Brief Reports

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dc Kapitza resistance between liquid ^{3}He and $Mn(NH_4)_2$ Tutton salt

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Thermal boundary resistance R_B between liquid ^{3}He and $Mn(NH_4)_2$ Tutton salt is measured by the dc method in the temperature region 0.09 to 0.5 K. $R_B = 131/T^{2.57}$ cm^2 K W^{-1} is obtained. Thermal conductivity κ of this salt is also measured in the region near the Néel temperature, $T_N = 0.176$ K. The temperature dependence of κ changes beyond T_N, but no characteristic change in R_B is observed throughout the ordering region.

In the recent comprehensive review Harrison[1] has pointed out that the observed temperature dependences of thermal boundary resistance R_B between liquid ^{3}He and powdered cerium magnesium nitrate (CMN) can be explained in terms of the spin-lattice resistance in CMN, rather than in terms of a magnetic coupling at the boundary.

As for the magnetic coupling[1,2] between ^{3}He nuclear spins and electron spins of a magnetic solid, relevant experimental results of R_B were all derived from the measurements of the thermal relaxation time necessary to attain the thermal equilibrium in the sample system after a heat pulse on the system. On the contrary, the R_B measured by the dc method (potentiometric method[3]) does not depend on the thermal relaxation time. Harrison and Pendrys[3] measured R_B between liquid ^{3}He and a single crystal of CMN by the dc method in the region 70 to 700 mK. They could observe no evidence of the magnetic coupling in their result. The lowest temperature of 70 mK in their work, however, seems to be too high to observe the effect due to the magnetic coupling in the ^{3}He-CMN system. The effect is possibly not important in that temperature region. In fact, R_B between liquid ^{3}He and powdered CMN derived from the measurement of thermal relaxation time, sometimes exhibited $R_B \propto T^{-1}$ dependence[4] below 20 mK, which was attributed to a contribution of the magnetic coupling.

Nevertheless, there still remain problems to be explored concerning the effect of the magnetic coupling on R_B. Particularly, the roles of electron spins for the energy transfer at the surface of magnetic solid and for the thermal relaxation in the solid should be explored. The former role would be effectively revealed by the investigation on the thermal boundary resistance R_B of a magnetic solid by the dc method when the solid passes through its paramagnetic state to an ordering state of electron spins. The latter role would be revealed by the investigation of the thermal conductivity κ of the solid beyond the magnetic ordering, since the spin-lattice relaxation in a magnetic solid changes usually near the ordering, and κ is generally related to the thermal relaxation time in the solid, $\tau_c \propto l^2 c/\kappa$, by the thermal diffusion process, where l is the dimension of the solid and c the heat capacity per unit volume. A magnetic salt, where the magnetic ions have larger magnetic moments, is practically suitable for such studies because magnetic interactions within the salt or on the surface are larger and its magnetic ordering takes place generally at relatively higher temperature.

In this report, I present new results of two series of experiments on manganese ammonium Tutton salt, $Mn(NH_4)_2(SO_4)_2 \cdot 6H_2O$ (abbreviated to Mn-T), which has an antiferromagnetic ordering[5,6] of Mn^{2+} spins ($S = \frac{5}{2}$) below $T_N = 0.176$ K. The first of them is the result of the κ measurement in the temperature region 0.11 to 0.45 K. The observed κ is anomalously small and its temperature dependence in the paramagnetic state is different from that in the ordered state of this salt. The second is the result of R_B between liquid ^{3}He and Mn-T measured by the dc method. The observed R_B is not so different from R_B of the liquid ^{3}He-CMN system. It is striking, however, that no change is observed in the temperature dependence of R_B throughout the paramagnetic and the ordered region.

Single crystals of Mn-T were grown from a saturated aqueous solution of 1:1 mol of manganese sulfate, $MnSO_4 \cdot 4-6H_2O$ and ammonium sulfate, $(NH_4)_2SO_4$ at ambient temperature. To obtain large single transparent crystals, the concentration of hydronium ion in the solution was kept near $p\mathrm{H} = 6.5$ to 7.0 by addition of a small amount of ammonia water. Single crystals were grown typically to a size of dimension $\sim 20 \times 25 \times 40$ mm^3. The samples used for the measurements were cut from these single crystals using a wet wire-string saw and then water polished to desired size.

The measurements of κ were done by the conventional steady-state, longitudinal heat-flow method.[7,8] The samples were 35 mm long with 4×8-mm^2 rectangular cross section. A more than 20-mm length of a sample was used for a contact to heat links made of silver plate which were connected to a mixing chamber of a dilution refrigerator. An electrical heater and two carbon resistance thermometers were arranged in the same way as in the work of Ref. 8. The results of the study of κ as a function of temperature are shown in Fig. 1. The observed κ value

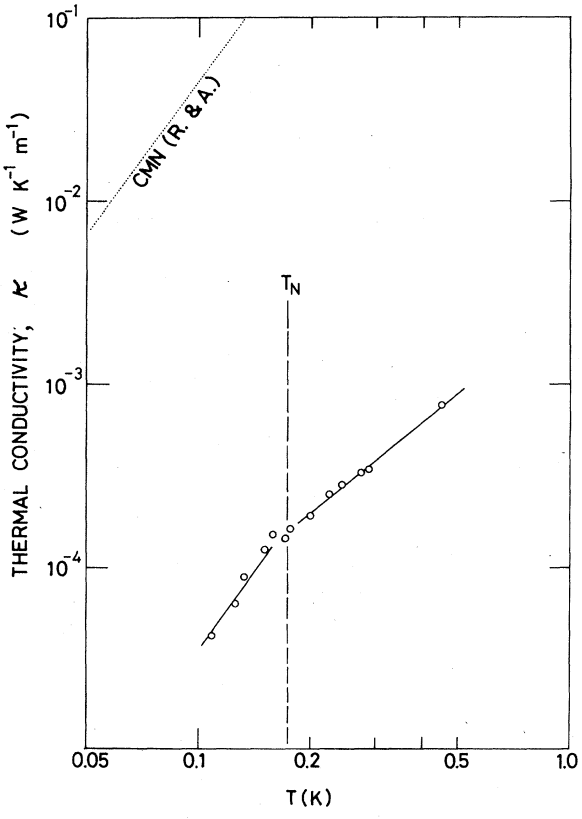

FIG. 1. Thermal conductivity κ of $Mn(NH_4)_2$ Tutton salt near the Néel temperature, $T_N = 0.176$ K. Dotted line shows κ of cerium magnesium nitrate (CMN) partly cited in Ref. 8.

is anomalously small as thermal conductivity of a single crystal of magnetic insulators, which is smaller by three orders than that of CMN,[8] as partly shown in Fig. 1. The values of κ of Mn-T can be approximated by $\kappa = (2.9 \times 10^{-3}) T^{1.66}$ in the region above T_N, while $\kappa = (4.9 \times 10^{-2}) T^{3.2}$ in the region below 0.16 K (in units of W K^{-1} m^{-1}). Those are shown by straight lines along the measured points in Fig. 1.

The thermal conductivities of magnetic insulators are generally influenced by the low-lying excitations of the spin system. These excitations affect the lattice thermal conductivity by scattering phonons, and also provide an additional mechanism for heat conduction through spin-lattice interactions. As for κ of Mn-T, it is considered that the temperature dependence of $T^{1.66}$ is due to an additional conductivity attributed to interactions between phonons and Mn^{2+} spins. This additional effect dominates in the region near T_N and diminishes in the region below T_N. Therefore, the dependence of $T^{3.2}$ might approach the standard temperature dependence of phonon conductance, T^3, in the region far below T_N. As a consequence of this variation of κ, thermal relaxation time τ_c is largely modified when temperature is lowered through T_N. In fact, the time necessary to attain the thermal equilibrium has been found to increase with decreasing temperature in the region below T_N. A long thermal relaxation time of Mn-T below T_N was also reported in the specific-heat measurement by Vilches and Wheatley.[5]

For the R_B measurement, a cylindrically shaped single crystal of 8.2-mm diameter with 17.2-mm height was used. Two small cavities were made in the sample, which were slightly larger than the carbon resistance thermometer and an electrical heater made of manganin wire, respectively. A resistance thermometer and an electrical heater were stuck on the walls of each cavity with GE 7031 varnish. The cavities were sealed with Apiezon N grease. The sample was set in a cylindrical Pyrex glass cell of 10-mm inner diameter. In the space between the crystal and the Pyrex wall, a brush made of silver wire (0.2-mm diam) was inserted to obtain a uniform distribution of the ^{3}He temperature. 0.03 mol of liquid ^{3}He containing a ^{4}He impurity less than 50 ppm, was condensed in the cell. A carbon resistance thermometer and a germanium sensor were used to measure the temperature of liquid ^{3}He. The outputs of three ac-resistance bridges for these thermometers were recorded on a multipen recorder. The experimental procedure to obtain the thermal balancing in the sample system was followed in the same way as reported in Ref. 3. Thermal equilibrium after the sample heater was turned on or off was attained within less than 40 min in the region well above T_N, while it took more than three hours in the region below T_N. The results for R_B are shown in Fig. 2, where the surface area of the Mn-T sample, 5.487 cm^2, is used

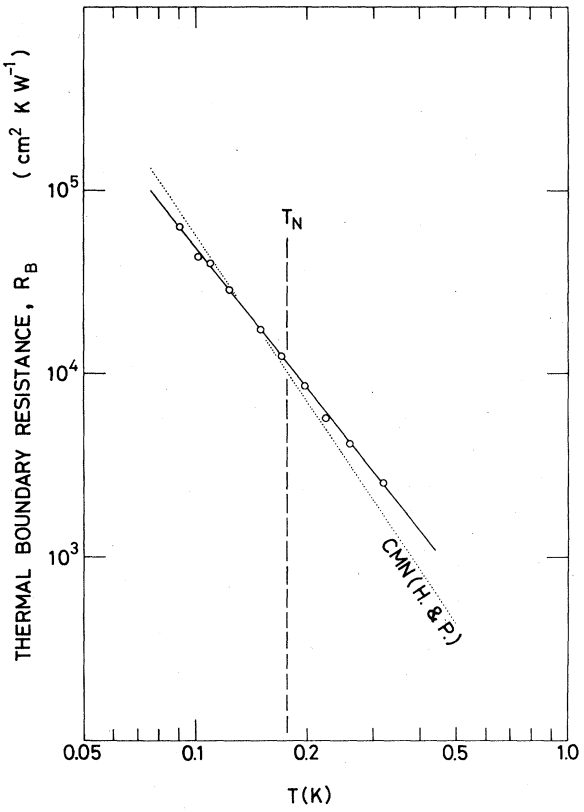

FIG. 2. Thermal boundary resistance R_B between liquid ^{3}He and $MN(NH_4)_2$ Tutton salt. Dotted line is R_B between liquid ^{3}He and cerium magnesium nitrate (CMN) cited in Ref. 3.

for the R_B estimation. The data can be well approximated by $R_B = 131/T^{2.57}$ cm^2 K W^{-1}. The actual numerical factor in this expression would be between 130 and 200 because there was an ambiguity coming from the surface condition of the crystal. The dotted line in Fig. 2 shows $R_B = 55/T^3$ between liquid ^{3}He and CMN, obtained by the dc method reported in Ref. 3.

The thermal boundary resistance given by the acoustic-phonon mismatch theory[9] is written as

$$R_B = \frac{15\rho_s (v_t^s)^3 \hbar^3}{2\pi^2 \rho_l v^l k_B^4 F_1^{ls} T^3} , \quad (1)$$

where F is a factor of order unity, ρ_l and ρ_s are densities of a liquid and solid, respectively, and v^l and v^s the velocities of sound in the liquid and in the solid, respectively. The present R_B result shown in Fig. 2 is qualitatively explained by Eq. (1). Now, let us focus our attention onto the temperature dependence of R_B at the magnetic ordering. As is seen in Fig. 2, there is no change of the temperature dependence near T_N. This means that any change of v_t^s in Eq. (1) does not occur at the magnetic ordering, in contrast to the κ, i.e., τ_c is modified by the ordering. In other words, the sound velocity in a magnetic solid is not modified by spin-phonon interactions even though the thermal conductivity is modified by them.

In the measurement of R_B, if the method of thermal relaxation time were taken for the sample consisting of liquid ^{3}He and powdered Mn-T, size-dependent R_B would be observed, because a small κ value of this salt and a large variation of the time necessary for thermal equilibrium in the ^{3}He–Mn-T system have actually been observed as mentioned above. For a sample system consisting of two specimens which have heat capacities c_1 and c_2, thermal resistance R derived from the thermal relaxation time τ is usually expressed[4] as $\tau = Rc_1c_2/(c_1+c_2)$, and if $c_1 \ll c_2$, it becomes $\tau = Rc_1$. In the case of the liquid ^{3}He–Mn-T system, c_1 is the specific heat of Mn-T. The above τ includes the thermal relaxation time in the Mn-T crystal, τ_c, so that R could not be independent of τ_c/c_1. Obviously, it is emphasized on the basis of the present results that the thermal relaxation time should be carefully examined when it is used for the estimation of R_B. In such a case, it is necessary to have independent evidence that the thermal relaxation time within the solid is negligibly small compared with the relaxation time at the boundary.

Another striking fact in Figs. 1 and 2 is seen by comparing the κ values and the R_B values for CMN and Mn-T. There we see that R_B's are scarcely dependent on κ values. This situation is also suggested by referring to the investigations concerning the R_B's between liquid helium and metals in the superconducting state. The recent investigations[10,11] have concluded that most results of the earlier experiments are spurious, and that they are explained as strain-induced bulk thermal resistance in the superconducting state.

Recently, a kind of strong magnetic coupling between liquid ^{3}He and a magnetic salt near the magnetic phase transition was proposed by the present author.[12] However, it has been revealed[13] that the anomaly depends on a thermal gradient in liquid ^{3}He along the crystalline surface. Therefore, the anomaly seems to be caused by another magnetic effect rather than the strong magnetic coupling at the boundary. Our experimental study for this problem is in progress.

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