Polar Phase of Superfluid ^{3}He in Anisotropic Aerogel

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We report the first observation of the polar phase of superfluid ^{3}He. This phase appears in ^{3}He confined in a new type of aerogel with a nearly parallel arrangement of strands which play the role of ordered impurities. Our experiments qualitatively agree with theoretical predictions and suggest that in other systems with unconventional Cooper pairing (e.g., in unconventional superconductors) similar phenomena may be found in the presence of anisotropic impurities.

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Introduction.—One of the examples of superfluid Fermi systems with unconventional Cooper pairing is superfluid ^{3}He where the pairing occurs with spin and orbital angular momentum equal to 1. In isotropic space the free energy and the superfluid transition temperature are degenerate with respect to spin and orbital momentum projections. This allows a variety of superfluid phases with the same transition temperature, but in zero magnetic field only two phases (A and B) with the lowest energy are realized [1–3]. Anisotropy of the space lifts the degeneracy and other phases can be stabilized. So, in the magnetic field the spin degeneracy is lifted and the A_1 phase appears in a narrow region below the transition temperature [4]. As shown in Ref. [5] and in further theoretical works [6–8], the orbital degeneracy may be lifted by anisotropic impurities, e.g., by globally anisotropic aerogel in which strands are aligned on average along the same direction ("stretching" anisotropy). In this case a new superfluid phase of ^{3}He, the polar phase, may be stabilized below the transition temperature. It was predicted that on further cooling a second-order transition into a polar-distorted A phase should occur and the distortion should decrease with cooling. At lower temperatures a first-order transition into a polar-distorted B phase was also expected.

Samples of silica aerogel are used in most experiments with ^{3}He in the aerogel. They consist of chaotic SiO_2 strands with diameters of ~3 nm and an average separation of ~100 nm. The superfluid coherence length at different pressures is in the range of 20–80 nm, so the strands play the role of impurities. It is established [9,10] that the superfluid transition temperature of ^{3}He in aerogel (T_{ca}) is lower than the transition temperature (T_c) of bulk ^{3}He but order parameters of the observed A-like and B-like phases correspond to those of the A and B phases [11–15]. Global anisotropy of silica aerogel may change T_{ca} and the A-B transition temperature [16,17], but no evidence of the polar distortion in the superfluid phases is found. Much larger "stretching" anisotropy is inherent to "nematically ordered" aerogel (N-aerogel) which strands are oriented along the same direction $\hat{\zeta}$. There are two types of N-aerogel: "Obninsk aerogel" and nafen [18]. In recent experiments with ^{3}He in Obninsk aerogel two superfluid phases were observed: the polar-distorted A phase at higher temperatures and the polar-distorted B phase at lower temperatures [19–21]. This qualitatively agrees with theoretical predictions, but the existence of the polar phase has not been proved.

Here, for the first time, we have investigated superfluid ^{3}He confined in nafen, which has a much larger overall density than Obninsk aerogels. We have found that in nafen the transition occurs into the pure polar phase. We have also done additional experiments which show that in Obninsk aerogel the polar phase is still not realized.

Samples and methods.—We have used 2 nafen and 2 Obninsk aerogel samples which all have a cuboid shape with sizes of ~4 mm. Experimental chambers were similar to those described in Ref. [19]. Nafen samples were produced by ANF Technology (Tallinn, Estonia). They consist of Al_2O_3 strands and have porosities of 97.8% (sample "nafen-90" with overall density 90 mg/cm^3) and 93.9% ("nafen-243" with density 243 mg/cm^3). Obninsk aerogel samples consist of AlOOH strands and were produced in the Leypunsky Institute (Obninsk, Russia). Their porosities are 97.9% ("Obninsk-50" with density 50 mg/cm^3) and 99.6% ("Obninsk-8" with density 8 mg/cm^3). Strands of all samples are nearly parallel to one another and have diameters 6–9 nm. For more information about the samples see Ref. [18]. In the limit of zero temperature the spin diffusion of ^{3}He in N-aerogel is anisotropic and the ratio of spin diffusion coefficients along and transverse to strands is an important parameter of the theory. For Obninsk-8 this ratio is ~1.5 [22], while for nafen-90 and nafen-243 it is 3.3 and 8.1, respectively [23].

Experiments were performed using continuous wave (cw) and pulse NMR in magnetic fields of 10–37 mT (NMR frequencies are 330–1200 kHz) and at pressures 0.2–29.3 bar. We were able to rotate the external steady magnetic field $\mathbf{H}$ by an arbitrary angle μ with respect to $\hat{\zeta}$. The necessary temperatures were obtained by a nuclear

demagnetization cryostat and measured by a quartz tuning fork calibrated by measurements of the Leggett frequency in bulk ^{3}He-B. To avoid a paramagnetic signal from surface solid ^{3}He, the samples were preplated by ~2.5 ^{4}He monolayers.

Spin dynamics of ^{3}He *in N-aerogel.*—The order parameter of polar, polar-distorted A, and A phases is

$$A_{\nu k} = \Delta_0 e^{i\varphi} d_\nu (a m_k + i b n_k), \quad (1)$$

where Δ_0 is the gap parameter, φ is the phase, $\mathbf{d}$ is the unit spin vector, $\mathbf{m}$ and $\mathbf{n}$ are mutually orthogonal unit vectors in orbital space and $a^2 + b^2 = 1$. For the A phase $a = b$, for the polar-distorted A phase $a^2 > b^2 > 0$, and for the polar phase $a = 1, b = 0$. The A phase and the polar-distorted A phase are chiral and their gap is zero along $\mathbf{l} = \mathbf{m} \times \mathbf{n}$, but the polar phase is not chiral and its gap is zero in the plane normal to $\mathbf{m}$. All these phases are equal spin pairing (ESP) phases in which the spin susceptibility is temperature independent.

We identify superfluid phases by measurements of the NMR frequency shift ($\Delta\omega$) from the Larmor value (ω_L). The shift is due to the dipole interaction of spins of the superfluid condensate and depends on the order parameter, its spatial distribution, and μ. In the polar-distorted A phase strands of N-aerogel destroy the long-range order in orbital space: $\mathbf{m}$ is along $\hat{\zeta}$, but vectors $\mathbf{n}$ are uniform only over ~1 μm and at larger distances form a static 2D Larkin-Imry-Ma (LIM) state [19,20,24] corresponding to a random distribution of $\mathbf{n}$ in the plane normal to $\hat{\zeta}$. This state is similar to the LIM state in the A-like phase in silica aerogel [14,25]. The vector $\mathbf{d}$ is normal to the magnetization $\mathbf{M}$ and must be uniform over a dipole length $\xi_d \sim 10$ μm determined by dipole and gradient energies. At larger distances $\mathbf{d}$ can be uniform ("spin nematic," SN, state) or random ("spin glass," SG, state). The SN state is favorable and usually appears in low excitation NMR. The SG state is metastable and corresponds to a local minimum of the total energy. It can be created in A and polar-distorted A phases by cooling through T_{ca} in the presence of high NMR excitation generating a random $\mathbf{d}$ distribution [14]. On further cooling this distribution is "frozen" and stabilized by the LIM state. For the 2D LIM state, the frequency shift in the SN state is given by [19,20,26]:

$$2\omega_L \Delta\omega = K\left[\cos\beta - \frac{\sin^2\mu}{4}(5\cos\beta - 1)\right]\Omega_A^2, \quad (2)$$

where

$$K = \frac{4 - 6b^2}{3 - 4a^2 b^2}, \quad (3)$$

β is the tipping angle of $\mathbf{M}$ and $\Omega_A = \Omega_A(T) \propto \Delta_0$ is the Leggett frequency of the A phase (if this phase existed and had the same transition temperature). The Leggett frequency grows from 0 up to ~100 kHz on cooling from the superfluid transition [1]. For linear cw NMR ($\cos\beta \approx 1$) and for $\mu = 0$ ($\mathbf{H} \| \hat{\zeta}$) we get

$$2\omega_L \Delta\omega = K\Omega_A^2, \quad (4)$$

while for $\mu = \pi/2$ the shift is zero. From Eqs. (3) and (4) it follows that if Ω_A is known then measurements of $\Delta\omega$ for $\mu = 0$ allow determination of the polar distortion value: in the A phase K should equal $1/2$ while for the polar phase $K = 4/3$. However, there are two problems. The first is that the Leggett frequency was measured only in bulk ^{3}He (we denote this value by Ω_{A0}) [27,28]. In aerogel Ω_A is smaller due to suppression of the transition temperature ($\Delta T_{ca} = T_c - T_{ca}$) and the corresponding decrease of Δ_0. Fortunately, in our experiments ΔT_{ca} is small (2%–10% of T_c depending on pressure). Then, to a first approximation, we can use Ω_A obtained by a rescaling of Ω_{A0}:

$$\Omega_A(T/T_{ca}) = \frac{T_{ca}}{T_c}\Omega_{A0}(T/T_c). \quad (5)$$

The second problem is that Eq. (3) is derived in the weak-coupling limit [1], which is believed to be a good approximation for bulk ^{3}He at low pressures. However, strong coupling corrections should not exceed ±5% [29]. Equation (3) also does not account for an influence of spin diffusion anisotropy on the dipole energy [29]. This correction is also expected to be small and this is proved by our results (see below).

Thus, measurements of $\Delta\omega$ for $\mu = 0$ allow estimation of the distortion with good accuracy, but to distinguish the pure polar phase from the polar-distorted A phase this is not enough. For this purpose we can compare NMR properties of the SN and SG states. Continuous wave NMR frequency shift in the SG state for $\mu = 0$ is the same as in the SN state but for $\mu = \pi/2$ it is negative and is given by [19,20]

$$2\omega_L \Delta\omega = -CK\Omega_A^2, \quad (6)$$

where $C = 1/2$ for an isotropic $\mathbf{d}$ distribution in the plane normal to $\mathbf{M}$. For $\mu = \pi/2$ the value of C may be smaller because the $\mathbf{d}$ distribution becomes anisotropic (see Supplemental Material [26]), but the shift remains negative. Note that the polar phase order parameter does not contain $\mathbf{n}$; i.e., the SG state cannot be stabilized and for $\mu = \pi/2$ the shift should always equal zero. Thus, the negative shift for $\mu = \pi/2$ indicates that it is the SG state and that it is not the pure polar phase.

Results and discussion.—In experiments with ^{3}He in Obninsk aerogel with density 30 mg/cm^3 [19] it was found that in the polar-distorted A phase the distortion is maximal near T_{ca} where, depending on pressure, values of K were within the range 0.6–1.07. The maximal K was obtained

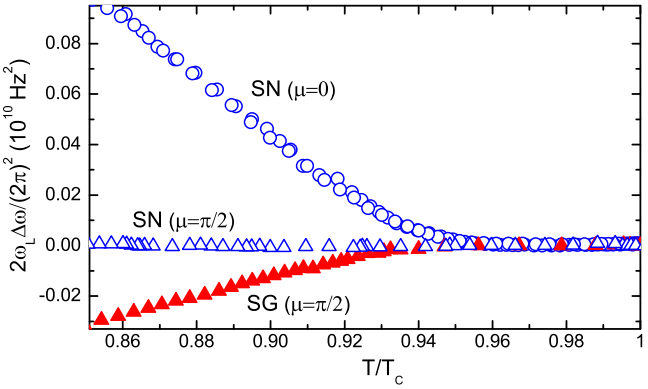

FIG. 1 (color online). The frequency shift versus temperature in ^{3}He in the Obninsk-50 sample. Circles—the SN state for $\mu = 0$, open triangles—the SN state for $\mu = \pi/2$, filled triangles—the SG state for $\mu = \pi/2$. $T_{ca} \approx 0.94T_c$, $P = 6.9$ bar. The x axis represents the temperature normalized to the superfluid transition temperature in bulk ^{3}He.

at low pressures and corresponded to $a^2 = 0.73$ and $b^2 = 0.27$. This shows that the strongly distorted A phase was definitely obtained, but, taking into account possible corrections, the question about the existence of the polar phase remained open. To clarify this, we have done experiments with Obninsk-8 and Obninsk-50 samples. At low pressures near T_{ca} we have obtained that values of K are ~1.06 and ~1.07, respectively. In contrast to Ref. [19], we have created SG states in both samples and found that the shift for $\mu = \pi/2$ is negative and disappears only at T_{ca} (Fig. 1). This means that the polar phase is still not stabilized in Obninsk aerogel.

The situation is different in ^{3}He in nafen, where the polar phase is realized in a wide temperature range [Figs. 2(a) and 2(b)]. The identification of observed superfluid phases is based on the following: (i) The superfluid transition occurs into the ESP phase, because the spin susceptibility does not depend on temperature (Insert in Fig. 3). (ii) Pulse NMR experiments for different μ and β show that in both samples the spin dynamics in the ESP phase is described by Eq. (2) (Fig. 3). (iii) In nafen-243 $K \approx 4/3$ and practically is independent of temperature, as it should be in the polar phase [5] [Fig. 4(a)]. (iv) In nafen-90 $K \approx 4/3$ only in a finite range of temperatures $T_p < T < T_{ca}$ and on further cooling K decreases as expected for the polar-distorted A phase [Fig. 4(b)]. (v) Near T_{ca} values of K are nearly the same in both samples although they have essentially different densities and values of the spin diffusion anisotropy. In particular, it means that corrections to Eq. (3) due to the spin diffusion anisotropy are small. (vi) In both samples we were not able to create the SG state by the same procedure which was successful in ^{3}He in silica or in Obninsk aerogel: after all of our attempts to create the SG state the shift for $\mu = \pi/2$ was absent [Figs. 4(a) and 4(b)].

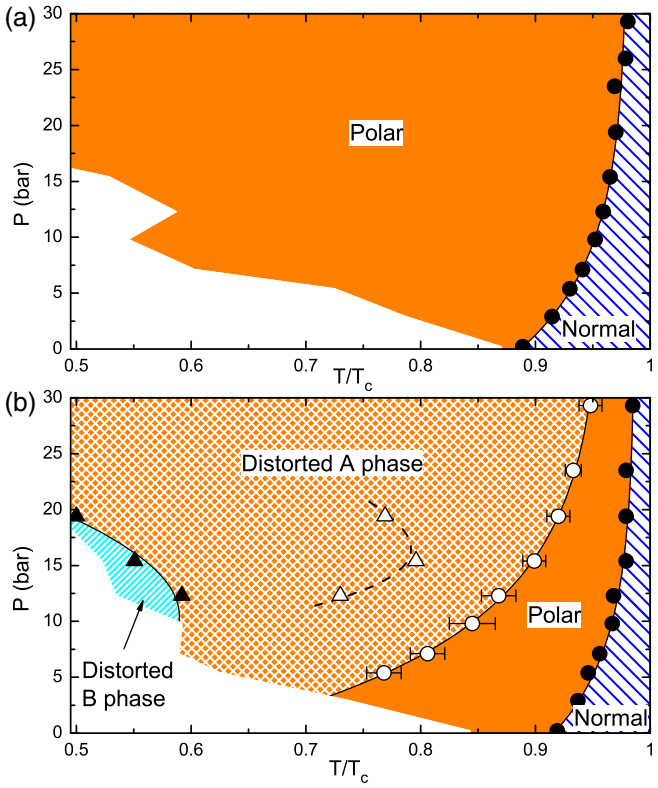

FIG. 2 (color online). Phase diagram of ^{3}He in nafen-243 (a) and in nafen-90 (b). Filled circles mark the superfluid transition of ^{3}He in nafen. Open circles mark the transition between polar and polar-distorted A phases. Filled triangles mark the beginning of the transition into the polar-distorted B phase on cooling. Open triangles mark the beginning of the transition into the distorted A phase on warming from the distorted B phase. The widths of the A-B and B-A transitions are $\sim 0.02T_{ca}$. The white area shows regions with no experimental data.

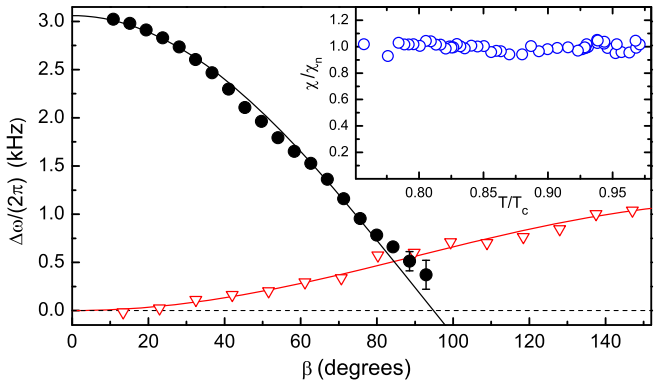

FIG. 3 (color online). Frequency shift versus β in ^{3}He in nafen-243. Filled circles: $\mu = 0$, $P = 19.4$ bar, $\omega/(2\pi) = 880.5$ kHz, $T \approx 0.78T_c$. Triangles: $\mu = \pi/2$, $P = 7.1$ bar, $\omega/(2\pi) = 359.5$ kHz, $T \approx 0.83T_c$. Solid lines correspond to Eq. (2) with $K\Omega_A^2$ measured by cw NMR. (Insert) Spin susceptibility (measured by cw NMR) normalized to the normal state value in ^{3}He in nafen-243. $\mu = 0$, $P = 7.1$ bar, $\omega/(2\pi) = 885.5$ kHz.

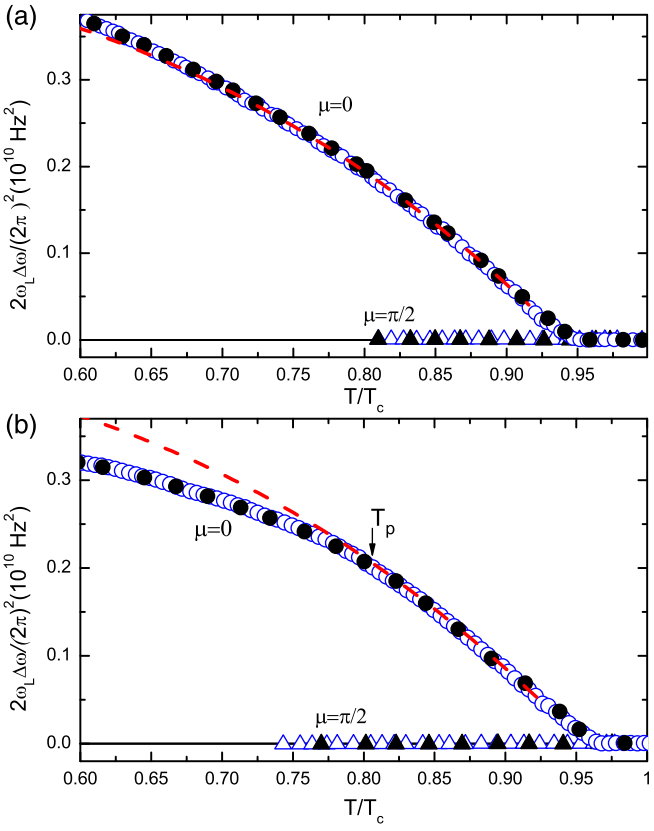

FIG. 4 (color online). Continuous wave NMR frequency shifts versus temperature in ^{3}He in nafen at $P = 7.1$ bar. Open symbols: the SN state; filled symbols: data obtained after attempts to create the SG state. $\mu = 0$ (circles), $\mu = \pi/2$ (triangles). (a) Nafen-243. $T_{ca} \approx 0.94 T_c$. The dashed line corresponds to Eq. (4) with $K = 1.245$. (b) Nafen-90. $T_{ca} \approx 0.955 T_c$. The dashed line corresponds to Eq. (4) with $K = 1.24$.

The first two of the points listed above show that in nafen we obtain one of the following phases: A, polar-distorted A, or polar. Points (iii) and (iv) exclude the A phase and points (v) and (vi) prove the existence of the pure polar phase. Thus, we conclude that in nafen-243 the polar phase exists down to the lowest attained temperatures, while in nafen-90 it exists down to $T = T_p$ where the second-order transition into the polar-distorted A phase occurs. On further cooling the distortion decreases and values of $\Delta\omega$ deflect from the curve expected for the polar phase. In Fig. 5 we present the pressure dependence of K determined in the regions of existence of the polar phase. At low pressures K is close to $4/3$, while at high pressures it is smaller by 10%–15% presumably due to corrections to Eq. (3), which grow with the increase of pressure.

In ^{3}He in nafen-90 at low enough temperatures the first-order transition into the B-like phase occurs, which is accompanied by a decrease of the susceptibility and by a change of $\Delta\omega$. Properties of this phase were not investigated in detail, but we assume, in analogy with Ref. [21], that it corresponds to the polar-distorted B phase.

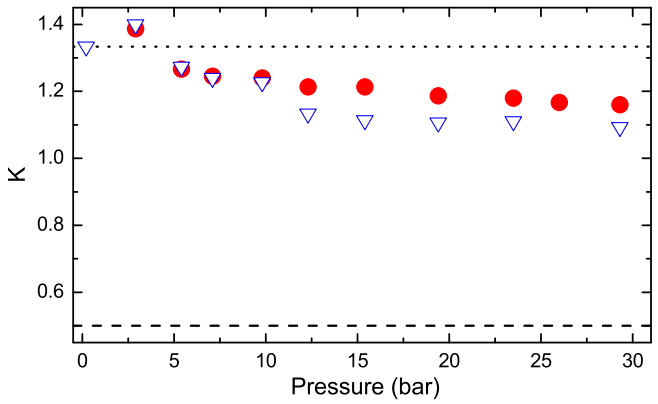

FIG. 5 (color online). K in the polar phase versus pressure. Open triangles—^{3}He in nafen-90, filled circles—^{3}He in nafen-243. Dotted and dashed lines correspond to K expected from Eq. (3) for polar and A phases, respectively. Depending on the temperature range used for determination of K, the obtained values vary by $\pm 2\%$ that limits the accuracy.

In conclusion, we have shown that in ^{3}He in nafen near T_{ca} the polar phase becomes more favorable than other phases. The polar phase is not stable in bulk ^{3}He, but it was obtained in our experiments by using nanoscale confinement. This is a new topological superfluid and new phenomena can be observed. For example, in this phase half-quantum vortices can be stable [29]. Our results also suggest that anisotropic impurities may influence the order parameter in other unconventional superfluids, e.g., in unconventional superconductors or in some quantum gases.

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