Homogeneous Equal-Spin Pairing Superfluid State of ^{3}He in Aerogel

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Pulsed nuclear magnetic resonance measurements of the transverse frequency and magnetization of superfluid ^{3}He in a 98.2% porous aerogel are observed at temperatures reduced significantly below the bulk ^{3}He superfluid transition and are discussed in terms of an isotropic impurity scattering model. Magnetization measurements suggest an equal-spin pairing superfluid. For NMR tipping angles, $\phi \gtrsim 40°$, the shifts drop abruptly to zero, unlike the known dependence of either the ^{3}He-*A* or ^{3}He-*B* superfluid phases.

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^{3}He was the first unconventional BCS superfluid to be discovered. Since then it has become the paradigm for unconventional pairing in a class that now includes some heavy fermion and cuprate superconductors. One of the key properties of these superconductors is their sensitivity to impurity scattering, now a subject of considerable current interest as an indication of non-s-wave ground states. In order to better understand such phenomena it is important to study impurities in the model system of ^{3}He where the ground state is well known. Unfortunately the impurity problem has not been experimentally tractable for ^{3}He. By this we mean that it is difficult to introduce inhomogeneity into ^{3}He of size less than the superfluid coherence length [1]. Walls and surfaces can be used to confine a sample, but they both suppress and distort the order parameter, producing a heterogeneous superfluid state [2,3].

In this Letter we present our observations of a transition to superfluidity of ^{3}He in 98.2% porous aerogel which homogeneously suppresses the order parameter from its bulk values. The structure of the aerogel is small compared to the bulk superfluid coherence length and it appears that the aerogel is a dilute impurity in the ^{3}He. We have found that the transition has a sharp onset consistent with this interpretation. Our results complement the recent discovery of superfluidity in a similar sample of aerogel reported by Porto and Parpia [4].

Aerogel has a tenuous open gel structure consisting of clusters of silica connected in strands approximately 5 nm in diameter with average strand separation approximately 100 nm [5]. The cluster size is much smaller than the superfluid ^{3}He coherence length, $\xi_T = \hbar v_F/\pi\Delta(T)$, which at zero temperature decreases with increasing pressure from 90 to 20 nm at 34 bars, and diverges near T_c as $(1 - T/T_c)^{-1/2}$. The void sizes in the aerogel are smaller than the typical bending lengths that can be tolerated by the bulk superfluid order parameter. Consequently, one is led to think of the silica strands as isolated impurities and of the aerogel-helium mixture as a realization of an impurity in an unconventional BCS superfluid. It is not expected that the order parameter can form textures to accommodate the aerogel strands since textural bending lengths exceed all relevant scales in the structure. Our experiments support this basic picture, although there are a number of intriguing failures of the predictions of a simple impurity scattering model.

We used a cylindrical sample of aerogel 1 cm long and 4 mm in diameter that fit within an epoxy cell such that its axis was perpendicular to the NMR field. The specimen was connected via a 1.6 mm diameter hole to a ^{3}He reservoir (4 mm in diameter and 10 cm in length) extending to a copper sinter heat exchanger at the demagnetization stage. Also connected to the ^{3}He reservoir was an open-volume NMR cell [6] positioned 1 cm above the aerogel cell and at the NMR magnet center, which was used for simultaneous measurement of bulk behavior of the superfluid. Pulsed NMR techniques were performed [6] in a field of 1.117 kG (3.62 MHz) using small tipping angles (10°). The absorption spectra were numerically integrated to obtain the average frequency $\langle\omega\rangle$, magnetization M, and linewidth (weakly temperature dependent and ~ 250 Hz at 2 mK).

Figure 1 shows a characteristic set of $\langle\omega\rangle$ as a function of temperature. The most notable feature is the sharp onset of large positive frequency shifts at a temperature T_c. For $T < T_c$ the NMR line shape is a symmetric Lorentzian consistent with the ^{3}He in the cell being in fast exchange [7]. Under these conditions ^{3}He atoms move freely between the solid and liquid phases during a free induction decay (FID) and only a single NMR line is observed. A liquid line distinct from the solid would otherwise be expected. At the highest pressures the shifts relative to $\langle\omega\rangle$ at T_c are largest and exceed the linewidth. Therefore a simple redistribution of spectral weight within the NMR line is not sufficient to explain the observed shifts; an additional spin torque [8] must be present such as was discovered in the bulk *A* phase of ^{3}He. Together with the observation of a mass unloading reported by Porto and Parpia [4], we associate the onset of shifts in $\langle\omega\rangle$ with a sharp superfluid transition at T_c. Since NMR is a microscopic probe of the ^{3}He state, we infer that the entire sample must undergo this phase change in unison at a well defined temperature.

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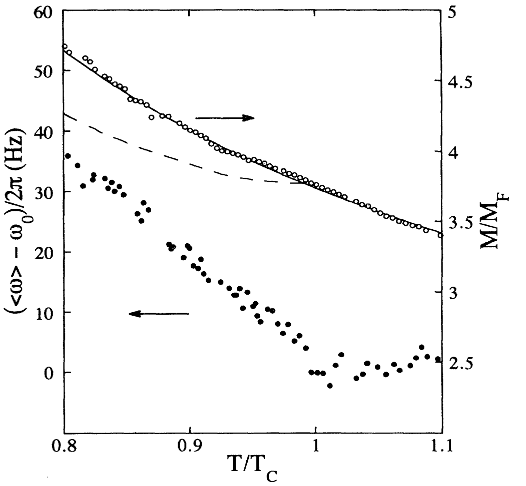

FIG. 1. Magnetization (open circles) and frequency (solid circles) against reduced temperature at 18.28 bars. The frequency shift onset defines T_c. The magnetization is normalized by its Fermi fluid value and a Curie-Weiss behavior is due to ^{3}He solid layers, as described in text. The solid curve corresponds to the liquid magnetization being constant at all temperatures, and the dashed curve indicates the magnetization expected if the ^{3}He liquid were to enter a B phase.

In other porous media such as packed silver powders [9] a bulklike order parameter is suppressed for a distance comparable to ξ_T due to quasiparticle scattering from surfaces [2]. The resulting heterogeneous superfluid state exhibits considerable rounding at the onset of the phase transition. Given that the length scale for the heterogeneity of the aerogel used here is of order, or less than, that of the coherence length itself, this medium can be expected to be in a different class. The NMR and superfluid density measurements [4] confirm this expectation.

The transition temperatures T_c are shown in Fig. 2 for pressures in the range $12.8 < P < 28.7$ bars. Also shown for reference are curves which separate the normal, ^{3}He-A, and ^{3}He-B bulk phases (in a field of 1.117 kG [10]). Since T_c crosses the AB phase boundary, the low temperature phase in aerogel is stable when in contact with both ^{3}He-A and ^{3}He-B phases outside of the cell. Below 12.8 bars no transitions were observed in the NMR experiment down to 0.9 mK. Both the amplitude of the frequency shifts and the transition temperatures are reduced markedly with decreasing pressure.

For a variety of porous media, it is known that several layers of solid ^{3}He adsorb to available surfaces and exhibit Curie-Weiss magnetization $M_s = C/(T - \theta)$, where the Curie constant C is proportional to the areal coverage of the surface solid and the ordering temperature, θ, is typically 0.5 mK. The total magnetization M is the sum of M_s and the liquid magnetization M_l. In our experiment we find that the total magnetization is dominated at low temperatures by the solid layer magnetization and at all temperatures fits $M = C/(T - \theta) + M_l$ with values of C and θ in good agreement with previous [7,11] measurements of ^{3}He in other porous materials. At all pressures M_l is temperature independent as expected for a Fermi liquid in the temperature range $T_c < T \ll T_F$, where T_F is the Fermi temperature, and $\theta = 0.4$ mK. At temperatures $T < T_c$ there is no evidence that M_l deviates from its normal state value, M_F. From these magnetization data we conclude that the superfluid phase in the aerogel must be an equal-spin pairing (ESP) state [1]. One example is the axial state which has been identified with ^{3}He-A [12].

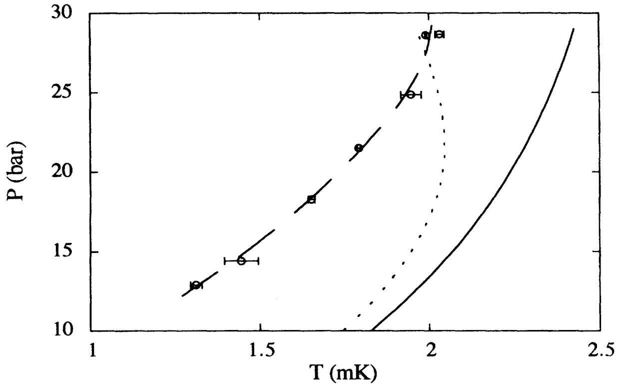

FIG. 2. Phase diagram of ^{3}He in aerogel showing pressures and temperatures of a superfluid transition in 1.117 kG field (circles). The ^{3}He is in an equal-spin pairing state to the left of this curve. The dashed curve through the circles is a guide to the eye. The solid and dotted curves are, respectively, the bulk A-phase and B-phase transition temperatures for a field of 1.117 kG taken from Ref. [10].

Figure 1 shows M/M_F in the vicinity of T_c along with the fit curve setting M_l a constant. The dashed curve shows the expected path if the ^{3}He were to enter an isotropic phase at T_c. It should be noted that the scatter evident in the data does not exceed more than 5% of M_F.

In order to determine the frequency shifts associated with the liquid alone we must account for the contribution to the NMR spectrum from the solid phase. As mentioned previously, the single NMR line indicates that ^{3}He in the adsorbed solid and liquid phases is in the fast exchange limit [7]. Since the two ^{3}He phases remain in thermal equilibrium no Zeeman energy is exchanged, and the average frequency is related to the characteristic frequency in the liquid, $\langle\omega_l\rangle$, by $\langle\omega\rangle = (\langle\omega_l\rangle M_l + \langle\omega_s\rangle M_s)/(M_l + M_s)$, where $\langle\omega_s\rangle$ is the precession frequency in the solid phase. The adsorbed ^{3}He solid layers have internuclear dipolar fields that produce frequency shifts dependent on the angle θ subtended by the surface normal and the external field, $\Delta\omega_s \sim 1 - 3\cos^2\theta$. However, all possible aerogel surface orientations are realized, so shifts from the Larmor precession frequency, ω_0, are averaged to zero. We take ω_0 to be $\langle\omega\rangle$ at T_c. Well above $T_{c,0}$ (~ 15 mK),

$\langle \omega \rangle$ differs from its value at T_c by no more than a few parts per million (ppm), but different choices of ω_0 do not alter the conclusions presented here.

In order to interpret frequency shifts attributed to the liquid we draw upon our understanding of NMR in spin-triplet p-wave superfluids. The dipolar torque causes a shift of the transverse NMR resonance [8], which can be expressed in the form $\omega^2 = \omega_0^2 + F\Omega^2$, where Ω^2 is the longitudinal resonance frequency. The factor F gives the dependence of ω^2 on the amplitude of excitation in the NMR experiment, i.e., the tip angle of the magnetization, as well as the orientation of the order parameter in the various spin pairing states. For small tip angles, as in the present discussion, $F = 0$ for the isotropic state and $F = 1$ for the axial state. In ^{3}He-A, Ω_A is proportional to the amplitude of the order parameter [13]. We use the above quadratic relation to compare the order parameter of ^{3}He in the aerogel and that of the A phase.

In Fig. 3(a) we show a selection of our data as a function of temperature compared with earlier measurements in the bulk A phase [14]. We find that aerogel data at all pressures can be collapsed onto a universal curve with a pressure dependent factor scaling Ω^2, as shown in Fig. 3(b). Furthermore, the aerogel and bulk frequency shifts also scale well together and this scaling is shown in the inset to Fig. 3. The magnitude of Ω^2 decreases with pressure, approaching zero as $P \simeq 10$ bars. This observation likely coincides with the absence of observable frequency shifts at pressures below 10 bars and is in qualitative agreement with the results of Porto and Parpia [4]. Differences in the aerogel samples may account for the quantitative differences in the phase diagram and degree of suppression of the superfluid order parameter.

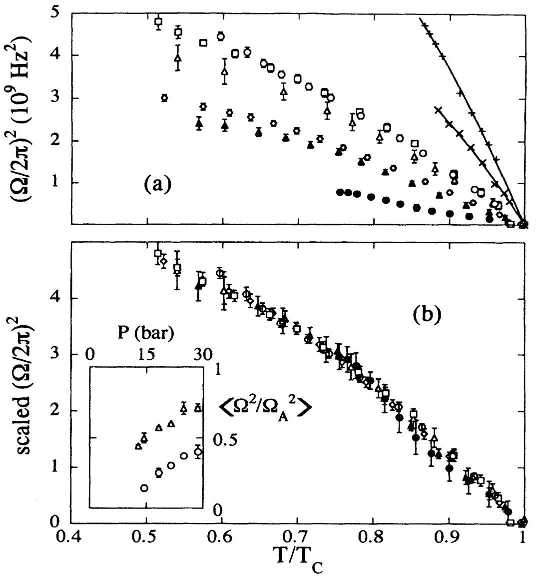

FIG. 3. (a) Longitudinal frequencies squared, Ω^2, and plotted against the reduced temperature at each pressure as described in the text for $P = 28.65$ (open circles), 28.58 (squares), 24.85 (open triangles), 21.49 (diamonds), 18.28 (solid triangles), and 14.40 bars (solid circles). Also shown are corresponding longitudinal resonance frequencies in the bulk A phase for 21.1 (+'s) and 29 bars (×'s) taken from Ref. [14]. The solid curves are a guide to the eye. (b) Ω^2 in (a) are scaled resulting in a universal function of the reduced temperature. Inset: Ω^2 scaled by the bulk A-phase value, Ω_A^2, measured at each pressure (circles) compared with that expected for each T_c calculated from the scattering model (triangles). See text for a detailed discussion.

As discussed earlier, a possible model for superfluidity in aerogel is to treat the aerogel strands as uncorrelated isotropic scatterers. It has long been known [15] that in an s-wave superconductor magnetic impurities greatly suppress T_c and the order parameter. In the case of a p-wave condensate, all scattering suppresses the condensate in a similar way. For impurities that scatter isotropically the reduction of T_c is a function of a single parameter, the scattering lifetime, τ, and is given by the Abrikosov-Gor'kov equation

$$\ln\left(\frac{T_{c,0}}{T_c}\right) = \Psi\left(\frac{1}{2} + \frac{\rho}{2}\right) - \Psi\left(\frac{1}{2}\right), \tag{1}$$

where $\rho = \hbar/2\pi\tau k_B T_c$, and Ψ is the digamma function. From our data we find the mean free path τv_F is in the range 208 to 297 nm, where v_F is the Fermi velocity. This compares well with the line-of-sight mean free path estimated from the geometry of the aerogel to be 278 nm. The magnitude of the order parameter is also suppressed in the presence of impurities, and in the Ginzburg-Landau limit and to lowest order in ρ can be shown [16] to be

$$\frac{\Delta^2(t_c)}{\Delta_A^2(t_{c,0})} \simeq \left(\frac{T_c}{T_{c,0}}\right)^2, \tag{2}$$

where we use the convention $t_\alpha = T/T_\alpha$. For the purpose of comparing our data to a suppressed axial state, we define the scaling factor $\langle \Omega^2(t_c)/\Omega_A^2(t_{c,0}) \rangle$ where the average is taken over the range of overlap between our aerogel and ^{3}He-A phase measurements taken simultaneously. The inset in Fig. 3 shows the comparison with the impurity scattering model, where we have made use of $\Omega(t) \propto \Delta(t)$ and Eq. (2). While the pressure dependence is similar, the predicted suppression of the order parameter is significantly less than that observed, suggesting that a simple isotropic scattering model is insufficient. In weak-coupling theory isotropic scattering favors the isotropic state [16]. This inconsistency may also reflect inadequacy in the model or that strong coupling effects are enhanced in aerogel as compared to the bulk.

Although an ESP state is indicated by the magnetization data and large frequency shifts are observed as in ^{3}He-A, there is no evidence presented here that unambiguously identifies the order parameter as the axial phase. One striking difference between ^{3}He-A and the ^{3}He phase in aerogel is found in the NMR tipping angle dependence of

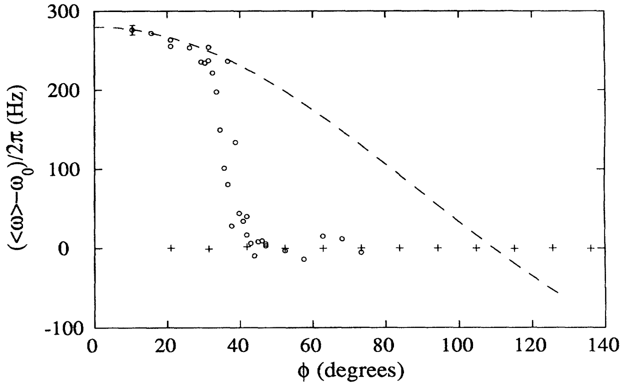

FIG. 4. Resonance frequency relative to the Larmor frequency at 21.49 bars as a function of the NMR tipping angle ϕ for $T < T_c$ (circles) and in the normal fluid (+'s). The dashed curve is proportional to the frequency of the bulk A phase showing its dependence on the tipping angle in comparison with that of the aerogel phase.

the frequency shift. Figure 4 shows the frequency shifts taken at a variety of tipping angles ϕ compared with the known dependence of ^{3}He-A; $F = (1 + 3\cos\phi)/4$. An abrupt drop to zero frequency shift occurs in the aerogel for tipping angles $\phi \geq \phi_c$, where the critical angle is nominally $40° \leq \phi_c \leq 50°$ and only depends weakly on temperature and pressure.

In conclusion, we have found suppression of T_c and the order parameter for superfluid ^{3}He in aerogel. Measurements of the magnetization indicate the order parameter to be an ESP state and the magnitude of the order parameter to be suppressed more than can be inferred from an isotropic impurity scattering model. Anomalous tip angle dependence is observed with a sudden disappearance of frequency shifts for tipping angles in excess of $\sim 40°$. The suppressed superfluid state discussed here is not the result of finite size effects associated with surface scattering of quasiparticles, but rather the ^{3}He-aerogel system should be thought of as a homogeneous superfluid with the aerogel as an impurity.

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[13] In ^{3}He-A, $\Omega_A^2 = (6/5)\gamma^2\lambda_D N_F \Delta^2(T)/\chi$, where Ω_A is the longitudinal resonance frequency, γ is the ^{3}He gyromagnetic ratio, λ_D is the dipolar coupling strength, N_F is the density of states at the Fermi surface, χ is the magnetic susceptibility, and Δ is the maximum gap at the Fermi surface. Only the numerical prefactor changes between the other bulk spin pairing states.

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