

Observation of a Superfluid He-3 *A-B* Phase Transition in Silica Aerogel

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New NMR studies of ^3He in high-porosity aerogel reveal a phase transition from an *A*-like to a *B*-like phase on cooling. The evidence includes frequency shift and magnetic susceptibility data, and similar behavior is found in two quite different aerogel samples. The *A*-like phase is stable only very near to T_c but can be supercooled to below $0.8T_c$. This behavior has been seen clearly at 32- and 24-bar pressures, and the presence of negative frequency shifts suggests that an *A*-like phase exists near T_c at pressures as low as 12 bars in a magnetic field of 28.4 mT.

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While all superconductors suffer depairing due to magnetic impurities, only in unconventional BCS states do nonmagnetic isolated impurities produce depairing and hence a suppression of both the energy gap and T_c . Superfluid ^3He is a desirable system in which to study such depairing by impurities: The net (nuclear) spin associated with the Cooper pairs produces complex NMR spin dynamics which are well characterized and able to probe the ordered states on a microscopic scale, and the absence of an underlying lattice allows the ordered states to better respond to the presence of the impurities. While no known substance is soluble in liquid ^3He at very low temperatures, recent work [1–10] has shown that high porosity silica aerogel may act as a pseudorandom impurity scatterer, since the diameter of the aerogel strands (≈ 5 nm) is small compared to the zero temperature coherence length, which varies from 80 to 15 nm for 0–34 bar sample pressures. Most experiments to date have utilized 98% porosity aerogel, although some recent work has involved higher porosities [3–6]. In this Letter, we discuss recent cw-NMR experiments on ^3He in two very different 98% porosity aerogel samples. In both samples we find strong evidence for the stability of an *A*-like phase near T_c and a *B*-like phase at lower temperatures. The first order transition between the two phases, similar to the *A-B* transition in the bulk, exhibits substantial supercooling.

The superfluid phases of ^3He are *p*-wave BCS states with both $S = 1$ and $l = 1$ [11]. In the *A* phase the orbital angular momentum of all the Cooper pairs is oriented in the same direction locally, \mathbf{l} , and the nuclear spins within a pair are parallel. This equal spin pairing results in a temperature independent susceptibility nearly equal to that of the normal phase. This state has nodes in the gap along \mathbf{l} , and depairing is minimized when \mathbf{l} is normal to surfaces. The nuclear dipole energy is minimized when the direction along which the spin projection is zero, \mathbf{d} , is parallel to \mathbf{l} . The Zeeman energy is minimized when \mathbf{d} is perpendicular to an applied magnetic field. In the *A* phase, the cw unsaturated NMR frequency

is given by $\omega^2(T) = \omega_{\text{Larmor}}^2 + \cos(2\phi)\Omega_{\text{long}}^2(T)$. Here $\omega_{\text{Larmor}} = \gamma H$ and Ω_{long} is a measure of the dipole-dipole energy, proportional to the energy gap, and ϕ is the angle between \mathbf{d} and \mathbf{l} . The *B* phase Cooper pairs have $S = 1$ and $l = 1$, but $J = 0$. The spin coordinate system must be rotated about an arbitrary axis \mathbf{n} with respect to the orbital coordinate system by an angle of $\cos^{-1}(-\frac{1}{4})$ to minimize the nuclear dipole-dipole energy. The *B* phase Cooper pairs include all three symmetric spin states, and hence that phase has a magnetic susceptibility less than that of the *A* phase and normal liquid. The rotation axis \mathbf{n} is oriented weakly by both magnetic fields and surfaces, and in the bulk, \mathbf{n} is parallel to \mathbf{B} . In the *B* phase one finds that $\omega^2(T) = \omega_{\text{Larmor}}^2 + \sin^2(\theta)\Omega_{\text{long}}^2(T)$. Here θ is the angle between \mathbf{n} and \mathbf{B} , and Ω_{long} in the *B* phase is substantially larger than that in the *A* phase. Note that only the *A* phase can have an NMR frequency less than ω_{Larmor} .

The reduction of T_c for superfluid ^3He in aerogel has been found to roughly follow the theory for impure superconductors first discussed by Abrikosov and Gor'kov [12], as expected. In the framework of this theory, the only effect aerogel should have on the superfluidity should be a suppression of the energy gap $\Delta(T)$. Recent theoretical work [13] suggests, however, that in the limit of weak coupling, the *B* phase is favored over the *A* phase by increasing the impurity scattering, although no new phases should appear.

Of the two aerogel samples employed in our studies, the first was grown by rapid supercritical extraction, a process developed by Poco *et al.* [14]. In this process, one proceeds with supercritical extraction soon after the precipitation of silica has occurred, with gelation taking place rapidly in the heated environment. In the second sample, the extraction process was not undertaken until after gelation had gone far enough that the sample was no longer a fluid. The aerogel was inserted into an NMR tailpiece machined from Stycast 1266 epoxy resin [15]. NMR coils above and below the aerogel probed the bulk superfluid, and another coil probed the ^3He inside the aerogel without sensing the bulk liquid (a small bulklike signal less than

1% of the total could be seen in some measurements). A powdered Pt NMR thermometer was also included below the aerogel. The tailpiece was attached to a pressed silver heat exchanger attached to a copper nuclear demagnetization stage. The majority of the measurements were performed in a static magnetic field of 28.4 mT and at a sample pressure of 32 bars.

Our first and second aerogel samples had superfluid transition temperatures at 32 bars of 2.25 and 2.05 mK, respectively, while at this pressure $T_c^{\text{bulk}} = 2.46$ mK. This variation is typical of what other investigators have observed. Because of the large surface area of aerogel (typically about 750 m²/gr for 98.2% porosity), the magnetization of the second localized layer of ³He adsorbed on the aerogel strands interferes with measurements of the liquid NMR properties. By adding small amounts of ⁴He to the cell, we could replace the localized ³He with spinless ⁴He. In studies with the first aerogel sample, Barker *et al.* [10] found that the average frequency shift in the liquid at low temperatures did not in any way depend upon the extent of this replacement, up to two monolayers of ⁴He. We thus focus here on new measurements made in the second aerogel, in which the localized ³He has been nearly completely replaced with ⁴He. We note that Sprague *et al.* [7] found, by contrast, that the presence of 3.4 layers of ⁴He stabilized the *B* phase over the *A* phase, which they had previously found to be stable over the entire temperature range for pure ³He samples.

A sample of NMR spectra obtained on cooling is shown in Fig. 1(a). Below $T_c = 2.05$ mK there is seen a narrowing and a shift in the NMR line. We use this shift to establish T_c . As T decreases, the NMR line becomes quite broad, with much of the signal shifted below ω_{Larmor} . Then, near 1.5 mK, the shape of the line changes dramatically. The spectral weight below ω_{Larmor} disappears, and a long high frequency tail appears. This new line shape is characteristic of the *B* phase in a confined geometry, such as would be defined by the cell walls containing the aerogel. Such spectra have been reported for ³He in aerogel by Alles *et al.* [4]. A simultaneous drop in the magnetization is also observed near 1.5 mK. Figure 1(b) shows a sample of the spectra upon warming. We never saw a portion of the NMR line shifted below ω_{Larmor} upon warming, and $M(T)$ rose continuously to its normal state value as the sample approached T_c . Similar hysteretic behavior was also observed in studies with the first aerogel sample [10].

We determined $M(T)$ by integrating the area under the NMR lines and used the first moment of the NMR lines to find the average frequency. Figure 2(a) shows $M(T)$ and the average frequency shift upon cooling. Just below 2 mK we see a small negative frequency shift develop while the magnetization remains unchanged. Near 1.6 mK we see a sudden drop in $M(T)$ and a simultaneous large increase in the average NMR frequency. This combination is very similar to what is found in mildly confined ³He at the *A* to *B* phase transition. By contrast, no abrupt changes in

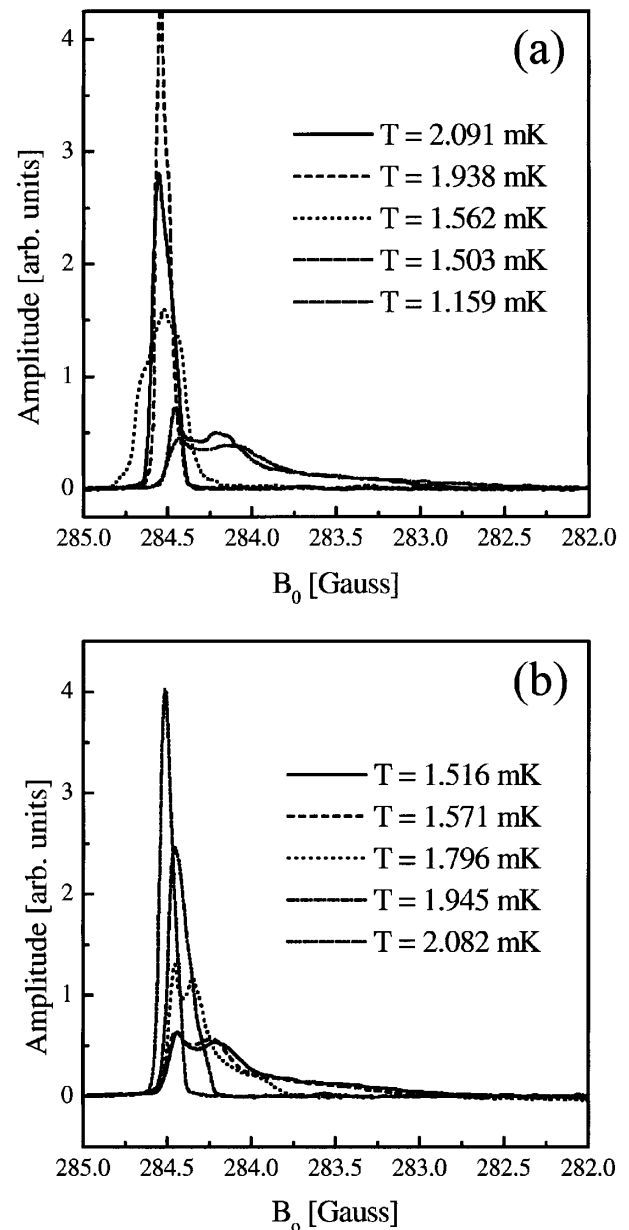


FIG. 1. cw NMR line shapes of ³He in aerogel at 32 bars on cooling (a) and on warming (b). Note the narrowing of the line shape just below $T_c^a = 2.05$ mK. Also note the change between 1.56 and 1.50 mK on cooling. In all cases, the localized layers of ³He on the aerogel surfaces have been replaced with ⁴He.

$M(T)$ or the average frequency shift were observed upon warming, as shown in Fig. 2(b). This disparity is consistent with a supercooled high temperature phase upon cooling which is actually stable only very close to T_c .

To make a comparison with the bulk, we use the fact that $\Delta^2(T) = C\Omega_{\text{long}}^2(T) \cdot \chi$, where C is a combination of temperature independent constants [16] and χ is the magnetic susceptibility. As stated earlier, the measured resonant frequencies in our experiment depend upon the orientation of the \mathbf{l} and \mathbf{n} anisotropy axes in the *A* and the *B* phase, respectively. Particularly in the *B* phase, one

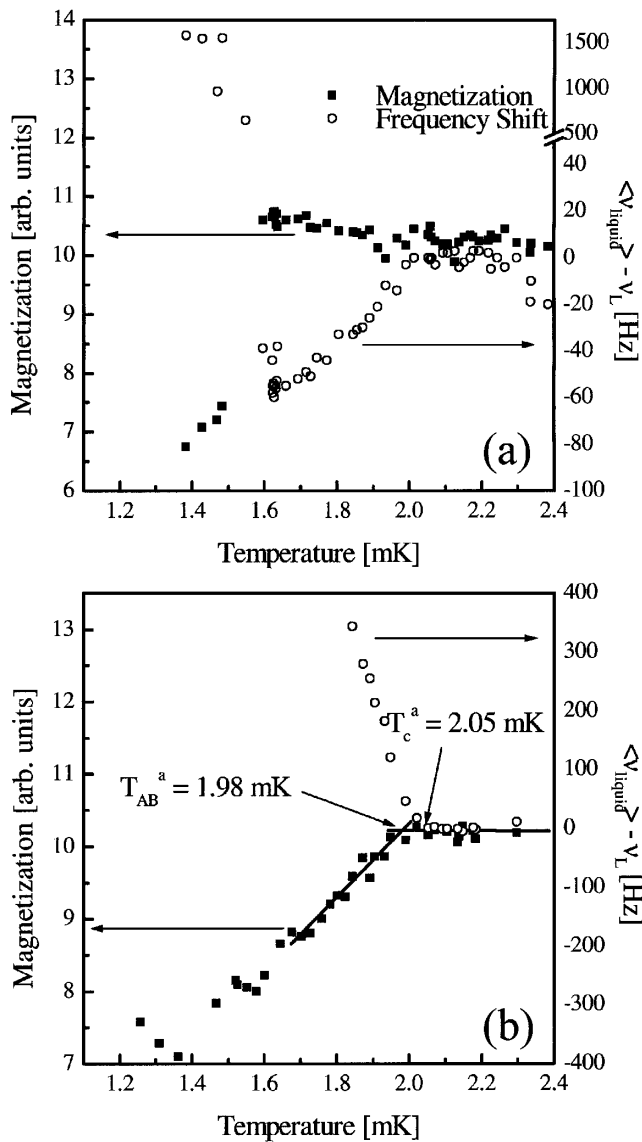


FIG. 2. Magnetization and frequency shift on cooling (a) and on warming (b) with two layers ^4He on the surface of the aerogel. The pressure is 32 bars. The large difference between cooling and warming is due to supercooling and is evidence that the phase transition is first order. Note the coexistence of constant magnetization with nonzero frequency shifts between ~ 1.98 and 2 mK on warming.

expects a broad distribution in orientation, and only when $\theta = 90^\circ$ is the frequency shift indicative of $\Omega_{\text{long}}^2(T)$ through the equation above. It is not clear that \mathbf{n} ever reached this extreme orientation in our cell, but experiments in less constricted geometries often find this to be the case, at least at low temperatures. We thus can estimate the ratio of the energy gap in our low temperature phase by assuming that $\theta = 90^\circ$ for the highest frequency shift we measure in the low temperature phase. At $T/T_c = 0.65$, this gives us a gap which is $0.53\Delta_{\text{bulk}}$. Note that we must also use $M(T)$ measured in the aerogel, which shows a drop of only about 45% as large as that measured in the

bulk and a different temperature dependence from the bulk B phase.

We cannot determine the gap in the A -like phase upon cooling because the broad nature of the line and the negative average frequency shift suggest a broad distribution of \mathbf{l} orientations. While this may result from a myriad of local “surfaces” in the aerogel which pin \mathbf{l} , the fact that we see no such broadening upon warming into the A -like phase suggests that this textural distribution may result from a quench condensation upon cooling, with different parts of the sample undergoing the transition independently. If this were the case, we should be able to estimate the A phase energy gap from the average frequency shift seen in the narrow region near T_c upon warming. To enhance this region, we applied a magnetic field of 110 mT, which suppressed T_{AB} . Comparing the slope of $\Omega_{\text{long}}^2(T)$ determined from the average frequency shift near T_c in the A phase to that found by Ahonen *et al.* [17] at the same pressure for bulk samples, we find a gap of about $0.5\Delta_{\text{bulk}}$. Thus the two estimates of the gap in the different phases agree, suggesting that the high temperature phase is most likely indeed the A phase, and not another equal spin pairing state.

Our best estimate of the equilibrium transition temperature between the two phases at 28.4 mT is 1.98 mK or $0.97T_c$. By contrast, upon cooling, the transition occurs near $0.76T_c$. This is an enormous degree of supercooling, compared to bulk behavior when T_{AB} approaches T_c . The metastable A -like phase in the aerogel remains upon cooling even after the B phase nucleates outside the aerogel. These observations all suggest that the A - B interface is strongly pinned by the aerogel.

We have made measurements at various pressures from 32 to 12 bars and find evidence for small negative frequency shifts upon cooling through T_c in the aerogel at all pressures. As low as 24 bars we find clear evidence for a phase transition upon cooling similar to what has been described above. While the first order phase transition and the extreme broadening upon cooling through T_c have never before been observed, they are features which have been seen in two very different aerogel samples and over a rather broad range in sample pressure and magnetic field. They add significantly to our knowledge of the response of a p -wave BCS state to quasirandom impurities.

The most striking feature of these new observations is unquestionably the strong evidence for an A - B phase transition upon cooling, combined with evidence for a small region of A phase stability upon warming. These observations show not only that scattering from the aerogel tends to stabilize the quasi-isotropic B phase, but they allow us to determine by how much. We propose that later experiments need to be carried out in lower density aerogel samples, so that the shift in $1 - T_{AB}/T_c$ from the bulk will be smaller, perhaps only a factor of 2.

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- [1] J. V. Porto and J. M. Parpia, Phys. Rev. Lett. **74**, 4667 (1995).
- [2] J. V. Porto and J. M. Parpia, J. Low Temp. Phys. **101**, 397 (1995).
- [3] H. Alles *et al.*, Physica (Amsterdam) **225B**, 1 (1998).
- [4] H. Alles *et al.*, Phys. Rev. Lett. **83**, 1367 (1999).
- [5] A. Matsubara *et al.*, Physica (Amsterdam) **284B**, 301 (2000).
- [6] G. Lawes, S. C. J. Kingsley, N. Mulders, and J. M. Parpia, Phys. Rev. Lett. **84**, 4148 (2000).
- [7] D. T. Sprague *et al.*, Phys. Rev. Lett. **75**, 661 (1995).
- [8] D. T. Sprague *et al.*, J. Low Temp. Phys. **101**, 185 (1995).
- [9] D. T. Sprague *et al.*, Phys. Rev. Lett. **77**, 4568 (1996).
- [10] B. I. Barker *et al.*, J. Low Temp. Phys. **113**, 635 (1998).
- [11] A. J. Leggett, Rev. Mod. Phys. **47**, 331 (1975).
- [12] A. A. Abrikosov and L. P. Gor'kov, Zh. Eksp. Teor. Fiz. **39**, 1781 (1960); Sov. Phys. JETP **12**, 1243 (1961).
- [13] E. V. Thuneberg, S. K. Yip, M. Fogelström, and J. A. Sauls, Phys. Rev. Lett. **80**, 2861 (1998).
- [14] J. F. Poco, P. R. Coronado, R. W. Pekala, and L. W. Hrubesh, Mater. Res. Soc. Symp. Proc. **431**, 297 (1996).
This paper discusses the techniques used to grow our samples. The second sample was grown using the traditional supercritical extraction technique.
- [15] *Emerson-Cummings*, Billerica, MA 01821.
- [16] A. J. Leggett, Ann. Phys. (N.Y.) **85**, 11 (1974).
- [17] A. I. Ahonen, M. Krusius, and M. A. Paalanen, J. Low Temp. Phys. **25**, 421 (1976).