PHYSICAL REVIEW B VOLUME 11, NUMBER 9 1 MAY 1975

Quantum-crystal alloys I: Mass-fluctuation waves*

Whittak Huang, H. A. Goldberg, and R. A. Guyer
Department of Physics and Astronomy, University of Massachusetts, Amherst, Massachusetts 01002

(Received 8 May 1974)

The mass fluctuations in a quantum-crystal alloy lead to a long-range interaction between the particles. For low ^{3}He concentrations in ^{4}He, this interaction requires a modification of the simple picture of mass-fluctuation waves. We determine the effect of this interaction on the motion of a ^{3}He particle through ^{4}He and give an alternate explanation of the data that is assumed to show the existence of these excitations. We set a limit at $x_3 \ll 10^{-3}$ for the onset of mass-fluctuation-wave behavior.

I. INTRODUCTION

"Mass-fluctuation waves" (MFW) were invented independently by Andreev and Lifshitz[1] and Guyer and Zane.[2] These excitations describe the coherent motion of an impurity atom through a crystal. Further theoretical discussion of them is found in Refs. 3–6. The early survey experiments by Myoshi, Cotts, Greenberg, and Richardson[7] on solid ^{3}He-^{4}He mixtures suggested that dilute ^{3}He-in-^{4}He solid mixtures are a suitable system in which to seek evidence for these excitations.[6] Subsequently Greenberg, Thomlinson, and Richardson[8,9] reported T_1 and T_2 measurements that showed compelling evidence for ^{3}He tunneling in ^{4}He. Richards *et al.*[10,11] have undertaken an extensive set of measurements on dilute ^{3}He-in-^{4}He mixtures designed to find evidence for the coherent motion of the impurity, i.e., for the existence of ^{3}He MFW. These measurements include diffusion data in addition to T_1 and T_2 data. Richards *et al.* have argued that only a diffusion measurement gives a clear signature for coherent motion. Recently Grigor'ev and co-workers[12–14] have verified and extended the diffusion measurements of Richards *et al.*

Many aspects of the data of Richards *et al.* are consistent with the qualitative features expected using the MFW description of the ^{3}He motion. But there are two serious difficulties in the quantification of such a description: (i) The rate of ^{3}He-^{4}He tunneling must be taken to be about two orders of magnitude below what is regarded as a plausible value[15]; (ii) the interaction between ^{3}He impurities must be taken to be about three orders of magnitude below what is regarded as a plausible value.[7,16]

This paper provides an explanation of the measurements of Richard *et al.* and Grigor'ev and co-workers in terms of ^{3}He tunneling in the presence of a strong ^{3}He-^{3}He interaction. This explanation in terms of an *interaction* model is offered as an alternative to the *MFW* model. It employs plausible magnitudes for the basic parameters describing the system. The interaction model yields a ^{3}He concentration limit below which MFW behavior should be seen in a diffusion experiment. The experiments of Richards *et al.* and Grigor'ev and co-workers reach down to this limit, $x_3 \ll 10^{-3}$.

In Sec. II we derive the Hamiltonian that we use to describe a ^{3}He-^{4}He mixture. We employ a single-particle representation of the particle motions using a few of the low-lying states of an average crystal, a crystal with an average mass at each lattice site. The real crystal has mass fluctuations that cause its Hamiltonian to depart from the average crystal Hamiltonian. These mass fluctuations lead to a mass-fluctuation–mass-fluctuation interaction which is important to the description of ^{3}He motion at low ^{3}He concentrations. We assess the magnitude and range of the mass-fluctuation–mass-fluctuation interaction in Sec. III. We find it to be of strength 10^{-2} K and to fall off as r^{-3}. In Sec. IV we examine the effect of the mass-fluctuation–mass-fluctuation interaction on the behavior of a ^{3}He MFW. We find that in the concentration range $10^{-3} \leq x_3 \leq 10^{-2}$ a ^{3}He particle moves incoherently because of the mass-fluctuation–mass-fluctuation interaction. The diffusion constant and T_2 that we calculate for this concentration range are in good qualitative and quantitative agreement with the data of Richards *et al.* and Grigor'ev and co-workers. We show that a rate of ^{3}He-^{4}He tunneling similar to the rate of ^{3}He-^{3}He tunneling is consistent with the data. We expect coherent ^{3}He motion, i.e., a ^{3}He MFW, at $x_3 \ll 10^{-3}$. Section V contains a brief summary of our results and conclusions. The details of many of the calculations that are required to support the arguments in the main body of the paper are contained in Appendices A–E. These include a reformulation of the small-polaron problem in Appendix D that may be of some interest.

II. THEORY OF THE HAMILTONIAN

In this section we derive the equation for the ground-state energy that leads to phase separation and to the interaction between impurity atoms. We formulate the theory, in second-quantized form, using the single-particle states of the average single crystal (average crystal), i.e., the single-particle states generated by the solution to the problem

$$\overline{\mathcal{H}}(x_3)=\sum_i \frac{p_i^2}{2}\langle m^{-1}\rangle+\frac{1}{2}\sum_{ij}{}' v(ij), \tag{1}$$

where

$$\left\langle\frac{1}{m}\right\rangle=x_3\frac{1}{m_3}+(1-x_3)\frac{1}{m_4}=\frac{1}{m_4}+x_3\left(\frac{1}{m_3}-\frac{1}{m_4}\right) \tag{2}$$

and $x_3=N_3/N$ is the ^{3}He concentration. A solid ^{3}He-^{4}He mixture is described by

$$\mathcal{H}=\sum_i \frac{p_i^2}{2m_i}+\frac{1}{2}\sum_{ij}v(ij)=\overline{\mathcal{H}}+\sum_i \frac{p_i^2}{2}(m_i^{-1}-\langle m^{-1}\rangle)\,. \tag{3}$$

We assume that the Einstein-oscillator ground-state problem associated with $\overline{\mathcal{H}}$ is solved with the results

$$\overline{\Psi}_0=E_0(x_3)\overline{\Psi}_0, \tag{4}$$

where

$$\overline{\Psi}_0=\prod_i \overline{\phi}_i^{(0)}(x_3,\ \vec{\mathrm{r}}_i)\prod_{i<j}\overline{g}(x_3;\ |\vec{\mathrm{r}}_{ij}|)\,. \tag{5}$$

Here $\overline{\phi}_i^{(0)}$ is a single-particle wave function $\{$perhaps of the form $\overline{\phi}_i^{(0)}=A_0\exp[-\frac{1}{2}\overline{\alpha}^2(\vec{\mathrm{r}}_i-\vec{\mathrm{R}}_i)^2]\}$ and $\overline{g}$ is the part of the pair wave function that, with v, yields the t matrix,

$$\overline{t}(x_3,\ \vec{\mathrm{r}}_{ij})=\overline{g}(x_3,\ \vec{\mathrm{r}}_{ij})v(\vec{\mathrm{r}}_{ij})\,. \tag{6}$$

The details of the solution of the ground-state problem with $\overline{\mathcal{H}}$ are described by Takemori and Guyer.[17] We assume that in addition to the ground-state single-particle wave functions $\overline{\phi}^{(0)}$ and the ground-state t-matrix elements, $\overline{t}_{00,00}$, we also have a knowledge of the low-lying excited states and corresponding t-matrix elements (see, for example, Appendix C). Using these we can write $\mathcal{H}$ in second-quantized form,

$$\begin{aligned}\mathcal{H}=&\sum_R[K(R)_{11}n(R)_1+K(R)_{22}n(R)_2+K(R)_{00}n(R)_0]+\frac{1}{2}\sum_{RR'}[t(RR')_{11,11}n(R)_1n(R')_1+t(RR')_{22,22}n(R)_2n(R')_2\\&+t(RR')_{00,00}n(R)_0n(R')_0]+\sum_{RR'}[t(RR')_{00,11}n(R)_0n(R')_1+t(RR')_{00,22}n(R)_0n(R')_2+t(RR')_{11,22}n(R)_1n(R')_2]\\&+\sum_{RR'}[t(RR')_{00,01}n(R)_0+t(RR')_{11,01}n(R)_1+t(RR')_{22,01}n(R)_2](b_{R'}^\dagger+b_{R'})\\&+\sum_{RR'}[t(RR')_{00,02}n(R)_0+t(RR')_{11,02}n(R)_1+t(RR')_{22,02}n(R)_2](d_R^\dagger+d_{R'})+\frac{1}{2}\sum_{RR'}t(RR')_{01,01}(b_R^\dagger+b_R)(b_{R'}^\dagger+b_{R'})\\&+\frac{1}{2}\sum_{RR'}t(RR')_{02,02}(d_R^\dagger+d_R)(d_{R'}^\dagger+d_{R'})+\sum_{RR'}t(RR')_{01,02}(b_R^\dagger+b_R)(d_{R'}^\dagger+d_{R'})+\sum_R[\Delta K(R)_{00}n(R)_0\\&+\Delta K(R)_{11}n(R)_1+\Delta K(R)_{22}n(R)_2+\Delta K(R)_{02}(d_R^\dagger+d_R)]\,,\end{aligned} \tag{7}$$

where $c_{R\nu}^\dagger$ creates a particle in state ν at lattice site R, $n(R)_\nu=c_{R\nu}^\dagger c_{R\nu}$, $b_R^\dagger=c_{R1}^\dagger c_{R0}$, and $d_R^\dagger=c_{R2}^\dagger c_{R0}$. For the states 1 and 2 we employ the approximate states

$$\overline{\phi}_R^{(1z)}=A_1\alpha z\exp[-\tfrac{1}{2}\alpha^2(\vec{\mathrm{r}}-\vec{\mathrm{R}})^2] \tag{8}$$

and

$$\overline{\phi}_R^{(2)}=A_2[1-\tfrac{2}{3}\alpha^2(r-R)^2]\exp[-\tfrac{1}{2}\alpha^2(\vec{\mathrm{r}}-\vec{\mathrm{R}})^2]\,. \tag{9}$$

The 15 potential energy terms in Eq. (7) are shown in Fig. 1. We understand $b_R^\dagger$ to create a *displacement* fluctuation and $d_R^\dagger$ to create a *width* fluctuation (see Fig. 2). The displacement-displacement coupling constant, $t(RR')_{01,01}$, is the random-phase-approximation (RPA) phonon coupling. The displacement-width coupling constant, $t(RR')_{01,02}$, is the RPA cubic anharmonic phonon coupling, etc. (see Appendix C). There are lots of terms in Eq. (7). Most of them describe properties of a perfect single crystal. The last four terms describe effects due to the mass fluctuations away from the average crystal. If we systematically go through Eq. (7) we can understand the physics in most of the terms and achieve an enormous simplification of the equation. To this end we use $n(R)_0+n(R)_1+n(R)_2=1$ to rewrite Eq. (7) (with rearrangement) in the form

$$\begin{aligned}\mathcal{H}=&\sum_R\overline{K}(R)_{00}+\frac{1}{2}\sum_{RR'}t(RR')_{00,00}+\sum_R\Delta\overline{K}(R)_{00}+\sum_R[\epsilon(R)_1-\epsilon(R)_0]n(R)_1+\frac{1}{2}\sum_{RR'}t(RR')_{01,01}(b_R^\dagger+b_R)(b_{R'}^\dagger+b_{R'})\\&+\sum_R[\epsilon(R)_2-\epsilon(R)_0]n(R)_2+\frac{1}{2}\sum_{RR'}t(RR')_{02,02}(d_R^\dagger+d_R)(d_{R'}^\dagger+d_{R'})+\sum_{RR'}t(RR')_{01,02}(b_R^\dagger+b_R)(d_{R'}^\dagger+d_{R'})\\&+\sum_{RR'}\{t(RR')_{00,01}+[t(RR')_{11,01}-t(RR')_{00,01}]n(R)_1+[t(RR')_{22,01}-t(RR')_{00,01}]n(R)_2\}(b_{R'}^\dagger+b_R)\end{aligned}$$

$$+\sum_{RR'}\{\overline{K}(R)_{02}+t(RR')_{00,02}+[t(RR')_{00,02}]n(R)_1+[t(RR')_{22,02}-t(RR')_{00,02}]n(R)_2\}(d^\dagger_{R'}+d_{R'})$$

$$+\frac{1}{2}\sum_{RR'}[t(RR')_{00,00}+t(RR')_{11,11}-2t(RR')_{00,11}]n(R)_1 n(R')_1$$

$$+\frac{1}{2}\sum_{RR'}2[t(RR')_{00,00}+t(RR')_{11,22}-t(RR')_{00,11}-t(RR')_{00,22}]n(R)_1 n(R')_2$$

$$+\frac{1}{2}\sum_{RR'}[t(RR')_{00,00}+t(RR')_{22,22}-2t(RR')_{00,22}]n(R)_2 n(R')_2+\sum_R \Delta\overline{K}(R)_{02}(d^\dagger_R+d_R)\,, \qquad (10)$$

where

$$\epsilon_\nu(R)=\overline{K}(R)_{\nu\nu}+\sum_{R'}t(RR')_{\nu\nu,00}+\Delta\overline{K}(R)_{\nu\nu}\,.$$

(i) The first term in this equation is the Einstein-oscillator ground-state energy. This contribution to the energy yields the phase separation curve to good approximation.[17] We define

$$E_0(x_3)=\sum_R\overline{K}(R)_{00}+\frac{1}{2}\sum_{RR'}t(RR')_{00,00}\,. \qquad (11)$$

(ii) The second term in Eq. (10) is the fluctuation in the kinetic energy from the average crystal. This contribution to the energy is zero:

$$\sum_R\Delta\overline{K}(R)\equiv\langle\langle\Delta\overline{K}(R)\rangle\rangle=0\,. \qquad (12)$$

(iii) The third term in Eq. (10) yields the RPA displacement fluctuations (phonons). We define

$$\mathcal{H}_D=\sum_R[\epsilon(R)_1-\epsilon(R)_0]n(R)_1+\frac{1}{2}\sum_{RR'}t(RR')_{01,01}$$
$$\times(b^\dagger_R+b_R)(b^\dagger_{R'}+b_{R'})\,. \qquad (13)$$

These phonons are discussed in this form by Guyer.[18] They are essentially the phonons of Nosanow and Werthamer.[19]

(iv) The fourth term in Eq. (10) yields the RPA width fluctuation. We define

$$\mathcal{H}_W=\sum_R[\epsilon(R)_2-\epsilon(R)_0]n(R)_2+\frac{1}{2}\sum_{RR'}t(RR')_{02,02}$$
$$\times(d^\dagger_R+d_R)(d^\dagger_{R'}+d_{R'})\,. \qquad (14)$$

This Hamiltonian will be discussed in some detail below.[20,21]

(v) The fifth term in Eq. (10) is the cubic anharmonic coupling of displacement fluctuations and width fluctuations, i.e., a part of the three-phonon process. We define

$$\mathcal{H}_{DW}=\sum_{RR'}t(RR')_{01,02}(b^\dagger_R+b_R)(d^\dagger_{R'}+d_{R'})\,. \qquad (15)$$

When the displacement fluctuation and width fluctuation are regarded as independent entities $\mathcal{H}_{DW}$ couples them.

(vi) The sixth and seventh terms in Eq. (10) are the displacement and width fluctuation at R' due to deviations of the particle at R from the Einstein-oscillator ground state. We note that[22]

$$\sum_{R\neq(R')}t(RR')_{\nu\nu,01}=0 \qquad (16)$$

and

$$\overline{K}(R)_{02}+\sum_{R'\neq(R)}t(RR')_{02,00}=0, \qquad (17)$$

using the orthogonality of the $\overline{\phi}$. Both the sixth and seventh terms are of order b^3 or b^2d. We drop them.

(vii) The eighth, ninth, and tenth terms in Eq.

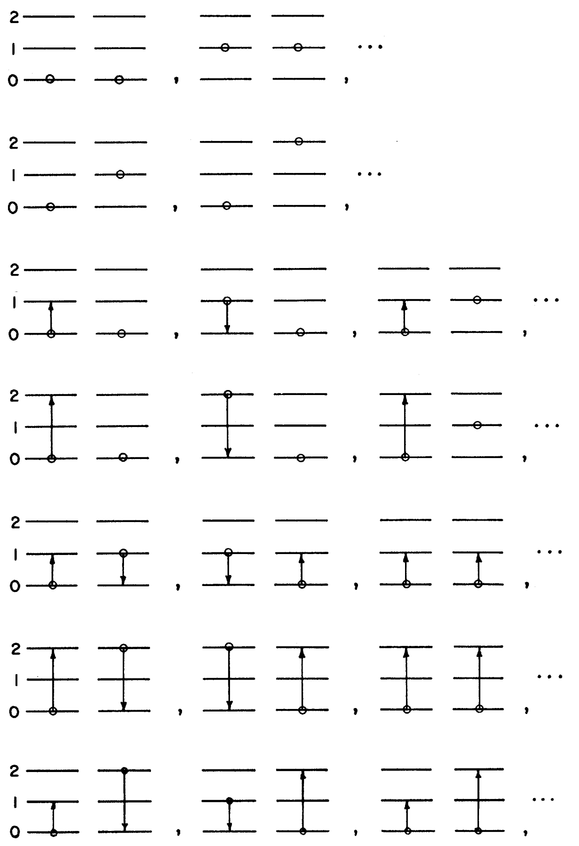


FIG. 1. Effect of the potential energy on the low-lying single-particle states. The various terms in the second quantized Hamiltonian, Eq. (7), are illustrated by what they do to the single-particle states.

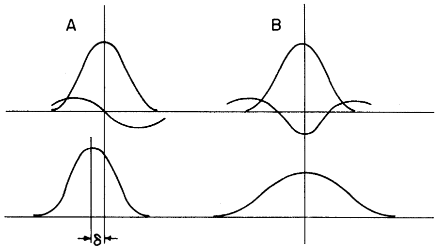


FIG. 2. Single-particle states. (a) Addition of the first excited state to the ground state causes the wave function to be displaced to the right or left. This type of admixture is called a displacement fluctuation. (b) Addition of the second excited state to the ground state causes the width of the wave function to change, without changing the position of the peak. This type of admixture is called a width fluctuation.

(10) are high-order fluctuations in the potential energy. They are proportional to n^2; we drop these terms.

(viii) The eleventh term in Eq. (10) is the transition 0 → 2 induced by the fluctuation in inverse mass at R away from $\langle m^{-1}\rangle$. This is the major term in Eq. (10) that results from the fluctuation in mass.

With the definitions in Eqs. (11)–(17) we may write the Hamiltonian in the form

$$\mathcal{H}(x_3) = E_0(x_3) + \mathcal{H}_D + \mathcal{H}_W + \mathcal{H}_{DW} + \sum_R \Delta\overline{K}(R)_{02}(d_R^\dagger + d_R)\,. \tag{18}$$

The terms $\mathcal{H}_D$, $\mathcal{H}_W$, and $\mathcal{H}_{DW}$ describe the propagation and interaction of the displacement fluctuations (DF's) and the width fluctuations (WF's). Such terms are present in the description of perfect single crystal. The last term in this equation brings about the effects due to the mass fluctuations.

For the DF Hamiltonian we have

$$\mathcal{H}_D = \sum_R [\epsilon_1(R) - \epsilon_0(R)]n(R)_1 + \frac{1}{2}\sum_{RR'} t(RR')_{01,01} \times (b_R^\dagger + b_R)(b_{R'}^\dagger + b_{R'})\,. \tag{19}$$

We take $\epsilon_1(\underline{R}) = \overline{K}_{\underline{11}} + \Delta\overline{K}(R)_{11} + \sum_{R'} t(RR')_{11,00} = \langle\langle\epsilon_1(R)\rangle\rangle$ and $\epsilon_0(R) = \overline{K}_{00} + \Delta\overline{K}(R)_{00} + \sum_{R'} t(RR')_{00,00} = \langle\langle\epsilon_0\rangle\rangle$. Both $\langle\langle\epsilon_1(R)\rangle\rangle$ and $\langle\langle\epsilon_0(R)\rangle\rangle$ are numbers independent of R. Further, we write $n(R)_1 = b_R^\dagger b_R$ (this is strictly correct if only the 0 and 1 states are involved, $n_0 + n_1 = 1$). Thus Eq. (19) takes the form

$$\mathcal{H}_D = \sum_R \Delta\epsilon_{10} b_R^\dagger b_R + \frac{1}{2}\sum_{RR'} t(RR')_{01,01} \times (b_R^\dagger + b_R)(b_{R'}^\dagger + b_{R'})\,. \tag{20}$$

It is straightforward to diagonalize this Hamiltonian employing a Bloch transformation and a Bogoliubov transformation, with the result (see Appendix A)

$$\mathcal{H}_D = \sum_q \hbar\omega_D(q)(b_q^\dagger b_q + \tfrac{1}{2})\,, \tag{21}$$

where

$$\hbar\omega_D(q) = \tfrac{1}{2}[\Delta\epsilon_{10}^2 + 2\Delta\epsilon_{10} m_D(q)]^{1/2}\,,$$

$$m_D(q) = \sum_{R' \neq (R)} e^{i\vec{q}\cdot\vec{R}-\vec{R}'} t(RR')_{01,01}\,,$$

and $\Delta\epsilon_{10} = \langle\langle\epsilon_1\rangle\rangle - \langle\langle\epsilon_0\rangle\rangle$.

For the WF Hamiltonian we have

$$\mathcal{H}_W = \sum_R [\epsilon_2(R) - \epsilon_1(R)]n_2(R) + \frac{1}{2}\sum_{RR'} t(RR')_{02,02} \times (d_R^\dagger + d_R)(d_{R'}^\dagger + d_{R'})\,. \tag{22}$$

As in the case of the DF's, we take $\epsilon_\nu(R) = \langle\langle\epsilon_\nu(R)\rangle\rangle$ and we assume that the WF's and DF's do not interfere with one another; i.e., we use $n_0 + n_2 = 1$ and write

$$\mathcal{H}_W = \sum_R \Delta\epsilon_{20} d_R^\dagger d_R + \frac{1}{2}\sum_{RR'} t(RR')_{02,02} \times (d_R^\dagger + d_R)(d_{R'}^\dagger + d_{R'})\,, \tag{23}$$

where $\Delta\epsilon_{20} = \langle\langle\epsilon_2\rangle\rangle - \langle\langle\epsilon_0\rangle\rangle$. Then, following the same steps as are employed in dealing with Eq. (20), we find

$$\mathcal{H}_W = \sum_k \hbar\omega_W(k)(d_k^\dagger d_k + \tfrac{1}{2})\,, \tag{24}$$

where

$$\hbar\omega_W(k) = \tfrac{1}{2}[\Delta\epsilon_{20}^2 + 2\Delta\epsilon_{20} m_W(k)]^{1/2}\,,$$

$$m_W(k) = \sum_{k' \neq (R)} e^{i\vec{k}\cdot\vec{R}-\vec{R}'} t(RR')_{02,02}\,.$$

The excitation created by $d_k^\dagger$ is a width fluctuation. It propagates with energy $\hbar\omega_W(k) = \frac{1}{2}[\Delta\epsilon_{20}^2 + 2\Delta\epsilon_{20} m_W(k)]^{1/2}$. For the DF we know $\lim_{q\to 0}\omega(q) \to q$; for the WF we *do not* know its $k \to 0$ behavior.

For the moment we drop the DF-WF interaction and consider the treatment of the mass-fluctuation term. We look at $\mathcal{H}_W + \mathcal{H}' \equiv \overline{\mathcal{H}}_W$,

$$\overline{\mathcal{H}}_W = \sum_R \Delta\epsilon_{20} d_R^\dagger d_R + \frac{1}{2}\sum_{RR'} t(RR')_{02,02} \times (d_R^\dagger + d_R)(d_{R'}^\dagger + d_{R'}) + \sum_R \Delta\overline{K}(R)_{02}(d_R^\dagger + d_R)\,. \tag{25}$$

We may eliminate the linear term in the WF operators d_R by introducing $\alpha_R^\dagger + \hat{a}_R$, where $\hat{a}_R$ is a number.[23] We find

$$\sum_{R'} [\Delta\epsilon_{20}\delta_{RR'} + 2t(RR')_{02,02}]\hat{a}_{R'} = -\Delta\overline{K}(R)_{02}\,,$$

$$\overline{\mathcal{H}}_W = \sum_{RR'} [\Delta\epsilon_{20}\delta_{RR'}\alpha_R^\dagger\alpha_{R'} + \tfrac{1}{2}t(RR')_{02,02}(\alpha_R^\dagger + \alpha_{R'})] + \sum_{RR'} \Delta\overline{K}(R)_{02} D_W(RR')\Delta\overline{K}(R')_{02}\,, \tag{26}$$

where $D_W(RR')$ is defined by

$$\sum_{R''} D_W(RR'')\,[\Delta\epsilon_{20}\delta_{R''R'} + 2t(R''R')_{02,02}] = -\,\delta_{RR'}\,. \tag{27}$$

We may solve Eq. (27) employing the WF dispersion relation. Since

$$[\hbar\,\omega_W(q)]^2 = \tfrac{1}{4}[\Delta\epsilon_{20}^2 + 2\Delta\epsilon_{20} m_W(q)] \tag{28}$$

and the Fourier transform of Eq. (27) is

$$(4/\Delta\epsilon_{20})\,D_W(q)[\hbar\,\omega_W(q)]^2 = -1, \tag{29}$$

we have

$$D_W(q) = -\frac{\Delta\epsilon_{20}}{4\hbar^2\omega_W(q)^2}\,. \tag{30}$$

Thus

$$D_W(RR'') = +\frac{\Delta\epsilon_{20}}{4\hbar^2 N}\sum_q \frac{e^{i\vec{q}\cdot\vec{R}-\vec{R}''}}{-\,\omega_W(q)^2}\,. \tag{31}$$

This latter formula is to be compared to the corresponding formula for the inverse of the phonon dynamical matrix (see below). Thus we can write

$$\bar{\mathcal{H}}_W = \mathcal{H}_W + \mathcal{H}_I^{(1)}, \tag{32}$$

where

$$\mathcal{H}_I^{(1)} = \sum_{RR'} \Delta\bar{K}(R)_{02} D_W(RR')\Delta\bar{K}(R')_{02} \tag{33}$$

is a direct interaction between the mass fluctuation at R and the mass fluctuation at R' communicated by the width fluctuation. For the Hamiltonian describing the system we take (we continue to ignore $\mathcal{H}_{DW}$)

$$\mathcal{H}(x_3) = E_0(x_3) + \mathcal{H}_D + \mathcal{H}_W + \mathcal{H}_I^{(1)}, \tag{34}$$

where $E_0(x_3)$, $\mathcal{H}_D$, $\mathcal{H}_W$, and $\mathcal{H}_I^{(1)}$ are given by Eqs. (11), (21), (24), and (33), respectively.

In order to estimate the consequences of $\mathcal{H}_I^{(1)}$ in Eq. (32) we need to know the strength of this interaction and its range. We discuss this question in Sec. III. From that discussion we learn that $\mathcal{H}_I^{(1)}$ is relatively weak and short ranged.

In addition to the direct interaction embodied in $\mathcal{H}_I^{(1)}$ there is a long-range interaction between mass fluctuations brought about by the displacement of the neighbors of a mass fluctuation. The term $\mathcal{H}_{DW}$ in the Hamiltonian couples the width fluctuation at R due to $\Delta K(R)_{02}$ to the displacement fluctuation at R'. Then, the displacement fluctuation at R' is communicated across the crystal by the phonons to interact with a similar displacement fluctuation at S' due to a width fluctuation at S. This process mixes b and d operators; it is most easily described in perturbation theory. We return to Eq. (18). The last term in Eq. (18) gives rise in second-order perturbation theory to a displacement fluctuation at R' described by

$$\mathcal{H}'' = \sum_{RR'} t(RR')_{01,02}\,\frac{1}{-\,\Delta\epsilon_{20}}\,\Delta\bar{K}(R')_{02}(b_R^\dagger + b_R)\,. \tag{35}$$

When $\mathcal{H}''$ is introduced into the Hamiltonian along with $\mathcal{H}_D$ we have $\mathcal{H}_D + \mathcal{H}'' = \bar{\mathcal{H}}_D$,

$$\bar{\mathcal{H}}_D = \sum_{RR'} \Delta\epsilon_{10}\delta_{RR'}\, b_R^\dagger b_R + \frac{1}{2}\sum_{RR'} t(RR')_{01,01} \times (b_R^\dagger + b_R)\,(b_{R'}^\dagger + b_{R'}) + \sum_R \Lambda(R)(b_R^\dagger + b_R)\,, \tag{36}$$

where

$$\Lambda(R) = \sum_{R'} t\,(RR')_{01,02}\,\frac{1}{-\,\Delta\epsilon_{20}}\,\Delta\bar{K}(R')_{02}\,. \tag{37}$$

We follow the same procedure in treating this Hamiltonian as we followed in treating Eq. (25). The result is

$$\bar{\mathcal{H}}_D = \mathcal{H}_D + \sum_{RR'} \Lambda(R)D_D(RR')\,\Lambda(R')\,, \tag{38}$$

where

$$\sum_{R''} D_D(RR'')[\Delta\epsilon_{10}\delta_{R''R'} + 2t(R''R')_{01,01}] = -\,\delta_{RR'}\,. \tag{39}$$

We have

$$D_D(RR'') = \frac{\Delta\epsilon_{10}}{4\hbar^2 N}\sum_q \frac{e^{i\vec{q}\cdot\vec{R}-\vec{R}''}}{-\,\omega_D(q)^2} \tag{40}$$

and

$$\hbar^2\omega_D(q)^2 = \tfrac{1}{4}[\Delta\epsilon_{10}^2 + 2\Delta\epsilon_{10} m_D(q)]\,. \tag{41}$$

We write Eq. (38) in the form

$$\bar{\mathcal{H}}_D = \mathcal{H}_D + \mathcal{H}_I^{(2)}, \tag{42}$$

where

$$\mathcal{H}_I^{(2)} = \sum_{RR'} \Lambda(R)D_D(RR')\Lambda(R')\,. \tag{43}$$

We see that both the *direct* width fluctuation coupling of $\Delta\bar{K}(R)_{02}$ to $\Delta\bar{K}(R')_{02}$ given by Eq. (33) and the *indirect* displacement fluctuation coupling given by Eq. (43) are of the same form. We write the total Hamiltonian as

$$\mathcal{H}(x_3) = E_0(x) + \mathcal{H}_D + \mathcal{H}_W + \mathcal{H}_I\,, \tag{44}$$

where

$$\mathcal{H}_I = \mathcal{H}_I^{(1)} + \mathcal{H}_I^{(2)}\,. \tag{45}$$

This is the total Hamiltonian we use in this paper. The two excitation terms $\mathcal{H}_D$ and $\mathcal{H}_W$ correct the Einstein-oscillator ground-state energy slightly and describe the thermal excitations. As we are at low temperature, we drop these terms and concentrate in the remainder of this paper on the mass-fluctuation–mass-fluctuation interaction given by Eq. (45).

III. MASS-FLUCTUATION–MASS-FLUCTUATION INTERACTION

In this section we discuss the quantitative effects of the MF-MF interaction and estimate its order of magnitude.

The MF-MF interaction is given by Eqs. (45), (43), and (33), where $D_D(RR'')$ and $D_W(RR')$ are given by Eqs. (40) and (31), respectively, and

$$\Delta\bar{K}(R)_{02} = -\frac{\hbar^2}{2}(\nabla^2)_{02}\left(\frac{1}{m(R)} - \left\langle\frac{1}{m}\right\rangle\right) . \tag{46}$$

If there is a ^{3}He atom at R we have

$$\Delta\bar{K}(R)_{02} = \Delta\bar{K}(1 - x_3)n_3(R) , \tag{47}$$

where $\Delta\bar{K} = -\frac{1}{2}\hbar^2(\nabla^2)_{02}[(1/m_3) - (1/m_4)]$. If there is a ^{4}He at R we have

$$\Delta\bar{K}(R)_{02} = -\Delta\bar{K}x_3 n_4(R) .$$

Thus we may write

$$\mathcal{H}_I^{(1)} = \sum_{RR'}\Delta\bar{K}^2 D_W(RR')[n_3(R) - x_3][n_3(R') - x_3] , \tag{48}$$

where we have used $n_3(R) + n_4(R) = 1$. Similarly, we have

$$\mathcal{H}_I^{(2)} = \sum_{RR'SS'} \vec{\Lambda}(RR') \cdot \overleftrightarrow{D}_D(R'S') \cdot \vec{\Lambda}(S'S)[n_3(R) - x_3] \times [n_3(S) - x_3] ,$$

where $\vec{\Lambda}(RR') = \Delta\bar{K}(1/\Delta\epsilon_{20})\vec{t}\,(RR')_{02,01}$. For a random distribution of ^{3}He in ^{4}He $\langle n_3(R)\rangle = x_3$ and these interactions lead to the energy

$$\langle\mathcal{H}_I\rangle = \left(\sum_R \Delta\bar{K}^2 D_W(RR) + \sum_{RR'S'} \vec{\Lambda}(RR') \cdot \overleftrightarrow{D}_D(R'S') \cdot \vec{\Lambda}(S'R)\right) x_3(1 - x_3) , \tag{49}$$

which makes a small additional contribution to the phase separation temperature calculated using $E_0(x_3)$ only.

As a consequence of the interaction described by $\mathcal{H}_I$, different arrangements of the particles in an isotopic mixture lead to different energy levels for the system. In principle these energy levels make themselves known in a variety of ways. For example, because of the quantum-mechanical tunneling of the particles in a mixture, various particle arrangements are accessible to take up the energy available from a thermal reservoir; $\mathcal{H}_I$ should be seen in thermodynamic measurements.[24,25] Further, at nondilute concentrations, an interaction between pairs of particles requires that one modify the simple picture of quantum tunneling to account for the effects of the interaction. At the very least the presence of an interaction sets a concentration threshold for the onset of MFW behavior. To quantify this discussion we require an estimate of the order of magnitude of the MF-MF interaction.

To estimate the order of magnitude of the MF-MF interaction we need to know $D_W(RR')$ and $D_D(RR')$. From Eqs. (31) and (40) we see that this in turn requires knowledge of $\omega_W(k)$ and $\omega_D(k)$. It is easy to learn as much as is necessary about $\omega_D(k)$; it is not easy to learn about $\omega_W(k)$. Thus we begin by discussing $D_D(RR')$. From Eq. (40) we have

$$\overleftrightarrow{D}_D(R'S') = \frac{3\alpha^2}{2mN}\sum_q \frac{\vec{e}_q\,\vec{e}_q}{-\omega_D(q)^2}\, e^{i\vec{q}\cdot(\vec{R}'-\vec{S}')} \tag{50}$$

or

$$\overleftrightarrow{D}_D(R'S') = \exp\left(\vec{\Delta}_{R'R} \cdot \frac{d}{d\vec{R}}\right)\exp\left(\vec{\Delta}_{S'S} \cdot \frac{d}{d\vec{S}}\right)\overleftrightarrow{D}_D(RS) , \tag{51}$$

where $\vec{\Delta}_{R'R} = \vec{R}' - \vec{R}$. Here in writing Eq. (50) we use the full three-branch generalization of the one branch $\overleftrightarrow{D}_D$ that we employed in developing the model Hamiltonian.[26] We write Eq. (51) in place of Eq. (50) to reference the point of the interaction on the location of the mass fluctuation. Using Eq. (51) in combination with Eq. (41) leads to

$$\mathcal{H}_I^{(2)} = -\sum_{RR'}\sum_{SS'}\sum_{\alpha\beta}\Lambda_\alpha(R'R)\left(\vec{\Delta}_{R'R} \cdot \frac{d}{d\vec{R}}\right)\Lambda_\beta(SS') \times \left(\vec{\Delta}_{S'S} \cdot \frac{d}{d\vec{S}}\right) D_D^{\alpha b}(RS)[n_3(R) - x_3]\,[n_3(S) - x_3] , \tag{52}$$

where we have expanded the exponentials in Eq. (51) and taken the first nonzero term, the dipole-dipole term. The sum on R' and S' has been isolated (it no longer involves $\overleftrightarrow{D}$) and can be done with the result (see Appendix B)

$$\mathcal{H}_I^{(2)} = -\tfrac{64}{9}\epsilon^2\Delta^2\sum_{RR'}\left(\frac{d}{d\vec{R}} \cdot \frac{d}{d\vec{R}}\right)D_D(RR')[n_3(R) - x_3] \times [n_3(R') - x_3] , \tag{53}$$

where $\epsilon = \Delta\bar{K}_{02}(1/\Delta\epsilon_{20})t(RR')_{02,01}$ measures the strength of the coupling of a width fluctuation at R to a displacement fluctuation at R'. (This result is achieved using the plausible assumption $\overleftrightarrow{D} = D\overleftrightarrow{I}$ that is verified by detailed calculation of $\overleftrightarrow{D}$ using the continuum approximation.) If $D_D(RR')$ is isotropic $\nabla^2 D_D$ is zero and $\mathcal{H}_I^{(2)}$ is zero.[27] But from the continuum approximation of Dederich and Leibfried[28] we know that $D(RR')$ is anisotropic and $\nabla^2 D_D$ nonzero. Using their results as described in Appendix B, we obtain

$$\nabla^2 D_D(\vec{r}) = \frac{\alpha^{-2}}{5\pi}\frac{1}{r^3}\frac{1}{C_{11}}, \tag{54}$$

where α^{-2} measures the width of the single-particle wave function [see Eq. (8)] and C_{11} is the elastic constant. We use Eq. (54) in Eq. (53) and the numerical estimates of ϵ, $\alpha^2\Delta^2$, and C_{11} and Appendix B to achieve

$$\mathcal{H}_I^{(2)} = -V_0 \sum_{RR'} \left(\frac{\Delta}{|R-R'|} \right)^3 [n_3(R) - x_3][n_3(R') - x_S] , \tag{55}$$

where $V_0 \approx 10^{-2}$ K. Thus the indirect MF-MF interaction communicated by the phonons is long range, r^{-3}, and relatively weak. There is some uncertainty in the estimate of V_0 that is not easily (or usefully) removed (see Appendix B).

The direct part of the MF-MF interaction is carried by the width fluctuations. We have made a study of this interaction employing a number of reasonable assumptions about the behavior of $\omega_W(k)$. We find the interaction to be weak, of order $\Delta \bar{K}^2 / \Delta\epsilon_{20} \approx 10^{-3} k_B \Theta_D$, and short ranged. If we had $\omega_W(k) \rightarrow 0$ as $k \rightarrow 0$ this conclusion would be invalidated. But we have no reason to believe $\omega_W(k) \rightarrow 0$ as $k \rightarrow 0$. Thus we expect $\mathcal{H}_I^{(1)}$ to contribute only to the short-range part of the interaction. The short-range part of $\mathcal{H}_I$, due to $\mathcal{H}_I^{(1)}$ and the $q \rightarrow q_D$ part of $\mathcal{H}_I^{(2)}$, is inadequately treated here. We believe it is not important to the physics we want to discuss. Thus we rewrite Eq. (45), using Eq. (55) in the form

$$\mathcal{H}(x_3) = \bar{E}_0(x_3) - V_0 \sum_{RR'} \left(\frac{\Delta}{|\vec{R} - \vec{R}'|} \right)^3 [n_3(R) - x_3] \times [n_3(R') - x_3] , \tag{56}$$

where $\bar{E}_0(x)_3$ is $E_0(x_3)$ corrected for the zero-point contributions from $\mathcal{H}_W$ and $\mathcal{H}_D$ and for the diagonal parts of $\mathcal{H}_I^{(1)}$ and $\mathcal{H}_I^{(2)}$, as in Eq. (49). We drop $\mathcal{H}_W$ and $\mathcal{H}_D$, as they now contain only thermal excitations.

IV. EFFECTS OF $\mathcal{H}_I$

The interaction discussed in Sec. III arises from deviations of the mass at a lattice site from the mass of the average crystal. This interaction is present over the entire concentration range for dilute ^{4}He in ^{3}He, for 50% mixtures, for dilute ^{3}He in ^{4}He, etc. In this section we will discuss the probable effect of the interaction at low concentrations of ^{3}He in ^{4}He on the diffusion constant, T_2 and T_1. We will attempt to determine criteria for the validity of the MFW picture in this concentration limit.

In a discussion of the *magnetic* properties of ^{3}He-^{4}He mixtures there are two extreme limiting cases that can be discussed. These are $x_3 = 1.0$, the pure-^{3}He case, and $x_3 \sim 0$, the MFW case. In this latter case the ^{3}He atoms are essentially isolated. They move by quantum-mechanical tunneling through their ^{4}He neighbors. Departures from either of these extremes lead to departures from the corresponding simple pictures. For example, as small concentrations of ^{4}He are added to pure ^{3}He a ^{4}He MFW enters the discussion. But this MFW is nonpropagating (it diffuses) by virtue of the fact that it moves in a spin-disordered medium.[6,16] The dominant mechanism for the motion of the magnetic moments continues to be the ^{3}He-^{3}He exchange process that occurs at $x_3 = 1$. But a new mechanism enters the discussion of T_1 data.[2] As the concentration of ^{4}He is further increased, the interaction between ^{4}He impurities described by $\mathcal{H}_I$ increases in importance and arrests the quantum tunneling motion that occurs in the MFW limit. For a ^{3}He at R and a ^{4}He at R' the potential energy given by $\mathcal{H}_I$ is

$$V(RR') = -V_0 \sum_{S \neq RR'} \left[\left(\frac{\Delta}{|\vec{R} - \vec{S}|} \right)^3 (1 - x_3) - \left(\frac{\Delta}{|\vec{R}' - \vec{S}|} \right)^3 x_3 \right] [n_3(S) - x_3] . \tag{57}$$

Upon exchange the potential energy of this pair is

$$V(RR') = V_0 \sum_{S \neq RR'} \left[\left(\frac{\Delta}{|\vec{R} - \vec{S}|} \right)^3 x_3 - \left(\frac{\Delta}{|\vec{R}' - \vec{S}|} \right)^3 (1 - x_3) \right] \times [n_3(S) - x_3]. \tag{58}$$

Thus the interchange leads to a change in potential energy of

$$V(R'R) - V(RR') = +V_0 \sum_{S \neq RR'} \left[\left(\frac{\Delta}{|\vec{R} - \vec{S}|} \right)^3 - \left(\frac{\Delta}{|\vec{R}' - \vec{S}|} \right)^3 \right] [n_3(S) - x_3]. \tag{59}$$

It is the comparison of this change in potential energy (essentially the gradient of $\mathcal{H}_I$) with the tunneling energy J_{34} that provides a measure of the ease with which the tunneling process can occur. Since J_{34} is expected to be of order J_{33}, $J_{33}/k_B \approx 5 \times 10^{-5}$ K, and $V_0/k_B \approx 10^{-2}$ K, we are dealing with a system in which the kinetic energy is much less than the potential energy. As a consequence the new T_1 mechanism above is modified upon increasing the concentration.[24,25] As the concentration of ^{4}He is further increased to $x_3 \approx 0.50$ the interaction comes to play a more important role; ^{3}He-^{4}He interchange is relatively slower than in the $x_3 \sim 1$ case so that ^{3}He-^{3}He exchange continues as the important motion. Further increases of the ^{4}He concentration lead to a region of hindered ^{3}He motion through a ^{4}He medium (below 1% ^{3}He in ^{4}He) and finally to MFW behavior in very dilute ^{3}He mixtures (see Fig. 3).

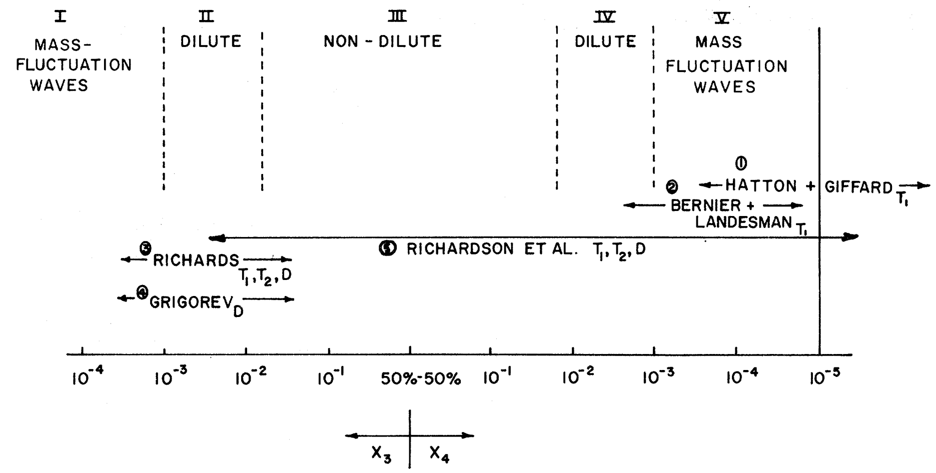


FIG. 3. Experiments on ^{3}He–^{4}He mixtures. We show the concentration regions examined in the literature. We are concerned here mainly with the dilute region, region II. The various regions have been explored in the experiments by (i) R. Giffard and J. Hatton [Phys. Rev. Lett. 18, 1106 (1967)]; (ii) Ref. 24; (iii) Refs. 10 and 11; (iv) Refs. 10–12; (v) Refs. 7–9.

A. Diffusion

Let us begin with a discussion of the very dilute ^{3}He in ^{4}He, the MFW range, of Fig. 3. In this concentration range, the ^{3}He are typically very far apart; we may assume that they form Bloch waves which propagate with dispersion relation (simple-cubic geometry)

$$\epsilon(\vec{q}) = 2J_{34}(\cos q_x\Delta + \cos q_y\Delta + \cos q_z\Delta), \tag{60}$$

where J_{34} is the rate of ^{3}He-^{4}He tunneling. The interaction between two MFW's, $+V_0(\Delta/|\vec{R}-\vec{R}'|)^3$, leads to a cross section for MFW-MFW scattering,

$$\pi\bar{r}^2 = \pi\Delta^2\left(\frac{3V_0}{J_{34}}\right)^{1/2}, \tag{61}$$

where we define $\bar{r}$ by

$$J_{34} = \left|(\vec{\Delta}\cdot\vec{\nabla})\,V_0\left(\frac{\Delta}{|\vec{r}|}\right)^3\right|_{r=\bar{r}}. \tag{62}$$

The physical idea here is that when two MFW's are closer than $\bar{r}$ from one another, they cannot continue to move by quantum-mechanical tunneling because the energy difference due to the motion of one relative to the other is greater than J_{34}. For two MFW's closer together than $\bar{r}$ relative motion can be energy conserving only upon absorption or emission of phonons. These phonon-assisted processes are incoherent (see Fig. 4). The MFW-MFW mean free path is

$$\lambda = \frac{1}{\pi}\frac{\Delta}{x_3}\left(\frac{J_{34}}{3V_0}\right)^{1/2} \tag{63}$$

and the diffusion constant in the MFW region is given by

$$D_I = \Delta J_{34}\lambda = \frac{1}{\pi}\,\Delta^2\,\frac{J_{34}}{x_3}\left(\frac{J_{34}}{3V_0}\right)^{1/2}. \tag{64}$$

Predictions for D_I based on our analysis of diffusion data in the dilute region (region II of Fig. 3) are shown in Fig. 7. A crude upper limit to the MFW region of Fig. 3 is given by requiring that the average distance between the ^{3}He particles be greater than $\bar{r}$; i.e., $\Delta/x_3^{1/3} > \bar{r}$ or

$$x_3 \ll \frac{\Delta^3}{\bar{r}^3} = \left(\frac{J_{34}}{3V_0}\right)^{3/4}. \tag{65}$$

For $J_{34}/3V_0 \approx 10^{-3}$ this is $x_3 \ll 6\times10^{-3}$. This estimate should be compared with that made below.

At concentrations greater than $\bar{x}$, which defines the transition from the MFW region to the dilute region, we have a situation in which a typical ^{3}He particle is continually strongly interacting with its neighbors. Thus the appropriate starting point is not Bloch waves, but states which have the ^{3}He localized on lattice sites, i.e., the eigenstates of the potential energy. The diffusion constant is obtained by treating the tunneling between these states in perturbation theory. Following the steps described in detail in Appendix E we find[29]

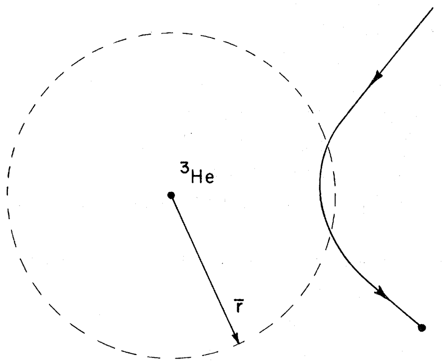


FIG. 4. MFW motion. When an ^{3}He moving through the lattice comes within a distance $\bar{r}$ of another ^{3}He, it is scattered because of the large interaction between the two particles.

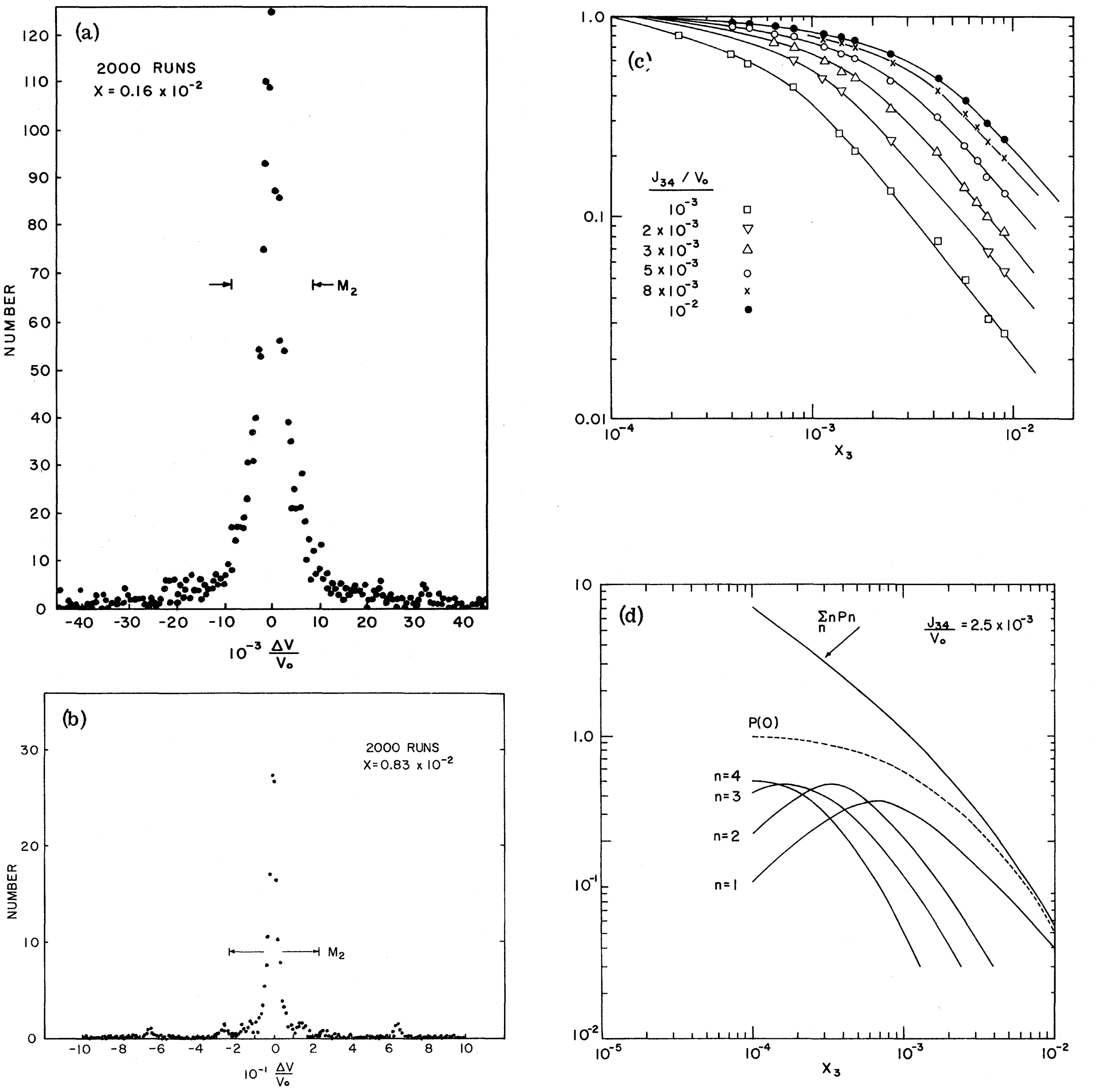


FIG. 5. Elements of the calculation of D. In (a) and (b) we show the spectra of $\Delta V(RR')$ from Eq. (57) for $x_3 = 0.0016$ and $x_3 = 0.0083$, respectively. These results are achieved by generating 2000 random configurations of particles at each concentration. In each figure we show the width of the spectrum that follows from a second-moment calculation. From spectra of this kind we can calculate $P(0)$ for various values of the ratio J_{34}/V_0. In (c) we show the results of such a calculation in the concentration range $10^{-4} \leq x_3 \leq 10^{-2}$ for $10^{-2} \leq J_{34}/V_0 \leq 10^{-3}$. In (d) we show the results of the generalization of the basic idea of the "one-step" process to include $2, 3, \ldots,$ steps. The quantities $nP_n(0)$ are plotted for $n = 1, 2, 3.$ and 4 as well as $\sum_n nP_n(0)$ for $1 \leq n \leq 10$. We also show $P(0)$, the "one-step"-only probability from (c), for comparison. All of these calculations are for the particular value of the ratio $J_{34}/V_0 = 2.5 \times 10^{-3}$. We note that at $x_3 \sim 6 \times 10^{-4} \times 1 \times P_1 \approx \sum_{n=2} nP_n(0)$, so that in $P(0)$ at this concentration half of the configurations that permit one step also permit more than one step. At $x_3 \approx 2.5 \times 10^{-3}$ at most 25% of the configurations that permit one step also permit more than one step. The diffusion results shown in Fig. 6 use $P(0)$ and $\sum_n nP_n(0)$ from this figure.

$$D = \tfrac{1}{6}\, z \Delta^2\, W_D(x_3), \tag{66}$$

where $W_D(x_3)$ is the average rate of exchange of a ^{3}He-^{4}He pair in a mixture at concentration x_3. Further, $W_D(x_3)$ is given by

$$W_D(x_3) = \pi (J_{34}/\hbar)\, P(0), \tag{67}$$

where J_{34} is the ^{3}He-^{4}He tunneling rate and $P(0)$ is the probability that the energy difference between the initial configuration (RR') and the final configuration ($R'R$) is of order J_{34} (see Appendix D). The rate $W_D(x_3)$ is the product of an attempt frequency $J_{34}/\hbar$ and the probability that the process is energetically possible. The result in Eq. (66) is the perturbation-theory result for D. It is derived in Appendix E from a Kubo formula for D so that one can see how to remove the specialization that produced it and to make improvements on it.

To calculate $P(0)$ we need to know the average number of $\Delta V(RR')$ states at zero energy. A moment calculation in which $\rho(0)$, the average density of $\Delta V(RR')$ states at zero energy, is approximated by the second moment of $\Delta V(RR')$ yields $\rho(0) = (1/V_0)[5/27x_3(1-x_3)]^{1/2}$. The calculation of higher moments contains little additional information about $\rho(0)$ even after substantial computational effort. The use of this second-moment $\rho(0)$ yields poor results for $P(0)$. We have undertaken Monte Carlo studies of the spectrum of $\Delta V(RR')$ and $P(0)$ for several values of J_{34}/V_0 as a function of concentration.[31] In Fig. 5(a) we show the spectrum $\Delta V(RR')$ for two concentrations, $x_3 = 0.0083$ and $x_3 = 0.0016$. The striking thing about these spectra is the relatively large central peak at zero energy. On each spectra we show the width given by a second-moment calculation. It is clear that such a calculation completely misses the essential feature of the spectrum for calculating $P(0)$. The central peak in the spectrum comes from configurations with particles near the plane perpendicular to the tunneling pair and bisecting their path. The edges of the spectrum are due to configurations having particles that neighbor the tunneling pair along the line joining their path. It is these configurations that dominate the second moment and lead to its inadequacy as a judge of $\rho(0)$. In Fig. 5(c) we show $P(0)$ as a function of x_3 for various choices of J_{34}/V_0. We find that for J_{34}/V_0 in the range 10^{-2}–10^{-3} $P(0)$ approaches 1 at x_3 about 10^{-3}. For all of the values of J_{34}/V_0 studied $P(0)$ is 1 at x_3 well below 10^{-3}. Thus we see that for $J_{34}/V_0 = 10^{-3}$ more than half of the configurations are within the energy interval $2J_{34}$ centered at zero for $x_3 < 7\times10^{-4}$. For $x_3 > 7\times10^{-4}$ more than half of the configurations are beyond the energy interval $2J_{34}$ and $P(0)$ is less than $\frac{1}{2}$. At $x_3 = 10^{-2}$ only 2% of the configurations are in the energy interval $2J_{34}$, $P(0) \approx 0.02$. We choose as a criterion for MFW behavior the condition that most configurations be in the energy interval $2J_{34}$. Thus for $J_{34}/V_0 = 10^{-3}$ we expect MFW behavior at $x_3 \ll 7\times10^{-4}$. We have used the results of the calculations shown in Figs. 5(a) and 5(b) to fit the diffusion data of Richards *et al.* and Grigor'ev and co-workers at 21.0 cm²/mole using Eqs. (66) and (67). We obtain the results shown in Fig. 6 using $V_0/k_B = 10^{-2}$ K and $J_{34}/k_B = 2.5\times10^{-5}$ K, i.e., $J_{34}/V_0 = 2.5\times10^{-3}$. In Fig. 6 we also show D_I as a function of x_3 calculated from Eq. (64) using $J_{34}/V_0 = 2.5\times10^{-3}$. For $J_{34}/V_0 = 2.5\times10^{-3}$ we expect MFW behavior at $x_3 \ll 1.3\times10^{-3}$.

We have attempted to improve on the "one-step" expression for D given by Eqs. (66) and (67) by writing

$$D = \frac{z\pi}{6}\,\frac{\Delta J_{34}}{\hbar}\sum_n n\Delta P_n(0),$$

where $\Delta J_{34}/\hbar$ is the velocity of the particle and $P_n(0)$ is the probability that particle is in a configuration that permits it to move distance $n\Delta$. Equations (66) and (67) are the $n=1$ approximation to this equation. We calculate $P_n(0)$ by finding the fraction of the configurations that permit a particle to make n steps (in one direction) before encountering a potential-energy barrier greater than $2J_{34}/n$. The results for $nP_n(0)$ are shown in Fig. 5(d) for $J_{34}/V_0 = 2.5\times10^{-3}$. The sum of the $nP_n(0)$ is also shown in this figure and the resulting calculation of D is shown in Fig. 6. It is clear that this generalization extends the concentration range over which the theory is valid to somewhat lower concentration, but certainly a static-configuration theory of the kind we have here must fail when sub-

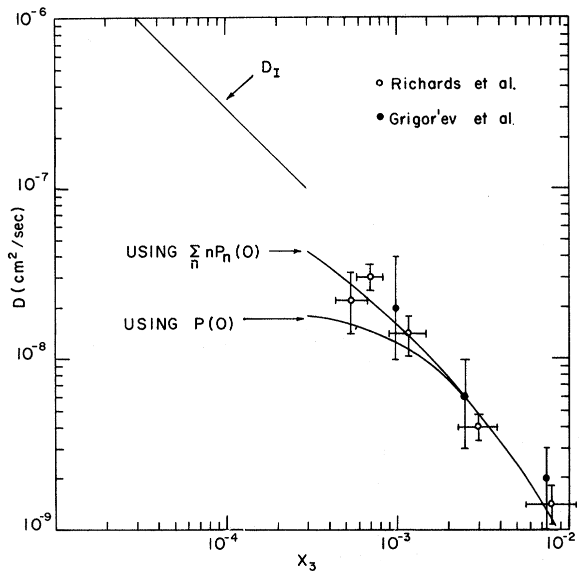


FIG. 6. Diffusion constant. Data of Richards *et al.* and Grigor'ev and co-workers are shown along with the results of the theory for two cases. The curve labeled $P(0)$ is the result using the one-step approximation that we believe to be valid at higher concentrations. The curve labeled $\sum_n nP_n$ is calculated with the many-step generalization of $P(0)$. In both cases we have fit the theory curve to the data at $x_3 = 2.5\times10^{-3}$. Since $P(0) \approx \sum_n nP_n(0)$ at this concentration, we have ignored the difference in J_{34}/V_0 that would result in using $\sum_n nP_n(0)$ or $P(0)$. The value $J_{34}/V_0 = 2.5\times10^{-3}$ comes from fitting the theory with $P(0)$ to the data at $x_3 = 2.5\times10^{-3}$.

stantial particle motion occurs. We expect this failure to occur at $x_3 \ll 10^{-3}$. The fit to the diffusion data in the range $10^{-3} < x_3 < 10^{-2}$ is quite good—but we chose J_{34} to make it good. The theory of D embodied in Eqs. (66) and (67) is valid only in this restricted concentration range. It appears that the high-concentration data, $x_3 \gtrsim 10^{-3}$, approach the theoretical D_I curve at low concentration. The virtue of our explanation of the diffusion data in the range $10^{-3} \leq x_3 \leq 10^{-2}$ is that we accomplish it using plausible values of the basic parameters. We employed the theoretical value of V_0 and found $J_{34} \approx \frac{1}{2} |J_{33}|$. A serious test of this explanation of the ^{3}He motion is made in attempting also to understand the T_2 data.

The result we have obtained for D is quite similar to that of Landesman and Winter.[30] Using a pseudospin-formulation of the diffusion problem Landesman and Winter achieve a formula similar to our equation (D4) with $\Lambda(t)$ given essentially by the first cumulant approximation of Eq. (D12). Unfortunately, the generalization of their computational procedure to handle the T_2 calculation was not carried through.

Before going on to this we note that $W_D(x_3)$ has relatively strong volume dependence. We have

$$W_D(x_3) = 2J_{34}^2 \rho(0;\, x_3), \tag{68}$$

where $\rho(0;x_3)$ goes as V_0^{-1}. Thus the volume dependence of $W_D(x_3)$ is

$$W_D(x_3) \propto J_{34}^2/V_0 \,. \tag{69}$$

This volume dependence is substantially greater than that of J_{33} alone[32] and in qualitative agreement with the observations of Richardson *et al.*[8] and Grigor'ev *et al.*[14]

B. T_2

The transverse relaxation time T_2 at zero Larmour frequency is given by[6]

$$\frac{1}{T_2(0)} = M_2(1)x_3\left[\sum_{R'\neq(R)}\left(\frac{\Delta}{|\vec{R}-\vec{R}'|}\right)^6 \int_0^{+\infty} dt\, \langle\langle n_3(R,0) \times n_3(R',0) n_3(R,t) n_3(R',t)\rangle\rangle \Big/ \sum_{R'\neq(R)}\left(\frac{\Delta}{|\vec{R}-\vec{R}'|}\right)^6\right] \tag{70}$$

where $M_2(1)$ is the second moment of a perfect single crystal of volume V at concentration $x_3 = 1$. The effect of dilution of the ^{3}He on the second moment is in the factor x_3. The important point illustrated in this formula is that a T_2 measurement senses two ^{3}He particles that must be *very close* to one another by virtue of the weighting factor r^{-6}. Thus in a T_1 and T_2 measurement we see the behavior of pairs of particles well within the interaction range $\bar{r}$. It is therefore not possible to see MFW behavior in T_1 and T_2 data. In contrast to T_1 and T_2, which see nearby pairs of particles, a diffusion measurement sees single particles. To calculate T_2 we make the approximation that the particles at R and R' move separately, i.e.,

$$\langle\langle n_3(R,0)n_3(R',0)n_3(R,t)n_3(R',t)\rangle\rangle = \langle\langle n_3(R,0)n_3(R,t)\rangle\rangle_{R'} \langle\langle n_3(R',0)n_3(R',t)\rangle\rangle_{R}\,, \tag{71}$$

where $\langle\langle n_3(R,0)n_3(R,t)\rangle\rangle_{R'}$ is the correlation function for the number operator at R, given that there is a spectator ^{3}He at R'. We further approximate this correlation function by

$$\langle\langle n(R,0)n(R,t)\rangle\rangle_{R'} = \exp[-t\overline{W}_2(x_3)_{RR'}]\,, \tag{72}$$

where $\overline{W}_2(x_3)_{RR'}$ is the rate of transition of a ^{3}He at R to a neighboring site R'' by interchange with a ^{4}He while a spectator ^{3}He is at R'. We write

$$W_2(x_3)_{RR'} = \sum_{R''} W_2(x_3;RR'')_{R'}\,. \tag{73}$$

We show examples of some of the geometries that are important to this calculation in Fig. 7. We assume that any step to R'' from R that a particle can take changes the local dipolar field at R' and contributes to the narrowing process. Thus we have

$$\overline{W}_2(x_3)_{RR'} = \sum_{R''} \overline{W}_2(x_3;RR'')_{R'}\,, \tag{74}$$

where $\overline{W}_2(x_3;RR'')_{R'}$ is the configuration average of the transition rate from R to R'' with a spectator at R'. Using Eqs. (72) and (74) in Eq. (70) leads to

$$\frac{1}{T_2(0)} = M_2(1)x_3\left[\sum_{R'\neq(R)}\left(\frac{\Delta}{|\vec{R}-\vec{R}'|}\right)^6 \frac{1}{2\overline{W}_2(x_3)_{RR'}} \Big/ \sum_{R'\neq(R)}\left(\frac{\Delta}{|\vec{R}-\vec{R}'|}\right)^6\right]. \tag{75}$$

Now, carrying out a calculation of the $\overline{W}_2(x_3;RR'')_{R'}$ following a procedure exactly analogous to that described in Appendix D for $\overline{W}_D(x_3)$ leads to

$$\overline{W}_2(x_3;RR'')_{R'} = \pi(J_{34}/h)P_{RR''}(0)_{R'}\,, \tag{76}$$

where $P_{RR''}(0)_{R'}$ is the probability that the poten-

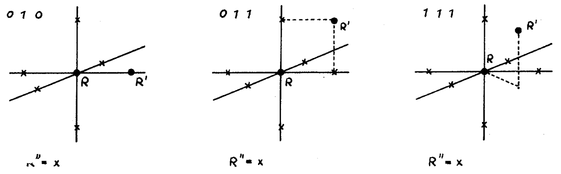
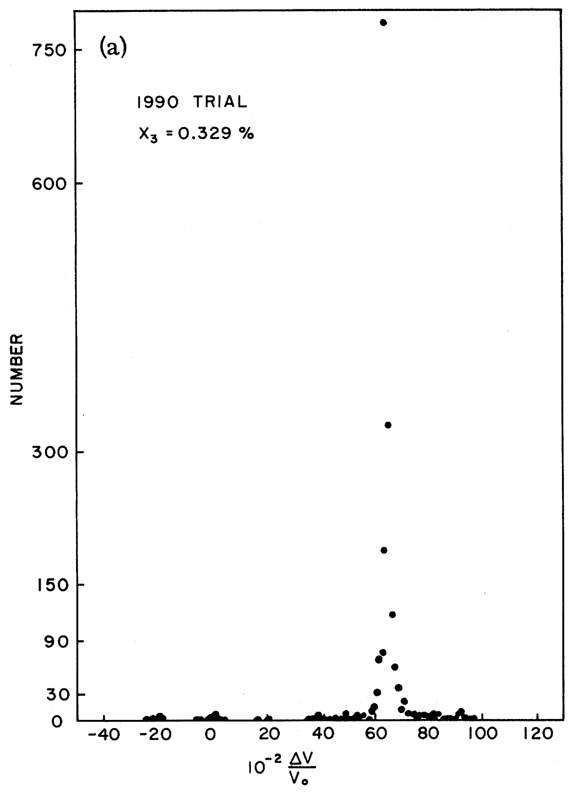
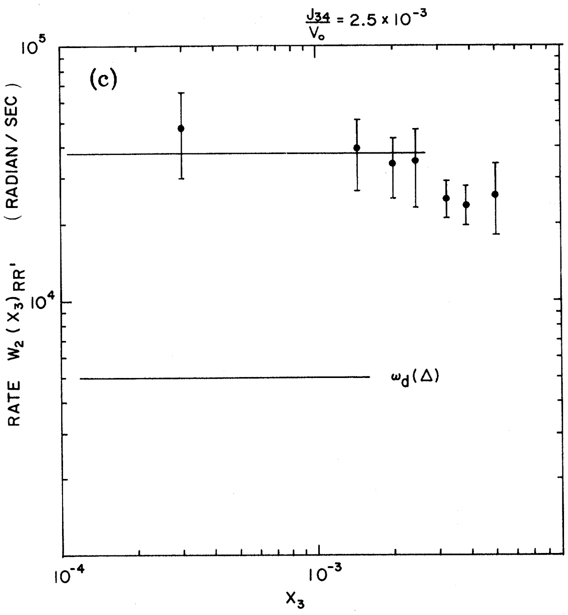


FIG. 7. Geometries. We show the three near-geometries that would be most important in a motional narrowing calculation of T_2. The spectator at R' watches the particle at R go to any of the near-neighbor sites R'' marked by an X.





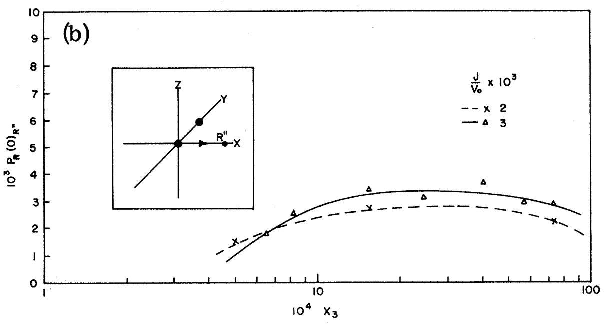


FIG. 8. Elements of the calculation of T_2. In (a) we show the spectrum of $\Delta V(RR'')$ for a spectator particle at R' for the (0, 1, 0) geometry of Fig. 7. We note that because the spectator is fixed nearby the spectrum peaks at a nonzero value. In (b) we show $P_{RR''}(0)_{R'}$ for the same geometry as (a) as a function of x_3 for two choices of the ratio J_{34}/V_0. In (c) we show $\overline{W}_2(x_3)_{RR'}$ for R and R' near neighbors. For the other geometries of Fig. 7, $\overline{W}_2(x_3)_{RR'}$ is slightly larger, and thus they will have a small contribution to T_2. In the figure we also show the dipolar frequency of a near-neighbor pair, 5×10^3 rad/sec^{-1}. It is therefore clear that a motional-narrowing picture is appropriate. The weak dependence of $\overline{W}_2(x_3)_{RR'}$ on x_3 can be understood in the following manner. In order for the ^{3}He at R' to move, two things must occur: A third ^{3}He must come nearby to balance the potential of the ^{3}He at R, and the other ^{3}He's in the system must remain far away. The first event occurs with a probability proportional to x_3, while the second occurs with a probability proportional to approximately $1/x_3$ (as indicated by the calculation of the diffusion constant). The probability that both events occur together is thus essentially independent of x_3.

tial-energy difference due to the exchange $RR''\to R''R$ is in the energy interval $2J_{34}$ centered at zero energy, given that a spectator ^{3}He particle is at R'. In Fig. 8(a) we show the spectrum of $\Delta V(RR'')_{R'}$ for $x_3 = 0.0033$. In Fig. 8(b) we show $P_{RR''}(0)_{R'}$ for the geometry shown in the inset of this figure for several values of J_{34}/V_0 as a function of x_3. In Fig. 8(c) we show the rates $\overline{W}_2(x_3)_{RR'}$ calculated from Eq. (73) for the value of the ratio J_{34}/V_0 obtained from the fit to the diffusion data above. In this figure we show the rate $\overline{W}_2(x_3)_{RR'}$ for pairs of particles in the near-geometries shown in Fig. 7. We also show in Fig. 8(c) the rigid-lattice dipolar rate of motion of each of these pairs of particles. For the rigid-lattice rate we use

$$\omega_d(RR')=\frac{\gamma\mu}{|R-R'|^3}\approx 5\times10^3\ \text{rad/sec},$$

where $\gamma = 2.04\times10^4$ rad/sec G, $\mu = 10^{-23}$ erg/G, and $|R-R'| = 3.5$ Å. We note that the frequency of particle motion given by $\overline{W}_2(x_3)_{RR'}$ is substantially greater than the frequency of motion due to the dipolar field. Thus we are justified in using the motional narrowing formalism implied by the basic equation we are employing to calculate T_2, Eq. (70). Using Eq. (76) for $\overline{W}_2(x_3)_{RR'}$ and taking only the near-neighbor contribution to the R' sum in

Eq. (75) leads to

$$\frac{1}{T_2(0)} = M_2(1) x_3 \frac{1}{\overline{W}_2(x_3)_{RR'}} . \tag{77}$$

For $M_2(1)$ we have

$$M_2(1) \approx 3.6 \omega_d (RR')^2 ,$$

so that Eq. (74) yields

$$\frac{1}{T_2} \approx 3.6 \frac{\omega_d (RR')^2 x_3}{\overline{W}_2(x_3)_{RR'}} .$$

Using the results for $\overline{W}_2(x_3)_{RR'}$ shown in Fig. 8(c) we obtain the results for T_2 shown in Fig. 9. From Fig. 9 we see that the order of magnitude and concentration dependence of the theory are in good agreement with the experimental results in the concentration range where we believe this theory to have validity.

In this paper we will not attempt to make a theory of T_1. But we note that in the dilute-concentration range on which we have focused attention there are two intrinsic frequencies J_{34} and J_{33} and a host of lower frequencies $W_2(x_3)_{RR'}$ that characterize the fundamental motions of the particles. A T_1 experiment will be sensitive to all of these motions and as a consequence its interpretation will be complex.

V. CONCLUSION

Using a single-particle picture of the particle motions we describe a ^{3}He - ^{4}He mixture using a Hamiltonian that contains an average single-crystal Hamiltonian (with an average mass at each lattice site) and a perturbation due to deviations of the mass at each site from the average mass. The mass-fluctuation perturbation is shown to lead to a mass-fluctuation–mass-fluctuation interaction. For dilute ^{3}He in ^{4}He this is essentially a ^{3}He-^{3}He interaction. This interaction is long ranged, r^{-3}, and relatively weak; its strength is about 10^{-2} K. But this energy is large compared to the energy associated with the quantum-mechanical tunneling of pairs of particles, e.g., $J_{34}/k_B \approx 10^{-5}$ K. Thus a description of the motions of ^{3}He particles through the ^{4}He medium must view the ^{3}He system as a strongly interacting system. We have explored some of the consequences of the long-range ^{3}He-^{3}He interaction on the propagation of a ^{3}He impurity. We find that the dilute concentration range $x_3 \lesssim 10^{-2}$ is broken up into two ranges: $10^{-4} \ll x_3 \lesssim 10^{-2}$, in which the interaction prevents coherent motion of a ^{3}He particle over a reasonable distance, and $x_3 \ll 10^{-3}$, in which MFW behavior is observed. We are able to give a qualitative and quantitative description of the D and T_2 data of Richards *et al.* and Grigor'ev and co-workers in the concentration range $10^{-3} \leq x_3 \leq 10^{-2}$. We conclude that data in this concentration range do not demonstrate the existence of MFN. They do provide numbers that permit a reasonable estimate of the concentration range in which MFW behavior should be seen. We estimate MFW behavior to appear in diffusion data at $x_3 \ll 10^{-3}$.

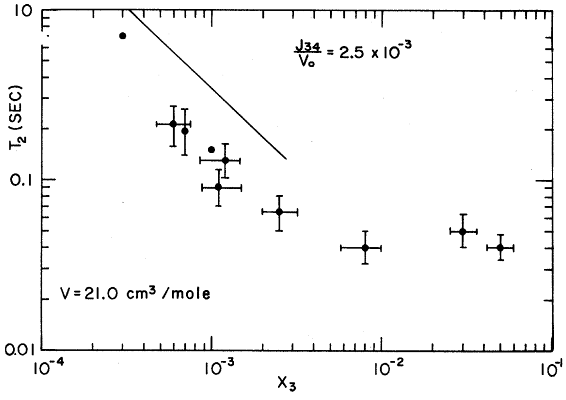


FIG. 9. T_2 as a function of concentration. The smooth curve is T_2 calculated from Eq. (77) using the results for $\overline{W}_2(x_3)_{RR'}$ from Fig. 8(c). The circles are the data of Richards *et al.* No attempt has been made to adjust either V_0 or J_{34} to improve the fit to the data. We expect that the magnitude of T_2 will depend on the crystal structure, and the strength of the ^{3}He-^{3}He interaction for near pairs. This strength will in general be different from the V_0 that appears in the diffusion constant, where only widely separated pairs are important. These considerations should not change the concentration dependence of T_2.

The calculations that we describe in this paper contain numerous approximations. Nonetheless, we believe that the essential features of the physics are described correctly. Unquestionably there is room for quantitative improvement of many aspects of the theory.

We have made a theory applicable to the dilute region $10^{-3} \leq x_3 \leq 10^{-2}$. For the MFW region, $x_3 \ll 10^{-3}$, we have only been able to make a theory of the diffusion constant. It remains then for a theory of T_1 and T_2 in the MFW region to be developed.

ACKNOWLEDGMENTS

We are grateful to Paul Tofts, M. G. Richards, and R. C. Richardson for very helpful conversations.

APPENDIX A: DIAGONALIZATION OF $\mathcal{H}_D$ AND $\mathcal{H}_W$

In this Appendix we diagonalize the Hamiltonians in Eqs. (21) and (24). These Hamiltonians are of the form

$$\mathcal{H} = \sum_{RR'} 2A(RR') a^\dagger_R a_{R'} + \sum_{RR'} B(RR') (a^\dagger_R a^\dagger_{R'} + a_R a_{R'}) , \tag{A1}$$

where for the displacement fluctuations we have

$$A_D(RR') = \tfrac{1}{2}(\epsilon_1 - \epsilon_0)\,\delta_{RR'} + \tfrac{1}{2}\,t(RR')_{01,01}\,, \qquad \text{(A2)}$$

$$B_D(RR') = \tfrac{1}{2}\,t(RR')_{01,01}\,, \qquad \text{(A3)}$$

and for the width fluctuations we have

$$A_W(RR') = \tfrac{1}{2}(\epsilon_2 - \epsilon_0)\,\delta_{RR'} + \tfrac{1}{2}\,t(RR')_{02,02}\,, \qquad \text{(A4)}$$

$$B_W(RR') = \tfrac{1}{2}\,t(RR')_{02,02}\,. \qquad \text{(A5)}$$

We employ a Bloch transformation to write

$$\mathcal{H} = \sum_k A(k)\,(a_k^\dagger a_k + a_{-k}^\dagger a_{-k}) + \sum_k B(k)\,(a_k^\dagger a_{-k}^\dagger + a_k a_{-k})\,, \qquad \text{(A6)}$$

and transform to the α_k defined by

$$\alpha_k^\dagger = u_k a_k^\dagger - v_k a_{-k}\,, \qquad \text{(A7)}$$

$$\alpha_k = u_k a_k - v_k a_{-k}^\dagger\,, \qquad \text{(A8)}$$

where $[\alpha_k, \alpha_k^\dagger] = 1$. Thus we obtain

$$\mathcal{H} = \sum_k 2\epsilon(k)\alpha_k^\dagger \alpha_k + \sum_k [\epsilon(k) - A(k)]\,, \qquad \text{(A9)}$$

where

$$\epsilon(k)^2 = A(k)^2 - B(k)^2\,. \qquad \text{(A10)}$$

For the displacement fluctuations we have

$$A_D(k) = \tfrac{1}{2}\Delta\epsilon_{10} + \tfrac{1}{2}\,m_D(k)\,, \qquad \text{(A11)}$$

$$B_D(k) = \tfrac{1}{2}\,m_D(k)\,, \qquad \text{(A12)}$$

and

$$\epsilon_D(k)^2 = \tfrac{1}{4}[\Delta\epsilon_{10}^2 + 2\Delta\epsilon_{10}\,m_D(k)]\,. \qquad \text{(A13)}$$

For the width fluctuations we have

$$A_W(k) = \tfrac{1}{2}\Delta\epsilon_{20} + \tfrac{1}{2}\,m_W(k)\,, \qquad \text{(A14)}$$

$$B_W(k) = \tfrac{1}{2}\,m_W(k)\,, \qquad \text{(A15)}$$

and

$$\epsilon_W(k)^2 = \tfrac{1}{4}[\Delta\epsilon_{20}^2 + 2\Delta\epsilon_{20}\,m_W(k)]\,. \qquad \text{(A16)}$$

In a displacement-fluctuation approximation to the ground-state energy of the average crystal we have

$$\mathcal{H} = E_0(x) + \sum_k [\epsilon_D(k) - A_D(k)]\,, \qquad \text{(A17)}$$

where $E_0(x) = N\epsilon_0$; thus

$$\langle \mathcal{H} \rangle_{T=0} = \sum_k [\epsilon_0 + \epsilon_D(k) - A_D(k)]\,. \qquad \text{(A18)}$$

Since we must have $\epsilon_D(k) \to 0$ and $k = 0$ we can write $m_D(k) = -\tfrac{1}{2}\Delta\epsilon_{10}S(k)$, where $S(k) \to 1$ as $k \to 0$. We also have $\epsilon_0 = u_0 \tfrac{3}{2}\bar{\epsilon}$ and $\epsilon_1 - \epsilon_0 = \bar{\epsilon}$; thus

$$\langle \mathcal{H} \rangle_{T=0} = E_0(x) + \frac{\bar{\epsilon}}{2}\sum_k \{[1 - S(k)]^{1/2} - [1 - \tfrac{1}{2}S(k)]\}\,. \qquad \text{(A19)}$$

The second term in this expression is the correction to the Einstein-oscillator ground-state energy due to improving that approximation to include the phonons. In the discussion of phase separation we do *not* include this correction.[17]

APPENDIX B: DETERMINATION OF THE MAGNITUDE OF $\mathcal{H}_I$

The ^{3}He-^{3}He interaction is given by $\mathcal{H}_I \approx \mathcal{H}_I^{(2)}$ and

$$\mathcal{H}_I^{(2)} = \sum_{RR'} \Delta\bar{K}_{02}^2\,\mathfrak{D}(RR')\,[n_3(R) - x_3][n_3(R') - x_3]\,, \qquad \text{(B1)}$$

where $\Delta\bar{K}_{02} = -\tfrac{1}{2}\hbar^2(\nabla^2)_{02}[(1/m_3) - (1/m_4)]$,

$$\mathfrak{D}(RR') = \sum_{SS'} \vec{t}(RS)_{02,01}\,\frac{1}{\Delta\epsilon_{20}} \cdot \overleftrightarrow{D}(SS') \circ \frac{1}{\Delta\epsilon_{20}} \times \vec{t}(S'R')_{01,02}\,, \qquad \text{(B2)}$$

and $\overleftrightarrow{D}(SS')$ is the inverse of the dynamical matrix, i.e.,

$$\sum_{\alpha\beta}\sum_{S''} D_{\alpha\beta}(SS'')\,\Phi_{\alpha\beta}(S''S') = -\delta_{SS'}\overleftrightarrow{I}\,. \qquad \text{(B3)}$$

We use the definition of ϵ from Eq. (C18) to write

$$\mathcal{H}_I = \sum_{RR'}\sum_{SS'} \vec{\epsilon}(RS) \cdot \overleftrightarrow{D}(SS') \cdot \vec{\epsilon}(S'R') \times [n_3(R) - x_3][n_3(R') - x_3]\,; \qquad \text{(B4)}$$

here $\vec{\epsilon}(RS)$ and $\vec{\epsilon}(R'S')$ are nonzero [see Eq. (C13)] for S and S' near neighbors of R and R'. To evaluate Eq. (B4) we need $\overleftrightarrow{D}(SS')$, given by

$$\overleftrightarrow{D}(SS') = -\frac{3\alpha^2}{2mN}\sum_q \frac{\vec{e}_q\vec{e}_q}{\omega(\vec{q})^2}\, e^{i\vec{q}\cdot\vec{S}-\vec{S}'} \qquad \text{(B5)}$$

in the form

$$\overleftrightarrow{D}(SS') = e^{\vec{\Delta}_{SR}\cdot\vec{D}_R}\, e^{\vec{\Delta}_{S'R'}\cdot\vec{D}_{R'}}\,\overleftrightarrow{D}(RR')\,, \qquad \text{(B6)}$$

where $\vec{\Delta}_{SR} = \vec{S} - \vec{R}$ and $\vec{D}_R = d/d\vec{R}$. If we expand $\overleftrightarrow{D}(SS')$ about $\overleftrightarrow{D}(RR')$ the first nonzero contribution to $\mathcal{H}_I$ comes from the Δ^2 term,[16]

$$\overleftrightarrow{D}(SS') = -\left(\vec{\Delta}_{RS} \cdot \frac{d}{d\vec{R}}\right)\left(\vec{\Delta}_{R'S'} \cdot \frac{d}{d\vec{R}}\right)\overleftrightarrow{D}(RR')\,, \qquad \text{(B7)}$$

since $d/d\vec{R} = -d/d\vec{R}'$. Using Eq. (B7) in Eq. (B4) leads to

$$\mathcal{H}_I = -\sum_{RR'}\sum_{SS'}\sum_{\alpha\beta} \epsilon_\alpha(RS)\left(\vec{\Delta}_{RS} \cdot \frac{d}{d\vec{R}}\right)\epsilon_\beta(R'S') \times \left(\vec{\Delta}_{R'S'} \circ \frac{d}{d\vec{R}}\right) D_{\alpha\beta}(RR')\,[n_3(R) - x_3][n_3(R') - x_3]\,. \qquad \text{(B8)}$$

The sum on S for fixed R is zero except for $\vec{\epsilon}(RS)$ parallel to $\vec{\Delta}_{RS}$; thus we have

$$\mathcal{H}_I = -g^2\epsilon^2\Delta^2 \sum_{RR'}\left(\frac{d}{d\vec{R}} \circ \frac{d}{d\vec{R}}\right) D(RR')\,[n_3(R) - x_3] \times [n_3(R') - x_3]\,, \qquad \text{(B9)}$$

where $g^2 = \sum_{R'}(e_{RR'}^z)^2$, $e_{RR'}^z$ is the z component of the unit vector from $\vec{R}$ to $\vec{R}'$, and we have assumed

$\overleftrightarrow{\mathrm{D}}(RR') = \overleftrightarrow{\mathrm{I}}D(RR')$.

To evaluate $\nabla^2 D(RR')$ we use the continuum approximation of Dederich and Leibfried.[28] Thus we have

$$D(\vec{\mathrm{r}}) = -(3\alpha^2/8\pi r)g(\Omega)\,, \tag{B10}$$

where $g(\Omega)$ is a function on the unit sphere. A factor $\frac{1}{2}3\alpha^2$ appears in this formula and in Eq. (B5) due to the difference in dimensions in the dynamical matrix in a single-particle picture and in a phonon picture; see Eq. (C16). For bcc ^{3}He we have to good approximation

$$g(\Omega) = (1/C_{11})(2.5 - 0.5\cos 4\theta)\,. \tag{B11}$$

We get Eq. (B11) from (a) noting that the ratio of the elastic constants for bcc ^{3}He is the same as for copper, (b) using an analytic approximation for the result for copper displayed in Fig. 6 of Leibfried and Dederich, and (c) ignoring the off-diagonal components of g, $g(\Omega) = g_{11}(\Omega) = g_{22}(\Omega) = g_{33}(\Omega)$. Now

$$\nabla^2 D(\vec{\mathrm{r}}) = -\left(\frac{3\alpha^2}{8\pi r^3}\right)\frac{0.5}{C_{11}}\,g''(\theta)\,, \tag{B12}$$

where $g''(\theta) = 16\cos^4\theta + 4\sin^4\theta(\cos\theta/\sin\theta)$. We replace $g''(\theta)$ in Eq. (B12) by its angular average. Then

$$\nabla^2 D(\vec{\mathrm{r}}) = \frac{\alpha^2}{5\pi}\left(\frac{1}{r^3}\right)\frac{1}{C_{11}}\,. \tag{B13}$$

Using Eq. (B13) in Eq. (B9) leads to

$$\mathcal{H}_I = -\frac{1}{5\pi}\frac{g^2\epsilon^2\alpha^2\Delta^2}{C_{11}\Delta^3}\sum_{RR'}\left(\frac{\Delta}{|\vec{\mathrm{R}}-\vec{\mathrm{R}}'|}\right)^3[n_3(R) - x_3]$$
$$\times[n_3(R') - x_3]\,. \tag{B14}$$

We have $g = \frac{8}{3}$, $\epsilon^2 = \frac{1}{2}\alpha^2\Delta^2 \times 10^{-4}\Delta\epsilon_{10}^2$, $C_{11}\Delta^3 \approx \Delta\epsilon_{10}$, and $\Delta\epsilon_{10} \approx k_B\Theta_D$. Thus

$$\mathcal{H}_I = -V_0\sum_{RR'}\left(\frac{\Delta}{|\vec{\mathrm{R}}-\vec{\mathrm{R}}'|}\right)^3[n_3(R) - x_3][n_3(R') - x_3]\,, \tag{B15}$$

where $V_0 \approx 10^{-2}$ K.

In this paper we are dealing with a displacement-displacement interaction in an hcp material. The calculation above is for a bcc material. But the order of magnitude of the effect we are describing is not strongly sensitive to this difference. Thus we use Eq. (B15) to characterize the ^{3}He-^{3}He interaction in hcp helium. We expect V_0 to be of order 10^{-2} K in this material and mildly volume dependent.

APPENDIX C: CALCULATION OF $t(RR')_{02,02}$, $t(RR')_{01,02}$, $t(RR')_{01,01}$, AND u_R

We employ the single-particle states (we drop the bar on α^2)

$$\overline{\phi}_R^{(0)} = A_0\exp[-\tfrac{1}{2}\alpha^2(\vec{\mathrm{x}}-\vec{\mathrm{R}})^2], \tag{C1}$$

$$\overline{\phi}^{(1\mu)} = A_1(x_\mu - R_\mu)\exp[-\tfrac{1}{2}\alpha^2(\vec{\mathrm{x}}-\vec{\mathrm{R}})^2], \tag{C2}$$

and

$$\overline{\phi}_R^{(2)} = A_2[1 - \tfrac{2}{3}\alpha^2(\vec{\mathrm{x}}-\vec{\mathrm{R}})^2]\exp[-\tfrac{1}{2}\alpha^2(\vec{\mathrm{x}}-\vec{\mathrm{R}})^2]. \tag{C3}$$

The general t-matrix element $t(RR')_{\alpha\beta,\gamma\delta}$ is given by

$$t(RR')_{\alpha\beta,\gamma\delta} = \int d\vec{\mathrm{x}}\int d\vec{\mathrm{x}}'\,\overline{\phi}_R^{\alpha}(\vec{\mathrm{x}})\overline{\phi}_R^{(\beta)}(\vec{\mathrm{x}})$$
$$\times t(\vec{\mathrm{x}}-\vec{\mathrm{x}}')\overline{\phi}_{R'}^{(\gamma)}(\vec{\mathrm{x}}')\overline{\phi}_{R'}^{(\delta)}(\vec{\mathrm{x}}'). \tag{C4}$$

The matrix element $t(RR')_{02,02}$ is given by

$$t(RR')_{02,02} = \int d\vec{\mathrm{x}}\int d\vec{\mathrm{x}}'\,\overline{\phi}_R^{(0)}(x)^*\overline{\phi}_R^{(2)}(\vec{\mathrm{x}})t(\vec{\mathrm{x}}-\vec{\mathrm{x}}')$$
$$\times\overline{\phi}_{R'}^{(0)}(\vec{\mathrm{x}}')\overline{\phi}_{R'}^{(2)}(\vec{\mathrm{x}}'). \tag{C5}$$

In order to calculate this matrix element it is useful to note that

$$\frac{d}{dx^\mu}(\overline{\phi}_R^{(0)})^2 = -A_0^2 2\alpha^2(x^\mu - R^\mu)\exp[-\alpha^2(\vec{\mathrm{x}}-\vec{\mathrm{R}})^2] \tag{C6}$$

and

$$\frac{d^2}{(dx^\mu)^2}(\overline{\phi}_R^{(0)})^2 = A_0^2 2\alpha^2[2\alpha^2(x^\mu - R^\mu)^2 - 1]$$
$$\times\exp[-\alpha^2(\vec{\mathrm{x}}-\vec{\mathrm{R}})^2]. \tag{C7}$$

Thus

$$\nabla^2(\overline{\phi}_R^{(0)}) = A_0^2 6\alpha^2[\tfrac{2}{3}\alpha^2(\vec{\mathrm{x}}-\vec{\mathrm{R}})^2 - 1]$$
$$\times\exp[-\alpha^2(\vec{\mathrm{x}}-\vec{\mathrm{R}})^2] \tag{C8}$$

and

$$\overline{\phi}_R^{(0)}\overline{\phi}_R^{(2)} = -\frac{1}{6\alpha^2}\frac{A_2}{A_0}\nabla^2(\overline{\phi}_R^{(0)})^2. \tag{C9}$$

We use this latter result to write

$$t(RR')_{02,02} = \frac{A_2^2}{36A_0^2\alpha^4}\int d\vec{\mathrm{x}}\int d\vec{\mathrm{x}}\,\nabla^2(\overline{\phi}_R^{(0)})$$
$$\times(\nabla')^2(\overline{\phi}_{R'}^{(0)})^2 t(\vec{\mathrm{x}}-\vec{\mathrm{x}}'). \tag{C10}$$

Now integration by parts and $\nabla = -\nabla'$ implies

$$t(RR')_{02,02} = \frac{1}{24\alpha^4}\cdot\int d\vec{\mathrm{x}}\int d\vec{\mathrm{x}}'\,|\overline{\phi}_R^{(0)}(\vec{\mathrm{x}})|^2$$
$$\times|\overline{\phi}_{R'}^{(0)}(\vec{\mathrm{x}}')|^2(\nabla^2)^2 t(\vec{\mathrm{x}}-\vec{\mathrm{x}}') \tag{C11}$$

or

$$t(RR')_{02,02} = \frac{1}{24\alpha^4}\langle(\nabla^2)^2 t(\vec{\mathrm{x}}-\vec{\mathrm{x}}')\rangle.$$

Before discussing this term in detail we calculate $t(RR')_{01,02}$ using a similar procedure. We have

$$t(RR')_{01,02} = \int d\vec{\mathrm{x}}\int d\vec{\mathrm{x}}'\,\overline{\phi}_R^{(0)}\overline{\phi}_R^{(1\mu)}$$
$$\times t(\vec{\mathrm{x}}-\vec{\mathrm{x}}')\overline{\phi}_{R'}^{(0)}\overline{\phi}_{R'}^{(2)}. \tag{C12}$$

Using Eq. (C6) we have

$$\overline{\phi}_R^{(0)}\overline{\phi}_R^{(1\mu)} = -\frac{A_1}{A_0}\frac{1}{2\alpha}\nabla_\mu(\overline{\phi}^{(0)})^2. \tag{C13}$$

We combine Eq. (C13) with Eq. (C9) to write Eq. (C12) in the form

$$t(RR')_{01,02} = \frac{1}{12}\frac{1}{\alpha^3}\frac{A_1A_2}{A_0^2}\int d\vec{x}\int d\vec{x}'\,\nabla^2(\overline{\phi}_R^{(0)})^2 \times t(\vec{x}-\vec{x}')\nabla'(\overline{\phi}_{R'}^{(0)})^2. \tag{C14}$$

Using integration by parts and $\nabla = -\nabla'$ leads to

$$t(RR')_{01,02} = +\frac{1}{12}\frac{1}{\alpha^3}\int d\vec{x}\int d\vec{x}'\,|\overline{\phi}_R^{(0)}|^2 \times |\overline{\phi}_{R'}^{(0)}|^2\,\nabla_\mu\nabla^2 t(\vec{x}-\vec{x}') \tag{C15}$$

or

$$t(RR')_{01,02} = \frac{1}{12\alpha^3}\langle\nabla_\mu\nabla^2 t(\vec{x}-\vec{x}')\rangle.$$

Similarly to calculate $t(RR')_{01,01}$ we use Eq. (C13) twice with the result

$$t(RR')_{01,01} = \frac{1}{4\alpha^2}\frac{A_1^2}{A_0^2}\int d\vec{x}\int d\vec{x}'\,\nabla_\mu(\overline{\phi}_R^{(0)})^2 \times t(\vec{x}-\vec{x}')\nabla'(\overline{\phi}_{R'}^{(0)})^2, \tag{C16}$$

$$t(RR')_{01,01} = -\frac{2}{3}\circ\frac{1}{\alpha^2}\int d\vec{x}\int d\vec{x}'\,|\overline{\phi}_R^{(0)}|^2 \times |\overline{\phi}_{R'}^{(0)}|^2\,\nabla_\mu\nabla_\nu t(\vec{x}-\vec{x}'), \tag{C17}$$

or

$$t(RR')_{01,01} = -\frac{2}{3\alpha^2}\langle\nabla_\mu\nabla_\nu t(\vec{x}-\vec{x}')\rangle.$$

Order-of-magnitude estimates of these matrix elements can be accomplished using phenomenological arguments. The spring constant that yields the phonons is $\gamma \approx z\langle\nabla^2 t\rangle$. Thus we have

$$\langle\nabla^2 t\rangle = \tfrac{1}{2}3\alpha^2\,t(RR')_{01,01} = \gamma/z$$

or

$$t(RR')_{01,01} \approx (3/2\alpha^2)(\gamma/z).$$

We also have

$$t(RR')_{01,02} \approx \gamma_D(1/\alpha\Delta)t(RR')_{01,01},$$

where γ_D is the phonon Grüneisen constant. Finally, for $t(RR')_{02,02}$ we would guess

$$|t(RR')|_{02,02} \approx [\gamma_D^2/(\alpha\Delta)^2]\,|t(RR')_{01,01}|.$$

For $\gamma_D = 3$ and $\alpha\Delta \approx 6$ (^{3}He at 20 cm^3/mole)

$$|t(RR')_{01,01}| \approx 2\,|t(RR')_{01,02}| \approx 4\,|t(RR')_{02,02}|.$$

All of these matrix elements are rapidly converging functions of $|R-R'|$ for $|R-R'|$ large; $t(RR')_{01,01} \sim |R-R'|^{-8}$, $t(RR')_{01,02} \sim |R-R'|^{-9}$, and $t(RR')_{02,02} \sim |R-R'|^{-10}$.

For the displacement at R' a neighbor of R (there is a mass fluctuation at R) we have

$$\vec{u}_{R'} = 2x_{01}\frac{1}{\Delta\epsilon_{10}}\vec{t}(R'R)_{01,02}\frac{1}{\Delta\epsilon_{20}}\Delta\overline{K}_{02} = \frac{2x_{01}}{\Delta\epsilon_{10}}\vec{\epsilon}, \tag{C18}$$

where we define

$$\vec{\epsilon} = \Delta\overline{K}_{02}\frac{1}{\Delta\epsilon_{20}}\vec{t}(RR')_{02,01}. \tag{C19}$$

We write Eq. (C18) in the form

$$\vec{\epsilon} = \vec{u}_{R'}\frac{\Delta\epsilon_{10}}{2x_{01}}. \tag{C20}$$

Using $x_{01} = 1/\sqrt{2\alpha}$ and $u_{R'} \approx 10^{-2}\Delta$ from the numerical studies of Mullin[33] and Glyde[34] we have

$$|\epsilon| = \frac{1}{\sqrt{2}}\frac{\alpha\Delta}{100}\Delta\epsilon_{10}. \tag{C21}$$

We use ϵ, a measure of the mass-fluctuation–displacement-coupling, to determine $\mathcal{H}_I$. See Appendix B and Eq. (53) of the text.

APPENDIX D: TRANSITION RATE

In this appendix we calculate the rate of tunneling of a ^{3}He-^{4}He pair that sits in the potential-energy field of their neighbors. We take the states of the system to be defined by the location of the pair of particles we study (they start at RR' and go to $R'R$) and the location of the background particles (denoted by a Greek index). The states we are concerned with are these "particle-configuration" states denoted by $(RR';\mu)$, $(R'R;\nu)$, etc. The rate of transition from $(RR';\mu)$ to $(R'R;\nu)$ is given by

$$W = \frac{2\pi}{\hbar^2}\sum_{\mu\nu}P_\mu\,|t(RR')_{\mu\nu}|^2\,\delta(E_{RR'}^\mu - E_{R'R}^\nu), \tag{D1}$$

where P_μ is the probability that the background is in state μ, $t(RR')_{\mu\nu}$ is the matrix element for the transition $RR' \to R'R$ as the background goes from μ to ν, and $E_{RR'}^\mu$ is the energy in the configuration $(RR';\mu)$. We assume that the matrix element for the transitions $(RR';\mu)$, $(RR';\nu)$ factor in the form

$$t(RR')_{\mu\nu} = t(RR')\langle\Phi_{RR'}^\mu|\Phi_{R'R}^\nu\rangle, \tag{D2}$$

where $t(RR')$ describes the motion of the pair from RR' to $R'R$ (it is $|J_{34}|$) and $\langle\Phi_\mu|\Phi_\nu\rangle$ is the overlap integral for the background wave functions. Thus we have

$$W = \frac{2\pi}{\hbar^2}|t(RR')|^2\sum_{\mu\nu}P_\mu\,|\langle\Phi_{RR'}^\mu|\Phi_{R'R}^\nu\rangle|^2\,\delta(E_{RR'}^\mu - E_{R'R}^\nu). \tag{D3}$$

Following the standard procedures for exponentiating the δ function and using the completeness of the $\Phi_{RR'}^\nu$ for description of the background we have

$$W = \frac{2\pi}{\hbar^2}|t(RR')|^2\frac{1}{2\pi}\int_{-\infty}^{+\infty}dt\,\Lambda(t), \tag{D4}$$

where

$$\Lambda(t) = \langle\langle RR' \,|e^{-i\mathcal{H}_{RR'}t}\, e^{i\mathcal{H}_{R'R}t}|RR'\rangle\rangle \tag{D5}$$

and $|RR'\rangle$ denote the complete set of background states based on the RR' configuration for the initial pair. Here we have

$$\mathcal{H}_{RR'} = \mathcal{H}_0 - V_0 \sum_{S\neq(RR')} \left[\left(\frac{\Delta}{|\vec{R}-\vec{S}|}\right)^3 (1-x_3) - \left(\frac{\Delta}{|\vec{R}'-\vec{S}|}\right)^3 x_3\right] [n_3(S) - x] \tag{D6}$$

and

$$\mathcal{H}_{R'R} = \mathcal{H}_0 - V_0 \sum_{S\neq(RR')} \times\left[-\left(\frac{\Delta}{|\vec{R}-\vec{S}|}\right)^3 x_3 + \left(\frac{\Delta}{|\vec{R}'-\vec{S}|}\right)^3 (1-x_3)\right][n_3(S) - x], \tag{D7}$$

where $\mathcal{H}_0$ represents the configuration-independent part of the Hamiltonian. If we write $\mathcal{H}_{RR'} = \mathcal{H}_0 + V_{RR'}$ and $\mathcal{H}_{R'R} = \mathcal{H}_0 + V_{R'R}$ we have $\mathcal{H}_{R'R} = \mathcal{H}_{RR'} + V_{R'R} - V_{RR'} = \mathcal{H}_{RR'} + \Delta V_{RR'}$. Now we use a mixed representation to write Eq. (D5); i.e., we write

$$\begin{aligned}\Lambda(t) &= \langle\langle RR'\,|e^{-i\mathcal{H}_{RR'}t}\, e^{i\mathcal{H}_{R'R}t}|\,RR'\rangle\rangle \\ &= \langle\langle RR'\,|\,Te^{iA(t)}\,|RR'\rangle\rangle,\end{aligned} \tag{D8}$$

where

$$A(t) = \frac{1}{\hbar}\int_0^t \Delta V_{RR'}(t')_1\,dt'. \tag{D9}$$

In this equation $\Delta V_{RR'}$ is given by

$$\Delta V_{RR'} = + \sum_{S\neq(RR')} V_0\left[\left(\frac{\Delta}{|\vec{R}-\vec{S}|}\right)^3 - \left(\frac{\Delta}{|\vec{R}'-\vec{S}|}\right)^3\right][n_3(S) - x_3] \tag{D10}$$

and the subscript one on $\Delta V_{RR'}(t')_1$ means that the time evolution of $\Delta V_{RR'}$ is due to $\mathcal{H}_{RR'}$. We can make an analytic approximation by employing a cumulant expansion of Eq. (D8), keeping the first few terms

$$\langle\langle Te^{iA(t)}\rangle\rangle = e^{iC_1(t)}\, e^{-(1/2!)C_2(t)}\ldots, \tag{D11}$$

where

$$C_1(t) = \frac{1}{\hbar}\left\langle\!\!\left\langle T\int_0^t dt'\,\Delta V_{RR'}(t')_1\right\rangle\!\!\right\rangle, \tag{D12}$$

$$C_2(t) = \frac{1}{\hbar^2}\left\langle\!\!\left\langle T\int_0^t dt'\int_0^t dt''\,\Delta V_{RR'}(t')_1 \Delta V_{RR'}(t'')_1\right\rangle\!\!\right\rangle, \tag{D13}$$

etc. To get an idea of the content of this low-order approximation consider the short-time behavior of $\langle\langle Te^{iA(t)}\rangle\rangle$ so that we can deal with Eqs. (D12) and (D13) quite simply. If we define $\Delta V(RR'; S)$ by

$$\Delta V_{RR'} = \sum_S \Delta V(RR'; S)\,[n_3(S) - x_3], \tag{D14}$$

we have

$$C_1(t) = \frac{1}{\hbar}\sum_S \Delta V(RR';\, S)\int_0^t dt'[\langle\langle n_3(S;\, t')\rangle\rangle - x_3] = 0$$

and for short times

$$C_2(t) = \frac{t^2}{\hbar^2}\sum_S \Delta V(RR';\, S)\sum_{S'} \Delta V(RR'; S') \times \langle\langle[n_3(S) - x_3][n_3(S') - x_3]\rangle\rangle. \tag{D15}$$

We can rearrange the sum SS' in the form

$$C_2(t) = \frac{t^2}{\hbar^2}\left(\sum_S V(RR'; S)^2 x_3(1-x_3) + {\sum_{SS'}}' \Delta V(RR'; S)\Delta V(RR'S')[\langle\langle n_3(S)n_3(S')\rangle\rangle - x_3^2]\right).$$

If we assume $n_3(S)$ is independent of $n_3(S')$, the second term is zero. If there is correlation between $n_3(S)$ and $n_3(S')$ we can write $\langle\langle n_3(S)\, n_3(S')\rangle\rangle = x_3^2 + x_3 g(SS')$ and obtain

$$C_2(t) = +\frac{t^2}{\hbar^2}\left(\sum_S \Delta V(RR';\, S)^2 x_3(1-x_3) + \sum_{SS'} \Delta V(RR'; S)\,\Delta V(RR';\, S')\,x_3 g(SS')\right). \tag{D16}$$

We drop the second term in this equation and write $C_2(t) = t^2(1/\hbar^2)\langle W^2\rangle x_3(1-x_3)$, where

$$\langle W^2\rangle = \sum_S \Delta V(RR';\, S)^2. \tag{D17}$$

It is not necessary to make these approximations.

Using Eqs. (D12)–(D17) we write Eq. (D11) in the form

$$\Lambda(t) = \exp\{-t^2/[2\langle W^2\rangle x_3(1-x_3)/\hbar^2]\}. \tag{D18}$$

We use this expression to do the time integral in Eq. (D4), with the result

$$W = \sqrt{2\pi}\;\frac{|t(RR')|}{\hbar}\;\frac{|t(RR')|}{[x_3(1-x_3)\langle W^2\rangle]^{1/2}}. \tag{D19}$$

We understand this result in terms of $|t(RR')|/\hbar$ being the attempt frequency for ^{3}He-^{4}He exchange and $|t(RR')|/[x_3(1-x_3)\langle W^2\rangle]^{1/2}$ being the probability that this energy difference between the two particle arrangements is of order $|t(RR')|$, i.e., that the transition $RR' \rightarrow R'R$ is energy conserving.

The numerical work we have done on the spectrum of $\Delta V(RR')$ suggests that the result in Eq. (D19) is qualitatively correct but quantitatively quite inaccurate. With the hindsight provided by Monte Carlo analysis we might proceed as follows. The average called for in Eq. (D8) or in the cumulants is an average over configurations. We divide the configurations into two groups, the "near" configurations and the "far" configurations. The configurations that belong to these two groups are determined from the Monte Carlo analysis discussed above. The near configurations involve particles near the trajectory of the tunneling pair that contribute to the background states in Fig. 5(a). The far configurations are those that contribute to the central peak. We break up $\langle\langle RR' | \cdots || RR' \rangle\rangle$ thus

$$\langle\langle RR' | \cdots | RR' \rangle\rangle = \langle\langle RR' | \cdots | RR' \rangle\rangle_N + \langle\langle RR' | \cdots | RR' \rangle\rangle_F \,, \tag{D20}$$

where the subscripts N and F mean that the averages called for are over near and far configurations. The near configurations span a large region of $\Delta V(RR')$ and dominate the second-moment approximation to the spectrum above, Eq. (D17). The far configurations are swamped in a second-moment calculation but they are precisely the configurations of importance in determining W. A cumulant expansion of (D20) carried to second order leads to

$$\Lambda(t) = P_N \exp\left(-\frac{t^2}{\hbar^2}\langle W_N^2\rangle\right) + P_F \exp\left(-\frac{t^2}{\hbar^2}\langle W_F^2\rangle\right) , \tag{D21}$$

where

$$\langle W_N^2\rangle = \left\langle\left\langle \sum_{S\neq(RR')} \Delta V(RR';\, S)^2 \right\rangle\right\rangle_N \tag{D22}$$

and

$$\langle W_F^2\rangle = \left\langle\left\langle \sum_{S\neq(RR')} \Delta V(RR';\, S)^2 \right\rangle\right\rangle_F \,. \tag{D23}$$

Here $P_N + P_F = 1$, P_N is the weight of the background states in Fig. 5(a), and P_F is the weight of the central peak. As the concentration is lowered $P_N \to 0$ and $P_F \to 1$. Equation (D21) is a far more satisfactory representation of $\Lambda(t)$ than Eq. (D18).

Regardless of the analytic approximations that are possible to represent the spectrum of ΔV, we can always write (as a lowest-order approximation)

$$W = \pi[\,|t(RR')|/\hbar\,]P(0) \,, \tag{D24}$$

where $P(0)$ is the number of states in energy interval $2|t(RR')|$ centered at energy zero. We can determine $P(0)$ analytically or by a Monte Carlo calculation. It is Eq. (D24) that we use in Sec. IV in conjunction with a Monte Carlo analysis of the spectrum of $\Lambda(t)$. In writing Eq. (D24) we are simply interpreting the basic perturbation-theory formula

$$W = 2\pi[\,|t(RR')|^2/\hbar\,]\rho(0) \,,$$

where $\rho(0)$ is the density of states. Our interpretation makes sense only so long as $\rho(E)$ changes slowly over interval $2|t(RR')|$ near $E=0$. From what we know about the spectrum of $\Delta V(RR')$ we know $\rho(E)$ is flat near $E=0$ until fewer than half of the $\Delta V(RR')$ are outside of the interval $2|t(RR')|$. But if fewer than half of the states are outside $2|t(RR')|$ our picture of hindered tunneling is not valid. Equation (D24) is valid in the concentration range in which we use the perturbation-theory formula. Note that the discussion here is independent of the specific character of the averaging procedure employed. Thus the entire discussion can be carried through for an averaging procedure carried out with a spectator particle, as is required for T_2 calculations.

APPENDIX E: DIFFUSION CONSTANT

The basic formula for the conductivity is given by

$$\sigma_{zz}(0) = \int_0^\infty dt \int_0^\beta d\lambda \left(\lim_{V\to+\infty} \frac{1}{V}\right) \frac{1}{Z} \mathrm{Tr}\,[\rho_0 I_z(-\lambda) I_z^*(t)] \,. \tag{E1}$$

Employing a sequence of manipulations similar to those illustrated by Brinkman and Rice[35] leads to

$$\sigma_{zz}(0) = +\frac{\beta}{\pi V}\int_{-\infty}^{+\infty} d\omega' e^{-\beta\omega'} \frac{1}{Z}\mathrm{Tr}[I_z(0)\,\mathrm{Im}G(\omega') \times I_z^*(0)\,\mathrm{Im}G(\omega')] \,, \tag{E2}$$

where

$$G(\omega') = \frac{1}{\omega' - \mathcal{H}}, \quad \mathrm{Im}G(\omega\ ) = \frac{1}{2}\left(\frac{1}{\omega' - \mathcal{H} + i\epsilon} - \frac{1}{\omega' - \mathcal{H} - i\epsilon}\right) . \tag{E3}$$

The current operator is given by (we use a ficititious charge e)

$$I_z(0) = -i\frac{e}{\hbar}\sum_{RR'} t(RR')(R_z - R_z')c_R^\dagger c_{R'} \,. \tag{E4}$$

Using Eq. (E4) in Eq. (E5) leads to

$$\sigma_{zz}^{(0)} = \frac{\beta}{\pi V}\frac{e^2}{\hbar^2}\sum_{RR'}\sum_{SS'} t(RR')t(SS')(R_z - R_z')(S_z' - S_z) \times \int_{-\infty}^{+\infty} d\omega' e^{-\beta\omega'}\frac{1}{Z}\mathrm{Tr}[c_{R'}^\dagger c_R\,\mathrm{Im}G(\omega')c_S^\dagger c_{S'}\,\mathrm{Im}G(\omega')] \,. \tag{E5}$$

We assume (a) that the current operator does not change the background state (see Appendix D) and (b) that the site diagonal component of $G(\omega')$ is adequate; i.e.,

$$\mathrm{Tr}[c_R^\dagger c_{R'}\,\mathrm{Im}G(\omega')c_S^\dagger c_{S'}\,\mathrm{Im}G(\omega')] = \delta_{RS'}\delta_{R'S}\,\mathrm{Tr}\,\mathrm{Im}G_{RR'}(\omega')\,\mathrm{Im}G_{R'R}(\omega') \,, \tag{E6}$$

where

$$G_{RR'}(\omega) = \frac{1}{\omega - \mathcal{H}_{RR'}} \,. \tag{E7}$$

Here $\mathcal{H}_{RR'}$ is given below Eq. (D7). Using

$$\mathrm{Im}G_{RR'}(\omega) = -\frac{i}{2}\int dt\, e^{-i(\omega-\mathcal{H}_{RR'})t} \tag{E8}$$

we can obtain

$$\sigma_{zz}(0) = \pi\frac{\beta}{V}\frac{e^2}{\hbar^2}\sum_{RR'} t(RR')^2(R_z - R_z')^2\frac{1}{2\pi}\int_{-\infty}^{+\infty} dt\,\Lambda(t)\ , \tag{E9}$$

where $\Lambda(t)$ is given by Eq. (D5). If we combine Eq. (E9) with Eq. (D4) we obtain

$$\sigma_{zz}(0) = \frac{\beta e^2}{2V}\sum_{RR'}(R_z - R_z')^2\, W\ , \tag{E10}$$

where W is given by Eq. (D4). Using the Einstein relation, $D_{zz} = (k_B T/e^2)(V/N)\sigma_{zz}(0)$, leads to

$$D_{zz}(0) = \frac{1}{2}\sum_{R'}(R_z - R_z')^2\, W\ . \tag{E11}$$

For z near neighbors at Δ we have (approximately)

$$D_{zz}(0) = \tfrac{1}{6} z\,\Delta^2 W\ . \tag{E12}$$

Using Eq. (D24) for W leads to

$$D_{zz}(0) = \frac{\pi z}{6}\Delta^2\,\frac{|t(RR')|}{\hbar}\,P(0)\ . \tag{E13}$$

---

*Supported in part by the National Science Foundation and the Alexander von Humboldt Foundation.

[1] A. F. Andreev and I. M. Lifshitz, Zh. Eksp. Teor. Fiz. 56, 2054 (1969) [Sov. Phys.–JETP 29, 1107 (1969)].

[2] R. A. Guyer and L. I. Zane, Phys. Rev. Lett. 24, 660 (1970).

[3] Radha Balakrishnan and Robert V. Lange, Phys. Rev. A 3, 496 (1971).

[4] V. Balakrishnan and Radha Balakrishnan, J. Low Temp. Phys. 9, 255 (1972).

[5] Radha Balakrishnan, thesis (Brandeis University, 1970) (unpublished).

[6] R. A. Guyer, R. C. Richardson, and L. I. Zane, Rev. Mod. Phys. 43, 532 (1971).

[7] D. S. Miyoshi, R. N. Cotts, A. S. Greenberg, and R. C. Richardson, Phys. Rev. A 2, 870 (1970).

[8] A. S. Greenberg, W. G. Thomlinson, and R. C. Richardson, Phys. Rev. Lett. 27, 179 (1971).

[9] A. S. Greenberg, W. C. Thomlinson, and R. C. Richardson, J. Low Temp. Phys. 8, 3 (1972).

[10] M. G. Richards, J. Pope, and A. Widom, Phys. Rev. Lett. 29, 708 (1972).

[11] M. G. Richards, J. Pope, and A. Widom, in Proceedings of the Thirteenth International Conference on Low Temperature Physics, Boulder, Colorado, 1972 (to be published).

[12] V. N. Grigor'ev, B. N. Essel'son, V. A. Mikheev, V. A. Slusarev, M. S. Strzhemechry, and Yu E. Shulman, J. Low Temp. Phys. 13, 65 (1973).

[13] V. N. Grigor'ev, B. N. Essel'son, V. A. Mikheev, and Yu. E. Shul'man, Zh. Eksp. Teor. Fiz. Pis'ma Red. 17, 25 (1973) [JETP Lett. 17, 16 (1973)].

[14] V. N. Grigor'ev, B. N. Essel'son, and V. A. Mikheev, Zh. Eksp. Teor. Fiz. Pis'ma Red. 18, 289 (1973) [JETP Lett. 18, 169 (1973)].

[15] R. A. Guyer and L. I. Zane, Phys. Rev. 188, 445 (1969).

[16] R. A. Guyer, J. Low Temp. Phys. 8, 424 (1972). An interaction of the kind we are discussing occurs between vacancies; such interactions are well known in solid-state systems. See Ref. 28 or the literature on hydrogen in metals for examples.

[17] M. T. Takemori and R. A. Guyer (unpublished). In this paper a modification of standard quantum-crystal methods is used to do the average crystal problem and discuss phase separation. See also B. Brandow, Ann. Phys. 74, 112 (1972).

[18] R. A. Guyer, in *Selected Topics in Physics*, edited by X. Laredo and Y. Jurisic (Reidel, Dordrect, Holland, 1973).

[19] L. H. Nosanow and N. R. Werthamer, Phys. Rev. Lett. 15, 618 (1965); see also W. Brenig, Z. Phys. 171, 60 (1963).

[20] H. Horner, Phys. Rev. Lett. 29, 556 (1972).

[21] W. C. Kerr and A. Sjolander, Phys. Rev. B 1, 2723 (1970).

[22] There are three degenerate $n=1$ levels of the Hartree states. We use only one of these—that which permits displacement in the z direction. Thus matrix elements like $t(RR')_{\nu\nu,01}$ have a vector property that we do not exhibit unless it is necessary to understand the detailed structure of a formula. See, for example, the discussion in Appendix B. For doing numerical work we use a phonon spectrum with three polarizations.

[23] This is a standard procedure; see Ref. 16 or H. Wagner and J. Swift, Z. Phys. 239, 182 (1970).

[24] M. Bernier and A. Landesman, Solid State Commun. 8, 2151 (1970).

[25] R. A. Guyer, Phys. Rev. A 5, 2541 (1972).

[26] A. A. Maradudin, in *Phonons and Phonon Interactions* (Benjamin, New York, 1964), p. 424.

[27] H. Wagner, Phys. Rev. Lett. 25, 31 (1970).

[28] P. H. Dederich and G. Leibfried, Phys. Rev. 188, 1175 (1969).

[29] R. Kubo, Can. J. Phys. 34, 1279 (1956).

[30] A. Landesman and J. M. Winter, in Ref. 11.

[31] We have done the numerical work for a simple-cubic geometry. This leads to possible numerical errors in $P(0)$ of order 1.

[32] S. B. Trickey, W. P. Kirk, and E. D. Adams, Rev. Mod. Phys. 44, 668 (1972).

[33] W. J. Mullin, Phys. Rev. Lett. 20, 254 (1968).

[34] H. R. Glyde, Phys. Rev. 177, 202 (1969).

[35] W. Brinkman and M. Rice, Phys. Rev. B 1, 1324 (1970).