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Mass-Spin-Fluctuation Waves in Solid Helium with Isotopic Impurities

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A Green's-function formalism is presented to study in a unified manner the propagation of mass-spin-fluctuation waves in solid helium with isotopic impurities. Based on this, consistent physical mechanisms are put forward for certain relaxation phenomena in impure solid helium.

INTRODUCTION

The motion of isotopic impurities in solid helium is a topic of current interest to which several theoretical[1-3] and experimental[4] papers have been devoted. The large zero-point motion of atoms in a quantum crystal such as solid helium causes them to tunnel from site to site in the lattice.[5] When a ^{3}He [^{4}He] impurity atom hops from site to site exchanging positions with the atoms in a crystal of solid ^{4}He [^{3}He], it creates at each of these sites a mass fluctuation with $|\Delta m| = m_4 - m_3$. A new type of many-body excitation (called[2] a mass-fluctuation wave) then arises,[1-3] and its effects have to be taken into account, for instance, in the explanation of NMR data in impure solid helium.

Such a tunneling motion of an isotopic impurity atom also creates spin fluctuations with $\Delta I = \pm\frac{1}{2}$ at the sites involved. In this sense, the excitation is a *mass-spin-fluctuation wave,* rather than just a mass-fluctuation wave: while the mass fluctuation affects the phonon spectrum in the crystal, the accompanying spin fluctuation will be coupled to spin-spin interactions such as the Heisenberg exchange term between two ^{3}He atoms. As we shall see, the inclusion of this aspect alters crucially the role of these excitations in the explanation of the effects of ^{4}He impurities on relaxation times in solid ^{3}He (or in solid ^{4}He with a large concentration of ^{3}He impurities).

The purpose of this paper is twofold. We first describe a Green-function formalism to study the motion of isotopic impurities in both solids, that treats the mass and spin fluctuations *simultaneously,* and enables us to judge when the latter are important. In particular, the procedure yields information, in various physical situations, on the lifetime[6] of the so-called mass-fluctuation excitations when spin fluctuations are also included, and on the essential differences between the tunneling of ^{3}He impurities in solid ^{4}He and that of ^{4}He impurities in solid ^{3}He. Using the method developed here, in the second half of the paper we discuss the role of mass-spin fluctuation waves in the understanding of low-temperature NMR data in impure solid helium of both types. We shall comment on the drawbacks of previous discussions of this aspect and then proceed to suggest alternative explanations of the underlying mechanism in various cases. The Green-function approach introduces the correlation between the motion of impurity atoms and that of the atoms of the medium in which they propagate in a very natural fashion. This enables us to base our discussion on simple physical arguments and interpretations throughout.

EFFECTIVE HAMILTONIAN

To describe the hopping of atoms from site to site in a solid ^{3}He-^{4}He mixture (total number of atoms $=N$), we extend the Hubbard model used in earlier work,[2,3] to derive the following effective Hamiltonian that describes the various tunneling processes in a ^{3}He-^{4}He mixture, to second order in the hopping energies of the atoms between neighboring sites i and j:

$$\begin{aligned} H &= \tfrac{1}{2}\sum_{ij} J_{ij}\vec{\mathrm{I}}_i\cdot\vec{\mathrm{I}}_j + \sum_{ij\sigma} K_{ij} n_{i\sigma}\rho_j \\ &\quad + \tfrac{1}{2}\sum_{ij\sigma} L_{ij} D^+_{i\sigma} D^-_{j\sigma} + \sum_{ij} M_{ij}\rho_i\rho_j \\ &\equiv H_X + E_{34} + H_{34} + E_{44}\,. \end{aligned} \tag{1}$$

Equation (1) is applicable when there is exactly one atom per site in the lattice. J, K, L, and M are quadratic in the hopping energies with *no* linear terms. Also $D^+_{i\sigma} = c^\dagger_{i\sigma} a_i$, $D^-_{i\sigma} = a^\dagger_i c_{i\sigma}$, $I^+_i = c^\dagger_{i\uparrow} c_{i\downarrow}$, etc., where $c^\dagger_{i\sigma}$ and $a^\dagger_i$ are ^{3}He and ^{4}He creation operators. The individual terms in H have simple physical interpretations (see Ref. 3). It is the term H_{34} (which refers to the *exchange* of positions of a ^{3}He atom and a ^{4}He atom) that gives rise to mass-fluctuation waves, but only under certain conditions, as we shall find. The D operators can be thought of, in crude way, as creation and destruction operators for a "particle" of mass Δm and spin $\frac{1}{2}$. However, the anticommutation relations between the D operators read

$$\{D^+_{i\sigma}, D^-_{j\sigma}\} = (n_{i\sigma}+\rho_i)\delta_{ij}, \{D^+_{i\uparrow}, D^-_{j\downarrow}\} = I^+_i\delta_{ij}, \{D^+_{i\sigma}, D^+_{j\sigma'}\} = 0 . \qquad (2)$$

If the right-hand side of the first of these relations had been a *c* number, and that of the second, zero, then D^+ and D^- would have been free-fermion operators. Note that even a redefinition of the D' s will *not* cause the second anticommutator to vanish, because of the boson parts of these operators.

GREEN FUNCTION

Let us now introduce the Zubarev double-time Green function[7]

$$G^\sigma_{ij}(t-t') = \langle\langle D^+_{i\sigma}(t); D^-_{j\sigma}(t')\rangle\rangle = -i\theta(t-t')\langle\{D^+_{i\sigma}(t), D^-_{j\sigma}(t')\}\rangle , \qquad (3)$$

where the expectation value for any operator A is given by

$$\langle A\rangle = \mathrm{Tr}[Ae^{-\beta(H-\mu_3\hat{n}-\mu_4\hat{\nu})}]/\mathrm{Tr}[e^{-\beta(H-\mu_3\hat{n}-\mu_4\hat{\nu})}] . \qquad (4)$$

Here $\hat{n}$, $\hat{\nu}$ are respectively ^{3}He and ^{4}He number operators ($\sum_{i\sigma} n_{i\sigma}$ and $\sum_i \rho_i$), and μ_3, μ_4 are the respective chemical potentials.[8] The complete equation of motion for the above Green's function reads, with $\sigma=\uparrow$,

$$i(d/dt)G^\uparrow_{ij}(t-t') = \delta(t-t')\delta_{ij}\langle n_{i\uparrow}+\rho_i\rangle + \sum_l [2(M_{li}-K_{li})G^\uparrow_{lj} + \tfrac{1}{2}L_{li}G^\uparrow_{lj} - \tfrac{1}{4}J_{li}(Q^\uparrow_{ilj} - Q^\downarrow_{ilj})$$

$$- 2\sum_\sigma (M_{li}-K_{li})Q^\sigma_{ilj} + \tfrac{1}{2}L_{li}Q^\downarrow_{lij} - \tfrac{1}{2}J_{li}\Gamma_{ilj} - \tfrac{1}{2}L_{li}\Gamma_{lij}] , \qquad (5)$$

where

$$Q^\sigma_{lij} = \langle\langle D^+_{l\uparrow}(t)n_{i\sigma}(t); D^-_{j\uparrow}(t')\rangle\rangle,$$

$$\Gamma_{lij} = \langle\langle D^+_{l\downarrow}(t)I^+_i(t); D^-_{j\uparrow}(t')\rangle\rangle . \qquad (6)$$

For a single ^{3}He[^{4}He] impurity atom, or for very low impurity concentrations, we may drop the terms involving J[M] in Eq. (5).

A. Impure Solid ^{4}He

When there is a *single* ^{3}He impurity atom present, the higher-order Green's functions Q and Γ vanish identically. The Fourier transform of $G^\uparrow$ is then found to be

$$\mathcal{G}^\uparrow(\vec{k}), \omega) = [\omega - 2M(0) - 2K(0) - \tfrac{1}{2}L(\vec{k})]^{-1} , \qquad (7)$$

where (specializing to nearest-neighbor interactions) $L(\vec{k}) = L\sum e^{i\vec{k}\cdot\vec{a}}$, $\vec{a}$ running over the nearest-neighbor vectors from a lattice site. We thus have a single undamped[6] mass-fluctuation wave. No decoupling approximations have been made in this case.

In the presence of more than one impurity atom, Q and Γ do not vanish identically, and this will in general introduce a width (in ω) for the excitations. However, since Q and Γ in Eq. (5) are multiplied by J, K, L, or M, such terms contribute only when ^{3}He atoms become nearest-neighbors at the same instant of time; for sparsely distributed impurities at low concentrations these contributions are negligible, and long-lived mass-fluctuation waves can thus occur once again.

This may happen even for somewhat higher concentrations of ^{3}He, under special circumstances. Adopting a decoupling scheme for higher-order Green functions[9] one stage beyond the level of G, we may decouple the Q's, but we must retain the Γ's and write equations of motion for them.[10] If a magnetic field is now applied (along either the ↑ or the ↓ direction), then, at sufficiently low temperatures, the ^{3}He spins are aligned parallel to the field and the Γ's vanish since there will be no ^{3}He pairs with opposite spins. We can then expect the excitations to be essentially undamped.

B. Impure Solid ^{3}He

We may consider bcc solid ^{3}He and divide the lattice into two sublattices A and B, and write down from Eq. (5) the coupled equations of motion for $G^\sigma_{\alpha\alpha'}(\alpha, \alpha' \in A)$ and $G^\sigma_{\beta\alpha'}(\beta \in B)$. It should be noted that Q and Γ do not vanish even if there is only one (^{4}He) impurity atom present in this case. A consistent decoupling procedure can be used to obtain a closed set of equations for the Green's functions.[10] The zeroth approximation in which all "higher-order" Green's functions such as Q, Γ, etc. are decoupled yields, for low ^{4}He concentrations, the dispersion relation

$$[\omega - (1-2\rho)K(0)]^2 = [\tfrac{1}{2}sJ(0)]^2 - [(1+\rho)^2 - 4s^2][\tfrac{1}{4}L(\vec{k})]^2 \qquad (8)$$

for the excitations, with $\rho = \langle\rho_i\rangle$ and $s = \langle I^z_\alpha\rangle = -\langle I^z_\beta\rangle$. Decoupling at a later stage (after writing down the equations of motion for Γ's) introduces imaginary parts in the solutions for ω *and leads to a finite lifetime for the excitations, even if there is only one impurity atom present.* We do not give here the rather lengthy expressions involved, as the details may be found in Ref. 10.

Comparing impurity-motion in solid ^{3}He and ^{4}He, it is in the exchange term H_X present in the former case that restricts the motion of a ^{4}He impurity atom. The site to which the atom can hop at any instant depends on the spin pattern around it at that instant, and the path chosen by it is such that the least number of antiferromagnetic bonds are broken in its journey. (If all the ^{3}He spins had been parallel, then of course the ^{4}He atom would have been equally free to take any path.[11] For a ^{3}He impurity atom in a ^{4}He lattice, however, one path is just as favorable as another, and the atom is free to hop to any neighboring site, the mass-spin fluctuation thus occurring with perfect periodicity. It should thus be easier to observe (for instance in neutron scattering experiments) the excitations of interest in impure solid ^{4}He as opposed to solid ^{3}He.

EXCHANGE - LATTICE RELAXATION IN IMPURE SOLID ^{3}He

We recall that the standard phenomenology used in the explanation of very-low-temperature NMR data in (pure) solid ^{3}He is the three-bath model,[12] in which the Hamiltonian in the presence of a constant external magnetic field is the sum of four terms: three of these are mutually commuting terms corresponding to the Zeeman (Z), exchange (X), and lattice (L) baths, coupled by the fourth term, the weak dipole-dipole interaction H_d. For application to impure solid ^{3}He, the model has been extended by Guyer and Zane[2] to a "four-bath" model, an extra, independent bath corresponding to the mass-fluctuation waves being introduced between X and L. In such a procedure, the original (three-bath model) calculations of relaxation times, etc., can be very easily modified to exhibit the ^{4}He concentration x_4 dependence of these quantities, as x_4 enters simply through the specific heat of the new bath.

While we agree with the over-all physical picture of Ref. 2 for the role played by the hopping of impurities in relaxation phenomena in impure solid ^{3}He, we feel that the four-bath model is not the natural way of incorporating this into the theory.[13,14] For one thing, the term H_{34} in H that is responsible for mass-fluctuation waves *does not commute with* H_X (although it commutes with H_Z and H_L). It is therefore inappropriate to associate an independent bath with this term. Doing so also *separates*, rather artificially, *the motion of the impurities from that of the spin patterns in the host crystal*. This brings us to a second point: Using the Green-function approach, which automatically treats the two motions together, we have seen that, in solid ^{3}He, mass-fluctuation waves are in fact generally not long-lived excitations. [The damping is essentially due to the last two terms on the right in Eq. (5). While the J term comes from H_X, as expected, the extra L term comes from H_{34} itself, and occurs because $D_{i\uparrow}^{+}$ and $D_{i\downarrow}^{-}$ do *not* anticommute, as would free-fermion operators.] These comments clearly apply also to the "five-bath" model proposed by Guyer[2] for higher ^{4}He concentrations.

We are thus left with the following *consistent* methods of handling the H_{34} term: (i) We can simply add it to H_X to form a common bath (H_{34} does commute with H_Z and H_L), or (ii) we can treat it as a term that couples the exchange-bath to the phonon-bath, just as H_d couples Z to X. If we adopt (i), we are then left with no mechanism for the relaxation of this common bath to the lattice. Further, it turns out that we must then accept the (rather unphysical) estimate[13] of $|L| \approx 18J$ in order to fit the observed x_4 dependence (at low concentrations) of the specific heat of this bath.[15] The more physical mechanism is thus that of (ii): H_{34} describes the hopping of ^{4}He atoms through the lattice by exchange of positions with ^{3}He atoms; even a single hop of an impurity atom can alter considerably the spin pattern, and hence the ^{3}He-^{3}He exchange bonds, in its neighborhood. *While the spin-fluctuation present in H_{34} couples to the exchange-bath, the mass-fluctuation inherent in this term couples to the phonons.* Using the standard density-matrix formalism,[16] the exchange-lattice relaxation time is then given by

$$[\tau_{X,L}(x_4)]^{-1} \sim \int_0^\infty d\tau\, \mathrm{Tr}[A(-\tau)A(0)]/\mathrm{Tr}(H_X)^2, \tag{9}$$

where $A(0)=[H_{34}, H_X]$ and $A(-\tau)=e^{-iH_X\tau}A(0)e^{iH_X\tau}$.

Parenthetically, we remark that our formalism also enables us to pinpoint a similar mechanism for $X \to L$ relaxation in pure solid ^{3}He. If there are vacancies in the lattice, it is found[3] that there occurs in the effective Hamiltonian H an extra term describing vacancy-hopping that is of *first order* in the hopping energy. This (spin-dependent) term can play the role of H_{34} in the preceding discussion, and couple X to L, leading to a relaxation time that depends on the vacancy concentration. This supports the qualitative conjecture of Guyer and Zane[17] in this connection.

RELAXATION IN IMPURE SOLID ^{4}He

Let us now consider the relaxation of (^{3}He) impurity spins in solid ^{4}He. For low values of the

^{3}He concentration x_3, the term H_X in H may be dropped, and the terms E_{34} and E_{44} do not change appreciably with the hopping of the atoms. We have also seen that mass-fluctuation waves are long-lived excitations in this case. In the presence of a constant external magnetic field, therefore, we are justified in describing the system in terms of three independent baths: Z, L, and an intermediate bath F for H_{34}. The relaxation "topology" is then $Z \rightarrow F \rightarrow L$. Now it is found experimentally[4] that there is a range of intermediate temperatures in which the relaxation time becomes temperature-independent. We explain this as follows: At the temperatures concerned, F and L are well-coupled, and $Z \rightarrow F$ relaxation occurs via the modulation of H_d by H_{34}, a quantum-mechanical exchange effect independent of temperature. We find that the relevant relaxation time $\tau_{Z,F}$ is given by

$$[\tau_{Z,F}(x_3)]^{-1} \sim \int_0^\infty d\tau \, \mathrm{Tr}([I^z, H_d][H_d(-\tau), I^z])/\mathrm{Tr}(I^z)^2 , \tag{10}$$

where $H_d(-\tau) = e^{-i\tau(H_Z + H_{34})} H_d e^{i\tau(H_Z + H_{34})}$.

Let us remark briefly on the situation for higher impurity concentrations in both solids. The terms E_{34} and E_{44} have *also* to be taken into account now, and the "topology" for relaxation will be more complicated. For instance, H_{34} can connect the E_{34} and E_{44} baths, and these in turn may couple to the lattice. It is conceivable that the high specific heat of the intermediate bath that is found experimentally,[15] at somewhat higher impurity concentrations in solid ^{3}He, is due to the contributions of these two additional baths. In the case of very impure solid ^{4}He, a further complication is the fact that H_X plays a significant role, and we will no longer be justified in assigning a separate bath to H_{34}.

In conclusion, the Green's-function formalism we have used has shown us precisely how the free propagation of a mass-fluctuation wave in impure solid helium is affected by the accompanying spin fluctuation when there are spin-spin interactions present. It has also enabled us to suggest consistent physical mechanisms, whose effects can be expressed quantitatively, for relaxation in NMR experiments in both solid ^{3}He and solid ^{4}He with low impurity concentrations.

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

[6] We shall not consider interaction with phonons here. The question of the lifetime of mass-fluctuation waves, with reference to phonon interactions alone, has been discussed qualitatively in Ref. 5.

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[8] μ_3 and μ_4 are introduced because of the presence of $D^\pm$. However, since the total number of atoms (N) is fixed, Eq. (4) can be simplified and written in terms of $\mu_3 - \mu_4$ and either $\hat{n}$ or $\hat{\nu}$. For instance, in the case of solid ^{3}He with a single ^{4}He impurity, the trace in general is over the states with $\nu = 0$ and 1, with all possible values of the total z component of the spin.

[9] N. N. Bogoliubov and S. V. Tyablikov, Proc. Acad. Sciences USSR 126, 53 (1959) [Sov. Phys.-Doklady 4, 589 (1959)]; see also Ref. 7.

[10] V. Balakrishnan and R. Balakrishnan, J. Low Temp. Phys. 9, 255 (1972). We may note that while Q does not involve spin flips, and is of "fourth order" in terms of the basic operators $D^\pm$ and $I^\pm$ (using $n_{i\sigma} = D^+_{i\sigma} D^-_{i\sigma}$), the "third-order" function Γ involves spin flips and is nonzero essentially because of the Heisenberg exchange interaction H_X present in H.

[11] The problem of the motion of a ^{4}He impurity in a ^{3}He lattice, formulated as we have done in terms of an underlying Hubbard-like model, is in some respects similar to that of the motion of a hole through an array of spins, discussed by W. F. Brinkman and T. M. Rice [Phys. Rev. B 2, 1324 (1970)]. These authors study the relevant Green's functions by examining, order by order in the hopping integral, the different closed paths that the hole can take leaving the spin configuration unchanged. A similar technique may be applicable to our problem too, as an alternative to the equation-of-motion method, on properly taking into account the complications that arise because our D operators do not satisfy free-fermion anticommutation relations. See also the relevant comments in Ref. 13 below.

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7, 529 (1969). On the other hand, at the low concentrations x_4 with which we are concerned, X should be left unchanged; the x_4 dependence of the measured relaxation time T_1 comes from that of the $X \rightarrow L$ relaxation time $\tau_{X,L}$, to which it is related by $T_1 = \tau_{X,L}(1 + C_X/C_Z)$. For such a parametrization of the x_4 dependence of the data, see R. P. Giffard and J. Hatton, Phys. Rev. Lett. 18, 1106 (1967).

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