Semisuperfluidity of ^{3}He in Aerogel?

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According to hydrodynamic, acoustic, and NMR studies the superfluid transition temperature of ^{3}He in aerogel (T_c^a) is significantly suppressed with respect to that of bulk ^{3}He. We have found in the range of temperatures between T_c and T_c^a a large and unexpected NMR satellite line attributable to the liquid inside the aerogel. We propose that this anomalous behavior of liquid ^{3}He corresponds to a new type of superfluid ordering related to magnetic and possibly orbital coherence.

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One of the current issues about superfluids and superconductors is the influence of a spatial disorder on the order parameter. Liquid ^{3}He is an absolutely pure superfluid at low temperatures. However, short-range inhomogeneities can be introduced by inserting materials of high porosity into the liquid. Perhaps the most interesting material for this sort of investigation is aerogel, a silica glass, which has a structure consisting of interconnected strands. For a typical 98% aerogel the strands are 5 nm in diameter and are separated by 100 nm. The mean free path of a ballistic particle in this structure is 200 nm and during this time it will be at most 30 nm from a surface. These lengths are comparable to the relevant parameter of superfluid ^{3}He, the coherence length.

The superfluid transition of ^{3}He in aerogel has been observed by several methods. Torsion oscillator measurements [1] give a clear indication of global superfluidity of ^{3}He in aerogel over a range of pressure. In acoustic measurements superfluidity manifests itself by a mode of second sound [2], and in NMR experiments by a frequency shift of the main resonance line [3,4].

In this article we address the question of the state of ^{3}He inside aerogel in the temperature region between the superfluid transition in the bulk and the observed global superfluid transition in the aerogel. For a simple system like superfluid ^{4}He or electrons in superconductors, which exhibit only phase coherence, the suppression of superfluidity leads to a normal liquid state. For ^{3}He the answer is not so simple because not only gauge symmetry but also spin and orbit rotation symmetries are broken. Consequently, in addition to phase coherence one must consider spin and orbit coherence. As has been shown theoretically by Volovik [5], impurities can stabilize states where the phase coherence is suppressed while spin and orbit coherence is preserved. In this article we report experimental observations, which can be explained by these exotic semisuperfluid states of ^{3}He in aerogel.

We have used two samples of nominal 98% aerogel in two different experimental setups. The first setup consists of two cylindrical towers of 5 mm diameter connected to a main reservoir of ^{3}He. One of the towers was almost filled by the aerogel sample, the gap left between the wall of the cell and the aerogel being smaller than 0.2 mm. The second tower was designed to force the angular momentum vector of bulk ^{3}He-B to be nearly horizontal, thus providing a reference signal. It simply contained a 1.5 mm diameter axial solid rod. The second setup had a single experimental tower filled with aerogel. This time the gap between the aerogel and the walls was made to be 0.1 mm. Two vibrating wire resonators (13.5 μm NbTi and 89 μm Nb) enabled us to determine accurately the ^{3}He temperature between 0.2 and 100 mK [6].

Continuous wave (cw) NMR measurements were made at a frequency of about 660 kHz. We checked that heating effects were negligible at the very low NMR applied power used. As usual, the measurements were made by sweeping the magnetic field (the Larmor frequency) but the results will be presented in terms of a frequency sweep. In order to suppress any coupling to bulk liquid at the open end, the NMR signal was detected with a saddle coil that covered only the lower part of the sample tower.

The measured NMR signal is the superposition of the signals from several different sources: the bulk liquid surrounding the aerogel, the liquid confined within the aerogel, and the atomic layers of ^{3}He adsorbed on the aerogel. We find that the adsorbed solid is in thermal equilibrium with the bulk liquid even at our lowest temperatures of about 200 μK provided by a copper nuclear demagnetization stage. It is generally believed that there is fast exchange between the solid and liquid ^{3}He inside the aerogel, leading to a common precession of the sum of the two components of the magnetization at an averaged frequency,

$$\langle\omega\rangle = \frac{\langle\omega_s\rangle M_s + \langle\omega_l\rangle M_l}{M_s + M_l}, \tag{1}$$

where $\langle\omega_s\rangle$ ($\langle\omega_l\rangle$) is the average precession frequency and M_s (M_l) the magnetization of the solid (and liquid, respectively). This condition, verified previously in experiments using Mylar [7] and aerogel [3] was generally well fulfilled in our experiment, except for one very important case, that is the subject of this Letter.

In Fig. 1 we show the main results of our NMR experiments with the first sample at 17.6 and 21.5 bar pressures. The area of the main NMR line is proportional to

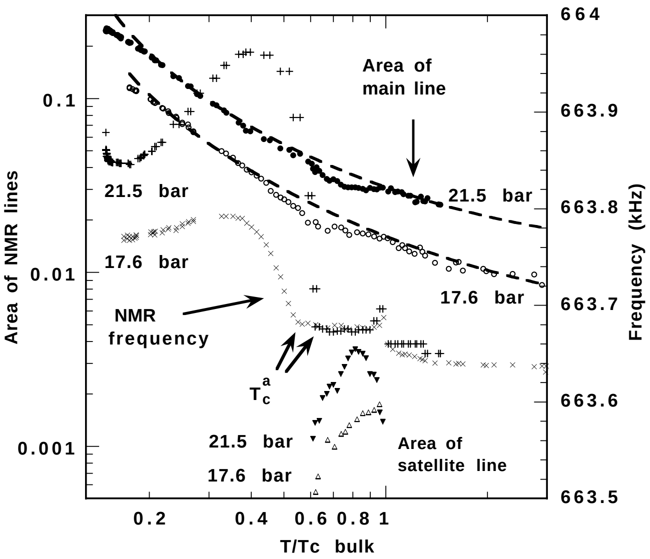

FIG. 1. The area (∘, •) and the frequency (×, +) of the main NMR line at 17.6 and 21.5 bar as a function of temperature. At the bottom is shown the area of the corresponding satellite lines. Dashed line: Curie-Weiss law.

the magnetization of the ^{3}He in the cell. Its analysis of over a wide temperature range allows a clear identification of the different contributions. At high temperatures, above 100 mK, we observe the well-known susceptibility of the Fermi liquid (out of scale of Fig. 1). At lower temperatures the magnetization of the solid increases, in accordance with the Curie-Weiss law, and becomes the major component below a few mK. At temperatures below 0.4 mK Curie-Weiss law fails to describe our results. However, the solid contribution can be described well over the entire temperature range by a 2D Heisenberg ferromagnet Hamiltonian as found for 2D solid films [8].

The dipole-dipole interaction of superfluid ^{3}He leads to a NMR frequency shift which depends on the structure and orientation of the order parameter. For superfluid ^{3}He-B the frequency shift is described by the equation

$$\Delta\omega = \cos\beta^2 \, \frac{\Omega_L^2}{2\omega_0}, \tag{2}$$

where Ω_L is the Leggett frequency which characterizes the dipole-dipole interaction, ω_0 is the Larmor frequency, and β describes the orientation of the order parameter $\vec{n}$ with respect to the external magnetic field. In the bulk, near the walls of the cell, the orientation of order parameter leads to a large frequency shift. Usually the order parameter forms a spatial texture which determines the NMR line shape observed in bulk ^{3}He-B.

It was found that below the superfluid transition inside the aerogel liquid ^{3}He has a frequency shift, which leads to a frequency shift of the common liquid-solid precession, as shown in Fig. 1. The onset temperature of this frequency shift T_c^a was found to be in good agreement with torsion and acoustic measurements of the superfluid transition temperature in aerogel. Our experiments show the same behavior of the main NMR line. With the second aerogel sample we find superfluid transition temperatures in excellent agreement with previous experiments [3] performed above 12 bars. We have observed also transitions at 900 μK (8 bars) and at 300 μK (5 bars). We did not find any transition at zero pressure with temperatures down to 200 μK. In the first sample we find that the transition temperatures T_c^a are systematically shifted to lower temperatures. We have observed transitions at 1.4 mK for 21.5 bars, 1.18 mK for 17.6 bars, and 760 μK for 12.5 bars. We have not seen any transition down to 300 μK for 8 and 5.4 bars.

With further cooling, the magnetization of solid ^{3}He increases, thus causing a decrease of the common frequency shift, as shown in Fig. 1. At a temperature of $0.15T_c$ at 21.5 bars we have observed an unexpected growth of the frequency shift. This last observation will be discussed in other publications.

In addition to the main NMR line, we see a satellite signal that appears at a temperature near T_c of bulk and moves away from the main resonance with decreasing temperature. The origin of this signal is puzzling. At first glance this signal could be due to bulk ^{3}He surrounding the aerogel sample. We were able to compare this signal with that received from the empty cell of the same dimensions, but with a solid rod in its axis, described above. The cell was filled by bulk ^{3}He. The NMR lines from both cells shown in Fig. 2 have been measured at exactly the same temperature using the same settings of the spectrometer. The signal from the cell with the rod can be explained well by a bent texture [9]. Its high frequency limit corresponds to a perpendicular orientation of the vector $\vec{L}$ with respect to the vertical walls and the magnetic field. The signal from the

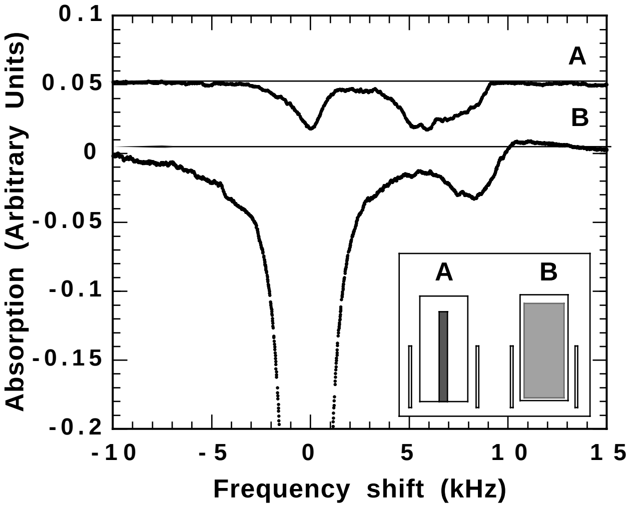

FIG. 2. The NMR satellite line measured in bulk ^{3}He at 17.6 bar and $0.8T_c$ in (A) the cell with an axial rod, compared to (B) the cell filled with aerogel.

aerogel cell shows about the same frequency shift, but a different form of broadening. To our surprise, the amplitude of the satellite lines from the two cells is comparable. One should remember that one of the signals is received from bulk ^{3}He filling up the entire cell, while the other originates from the cell with aerogel, where bulk superfluid ^{3}He exists only in a narrow gap between the wall and the aerogel sample. In Fig. 3 are shown the satellite lines at different temperatures. In all cases the frequency shift is smaller, but comparable with that for ^{3}He-B with $\vec{L}$ perpendicular to $\vec{H}$.

If indeed the satellite peak has its origin in the bulk ^{3}He contained between the cell's walls and the aerogel, then its magnitude should simply be proportional to the susceptibility of the B-phase superfluid and the volume. Given the geometry of the cell, we can estimate the maximum volume of bulk liquid contained between the aerogel sample and the walls in the region seen by the NMR coils to be between 5 and 15% of the total volume of the cell. The magnetization of the measured satellite line, which varies with both temperature and pressure, can be compared to that tabulated for superfluid ^{3}He-B, normalized by the NMR signal measured in our setup above T_c. We show in Fig. 4 the magnitude of the satellite signal as a percentage of the signal expected from the total amount of liquid ^{3}He inside the cell. This value is found to be as large as 50%.

Such a large value of the satellite signal simply cannot be explained by the bulk ^{3}He surrounding the aerogel. Therefore, we have to consider the possibility that it originates from the ^{3}He liquid confined within the aerogel. This conclusion is further supported by measurements of the area of the main NMR line, shown in Fig. 1. We observe that the magnetization of the main line begins to deviate from the Curie-Weiss law below the bulk transition T_c. The deficit in magnetization roughly corresponds to the magnetization of the satellite line. At around T_c^a the amplitude of the satellite line decreases, as seen in Fig. 1. At a lower temperature this magnetization returns to the main line. Since bulk ^{3}He outside the aerogel does not change in this region of temperature, this observation supports the conclusion that the satellite signal is emitted by ^{3}He confined within the aerogel. These results have been confirmed by the experiments made with the second sample.

Our measurements show that ^{3}He inside aerogel in the region of temperatures between T_c and T_c^a is not in a simple normal state. Keeping this in mind we have reexamined the results of previous experiments. The observation of a satellite NMR line of intensity of about 12% of liquid has been mentioned in [10]. In a recent discussion of this experiment we have learned that adding some amount of ^{4}He had increased the amplitude of the satellite line up to 24% [11]. Since ^{4}He does not change any bulk parameter, this observation definitely supports our conclusion that the satellite NMR signal is emitted from ^{3}He confined within the aerogel. Acoustic measurements [2] also show a signal in the region between T_c and T_c^a, which has been attributed, without quantitative analysis, to the influence of external ^{3}He. An unexpected anisotropy of the NMR signal from aerogel at the same temperatures was reported [4]. Our work suggests that this anisotropy is the result of a texture inside the aerogel, the orientation of which is determined by the cell wall.

As a first tentative explanation of satellite signals we considered a proximity effect between the bulk superfluid and the "dirty" ^{3}He contained inside the aerogel. In order to explain the amplitude of the satellite line, we should suppose that all the ^{3}He magnetization (liquid and solid) on a distance of about 0.1 mm inside the aerogel precesses at a frequency related to the order parameter of the external superfluid ^{3}He. This would mean a giant penetration, over

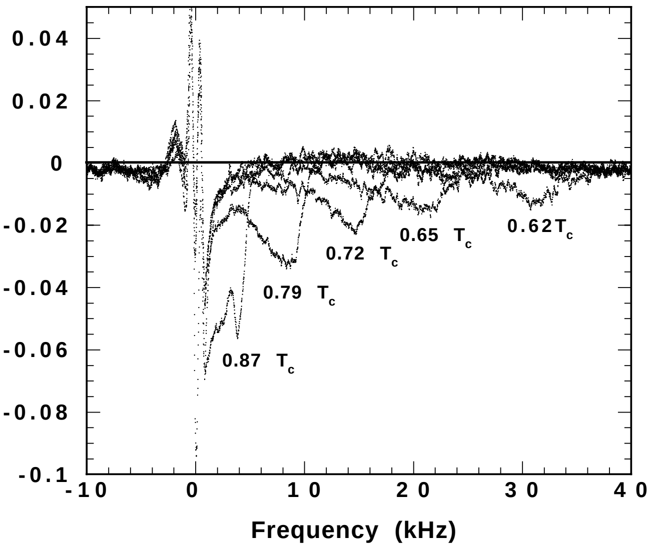

FIG. 3. The satellite lines in aerogel at 21.5 bar. The main line has been subtracted.

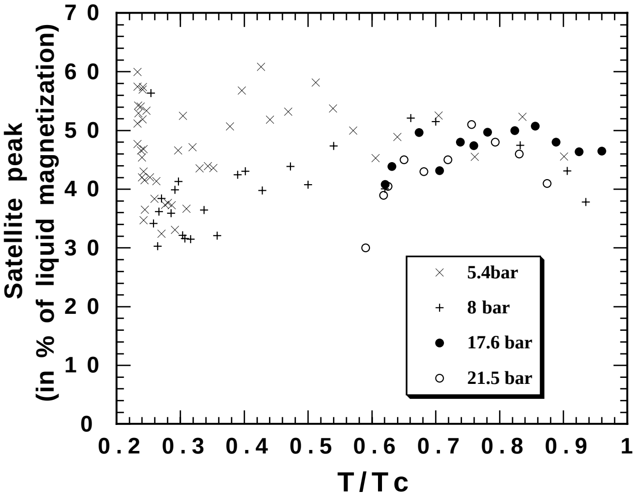

FIG. 4. The magnetization of the satellite signal in units of that of an equal amount of liquid ^{3}He-B filling the cell.

1000 coherence lengths. However, this hypothesis can be applied in our case only if the satellite line frequency corresponds to the averaged frequency of liquid and solid magnetization precession, as described by Eq. (1). This should lead to a broadening of the main part of the satellite line down to the main line frequency and cannot explain our relatively sharp satellite line.

If we assume that the aerogel sample has relatively large voids where the liquid ^{3}He is in a local superfluid B state and its precession is decoupled from the solid one, then the volume of these pores should be as large as about 30% of the aerogel volume, which is unlikely. But if it is a case, then the orientation of the order parameter in these pores would be correlated with that of the superfluid ^{3}He outside of the aerogel. In other words, spin and maybe orbital supercurrents transport the texture through the aerogel derby, while the global phase coherence is suppressed. Consequently, ^{3}He inside the aerogel should be in a magnetically ordered and in a liquid crystal state.

It is more likely that the satellite signal is emitted by the liquid ^{3}He confined within the aerogel. Clearly, it is difficult to explain the existence of satellite lines without invoking a substantial modification of the state of ^{3}He in aerogel. The state of ^{3}He in the region of temperature considered here may be very unusual. Generally speaking, there are many interesting exotic states related to the p-wave pairing of Cooper pairs. It was Volovik [5], who suggested that impurities tend to destroy the anisotropic correlation of the order parameter $\langle A_{\alpha i}\rangle$, while the correlations of the order parameter, which in principle are more symmetric than the order parameter itself, may still survive. These correlations correspond to superfluidity with four-particle correlated states $\langle A_{\alpha i}A_{\alpha i}\rangle$, $e_{\alpha\beta\gamma}\langle A_{\alpha i}A_{\beta i}\rangle$ (state with spin anisotropy), $e_{ijk}\langle A_{\alpha i}A_{\alpha j}\rangle$ (state with orbital anisotropy), as well nonsuperfluid states with magnetic or/and orbital ordering: $e_{\alpha\beta\gamma}\langle A^*_{\alpha i}, A_{\beta i}\rangle$, $e_{ijk}\langle A_{\alpha i}A^*_{\alpha j}\rangle$, and $e_{ijk}e_{\alpha\beta\gamma}\langle A^*_{\alpha i}A_{\beta j}\rangle$. Some of these states are natural candidates for ^{3}He ordering in the conditions considered here.

The existence of a ^{3}He state with spin and maybe orbit coherence should have important consequences on the spin dynamics. Liquid ^{3}He undergoes an antiferromagneticlike ordering, characterized by two NMR branches. In bulk superfluid ^{3}He the longitudinal branch of NMR has a frequency of about Ω_L. What should be with this branch owing to the interaction with the highly magnetized surface solid ^{3}He? As a tentative hypothesis we can suggest that the satellite signal is the signature of the second branch of liquid NMR in the conditions of strong interaction with magnetized solid ^{3}He.

The state considered here would have similar characteristics to ^{3}He-A modified by the influence of the aerogel; in particular, we would expect the mass dynamics to have similar dissipative properties. Thus a torsion oscillator experiment may fail to detect the superfluid signal. The transition seen previously by torsion oscillator experiments as well as by the NMR frequency shift of the main line would then be the analog of the A-B transition in the bulk. In support of this hypothesis, we can mention that there is a similarity between the transition observed in aerogel and the A-B transition in bulk at high magnetic field. The surface of the aerogel may play a role similar to a magnetic field, which shifts the A-B transition to lower temperatures.

In conclusion, in this article we report for the first time the unusual properties of ^{3}He in aerogel in the region of temperature between T_c in the bulk and T_c^a in aerogel, which we named "semisuperfluidity." This phenomenon is characterized by formation of a satellite NMR line which cannot be explained by external bulk ^{3}He. This article can be considered as a call for theorists to investigate in more detail the possible magnetic and orbital states of ^{3}He inside aerogel.

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