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Orbital-Flop Transition of Angular Momentum in a Topological Superfluid

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The direction of the orbital angular momentum of the B -phase of superfluid ^3He can be controlled by engineering the anisotropy of the silica aerogel framework within which it is imbibed. In this work, we report our discovery of an unusual and abrupt ‘orbital-flop’ transition of the superfluid angular momentum between orientations perpendicular and parallel to the anisotropy axis. The transition has no hysteresis, warming or cooling, as expected for a continuous thermodynamic transition, and is not the result of a competition between strain and magnetic field. This demonstrates the spontaneous reorientation of the order parameter of an unconventional BCS condensate.

Superfluid ^3He can survive in a porous medium provided it is sufficiently tenuous, such as for 98% porosity silica aerogel. Recently it has been shown that uniform strain imposed on this type of aerogel imprints anisotropy on the superfluid resulting in states with either chiral or time-reversal symmetry depending on the sign of the strain.[1–3] In the present work we report a spontaneous reorientation of the direction of the B -phase Cooper pair angular momentum as a function of temperature in a negatively strained anisotropic aerogel.

Unconventional superconductivity and superfluidity break symmetries of the normal Fermi liquid state beyond gauge symmetry. In the case of superfluid ^3He , the p -wave, spin-triplet condensate has angular momentum $L = 1$ and spin angular momentum $S = 1$, and is described by a 3×3 complex matrix dependent on spin and orbital space vectors.[4, 5] The spin degrees of freedom couple to the magnetic field, while the orbital degrees of freedom interact strongly with physical boundaries. Spin and orbit are then coupled through the nuclear dipole-dipole interaction. Here, we investigate the effects of impurities on superfluid ^3He -B relevant to understanding disorder in quantum condensed states with a vector order parameter. In the present work, these impurities are in the form of high-porosity silica aerogel which consists of microscopic particles, $\sim 3\text{nm}$ diameter, in a limited fractal distribution.[6] In the presence of anisotropic disorder from strained aerogel, we have discovered a transition in orientation of the orbital angular momentum in superfluid ^3He . We detect this transition through the orientation of the angular momentum axis relative to the magnetic field from nuclear magnetic resonance (NMR) spectra shown in Fig. 1.

Shortly after the discovery of superfluidity of ^3He imbibed in silica aerogel,[7, 8] the experimental observations of this new class of coherent superfluid states were accounted for with a Ginzburg-Landau theory.[9, 10] The principal parameters of the model are the Fermi quasiparticle mean-free-path, λ , and a silica particle-particle correlation length, ξ_a . An important prediction was that anisotropic (isotropic) Fermi quasiparticle scattering would tend to favor the stability of anisotropic (isotropic) superfluid states. This provoked development

and characterization of aerogel samples with uniform, global anisotropy, as well as ‘isotropic aerogels’ with no preferred direction on length scales greater than a few microns.[6] NMR experiments on the superfluid imbibed in isotropic samples were consistent with these predictions.[11] This was followed by a series of experiments and theoretical works focusing on anisotropic superfluids and anisotropic disorder[1, 12, 13] that has recently been summarized.[3]

Although the order parameter amplitude for the pure superfluid B -phase is completely isotropic, in the presence of an external field that breaks this perfect orbital rotation symmetry (sample boundaries, phase boundaries, superfluid flow, magnetic fields, and anisotropic disorder from impurities on length scales much larger than the superfluid coherence length) its orbital degrees of freedom are constrained, and can be characterized by an orbital angular momentum axis, \mathbf{l}_B . [14] This axis is characterized by a large bending energy, resulting in order parameter textures.[5] In particular, spatial constraints from sample cell walls and external surfaces produce a “flare-out” texture for ^3He -B in its pure form,[14] as well as in isotropic aerogel.[11] For negatively strained aerogel the situation is very different.

Contrary to expectation, a distorted version of the B -phase becomes progressively more stable with increasing negative strain, effectively competing with the A -phase, persisting up to T_c and extending to non-zero magnetic fields.[2] In the present work, we have exploited this fact to explore the orientation effects of anisotropic disorder on the orbital components of the B -phase order parameter over our full temperature range up to T_c , Fig. 1, and as a function of pressure, Fig. 2. We find that the orientation of \mathbf{l}_B is controlled by the strain axis, ϵ , as was first shown in NMR measurements by Li *et al.*[15], Fig. 1 a), b) at 26 bar. Here, we have carried out a thorough investigation of this transition, including increasing our NMR spectral resolution by almost two orders of magnitude, Fig. 1 c), by eliminating two monolayers of paramagnetic solid ^3He adsorbed on the aerogel surface with the substitution of solid ^4He , so-called pre-plating. When present, adsorbed solid ^3He is in fast exchange with the bulk liquid, decreasing the frequency shift for the entire sample

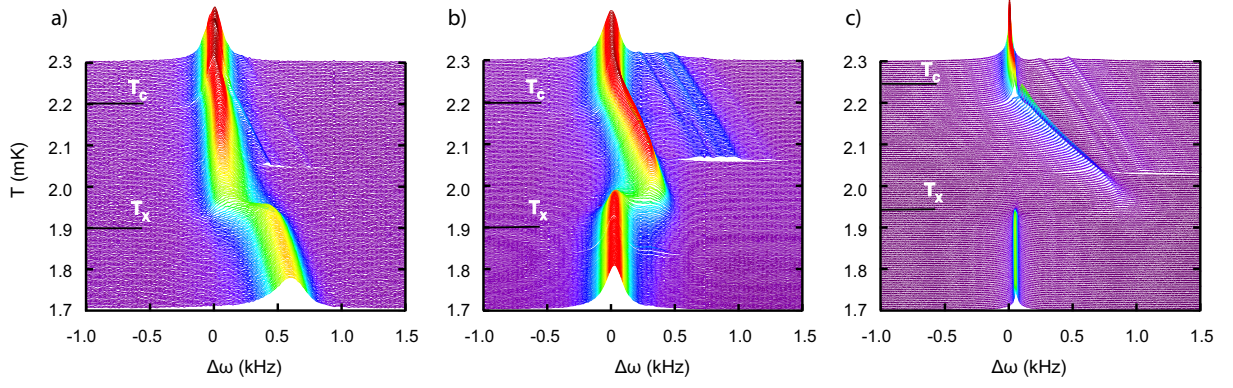


FIG. 1. (Color online). Temperature dependence of the NMR spectra at small tip angle $\beta \sim 10^\circ$ showing an orbital reorientation transition at T_x . Aerogel samples with $\varepsilon \sim -20\%$ strain at $\hat{H}_0 = 0.096$ T; a) $\varepsilon \sim -23\%$, $\varepsilon \perp \mathbf{H}_0$, 26 bar; b) $\varepsilon \sim -19\%$, $\varepsilon \parallel \mathbf{H}_0$, 26 bar; both a) and b) without ^4He pre-plating; and c) the same sample as b) but with ^4He pre-plating at 27 bar. The superfluid transition temperatures are $T_c = 2.19, 2.19$ and 2.24 mK for a) through c), respectively. The large frequency shift below T_x in a) continues smoothly from that above T_x in b). The ripples at high frequency shift are from small bulk superfluid components in the A-phase.

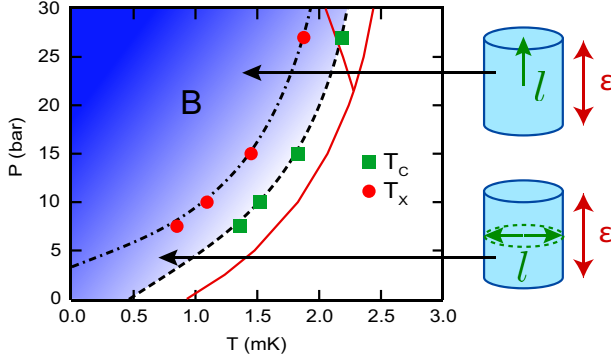


FIG. 2. (Color online) Pressure-temperature phase diagram at low magnetic field, extrapolated to zero, for an aerogel compressed by $\sim 20\%$ from an isotropic sample reported upon earlier.[11] The green squares correspond to the superfluid phase transition, T_c , in aerogel, and the solid red circles to the reorientation transition, T_x , without ^4He pre-plating. The dashed lines are guides to the eye. The solid red lines are the phase diagram of pure superfluid ^3He .

by the ratio of liquid-to-solid magnetization as well as increasing the spectral linewidth.[8, 16] With our increased frequency resolution, it appears that there is a very well-defined temperature T_x above which the order parameter is oriented with $\mathbf{l}_B \perp \varepsilon$, while below, it reorients with $\mathbf{l}_B \parallel \varepsilon$. This transition is sharp and reversible, while the magnetic susceptibility is continuous with temperature throughout, as is characteristic of the pure B-phase. It appears that we can flip the angular momentum orientation 90° by changing the temperature by as little as $15 \mu\text{K}$, Fig. 3, or by what should be equivalent, changing the pressure, Fig. 2.

Pulsed ^3He NMR measurements were conducted with a magnet having a homogeneity of ≈ 2 ppm over the sample volume and magnetic fields from 50 to 200 mT.

Temperature was measured with ^{195}Pt NMR and melting curve thermometry, both with reference to the known NMR signatures of the superfluid transitions from small amounts of bulk ^3He in the sample cell, evident in Fig. 1. Cylindrical silica aerogel samples, 4 mm diameter \times 5 mm length, were grown in glass tubes with nominal 98% porosity. Following a one-step procedure with rapid supercritical extraction, we used optical birefringence[6] to obtain a quantitative measure of its anisotropy.[13] After investigation of superfluid ^3He imbibed in one isotropic sample,[11, 17] we compressed it by $\sim 20\%$ to provide the data in Fig. 1 b), c), and also in Fig. 2-4. The early work

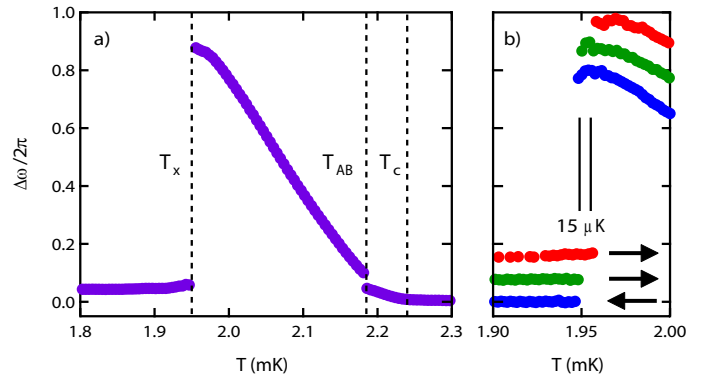


FIG. 3. (Color online). a) NMR frequency shift, $\Delta\omega$, as a function of temperature with $\varepsilon \parallel \mathbf{H}_0$ and ^4He pre-plating at $\hat{H}_0 = 0.096$ T and $P = 27$ bar. This data corresponds to the spectra in Fig. 1c). The large $\Delta\omega$ for $T > T_x$ indicates $\mathbf{l}_B \perp \mathbf{H}_0$ and the small $\Delta\omega$ at low temperatures, $\mathbf{l}_B \parallel \mathbf{H}_0$. There is a small window of the superfluid A-phase close to T_c . b) (Traces are offset vertically for clarity.) Tracking $\Delta\omega$ back and forth across T_x in directions indicated by arrows, shows no hysteresis within the reproducibility of the measurement of temperature, $\sim 15 \mu\text{K}$.

with this sample was performed without ^4He pre-plating, Fig. 1 b), as were experiments with other similarly prepared samples, such as Fig. 1 a).

The order parameter orientation in the B -phase was determined from the peak frequency of the NMR spectra.[5] The coupling of spin and orbital degrees of freedom results in a frequency shift, $\Delta\omega$, away from the normal-state precession at the Larmor frequency, ω_L . [18, 19] This shift is dependent on both the magnitude and orientation of the order parameter, as well as the amplitude of the applied radio frequency pulsed excitation, which tips the nuclear magnetization by an angle β with respect to the static magnetic field, \mathbf{H}_0 . For ^3He -B there are two experimentally realized limits which illustrate the effect of the orientation of \mathbf{l}_B on the NMR response to this excitation. If $\mathbf{l}_B \parallel \mathbf{H}_0$, the frequency shift is given as a function of β as, [20, 21]

$$\Delta\omega_{\parallel} \approx 0, \quad \beta < 104^\circ \quad (1)$$

$$\Delta\omega_{\parallel} = -\frac{4}{15} \frac{\Omega_B^2}{\omega_L} (1 + 4 \cos \beta). \quad \beta > 104^\circ, \quad (2)$$

where Ω_B is the longitudinal resonance frequency of the B -phase and is proportional to the amplitude of the order parameter.[4] When β is small, this results in near-zero measured frequency shift, with the onset of larger shifts for tipping angles greater than the Leggett angle, 104° . This is the expected behavior of the bulk superfluid in a uniform magnetic field, in the absence of any external constraints. The other limit, $\mathbf{l}_B \perp \mathbf{H}_0$, is not the minimum of the dipole energy, and so requires some external constraint imposed on a higher energy scale to orient the angular momentum. If $\mathbf{l}_B \perp \mathbf{H}_0$, the resulting frequency shift is, [22, 23]

$$\Delta\omega_{\perp} = \frac{\Omega_B^2}{2\omega_L} (\cos \beta - \frac{1}{5}), \quad \beta < 90^\circ, \quad (3)$$

$$\Delta\omega_{\perp} = -\frac{\Omega_B^2}{10\omega_L} (1 + \cos \beta). \quad \beta > 90^\circ. \quad (4)$$

In this case, there is a large positive frequency shift measured at small tip angles. This type of behavior has been reported for superfluid ^3He -B in both its pure form[5] and in isotropic silica aerogel,[11] where \mathbf{l}_B is constrained to be normal to a surface. These two limits are displayed in Fig. 4, where the dashed black curve corresponds to $\Delta\omega_{\parallel}$, Eq.'s 1, 2, and the solid black curve for $\Delta\omega_{\perp}$ from Eq.'s 3, 4.

The configuration $\mathbf{l}_B \parallel \mathbf{H}_0$ corresponds to the behavior we observe for $T > T_x$ with $\varepsilon \perp \mathbf{H}_0$ (Fig. 1 a) and for $T < T_x$ when $\varepsilon \parallel \mathbf{H}_0$ (Fig. 1 b), c). For the complementary ranges of temperature in these two cases, the spectra indicate that $\mathbf{l}_B \perp \mathbf{H}_0$. The tip angle dependence of the frequency shift is shown for both regions in Fig. 4, showing qualitative agreement. This is one example from among many measurements, confirming our identification of these orientations of the angular momentum axis relative to the magnetic field. The NMR linewidths of the

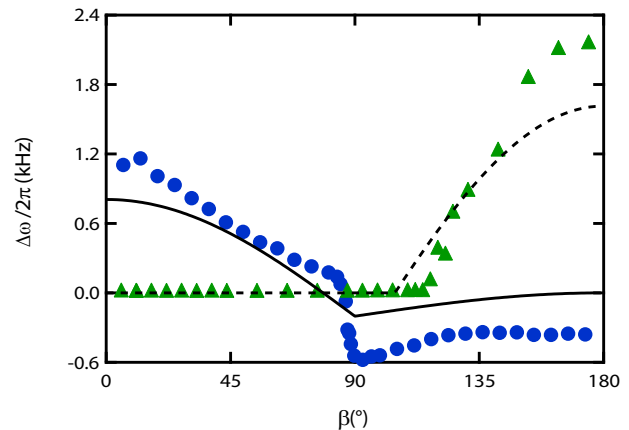


FIG. 4. (Color online). Typical tipping angle dependence of the frequency shift of data above T_x at $T = 2.03$ mK (blue circles) for the sample in Fig. 1 c), and below T_x at $T = 1.76$ mK (green triangles), scaled to 2.03 mK. Measurements were taken with $\varepsilon \parallel \mathbf{H}_0$ and ^4He pre-plating at 0.05 T. The solid (dashed) curve shows the theoretical dependence for $\Delta\omega_{\perp}$ ($\Delta\omega_{\parallel}$) scaled to the frequency shift of isotropic aerogel.[11] The B -phase is distorted by strain resulting in a larger energy gap and frequency shift compared to the isotropic case.[13]

spectra in Fig. 1 a), b) are approximately the same as the normal state, indicating that the entire sample has a uniform orientation of \mathbf{l}_B . For the higher resolution data, Fig. 1 c), the linewidth increases slightly at high temperatures, $T > T_x$, possibly indicating a small deviation from a uniform distribution, since this configuration is not a minimum of the dipole energy. We note that the linewidth is always smaller than that associated with the textures in pure ^3He -B or the B -phase of isotropic aerogel. The temperature at which the transition occurs, T_x , is very well-defined and non-hysteretic within experimental error, Fig. 3b).

This behavior has been investigated in samples with four values of strain: 0%, $\sim -12\%$, $\sim -20\%$, and $\sim -30\%$. We find that T_x is approximately independent of strain between -20% and -30% , Fig. 5 b). The transition is not present in isotropic samples, or for small strain $\sim -12\%$, indicating that there is an anisotropy threshold for the transition to exist above which it is independent. Similarly, T_x is independent of the magnitude of magnetic field and its orientation, Fig. 5, indicating that the reorientation of the order parameter cannot be the result of competition between the strain and magnetic field. T_x is modified only by aerogel surface conditions and pressure, Fig. 2 and 6, demonstrating that the transition is driven by the impurity. The NMR results indicate that for $T > T_x$ the angular momentum is perpendicular to the strain axis, $\mathbf{l}_B \perp \varepsilon$, and that for $T < T_x$ it reorients to be parallel to the strain axis, $\mathbf{l}_B \parallel \varepsilon$. We do not cover a sufficiently large range of magnetic field to stabilize the A phase at temperatures below T_x ; however, we note that above this temperature the angular momentum

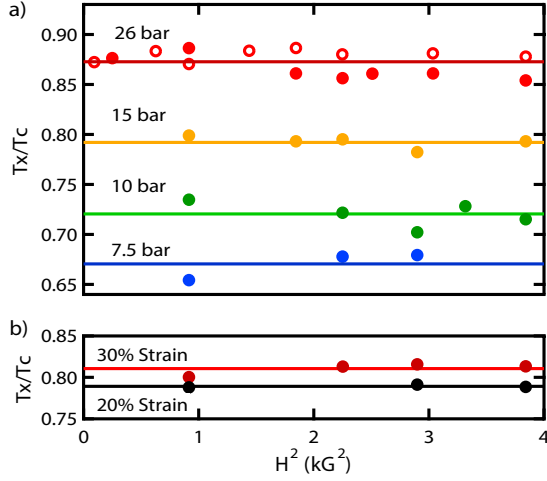


FIG. 5. (Color online) a) Magnetic field independence of T_x/T_c for different pressures without pre-plating. At $P = 26$ bar with $\sim -19\%$ strain, $\epsilon \parallel \mathbf{H}_0$ (open circles), and with $\sim -23\%$, $\epsilon \perp \mathbf{H}_0$ (closed circles). b) Strain dependence is very small between $\sim -20\%$ and $\sim -30\%$ at 27 bar with $\epsilon \perp \mathbf{H}_0$.

in the A -phase is oriented with $\mathbf{l}_A \perp \epsilon$, the same as that of the B -phase at the same temperatures, but at lower magnetic fields.[2]

Various physical systems with vector order parameters exhibit transitions involving their reorientation, most notably the Fréedericksz transition in liquid crystals[24], important in flat screen displays, and the spin-flop transition in antiferromagnets[25], as well as multiferroics[26], which have a variety of electronic switching applications. Additionally, a reorientation Fréedericksz-type transition of \mathbf{l}_A in superfluid $^3\text{He-A}$ was reported, a balance between magnetic field and superfluid flow from rotation.[27] All of these transitions are characterized by a competition between two orienting fields. The transition at T_x is quite unique, appearing spontaneously as a function of temperature.

The interaction of a superconductor with disorder is determined by the superconducting coherence length, ξ_0 . To describe a conventional, $L = 0$ superconductor in the presence of isotropic disorder, Abrikosov and Gorkov[29] developed a theory that expressed the superconducting transition in terms of a pair-breaking parameter, x , given by the ratio of the pure sample coherence length to the quasiparticle mean-free-path associated with quasiparticle scattering from magnetic impurities, $x = \xi_0/\lambda$. The theory was extended to isotropic impurity in unconventional superconductors, by Larkin[30], where both magnetic and nonmagnetic scattering play a role, and later specialized to superfluid ^3He . [9, 31] In the limit of small suppression of T_c relative to T_{c0} , the transition tempera-

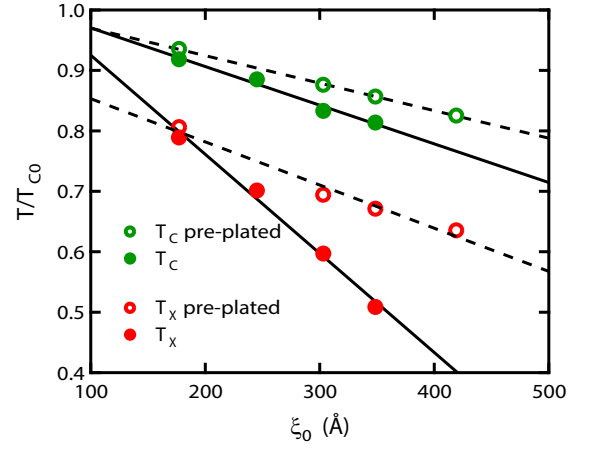


FIG. 6. (Color online) Transition temperatures for T_c and T_x scaled to the pure transition temperature as a function of the pure superfluid coherence length. Closed (open) circles indicate measurements without (with) ^4He pre-plating. The linear dependence suggests that both transitions are driven by impurity and can be characterized by a length scale in the aerogel. Without pre-plating, the extracted length scales are $\lambda = 385$ nm and $\lambda_x = 150$ nm. With pre-plating, they are $\lambda = 550$ nm and $\lambda_x = 347$ nm. For all data, we find $\xi_a \sim 3 - 6$ nm. These should be compared with estimates for the length scales for 98% porosity isotropic aerogel, $\lambda \approx 150$ nm and $\xi_a \approx 40$ nm.[28]

ture is linear in x ,

$$\frac{T_c}{T_{c0}} = 1 - \frac{\pi^2}{4} \frac{\xi_0}{\lambda}, \quad (5)$$

where T_{c0} is the impurity-free transition temperature.

This expression must be modified due to the fractal structure of aerogel by taking correlations into account. Sauls and Sharma[32] redefined the pair-breaking parameter to allow for aerogel silica particle-particle correlations with a correlation length ξ_a by introducing an effective pair-breaking parameter $\tilde{x} = x/(1 + \xi_a^2/(\lambda^2 x))$. A similar result was derived by Fomin by different methods.[31] In the clean limit as $T_c \rightarrow T_{c0}$,

$$\frac{T_c}{T_{c0}} = 1 - \frac{\pi^2}{4} \frac{\xi_0}{\lambda} + \frac{\pi^2}{4} \left(\frac{\xi_a}{\lambda} \right)^2. \quad (6)$$

In Fig. 6, we show that both T_c and T_x vary linearly with ξ_0 for superfluid ^3He in aerogel, suggesting that both are described by quasiparticle scattering from impurities. The different slopes for T_c and T_x indicate that a new length scale, λ_x , must be introduced to describe T_x , taking the place of λ in Eq. 6. After pre-plating, different values of λ and λ_x are extracted due to changes in the scattering conditions. The ratio $\lambda/\lambda_x = 1.6$ with pre-plating and 2.6 otherwise. It is much larger than the aerogel anisotropy of $\sim 20\%$. This, together with the strain independence in Fig. 5 appears to rule out the simple identification of λ and λ_x as the mean free path

perpendicular and parallel to strain. However, it is clear that λ_x is a signature of the strain-induced broken rotational symmetry from the aerogel structure. Determining the exact nature of this new length scale and the mechanism for the angular momentum reorientation will require further investigation.

In summary, we have discovered a unique ‘orbital flop’ transition in superfluid $^3\text{He-B}$ in negatively strained silica aerogel. At a well-defined temperature the transition appears as a spontaneous reorientation of the angular momentum of the Cooper pair by 90° relative to the direction of strain. This transition is a coherent and uniform response of the order parameter to the existence of anisotropy in the aerogel. We infer that the transition temperature corresponds to a specific length scale that is determined by superfluid coherence length.

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