Editors' Suggestion

Magnetic Susceptibility of Andreev Bound States in Superfluid ^{3}He-B

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Nuclear magnetic resonance measurements of the magnetic susceptibility of superfluid ^{3}He imbibed in anisotropic aerogel reveal anomalous behavior at low temperatures. Although the frequency shift clearly identifies a low-temperature phase as the B phase, the magnetic susceptibility does not display the expected decrease associated with the formation of the opposite-spin Cooper pairs. This susceptibility anomaly appears to be the predicted high-field behavior corresponding to the Ising-like magnetic character of surface Andreev bound states within the planar aerogel structures.

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Unconventional superconductors break symmetries beyond gauge symmetry and are classified by the symmetries of the order parameter. The paradigm of unconventional superconductors is ^{3}He, a spin-triplet p-wave BCS superfluid [1,2] with two zero field phases A and B, breaking and preserving time-reversal symmetry, respectively. One identifying characteristic of unconventionality is the strong suppression of the transition temperature, and concomitantly, the amplitude of the order parameter induced by nonmagnetic impurities [3]. This is in stark contrast to conventional superconductivity [4]. In the case of ^{3}He, dilute nonmagnetic impurities of silica aerogel particles that are much smaller than the coherence length reduce both the amplitude of the order parameter and the transition temperature [5,6], just as expected for an unconventional superconductor [7,8]. The ^{3}He quasiparticle scattering from these impurities produces surface bound states, Andreev bound states [9], irrespective of whether the surface scattering of ^{3}He quasiparticles is specular or diffuse. Specularity can be achieved by coating the surfaces with at least two atomic layers of ^{4}He [10]. Under those conditions the bound states in ^{3}He-B are expected to be Majorana fermions [11].

The evidence for the existence of these Andreev bound states, both theoretical and experimental, and their relation to the topological character of the B phase have been reviewed by Mizushima *et al.* [11]. Acoustic impedance measurements have resolved a density of states consistent with the existence of a Majorana cone as specularity is increased [12]. The bound states are also responsible for dissipation from vibrating wire devices in the low-temperature limit of the B phase [13]. Theoretical interpretation of ion mobility measurements at the specular free surface are also consistent with their Majorana character [14,15]. Particularly relevant for the present work are theoretical predictions for the enhancement of the magnetic susceptibility [16–18] of the bound states. Here, we report the detection of an anomalous enhancement to the magnetic susceptibility in the B phase within a 98% porosity silica aerogel with specular boundary conditions which we attribute to the magnetic susceptibility of ^{3}He Andreev bound states.

The application of aerogels for investigation of ^{3}He superfluid phases is now widespread [19]. It is significant that silica aerogels with global anisotropy [20] couple directly to the ^{3}He orbital angular momentum, producing well-defined, uniform order parameter textures [21,22]. The aerogel structure also determines the stability of different superfluid phases. Silica aerogel samples can be either grown or mechanically strained to produce uniaxial anisotropy [23], stabilizing the A phase when stretched (positive strain) [24] and the B phase under compression (negative strain) [25]. In both phases there is a second transition on cooling at $T_x < T_c$ from a uniform texture with angular momentum $\boldsymbol{\ell}$ parallel to the anisotropy axis to a uniform texture with $\boldsymbol{\ell}$ perpendicular to the anisotropy axis [20]. We refer to this as the orbital flop transition [22], clearly evident in Fig. 1(a) at $0.77T_c$, where T_c is the superfluid transition temperature. A different class of nematic alumina aerogels, nafen, are extremely anisotropic; they stabilize the polar phase [26] which is a new p-wave superfluid phase that hosts half-quantum vortices [27]. In the present work we find that positive strain and specular boundary conditions for ^{3}He quasiparticle scattering are requirements for the enhanced magnetic susceptibility that we observe in the superfluid B phase.

We determine the magnetic susceptibility χ relative to the normal Fermi liquid magnetic susceptibility χ_N from the nuclear magnetic resonance (NMR) spectra of superfluid ^{3}He imbibed in an anisotropic 16% stretched aerogel (see the Appendix for details). Computational studies with a biased diffusion-limited cluster aggregation algorithm show that the microscopic structure of stretched aerogel has an anisotropic mean-free path and planar voids [20].

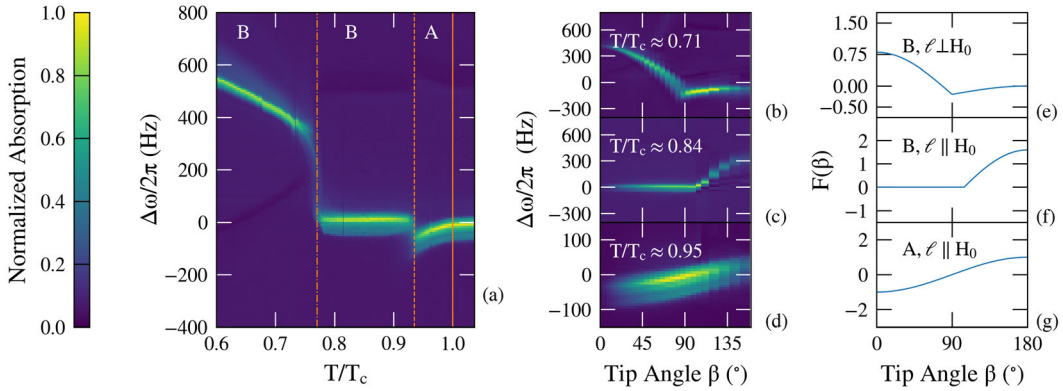

FIG. 1. Temperature dependence and tip angle dependence of NMR absorption spectra of ^{3}He imbibed in 16% stretched aerogel compared with pure ^{3}He. In (a)–(d) the absorption spectrum is shown at each temperature and tip angle as a function of frequency by a color scale representing the spectral amplitude normalized to the maximum absorption amplitude in each panel. (a) Temperature dependence of spectra at tip angle, $\beta = 8°$, $P = 26.6$ bar, $H_0 = 74.5$ mT. The configurations of the order parameter in order on warming; below the orbital flop transition $T_x = 0.77T_c$, $\boldsymbol{\ell} \perp \boldsymbol{H_0}$ [22]; above $T_x = 0.77T_c$, $\boldsymbol{\ell} \| \boldsymbol{H_0}$; above $T_{ab} = 0.93T_c$ the tip angle dependence is characteristic of the A phase with $\boldsymbol{\ell} \| \boldsymbol{H_0}$. (b)–(d) Tip angle dependence of spectra in (a). (e)–(g) Theoretical tip angle dependence of spectra for pure ^{3}He corresponding to (b)–(d) [28].

Unlike in our prior experiments on similarly stretched aerogel, we have added ≈3.5 layers of ^{4}He to the surface, changing the quasiparticle scattering potential [10,29] and eliminating the contribution of surface solid ^{3}He to the spectral weight of the NMR signal [30]. The addition of surface ^{4}He has also been previously shown to alter the stability of superfluid phases imbibed in aerogel [31–33].

We identify a B phase from the NMR frequency shift, Fig. 1. In the superfluid, macroscopic coherence of the nuclear dipole-dipole interaction causes a frequency shift $\Delta\omega$ away from the Larmor frequency $\omega_L = \gamma H_0$ [1]. The frequency shift $\Delta\omega$ is given by

$$2\omega_L \Delta\omega = \Omega(P,T)^2 F(\beta). \tag{1}$$

The magnitude of the frequency shift is set by the square of the longitudinal resonance frequency $\Omega^2 \propto \Delta(P,T)^2/\chi$, where $\Delta(P,T)$ is the pressure- and temperature-dependent order parameter amplitude. The frequency shift dependence $F(\beta)$ on the NMR tip angle β is set by the structure and orientation of the spin and orbital degrees of freedom of the order parameter [28]; further discussion of the NMR experiment, including the tip angle, is given in the Appendix. The theoretical behavior of $F(\beta)$ for pure ^{3}He is displayed in Figs. 1(e)–1(g), and can be compared with our data in Figs. 1(b)–1(d). The orbital degrees of freedom are defined by the orbital angular momentum axis $\boldsymbol{\ell}$, which is oriented by the aerogel anisotropy axis. In the present work the static magnetic field $\boldsymbol{H_0}$ is aligned with the axis of anisotropy of the aerogel.

At temperatures between T_c and T_{ab}, as seen in Fig. 1(a), the nuclear magnetic resonance frequency decreases with decreasing temperature. This negative frequency shift is characteristic of the A phase with $\boldsymbol{\ell} \| \boldsymbol{H_0}$. Its tip angle dependence is shown in Fig. 1(d), consistent with theory Fig. 1(g) [28]. This configuration results from anisotropic quasiparticle scattering induced by the planar aerogel structure [20] in a manner analogous to confinement in a slab [34] and consistent with results from other anisotropic aerogels [21,35]. For comparison, in the pure bulk superfluid the energetically favored configuration is $\boldsymbol{\ell} \perp \boldsymbol{H_0}$ for which the frequency shifts are positive.

At lower temperatures, the superfluid enters the B phase which we identify from its characteristic NMR frequency shifts, shown in Fig. 1(a). The shifts above and below $T_x = 0.77T_c$ are characteristic of the B phase with $\boldsymbol{\ell} \| \boldsymbol{H_0}$, Figs. 1(c) and 1(f), and $\boldsymbol{\ell} \perp \boldsymbol{H_0}$, Figs. 1(b) and 1(e), respectively, where T_x is the orbital flop transition [22], referred to earlier. The magnetic susceptibility in the B phase is temperature independent within our measured temperature range, Fig. 2, in striking contrast to the temperature-dependent magnetic susceptibility in the pure superfluid.

In pure ^{3}He B, the reduction of the magnetic susceptibility with decreasing temperature corresponds to the formation of opposite-spin Cooper pairs [39,40]. This is observed in both pure and impure superfluids [31,36]. The extent of the reduction is less in a large magnetic field [38,41], or in confinement [37], which tends to suppress the formation of opposite-spin pairs. The effect corresponds to a polar distortion of the B phase order parameter that can be measured independently. We follow the method of Rand *et al.* [38], finding that the distortion varies smoothly from ∼0.34 to 0.09 in the region below $0.77T_c$, as shown in Fig. 3 (see Appendix). The polar distortion ≤ 0.09 increases the susceptibility in the pure B

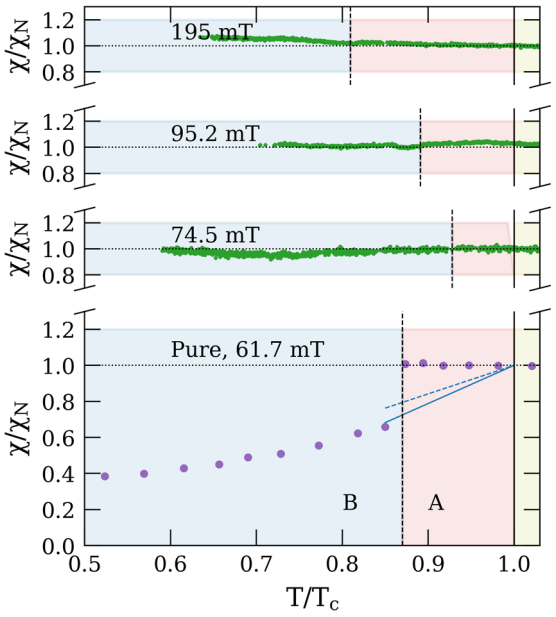

FIG. 2. Magnetic susceptibility χ/χ_N (green circle) over a range of fields at $P = 26.6$ bar. In contrast to the behavior expected, the magnetic susceptibility is temperature independent in the B phase (blue background) for all fields, different pressures, temperatures below T_{ab}, and different orbital configurations. For comparison, χ/χ_N of the pure superfluid at $P = 27.0$ bar is shown in the lowest panel (purple circle), taken from Ref. [36]. The blue solid (dashed) lines are the Born (unitary), Ginzburg-Landau results from the homogeneous isotropic scattering model for ^{3}He in aerogel [9].

phase by $\lesssim 0.15\chi/\chi_N$ [38,41]. Polar distortion cannot account for the excess susceptibility. Similarly, the effect of quasiparticle scattering from impurities is also too small, as shown in Fig. 2.

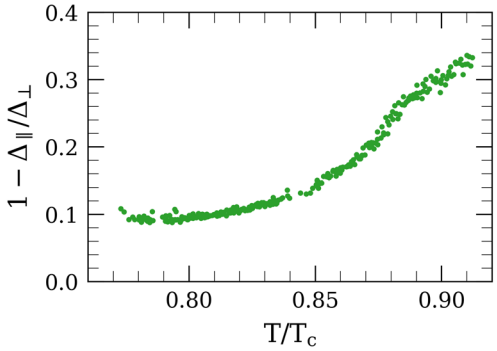

FIG. 3. Measured order parameter distortion. The order parameter amplitude distortion in the direction of the magnetic field, $1 - \Delta_\parallel/\Delta_\perp$ at $H_0 = 74.5$ mT for the temperature range between T_{ab} and $T_x = 0.77T_c$. The degree of order parameter distortion $\approx$0.34 close to T_{ab} is comparable to that seen in nanoconfined planar slabs [37]. At lower temperatures the distortion we measure $\approx$0.09 approaches the value for bulk, pure ^{3}He $\approx$ 0.005 in a 112 mT magnetic field [38].

The earliest magnetic susceptibility experiments on the impure B phase in a nominally isotropic aerogel were performed by Sprague *et al.* [31] with ^{4}He covering the surface of the aerogel, similar to the experiments we report here. They found a small increase in the susceptibility compared to the pure superfluid state, subsequently accounted for by microscopic theories for isotropic impurities including polarization of Andreev bound states [9,42]. In the treatment by Sharma and Sauls [9], the aerogel was modeled as uncorrelated impurities with a single mean-free path, from which both susceptibility and suppression of the critical temperature were determined. This theory has also been used to account for the temperature-dependent susceptibility observed in both isotropic [43] and compressed aerogel samples [25] without ^{4}He preplating, which do not exhibit the anomalous behavior we report here. The susceptibility in the Ginzburg-Landau limit of the quasiclassical theory is shown in Fig. 4 as a blue solid (dashed) line for the Born (unitary) scattering limits.

Theoretical studies of the magnetic susceptibility from bound states in the B phase near a solid plane boundary have found two effects, the emergence of a spontaneous polarization normal to the surface of the plane [16] and the enhancement of the magnetic susceptibility in a confined slab [17]. In the latter case, it was predicted that confining the B phase to a slab causes an increase in the susceptibility, with the highest levels of confinement recovering the susceptibility of the normal liquid [17,47]. A detailed study of the field dependence of Andreev bound states found that the susceptibility increase is associated with a gap in the Andreev bound state dispersion spectrum induced by magnetic field [48].

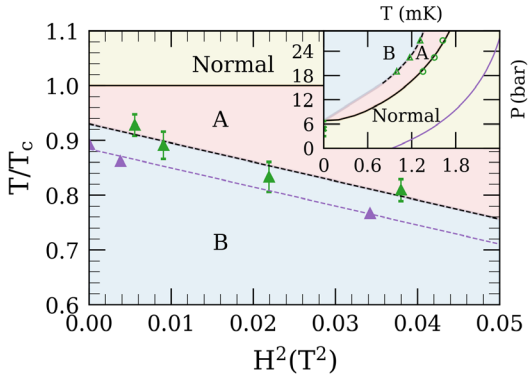

FIG. 4. Magnetic phase diagram at $P = 26.6$ bar. The green trace (green shaded triangle) denotes the experimentally observed T_{ab} in the aerogel. A linear fit shows the extrapolated T_{ab} at zero field to be $T_{ab} = 0.93T_c$. The purple trace (purple shaded triangle) denotes the $A - B$ transition in pure bulk superfluid at $P = 27.0$ bar [36,44] for comparison. Inset: the pressure-temperature phase diagram at $H_0 = 195$ mT, with critical temperature T_c (green open circle), $A - B$ transition temperature T_{ab} (green open triangle). The black curve is a fit for the observed T_c based on an impurity model [7,45,46]. The purple line denotes the pure liquid T_c, the dashed black line is a guide to the eye.

The NMR frequency shift of the superfluid below T_{ab} precludes the identification of the superfluid state as any of the experimentally observed or theoretically well-established equal spin pairing states, such as the polar or planar phases. The magnetic susceptibility also does not display the decrease characteristic of the isotropic impure superfluid models, and is inconsistent with the Ginzburg-Landau limit of these theories. Simulations of the growth of anisotropic aerogels [20], together with small angle x-ray scattering [23], provide strong evidence for the existence of planar structures in stretched aerogels. As a consequence, we propose that planar structures in the aerogel are responsible for the enhanced susceptibility in a manner analogous to the predictions for Andreev bound states in confined slabs.

According to theory, a slab of thickness of $\approx 5\xi_0$ is required to recover the full normal state susceptibility from bound states [17]. In the pressure range of the current work this corresponds to 130 nm taking the zero temperature cohrence length to be $\xi_0 = (\hbar v_F)/(2\pi k_B T_c)$, where v_F is the Fermi velocity. Numerical simulations of the stretched aerogel structure indicate an approximately planar mass distribution with plane separations of ~50–100 nm. This is similar to the calculated quasiparticle mean-free path of 90–120 nm [20]. Although aerogel and slab confinements are very different, the slab thickness for the expected enhanced magnetic susceptibility is roughly the same as that of the planar structure of the stretched aerogel.

The A to B transition temperature T_{ab} decreases with a quadratically increasing field with a slope comparable to that of the pure superfluid and similar for all isotropic aerogels and anisotropic silica aerogels with ^{4}He preplating including the present work, and shown in Fig. 4 [33,36,49–52]. This slope taken at comparable pressures in aerogel samples with and without preplating is included in Supplemental Material as Table 1 [53]; see also Refs. [25,33,43]. Since this transition is first order its field dependence is given by a Clausius-Clapeyron relation between the differences in susceptibility χ and entropy S for the two superfluid phases:

$$\frac{dT_{ab}}{dH^2} = -\frac{1}{2}\frac{\chi_A - \chi_B}{S_A - S_B}. \tag{2}$$

Our measurements of the magnetic susceptibility χ_B are in stark contrast with the Clausius-Clapeyron relation.

In summary, we have discovered an anomalous contribution to the susceptibility of superfluid ^{3}He-B imbibed in a stretched aerogel. This susceptibility appears to be a consequence of planar regions in the aerogel structure and is surprisingly large, essentially identical to that of the normal state, similar to predictions for the susceptibility of ^{3}He-B confined to a planar slab with specular boundary conditions [17]. How this can be reconciled with the field dependence of the AB transition remains an open question.

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Appendix: Experimental methods.—We grow the 16% stretched aerogel with the rapid supercritical extraction method presented in Pollanen *et al.* [23]. The aerogel is grown in a 4.20 mm inner diameter glass NMR tube; however, during the growth process the aerogel shrinks radially inward from the walls of the NMR tube. The NMR sample is approximately 3.53 mm in diameter and it measures 5.5 mm long. Density measurements show the sample to be approximately 97.5% porous. Optical birefringence measurements reveal an axis of structural anisotropy aligned with the cylindrical axis of the aerogel. The cylindrical axis is oriented along the direction of the static NMR field H_0 and connected by a fill line to the silver heat exchanger of the cryostat.

The NMR experiments require a radio frequency pulse of length $\tau_p \propto \beta$, the proportion calibrated in the normal Fermi liquid by observing the dependence of the intensity of the NMR signal on the length of the pulse τ_p; maximal intensity corresponds to $\beta = 90°$. After cooling by nuclear demagnetization, intermittent NMR pulses of several fixed lengths are delivered as the sample warms, producing the color map in Fig. 1(a). During this process we measure temperature via a capacitive pressure transducer ^{3}He melting curve thermometer [44] and a ^{195}Pt susceptibility thermometer. The cryostat can also be held at a roughly fixed temperature while spectra are captured for a variety of pulse lengths, producing the color maps in Figs. 1(b)–1(d). Each NMR spectrum in the temperature sweep data is accumulated over a small region of temperature, typically ~8 μK.

Measurements were performed between pressures of 19.0 and 26.6 bar and magnetic fields from 74.5 to 195 mT. The pressure-dependent critical temperature suppression of the 16% stretched aerogel is consistent with a quasiparticle mean-free path $\lambda = 131$ nm and aerogel correlation length $\xi_a = 23$ nm [45].

We determined the polar distortion of the amplitude of the order parameter, which is the suppression of $\Delta_\parallel$ aligned with the magnetic field relative to the perpendicular component $\Delta_\perp$, following the method of Rand *et al.* [38]. The frequency shift was measured at two tip angles above and below the critical tipping angle [nominally $\arccos(-1/4) \approx 104°$], separating the regions where $\Delta\omega$ is small and large [e.g., 10° versus ≈135° in Fig. 1(c)]. The frequency shifts for two different pulse lengths taken on warming determine the distortion of the order parameter as a function of temperature. This gap distortion is given by

$$\frac{\Delta_{\parallel}}{\Delta_{\perp}} = \frac{1 + \frac{\Delta\omega_1}{\Delta\omega_2} + 2\frac{\Delta\omega_1}{\Delta\omega_2}\cos\beta_2}{1 - 2\frac{\Delta\omega_1}{\Delta\omega_2}\cos\beta_2}, \tag{A1}$$

with $\Delta\omega_1$ and $\Delta\omega_2$ the frequency shifts of the small and large pulses, respectively, and β_2 the tipping angle of the large pulse [54]. This ratio is independent of the magnetic susceptibility of the superfluid and the B phase longitudinal resonance frequency Ω_B.

The susceptibility is proportional to the integral of the absorption spectrum of the NMR signal. At temperatures above those shown in Fig. 2 there is an additional contribution to the susceptibility from the ^{3}He in the fill lines and from that in the near vicinity of the aerogel, which we refer to as the ^{3}He bath. At temperatures substantially higher than T_c in the aerogel, the bath is in the superfluid A phase. As a consequence, the contribution of the bath to the signal, ~20% of the total spectral weight is shifted in frequency away from the signal of the normal liquid in the aerogel, as shown in Supplemental Material Fig. 1 [53]. At and below T_{ab} in the pure superfluid, the B phase bath signal abruptly becomes very broad. At lower temperatures, below T_c in the aerogel as shown Fig. 2, this contribution cannot be distinguished from the background noise. The influence of the superfluid bath on the susceptibility in both stretched and compressed aerogel can be seen in Supplemental Material Fig. 1 [53]; see also Refs. [22,55].

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