Magnetic channel of the Kapitza resistance for a dilute ³He-⁴He solution at temperatures below 1 mK

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A mechanism for the heat transfer between a dilute ³He-⁴He solution and a sintered powder, leading to specific characteristics of the Kapitza resistance, is studied theoretically. It is found, in contrast with the acoustic mechanism, the magnetic dipole coupling between the nuclear spins of ³He dissolved in ⁴He and magnetic impurities at the surface of the sintered powder plays an important role for the heat transfer. In particular, it is remarkable that the magnetic coupling provides a new explanation of the observed features of the temperature ($\sim T^{-2}$) and Fermi-energy ($\sim T_F^{-1}$) dependence of the Kapitza resistance.

The cooling of dilute ³He-⁴He solutions down to a few tens of μK has been of great interest because of the possible discovery of the superfluid transition in the dissolved ³He. This cooling is, however, very difficult due to the problem of the Kapitza resistance between the refrigerand and the helium sample. To date, the lowest temperature of the ³He-⁴He solution achieved using sintered Ag powder is close to 200 μ K.^{1,2} The Kapitza resistance R_K between a dilute solution of ³He in ⁴He and sintered Ag powder at temperatures below 1 mK has been reported by several groups.³⁻⁶ The experimental results indicate that the Kapitza resistance values are proportional to T^{-2} . In addition to the above temperature dependence, Ritchie, Saunders, and Brewer⁶ and Chocholacs et al.¹ have investigated the effect of varying the ³He concentration in the solution on the R_K values, that is, the Fermi temperature T_F . They found that the R_K values are proportional to T_F^{-1} . The underlying mechanism to explain these experimental features has not yet been clarified.

This Rapid Communication reports a theoretical investigation on the heat-transferring mechanism between a dilute solution of ³He in ⁴He and sintered powder. We show that magnetic coupling provides a new explanation of the observed features of the heat transfer for this system.

It is natural to consider two independent channels for our system: the acoustic and the magnetic mechanism. Let us discuss, at first, the acoustic channel. At temperatures at or below 1 mK, the collective modes (phonons) in a ³He-⁴He solution do not couple with the vibrational modes of sintered powder, since at such low temperatures the metal particles of micrometer size in the sinter vibrate without volume change of individual particles (due to the size effect), i.e., the low-energy vibrational modes of the sinter do not excite phonons into the ³He-⁴He solution due to the slip condition at the boundary. (In the case of liquid ³He, the nonslip condition leads to the excitation of zero sound.¹⁵) The plausible acoustic channel is via the direct interaction between ³He quasiparticles and the lowenergy vibrational modes in the sinter.⁷ Sintered powders with packing fraction f of 0.4-0.5 are normally used for facilitating the cooling of the dilute ³He-⁴He solution into the temperature range concerned, where the sinter forms a three-dimensional percolating network and takes a fractal structure at short-length scales.⁸

The vibrational density of states (DOS) for a percolating network, in the sense of the effective medium theory,⁹ is proportional to ω^2 below some characteristic frequency ω_c and becomes constant above ω_c . This is expressed as

$$D(\omega) = \left(\frac{\omega}{\omega_c}\right)^2 \frac{9N}{3\omega_D - 2\omega_c}, \quad \omega < \omega_c \quad , \tag{1}$$

$$D(\omega) = \frac{9N}{3\omega_D - 2\omega_c}, \quad \omega > \omega_c \quad , \tag{2}$$

where the frequency ω_D is the cut-off frequency corresponding to the mode with the wavelength of the interparticle distance and N the number of metal particles per unit volume, respectively. Thus, the characteristic frequency ω_c of the sintered powder is an important factor for predicting the power of the temperature dependence of the Kapitza resistance R_K .

We can estimate the characteristic frequency ω_c for the typical sintered powder with packing fraction f = 0.5, which is assumed to be composed of particles a = 1000 Å in diameter. The above choice of the numerical values is quite reasonable because the sintered powders used in the experiments^{1,2,4-6} are made from silver particles with the nominal size ~ 700 Å. Provided that the system takes a simple cubic composed of spheres (1000 Å in diameter), this system has the filling factor $f_0 = 0.52$. The relation between the filling factor f_0 and packing fraction f is given by

$$f = nvp = f_0 p , \qquad (3)$$

where v and n are the volume of one particle and the occupied number of particles in unit volume. The percolation density is defined by p. From Eq. (3), p is estimated to be 0.96 for the sinter of f=0.5. Using this value of p and taking into account the dimensionality of the system, the average distance between vacant sites becomes $\sim 3a$. [This was estimated from the relation $a(1-p)^{-1/3} \sim 3a$.] As a result, the approximately estimated characteristic length ξ_c becomes of the order of 5000 Å. This length is related to the characteristic frequency v_c by $v_c = v_s/\xi_c$.

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Since the sound velocity of the sinter with packing fraction $f \sim 0.5$ is $v_S \simeq 10^5$ cm/sec, ¹⁰ we have $v_c \sim 2$ GHz, which corresponds to a few tens of mK in a temperature scale. We should note here that the value of $T_c = 15$ mK recovers well the acoustic channel of the Kapitza resistance for pure ³He as shown in Ref. 15. Thus, at temperatures at or below 1 mK, the Debye-phonon picture [Eq.(1)] is valid for sintered powders composed of the particle size of around 1000 Å.

The dissolved ³He atoms in ⁴He confined in pores can diffuse into the adjacent pore, by interaction with lowenergy vibrational modes of the sinter through the narrow channel. In this process, the sizes of pores and channels are important factors for the diffusion of ³He atoms. For instance, consider the *special* case where ³He atoms are confined in small pores of 100 Å in diameter which are connected by cylindrical channels of 7 Å in diameter and 10 Å in length. The zero-point energy E_0 of ³He quasiparticles in the channel becomes much higher than that in a pore, namely $E_0 = 1.41$ K for d = 7 Å. Since the Fermi energy E_F of a 5% ³He-⁴He solution becomes ~ 0.33 K, ³He atoms must *tunnel* through the potential barrier with height of 1 K in order to diffuse into the adjacent pore (see Fig. 1).¹¹

In actual sintered powders used in the experiments, $^{1,2,4-6}$ the average pore size D is to be of the order of 1000 Å, and the size of channels connecting pores is of the order of one tenth of D, i.e., 100 Å. For this situation, ³He atoms can move freely through the narrow channels in contrast with the above case. Thus, dissolved ³He atoms in the sintered powder can be treated as free particles with the *continuous* spectrum and are scattered inelastically by a vibrating interface. Since we are interested in the temperature range at or below 1 mK, the DOS expressed by Eq. (1), which is proportional to ω^2 , should be taken into account. In this case, the Kapitza conductance becomes, using the formula for the Fermi "gas" obtained by Toombs, Sheard, and Rice, ¹²

$$h_{K} = \frac{ck_{F}^{4}k_{B}^{4}T^{3}}{15\hbar^{2}\rho_{S}v_{S}^{3}} , \qquad (4)$$

where ρ_S is the mass density of the sinter and c is a number of order unity. The above Kapitza resistance $(R_K - 1/h_K)$ is proportional to T^{-3} and T_F^{-2} in contradiction with the experimental facts. The magnitude of Eq.



FIG. 1. Conceptional illustration of potential wells for 3 He quasiparticles confined in small pores 100 Å in diameter. The barriers correspond to the narrow channels.

(4) at 1 mK is also too small ($\sim 10^{-2}$) to explain the experimental data.

As a next step, let us discuss another heat-transferring mechanism between dissolved ³He in ⁴He and sintered powders. This is attributed to the magnetic coupling between ³He quasiparticles and magnetic impurities (i.e., O_2 , O_2^- , O, O^- , etc.) in the vicinity of the surface of sintered powder, ¹³ which has played an important role in the case of the pure ³He-sintered powder interfaces. ^{14,15,19}

One might suppose, at first glance, that the magnetic coupling is irrelevant due to the fact that ⁴He atoms are preferentially adsorbed at the surface of the sinter, and interrupt the effective coupling. This is not always true for the following reasons. Even for the pure ³He-sintered powder interface, the first few ³He-adsorbed layers are localized at the interface and they are not important for the direct coupling between ³He quasiparticles in bulk liquid ³He and localized magnetic impurities. That is, though adsorbed ³He atoms should play a role as a second-order process in the perturbation theory, they do not contribute to the direct energy exchange (the first-order process). In addition, the observation of the T_F dependence of R_K for ³He-⁴He solutions^{1,6} is the clear evidence of the irrelevance of the first few adsorbed ⁴He layers to the heat transfer. These results are reasonable because the magnetic dipole coupling is proportional to the distance r^{-3} and is not a short-range one.

When a ³He quasiparticle with momentum $\hbar k$ approaches the interface and is scattered by flipping the ³He nuclear spin due to the magnetic interaction with localized magnetic impurities near the surface, the Kapitza conductance $h_K = 1/R_K$ is expressed as follows, in a manner entirely analogous to that used for the case of pure ³He:^{14,15}

$$h_{K} = \sum_{\mathbf{k},\mathbf{k}'} \int_{0}^{\infty} \frac{\Delta^{2} f(\mathbf{k}) [1 - f(\mathbf{k}')]}{1 + \exp(-\Delta/k_{B}T)} \frac{n(\Delta) W_{\mathbf{k}\mathbf{k}'}}{k_{B}T^{2}} d\Delta , \quad (5)$$

where $f(\mathbf{k})$ is the Fermi distribution function for the ³He quasiparticles and $W_{kk'}$ is the transition rate of a ³He atom from an occupied state k to an empty state k'. In Eq. (5), the localized spins contributing to the transition are expressed by the two-level system with an energy splitting Δ with the distribution $n(\Delta)$, in which the nature of randomly distributed localized spins (dipole spin glass) is involved. It has been shown 16,17 that the dipole interaction behaves like an effective contact interaction when the heat exchange is dominated by scattering with the momentum transfer of the order of the ³He Fermi momentum $p \sim p_F$. Thus, we can take the dipole interaction d(x) to be the contact type by setting $d(x) = J\delta(x)$, where J takes the value of 0.99×10^{-43} erg cm³. As a result, the Kapitza conductance $h_K = 1/R_K$ is obtained as, by omitting the Stoner's enhancement factor K_{eff} introduced in the case of pure ³He,¹⁵

$$h_{K} = \frac{J^2 m^{*2} k_{F}^2}{8\hbar^5 \pi^3 k_B T^2} \int_0^\infty \frac{N(\Delta) \Delta^3 d\Delta}{\exp(\Delta/k_B T) - \exp(-\Delta/k_B T)} ,$$
(6)

 m^* being the effective mass of ³He quasiparticle. The width of the energy distribution function $n(\Delta)$ can be es-

timated as $\overline{\Delta} \simeq z \mu_e^2 / \overline{r}^3$, where z is a number, of order unity representing the effective coordination number, and \overline{r} is the mean distance from any localized spin to the nearest one. This estimation of $\overline{\Delta}$ is reasonable since the effective field acting on any given spin is dominated by the few spins which happen to be the closest.

Following the discussion by Villain,¹⁸ the nonvanishing distribution function at $\Delta = 0$ is assumed. The energy distribution function $n(\Delta)$ [Eq. (6)] is taken as a Gaussian as the simplest one:¹⁵

$$n(\Delta) = \frac{n_0}{\overline{\Delta}\sqrt{\pi}} \exp(-\Delta^2/\overline{\Delta}^2) .$$
 (7)

Here n_0 is the areal density of the two-level system near the surface in a projected mean. Combining the density n_0 and the width $\overline{\Delta}$, we can write down the distribution function $n(\Delta)$ as a function of one variable n_0 (or $\overline{\Delta}$) from the relation $\overline{\Delta} = z\mu_e^2 n_0^{3/2}$.

By using the same value of the areal density of magnetic impurities n_0 at the surface of sintered silver, which was estimated of $n_0 \approx 1/(5 \text{ Å})^2$, ^{13,19} we have $\overline{\Delta} \sim 5 \text{ mK}$. At sufficiently low temperatures $T < \overline{\Delta}/k_B$, the integration in Eq.(6) can be easily performed and one has the Kapitza resistance

$$R_{K} = \frac{16\pi^{1/2}\hbar^{7}\overline{\Delta}}{J^{2}m^{*}T_{F}k_{B}^{4}n_{0}T^{2}} .$$
(8)

It should be emphasized that Eq.(8) varies as T^{-2} and T_F^{-1} , whose dependences are in accord with the experimental features.^{1,4-6} Note here that the concentration (T_F) dependence of the effective mass of m^* of ³He quasiparticles is very weak. By taking the suitable values for the factors for 5.0% solution of ³He-⁴He solution: $J = 0.99 \times 10^{-43}$ erg cm³, $m^* = 2.46m_3$, $E_F = 4.57 \times 10^{-17}$ erg, and $n_0 = 4.6 \times 10^{14}$ cm⁻², we find $R_K = 32T^{-2}$ (m²K/W) at sufficiently low temperatures. This result is plotted by the straight line (below) as well as the experimental data (Fig. 2). The upper line in Fig. 2 is for 1.3% solution: $R_K = 80T^{-2}$ (m²K/W).

It is remarkable that the observed T^{-2} and T_F^{-1} dependences of R_K are recovered by the *magnetic* coupling. In addition, one should note that the magnitude of R_K obtained theoretically agrees well with the observed one as found in Fig. 2, which was calculated by using the *same* areal density of magnetic impurities n_0 with that used in the analysis for the Kapitza resistance of pure ³He (Refs. 13 and 15) and the NMR experiments.¹⁹



FIG. 2. The Kapitza resistance R_K of dilute ³He-⁴He solutions as a function of temperature. Data are taken from Refs. 4-6. Solid lines are the theoretical ones calculated from Eq. (8). The lower line is for 5.0% solution, and the upper one is for 1.3% solution, respectively.

If the magnetic coupling is a dominant channel between ³He-⁴He solution and sintered powder at sufficiently low temperatures, the similar magnetic-field dependence on R_K with that for pure ³He (Ref. 13) will be observed. Although it has been reported by Ritchie *et al.*⁶ that the observed R_K for ³He-⁴He solution is insensitive to the magnetic field (\sim 3 T) *above* 5 mK, this point remains to be clarified by further experimental investigations, especially the data at submillikelvin temperatures are interesting. Finally, I hope that the present work would be helpful in searching for the way of cooling dissolved ³He atoms in ⁴He down to the "superfluid" transition temperature.

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