Modification of the Superfluid ^{3}He Phase Diagram by Impurity Scattering

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The effect of impurity scattering on the phase diagram of pure superfluid ^{3}He has been investigated by acoustic techniques near the bulk polycritical pressure. Impurities were introduced with a 98% porous silica aerogel. In zero applied field, the equilibrium phase is a B phase, consistent with predictions from isotropic scattering. The superfluid transition in a magnetic field at 25 bars is from normal to A phase and is independent of the magnetic field up to 2.9 kG. A first-order phase transition between A and B superfluids is observed with surprisingly strong supercooling in both zero and nonzero applied field.

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It was recently discovered that ^{3}He confined in ultralight aerogels provides a model system for the study of the effects of impurity scattering on an unconventional superfluid [1–3]. The silica aerogels typically have a porosity near 98%, as in the work we report here. The aerogel is formed by a network of strands of approximately 3 nm in diameter producing a mean-free path for quasiparticle scattering of approximately 200–300 nm [4,5]. Since the mean-free path is much larger than the coherence length in bulk ^{3}He, superfluidity is not completely destroyed. The effect of impurity scattering is to inhibit phase coherence of the Cooper pairs and reduce the superfluid transition temperature. It is also possible that impurity scattering can stabilize superfluid states not observed in the clean limit. In fact, theoretical models [6] predict that in zero magnetic field, isotropic scattering can stabilize the isotropic state (B phase) over the axial state (A phase), possibly overcoming strong coupling effects in the pairing interaction that are responsible for the existence of the bulk superfluid A phase at pressures above 21 bars. Today we have an excellent understanding of the bulk ^{3}He superfluid phase diagram. In contrast to this situation, a characterization of the "dirty" superfluid state is controversial, and the field dependence of the phase diagram of pure ^{3}He in aerogel is largely unknown. A fingerprint of the various possible triplet spin states of a superfluid can be determined from measurement of their stability in a magnetic field. In this Letter, we report acoustic measurements as a function of magnetic field that determine phase transitions from normal fluid to A phase and A to B phase clarifying the nature of the aerogel-superfluid state at pressures near 21 bars.

Superfluidity of ^{3}He in aerogel was first observed by torsional oscillator measurements [1] that probe the superfluid density, and pulsed nuclear magnetic resonance (NMR) [2,3] from which information on spin susceptibility and pairing amplitude were obtained. From the latter it was inferred that an equal spin-pairing state was being observed. Since then additional experiments have been performed including cw-NMR [7–9], torsional oscillator work [7,10,11], and acoustics [4,12,13]. There is general agreement on the extent of the suppression of the superfluid transition temperature by impurity scattering from the aerogel and that this suppression is more pronounced at low pressure. A critical pressure near 6 bars was found [10] for a 98% porous aerogel. Theoretical investigations have provided a reasonable understanding of the role of impurity scattering in aerogel [6,14–16].

As is now the custom, the terms A and B phases in aerogel are used in analogy to bulk ^{3}He, although in reality phase identification has been limited to magnetic measurements which distinguish equal spin-pairing (such as the A phase) and nonequal spin-pairing states (such as the B phase). NMR probes the spin state of the p-wave Cooper pairs from both frequency shifts of the resonance and from the magnetization. However, the magnetization experiments for pure ^{3}He in aerogel are complicated by the large contribution from a ^{3}He solid layer that forms on the aerogel strands. The first NMR work in a magnetic field of 1.2 kG suggested an A-like superfluid [2]. Subsequent NMR measurements of Alles *et al.* [7] at very low fields, near 50 G, suggested a B-like phase based on a textural analysis of the NMR line shape. Further NMR measurements by Barker *et al.* [8] at 284 G, with ^{4}He preplating, showed a transition between the two phases and a narrow region of equilibrium for an A-like superfluid at high pressure, 32 bars. They reported that the stable region for the A superfluid is greatly reduced by impurity scattering for the case where the solid ^{3}He on the surface was replaced with ^{4}He. In brief, we do not have a good understanding of the phase diagram of the superfluid phases of ^{3}He in aerogel, in particular in zero applied magnetic field. Our acoustic measurements have the great advantage that they can be performed with equal facility in zero and nonzero applied fields.

The acoustic technique is similar to that used previously in Refs. [4,17] and a schematic of our acoustic cavity is depicted in Fig. 1. The cavity was formed with two quartz transducers separated by two stainless steel parallel wires of diameter $d = 250\ \mu$m. One transducer was AC-cut for transverse sound, and the other X-cut for longitudinal sound, with a diameter of 9.5 mm. Their fundamental frequencies were 4.8 and 2.9 MHz, respectively. This

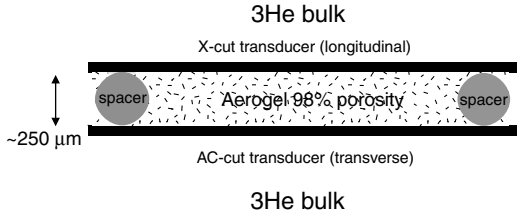

FIG. 1. Schematics of the acoustic cavity. The X-cut (longitudinal sound) and AC-cut (transverse sound) transducers are separated by 250 μm spacers and the 98% porous aerogel was grown *in situ*. Each transducer has two active sides; one probes the aerogel-filled cavity while the other probes the bulk liquid outside the cavity.

arrangement permitted experiments with either transverse or longitudinal sound waves. The aerogel was grown *in situ* in the volume between the transducers. Each transducer has two active sides; one probes the aerogel-filled cavity, and the other the bulk liquid ^{3}He outside the cavity. All experiments were performed with pure ^{3}He verified to contain less than 250 ppm of ^{4}He, much less than the amount required to cover the aerogel strands with one atomic layer of ^{4}He.

The electrical impedance of the transducers was measured using a continuous wave spectrometer. The measurements were performed at a fixed frequency corresponding to odd harmonics of the fundamental resonance, with a frequency modulation of 400 Hz and modulation amplitude of 3 kHz. For longitudinal sound, small changes in the attenuation and velocity produce changes in the electrical impedance of the transducers detected by the spectrometer. For transverse sound waves, the situation is more complex, and we do not have a quantitative framework for interpretation of the impedance measurement. It is not possible with this technique to separate contributions from attenuation, sound velocity, or coupling to collectives modes. However, we have found that, at low frequencies, the impedance changes abruptly at the known phase transitions in each of the bulk or aerogel superfluids, Fig. 2.

The transverse acoustic response at 8.691 MHz and 25 bars pressure is shown in Fig. 2 on cooling (upper trace) and warming (lower trace) and at zero (upper panel) and 2.1 kG applied magnetic field (lower panel). The phase transitions in bulk liquid and the aerogel are denoted by arrows. On cooling we successively observed the bulk superfluid (T_{c0}), bulk A-B (T_{AB0}), aerogel superfluid (T_{ca}), and aerogel A-B (T_{ABa}) transitions. The thermometry scales for cooling and warming are different. The scale shown in the figure is for slow warming such that equilibrium is assured between the lanthanum-doped cerium magnesium nitrate thermometer used in low field, the melting curve thermometer, and the aerogel sample. For the results in Figs. 3 and 4, we corrected the thermometry for the more rapid cooling experiments using the temperature dependence of the acoustic impedance established in equilibrium during warming. We also observed the bulk A_1 transition as a "knee" in the acoustic signature at T_{c0}

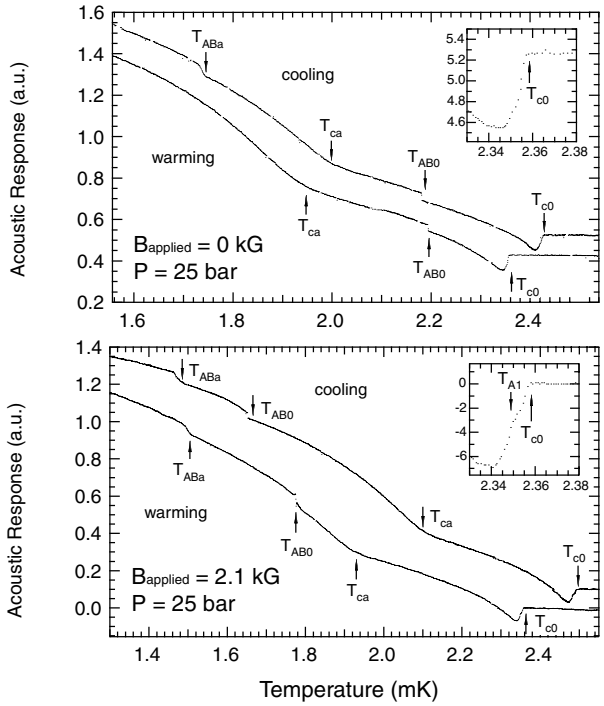

FIG. 2. Transverse acoustic response at 25 bars and zero applied magnetic field (upper panel) and 2.1 kG (lower panel). In each panel, the data in the upper (lower) trace are taken on cooling (warming). The temperature scale is for warming only. For cooling, the temperature of the transitions was determined from the impedance trace calibrated from the warming experiment. The arrows indicate the bulk and aerogel phase transitions. The inset is an enlarged view of the bulk superfluid transition at T_{c0} which, at 2.1 kG, shows the bulk A_1 transition.

shown in the inset of the lower panel which scales with field from 2.1 to 2.9 kG. Note that the bulk A-B transition was highly supercooled at 2.1 kG, but not at zero field.

We verified that the change of slope in the trace labeled T_{ca} corresponds to the superfluid transition in aerogel by comparing with attenuation of longitudinal sound in the same sample where the attenuation is known to drop on cooling through the superfluid transition [4]. The transition temperatures we observe in zero field are in excellent agreement with those reported elsewhere [2,7,8,10] for similar density aerogels. However, one might ask if the A-B aerogel transitions might arise from a local effect near the surface of the transducer. The comparison with longitudinal sound shows that this is not the case. The longitudinal sound mode is of sufficiently low attenuation that acoustic standing waves are established in the cavity. At high fields we observed coincident acoustic signatures of T_{ABa} in the longitudinal acoustic trace which confirms that the signatures from transverse impedance reflect the superfluid behavior of the entire aerogel sample.

The magnetic field dependence of the aerogel superfluid transitions is shown in Fig. 3 at a pressure of 25 bars using transverse acoustics at 8.691 MHz. The triangles are the aerogel superfluid transition. The filled circles are the equilibrium aerogel B-A transition (on warming) and

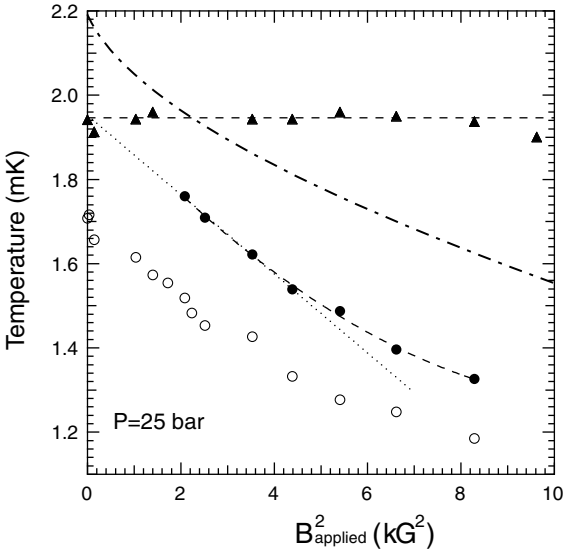

FIG. 3. Phase diagram of the superfluid phases in aerogel at 25 bars in a magnetic field. The triangles are the superfluid transition temperature, T_{ca}, determined from transverse acoustics. The supercooled aerogel A-B transition, T_{ABa}, is shown on cooling (empty circles) and in equilibrium on warming (filled circles). The dot-dashed line is the field dependence of the bulk A-B transition at the same pressure [18]. The dashed lines are guides to the eye, and the dotted line is an extrapolation of T_{ABa} to zero field.

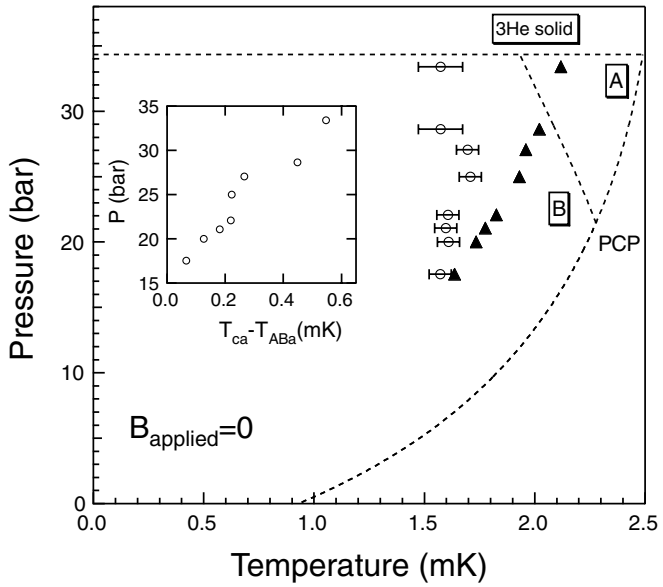

FIG. 4. Phase diagram of ^{3}He in 98% porous aerogel in *zero applied magnetic field*. The triangles are the aerogel superfluid transition and the open circles the aerogel A-B transition on cooling. The thermodynamic B-A transition (on warming) was not observed for any of the pressure shown in this plot. The inset shows the magnitude of supercooling of the aerogel A-B transition as a function of pressure. The dotted line is the bulk ^{3}He superfluid phase diagram and the bulk A, B superfluid and solid phases are labeled.

the empty circles are the supercooled A-B taken on cooling. The dot-dashed line is the bulk superfluid A-B transition [18]. The dashed lines are guides to the eye, and the dotted line is an extrapolation of T_{ABa} to zero field. The aerogel superfluid transition T_{ca} is clearly field independent [19] over the range of magnetic fields used in this work, 0 to 2.9 kG. The fact that the normal-to-superfluid aerogel transition is independent of magnetic field is a strong indication that the superfluid is an equal spin-pairing state as for the bulk A phase in nonzero magnetic field. The aerogel superfluid transition is sufficiently broad that we would not expect in this field range to observe changes owing to the appearance of an A_1 transition as shown, for example, in Fig. 2 for the bulk superfluid. The remarkable supercooling of T_{ABa}, $\sim$200 μK, even in zero applied field is shown as a function of pressure in Fig. 4 and will be discussed later in greater detail. Supercooling indicates that the transition is thermodynamically of first order and its strong field dependence suggests that it is a transition from an equal spin-pairing state to a nonequal spin-pairing state, similar to the bulk A-B transition. These results are consistent to that reported at 32 bars by Barker *et al.* [8] for aerogel preplated with ^{4}He, suggesting that ^{4}He does not greatly affect the superfluid phase diagram. With our technique we were unable to observe the aerogel B-A transition on warming at fields below 1.4 kG, that is, for $T_{ABa}/T_{ca} \geq 0.92$. However, we can make a linear extrapolation to low fields in the manner shown in the figure with a dotted line. This extrapolation is consistent with our observed field dependence of the supercooled A-B transition given as open circles. We find that at zero field $T_{ABa} = 1.95 \pm 0.01$, collapsing to the superfluid transition $T_{ca} = 1.946 \pm 0.009$ mK within the accuracy of our data. This indicates that the equilibrium phase space of the A phase at 25 bars must be extremely narrow, $\leq$20 μK, a behavior quite different from the bulk superfluid.

The linearity of the data at low fields as a function of B^2, Fig. 3, suggests that a Ginzburg-Landau theory near T_{ca} can be applied. To first order in B^2, the field dependence of the aerogel A-B transition T_{ABa} is given by

$$1 - \frac{T_{ABa}}{T_{ca}} = g_a\left(\frac{B}{B_0}\right)^2 + \mathcal{O}\left(\frac{B}{B_0}\right)^4, \qquad (1)$$

where the magnetic field scale B_0 is defined as

$$B_0 = \sqrt{\frac{8\pi^2}{7\zeta(3)}}\,\frac{k_B T_{ca}}{\gamma\hbar}\,(1 + F_0^a), \qquad (2)$$

and γ is the gyromagnetic ratio of ^{3}He, F_0^a is a Fermi liquid parameter, and g_a is a strong-coupling parameter [18] defined similarly as for bulk ^{3}He and depends on combinations of the five coefficients of the fourth-order invariants in the Ginzburg-Landau theory [6]. The g_a parameter in aerogel can be extracted directly from our data. We find that $g_a = 4.3 \pm 0.3$ at 25 bars in aerogel. The bulk value

of the parameter, g_0, is 1 in weak coupling theory but is observed to vary strongly with pressure, 1.61 at zero pressure, 4.41 at 12.8 bars, and it increases markedly at the polycritical pressure (PCP) of 21.45 bars [18]. The small value for g_a at 25 bars in aerogel, above the bulk polycritical pressure, is a significant effect of impurity scattering.

In Fig. 4, the phase diagram at *zero applied magnetic field* is shown for the aerogel and bulk ^{3}He. On cooling, the aerogel *A*-*B* transition is observed for pressures above and below the bulk polycritical pressure, but was not observed on warming, and in particular at 25 bars we have shown that the region of stable *A* phase in the aerogel must be very small. The extent of supercooling does not differ much from run to run, nor is it strongly field dependent. However, the supercooling is highly pressure dependent as is clear from the inset of Fig. 4. The observation of the aerogel metastable *A* phase in zero applied field is striking, particularly below the polycritical pressure where metastability in the bulk superfluid has never been reported. The supercooling seems to vanish in the vicinity of 15 bars. Nonetheless, the existence of an aerogel *A*-*B* transition on cooling implies that there must be a nonzero region of *A* phase in equilibrium. In an applied magnetic field it is straightforward to account for an *A* phase, as, for example, was observed by Barker *et al.* [8] in 284 G and with ^{4}He preplating. They observe a similar phase diagram as Fig. 4 under these conditions. However, in our case in zero applied field, supercooling might arise for several reasons. Small residual fields from the demagnetization magnet, the Earth's magnetic field, or from the nuclear dipolar field of the liquid itself might give rise to a stable but narrow *A*-phase region. We estimate that this is ~ 0.1 μK for a background field of ~ 10 G. It might also be possible that even in zero magnetic field there is an equilibrium region of *A* phase but from our measurements at 25 bars this should be less than ~ 20 μK. Future work is needed to resolve this issue.

Using transverse acoustic impedance experiments we have observed that impurity scattering from a 98% porosity aerogel produces a significant modification of the bulk phase diagram of superfluid ^{3}He. We have concentrated our attention to a region at high pressure near the bulk polycritical point. The transition temperature to an aerogel superfluid is field independent up to 3 kG. Most of the observable pressure-temperature plane of the aerogel superfluid in zero applied magnetic field is a *B* phase, in contrast to the bulk superfluid which exhibits a polycritical point with a large region of stable *A* phase. We believe that, in equilibrium, there also must be a thin, but unobserved, sliver of stable *A* phase which accounts for metastability of this phase on cooling. Finally, the magnetic field dependence of the *A*-*B* phase boundary at 25 bars can be described by Ginzburg-Landau theory including the effects of impurity scattering.

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