Equal-spin-pairing superfluid phase of ^{3}He in an aerogel acting as an impurity

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The superfluid phases of ^{3}He in a 97.5% porosity aerogel have been studied using cw and pulsed NMR techniques at several pressures. The equilibrium equal-spin-paring superfluid phase, *A*-like phase region is observed very near T_c^{aero}, and that region normalized with T_c^{aero} is found to be independent of pressure in contrast with that of the bulk *A* phase. Tipping angle dependent frequencies in the free induction decay signal of the *A*-like phase are quite different from those of the bulk *A* phase. On these grounds, we propose that the *A*-like phase in aerogel is a different superfluid phase and can be described as a robust state introduced by Fomin [JETP Lett. **77**, 240 (2003)].

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The effect of impurity scattering is a central and very interesting problem in condensed matter; yet, few systems are available where the phenomenon can be studied in detail. Superfluid ^{3}He is an ideal candidate for such a study because the bulk liquid properties are well understood. Since the superfluid transition is at millikelvin temperatures, it exhibits the purest BCS type of coherent wave function with a spin triplet *p*-wave Cooper pair state, making the analysis relatively straightforward. Impurity scattering can easily be generated by immersing aerogel, a highly porous lattice of silica strands, within the fluid. The aerogel has tipically high open volume fraction of more than 98%: the aerogel strands occupy with only 2% of the volume fraction. The strands are a few nanometers in diameter, much smaller than the superfluid coherence length ξ_0, which varies from 65 nm at zero pressure to 13 nm at the melting pressure. The mean distance between the strands is comparable to ξ_0 with a distribution over the range from a few to a few hundred nanometers. A number of experiments on this system using more than 98% porosity aerogel have shown that the superfluid transition temperature T_c^{aero}, and the superfluid density are suppressed as expected for impurity scattering.[1–4] The T_c^{aero} suppression and the magnetization are explained by the isotropic inhomogeneous scattering model[5] and magnetic response of low energy excitation.[6] However, the detailed nature of the superfluid phases within the aerogel has not yet been determined. So far, only a few studies have been done using lower porosity aerogel: higher concentration of impurities.[7] Interest is aroused how superfluid phases of ^{3}He is affected in dense impurities.

For magnetic fields below 3 T, an *A*-like and a *B*-like states have been observed, similar to the *A* and *B* states of the bulk. Earlier NMR experiments[1,3,8,9,14] found that the *A*-like phase is an equal-spin-pairing (ESP) state whose magnetization is nearly equal to that of the normal phase. The *B*-like phase was found to be a non-ESP state with a magnetization that falls with temperature. At the first order phase transition between the *A*-like and the *B*-like phase, the *AB* coexisting phenomena were observed.[9–13] Results from tipping-angle-dependent frequency measurements via free induction decay (FID), signals, the homogeneous precession domain,[14] and superfluid flow measurements during rotation[15] suggested that the *B*-like phase could be explained using the same order parameter as the bulk *B* phase. Recently, acoustic experiments in 98% aerogel[12] suggest that the *A*-like phase has a different order parameter than the bulk, and NMR experiments[16] indicate that it may be a mixed state with two different spin states. Thus, it appears that the order parameter in the *A*-like phase has not yet been conclusively identified. A "robust state"[17] with the orientation of the order parameter insensitive to the presence of the aerogel and an Anderson-Brinkman-Morel (ABM) state with high spatial randomness[18] have been proposed. In this Brief Report, we report studies of the superfluid state of ^{3}He in 97.5% aerogel using cw and pulsed NMR techniques at several pressures. The results suggest that the *A*-like phase corresponds to the robust state.[17]

The small angle neutron scattering experiments found that differential cross section in the 97.5% aerogel has one broad peak as a function of wave number.[13] This means that the aerogel has a strongly correlated structure and the aerogel strands are distributed inhomogeneously. The corresponding correlated length is 53.6 nm.[19] We used the aerogel directly made in a glass cylinder with 4 mm in diameter without thin bulk space between the aerogel and the wall of cylinder. As a result, any signals from the bulk have not been observed. This aerogel is the same batch used in the above structural analysis. We have performed cw NMR experiments at the operating frequency of 923 kHz at pressures of 1.0, 1.3, 1.6, 1.9, 2.4, and 3.2 MPa. The aerogel strands were coated with about 2.5 layers of ^{4}He thin film in advance to eliminate the surface magnetization of ^{3}He. We did not observe any surface solid ^{3}He contributions except at the highest pressure.[20] Here, we argue about superfluid phases of ^{3}He in the aerogel using mainly the results at 2.4 MPa without any solid contributions. The NMR static magnetic field of 28.4 mT was parallel to the cylinder axis. We have also measured tipping angle dependence of frequency in the FID signal of both the *A*-like and the *B*-like phases using the pulsed NMR method at 2.4 and 3.2 MPa. Tipping angle β was calibrated with the

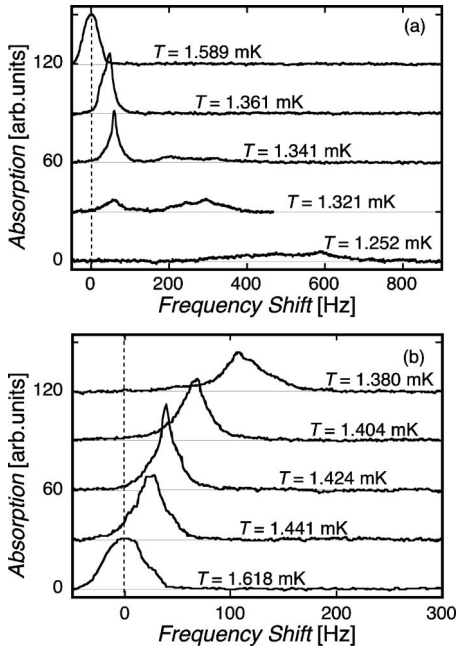

FIG. 1. NMR spectra of ^{3}He in the 97.5% aerogel at 2.4 MPa (a) on cooling and (b) on warming at several temperatures as a function of frequency shift from the Larmor frequency. Note that (a) and (b) are represented with different frequency scales.

normal phase in the aerogel. We used a Pt-NMR thermometer immersed in the liquid with the Greywall temperature scale.[21]

At all pressures, the superfluid phase transitions were observed showing the changes of NMR signal shape and resonance frequency. Figure 1(a) shows typical NMR spectra at 2.4 MPa on cooling from the normal phase. The superfluid phase transition to the *A*-like phase occurred at T_c^{aero} $=1.460$ mK ($T_c^{bulk}=2.339$ mK). In the *A*-like phase, the resonance peak always showed positive frequency shift from the Larmor frequency. The magnetization of the *A*-like phase was nearly equal to that of the normal phase. On further cooling, the *B*-like phase appeared at $0.921T_c^{aero}$, with the large positive frequency shift and the total magnetization decreased through the *AB* coexisting state. On cooling, the phase conversion from the *A*-like to the *B*-like phase progressed continuously and the coexisting state remained down to $0.892T_c^{aero}$. As shown in Fig. 1, NMR signals from the two phases are clearly separated at low temperature. Thus, we can trace each spectrum as a function of temperature and can estimate directly each fraction of the two phases by integrating the area under NMR spectra. We refer to this *AB* coexisting temperature region as T_{low}^{AB} band. We determine that the transition temperature to the *B*-like phase on cooling $T_{AB,c}^{aero}$ is $0.921T_c^{aero}$. Only the *B*-like phase existed below T_{low}^{AB} band. The characteristic features mentioned above were observed at all pressures. The coexisting state at 1.0 MPa, however, remained even at the lowest temperature of 0.320 mK.

NMR spectra on warming from 1.20 mK are shown in Fig. 1(b). No *A*-like phase appears on passing through T_{low}^{AB} band. As increasing temperature, the peak frequency of the *B*-like phase approached the Larmor frequency and the linewidth became narrower. The width was the narrowest at $0.975T_c^{aero}$. On further warming to T_c^{aero}, the spectra became broad. With cooling it again from any temperature between $0.975T_c^{aero}$ and T_c^{aero}, the *AB* coexisting state with separated signals appeared clearly. We call this a turn-around experiment and refer to this temperature region as T_{high}^{AB} band. The broad signal in T_{high}^{AB} band is due to the superposition of resonance signals from two phases. We determine that the transition temperature to the *A*-like phases on warming $T_{AB,w}^{aero}$ is $0.975T_c^{aero}$ at which the *A*-like phase first appears. At all pressures with warming, we observed narrowing the NMR signal of the *B*-like phase followed by broadening the signal near T_c^{aero}, indicating the appearance of the *A*-like phase. The phase conversion process in the two bands is very interesting, because the first order phase transition in normal materials occurs at the transition temperature not in the finite temperature width.[22] These phenomena are related to a pinning mechanism of phase boundary in coexisting states inside aerogel.[10,23] In previous experiments, the *AB* coexisting state was observed on only cooling process[9,12] or both on cooling and on warming process.[10,11] This discrepancy can be caused by the difference of the internal structure of aerogel. It should be noted that no *AB* coexisting state was observed without ^{4}He coating.[19] Moscow group reported that the coexisting state was more stable in terms of time in aerogel coated with thin ^{4}He film than in that with surface solid ^{3}He.[9] These suggest that the first order phase transition between the *A*-like and the *B*-like phase may be influenced by not only the microscopic structure of the aerogel but also the surface solid ^{3}He on the aerogel strands.

The *A*-like phase often shows the positive frequency shift[1,10,15] like our results but sometimes shows the negative frequency shift on cooling.[3,9] Two theoretical models giving only positive frequency shift[18,24] explains the former results. An angular momentum vector may be pinned by temporary boundaries (defects) due to independent local superfluid nucleations on cooling, which may give rise to the negative shifts,[25] but there is no clear explanation for pinning mechanism and for texture of angular momentum. The signals of the *B*-like phase were quite different from previous ones showing that the main peak was almost at the Larmor frequency.[3,8,9,15] Those spectra can be explained with the spatial distribution of **n** vector (the order parameter of the bulk *B* phase) like the flare-out texture in the bulk liquid where most of **n** vectors except at the edge part are aligned parallel to the magnetic field by the field orientation energy.[26] On the other hand, the positive frequency shifts in Fig. 1 show that most of **n** vectors have the finite angle with respect to the magnetic field. Such a configuration can be given by the surface field energy[26] and a longer healing length of **n** texture than that in the bulk liquid, indicating the small field orientation energy in the aerogel.

The bulk *A* phase exists above the polycritical point (PCP) pressure of 2.15 MPa in zero magnetic field and the region of thermal equilibrium *A* phase depends on the pressure. By applying a magnetic field, the PCP no longer exists and the *A* phase region spreads and appears even at lower pressures near the superfluid transition temperature.[27] It has been reported that the equilibrium *AB* transition temperature

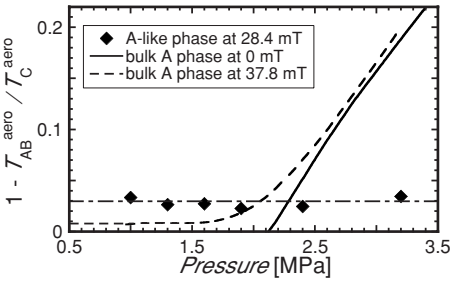

FIG. 2. Comparison of the temperature region of the A-like phase with that of the bulk A phase (Ref. 27) as a function of pressure. Solid and broken lines show equilibrium AB transition of the bulk liquid without and with magnetic fields, respectively. The dash-dot line is a guide for the eyes.

in a 99.3% aerogel was very close to $T_{AB,w}^{aero}$.[28] In Fig. 2, we plot the $T_{AB,w}^{aero}$, by the reduced temperature of $1-T_{AB,w}^{aero}/T_c^{aero}$, as a function of pressure.[29] Those of the equilibrium bulk A phase are also shown in Fig. 2 by $1-T_{AB}/T_c$ with solid and broken lines, below which the bulk A phase exists. We found that the reduced region of the A-like phase is nearly independent of pressure in contrast with that of the bulk A phase. In the 99.3% aerogel, the A-like region was suppressed but showed weak pressure dependence.[30] This difference can be due to the concentration of impurities, high concentration of impurities reduces the strong coupling effect which stabilizes the ESP state more than low concentration does even at high pressure. Here, it is very important that the A-like phase is strongly suppressed very near T_c^{aero} at higher pressures, where the bulk A phase has large stable region. Accordingly, we think that the A-like phase must be a different superfluid state from the bulk A phase.

It is well understood in the bulk A phase that frequencies in the FID signal depends on β given by Leggett,[31]

$$\Delta f(\beta) = \Delta f(0)(1 + 3\cos\beta)/4, \tag{1}$$

where $\Delta f(\beta)$ represents the frequency shift from the Larmor frequency, $\Delta f(0)$ is the frequency shift in cw NMR experiments. This is a unique feature in superfluid ^{3}He as a result of broken spin-orbit symmetry. In Fig. 3, we illustrate frequency shifts in the FID signal of the A-like phase at 2.4 MPa, 1.420 mK as a function of β. Frequencies of the A-like phase show the tipping angle dependence a bit different from that of the bulk A phase of Eq. (1) represented with broken curve. This provides another evidence that the A-like phase is not the bulk A phase. The frequency shifts are found to be well explained by theoretical calculation using the order parameter of the robust state proposed by Fomin.[17] Based on the robust state, Miura *et al.*[24] gives

$$\Delta f(\beta) = \Delta f(0)(1 + \cos\beta)/2, \tag{2}$$

shown with solid curve in Fig. 3. The tipping-angle-dependent frequencies at 3.2 MPa were also explained with Eq. (2).[20] Recently, Volovik obtained the same tipping angle dependence $(1+\cos\beta)$ of frequency shift as Eq. (2) for the

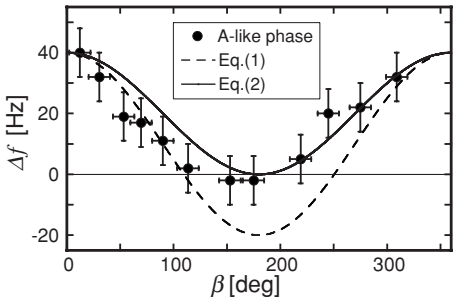

FIG. 3. Δf, frequency shifts from the Larmor frequency in the FID signal of the A-like phase at $T=1.420$ mK, and $p=2.4$ MPa as a function of tipping angle β. The broken curve and the solid curve are theoretical results based on the bulk A phase (Ref. 31) and the robust state (Ref. 24), respectively.

ABM state in the presence of random anisotropy disorder.[18] So we cannot conclude which state is the A-like phase, a robust state, or the ABM state from only the observation of the tipping angle dependence of frequencies. However, it is clear that the A-like phase does not show the bulk A-phase behavior in pulsed NMR experiments. That the equilibrium A-like phase exists at the vicinity of T_c^{aero}, as shown in Fig. 2, is consistent with the theory based on the robust state.[17,32] The above two observations suggest that the A-like phase can be described with the robust state. The tipping angle dependence of frequencies of the A-like phase qualitatively agrees with pulsed NMR results in 98% aerogel below $\beta=140°$.[16]

We also plot frequencies in the FID signal of the B-like phase at 0.93 mK in Fig. 4. It was found that frequencies have tipping angle dependence and they are fairly similar to those of the bulk B phase confined between a pair of parallel plates.[33] The **n** vector between them was in the non-Leggett configuration uniformly and had the finite angle with respect to the magnetic field, giving the large positive frequency shift in cw NMR measurements. Frequencies in the FID after an rf pulse depended on tipping angles such that they approached the Larmor frequency with increasing β and were almost equal to the Larmor frequency above a characteristic tipping angle. Such behaviors were explained by a numerical calculation using the order parameter of the B phase with intrinsic relaxation mechanism and non-Leggett configuration as an initial state.[33] These behaviors are also seen in Fig.

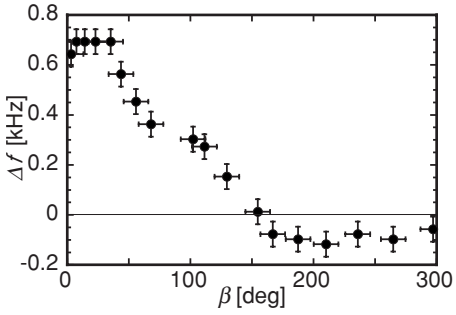

FIG. 4. Δf, frequency shifts from the Larmor frequency in the FID signal of the B-like phase at $T=0.93$ mK, and $p=2.4$ MPa as a function of tipping angle β.

4 except a little negative shift at large tipping angles.[34] So pulsed NMR frequencies on the *B*-like phase seem to be explained qualitatively by using the *B* phase order parameter as earlier NMR experiments.[8,14,15] The non-Leggett configuration also explains the positive frequency shift of the *B*-like phase in Fig. 1.

In conclusion, we have studied superfluid phases of ^{3}He by NMR methods in the 97.5% aerogel coating with ^{4}He thin film. The *A*-like phase on warming is observed very close to T_c^{aero} at all pressures and the reduced region is nearly independent of pressure. Tipping angle dependence of frequencies in the FID of the *A*-like phase differs significantly from that of the bulk *A* phase. We see that the robust state proposed by Fomin explains last two properties of the *A*-like phase quite well. On the other hand, the order parameter of the *B*-like phase seems to be same as in the bulk *B* phase.

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