Magnetic coupling in thermal-boundary resistance between thin silver films and liquid ^{3}He in the millikelvin regime

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Using a glass-capacitance technique, we have measured the thermal-boundary resistance R_K between liquid ^{3}He and thin silver films on a flat glass substrate in the millikelvin regime. For $T>30$ mK, the experimental observations are in excellent agreement with the acoustic mismatch theory. For $T<30$ mK, we find concrete evidence supporting the magnetic coupling mechanism: (i) observation of a minimum in R_K as a function of magnetic field, (ii) a decrease in R_K as ^{3}He pressure is increased, and (iii) a nearly order-of-magnitude increase in R_K resulting from preplating the silver surface with monolayers of ^{4}He (with no such effect at $T>30$ mK). [S0163-1829(96)50938-0]

Efficient heat transfer between solids and liquid helium, despite becoming increasingly difficult to achieve at lower temperatures, is a crucial element in experiments at millikelvin and still lower temperatures. For example, the performance of a dilution refrigerator depends critically on efficient energy exchange between the warm incoming and the cold outgoing ^{3}He streams, typically across a metal interface. Cooling and measuring the temperature of the sample also require good thermal contact between the liquid helium and solids.

As essential as this energy exchange between liquid He and solids is, however, the physical process responsible for it remains poorly understood and represents a long-standing problem in low-temperature physics. At these low temperatures, the bottleneck in the energy transfer typically occurs at the solid-liquid helium interface. This is known as the Kapitza resistance and is defined as

$$R_K=\frac{\Delta T}{\dot{Q}/A}, \tag{1}$$

where $\dot{Q}/A$ and ΔT are the heat flux and temperature discontinuity across the solid-liquid interface.

In this paper, we limit our discussion to R_K between metals and liquid ^{3}He. We present precise measurements of R_K as a function of temperature, magnetic field, ^{3}He pressure, and ^{4}He impurity and report direct evidence for a magnetic energy transfer mechanism that is in close qualitative agreement with existing theories.

At $T<100$ mK, the specific heat of liquid ^{3}He and that of metal are dominated by single-quasiparticle excitations and by the electron gas, respectively, so that sufficient energy exchange between the quasiparticles and electrons is essential for good thermal contact between the two materials. It has been shown,[1] however, that direct heat transfer between ^{3}He quasiparticles and conducting electrons is very inefficient because the degeneracy temperature of a metal is greater than the electron spin fluctuation temperature by a factor of 10^8.

A possible indirect path for heat flow is from quasiparticles to zero-sound phonons in ^{3}He, then to acoustic phonons in metal through surface coupling, and finally to the electrons. Because of the large difference in the speeds of sound in the two media (2×10^2 m/s and 5×10^3 m/s, respectively), only phonons within a small ($\sim3°$) critical cone normal to the interface can propagate across the interface, while others are reflected, and this results in a large R_K.[2] Later enhancements[3–5] to the acoustic mismatch theory (AMT) consider both the longitudinal and transverse zero-sound modes in ^{3}He. Using the notation of Nakayama,[6] R_K due to phonons is

$$R_K=\frac{15\hbar^3\rho_S v_T^3}{2\pi^2\rho_L k_B^4 T^3(a_1c_{L0}+a_2c_{T0})F}, \tag{2}$$

where ρ_S is the solid mass density, v_T the transverse speed of sound in the solid, ρ_L the liquid ^{3}He mass density, c_{L0} and c_{T0} the longitudinal and transverse speeds of zero sound in ^{3}He, and F a material-dependent function which is calculated numerically. The remaining parameters, a_1 and a_2, are of order 1 and are functions of the Landau parameters F_0^S and F_1^S.[6] The predictions of AMT usually agree with experimental observations in the temperature range of 100 mK to 1 K.[7]

At $T<30$ mK, however, R_K is widely recognized to be anomalously lower than predicted by AMT, and the temperature dependence deviates from T^{-3}.[7] This indicates a more efficient heat transfer channel at the boundary than that provided by phonons. One possible explanation is the inelastic scattering of the nuclear spins of ^{3}He quasiparticles by localized electron spins on the surface of the solid, where magnetic dipole-dipole interactions cause a direct energy transfer that bypasses the phonons.[7]

A number of theories for magnetic coupling have been proposed to explain the anomalous R_K. Several different states of localized electron spins have been considered—noninteracting spins,[8] spins in random internal fields,[9] ferromagnetically aligned spins,[9] and spin glass.[10] All of these theories predict an R_K that is drastically different from the AMT prediction. Below the electron spin ordering temperature T_o, R_K decreases as T increases, with an explicit temperature dependence that is model dependent. At $T>T_o$, however, R_K *increases* linearly with T.

Relying on temperature dependence to determine whether the coupling is magnetic or acoustic can be problematic, however, because when soft-phonon modes and size effects are taken into consideration,[6,11] AMT also produces a thermal resistance that deviates from $R_K \propto T^{-3}$. On the other hand, a dependence of R_K on magnetic field is only possible with magnetic coupling, since phonon properties are independent of magnetic field. The presence of ^{4}He monolayers on the solid surface also produces drastically different effects in the two models. In the magnetic coupling model, ^{4}He monolayer coverage greatly suppresses the energy transfer because the dipole-dipole interactions at the interface are sensitive to the distance between the dipoles. No such effect is expected in AMT, however, because ^{3}He phonon wavelengths (~ 3 μm at 1 mK) are greater than the thickness of ^{4}He monolayers.

Experimental work on the dependence of R_K on magnetic field strength and ^{3}He pressure, however, has yielded seemingly conflicting findings. For example, in a ^{3}He-Pt powder system,[12] R_K is insensitive to magnetic field strength up to 0.2 T and decreases steadily for higher fields, while changes in ^{3}He pressure have no effect on R_K. The ^{3}He-Ag sinter system,[13] on the other hand, behaves quite differently. R_K increases with magnetic field strength up to 0.2 T but becomes insensitive to higher fields up to 9.4 T, while increasing ^{3}He pressure causes an increase in R_K. Meanwhile, other measurements of R_K between ^{3}He and Ag and Cu powders reveal no dependence of R_K on magnetic field strength and ^{3}He pressure.[14–16]

The effect of ^{4}He monolayers is equally ambiguous. Some experiments show an order-of-magnitude increase in R_K when monolayers of ^{4}He are present,[17,18] while other experiments show no such effect.[14,16] Therefore, no conclusive experimental evidence has so far emerged for the mechanism of heat transfer between metals and liquid ^{3}He below 30 mK.

A possible reason for the difficulties in obtaining consistent experimental results may lie in the geometry of the powder or sinter samples.[7] Typical grain size for the powders is 70 nm, while a typical mean free path for electrons in bulk metal is about 5 μm and the mean free path of quasiparticles is 0.3 μm for pure ^{3}He at 10 mK. Consequently, the mean free paths of both the electrons and the ^{3}He quasiparticles are boundary limited. Therefore, the thermal resistance of the sinter and liquid ^{3}He can be significantly higher than that of bulk metal and ^{3}He, and this makes accurate temperature measurement difficult. Size effects also make comparison with theory difficult, because most theories were developed for bulk solids and bulk liquid ^{3}He. Properties of ^{3}He in pores of powders are different from those of bulk ^{3}He,[7] while the soft-phonon modes associated with the fractal geometry of sinters give rise to a different density of states than that found in a bulk solid.[6]

To avoid complications from size effects, solids larger than the ^{3}He mean free path are desirable.[19] We have chosen a geometry where the solid-liquid interface is flat. Our sample (Fig. 1) consists of thin silver films deposited on two sides of a thin glass slide, forming a parallel-plate capacitor with silver films as the electrodes and glass as the dielectric medium. Because the dielectric constant of glass is sensitive to temperature changes in our temperature region,[20] the glass

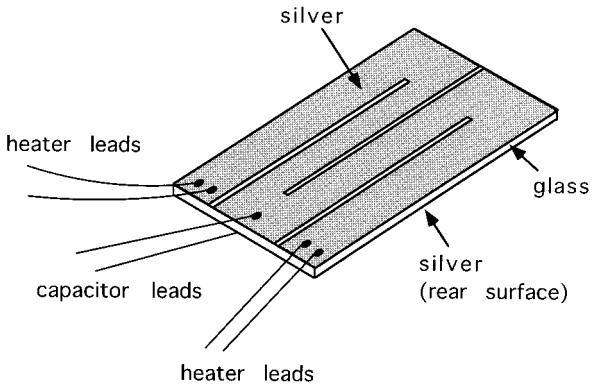

FIG. 1. A schematic drawing of a glass capacitor used for Kapitza resistance measurements. The thin silver films serve as electrodes for the parallel-plate capacitor, and one of the electrodes, with grooves cut into the metal to increase the resistance, serves as a heater.

capacitor serves as a thermometer for measuring the temperature of the silver films. There are several advantages here: (i) The flat interface eliminates size effects of ^{3}He confined in the pores of powders. (ii) Comparison with theory is easier since the experimental and theoretical configurations are now compatible. (iii) The small thermal mass of the glass capacitor ensures good thermal equilibrium within the solid phase and a fast thermal response time—both of which are desirable for accurate temperature measurements.

Two sample cells were constructed and used. sample I was an ordinary microscope cover glass (Corning Brand, $7.5\times 18\times 0.24$ mm^3). Silver films of 0.5 μm thickness were thermally evaporated onto both sides of the glass. The sample was put inside a superconducting solenoid that provided magnetic fields parallel to the solid-^{3}He interface. Sample II was made with a larger piece of glass (Fisher Brand microscope slide, $38\times 25\times 1$ mm^3). Silver films of 0.5 μm thickness were sputtered onto the glass. The silver films of sample I were examined under an electron microscope and were found to be smooth to at least 0.1 μm. Because of the loss in sensitivity of the glass thermometers at the lowest temperatures, our measurements were limited to $T>3$ mK for both samples.

The sample cell was fitted inside a silver stage that was thermally anchored to a $PrNi_5$ demagnetization stage in a dilution refrigerator. The capacitor was immersed in liquid ^{3}He, and the ^{3}He temperature was measured with a ^{3}He melting curve thermometer. A heat flux from the silver films to the liquid ^{3}He was introduced by passing a direct current I through one of the electrodes, causing Joule heating due to the resistance of the silver film. The temperature rise of the silver films was measured by monitoring the impedance of the glass capacitor with a symmetric ratio-transformer bridge. The heating power was controlled to ensure $\Delta T/T<10\%$, and the thermal response time of the glass capacitor was less than 1 min in the absence of magnetic fields, even at the lowest temperatures. R_K was determined by Eq. (1). (More information on sample preparation and the measurement technique is given elsewhere.[21])

For accurate measurement of R_K between ^{3}He and the

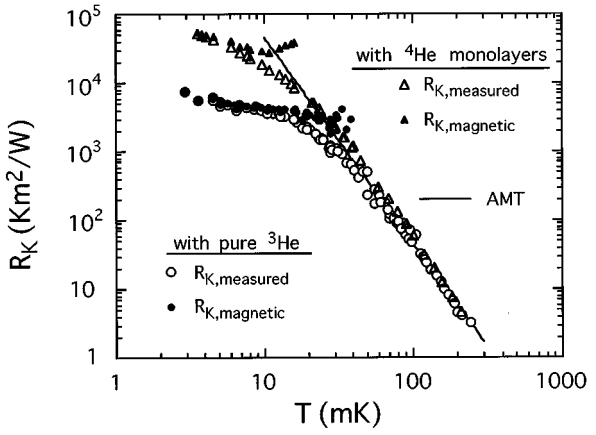

FIG. 2. Temperature dependence of R_K at saturated vapor pressure of ^{3}He (sample II). The long line represents the zero-free parameter predictions of AMT.

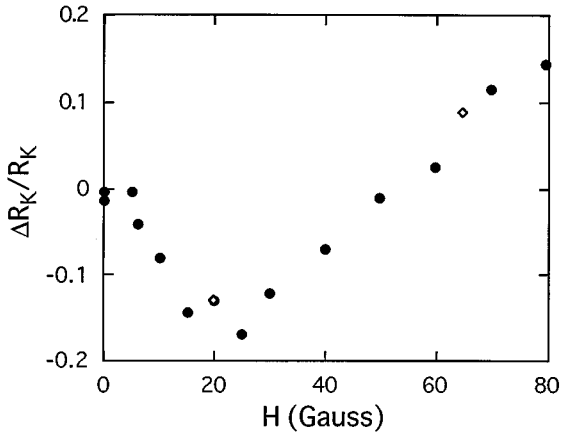

FIG. 3. The fractional change of R_K in small magnetic fields (sample I). The direction of the field is parallel to the solid-liquid interface. A minimum is seen at $\mu_B H/k_B T \approx 1/4$. The open diamonds are data taken with the magnetic field reversed in direction.

silver films, the applied heat $\dot{Q}$ had to flow from the surface of the silver films to the surrounding ^{3}He but not through the electrical wires. The thermal conductance through the wires was measured when the cell was empty of liquid ^{3}He and was found to be at least two orders of magnitude smaller than the conductance through the ^{3}He-silver interface, so that it could be ignored.

Figure 2 shows the temperature dependence of R_K of sample II at saturated ^{3}He vapor pressure for both pure ^{3}He and ^{3}He mixed with 1.3% ^{4}He. Because ^{4}He preferably plates the solid surfaces, the amount of ^{4}He in the ^{3}He-^{4}He mixture accounts for about six monolayers of ^{4}He on the surface of the silver films. Data in Fig. 2 show that for $T>30$ mK, measured values of R_K for both pure and impure ^{3}He agree well with AMT, not only in the exponent of the temperature dependence, but also in magnitude. This strongly supports the main heat transfer channel above 30 mK being phonon mediated.

For $T<30$ mK, measured values of R_K for pure ^{3}He are much smaller than predicted by AMT. However, this improvement of heat transport is hindered by a factor of 10 or so when monolayers of ^{4}He are present between ^{3}He and the silver films. This rules out the possibility that the dominant heat transport mechanism is acoustic and at the same time provides evidence for a magnetic coupling mechanism for reasons described earlier.

Figure 3 shows the dependence of R_K (sample I) on magnetic field at a constant temperature of 6.7 mK. As the field increases, R_K first decreases and then increases, with a minimum at a field strength of about 2.5 mT. (Two data points taken with reversed fields are also included to exclude the possibility of a residual trapped field.) The theoretical analysis of R_K in magnetic fields by Leggett and Vuorio[8] also shows a minimum of R_K at $H \approx 3k_B T/2\mu_B$, where μ_B is the Bohr magneton. The physical origin of this is that at zero field all excitation energies of the electron spins are small compared to $k_B T$. When a small field is applied, the excitation energies are raised toward $k_B T$, where the energy transfer becomes more efficient and R_K decreases. As $\mu_B H$ becomes larger than $k_B T$, however, the energy transfer becomes less efficient again, and R_K increases. The minimum of R_K in our experiment occurs at $\mu_B H/k_B T \approx 1/4$, in close agreement with theory.

R_K also depends on the pressure of ^{3}He, because as ^{3}He pressure increases, the ^{3}He quasiparticle mass m^* also increases. (From saturated vapor pressure to 5 and 10 bars, m^* increases by factors of 1.23 and 1.38, respectively.[22]) This results in a higher density of states at the Fermi level and smaller R_K. Theoretical calculations[8–10] indicate $R_K \propto (m^* k_F)^{-2}$, where k_F is the Fermi wave number of ^{3}He, which also increases with increasing ^{3}He pressure. Even though the experimental observations show this general trend, the pressure dependence seems weaker than theoretical predictions. Figure 4 shows that to within 5% experimental uncertainty, $R_K m^*$ is independent of ^{3}He pressure for $T<30$ mK and for ^{3}He pressures up to 10 bars.

The discrepancy between the theories and our empirical scaling law ($R_K \propto m^{*-1}$) does not come as a surprise, however. The reason could be that the theories all treat the liquid

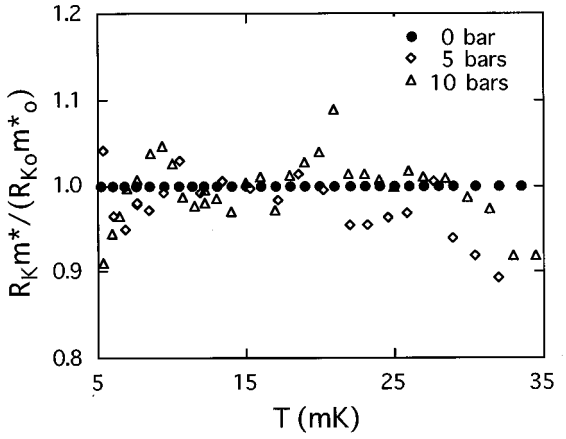

FIG. 4. The ratio of $R_K m^*$ at 0, 5, and 10 bars of ^{3}He pressure to the values at saturated vapor pressure (sample I). The data show an empirical scaling law $R_K \propto m^{*-1}$.

^{3}He as a bulk Fermi liquid up to the solid-liquid interface, while the first two monolayers of ^{3}He on the solid are really solid and dense liquid, respectively.[23] Assuming that it is the contact interaction between the ^{3}He quasiparticles and electron spins on the solid surface that determines the heat transfer, we expect the first layers of ^{3}He on the solid surface to play an important role, so that they cannot be treated as bulk ^{3}He fluid.

Finally, we examine the temperature dependence of R_K at low temperatures. Assuming the magnetic and acoustic thermal conduction channels to be in parallel, the measured R_K can be expressed as

$$R_{K,\text{measured}}^{-1} = R_{K,\text{acoustic}}^{-1} + R_{K,\text{magnetic}}^{-1}. \tag{3}$$

We use Eq. (2) for $R_{K,\text{acoustic}}$ and plot $R_{K,\text{magnetic}}$ in Fig. 2. We see that in the absence of the ^{4}He monolayers, the magnetic channel provides the dominant thermal conductance for $T<15$ mK. If fitted to a power law $R_{K,\text{magnetic}} \propto T^{-n}$, the experimental value for n is ~ 0.4, while the theoretical prediction is 2 for randomly distributed localized electron spins below T_o.[8–10] This discrepancy may reflect our lack of knowledge of the state of the localized electron spins or may reflect the inadequacy of treating ^{3}He as a bulk Fermi liquid at the interface.

It is worth noting that below 30 mK, R_K values in our experiments are similar in magnitude to values in a $PrNi_5$-^{3}He system[19] and a Pt-^{3}He system,[24] where in both cases size effects have been eliminated by using larger-diameter particles. The temperature dependence of R_K in these two powder systems, however, is T^{-2}.

At present, the question of what the localized electron spins might actually be remains to be fully addressed. We could not address this in our experiments, because the surface conditions were not controlled. It has been suggested[6,25] that adsorbed oxygen on the surface of the solid could form a paramagnetic submonolayer and the magnetic coupling could be between oxygen impurities and ^{3}He quasiparticles. Future investigation of the effects of oxygen and how oxygen couples to free electrons in metals is therefore desirable for gaining a complete understanding of Kapitza resistance at low temperatures.

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