NMR Studies of Superfluid ^{3}He in Low Density Silica Aerogels

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Abstract

Continuous wave NMR studies of superfluid ^{3}He in 99.4%, 98.7% and 98.3% porosity silica aerogels show similar phase diagrams, with an equal spin pairing (ESP) A-like state stable near $T_{c,a}$, and a B-like phase stable at lower temperatures. On cooling through $T_{c,a}$, the sample magnetization and frequency shift show substantial supercooling of the A-like phase, with discontinuous changes to B-like behavior seen at about $0.64T_{c,b}$ (bulk) for all samples. On warming, both the frequency shift and magnetization change *continuously* as the sample enters an ESP state, suggesting that this phase transition must be second order, with the components of the order parameter varying smoothly across the transition.

Key words: superfluid ^{3}He; aerogel; unconventional BCS states; impurity depairing

1. Introduction

The response of the superfluid ^{3}He order parameter to the presence of a low density silica aerogel has been shown to be a valuable tool for understanding how unconventional BCS states ($l > 0$ for the Cooper pairs) respond to scattering sites which are small compared to the zero temperature coherence length, ξ_0. It is particularly useful in this regard because of the strong pressure dependence to ξ_0, and the existence of two very different condensate wave functions, stable over different temperature regions. Near melting pressure, the difference in the free energies of these two states is never more than about 1 erg/cm^3, only about 1% of the condensate free energy [1]. By studying how the equilibrium transition between these two states depends upon the density of the aerogel containing the superfluid, we learn how condensates of different *symmetry* respond to scattering sites.

The response of the superfluid ^{3}He order parameter to the presence of a low density silica aerogel has been studied using several different probes. Torsional oscillator studies [2–4] show substantial supressions in both T_c and ρ_s, which increase as the sample pressure is lowered, due to the progressive increase in ξ_0. These measurements do not seem to be sensitive to the specific phase (A vs. B for example) present in the cell, however, as would be expected to be the case if the A phase had its macroscopic angular momentum vector, $\mathbf{l}$, randomly pinned by the presence of the aerogel. NMR studies give additional information, by allowing one to measure the distribution of local NMR frequencies in the superfluid as well as its magnetization within the aerogel. The first studies of this sort, all using nominally 98.3% porosity aerogel, were pulsed NMR studies at Northwestern University [5–7]. In those studies, it was concluded that only one phase was seen in the aerogel, and that phase was A-like provided the aerogel strands were covered with localized ^{3}He atoms, and B-like if one replaced the localized ^{3}He atoms with 3.4 monolayers of ^{4}He. Later, continuous wave NMR studies at Stanford [8] found that the average NMR fre-

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quency shift in the liquid did not depend upon the presence of localized ^{3}He atoms on the aerogel, and hence the stable phase could not depend upon the isotopic identity of the localized atoms covering the aerogel. This work also reported a sharp change in the distribution of NMR frequencies within the sample from a very broad spectrum showing both positive and negative frequency shifts to one with only positive frequency shifts at a sharp transition near $0.75T_{\rm c,a}$ ($T_{\rm c,a}$ refers to $T_{\rm c}$ in the aerogel). This change was not interpreted as a transition between phases, but rather a sudden change in the texture of the $\mathbf{l}$-vector in an A-like phase. It was John Hook's group at Lancaster University that first suggested that the stable phase at low temperatures for a pure ^{3}He sample was B-like [9], at lower pressures and very low magnetic fields.

In a second experiment using a different 98.3% porosity aerogel sample, Barker *et al.* [10] measured both the average NMR frequency of their sample and its magnetization for a sample in which the localized ^{3}He atoms were replaced by just two layers of ^{4}He atoms. In this experiment, it was found that the sudden change in the NMR spectrum of the sample on cooling at about $0.75T_{\rm c,a}$ was accompanied by a sudden drop in sample magnetization. This showed clearly and for the first time the presence of a *first order phase transition* on cooling from an equal spin pairing (ESP) A-like phase to a phase with a reduced magnetization and a long-range texture, consistent with the B-phase identification made by Alles *et al.* [9], but now near the melting pressure. In addition, on warming slowly toward $T_{\rm c}$ from below the A to B phase transition on cooling, Barker *et al.* [10] found evidence suggestive of an ESP state stable only within $0.98T_{\rm c,a}$ in a field of 28.4 mT. This region of stability increased down to about $0.97T_{\rm c,a}$ when the magnetic field was increased from 28.4 mT to 100 mT.

2. Results

In the new experiments reported here, we have extended the NMR studies carried out by Barker *et al.* to lower density aerogel samples of 98.7% and 99.4% porosity. Our samples have all been grown by Poco and Hrubesh at Lawrence Livermore National Laboratories. At all aerogel densities, we observe behavior similar to that seen in the 98.3% porosity aerogel, although the values of $T_{\rm c,a}/T_{\rm c,b}$ and $T_{\rm AB,a}/T_{\rm c,a}$ depend monotonically on the aerogel density. Here $T_{\rm c,b}$ refers to $T_{\rm c}$ in the bulk, and $T_{\rm AB,a}$ refers to the A to B transition temperature for ^{3}He within the aerogel. So far, these new data have only been taken at a pressure of 34 bars, somewhat higher than the 32 bar pressure used for the high pressure studies by Barker *et al.* [10]. In addition,

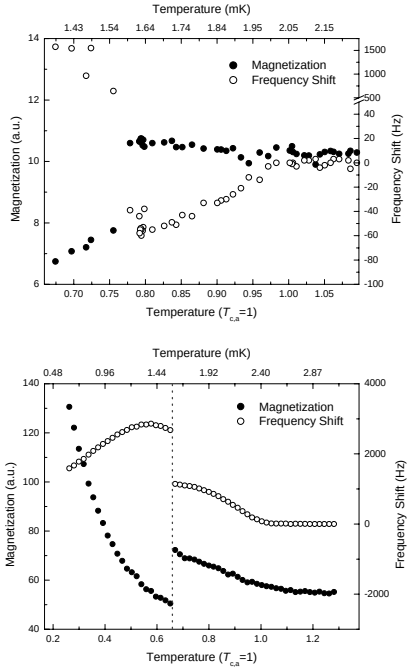

Fig. 1. *Top*: Magnetization and average frequency shift upon cooling for ^{3}He in 98.3% porosity silica aerogel with p = 32 bars, B = 28.4 mT, and two layers of ^{4}He on aerogel surfaces. ESP on cooling shows negative frequency shift. Note the sharp transition at 1.6 mK. *Bottom*: The same, but for pure ^{3}He in 99.4% porosity aerogel with p = 34 bars and B = 28.4 mT. At low temperatures, M is dominated by solid, which causes a decrease in the frequency shift.

the new data so far have all been taken only with pure ^{3}He in the sample cell, and thus we must subtract the magnetization of the localized ^{3}He atoms on the aerogel strands to get that due only to the liquid. This subtraction is not as difficult for the lower density aerogel samples, as they have less surface area and the value of $T_{\rm c,a}$ is higher in these samples, both making the magnetization of the localized spins less at $T_{\rm c,a}$. The magnetization due to the localized spins amounts to 183%, 131%, and 64% of the liquid magnetization at $T_{\rm c,b}$ for the 98.3%, 98.7%, and 99.4% porosity aerogel samples respectively. Well below $T_{\rm c,a}$, we do not consider our liquid magnetization values very accurate, although all the features we need to understand can be seen quite clearly in the present data.

Fig. 1 shows typical behavior of the magnetization and average frequency shift in the liquid upon cooling for the earlier 98.3% porosity aerogel data by Barker *et al.* [10] and the new 99.4% porosity aerogel data, uncorrected for the presence of localized ^{3}He atoms on the aerogel in the new data. In *all* aerogel samples, a sharp change in both the average NMR frequency shift $(\nu - \nu_{\rm Larmor})$ and M, the magnetization of the liquid, is seen at a temperature of about 0.63 to 0.64 of $T_{\rm c,b}$. We

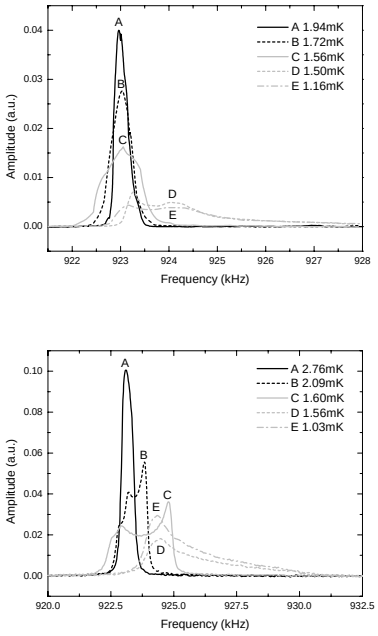

Fig. 2. *Top*: NMR spectra for ^{3}He in 98.3% aerogel on cooling. Sample has two layers of ^{4}He on aerogel surfaces. NMR lines broaden just below T_c almost symmetrically about the Larmor frequency. Below 1.6 mK, the line structure changes to one with a long high frequency tail, as in a B-like state in a confined geometry. *Bottom*: NMR spectra for pure ^{3}He in 99.4% aerogel on cooling. Behavior shown is nearly identical to that seen in the denser aerogel with ^{4}He films on the aerogel surfaces.

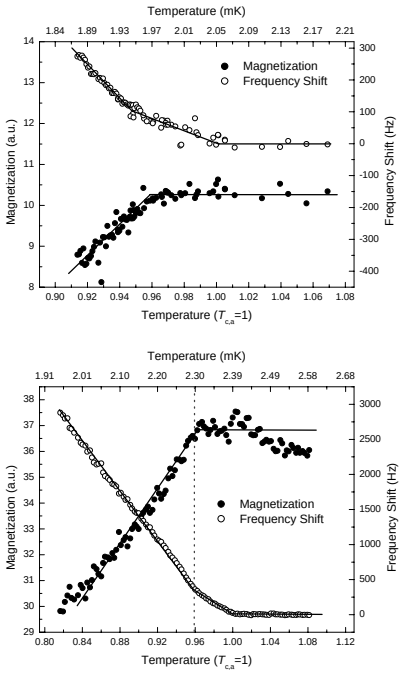

Fig. 3. *Top*: Magnetization and frequency shift data for ^{3}He in 98.3% porosity aerogel on warming, with p = 32 bars, B = 100 mT, and 2 layers of ^{4}He on aerogel surfaces. Magnetization reaches normal value before frequency shift reaches zero, showing existence of ESP state near T_c. *Bottom*: Similar behavior seen in pure ^{3}He within 99.4% porosity aerogel, after correcting for solid. The change in $d\nu/dT$ at $0.965T_{c,a}$ is clearly coincident with the change in dM/dT. For these data, the RF NMR field was a bit high, and one sees effects of saturation above $T_{c,a}$.

conclude that this represents a first order phase transition in the aerogel-born liquid. From the fact that the magnetization of the liquid is equal to the normal state value above this transition temperature and below the normal state value below this transition, we conclude that this must be a first order transition between an ESP state and a B-like state. The temperature of this transition is fairly reproducible for a given aerogel sample, and for the 99.4% porosity sample, the transition of the ^{3}He in the aerogel is *simultaneous* with the bulk A-B phase transition, suggesting that the transition in the aerogel is indeed the A-B phase transition. For the other two samples, the A-B transition in the bulk occurs at a higher temperature than the transition seen in the aerogel samples. This suggests that density variations in the aerogel represent effective pinning sites for the A-B interface. This seems plausible, given the width of the interface [1] and the length scale of the largest density variations in the aerogel [3].

The NMR spectra of the ESP state seen on cooling do not resemble that of bulk A phase for any of our samples, but are similar to each other, whether or not the aerogel surfaces have been coated with two monolayers of ^{4}He. The NMR line broadens continuously upon cooling, with portions of the line shifted both above and below the Larmor frequency. The average frequency shift in the ESP state upon cooling can be either positive or negative, and the value depends upon the orientation of the magnetic field, which we can rotate about the axis of our cylindrical samples. This seems to suggest that the **l**-vector in the ESP state is pinned by density variations in the aerogel, as was suggested above. Below the first order transition, the NMR lineshape is reminiscent of that seen in B-phase contained within a cylindrical tube [11]. Fig. 2 shows a typical series of NMR lineshapes upon cooling for 98.3% and 99.4% porosity aerogel samples. The 98.3% data were taken with two monolayers of ^{4}He coating the aerogel surfaces, while the 99.4% data were from a pure ^{3}He sample. Note that the lineshapes at the same value of $T/T_{c,a}$ are very similar for the two sets of curves. This suggests that the states involved are also similar, both above and below the first order transition.

As Barker *et al.* found [10], the B-like state produced by the first order phase transition remains stable as the sample is warmed almost to $T_{c,a}$. In Fig. 3 are shown

Porosity	99.4%	98.7%	98.3%
$T_{c,b}$	2.49mK	2.49mK	2.46mK
$T_{c,a,w}/T_{c,b}(\Delta\nu)$	0.962	0.942	0.833
$T_{AB,a,w}/T_{c,b}(\Delta\nu)$	0.922	0.911	
$T_{AB,a,w}/T_{c,b}(\Delta M)$	0.924	0.918	0.813
$T_{AB,a,c}/T_{c,b}$	0.636	0.636	0.637
$T_{AB,b,c}/T_{c,b}$	0.636	0.658	
$T_{AB,a,w}/T_{c,a}$	0.958	0.967	0.976

Fig. 4. Transition temperatures for the three aerogel samples.

the magnetization and average frequency shift vs. temperature in the 98.3% and 99.4% porosity aerogel samples upon warming. For clarity, the data shown for the 98.3% porosity sample were taken in a field of 100 mT, while the 99.4% data were obtained in a field of 28.4 mT. Notice that in both cases the liquid magnetization upon warming reaches the normal state value below the temperature at which the frequency shift goes to zero. This suggests that very near $T_{c,a}$, the superfluid is in an ESP state, not a B-like state. Similar behavior is also found in the 98.7% porosity aerogel data. Fig. 4 shows the values of $T_{c,a}$ (T_c in the aerogel), $T_{AB,a,c}$ (T_{AB} in the aerogel upon cooling), and $T_{AB,a,w}$ (T_{AB} in the aerogel upon warming) for the three aerogel samples at 28.4 mT. The 98.3% porosity data were taken at p = 32 bars, and the other two sets of data were taken at p = 34 bars. Notice the gradual variation of all quantities with aerogel porosity. It is perhaps a coincidence, but we find that when expressed as a fraction of the bulk T_c, the values of $T_{AB,a,c}$ are almost identical for the three samples.

3. Conclusions

The presence of the aerogel has a much larger effect on the equilibrium value of T_{AB} (the equilibrium value as measured upon warming) than it has on T_c. Even in the highest porosity aerogel, T_{AB} is shifted from $0.8T_c$ in the bulk phase to $0.96T_c$ in the aerogel, while T_c itself is shifted by only $0.04T_c$. From this we conclude that the presence of the aerogel strongly stabilizes the B-like phase with its isotropic energy gap.

Neither the superfluid frequency shift nor its magnetization show discontinuities at $T_{AB,a,w}$ in any of the aerogel samples. The magnetization gradually rises to the normal state value with a nearly constant slope while the frequency shift is still non-zero. There is a change in the rate of change of the frequency shift with temperature at $T_{AB,a,w}$ but no discontinuity in the frequency shift itself. The observed behavior would be very different if the stable phase were B-like up to $T_{c,a}$ but the transition temperature varied throughout the sample due to density variations in the aerogel, or if there were a thermal gradient across the aerogel sample. In both these cases, one would observe a decrease in the rate the magnetization rose with temperature at the point one first reached $T_{c,a}$ somewhere inside the aerogel. However, the magnetization would remain below the normal state value until the average frequency shift reached zero. In addition, we see no discontinuity in ΔM and $\Delta\nu$ at $T_{AB,a,w}$ for three very different aerogel densities, and the magnetization and frequency shifts appear to vary *linearly* with temperature on both sides of this apparent transition. From this we conclude that the transition we observe upon warming must be second order, not first order. This seems quite remarkable.

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