Evidence for Magnetic Coupling in the Thermal Boundary Resistance between Liquid ^{3}He and Platinum

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The first measurements are reported showing that the thermal boundary resistance between a metal and liquid ^{3}He depends on the applied magnetic field. The resistance, which is not affected by pressure changes or superfluidity of the liquid, is insensitive to fields less than 2 kG, but decreases by a factor of 3 when the field is increased to 8 kG. The experimental results are explained by a model which assumes that the thermal transfer between the Pt and ^{3}He is due to a magnetic coupling between the magnetic moments of conduction electrons in the metal and the nuclear spins of the solid ^{3}He layer on the Pt surface.

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A long-standing unsolved problem in low-temperature physics is the unexplained thermal boundary resistance R between liquid ^{3}He and solids.[1] The acoustic mismatch theory[2] overestimates the resistance and cannot explain the observed temperature dependence, $R \propto 1/T$, for heat conduction between liquid ^{3}He and metals at temperatures below 20 mK.[3] Several authors have proposed that heat transport across the boundary is due to magnetic coupling between ^{3}He nuclear spins and magnetic impurities in the metal,[4] similar to the proposed coupling between ^{3}He and the paramagnetic salt cerium magnesium nitrate.[5-7] The major objection to this suggestion[3] has been the absence of any observed magnetic field dependence of R for fields up to 1.7 kG.[8] In this Letter we report the first measurements that show a magnetic field dependence of the boundary resistance between liquid ^{3}He and a metal. We also propose a theoretical model for the resistance, based on a magnetic coupling between the conduction-electron spins and the nuclear spins of the solid ^{3}He surface layer. This model accounts for all of the experimental observations.

The measurements we report here were made on one sample of platinum powder, with a specific surface area of 0.10 m^2/g as measured using argon adsorption isotherms. Transmission-electron micrographs of the powder show irregularly shaped particles with sizes between 1 and 10 μm. The powder, which was not cleaned, was mixed with an equal volume of 1-μm graphite powder and packed inside a coil with a platinum packing fraction of 11%. The cell containing the coil held 6 cm^3 of ^{3}He (^{4}He impurities less than 50 ppm), and contained a sintered silver heat exchanger with a surface area of 128 m^2 attached to a copper demagnetization stage. The temperature of the powder was determined from its nuclear susceptibility, as measured using cw nuclear magnetic resonance (NMR). This thermometer was calibrated with both the superfluid transition temperature and the Pt spin-lattice Korringa relaxation time. The platinum was heated slightly above the ^{3}He by applying an off-resonance radio-frequency voltage to the NMR coil.

Two different methods were used to determine the boundary resistance. In one method, pulses of eddy-current power were used to heat the Pt. When the heating field was turned off, the temperature difference relaxed exponentially with a time constant τ. Above a few millikelvins, τ is the spin-lattice relaxation time T_1 due to the Korringa process in Pt, but below 5 mK τ is longer than T_1, suggesting that there are two thermal resistances between the Pt nuclei of the thermometer and the ^{3}He. One is the resistance between the Pt nuclei and conduction electrons, characterized by T_1, and the other is the thermal boundary resistance R. Using this analysis, we have $R = A(\tau - T_1)/C$, where C is the magnetic heat capacity of the Pt nuclei ($C \propto H^2/T^2$). The second method for measuring the resistance used a steady rf field to heat the Pt to a temperature greater than that of the ^{3}He by an amount ΔT; the boundary resistance was then calculated from $R = A\Delta T/\dot{Q}$, where A is the surface area and $\dot{Q}$ is the heat flow across the boundary. The proportionality factor between $\dot{Q}$ and the rf power applied to the coil was determined from a comparison of R with the pulsed heating measurements, but even without this factor, the temperature and field dependence of R can be obtained by this steady-state method.

Both methods of measurement give the same results, as shown in Fig. 1. At each field value, we find $R \propto 1/T$. Measurements made at a pres-

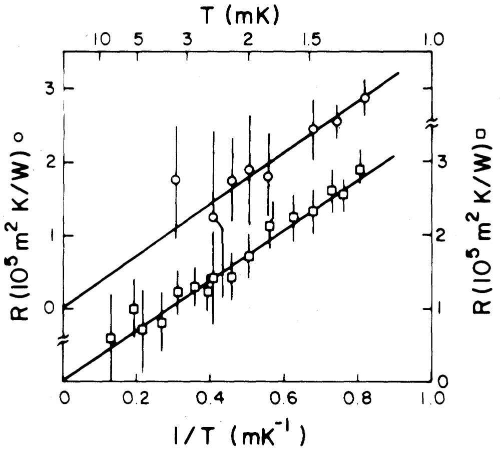

FIG. 1. Thermal boundary resistance R at 2.61 kG, obtained by two different methods: pulsed (circles, left scale) and steady-state (squares, right scale). Solid lines represent least-squares fits.

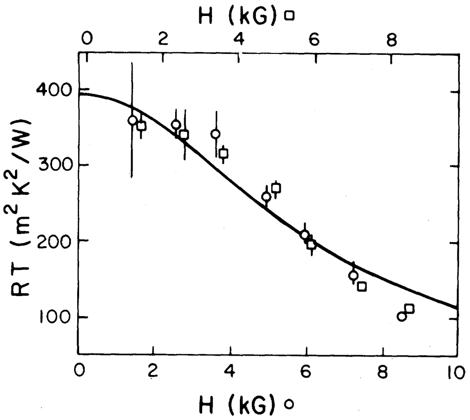

FIG. 2. The product of thermal boundary resistance and the temperature plotted as a function of the field. Data taken by the steady-state method (squares) are offset to the right of those taken by the relaxation-time method (circles) in order to show the error bars clearly. The solid curve results from the theory when $\hbar\omega_e/k_B = 0.1$ mK, $T = 2$ mK, and $g_0 = 10^{-6}$ eV.

sure of 20 bars are indistinguishable from those made at less than 1 bar, and the superfluid transition did not alter the resistance. The product RT, which is independent of area and temperature, is plotted in Fig. 2 as a function of the static magnetic field. The resistance decreases by a factor of 3 from its low-field value as the field increases to 8 kG. This is the first observation of any field dependence of the thermal boundary resistance between ^{3}He and a metal.

To date, no theoretical model has been proposed which accounts for most of these observations. We propose that at very low temperatures the thermal boundary resistance between the metal and the liquid ^{3}He is dominated by a weak magnetic coupling of the conduction electrons to the localized nuclear moments in the first atomic layer of ^{3}He that plates the Pt surface. The conductivity between the solid ^{3}He layer and the liquid ^{3}He is much larger than that between solid ^{3}He and Pt primarily because of the much larger liquid ^{3}He density of states compared with the Pt electronic density of states.

The conduction electron–solid ^{3}He coupling explains all of the qualitative experimental observations. With this mechanism, the boundary resistance is independent of the liquid pressure, is unaffected by the superfluid transition, and increases when ^{4}He impurities are added to replace the ^{3}He atoms at the helium-metal interface.[9] The absence of any field dependence of the boundary resistance below 2 kG rules out a coupling of localized electronic moments to the ^{3}He because the electronic Zeeman energy is much larger than the thermal energy. Furthermore, the field dependence cannot be due to the conduction-electron or ^{3}He Fermi liquids because their susceptibilities are field independent unless H is comparable to the Fermi energy, which corresponds to fields of about 10^4 kG in both cases. However, the magnetic coupling to the localized ^{3}He nuclei in the solid layer is affected by the field when the Larmor frequency becomes comparable to the exchange frequency in solid ^{3}He, which corresponds to a field of order 1 to 10 kG.

To quantify these remarks on the field and temperature dependence of the thermal boundary resistance, we use a general expression for the thermal conductance due to magnetic coupling derived by Leggett and Vuorio[6] from perturbation theory:

$$R^{-1} = \frac{\pi}{2}\hbar^2 k_B^{-1} g_0^2 \beta^2 n_A \times \sum_\alpha \int_0^\infty \frac{\omega^2 \chi_\sigma(\omega)\chi_{s_\alpha}(\omega)}{\sinh^2(\beta\hbar\omega/2)}\,d\omega, \qquad (1)$$

where $\beta = 1/k_B T$, g_0 is the coupling strength between conduction-electron spins and the solid ^{3}He spins (to be discussed below), $n_A = 10^{19}$ m^{-2} is the areal density of nuclear spins in the boundary layer, $\chi_\sigma(\omega)$ is related to the conduction-electron spin-fluctuation spectral function, and $\chi_{s_\alpha}(\omega)$ is the same quantity for a ^{3}He nucleus at site α in the solid layer. For experimentally accessible fields and temperatures, $\chi_\sigma(\omega)$ is determined by the dynamic susceptibility of a free Fermi gas of conduction-electron quasiparticles, so that Eq. (1) can be expressed as[10]

$$R^{-1}=\pi(k_B g_0)^2\hbar^{-1}N_e^2(E_F)K_{\mathrm{eff}}^2 n_A T Q_s(T,H), \tag{2}$$

with

$$Q_s(T,H)=\tfrac{1}{4}\beta^3\hbar^4\sum_\alpha\int_0^\infty \frac{\omega^3\chi_{s_\alpha}(\omega)}{\sinh^2(\beta\hbar\omega/2)}\,d\omega, \tag{3}$$

where $N_e(E_F)=1.7$ eV^{-1} is the electronic density of states at the Fermi energy, $K_{\mathrm{eff}}^2(\sim 7)$ is the electron-spin susceptibility enhancement factor, and $Q_s(T,H)$ depends only on T,H, and the properties of the solid layer of helium.

The spectral density $\chi_{s_\alpha}(\omega)$ is proportional to the imaginary part of the solid ^{3}He magnetic susceptibility at a single site on the surface. To determine its form, we note that the coupling to liquid ^{3}He atoms is strong because of exchange between the liquid and the solid, and this exchange frequency ω_e is the characteristic frequency for $\chi_{s_\alpha}(\omega)$. For exchange between a localized spin and a Fermi liquid, the exchange frequency is given by

$$\omega_e=4\pi\hbar^{-1}J^2N_3^2(E_F)k_BT, \tag{4}$$

where J is the exchange energy and $N_3(E_F)$ is the density of states at the liquid ^{3}He Fermi energy. We estimate $J/k_B\sim 0.1$ K, which is one order of magnitude smaller than the exchange energy within the liquid, so that $\omega_e\sim 10^7$ T sec^{-1} mK^{-1}. We assume a Lorentzian spectral function with width ω_e for the localized ^{3}He moments,

$$\chi_{s_\alpha}(\omega)=\frac{1}{\pi}\left(\frac{M/H}{n_A(\gamma\hbar)^2}\right)\omega_e\omega \times\left[\frac{1}{(\omega-\omega_0)^2+\omega_e^2}+\frac{1}{(\omega+\omega_0)^2+\omega_e^2}\right], \tag{5}$$

where M is the surface magnetization in a static external field H, ω_0 is the corresponding Larmor frequency, and $\gamma\hbar$ is the ^{3}He nuclear moment. For small H, $M/H=\chi$, the Curie susceptibility. In zero field, the temperature dependence of $Q_s(T,H=0)$ can be scaled out of Eq. (3), so that $R\propto 1/T$, as observed experimentally.

The magnetic field dependence of the resistivity is determined by Eqs. (2)–(5). For $\omega_0\lesssim\omega_e$, corresponding to $H\lesssim 1$ kG, $Q_s(T,H)$ is approximately constant. For larger fields, R decreases until $\hbar\omega_0$ is of order k_BT; for still larger fields the resistivity increases with H. Numerical integration of Eq. (3), using $\omega_e=0.1$ mK, $T=2$ mK, and $g_0=10^{-6}$ eV (see below), produces the solid curve shown in Fig. 2. With these values, this model predicts a minimum resistivity, with $R_{\min}/R(H=0)=0.05$, at a magnetic field of 40 kG.

The interaction between the conduction electrons and the ^{3}He nuclei is

$$H_{\mathrm{int}}=g_0\sum\nolimits_\alpha\vec{\sigma}_\alpha\cdot\vec{S}_\alpha, \tag{6}$$

where $\vec{\sigma}$ and $\vec{S}$ are the Pauli spin operators for the conduction electron and the localized ^{3}He nucleus, respectively. One way to estimate g_0 is to assume that the coupling is due to exchange between Pt conduction electrons and the ^{3}He electrons.[11] The repulsion that prevents ^{3}He atoms from entering the metal is due to overlap of the Pt and ^{3}He electronic wave functions, and so that exchange rate should be large. To estimate the perturbation of the ^{3}He electronic state we note that the adsorbed atom has an electric dipole moment, as shown by field-emission studies of inert-gas atoms adsorbed on metallic surfaces.[12] From these measurements and the polarizability of ^{3}He, we estimate the dipole moment to be 0.1 D. This dipole moment results from a mixing of the $1s2p$ state with the $1s^2$ ground state. Using hydrogenic wave functions, we find that the mixing fraction is 0.05. The interaction strength between the ^{3}He electronic and nuclear moments is then $g_0\sim\mu_B\mu_N|\Psi(0)|^2$, where $|\Psi(0)|$ is the amplitude of unpaired electron spin at the nucleus. We find $|\Psi(0)|^2\sim 2\times 10^{24}$ cm^{-3}, giving $g_0\sim 2\times 10^{-6}$ eV, and $RT\sim 10^2$ m^2 K^2 W^{-1}, in accord with experiment. If this estimate is correct, the interaction should also produce a Knight shift[13] for the ^{3}He nuclei in the first solid layer with $\Delta H/H\sim 5\times 10^{-3}$.

In this Letter we have shown that the low-temperature thermal boundary resistance between liquid ^{3}He and Pt depends on the applied magnetic field. This magnetic field dependence establishes the importance of the solid ^{3}He layer,[14] and, when combined with other experimental observations, indicates that the magnetic interaction between conduction electrons and ^{3}He nuclei in the solid surface layer provides the dominant path for heat flow across the interface.

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Effective Mass of the Normal Negative-Charge Carrier in Bulk He II

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The first acceleration measurements are reported for negative ions in bulk superfluid ^{4}He. Data recorded at 70 mK, 25 bars for electric fields in the range $80 < E < 150$ V m^{-1} are consistent with an ionic dispersion relation identical to that of a free particle with mass $(87 \pm 5)m_4$, where m_4 is the ^{4}He atomic mass.

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Studies of the motion of positive and negative charge carriers have provided a particularly fruitful means of probing the properties of the (magnetically inert) superfluid ^{4}He. During the two decades following the pioneering experiments of Reif and Meyer[1] some hundreds of scientific papers have been published which describe work carried out on this basis[2] and which yield extensive and valuable information concerning, for example, the elementary excitations, critical velocities, surface properties, and macroscopic quantum states of the liquid. A quantity which is frequently of relevance to the analysis of experimental data is the effective mass m^* of the charge carrier (ion). It seems at first sight remarkable, therefore, that only one precise experimental determination[3] of m^* has so far been reported, and especially so considering that this measurement, being based on the resonance of ions trapped in a surface potential well, was entirely confined to the saturated vapor pressure. The latter restriction has given rise to what must be regarded as a serious lacuna in the experimentally determined properties of the negative ion. This is because of the expectation,[2] on sound theoretical grounds, that m^* for the negative ion should be strongly dependent on pressure: A decrease in m^* by a factor of circa 3 is to be anticipated between the saturated vapor pressure and the solidification pressure at 25 bars. It has not hitherto been possible to test this prediction experimentally. The purpose of the present Letter, however, is to report the development of a new technique which has enabled us to measure, for the first time, the effective mass of negative ions freely moving in bulk He II under elevated pressures.

The technique is a particularly straightforward and direct one, at least in principle, in that we simply measure the acceleration of the ion under a weak electric field E in the "mechanical vacuum" provided by isotopically pure He II at 70 mK, 25 bars. Under these conditions, drag due