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Magnetic Kapitza resistance and surface random spins

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It is emphasized that the randomly distributed spins near the surface play an important role on the magnetic Kapitza resistance. Under the assumption that dipolar spin-glass is formed in the vicinity of the surface of magnetic salt, the formulation of the Kapitza resistance is presented. The temperature dependence of magnetic Kapitza resistance is found to be proportional to T^{-2} at sufficiently low temperatures.

The problem of magnetic Kapitza resistance is of interest not only from practical implications in low-temperature physics but also in its own right.[1] In 1966, Abel, Anderson, Black, and Wheatley[2] discovered that below 20 mK the Kapitza resistance R_K between liquid ^{3}He and cerium magnesium nitrate (CMN) shows a different temperature dependence varying approximately as T rather than T^{-3} dependence expected from the normal Kapitza resistance. Legget and Vuorio[3] have developed a theory to explain these observations in which the heat exchange is assumed to occur through the magnetic coupling between the cerium spins and the nuclear ^{3}He spins. This theory derived the resistance R_K proportional to T as observed by Abel *et al.*[2] above the magnetic ordering temperature ($T_c \simeq 2$ mK) of CMN. It remains, however, an open question[1] whether or not the magnetic coupling is necessarily important to understand various experiments[4-8] performed after that of Abel *et al.*,[2] and more detailed study on the effects of magnetic coupling has been called for both experimentally and theoretically.

The motivation of this Brief Report is based on the possible existence of dipolar spin-glass in the vicinity of the surface of the salt. This should be due to the unavoidable surface inhomogeneity of the magnetic substance exposed to air and due to the magnetic impurities such as O_2 molecules or O atoms with spin which are adsorbed randomly at the surface. As a consequence, the magnetic Kapitza resistance can be drastically different from that expected in an ordinary magnetic salt.

The viewpoint is described as follows: since the magnetic dipole interaction falls off as r^{-3}, the effective magnetic coupling seems likely to take place from the localized spins located in the vicinity of the surface of the salt. In particular, the surface of magnetic substances might be covered with adsorbed impurities with the magnetic moments such as O_2 molecules or O atoms as pointed out by Potter.[9] The adsorbed magnetic impurities and the magnetic ions near the surface of the salt are randomly distributed resulting from unavoidable surface inhomogeneity and adsorption site irregularity. The possible existence of dipolar spin-glass in such a system has been pointed out[10] from the analogy between the Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange potential and the magnetic dipole-dipole interaction. Dipole interactions are in many respects similar to the RKKY interactions: competition between ferro- and antiferromagnetic interactions, and r^{-3} behavior. In fact, this expectation is supported experimentally.[11,12]

As is well known, the magnetic Kapitza resistance R_K is proportional to τ/C_M, where C_M is the heat capacity of magnetic substance and τ is the relaxation time between two systems to reach the equilibrium. For instance, in the temperature regime above the magnetic ordering temperature T_c, the heat capacity C_M is proportional to T^{-2}, and $1/\tau$ varies as T reflecting the sharpness of the Fermi distribution function of normal liquid ^{3}He at low temperatures. As a result, the R_K behaves as T above T_c as obtained by Legget and Vuorio.[3] From the similar arguments, if a spin-glass state is present in the vicinity of the surface where the magnetic moments are frozen in randomly oriented local fields, the heat capacity will be proportional to T in most cases at sufficiently low temperatures. As seen later, this rather general feature of the heat capacity is well interpreted by introducing the magnetic two-level systems with the broadly distributed energy difference.[13] In such a case, the Kapitza resistance R_K is expected to be proportional roughly to T^{-2} at sufficiently low temperatures. Now a quantitative treatment of the magnetic Kapitza resistance between a spin-glass state and liquid ^{3}He is given below. Let us consider the situation where the ^{3}He quasiparticle with momentum $\vec{k}$ approached to the interface and is scattered by flipping the ^{3}He nuclear spin due to the magnetic interaction with localized electronic spins near the surface. Under this situation the Kapitza resistance R_K is expressed as

$$R_K^{-1} = \sum_{\vec{k}} \sum_{\vec{k}'} \int_0^\infty d\Delta\, n(\Delta) \frac{\Delta^2 f(\vec{k})[1-f(\vec{k}')]}{1+\exp(-\Delta/k_B T)} \frac{W_{\vec{k}\vec{k}'}}{k_B T^2} , \tag{1}$$

where $f(\vec{k})$ is the Fermi distribution function for the ^{3}He quasiparticles and $W_{\vec{k}\vec{k}'}$ is the transition rate of a ^{3}He atom from an occupied state $\vec{k}$ to an empty state $\vec{k}'$. In Eq. (1), the localized spins contributing to the transition are expressed by the two-level system with a energy splitting Δ with the distribution $n(\Delta)$. The expression for $n(\Delta)$ is very important in the present analysis and we shall discuss it fully later. Now the transition rate $W_{\vec{k}\vec{k}'}$ from the state $\vec{k}$ is written down as

$$W_{\vec{k}\vec{k}'} = \frac{2\pi}{\hbar} |\langle \vec{k}' | H' | \vec{k} \rangle|^2 \delta(\epsilon_{\vec{k}'} - \epsilon_{\vec{k}} - \Delta) , \tag{2}$$

where H' is the interaction Hamiltonian of the two-level system. The magnetic coupling between ^{3}He nuclear spins and the electronic spins is taken to be dipolar type. Then we can write down the dominant interaction Hamiltonian in the second quantized form as

$$H' = \frac{1}{2V} \sum_{\vec{k}\vec{k}'} \sum_n \exp[i(\vec{k}-\vec{k}')\cdot\vec{R}_n] \times J(\vec{k},\vec{k}') S_n^+ a_{\vec{k}'}^\dagger a_{\vec{k}} , \tag{3}$$

where $a^{\dagger}_{\vec{k}}$ and $a_{\vec{k}}$ are the creation and annihilation operator for the ^{3}He atoms with the momentum $\vec{k}$ and spin σ. The factor V is the volume of a half space occupied by liquid ^{3}He. The symbol S_n^+ expresses the rising operator for the spin state characterizing localized state at site $\vec{R}_n$ in the effective mean near the surface. The factor $J(\vec{k},\vec{k}')$ is the Fourier transform of dipole interactions $d_{\alpha\beta}(\vec{x}) = (r^2\delta_{\alpha\beta}-3r_\alpha r_\beta)r^{-5}$ in a half space. It is shown[14] 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 ^{3}He Fermi momentum p_f.[15] Therefore we can take $J(\vec{k},\vec{k}')$ to be the contact type by setting the dipole interaction $d(\vec{x})=J\delta(\vec{x})$, thus we have $J(\vec{k},\vec{k}')=J$ with the magnitude of 10^{-43} ergs cm^3.

Substituting Eq. (3) into Eq. (1), we obtain the Kapitza resistance R_K as

$$R_K^{-1}=\frac{J^2m^{*2}k_F^2K_{\mathrm{eff}}^2}{2\hbar^5\pi^3k_BT^2}\int_0^\infty d\Delta\frac{n(\Delta)\Delta^3}{\exp(\Delta/k_BT)-\exp(-\Delta/k_BT)}\ , \tag{4}$$

where the use is made of

$$f(\vec{k})[1-f(\vec{k}')]=\delta(\epsilon_{k'}-\epsilon_f)/[\exp(\epsilon_k/k_BT)-1]\ .$$

The exchange enhancement effect K_{eff} of quasiparticles is included through in Eq. (4) which increases the conductance $h_K=R_K^{-1}$ by one order of magnitude for pure ^{3}He.

Now let us describe the nature of the distribution function $n(\Delta)$ in Eq. (4). The important point in the spin-glass behavior is the potential energy as a function of the simultaneously specified orientations of all of the spins. In this connection, the specific heat of dipolar spin-glass is well described by assuming the two-level system as shown by Villain,[13] which implies that the transition involves the simultaneous rearrangement of a small number of spins. The width $\bar{\Delta}$ of the energy distribution function $n(\Delta)$ of the above-mentioned two-level system can be estimated as $\bar{\Delta}=z\mu_e^2/\bar{a}^3$, where z is a number of order unity representing the effective coordination number and $\bar{a}$ is the mean distance from any localized spin to the nearest one. This estimation of $\bar{\Delta}$ 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 of Villain,[13] the nonvanishing distribution function at $\Delta=0$ is assumed. We take the energy distribution function $n(\Delta)$ in Eq. (4) as a Gaussian such as

$$n(\Delta)=n_0\exp(-\Delta^2/\bar{\Delta}^2)/\sqrt{\pi}\bar{\Delta}\ ,$$

where 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 $\bar{\Delta}$, we can write down the distribution function $n(\Delta)$ as a function of one variable from $\bar{\Delta}=z\mu_e^2n_0^{3/2}$.

By changing the variable Δ in the integral of Eq. (4) to the dimensionless one $x=\Delta/k_BT$, we can find the characteristic feature of the temperature dependence of the resistance as follows. Using a dimensionless variable x, Eq. (4) becomes

$$R_K^{-1}=\frac{J^2m^{*2}k_F^2K_{\mathrm{eff}}^2k_B^3T^2}{2\hbar^5\pi^3}\int_0^\infty dx\frac{n(\Delta)x^3}{e^x-e^{-x}}\ . \tag{5}$$

The width of the distribution function $\bar{\Delta}/k_BT$ in Eq. (5) varies as the temperature. Combining this function $n(\Delta)$ with the factor $x^3/(e^x-e^{-x})$ in Eq. (5), we see that at sufficiently low temperatures $T \ll \bar{\Delta}/k_B$ the integral can be easily obtained as $n_0\pi^3/(4\sqrt{\pi}\bar{\Delta})$ and the resistance R_K behaves as T^{-2}. On the other hand, at high temperatures $T \gg \bar{\Delta}/k_B$ we can also integrate Eq. (4) as $n_0\bar{\Delta}^2k_BT/8$. As a result, the resistance R_K becomes proportional to T, whose temperature dependence is the same with the Legget and Vuorio[3] reflecting free-spin states. At the intermediate temperature range, the theoretical curve of R_K is connected smoothly with the minimum around $T_m \simeq \bar{\Delta}/2.5k_B$.

Let us discuss the ratio of the magnetic Kapitza resistance to the resistance due to zero-sound excitation. The explicit form of the Kapitza resistance due to zero-sound excitation was derived by Bekarevich and Khalatnikov[16] and Gavoret,[17] which is expressed as

$$R_K^{-1}=\frac{2\pi^2k_B^4\rho_Lc_0F_1T^3}{15\hbar^3\rho_Sc_t^3}\ . \tag{6}$$

Here, ρ_L and ρ_S are the mass densities of liquid ^{3}He and a solid, and c_0 and c_L are the velocities of zero-sound and transverse acoustic phonons, respectively. F_1 is the numerical factor of about 1.5. This formula can be applied to the whole scale of temperature. The ratio of Eq. (6) to the magnetic Kapitza resistance at high temperatures derived from Eq. (4) becomes

$$\gamma=\frac{R_K(\text{magnetic})}{R_K(\text{zero sound})}=\frac{32\pi^5\rho_Lc_0k_B^4\hbar^2F_1T^4}{15\rho_Sc_t^3J^2m^{*2}k_f^2K_{\mathrm{eff}}^2n_0\bar{\Delta}^2}\ . \tag{7}$$

Using the known values of physical quantities used in Eq. (7), we find that the ratio can be expressed as

$$\gamma \sim 10^{51}n_0^{-4}T^4\ . \tag{8}$$

From Eq. (8), it is concluded that the heat transfer due to the magnetic coupling is a dominant channel below about 1 K compared with the heat transfer due to the zero-sound excitation, as far as we consider the reasonable areal density of states of spins of the order of $n_0 \sim 10^{13-14}$ cm^{-2}.

It is interesting to note that the temperature dependence ($\sim T^{-1.5}$) derived from Eq. (5) is close to that observed by Hebral *et al.*[7] in which the observed resistance decreased with increasing temperature up to around 10 mK. Recently, Fujii and Shigi[18] have used the potassium tutton salt (CPS):$CuK_2(SO_4)_2 6H_2O$ in order to clarify the role of the magnetic coupling for the Kapitza resistance. This substance is, in fact, suitable to reveal the role of magnetic coupling because its magnetic ordering temperature $T_c \simeq 29.6$ mK is tractable. Fujii and Shigi[18] obtained similar tendency of the resistance for CPS with those of Hebral *et al.*,[7] i.e., the observed resistance[18] decreased rapidly from 15 up to 60 mK and its temperature dependence varies rather like $T^{-1.5}$ below 50 mK. This temperature dependence can be recovered from Eq. (5) in addition to the agreement on the magnitude of R_K by taking $\bar{\Delta}=130$ mK.

To summarize, the present work suggests an important mechanism together with the suggestion of the existence of dipolar spin-glass in the vicinity of the surface of the salt causing from the surface inhomogeneity and magnetic adsorbed impurities. Finally, I predict the temperature dependence of the resistance when liquid ^{3}He is solidified in the present system. As shown by Legget and Vuorio,[3] the conductance h_K is proportional to the product of susceptibilities of two systems: $h_K \propto \chi_M \chi_{He}$ where χ_M is the suceptibility for the magnetic substance, and χ_{He} for the helium system. For solid ^{3}He, the magnetic susceptibility χ_{He} follows a Curie-Weiss law T^{-1} above the ordering temperature $T_N \sim 1$ mK and the suceptibility for the spin-glass at sufficiently lower than freezing temperature should be roughly proportional to T^2 if the specific heat behaves as T. As a result, we have the conductance h_K proportional to T in the temperature range considered above. This observation will confirm the model proposed in the present work.

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