Magnetic Coupling between ^{3}He and ^{19}F at Low Temperatures

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Measurements of relaxation phenomena between liquid ^{3}He and ^{19}F nuclei in small fluorocarbon particles are reported. Magnetic cross relaxation between the ^{19}F in the substrate and the liquid is observed in measurements between 1 K and 0.6 mK. The spin temperatures remain strongly coupled in magnetic fields up to 125 mT. Moreover it is observed that there is a decrease in magnetic relaxation time at the onset of superfluidity in the liquid ^{3}He and that the thermal relaxation times are remarkably short at all temperatures.

The nature of the coupling of excitations at the interface between liquid ^{3}He and solids remains one of the most significant unsolved problems in very-low-temperature physics. At temperatures less than 10 mK, heat is conducted from most materials into liquid ^{3}He more rapidly than can be understood in terms of lattice vibration models, and spin excitations in liquid ^{3}He in confined spaces relax much more rapidly than is expected for bulk liquid.[1]

In the work reported here, we have studied the system of liquid ^{3}He filling the pores of a loosely packed powder of small fluorocarbon particles at low temperatures. We have measured the equilibrium magnetization of the ^{3}He and of the ^{19}F in the particles, the rates of recovery to thermal equilibrium of both spin species, and the behavior of the ^{3}He magnetization when the ^{19}F magnetization is perturbed from the equilibrium. The experiments extend from 1 K to below 0.6 mK and were performed in applied magnetic fields up to 125 mT.

The particles are composed of a fluorocarbon polymer similar to Teflon and are sold by Dupont under the trade name DLX6000. They are approximately spherical with a rather uniform radius of about 0.1 μm. They were packed in a cylindrical coil form with a volume filling factor of approximately 25%. The coil form was immersed in liquid ^{3}He in a tower mounted on a silver chamber containing a sintered silver heat exchanger and the liquid ^{3}He. The apparatus was cooled by adiabatic demagnetization of PrNi$_5$. Temperatures were measured by a small ^{3}He melting-curve thermometer similar to that described by Greywall and Busch,[2] attached to the silver chamber. A paramagnetic salt thermometer immersed in the liquid ^{3}He was used in the submillikelvin regime. Further details about the apparatus will be given elsewhere.[3] We have measured the

equilibrium susceptibility of both the ^{19}F and the ^{3}He at temperatures down to 0.6 mK, and the relaxation behavior of the two spin species under various perturbations. In addition to conventional measurements of the nuclear relaxation times of the ^{19}F and ^{3}He spins we have observed the ^{3}He magnetization during ^{19}F relaxation. The ^{3}He is strongly affected by the strong coupling across the interface separating the substrate and the liquid. An unexpected result in the relaxation measurements was the quite marked effect of the superfluid transition on these relaxation times.

The ^{19}F susceptibility was measured between 1 K and 0.6 mK by use of both cw and pulsed NMR techniques and was found to follow Curie's law down to the lowest temperatures. The measurement of the ^{19}F susceptibility in the fluorocarbon substrate would provide a very useful means of determining the inverse temperature at very low temperatures since the T_1's are short down to the lowest temperatures we could achieve. The ^{3}He susceptibility was independent of temperature between 100 and 20 mK as one would expect for bulk liquid. Down to 1 mK the susceptibility could be fitted to

$$\chi / \chi_{\text{bulk}} = 1 + A/(T - \Delta) \qquad (1)$$

where χ_{bulk} is the susceptibility measured at temperatures between 20 and 100 mK. We found A to have a value of 1.8 mK and Δ to be 0.5 mK. This form for the susceptibility has been observed by earlier workers[4] and is thought to reflect a Curie-Weiss susceptibility due to ferromagnetic tendencies of the solid layer of ^{3}He on the surface of the particles which adds to the bulk susceptibility of the remaining liquid.

We show in Fig. 1 a plot of the ^{19}F relaxation time as a function of temperature at a pressure of 0.75 bar and at two values of the applied magnetic field. The measurements were made in the
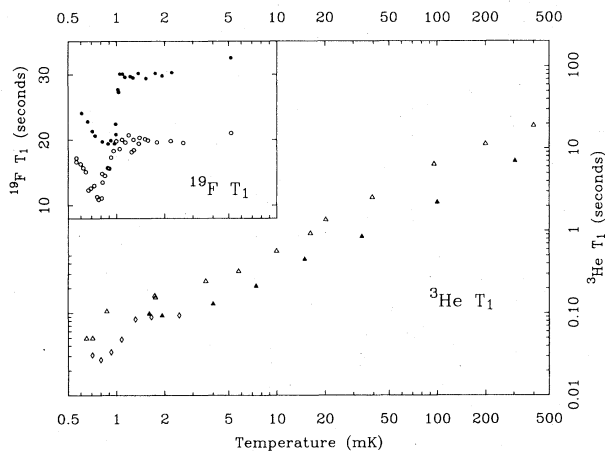
FIG. 1. Relaxation times at low temperatures. The triangles show the ^{3}He relaxation times at 63 mT (closed triangles) and 125 mT (open triangles), both taken at a pressure of 0 bar. The open diamonds show ^{3}He relaxation times taken at 63 mT and at 0.75 bar. The inset shows the longer ^{19}F relaxation times at applied magnetic fields of 50 mT (closed circles) and 100 mT (open circles). Both ^{19}F T_1 measurements were made at a pressure of 0.75 bar.

FIG. 2. Magnetic coupling between ^{19}F and ^{3}He. The ^{3}He magnetization drops rapidly following an initial pulse at the ^{19}F resonant frequency, then relaxes on a time scale long compared to the ^{3}He T_1. The three curves show the response at three temperatures. All were taken in an applied magnetic field of 125 mT and at a pressure of 0 bar. M_∞ is the ^{3}He magnetization before the ^{19}F magnetization is disturbed. The lines are guides to the eye only.

usual manner by monitoring the ^{19}F magnetization after tipping the ^{19}F spins with a pulse at their resonant frequency. We also show for comparison the much faster relaxation times for the ^{3}He spins at 0 and 0.75 bar. The ^{19}F relaxation time shown is nearly independent of temperature between 200 and 1.5 mK; this is also true at 0 bar and 1.44 bar. At about 1 mK the relaxation time decreases rapidly. As the pressure is varied the temperature at which this sudden change occurs varies in a way that suggests that it is associated with the superfluid transition in the pores of the powder. The transition temperature is expected to be suppressed by roughly 30% in this confined geometry because the coherence length of the superfluid (of order $h v_f / \pi k T$) is comparable to the dimensions of the pores.[5,6] Below T_c the relaxation times begin to increase as temperature decreases with a temperature dependence which is the same at both 0.75 and 1.44 bar; we are unable to cool sufficiently far below T_c at 0 bar to see the minimum in T_1. Figure 1 shows the relaxation times in magnetic fields of 100 and 50 mT. The relaxation times are reduced by a factor of 0.7 above T_c and slightly more below T_c when the field is reduced by a factor of 2.

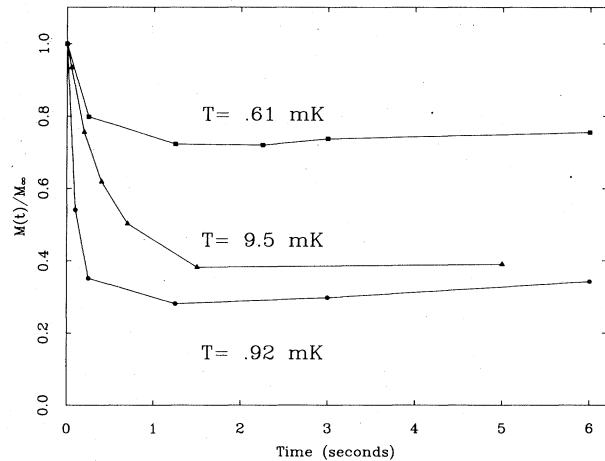The relaxation time of the ^{3}He was determined by measuring the height of the ^{3}He spin echo at

varying times after the ^{3}He magnetization was tipped through 135 degrees. A 1-h wait between measurements was required for the system to return to thermal equilibrium after the pulses. The relaxation time of the ^{3}He is approximately three orders of magnitude shorter than that of the ^{19}F at the lowest temperatures. Above T_c, T_1 was measured to be proportional to temperature in agreement with earlier work.[4,7] Near T_c the ^{3}He T_1 drops, but less rapidly than the ^{19}F T_1. Depending on the pressure, T_1 became temperature independent or even passed through a minimum below T_c. Also in agreement with earlier work[4,7] the ^{3}He T_1 is proportional to applied magnetic field above T_c.

We were able to observe the behavior of the ^{3}He spins as the ^{19}F relaxed by monitoring the ^{3}He magnetization after applying a pulse which rotated the ^{19}F magnetization through 135 degrees. Figure 2 shows the response at three temperatures in an applied magnetic field of 125 mT. The response is rather insensitive to temperature between 20 mK and T_c. The ^{3}He magnetization initially drops on a time scale similar to that characterizing the ^{3}He T_1 to a minimum value approximately 30% of the equilibrium value and then relaxes to equilibrium on a time scale similar to the ^{19}F relaxation times. It is useful to remember that the energy of the ^{19}F spins is 250

times greater than that of the ^{3}He spins at the lowest temperatures. During the ^{3}He relaxation only a small amount of energy is added to the cold reservoir. When the ^{19}F magnetization is inverted and relaxes, taking advantage of the strong coupling to ^{3}He spin bath, the ^{3}He spin bath is heated and the spins driven far from equilibrium for as long as is required for the ^{19}F enthalpy to finally be absorbed by the quasiparticle bath.

Earlier work has shown a strong coupling to exist between the two spin baths at higher temperatures.[8-10] Here, we have observed that this coupling persists undiminished down to the lowest temperatures that we can achieve. The ^{3}He magnetization can be altered by manipulating the ^{19}F magnetization because of an interface interaction with ^{19}F. Although we do not know the spatial distribution of the ^{3}He magnetization it is clear that the ^{3}He magnetization must be far from equilibrium with the quasiparticle reservoir for times comparable to the ^{19}F T_1. In the curve shown in Fig. 2 at 9.5 mK the minimum ^{3}He magnetization is 30% of the equilibrium value. If we use Eq. (1) and assume that all of the excess magnetization associated with the surface is also tipped by 135 degrees, this response curve shows that the magnetization of the bulk liquid must still be reduced by 35% for times long compared to the ^{3}He T_1. The total enthalpy of the ^{19}F spins is sufficient to increase the temperature of the liquid ^{3}He by less than 0.1 mK at this temperature. Yet the magnetization is suppressed to a value which is equal to the equilibrium value at a temperature comparable to the magnetic Fermi temperature ($T_F^* = 390$ mK). The degree to which the magnetization is suppressed is nearly independent of temperature above T_c. Below T_c the magnetization is suppressed by only 25%. At all temperatures the recovery time is roughly equivalent to the ^{19}F relaxation times at the same temperature.

We have no models which will readily explain all of these observations. Because bulk processes in both the ^{3}He and the fluorocarbon substrate due to the magnetic dipole interaction are many orders of magnitude slower than times measured here, surface processes must be dominant. The relaxation process must involve at least two steps: the removal of the magnetization from the ^{19}F spin bath and the relaxation of the spin disequilibrium to the quasiparticle reservoir. The response of the ^{3}He spins to an inversion of the ^{19}F spins is probably a complex process involving the competition of these two processes and possibly others. In a ^{19}F T_1 measurement the energy in the inverted ^{19}F spins must eventually be absorbed by the quasiparticle bath. We are observing an intermediate step in the ^{19}F relaxation process. Apparently energy is absorbed by the ^{3}He spin bath by allowing the Fermi energies of the up- and down-spin baths to differ. These energies equilibrate by means of ^{3}He spin flips which give up energy to quasiparticle bath. The suppressed magnetization which we observe is that at which the energy flow out of the ^{3}He spins balances the energy flow in from the hot ^{19}F spin bath.

Two processes suggest themselves as being potentially responsible for the relaxation of the ^{3}He spin disequilibrium to the quasiparticle bath. The fact that the superfluid transition strongly affects the relaxation times suggests that spin transport in the liquid is an important element in the relaxation process. If this is the case it is difficult to understand the exact form of the temperature and field dependence of the relaxation times because the transport properties have not been seen to be sensitive to applied magnetic fields of the magnitude used here. A second possibility is that the ^{3}He spins relax at the surface of the particles and that the superfluid transition has an effect on this process.

We can regard the ^{19}F T_1 as a measure of the thermal relaxation time which characterizes heat flow from the fluorocarbon solid into the liquid. This time can be related to the boundary resistance through $\tau = RC$ where C is the heat capacity, R is the boundary resistance, and τ is the measured thermal time constant. The heat capacity of the ^{19}F spins dominates the fluorocarbon sphere heat capacity and is proportional to $(H/T)^2$. Thus the very weak temperature dependence of the relaxation time constant above T_c implies that the rate-limiting resistance in transmitting the heat from the ^{19}F spins to the quasiparticle reservoir is decreasing as T^2. Most measurements of the boundary resistance reveal a resistance which increases as temperature decreases. Another exception to this generally observed behavior is in measurements on the paramagnetic salt cerium magnesium nitrate,[11,12] whose boundary resistance also falls with temperature. The magnitude of the resistance which we observe is smaller than most other materials reported.[1]

Chapellier[9] has suggested that the source of the coupling between the ^{3}He and ^{19}F in these beads is likely to be static charges or defects within

the particles. In his model the hyperfine interaction between the nuclei and the electronic charges allows a mixing between the nuclear spin species. This mechanism should be ineffective when the electronic moments become highly polarized. In this experiment with such low temperatures in a field of 100 mT such electronic moments should be completely polarized if they have a g value of 2. Chapellier has also recently reported[13] a coupling in these particles in experiments down to 30 mK and in a field as large as 8 T where the electronic moments should be highly polarized. The microscopic origin of the coupling between the ^{3}He and the ^{19}F remains unknown.

With regard to the relationship between the ^{3}He relaxation rate and the boundary resistance, Beal-Mond and Mills[14] and Guyer[15] have suggested that for such systems the boundary resistance should be proportional to the temperature times the ^{3}He relaxation time. Although our results agree with this prediction, the agreement is probably fortuitous because the times we measure are determined by relaxation to the lattice as well as to the ^{19}F spin baths.

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