Effect of Magnetic Scattering on the ^{3}He Superfluid State in Aerogel

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Pure ^{3}He in highly porous aerogel forms an equal-spin pairing superfluid with transition temperatures suppressed from bulk values. We have measured the magnetic field dependence of the transition temperature from which it can be inferred that both magnetic and nonmagnetic quasiparticle scattering from solid ^{3}He at the aerogel surface is important to the superfluid state. Replacement of the solid ^{3}He on the surface of the aerogel with ^{4}He gives rise to a unequal spin pairing superfluid phase. [S0031-9007(96)01769-3]

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It has been shown recently that liquid ^{3}He confined in high porosity aerogel undergoes a transition [1,2] to a homogeneous superfluid state, but with significant suppression of both the order parameter and transition temperature, T_c, as compared with the bulk superfluid at the same pressure. Superfluid ^{3}He in aerogel is a unique system because inhomogeneity introduced by the gel is on length scales less than the superfluid coherence length. In this sense it is a "dirty superfluid" where the gel appears to the ^{3}He as a dilute impurity of quenched disorder in analogy with the dirty limit in superconductors. Dirty superfluid ^{3}He is unlike any superfluid previously studied in confining media, such as Vycor glass and packed metal powders [3], where an inhomogeneous order parameter exists, but is locally suppressed near surfaces [4,5] owing to quasiparticle scattering. Recent theoretical work has developed these ideas taking into account quasiparticle scattering anisotropy and orientational disorder of the order parameter [6].

In previous work ^{3}He superfluidity in 98.2% aerogel has been observed in two different experimental arrangements. Torsional oscillator measurements [1] of the superfluid density in zero magnetic field give a clear indication of a sharp onset of superfluidity over a range of pressures. Similar results were found in nuclear magnetic resonance (NMR) measurements of resonance frequency shifts in 1.117 kOe [2] indicating the onset of dipolar order from which significant suppression of the order parameter was determined. Differences in T_c found in these two experiments have been attributed to variations between aerogel samples. The magnetization was found from the NMR experiment to be temperature independent as expected for an equal-spin pairing phase. An equal-spin pairing (ESP) superfluid consists of bound pairs of spin-1/2 quasiparticles in the same spin state. Previous experience with ESP states has been limited to the A phase and A_1 phase of bulk ^{3}He for which the transition temperature is only very weakly and linearly dependent on magnetic field, owing to particle-hole asymmetry. In this Letter we report the apparently contradictory observation that the transition temperature of dirty superfluid ^{3}He, thought to be an ESP state, depends quadratically on the magnetic field, and as a resolution to this problem we propose that the field dependence comes from magnetic quasiparticle scattering.

Aerogels are a class of highly dilute porous glasses composed of interconnected silica strands approximately 5 nm in diameter and separated by an average distance of 200 nm [7]. In the open geometry of aerogel the ^{3}He quasiparticle mean free path is long, $\lambda > 200$ nm, even though all points in the liquid are within $\simeq$50 nm of a scattering surface [1,2]. The strand diameter is small compared with the superfluid coherence length, $\xi_T = \hbar v_F/\pi\Delta(T) \simeq 90$ nm (at 0 bar), and much smaller than the textural bending length, $\xi_B \simeq 10$ μm. Thus, the order parameter cannot form textures to minimize strain energy near the surface of the aerogel strands. It is for this reason the gel strands act as dilute impurities embedded in a homogeneous superfluid. Such systems have been studied extensively in the context of superconductivity [8]; the superfluid phase in aerogel should consist of a spatially homogeneous order parameter, but one that is suppressed by pair-breaking scattering which occurs at the strand surfaces.

An NMR cell containing a sample of aerogel [2] was connected through a 1.6 mm diameter hole to a ^{3}He reservoir of 12 cm^3 extending to a 35 m^2 copper sinter heat exchanger in thermal contact with a $PrNi_5$ demagnetization stage. Also connected to the ^{3}He reservoir was an open-volume NMR cell [9] for the simultaneous measurement of the frequency and magnetization in bulk superfluid. Data in fields $0.385 \leq B \leq 2.047$ kOe were taken at roughly the same pressure, $18.2 \leq P \leq 19.0$ bar. All data with pure ^{3}He were taken before introducing ^{4}He. Addition of ^{4}He was accomplished by warming the cryostat and sample cell above 4.2 K and removing the sample gas to which the ^{4}He was added before the new mixture was recondensed in the cryostat. ^{4}He has a larger Van der Waals attraction than ^{3}He, and so will preferentially adsorb to the aerogel surfaces. Assuming uniform coverage of all available surface area, the ^{4}He concentration, χ_4, may be expressed in equivalent layers, $D_4 = 930\chi_4$ layers.

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Pulsed NMR measurements with 9° tipping pulses were performed [9] with data accumulation beginning 40 μs after the pulse. The absorption spectra were obtained by Fourier transformation and were numerically integrated to obtain the average nuclear spin resonance frequency, the linewidths, and the nuclear magnetization, M. Three sources of magnetization were identified in the NMR signal from the aerogel cell: liquid ^{3}He in the aerogel, solid layers of ^{3}He adsorbed to the aerogel surfaces, and a stray signal from bulk ^{3}He, presumably from the ^{3}He reservoir. The part of the spectrum from bulk ^{3}He was easily identified from its characteristic frequency shift. The ^{3}He solid layers exhibited Curie magnetization with a Curie-Weiss temperature of 0.4 mK, found previously to be independent of pressure [2]. The liquid and solid signals from the ^{3}He within the cell were in fast exchange [10] resulting in a single narrow NMR line. However, the shift of the precession frequency of the liquid spin population alone, $\langle \Delta\omega \rangle$, expressed relative to the normal fluid Larmor frequency, ω_0, can be recovered in a straightforward way [2,10]. It is likely that there also exists a dense first layer of solid ^{3}He adjacent to the aerogel which does not appear in our NMR spectrum [11]. Such a layer can be expected to be significantly dipolar broadened and to be uncoupled from the rest of the ^{3}He in the system.

As in Ref. [2], the onset of superfluidity is identified by the sudden onset of frequency shifts, $\langle \Delta\omega \rangle$, measured with respect to the normal state values. In the spin-triplet p-wave superfluid, the magnetic dipolar interaction between Cooper pairs causes a shift of the transverse resonance [12], $\omega^2 = \omega_0^2 + F(\phi, \theta)\Omega^2$, where Ω is the temperature dependent longitudinal resonance frequency and is proportional to the amplitude of the order parameter, often referred to as the energy gap. $F(\phi, \theta)$ is a temperature independent factor that depends on the amplitude of excitation in the NMR experiment through the spin tipping angle, ϕ, and on the orientation of textures of the order parameter to the external field, given by the angle θ, as may be appropriate for the particular spin pairing state. $F(\phi, \theta)$ is unknown for the aerogel superfluid phase. However, the limiting behavior at small tipping angles is qualitatively similar to the equilibrium bulk superfluid phase ^{3}He-A. In both systems, $\langle \Delta\omega \rangle$ is a maximum at $\phi = 0$ [2] and the pairing is equal spin. Thus we take the small tip angle limit of dipole-locked ^{3}He-A for the analysis presented here, $F = 1$.

Figure 1 shows the square of the longitudinal resonance, Ω^2, for three magnetic fields as a function of the reduced temperature, $T/T_c(H)$ (see Fig. 2). In our largest field Ω^2 appears to saturate at low temperature at a significantly smaller value as compared with that observed in lower fields. The suppression of the order parameter by magnetic field at low temperatures is inconsistent with the one parameter isotropic scattering model (ISM) [26]. The suppression of T_c by the presence of aerogel has a quadratic dependence on applied magnetic field, $\Delta T_c = \alpha + \beta H^2$, where at 18.7 bars, $\alpha = 440 \pm 40$ μK and $\beta = 60 \pm 12$ μK/kOe^2 (see Fig. 2). The values of T_c shown in Fig. 2 were adjusted to a common pressure of 18.7 bars using the pressure dependence measured in Ref. [2] of 51.6 μK/bar near 18.7 bars. The field dependence observed is comparable in magnitude to the quadratic field suppression of the bulk ^{3}He-A to ^{3}He-B phase transition (AB) at the same pressure.

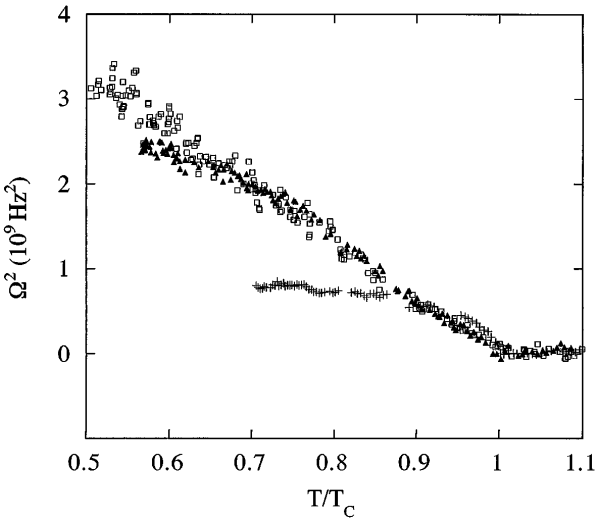

FIG. 1. The square of the longitudinal resonance frequencies in the superfluid plotted against the reduced temperatures at representative fields 0.382 kOe (squares), 1.12 kOe (triangles), and 2.18 kOe (crosses).

The mechanism for magnetic field dependence of the transition temperature in bulk ^{3}He superfluid takes two forms. The breaking of unequal-spin pairs in ^{3}He-B, $|\uparrow\downarrow\rangle$, modifies the relative free energies of ^{3}He-A and ^{3}He-B and gives rise to the first order AB transition. This effect is quadratic in field. However, the magnetization behavior in aerogel [2] indicates that the equilibrium phase of the dirty superfluid is an ESP state with no population of $|\uparrow\downarrow\rangle$ pairs. Such states will have a weak linear field dependence associated with Zeeman splitting of the Fermi surface, as is the case for the linear magnetic splitting of the bulk ^{3}He-A_1 phase ($\simeq 6$ μK/kOe) [13]. Neither of these mechanisms can explain the large quadratic magnetic field suppression of superfluidity in aerogel. Consequently we consider the role of magnetic scattering from the polarized layers of solid ^{3}He. Our hypothesis is that the field independent suppression of T_c, given by α, comes from the potential scattering of quasiparticles while the magnetic field dependent part, given by β, comes from magnetic scattering. To illustrate this in the context of the ISM, we take as a model for an elastic scattering potential, $u = v + a\vec{P} \cdot \vec{\sigma}$, where v and a are scalar amplitudes, $\vec{\sigma}$ is a quasiparticle spin, and $\vec{P}$ is the polarization of the scattering surface, with $P \propto H$. This potential naturally produces the desired H^2 dependence

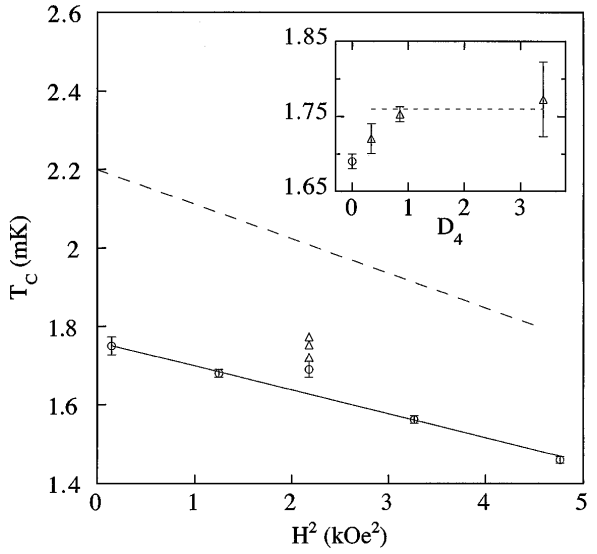

FIG. 2. The suppression of T_c with magnetic field (circles) is quadratic, and comparable in magnitude to the suppression of the B phase in bulk (dashed line). The values of T_c have been adjusted to a common pressure of 18.7 bars, as described in the text. The triangles correspond to the ^{4}He additions in the inset. Inset: T_c dependence on ^{4}He coverage in 98% aerogel. Data are taken in a field of 1.47 kOe. The dashed line at 1.76 mK is the zero field extrapolated value of T_c for pure ^{3}He.

in the ISM. The suppression of T_c is given by $k_B \Delta T_c \simeq 8\hbar/\pi\tau$ [8] where the quasiparticle scattering rate, $1/\tau$, is proportional to the square of the matrix element, $|\langle u \rangle|^2$, and hence H^2.

To test this hypothesis we removed the magnetic surface solid incrementally by replacement with nonmagnetic ^{4}He, and we monitored the process through measurement of the magnetization. At the largest ^{4}He coverage, $\sim$3.4 layers, all of the solid layer magnetization was extinguished, and we recovered the zero field value of T_c. It is likely that the aerogel surface was covered with both solid and a superfluid film of ^{4}He. We interpret this behavior as an indication that the magnetic scattering channel is shut off by the addition of ^{4}He, confirming the importance of the surface solid in the field dependence of T_c.

At the ^{4}He coverage of 0.8 layer only 20% of the ^{3}He surface solid was replaced by ^{4}He. There are several possible scenarios for placement of the ^{4}He consistent with this result. We suspect that the ^{4}He first replaces most of the solid ^{3}He in the first layer at the aerogel surface [11]. The first solid layer of ^{3}He is not expected to couple to the overlayers of ^{3}He [14]. Subsequent additions of ^{4}He replace the solid ^{3}He that is observable by NMR until all of the surface solid NMR signal is eradicated. Another possibility rests on the proposal of enhanced solubility of ^{4}He near the solid-liquid interface [15–17]. In this case the first additions of ^{4}He occupy surface states in the liquid adjacent to the solid surfaces.

The shift of T_c for the submonolayer ^{4}He coverage must be explained in the context of these two scenarios. The first solid layer is not believed to exchange couple with the rest of the ^{3}He in the system so, in the first case, a purely magnetic interaction would be required for the shift seen in T_c, either via dipolar coupling or indirectly through local disturbances of the quasiparticle Fermi surface. In the second case the ^{4}He trapped in surface states near the solid-liquid interface could interrupt surface scattering of ^{3}He quasiparticles from solid ^{3}He.

After the addition of 3.4 layers of ^{4}He, the coverage for which all surface solid ^{3}He is removed, we see dramatic changes in the superfluid behavior in aerogel. A new superfluid phase is stabilized, characterized by a decreasing magnetization with decreasing temperature as shown in Fig. 3. This is in contrast with the other lower ^{4}He coverages and all other experiments with pure ^{3}He in aerogel at different fields and pressures where the superfluid magnetization is temperature independent. The new phase must be a non-ESP phase and so in Fig. 3 we compare with the bulk B phase. We find that Ω^2, deduced from the frequency shifts in this new phase, is 60% larger than observed before and that the NMR linewidths are also much larger than at lower coverages and temperature dependent, given approximately by $1.05\langle\Delta\omega\rangle$. In our other experiments, the linewidth was approximately the same in normal and superfluid states except for a small bump just below T_c [18]. The similar magnitude of shifts and linewidth indicate textural broadening. For a random texture in the B phase we calculate that the

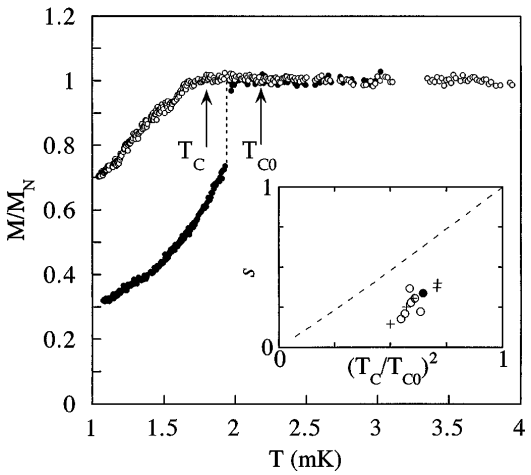

FIG. 3. Total magnetization plotted against reduced temperature in aerogel plated with 4 layers ^{4}He. The magnetization drops below the Fermi liquid value at temperatures $T < T_c$ as indicated by the arrow. For comparison the simultaneous measurement of the bulk ^{3}He magnetization is shown with the AB transition indicated as a dashed line and the bulk transition temperature, T_{c0}. The inset gives the suppression of the square of the order parameter as a function of the square of the suppression of T_c. The ISM for the unitary limit is a dashed line. Earlier frequency shift data [2] (crosses) and the present work (circles) are shown for the ESP phase. The frequency shift for the non-ESP phase (solid circle) is also shown.

linewidth should be $1.10\langle\Delta\omega\rangle$, determined as the ratio of first moment to the square root of the second moment of the spectrum. This is close to what we observe and so we infer that the new phase is qualitatively similar to a suppressed B phase, but having a random texture, under the condition of coating the aerogel surface with 3.4 layers of ^{4}He. The large values for frequency shifts also suggest a B-like phase since Ω^2 is larger for the bulk B phase compared to the A phase.

The suppressions of the order parameter,

$$S \equiv \frac{\partial\Omega^2/\partial(T/T_c)}{\partial\Omega^2_{\rm bulk}/\partial(T/T_{c0})}, \tag{1}$$

are intercompared as a function of $(T_c/T_{c0})^2$ in the inset of Fig. 3. The results from analysis of the initial slopes of the frequency shifts for data presented in Fig. 2 are shown as open circles. For the data taken at 3.4 layers of ^{4}He, we normalize the slope relative to the bulk B phase, including a textural average ($\langle F\rangle = 2/3$), and we plot this as a solid circle in the inset to Fig. 3. These are compared with the earlier work of Sprague *et al.* [2], taken at fixed magnetic field and varying pressure, shown as crosses in this inset. The remarkable agreement between these various measures of suppression of the order parameter shown in this inset gives confidence in our having identified the superfluid states correctly. In the ISM for the case of unitary scattering, S is uniquely determined by and approximately proportional to the single parameter $(T_c/T_{c0})^2$, shown as the dashed curve.

In conclusion, the suppression of the superfluid transition of pure ^{3}He in aerogel has been found to be quadratically dependent on an externally applied magnetic field which we attribute to magnetic quasiparticle scattering in the dirty superfluid. Replacement of the solid ^{3}He by nonmagnetic ^{4}He supports this picture. With enough ^{4}He preplating of the available surface area to replace all the solid ^{3}He, the stable superfluid phase switches from an ESP to a non-ESP state exhibiting the same degree of order parameter suppression.

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