MASS FLUCTUATION WAVES

R. A. Guyer*
Department of Physics and Astronomy, University of Massachusetts, Amherst, Massachusetts 01002

and

L. I. Zane
Department of Physics, Duke University, Durham, North Carolina 27706

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The motion of a ^{4}He particle through a ^{3}He lattice leads to an excitation (a mass fluctuation wave) which strongly couples to the phonons and provides a simple explanation for the NMR data in solid ^{3}He-^{4}He mixtures.

There is a large body of NMR data on solid ^{3}He-^{4}He mixtures at low ^{4}He concentration[1-4] which are difficult to understand within the framework of the conventional model of the solid system.[5] The purpose of this paper is to suggest the existence of a new excitation in solid mixtures, the mass fluctuation wave, which provides an explanation for part of the observed data.

Consider a pure ^{3}He lattice containing a single ^{4}He atom. This ^{4}He atom can move through the lattice by a tunneling process in which it and a ^{3}He atom change place. This tunneling process occurs in a ^{3}He lattice of volume V at a rate which is comparable with the rate of the tunneling motion of two ^{3}He atoms. Thus it is characterized by an energy on the order of the exchange energy in the pure ^{3}He lattice, $J(V)$. We construct a Hamiltonian which describes this motion. To describe pure solid ^{3}He we employ a complete set of harmonic oscillator (HO) wave functions at each lattice site, the $\varphi_R{}^\alpha(\vec{x})$ given by

$$[T(x)+\tfrac{1}{2}k(\vec{R}-\vec{x})^2]\varphi_R{}^\alpha(\vec{x})=E_\alpha\varphi_R{}^\alpha(\vec{x}), \tag{1}$$

where we choose the single-particle potential, i.e., k, such that the ground-state wave function $\varphi_R{}^0(\vec{x})=A_0\exp[-(\alpha^2/2)(\vec{x}-\vec{R})^2]$ agrees well with the ground-state single-particle wave function for the solid. We will take the solid to be described by

$$H_e=\sum_i T(\vec{x}_i)+\tfrac{1}{2}\sum_{ij}{}'\nu_e(|\vec{x}_i-\vec{x}_j|), \tag{2}$$

where $\nu_e(\vec{x}_i-\vec{x}_j)$ is an appropriate "t-matrix" interaction potential for particles in the low-lying HO states of Eq. (1).[6] For a pure ^{3}He lattice in the ground state, the interesting motions of the particles are found by second quantizing H_e using the field operator

$$\psi^\dagger(\vec{x})=\sum_R\varphi_R{}^0(\vec{x})b_{R0}{}^\dagger, \tag{3}$$

where $b_{R0}{}^\dagger$ creates particle at lattice site R in the ground state, $\alpha=0$. The resulting Hamiltonian is

$$H=\sum_R\epsilon_0 b_{R0}{}^\dagger b_{R0}+\sum_{RR'}t(RR')b_{R0}{}^\dagger b_{R'0}+\varphi\sum_R b_{R0}{}^\dagger b_{R0}{}^\dagger b_{R0}b_{R0}, \tag{4}$$

where ϵ_0 is a single-particle energy ($\langle H_0\rangle\approx N\epsilon_0\approx$ ground-state energy), $t(RR')=\int dx\,\varphi_R{}^0(x)T(x)\varphi_{R'}{}^0(x)$ is a tunneling energy, and $\varphi=\int d\vec{x}d\vec{x}'|\varphi_R{}^0(x)|^2|\varphi_R{}^0(x')|^2\nu_e(\vec{x}-\vec{x}')$ is a hard-core energy associated with the double occupation of lattice site.[7] When spin variables are associated with the states the tunneling term and hard-core term admit particle motion through the lattice which is identifiable by the spin variables. This particle motion is described by

$$H_x=\sum_{RR'}{}'\frac{t(RR')^2}{-\varphi}(b_{R\uparrow}{}^\dagger b_{R'\uparrow}+b_{R\downarrow}{}^\dagger b_{R'\downarrow})(b_{R'\uparrow}{}^\dagger b_{R\uparrow}+b_{R'\downarrow}{}^\dagger b_{R\downarrow}), \tag{5}$$

an exchange Hamiltonian[8] for the cooperative tunneling motion in which the intermediate state is a virtual vacancy state.[9] (We have dropped the state index.) See Fig. 1.

Consider the addition of a single ^{4}He atom at lattice site R. If the ^{4}He atom does not tunnel, the ground state of the crystal is N-fold degenerate (each of the degenerate states is designated by the latice site at which the ^{4}He is found). If the ^{4}He atom tunnels through the lattice this degeneracy is removed. The tunneling motion of the ^{4}He atom through the lattice is described by

$$H_4=(-1/\varphi)\sum_{RR'}{}'[t(RR')_3 b_{R3}{}^\dagger b_{R'3}+t(RR')b_{R4}{}^\dagger b_{R'4}][t(RR')_3 b_{R'3}{}^\dagger b_{R3}+t(RR')_4 b_{R'4}{}^\dagger b_{R4}], \tag{6}$$

where $b_{R3}^\dagger$ creates a particle at R in the ^{3}He ground state, $b_{R4}^\dagger$ creates a particle at R in the ^{4}He ground state, and $t(RR')_3$ and $t(RR')_4$ are the tunneling energies for ^{3}He and ^{4}He particles, respectively.[10] The important motion for splitting the degenerate ground state is given by the operator combination

$$b_{R3}^\dagger b_{R'3} b_{R'4}^\dagger b_{R4} = b_{R3}^\dagger b_{R4} b_{R'4}^\dagger b_{R'3} = a_R^- a_{R'}^+,$$

where $a_R^+ = b_{R4}^\dagger b_{R3}$ is an operator which creates a mass fluctuation at R. The equation of motion for $a_k^+ = \sum_R a_R^+ \exp(i\vec{k}\cdot\vec{R})$ is determined from Eq. (6) and leads to the dispersion relation which in the $k \to 0$ limit is

$$h\omega(k) = \Delta\epsilon_4 + \tfrac{1}{2}Z(t_3 t_4/\varphi)(k\Delta)^2, \qquad (7)$$

where $\Delta\epsilon_4$ is a finite energy shift, Z = the number of near neighbors, Δ = the near-neighbor distance, and the crystal has been assumed to have simple-cubic structure. We regard a_k^+ as the creation operator for a mass fluctuation wave of wave vector $\vec{k}$; Eq. (7) gives its dispersion relation.

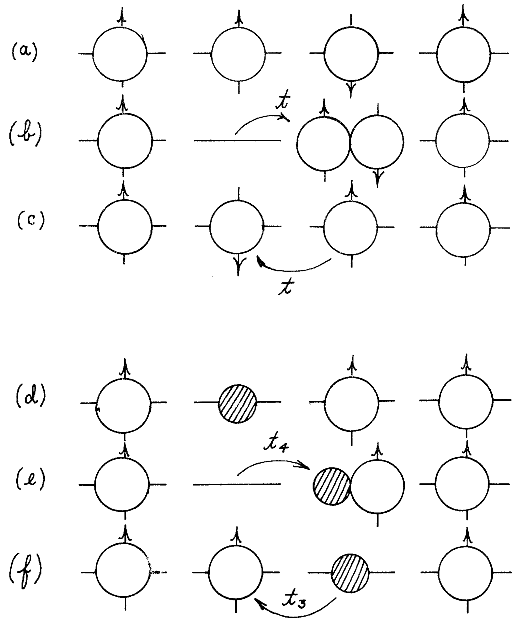

FIG. 1. Tunneling in pure ^{3}He and mixtures. In (a)-(c) the elemental tunneling motion involving a pair of ^{3}He atoms is shown schematically; $b_{R\downarrow}^\dagger b_{R'\downarrow}$ takes the system from (a) to (b) with matrix element t; $b_{R'\uparrow}^\dagger b_{R\uparrow}$ returns the system to the original state with a reversal of spins. The intermediate state (b) is termed a virtual vacancy state; it becomes a full fledged vacancy state when the hole (empty lattice site) and particle (doubly occupied lattice site) wander away from one another. In (d)-(f) the elemental tunneling motion by which a ^{4}He atom moves through the ^{3}He medium is drawn.

We may understand this result in analogy to the Li impurity in KCl[11] or to a single inverted spin in a ferromagnetic system.[12] In all of these cases a tagged particle (^{4}He, Li, or $\downarrow$) has n equivalent sites available to it. The tagged particle moves among these equivalent sites via a tunneling process and splits the degeneracy that would exist in the absence of that process. The excitations which split the degeneracy are the mass fluctuation waves, tunneling states, and spin waves, respectively.

The mass fluctuation waves interact strongly with the phonons. An estimate of the strength of this interaction can be given by calculating the rate at which energy is transferred from the phonons to the mass fluctuation wave due to the perturbation

$$H' = \sum_R \tfrac{1}{2}\Delta M_R(t)\dot{\vec{U}}_R^{\,2} \qquad (8)$$

in the phonon Hamiltonian; here $M_R(t) = (M_R - M_3)A(t)$ where $A(t)$ is taken to be a noise source with characteristic frequency $zJ' = zt_3t_4/\varphi \approx zJ$. The rate of transfer of energy between the phonons and the mass fluctuation waves is

$$de_p/dt = (Nx)(1.2\times10^3)(\delta M/M)^2\hbar(zJ')^2 \times (T/\Theta_D)^6 k_B T(\beta_4 - \beta_L), \qquad (9)$$

where $\beta = (k_B T)^{-1}$, x is the concentration of ^{4}He atoms, and Θ_D is the Debye temperature of the solid. Equating this energy change to the change in energy of the ^{4}He system,

$$dE_4/dt = (1/T_1^{\,0})C_4 T_L^{\,2}(\beta_4 - \beta_L), \qquad (10)$$

we find

$$\frac{1}{T_1^{\,0}} \approx 256\left(\frac{\delta M}{M}\right)^2 \frac{k_B T}{\hbar}\left(\frac{T}{\Theta_D}\right)^6. \qquad (11)$$

Here we have taken the specific heat of the He^4 system to be that of a gas of excitations with the dispersion relation given by Eq. (7):

$$\frac{C_4}{N} = x\lambda \tfrac{3}{8} z k_B \left(\frac{\hbar J'}{k_B T}\right)^2, \qquad (12)$$

where $\lambda = 12z$. At $V = 20.0$ cm^3/mole and $T \approx 0.5$°K we find $T_1{}^0 \sim 10^{-2}$ sec. This magnitude for the intrinsic process is on the order of the T_1 required to explain the data of Hatton and Giffard.[3] We make the following conjecture. At low temperatures the presence of ^{4}He impurities leads to relaxation of the ^{3}He exchange system according to the topology[13] shown in Fig. 2.[5] The Zeeman system, exchange system, and ^{4}He system come to equilibrium in times short compared with the time required for the energy in these coupled systems to be dumped into the phonons by the mass fluctuation waves. For this topology the experimentally observed long time is[14]

$$\frac{1}{T_1} = \frac{1}{T_1{}^0} \frac{k_4}{k_Z + k_3 + k_4}, \tag{13}$$

where $k_4 = x(\frac{3}{8})\lambda z k_B (J')^2$, $k_3 = \frac{3}{8} z k_B J^2$, and $k_Z = \frac{1}{4} k_B (g\mu_B H_0)^2$. $T_1{}^{-1}$ given by Eq. (13) has (1) the correct Zeeman frequency and ^{4}He concentration dependence due to the topology, (2) the correct temperature and volume dependence due to the intrinsic relaxation time $T_1{}^0$, and (3) the order of magnitude of the experimentally observed $T_1{}^{-1}$.

The fast time coupling the ^{3}He bath to the ^{4}He bath is

$$\frac{1}{\tau_{34}} \approx \frac{1}{\hbar}\left(\frac{t_3 t_4}{\varphi}\right) x \approx Jx, \tag{14}$$

a time much shorter than T_1, as it must be. The topology in Fig. 2 explains why τ_{34} has been observed in the NMR experiments where there was every reason to expect it.[6]

In conclusion:

(1) We suggest that ^{4}He impurities in ^{3}He move by a tunneling process and that these motions are manifested as mass fluctuation waves.

(2) When the energy associated with the ^{4}He motion and the attendant phonon perturbation are incorporated into the description of NMR relaxation phenomena, a quantitative and qualitative description of the low-concentration ($x < 500$ ppm) and low-temperature NMR data results.

(3) The ^{4}He bath introduced above provides a qualitatively correct explanation for the anomalous specific heat results of Bernier and Landesman[4] and others.[1] We do not wish to discuss this subject at length here.[15] However, we note that the extra specific heat observed in the experiments of Bernier and Landesman[4] is of the order of magnitude of those we find for the ^{4}He bath with $J' \approx J$.

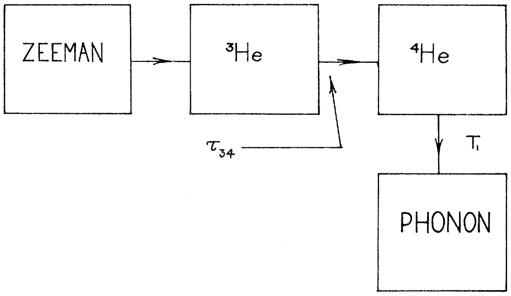

FIG. 2. Energy flow for NMR relaxation at $T < 0.5$°K. An energy bath associated with the mass fluctuation waves is introduced. This bath comes rapidly to equilibrium with the ^{3}He (exchange system) and provides a strong link by which energy involved in particle motion (i.e., the ^{3}He bath and ^{4}He bath) can be transferred to the phonons.

(4) There is every reason to expect similar effects, which however would be more difficult to observe, for ^{3}He in a nearly pure ^{4}He lattice.

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tism (Wiley, New York, 1965).

[13] The basic idea of this topology is strongly suggested by the data in Refs. 3 and 4 and has been discussed by R. C. Richardson, private communication, and C. M. Varma, private communication, among others.

[14] The system of equations giverning the behavior of a multiple-bath system is discussed carefully in Ref. 1.

[15] A quantitative discussion of the specific-heat anomaly in mixtures depends upon the magnitude of the ^{3}He-^{4}He tunneling rate J'. Experimental information on J' will be available from the experiments of Richardson and co-workers on mixtures. We will discuss the specific-heat anomaly in detail when the mixture experiments have been completed.

"SHAPE-DEPENDENT" SPECIFIC HEAT OF CEROUS MAGNESIUM NITRATE*

B. M. Abraham and Y. Eckstein
Argonne National Laboratory, Argonne, Illinois 60439

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The specific heat of two spheres and of one cylinder of compacted powdered cerous magnesium nitrate has been measured from $T^* = 3.5$ to 20 mK. Above 6.5 mK a correction of +0.3 mK must be added to convert T^*(sphere) to the thermodynamic temperature T, and +1.1 mK to convert T^*(cylinder) to T. Below 6.5 mK, T^*(sphere of powder) + 0.3 = T^*(cylinder of powder) + 1.1 mK = T^*(single-crystal sphere) + Θ(Weiss constant). The specific heat of cerous magnesium nitrate is given by $C/nR = 6.4/T^2 - 12/T^3$.

Although nuclear magnetic resonance and Josephson junctions are promising techniques for temperature measurement in the millikelvin range, the susceptibility of cerous magnesium nitrate (CMN) is still the primary thermometric parameter because of ease and precision of measurement. The latter feature is essential where small temperature increments are being measured. In spite of the increased activity in this temperature region and of the importance of the temperature scale, the T-T^* correlation[1-5] (thermodynamic-magnetic temperature) below 6 mK has not been established with certainty nor has the demagnetization correction for shapes other than ellipsoids of revolution been evaluated. Hudson[6] has recently published an excellent article which summarizes the present status of the CMN temperature scale and clearly points up the confusion that exists. In the attempt to bring a little order into the chaos, we have measured the specific heat of two spheres and of one cylinder, the three specimens machined from compacted powdered CMN. By using the specific heat as a transfer parameter, we have established the shape correction from a cylinder and a sphere of powder to single-cyrstal sphere, as well as the coefficient of the second term in the specific-heat expansion.

A mixture of 80% by volume powdered CMN and 20% by volume powdered AgCl was compacted around a fan of silver wires. The resultant cylinder (2.54 cm diam × 2.54 cm high) was machined into either a sphere (1.892 cm diam) or a cylinder (1.892 cm diam × 1.892 cm high). The specimens were inserted, in turn, into the same coil form for measurement of the specific heat. The coil form was made of epoxy and was wound with an inner secondary of approximately 2000 turns, a compensating secondary of approximately 1000 turns, and a shimmed primary to produce a uniform field around the specimen. A heater (100 Ω nominal resistance) of Pt-5%W alloy was wound on the silver wire tail which was used to provide thermal contact to a chrome-alum guard pill through a Pb superconducting switch.

The temperature scale for each specimen was set up by calibrating the mutual-inductance bridge[7] against the vapor pressure of pure ^{3}He with the specimen in situ. Calibration commenced at 600 mK and proceeded in five equal intervals of $1/T$ to 1.6 K, the limit set by the quantity of ^{3}He in the system. Cathetometer readings for each manometer leg and the bridge readings were plotted as a function of time; the calibration point was interpolated to the midpoint of the measurement period. Pressures were corrected to Torr; drifts during calibration were never more than 1 mK/min. A least-squares fit to the form $M - M_0 = A/T$ was made for the constants M_0 and A which now determined the temperature scale; M is the mutual-inductance bridge reading.

The experiment was performed by raising a superconducting solenoid around the chrome alum and energizing the magnet to produce a field of 18 kOe. The initial cooldown to 1.2 K and the dissipation of the heat of magnetization were achieved