_2(1)\rangle_{\text{ot}} P$,

where $$p = \int d\vec{x}_1 \int d\vec{x}_2\, \psi_{12}(12)\psi_{12}(21)/\langle 1|1\rangle \tag{64}$$

is the overlap integral. Large contributions to p come from regions of space where $\varphi_1(1)$ overlaps $\varphi_2(1)$ at the same time that $\varphi_2(2)$ overlaps $\varphi_1(2)$. Because of the factor $g_{12}(12)^2$ this overlap must occur with $\vec{x}_1$ and $\vec{x}_2$ separated by a distance on the order of σ, $|\vec{r}_{12}| \gtrsim \sigma$. When the single-particle wave functions are taken to be

Gaussians, the maximum overlap occurs in a spherical shell of approximate radius $\frac{1}{2}\sigma$ centered at the midpoint between R_1 and R_2. We illustrate this in Fig. 8. This region of space is called the "overlap trajectory." Equation (64) can be written in the form,

$$p=\left(\frac{2}{\pi}\right)^{1/2}\alpha^3 e^{-\alpha^2\Delta^2/2}\int_0^\infty r^2 g(r)^2 e^{-\alpha^2r^2/2}dr\,. \tag{65}$$

If we put $g(r)=1$ we have $p\approx e^{-\alpha^2\Delta^2/2}$, which results from having both $\varphi_1(1)$ overlap $\varphi_2(1)$, and $\varphi_2(2)$ overlap $\varphi_1(2)$, at the midpoint between R_1 and R_2, i.e.,

$$p=p_0\approx\varphi_1(\vec{x}_1=\tfrac{1}{2}\Delta)\varphi_2(\vec{x}_1=\tfrac{1}{2}\Delta)$$

$$\times\varphi_2(\vec{x}_2=\tfrac{1}{2}\Delta)\varphi_1(\vec{x}_2=\tfrac{1}{2}\Delta)=e^{-\alpha^2\Delta^2/2}\,.$$

For $g(r)$ behaving as a cutoff function at $\lambda\approx\sigma$, we have

$$p=\left(\frac{2}{\pi}\right)^{1/2}\alpha^3 e^{-\alpha^2\Delta^2/2}\int_x^\infty r^2 e^{-\alpha^2r^2/2}dr\,.$$

We may use the asymptotic formula[30]

$$\int_x^\infty e^{-t^2}dt=\frac{1}{2x}e^{-x^2}$$

and the approximation

$$\int_x^\infty t^2e^{-t^2}dt=x^2\int_x^\infty e^{-t^2}dt$$

to write p in the form

$$p=\left(\frac{2}{\pi}\right)^{1/2}\alpha\lambda e^{-\alpha^2\Delta^2/2}e^{-\alpha^2\lambda^2/2}\,.$$

Thus, with $g(r)$ present p is less than p_0 by a factor $e^{-\alpha^2\lambda^2/2}$ This extra factor arises because as the particles tunnel they are driven out of the straight-line trajectory between lattice sites by the hard-core interaction. They pass one another at relative distance λ (see Fig. 9). We have

$$p\approx\varphi_1(\vec{x}_1=d)\varphi_2(\vec{x}_1=d)\varphi_2(\vec{x}_2=d)\varphi_1(\vec{x}_2=d)$$

$$=e^{-\alpha^2(\Delta^2+\lambda^2)/2}\,,$$

where $d=[(\Delta/2)^2+(\lambda/2)^2]^{1/2}$

is the distance the particles are from their lattice sites when they pass one another. The tunneling integral is proportional to the overlap integral. The particle dynamics in the overlap integral describe the tunneling process. Let us reemphasize the most important point: Overlap does not occur at $r_{12}\to 0$; overlap occurs when $\varphi_1(1)$ and $\varphi_2(1)$ overlap and $\varphi_2(2)$ and $\varphi_1(2)$ overlap while $r_{12}\approx\sigma$.[31]

B. Interaction Integral

The interaction integral is given by

$$\Delta\epsilon_x=-\frac{\hbar^2\alpha^2}{m}\tfrac{1}{2}\Delta\int d\vec{r}\,g'(r)g(r)\,e^{-\alpha^2r^2/2}\,. \tag{66}$$

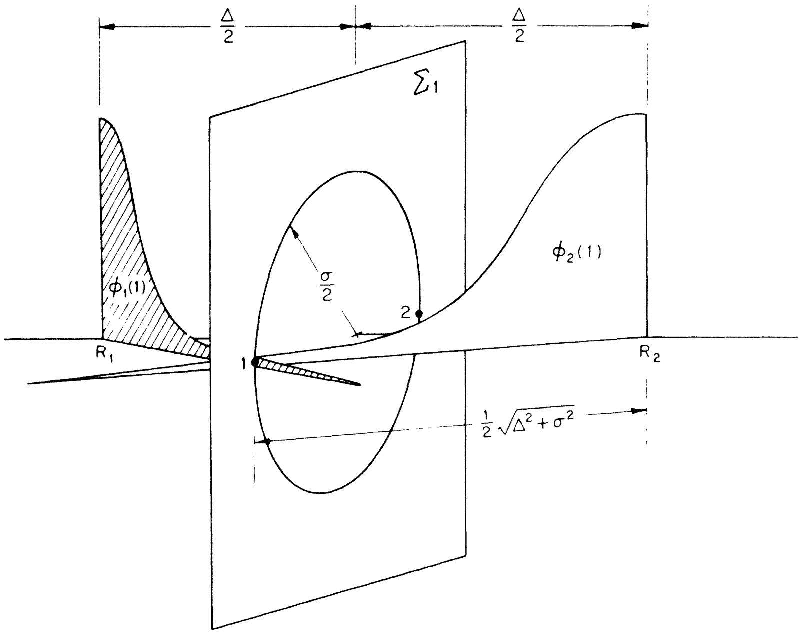

FIG. 8. The overlap trajectory: For Gaussian single-particle wave functions the maximum overlap occurs on a spherical surface at radius $\frac{1}{2}\sigma$ centered half-way between $\vec{R}_1$ and $\vec{R}_2$. This surface is at distances on the order of 0.6 Δ from the lattice sites.

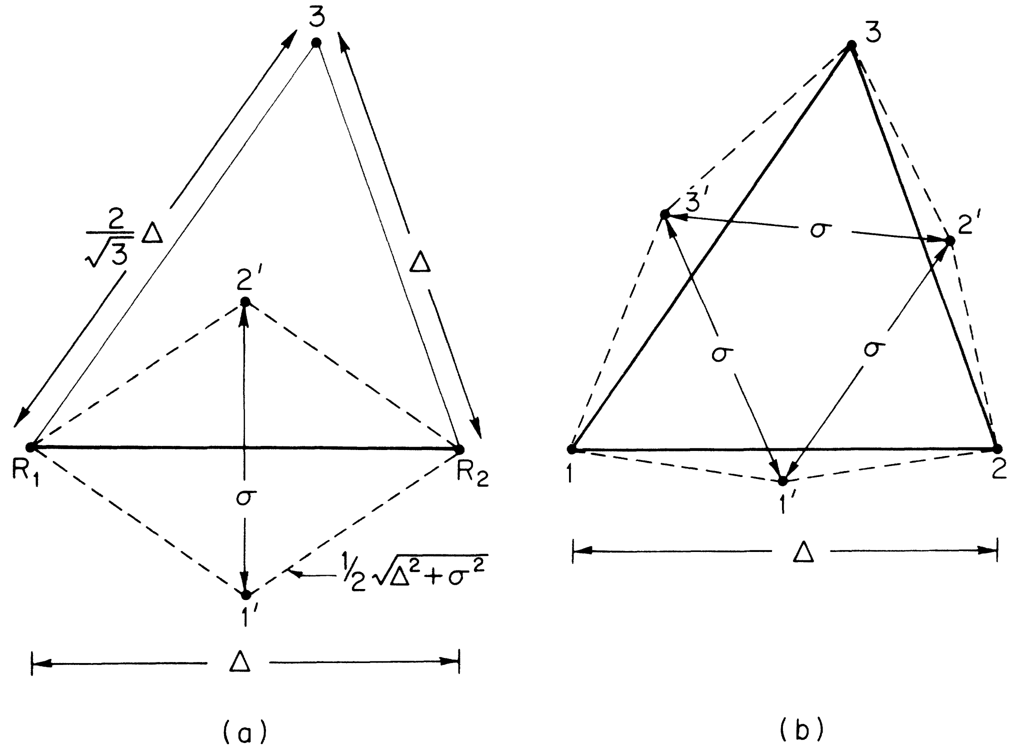

FIG. 9. Pair and triple exchange: (a) For pair exchange the particles must pass around one another at relative distance σ, $1 \rightarrow 1' \rightarrow 2$, $2 \rightarrow 2' \rightarrow 1$. (b) For triple exchange the particles cycle. They do not move far from the direct line of flight from one lattice site to another.

Since $g(r)$ behaves like

$$g(r) = \exp[-\gamma(r)], \tag{67}$$

where $\gamma(r) = \beta(\sigma/r)^n$

we expect $g'(r)$ to be largest in the vicinity of $r \approx \sigma$. Thus, $\Delta\epsilon_x$ is given by an integral like

$$\Delta\epsilon_x = \frac{\hbar^2\alpha^2}{m}\,\tfrac{1}{2}\Delta\int d\vec{r}\,\gamma'(r)g(r)^2 e^{-\alpha^2 r^2/2}.$$

In the spirit of the approximation we used in Eq. (63) we can write

$$\Delta\epsilon_x \approx \frac{\hbar^2\alpha^2}{m}\frac{\Delta}{2}\overline{\langle\gamma'(r)\rangle}_{\text{ot}}\,p, \tag{68}$$

where $\overline{\langle\gamma'(r)\rangle}_{\text{ot}}$ is $\gamma'(r)$ averaged over the overlap trajectory. Thus $\Delta\epsilon_x$ like $\Delta\epsilon_T$ is proportional to p. The physical description of the tunneling process given above applies equally well to the interaction process, except for the fact that the magnitude of the interaction process depends upon the potential energy the particles exert on one another as they pass near $x = 0$ at relative distance σ.

In Appendix A we discuss in detail the evaluation of Eqs. (61) and (62) for $\Delta\epsilon_T$ and $\Delta\epsilon_x$ using three different correlation functions. Here, we quote the results of that evaluation.

(1) For three correlation functions [(a) $g(r) = f(r)$ from the ground-state calculations of Nosanow and co-workers[6, 7]; (b) $g(r)$ from the ground-state calculations of Guyer and Sarkissian[12, 13]; and (c) $g(r) = \exp[-\beta(\sigma/r)^5]$, $\beta = 2/5\sigma \times (Vm\epsilon)^{1/2}/\hbar$, the WKB solution for $g(r)$ as $r \rightarrow 0$] we find $(\Delta\epsilon_T/\Delta\epsilon_x) \approx 2.5$ at all molar volumes from $V = 18.0$ to $24.7\ \text{cm}^3/\text{mole}$. These calculations were done with the values of α^2 and Δ shown in the table in Appendix A. The ratio $\Delta\epsilon_T/\Delta\epsilon_x$ is not strongly sensitive to the details of the single-particle wave function or the correlation function. This is demonstrated in Appendix A where we show, using quite general arguments, that $(\Delta\epsilon_T/\Delta\epsilon_x) \approx 4$ independent of α^2, Δ, and the correlation function.

(2) The magnitude of $\Delta\epsilon_T$ (or $\Delta\epsilon_x$) is most sensitive to how far into the hard core $g(r)$ permits a particle to go. In Fig. 5, we have plotted the three correlation functions which we have used in our calculations. It is clear that the Nosanow $f(r)$ permits the closest approach for a pair of particles; the WKB $g(r)$ keeps a pair furthest

apart. The magnitude of $\Delta\epsilon_T$ and $\Delta\epsilon_x$ calculated using Nosanow's $f(r)$ is about a factor of 2 greater than that obtained using the $g(r)$ of Guyer and Sarkissian. The magnitude of $\Delta\epsilon_T$ and $\Delta\epsilon_x$ calculated using $g(r)_{\mathrm{WKB}}$ is about a factor of 2 less than that obtained using the $g(r)$ of Guyer and Sarkissian. The rather wide range of variation in correlation functions shown in Fig. 5 leads to at most a factor of 4 in the behavior of $\Delta\epsilon_T$ and $\Delta\epsilon_x$.

Exchange in solid ^{3}He is principally due to a tunneling process. We can describe the effect of this process on the system by adding to the Hamiltonian $\mathcal{H}$, an exchange Hamiltonian,

$$\mathcal{H} = -2J(\Delta)\sum_i \sum_{j\neq(i)} \vec{\sigma}_i \cdot \vec{\sigma}_j ,$$

where $J = \Delta\epsilon_T + \Delta\epsilon_x$

is given approximately by

$$J(\Delta) \approx -\tfrac{3}{5}(2\pi)^{1/2}(\hbar^2\alpha^2/m)\alpha^3\sigma^2\Delta \times e^{-\alpha^2(\Delta^2+\sigma^2)/2} . \qquad (69)$$

This last equation follows from the analytic approximation to the overlap integral developed above and in Appendix A. It has a very simple meaning. Since $\hbar^2\alpha^2/m \approx k_B\Theta_D$ we write

$$|J(\Delta)|/\hbar = \omega_T \approx \omega_D e^{-\alpha^2(\Delta^2+\sigma^2)/2} , \qquad (70)$$

where $\hbar\omega_D = k_B\Theta_D$ is the frequency of a ^{3}He atom in its potential well (the number of times per second it approaches the barrier), and the factor $\exp[-\alpha^2(\Delta^2+\sigma^2)/2]$ is the probability of tunneling through the barrier.

In Fig. 10, we have plotted $J(\Delta)$ from the exact evaluation of $\Delta\epsilon_T$ and $\Delta\epsilon_x$ (Appendix A) against molar volume. On the same figure we have plotted the data from both NMR and thermostatic measurements of $J(\Delta)$.[3] We note that the computed values of $J(\Delta)$ have essentially the molar volume dependence of the data. The computed values of $J(\Delta)$ are below the experimental values as they should be, since $J(\Delta)$ is sensitive to the overlap at distances $d = \frac{1}{2}(\Delta^2+\sigma^2) \approx 0.6\Delta$, far from the lattice sites, where the Gaussians give too little wave function.

VI. CONCLUSION

In this paper, we have attempted to calculate the magnitude and sign of the exchange interaction in solid ^{3}He. The principal results we have obtained are these.

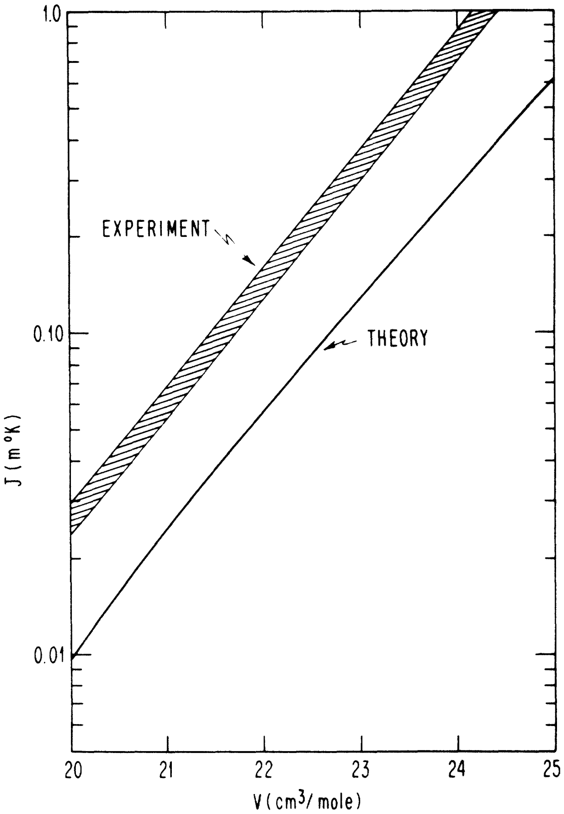

FIG. 10. J versus molar volume: The shaded area is the region in which the results of the NMR and thermostatic measurements of J fall. See Ref. 3.

(1) A Hamiltonian of the form

$$\mathcal{H}_x = -2J(\Delta)\sum_i \sum_{j\neq i} \vec{\sigma}_i \cdot \vec{\sigma}_j$$

correctly describes the exchange system. The exchange energy $J(\Delta)$ arises from two sources, tunneling and interaction.

(2) The tunneling process in which particle 1 tunnels through the potential barrier of the lattice medium from lattice site $\vec{R}_1$ to lattice site $\vec{R}_2$, at the same time that particle 2 tunnels from $\vec{R}_2$ to $\vec{R}_1$, leads to manifest antiferromagnetism for the ground state of the solid.

(3) The interaction process (which like the tunneling process depends on the overlap integral) is sensitive to the nature of the interaction between the exchanging pair. The interaction process leads to manifest ferromagnetism for the ground state of the solid.

(4) For a system in which there are strong dynamical correlations in the motion of a pair of particles, i.e., for a system in which the pair wave function must be taken of the form

$$\Psi_{12}(12)=\varphi_1(1)\varphi_2(2)g_{12}(12),$$

the effects of the interaction process is embodied solely in the dependence of the correlation function $g_{12}(12)$ upon the spatial symmetry of the pair wave function. *If the dynamical correlations do not depend on the symmetry of the pair wave function, there is no coupling between the interaction and symmetry.*

(5) For bcc ^{3}He over the full range of molar volumes at which it exists, the tunneling process leads to an antiferromagnetic energy shift which is greater by a factor of 2.5 than the ferromagnetic energy shift due to the interaction process. This result does not depend upon (a) the details of the single particle wave functions; (b) the details of the pair correlation function; (c) the molar volume; and (d) the spacing between the exchanging pairs (it is equally true to next-neighbor exchange). Therefore, the ground state of bcc ^{3}He is antiferromagnetic. The exchange interaction energy is principally due to a tunneling process.

(6) We have calculated the magnitude of $J(\Delta)$ using a wide variety of correlation functions. We find $J(\Delta)$ to be substantially independent of the details of the correlation function and in good agreement with experiment. The principal molar volume dependence in $J(\Delta)$ comes from the dependence of the overlap of the single-particle wave functions on molar volume.

(7) The physics of the tunneling process and the interaction process are the same as the physics which is described by Anderson[32] in the treatment of excitons and spin waves. A Hubbard-like Hamiltonian may be employed to describe solid helium and to quantify the relationship of the solid-helium magnetism problem to the more familiar treatments of magnetism.[33]

(8) Finally, we comment on the relation of our calculation of J to the work of Thouless, and of Nosanow and co-workers. The calculation of Thouless is a tunneling calculation and leads to manifest antiferromagnetism. The calculations of Nosanow and co-workers are of a quantity whose relationship to J is unknown. Under a special set of assumptions (which are not those employed by Nosanow and co-workers) the quantity calculated by Nosanow and co-workers is a tunneling approximation to J.

We will complete this section by commenting on two problems, related to the one we have dealt with above, which are the subject of our current investigations.

A. Triple Exchange

When a triple of particles tunnel in a cyclic way $[\varphi_1(1)\to\varphi_2(1),\ \varphi_2(2)\to\varphi_3(3)\to\varphi_3(3)\to\varphi_1(3)]$ it can be shown that the ground state for the triple is ferromagnetic. This result is in agreement with that obtained earlier by Thouless.[10] Like the pair tunneling process, triple tunneling is proportional to the overlap integral:

$$J(\Delta)_3\approx k_B\Theta_D p_3,$$

where
$$p_3=\frac{\int d\vec{x}_1\,d\vec{x}_2\,d\vec{x}_3\,\Psi_{123}(123)\Psi_{123}(312)}{\int d\vec{x}_1\,d\vec{x}_2\,d\vec{x}_3\,\Psi_{123}(123)\Psi_{123}(123)}$$

and
$$\Psi_{123}(123)=\varphi_1(1)\varphi_2(2)\varphi_3(3)g_{123}(123)$$
$$\approx\varphi_1(1)\varphi_2(2)\varphi_3(3)g_{12}(12)g_{23}(23)g_{31}(31).$$

When the triple of particles cycle they do not have to pass around one another as they do in pair tunneling. Thus, the factor $\exp[-\alpha^2\sigma^2/2]$ is absent (see Fig. 9). But p_3 requires the overlap of three pairs of wave functions. We estimate p_3 to be

$$p_3\leqslant\left(e^{-\alpha^2/2)(\Delta/2)^2}\right)^4\left(e^{-(\alpha^2/2)(\Delta_1/2)^2}\right)^2$$
$$=e^{-(5/3)\alpha^2\Delta^2/2},$$

where $\Delta_1=\frac{2}{3}\sqrt{3}\,\Delta$, the next-neighbor distance. Since p_2 is approximately

$$p_2\approx e^{-(\alpha^2/2)(\Delta^2+\sigma^2)},$$

we have $p_3/p_2\leqslant e^{-(\alpha^2/2)(\frac{2}{3}\Delta^2-\sigma^2)}\approx 0.14.$

Triple tunneling is down by about an order of magnitude from pair tunneling.

We expect the same inequality which related pair tunneling and pair exchange to relate triple tunneling and triple exchange. We have not investigated this point in detail.

B. Phonons and the Exchange System

The exchange system arises because of a pair tunneling process. The coupling of phonons to the exchange system occurs because of the modulation of the double well by the presence of a phonon in the lattice, i.e., $U_{12}(1;t)$ given by something like

$$U_{12}(1;t)=\sum_{j\neq 1,2}\int d\vec{x}_j\,v(\vec{x}_1\vec{x}_j)\,|\varphi_{R_j(t)}(j)|^2,$$

where $\vec{R}_j(t)=\vec{R}_j+\vec{e}\exp[i(\vec{k}\cdot\vec{R}_j-\omega t)]$ replaces $U_{12}(1)$ in Eq. (50). This modulation is most potent in the region of space midway between lattice sites. The pair of particles are tunneling through a barrier whose height is modulated by the presence of phonons in the lattice. This process may be studied using time-dependent perturbation theory; the perturbation is

$$U_{12}(1;t) + U_{12}(2;t) - U_{12}(1) - U_{12}(2).$$

The obvious result of such a calculation is that if the modulation is at frequency $J(\Delta)$ there is a resonant absorption of energy and a transition from $\Psi_{12}^{+}(12)$ to $\Psi_{12}^{-}(12)$. Such a modulation can be due to a phonon of frequency $\omega = J(\Delta)/\hbar$ (a 1-phonon process) or to a superposition of phonons whose beat frequency is $J(\Delta)/\hbar$ [a 2-phonon process with $\omega - \omega' = J(\Delta)/\hbar$].

It is clear that the perturbation

$$P_{12}(12;t) = U_{12}(1;t) + U_{12}(2;t) - U_{12}(1) - U_{12}(2)$$

is not easily represented by a simple power series in the displacements from the lattice sites. That is, we do not expect that there exists a useful expansion of $P_{12}(12;t)$ in the form

$$P_{12}(12;t) = P_{12}(\vec{R}_1\vec{R}_2) + \nabla_1 P_{12}(\vec{R}_1\vec{R}_2) \cdot \vec{u}_1(t) + \nabla_2 P_{12}(\vec{R}_1\vec{R}_2) \cdot u_2(t) + \cdots,$$

since $P_{12}(12;t)$ is most important in the region of space where $|\vec{u}_1| \approx |\vec{u}_2| \approx \frac{1}{2}\Delta$ and where such a power series breaks down. Therefore, we do not believe that the usual procedures[9] for treating 1- and 2-phonon processes are meaningful for treating phonon modulated tunneling. Nonetheless, the qualitative results of the usual treatment, (e.g., the temperature dependence, etc.) can be carried over to this problem since they are invariant to the details of the perturbation and depend principally on the density of states available to effect 1- and 2-phonon assisted transitions.

Varma,[9] and Nosanow and Varma[8] have developed a theory of the coupling of the phonons and the exchange system. In their theory the phonons appear in the off-diagonal matrix elements of the exchange operator. Explicit expressions are given for these off-diagonal matrix elements. But, when calculated properly, these off-diagonal matrix elements vanish identically. They are nonzero in the theory of Nosanow and Varma because these authors replace the matrix elements like

$$\langle \Psi_{12}^{+}(12) | H(12) | \Psi_{12}^{-}(12) \rangle\rangle,$$

where $H(12) = T(1) + T(2) + v(12)$,

by $\langle \Psi_{12}(12) | H(12) | \Psi_{12}^{-}(12) \rangle\rangle$,

which are nonzero.

Associated with $\Psi_{12}^{+}(12)$ and $\Psi_{12}^{-}(12)$ are spin wave functions which are orthogonal. There can be no matrix elements between $\Psi_{12}^{+}(12)$ and $\Psi_{12}^{-}(12)$ for spin-independent quantities; $H(12)$ is spin-independent. This latter objection also applies to our statement that time-dependent perturbation theory can be used to treat $P_{12}(12;t)$. There are no matrix elements of $P_{12}(12;t)$ between $\Psi_{12}^{+}(12)$ and $\Psi_{12}^{-}(12)$, since $P_{12}(12;t)$ does not involve σ_1 and σ_2. We believe that the appropriate microscopic mechanism for coupling the phonons to the exchange system has yet to be identified.

This latter remark leads us to consider the general question of the phonon-exchange interaction. Is there a mechanism for phonon-exchange coupling? We believe that neither the existing calculations[9, 34] nor our calculation by conversation (above) provides an explanation for the mechanism of coupling between the exchange system and the phonons. We are led to look at the experimental data which suggests the existence of phonon-exchange processes in the solid. The strongest such data is that of Giffard and Hatton.[34] In that data no evidence for a phonon-exchange interaction appears in the data on pure ^{3}He. When small concentrations of ^{4}He impurities are introduced a 2-phonon process occurs, i.e., Hatton and Giffard find an extra relaxation process which has a very strong temperature dependence. They suggest that this extra relaxation process is a 2-phonon process. We believe that this extra relaxation process is *not* a 2-phonon process. We make the following argument: (a) At $T^{-1} = 2.5$ K^{-1} in pure ^{3}He there is no evidence for a 2-phonon process, $T_1^{(2\text{ phonon})}(x=0) > 10^3$ sec. Therefore, the intrinsic relaxation rate for the 2-phonon process in bulk ^{3}He is less than 10^{-3} sec^{-1}. (b) The addition of 15 ppm ^{4}He impurities leads to $T_1^{(2\text{ phonon})}(x = 15\text{ppm}) \approx 10^2$ sec. Each impurity (or the region of space around each impurity) must relax about 10^5 ^{3}He atoms in 10^2 sec. An intrinsic relaxation rate at an impurity site of 10^3 sec^{-1} is required. (c) Thus, if the extra relaxation mechanism is a 2-phonon process in the vicinity of a ^{4}He impurity, this process must go six order of magnitude more rapidly than it does in bulk ^{3}He. We do not believe it is reasonable to expect this. Therefore, we argue that the extra relaxation is not due to a 2-phonon process.

As an alternative to the 2-phonon process we make the following speculation. In pure ^{3}He in the temperature range $1.0 \leqslant T^{-1} \leqslant 2.5$, the relaxation mechanism is due to the presence of vacancies. Relaxation occurs because one of a tunneling pair of ^{3}He atoms finds that it has a vacancy for a neighbor and tunnels to the vacancy lattice site leaving its tunneling partner behind and confronted with a magnetically inert vacancy. The original tunneling partnership is destroyed. We suggest that the presence of ^{4}He impurities in a ^{3}He lattice leads to a qualitatively similar relaxation mechanism. One of a tunneling pair of ^{3}He atoms finds it has a ^{4}He atom for a neighbor and tunnels to the ^{4}He lattice site leaving its original tunneling partner behind and confronted with a magnetically inert ^{4}He particle. The physics is the same; the original tunneling partnership is destroyed.

A few estimates of magnitudes are necessary to support this suggestion. First, we know that at $T^{-1} = 2.5$ in pure ^{3}He T_1 is greater than 10^3. This T_1 is due to a concentration $x \approx e^{-13.6\times 2.5} \approx 10^{-14}$ of vacancies. It requires a jump frequency (or tunneling frequency) of about 10^{10} sec^{-1}. This is a reasonable time for quantum mechanical tunneling of a ^{3}He atom into a neighboring vacancy lattice site. We estimate

$$\omega_T^{(v)} \approx \frac{k_B \Theta_D}{\hbar} \int d\vec{x}_1\, \varphi_1(1)\, \varphi_2(1) \approx \frac{k_B \Theta_D}{\hbar} e^{-\alpha^2 \Delta^2/4}$$

or $\omega_T^{(v)} \approx 2\times 10^5 \omega_T \approx 5\times 10^9 \text{ sec}^{-1}$.

Here ω_T is the quantum mechanical tunneling frequency computed above for two ^{3}He atoms. To make this numerical estimate we have used α^2 from Table I ($v = 20$ cm^3/mole) and $\Theta_D \approx 30$°K. A vacancy concentration of 10^{-5} would lead to a T_1 on the order of 10^{-5} sec. A ^{4}He impurity concentration of 10^{-5} leads to $T_1 \approx 10^2$ sec. Thus, the tunneling process involving a ^{3}He atom, and a ^{4}He atom is very slow compared to the vacancy tunneling process. In fact, we expect that $\omega_T^{(3,4)}$ is about

$$\omega_T^{(3,4)} \approx \frac{k_B \Theta_D}{\hbar} e^{-[\alpha_3^2(\Delta^2+\sigma^2)]/4} \times e^{-[\alpha_4^2(\Delta^2+\sigma^2)]/4} .$$

Since $\alpha_4^2 = \left(\frac{m_4}{m_3}\right)^{1/2} \alpha_3^2$, we have[35]

$$\omega_T^{(3,4)} \approx \omega_T e^{-\{[\alpha^2(\Delta^2+\sigma^2)/4][(m_4/m_3)^{1/2}-1]} \approx \tfrac{1}{3}\omega_T$$

or $\omega_T^{(3,4)} \approx 10^4 \text{ sec}^{-1}$.

This tunneling frequency is of the right order of magnitude.

Although the estimates of tunneling frequencies above tend to confirm our speculation about the ^{3}He impurity, the process we are describing has no temperature dependence. The extra relaxation process found by Giffard and Hatton depends on T as T^{-8} or $\exp(-3.5/T)$. This latter temperature dependence suggests either that the dynamics of the combined vacancy and ^{4}He impurity system is more complicated than simple superposition or that phonons may be aiding in the ^{4}He-^{3}He tunneling process. We do not understand the details of either of these processes.

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APPENDIX A.

1. Overlap Integral

Consider

$$I_0 = \frac{\int d\vec{x}_1\, d\vec{x}_2\, \psi_{12}(12)\psi_{12}(21)}{\int d\vec{x}_1\, d\vec{x}_2\, \psi_{12}(12)\psi_{12}(12)} , \tag{A1}$$

where $\psi_{12}(12) = \varphi_1(1)\varphi_2(2) g(r_{12})$ (A2)

and $\varphi_1(1) = e^{-[\alpha^2(\vec{x}_1 - \vec{R}_1)^2]/2}$. (A3)

Using the center of mass and relative coordinates we have

$$I_0 = 4\pi e^{-\alpha^2\Delta^2/2} \frac{\int_0^\infty r^2 g(r)^2 e^{-\alpha^2 r^2/2} dr}{\int d\vec{r} g(r)^2 e^{-\alpha^2(\vec{r}-\vec{\Delta})^2/2}} = N/D . \tag{A4}$$

The denominator is approximated adequately by putting $g(r) = 1$. We have

$$D = (2\pi)^{3/2}/\alpha^3 . \tag{A5}$$

For the numerator, we use a cutoff approximation in which $g(r)^2$ is replaced by $\theta(r-\lambda)$ for a suitable λ. We choose λ to be that r at which the exponential part of the integrand in Eq. (A4) has a maximum. For $g(r)$ given by

$$g(r) = e^{-\beta(\sigma/r)^n} , \tag{A6}$$

we have the condition

$$\frac{d}{dr}\left[\tfrac{1}{2}\alpha^2 r^2 + 2\beta(\sigma/r)^n\right]_{r=\lambda} = 0 . \tag{A7}$$

Thus, we have

$$I_0 = \frac{1}{D} 4\pi e^{-\alpha^2\Delta^2/2} \int_\lambda^\infty r^2 e^{-\alpha^2 r^2/2} dr$$

$$= e^{-\alpha^2\Delta^2/2} J_0(\lambda) \ . \qquad \text{(A8)}$$

We approximate $J_0(\lambda)$ thus:

$$J_0(\lambda) \equiv \frac{4\pi}{D} \int_\lambda^\infty r^2 e^{-\alpha^2 r^2/2}\, dr$$

$$\approx \frac{4\pi}{D} \lambda^2 \int_\lambda^\infty e^{-\alpha^2 r^2/2}\, dr \qquad \text{(A9)}$$

Further, we use the inequality

$$\frac{1}{x+(x^2+2)^{1/2}} < e^{x^2} \int_x^\infty e^{-t^2}\, dt \leq \frac{1}{x+(x^2+4/\pi)^{1/2}}$$

to approximate the remaining integral by

$$J_0(\lambda) \approx \frac{4\pi}{D} \lambda^2 \frac{\sqrt{2}}{\alpha} \int_{\alpha\lambda/\sqrt{2}}^\infty e^{-t^2}\, dt = \frac{4\pi}{D} \frac{\lambda}{\alpha^2} e^{-\alpha^2\lambda^2/2} \ . \qquad \text{(A10)}$$

Thus, we have

$$I_0 = (2/\pi)^{1/2} \alpha\lambda\, e^{-\alpha^2(\Delta^2+\lambda^2)/2} \ . \qquad \text{(A11)}$$

The cutoff λ is at

$$\lambda = \sigma(2\beta n/\alpha^2\sigma^2)^{2+1/n} = \sigma\kappa \ .$$

Since $\beta \approx 1$, $\alpha^2\sigma^2 \approx 8$, for $n = 4, 5, \ldots,$ κ is not substantially different from 1. The cutoff is near σ for all reasonable choices of n. It should be!

2. Interaction Integral

The relative coordinate part of the interaction integral is

$$I_1 = \frac{4\pi}{D} e^{-\alpha^2\Delta^2/2} \Delta \int_0^\infty r^2 g'(r) g(r) e^{-\alpha^2 r^2/2}\, dr \ . \qquad \text{(A12)}$$

Using the analytic approximation to $g(r)$,

$$g(r) = e^{-\beta(\sigma/r)^n} \ ,$$

we have

$$I_1 = \frac{4\pi}{D} e^{-\alpha^2\Delta^2/2} \Delta \times \int_0^\infty \frac{n\beta}{\sigma} \left(\frac{\sigma}{r}\right)^{n+1} r^2 g(r)^2 e^{-\alpha^2 r^2/2}\, dr \ .$$

We use the same cutoff procedure as above to write this in the form

$$I_1 = \frac{4\pi}{D} e^{-\alpha^2\Delta^2/2} \Delta \frac{n\beta}{\sigma} \times \int_\lambda^\infty \left(\frac{\sigma}{r}\right)^{n+1} r^2 e^{-\alpha^2 r^2/2}\, dr \ . \qquad \text{(A13)}$$

In the spirit of the approximation above to $J_0(\lambda)$, we write

$$J_1(\lambda) \equiv \frac{4\pi}{D} \frac{\Delta}{\sigma} n\beta \int_\lambda^\infty \left(\frac{\sigma}{r}\right)^{n+1} r^2 e^{-\alpha^2 r^2/2}\, dr$$

$$\approx \frac{4\pi}{D} \frac{\Delta}{\sigma} n\beta \left(\frac{\sigma}{\lambda}\right)^{n+1} J_0(\lambda) \ . \qquad \text{(A14)}$$

Thus, we have

$$I_1 = \frac{\Delta}{\sigma} n\beta \left(\frac{\sigma}{\lambda}\right)^{n+1} I_0 \ , \qquad \text{(A15)}$$

or

$$\frac{I_1}{I_0} = \frac{\Delta}{\sigma} n\beta \left(\frac{\sigma}{\lambda}\right)^{n+1} \ . \qquad \text{(A16)}$$

Now from the definition of λ, we have

$$\left(\frac{\sigma}{\lambda}\right)^{n+2} = \frac{\alpha^2\sigma^2}{2n\beta} = \left(\frac{\sigma}{\lambda}\right)^{n+1} \frac{1}{\kappa} \ , \qquad \text{(A17)}$$

so that we may write

$$\frac{I_1}{I_0} = \frac{1}{2}\alpha^2\Delta\sigma\kappa \ . \qquad \text{(A18)}$$

The interaction integral, defined by Eq. (62), is proportional to I_1; the tunneling integral, defined in Eq. (61), is proportional to I_0. We show in Appendix B that the tunneling integral is given by

$$\Delta\epsilon_T = 4\langle\Delta U_2(1)\rangle_{\text{ot}} I_0 \ ,$$

where $\langle\Delta U_2(1)\rangle_{\text{ot}} = (\hbar^2\alpha^2/m)\alpha^2\sigma\Delta/4$. From Eq. (62) we have

$$\Delta\epsilon_x = (\hbar^2\alpha^2/m)\tfrac{1}{2} I_1 \ . \qquad \text{(A19)}$$

Thus, we have

$$\Delta\epsilon_T/\Delta\epsilon_x \approx 4 \ . \qquad \text{(A20)}$$

We have calculated I_0 and I_1 and $\Delta\epsilon_T$ and $\Delta\epsilon_x$ exactly for the values of α^2 and Δ from the solution to the ground-state problem by Guyer and Sarkissian. We have done this for three correlation functions; (i) Nosanow's $f(r)=\exp[-Kv(r)]$, (ii) the correlation function of Guyer and Sarkissian, and (iii) the WKB correlation function. In Table I, we list the value of J_0 and the ratio J_1/J_0 for all three correlation functions. We note:

(a) The ratio J_1/J_0 is essentially the same at all molar volumes for all three correlation functions.

(b) The ratio J_1/J_0 agrees quite well with the result for that ratio using the cutoff approximation, i.e., compare J_1/J_0 with column four of the table.

(c) The ratio $\Delta\epsilon_T/\Delta\epsilon_x$ computed using the values of J_0 and J_1 from the table is $\Delta\epsilon_T/\Delta\epsilon_x \approx 2.5$ [independent of molar volume and in reasonable agreement with the estimate above, Eq. (A20)].

(d) Both J_0 and J_1 are largest using the Nosanow correlation function and smallest for the WKB correlation function. This is as it should be since the Nosanow correlation function lets the pair furthest into the hard core, whereas the WKB correlation function keeps them furthest away (see Fig. 5). By equating J_0 calculated from the exact formula [Eq. (A4)] with J_0 from the approximate formula we can find a λ for each molar volume and correlation function. We find: (i) the Nosanow correlation function acts like $\theta(r-2.22)$ and is independent of molar volume, (ii) the WKB correlation function acts like $\theta(r-2.65)$ at $v=24.7$ cm^3/mole and $\theta(r-2.45)$ at $v=18.0$ cm^3/mole, and (iii) the correlation function of Guyer and Sarkissian behaves like $\theta(r-2.45)$ at $v=24.7$ cm^3/mole and $\theta(r-2.30)$ at $v=18.0$ cm^3/mole. The Nosanow correlation function goes from 0 to 1 rapidly in the vicinity of σ. Its cutoff equivalent is less sensitive to molar volume than those of the WKB correlation function and the Guyer and Sarkissian correlation function.

APPENDIX B

The overlap trajectory is defined as that region of real space on which the overlap of $\varphi_1(1)\varphi_2(2)\times g(r_{12})$ and $\varphi_2(1)\varphi_1(2)g(r_{12})$ is a maximum. For Gaussian single-particle wave functions, this region of space is a spherical shell of radius $\frac{1}{2}\sigma$ centered midway between $\vec{R}_1$ and $\vec{R}_2$ (see Fig. 9). A quantity like

$$\langle X|F(\vec{x}_1\vec{x}_2)|1\rangle/\langle 1|1\rangle ,$$

where $F(\vec{x}_1\vec{x}_2)$ are mild functions of $\vec{x}_1$ and $\vec{x}_2$ is well approximated by

$$\langle X|F(\vec{x}_1\vec{x}_2)|1\rangle/\langle 1|1\rangle \approx \langle F(\vec{x}_1\vec{x}_2)\rangle_{\mathrm{ot}}p .$$

Here $\langle F(\vec{x}_1\vec{x}_2)\rangle_{\mathrm{ot}}$ stands for the average of $F(\vec{x}_1\vec{x}_2)$ over the overlap trajectory. [We have verified this approximation for $F(\vec{x}_1)=(\vec{x}_1-\vec{R}_1)^2$, where it is good to better than 2%.]

$\Delta U_2(1)$ is defined in Eq. (55) [see Fig. 6]. We take $U_{12}(1)$ to be the sum of clipped harmonic oscillator potentials

$$U_{12}(1)=\tfrac{1}{2}k(\vec{x}_1-\vec{R}_1)^2, \quad x<0;$$

$$U_{12}(1)=\tfrac{1}{2}k(\vec{x}_1-\vec{R}_2)^2, \quad x>0.$$

The biased single-particle potentials are

$$U_1{}^B(1)=\tfrac{1}{2}k(\vec{x}_1-\vec{R}_1)^2 ,$$

and $U_2{}^B(1)=\tfrac{1}{2}k(\vec{x}_1-\vec{R})^2$.

The average value of $\Delta U_2(1)$ over the overlap trajectory is

$$\langle\Delta U_2(1)\rangle_{\mathrm{ot}}=\tfrac{1}{2}k\int d\Omega[\vec{r}-\vec{R}_1)^2-(\vec{r}-\vec{R}_2)^2]$$

$$=-k\frac{\sigma\Delta}{4} , \tag{B1}$$

where $|\vec{r}|=\frac{1}{2}\sigma$ and the $d\Omega$ integral is over the half-space $0\leqslant\theta\leqslant\frac{1}{2}\pi$. Finally, we use the identity $k=\hbar^2\alpha^4/m$ to write

$$\langle\Delta U_2(1)\rangle_{\mathrm{ot}}=(\hbar^2\alpha^2/m)\alpha^2\sigma\Delta/4 . \tag{B2}$$

TABLE I. Values of J_0 and J_1/J_0 for the three correlation functions.

Molar volume	Δ	α^2	$\alpha^2\Delta\sigma/2$	g_{gs} J_0	g_{gs} (J_1/J_0)	$f(r)$ J_0	$f(r)$ (J_1/J_0)	g_{WKB} J_0	g_{WKB} (J_1/J_0)
18	3.39	2.48	10.8	3.57(−3)	8.5	5.81(−3)	8.6	1.53(−3)	8.9
19	3.45	2.23	9.9	5.90(−3)	7.8	1.02(−2)	7.9	2.71(−3)	8.2
20	3.51	2.03	9.2	9.06(−3)	7.3	1.62(−2)	7.2	4.41(−3)	7.6
21	3.56	1.84	8.4	1.34(−2)	6.8	2.47(−2)	6.7	6.90(−3)	7.1
22	3.62	1.69	7.9	1.86(−2)	6.3	3.48(−2)	6.2	9.99(−3)	6.6
23	3.67	1.55	7.3	2.53(−2)	5.9	4.79(−2)	5.7	1.42(−2)	6.2
24.7	3.76	1.32	6.4	4.20(−2)	5.2	8.08(−2)	4.9	2.58(−2)	5.5

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[1] In this early part of the discussion we shall ignore correlations.

[2] This terminology arises because a Hamiltonian like that in Eq. (6) correctly describes exchange in electron systems. We will show in this paper that exchange is due to a tunneling process and an interaction process. We will use the term exchange generally to refer to the piece of the Hamiltonian which embodies the effect of the tunneling process and the interaction process.

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[14] We adopt a notation in which subscripts refer to lattice sites as illustrated here.

[15] The Hamiltonian in Eq. (15) contains a single-particle potential $U_i(i)$ which is shared by i and all of its neighbors. Thus, in Eq. (13) a factor $\frac{1}{2}$ appears. We have in mind using the Hartree solution to the $\mathcal{H}_0$ problem to generate a complete set of wave functions with which to do perturbation theory.

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[26] There are two possible correlation functions. The function $g_{12}(12)$ solves the problem

$$[T(1) + T(2) + U_1(1) + U_2(2) + \tilde{v}(12)]\,\varphi_1{}^0(1) \times \varphi_2{}^0(2)\,g_{12}(12) = \epsilon_{12}\,\varphi_1{}^0(1)\,\varphi_2{}^0(2)\,g_{12}(12),$$

where $[T(1) + U_1(1)]\,\varphi_1{}^0(1) = \epsilon_0\varphi_1{}^0(1)$. The function $\bar{g}_{12}(12)$ solves the problem

$$[T(1) + T(2) + \bar{U}_1(1) + \bar{U}_2(2) + v(12)]\,\bar{\varphi}_1(1) \times \bar{\varphi}_2(2)\,\bar{g}_{12}(12) = \epsilon_{12}\,\bar{\varphi}_1(1)\,\bar{\varphi}_2(2)\,\bar{g}_{12}(12)_1,$$

where $[T(1) + \bar{U}_1(1)]\,\bar{\varphi}_1(1) = \bar{\epsilon}_0\,\bar{\varphi}_1(1)$. $g_{12}(12)$ and $\bar{g}_{12}(12)$ are related since $\varphi_1(1)\varphi_2(2)\,g_{12}(12) = \bar{\varphi}_1(1)\bar{\varphi}_2(2)\,\bar{g}_{12}(12)$. For the purpose of a computer solution to Eq. (30) it is most convenient to find $\bar{g}_{12}(12)$ and construct $g_{12}(12)$ from it. We will interchangeably use both without the notation change $g_{12}(12) \longleftrightarrow \bar{g}_{12}(12)$.

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Velocity of Second Sound in Dilute He^3-He^4 Mixtures

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Using the theory of Bardeen, Baym, and Pines (BBP) for dilute He^3-He^4 mixtures, the second-sound velocity has been calculated for concentrations of He^3 below 10% for temperatures below 0.5 °K and pressures below 20 atm. In addition, the phase diagram at $T=0$ for phase separation and solidification has been constructed. The changes in the second-sound velocity and the phase diagram caused by variations in the BBP parameter α are also studied.

I. INTRODUCTION

It is well known that the presence of a small amount of He^3 in an He^4 solution has a dramatic effect on the second-sound velocity c_{II} at very low temperatures. The qualitative explanation is that at these low temperatures the He^4 contribution to the normal fluid density ρ_n (as due to the phonons) is so small that the He^3 contribution to ρ_n becomes dominant.

A quantitative understanding of this phenomenon was obtained by Pomeranchuk and Landau,[1] who regarded the He^3 particles as independent excitations, with excitation spectrum $E_0 + \hbar^2k^2/2m^*$, where m^* is the (effective) mass of the excitations. It appears that there is a regime in concentrations x of He^3 where the second-sound velocity depends only on m^*. Thus m^* can be determined from measurement of c_{II} and is found[2] to be of order $2.5m_3$, where m_3 is the He^3 atomic mass. This regime is limited on the low-concentration side by the condition that He^3 excitations still dominate the phonons. For temperatures below $T=0.5$ °K this is already so for $x > 10^{-5}$. On the high-concentration side of the regime one has the limiting condition that the He^3 excitations are sufficiently dilute so that they can still be considered as independent.

Recently, Bardeen, Baym, and Pines[3] (BBP) have extended Pomeranchuk's idea and determined the effective interaction between the He^3 excitations. The interaction in dilute mixtures is weak and attractive and can be taken pairwise.

It is the aim of this paper to extend the second-sound theory in He^3 – He^4 mixtures to higher concentrations using the BBP interaction. We estimate the limit of concentrations, adequately described by the BBP interactions, to be of the order of 10%. Since BBP's potential is determined at $T=0$, we are restricted to temperatures for which the He^4 background is essentially in its ground state, i.e., to temperatures where no appreciable amount of phonons are present, which we estimate to be so below $T=0.5$ °K.

Also, the BBP potential is determined from specific-heat and spin-diffusion data at vapor pressure. Accordingly, the shape of the potential is known only at $p=0$ and there only approximately. However, since the shape of the potential enters into our results for c_{II} in a minor way, we have assumed that the shape is pressure-independent. In fact, the uncertainties due to the lack of knowledge of the pressure dependence of the shape seem small as compared to the uncertainties in the measured strength parameters, in particular, the so-called BBP parameter α.

The phase-separation curve is more sensitive to the shape of the BBP potential than c_{II}. Therefore, we have included in Appendix A a calculation