Phase-Transition Phenomena of 0.8 μm Superfluid ^{3}He Slab

Kenji Kawasaki, Tomohiro Yoshida, Miki Tarui, Hisashi Nakagawa, Hideo Yano, Osamu Ishikawa, and Tohru Hata

Graduate School of Science, Osaka City University, Osaka 558-8585, Japan

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We have investigated the transition phenomena of superfluid ^{3}He in thin 0.8 μm slabs with a cw-NMR method. We found that, just below the phase-transition temperature, only the A phase appeared at any pressure. At lower temperatures, the phase transition to the B phase occurred between 0.3 and 2.74 MPa. We obtained a universal critical thickness δ as a function of pressure. When the reduced slab thickness, $d/\xi(T)$, is smaller than δ, only the A phase becomes stable.

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Superfluid ^{3}He is understood as a p-wave BCS-type condensation. A characteristic length of such condensation is the coherence length $\xi(T)$, which diverges as $(1 - T/T_c)^{-1/2}$ near the superfluid transition temperature T_c, and $\xi(T)$ at $T = 0$ K is about 10 nm at 3.4 MPa and 50 nm at 0 MPa. It is thought that the long coherence length will cause the suppressions of both the superfluid transition temperature and the superfluid density in a restricted geometry or in a thin slab [1–3]. The suppression of T_c has been actually observed in thin films and in narrow pores [4,5], while the suppression of superfluid density has been observed in porous media [6].

In the slab geometry consisting of parallel plates, the reduction of the A-B phase-transition temperature was observed [7,8]. Such a stabilization of A phase was studied theoretically [2,3,9,10]. Two points are crucial for understanding. One is that the Cooper pairs of ^{3}He atoms form a constraint that their angular momentum should be perpendicular to the boundary surface. The other point is that the free energy is increased by the spatial variation of the order parameter occurring near the surface. For the A phase, this constraint does not change the condensation energy since the energy gap of the A phase has a node along the angular momentum. For the B phase with the isotropic energy gap, however, this constraint gives rise to the loss of condensation energy. So the A phase will be stable in a narrow slab geometry in the long coherence length region. Therefore, it has been highly desired to find what size of slab geometry will favor the A phase even at $T = 0$ K.

In 0.3 μm slab geometry, only the A phase was observed by the nuclear magnetic resonance (NMR) method even at 0.30 mK [11], which was inconsistent with the theoretical speculation for this thickness [3]. It was thought that the supercooling phenomena occurred. Recently, A-B transitions at 1.0, 2.4, and 2.7 MPa have been observed in a 1.1 μm slab geometry [8]; however, the observed A-B transition temperatures were slightly lower than the expected ones [12]. In this Letter, we report the measurement of A-B transition in 0.8 μm slab geometry. We derive a universal critical thickness as a condition of occurring A-B transition and also discuss the effect of ^{4}He coating on the phase transition.

To make the sample cell, we use nominal 0.825 μm diameter polystyrene spheres as spacers, whose spheres are dispersed on 12 μm thick polyethylene sheets [13]. The surface density of spheres on the sheets is nonuniform; some spheres aggregated together, resulting in sheet surfaces being studded with numerous aggregations, with the mean distance between them ranging from 10 to 100 μm. By stacking 400 sheets we made a 0.8 μm cell whose spacing was 0.808 ± 0.038 μm. These were measured by an electron probe microanalyzer. Because the filling factor of liquid ^{3}He was small, we adopted the resonance circuit with a high Q value [14]. By using a low temperature amplifier together with the superconducting wire as an NMR coil, the measured Q value was 11 000 in the millikelvin region. In this work, the NMR static field was 22.4 mT and the Larmor frequency was 717 kHz. The NMR coil was wound with a niobium wire of 76 μm diameter. The broad line width of about 2.5 kHz in the normal phase was due to these superconducting wires.

The NMR magnetic field was applied perpendicular to the sheet surface. In this situation, the $\boldsymbol{l}$ vector in the A phase is expected to be parallel to the surface normal of sheets and perpendicular to the $\boldsymbol{d}$ vector since the dipole healing length is much longer than 0.808 μm and the magnetic field is larger than the dipole field of a few mT. For this textural state, the negative shift of resonance frequency was expected by S. Takagi [15] and was actually observed in a thin slab as a fingerprint of the A phase [7,11]. For the B phase in this situation, the $\boldsymbol{n}$ vector is expected to be parallel to the surface normal and parallel to the magnetic field, where no frequency shift is expected. In confined geometries, it is well known that both surface solid and liquid magnetizations show the single absorption in NMR experiments due to the rapid spin exchange mechanism [8,11].

Figure 1 shows that the peak frequency and the magnetization of the broad absorption signal as a function of temperature at 2.74 MPa. Here the magnetization is obtained by integrating each absorption signal and subtract-

ing the Lorentzian shaped signal coming from outside of the cell, whose frequency stayed at Larmor frequency at any temperature [16]. With cooling to 1.5 mK, the peak frequency shifted gradually to the lower frequency side below 2.35 mK. Below 1.5 mK, it turned upward and became almost constant below 0.8 mK. The magnetization was almost constant down to 1.5 mK then abruptly decreased. Below 1.3 mK, the magnetization increased. On warming up, data show a hysteresis, where the magnetization during warming was smaller than that of cooling and coincided with that of cooling above 1.8 mK, and the peak frequencies during warming were higher than those during cooling. Above 1.8 mK, frequencies are the same in both processes. It is clear that the phase transition from normal phase to superfluid A phase occurred at 2.35 mK based on the behavior of the negative frequency shift and the constant magnetization.

In the previous experiment [8], the gradual increase of the magnetization at lower temperatures was observed with pure ^{3}He and almost disappeared by precoating 2.5 layers of ^{4}He. This means the gradual increase of the magnetization is attributed to the Curie-Weiss behavior of the surface solid ^{3}He on polyethylene sheets. In this experiment we also found that such an increase at low temperatures disappeared when a small amount of ^{4}He was precoated on the sheet. The surface solid magnetization may cause a small and gradual frequency shift in an absorption signal [11] but its frequency does not change, as shown in Fig. 1. Therefore, the abrupt changes in magnetization and in frequency slope in Fig. 1 are attributed to the phase transition in the superfluid state. The decrease in the magnetization implies that the lower temperature phase is the B phase. The cause of the hysteresis loop is explained as follows. It is thought that a part of A phase changed to the B phase at 1.5 mK in cooling. On further cooling, the gradual change from the remaining A phase to the B phase occurred; the A phase, however, still remained at the lowest temperatures. The existence of the larger amount of B phase on warming can explain not only a smaller magnetization but also a higher frequency than those in cooling down process qualitatively. The remaining A phase might be in the supercooled state. No observation of the discontinuous change in resonance frequency at the A-B phase transition is attributed to the coexistence of A phase and B phase. We maintained the temperature at 1.3 mK for two days at 2.74 MPa to see whether the hysteresis behavior would disappear and we observed no change. The existence of such hysteresis is also the evidence of the first order phase transition between A and B phase.

Resonance frequencies at several pressures are shown in Fig. 2 as a function of temperature. We defined the A-B transition temperature at which the B phase fraction disappeared in the warming up process. The obtained phase-transition temperatures are 1.79, 1.76, 1.78, 1.32, 0.95, and 0.7 mK at 2.74, 2.45, 1.87, 1.0, 0.5, and 0.3 MPa, respectively. Frequencies at 0.14 MPa in Fig. 2 did not show any rapid bend down to 0.33 mK, which meant only the A phase existed at such a low pressure. However, it will be explained below that this behavior may be the supercooling phenomena. The superfluid transition temperatures T_c were slightly suppressed at all pressures for pure liquid ^{3}He. The suppression was about 130 μK at lower pressures and 40 μK at higher pressures. This pressure dependence is explained qualitatively with the coherence length but the observed suppressions are a bit larger than the expected ones [1].

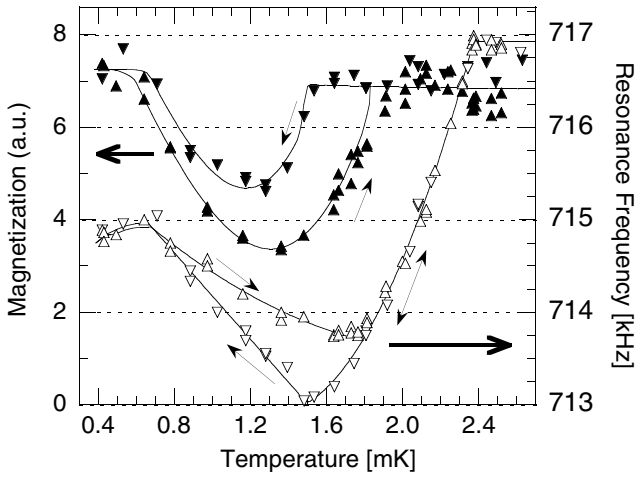

FIG. 1. Magnetization and resonance frequencies of cw-NMR at 2.74 MPa. Open inverse triangles and filled ones are values of the resonance frequency and the magnetization, respectively, during the cooling down process. Similarly, normal triangles are data during the warming up process. Solid lines are guides for the eye.

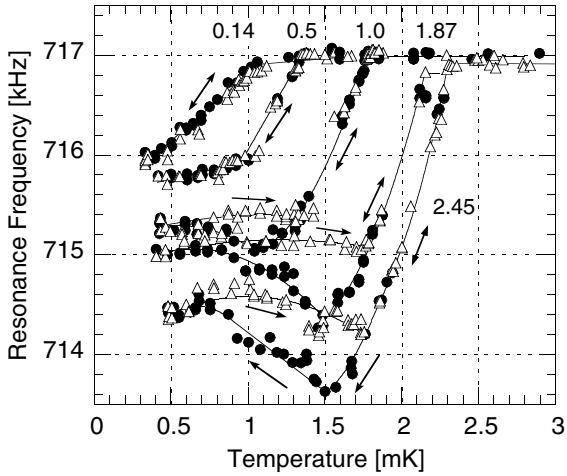

FIG. 2. Resonance frequency as a function of temperature. Solid circles are the resonance frequencies during cooling down and open triangles are frequencies during warming up. Figures indicate pressures in MPa unit. Solid lines are guides for the eye.

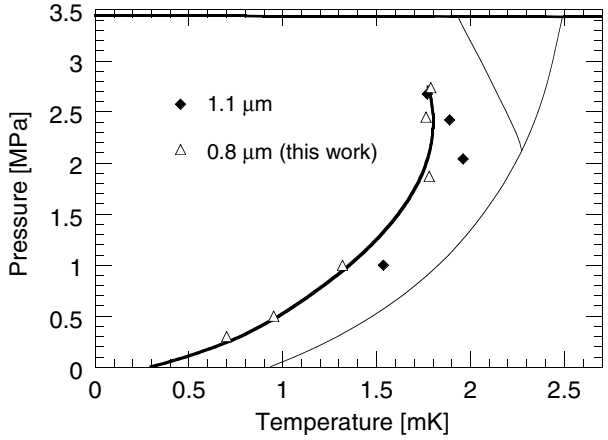

FIG. 3. Phase diagram of the *A-B* transition in slab geometries. Thin solid curves are *A-B* transition and superfluid transition in bulk liquid. Diamonds show the *A-B* transition in 1.1 μm [8] and triangles show the temperatures for completion of the *B* to *A* transition on warming in 0.8 μm. The bold curve is a guide for the eye.

It is interesting that the frequencies at the lowest temperatures depend on the liquid pressure. The negative shift from the Larmor frequency is 1 kHz at 0.5 MPa and 2.5 kHz at 2.45 MPa. At the lowest temperatures, the absorption signal mostly involved only information on the solid because of its large magnetization. Such a negative frequency shift can be explained as the result of partly aligned magnetic dipoles on the sheet qualitatively. From these measurements it might be possible to derive the exchange interaction between atoms in the two-dimensional solid under pressure. It is also interesting that the hysteresis loop becomes small at lower pressures. This indicates that the long coherence length at lower pressures may impede the gradual *A-B* transition.

Figure 3 shows the temperature for completion of the *B* to *A* transition on warming in the *p-T* phase diagram. A critical thickness δ was introduced to analyze *A-B* transition phenomena [9], which is defined as $\delta = d/\xi(T_{AB})$, where d is the distance between parallel plates and $\xi(T_{AB})$ is the coherence length at the *A-B* transition temperature T_{AB} in slab geometry. In calculations, we used the temperature dependent coherence length $\xi(T) = (3/5)^{1/2}\xi_0(1 - T/T_c)^{-1/2}$, where ξ_0 is the coherence length at 0 K defined as Eq. (2) in Ref. [8], and T_c is the superfluid transition temperature in bulk liquid. So the *A* phase appeared at temperatures where $d/\xi(T)$ was smaller than δ. In Table I, we show the critical thickness from experimental results and the theoretical calculation [9]. The critical thickness δ is about 20 and 30 at 1.0 and 2.0 MPa, respectively, in both 1.1 and 0.8 μm slab geometry. Considering the distribution of spacing, these values might be a bit smaller ones. These agree closely with the theoretical calculations of Fujita *et al.* [9]; δ is $\sim$10 at 0 MPa and $\sim$29 at 2.0 MPa by assuming a linear dependence of strong coupling parameter on pressure. In Fig. 4, the obtained δ is plotted with the calculated $d/\xi(0)$ for $d = 0.8$ and 0.3 μm as a function of pressure. Here, $d/\xi(0)$ of 0.3 μm is less than the critical thickness δ at any pressure, which means that only the *A* phase exists in the 0.3μm thick slab at any pressure down to $T = 0$ K. This expectation is consistent with the experimental results in Ref. [11], where only the *A* phase is observed at all pressures in a 0.3 μm thick slab. By extrapolating the critical thickness δ to 0 MPa in the linear approximation, we obtained a universal critical thickness depending on pressures as

$$\delta = 13.7 + 7.37p, \tag{1}$$

with p in MPa. We found that $d(0.8\ \mu\text{m})/\xi(0.33\ \text{mK}) = 16.8$ for 0.14 MPa is larger than $\delta = 14.7$ at 0.14 MPa derived from Eq. (1). This implies that the observed *A* phase below about 0.5 mK at 0.14 MPa may be in the supercooled state.

Finally we describe the effect of ^{4}He coating. It has become clear that with more than 2.5 ^{4}He layers, a quasi two-dimensional ^{4}He system on the surface solid ^{4}He shows the superfluidity which alters the microscopic pro-

TABLE I. Critical thickness

P [MPa]	δ_{calc} [a]	$\delta(1.1\ \mu\text{m})$ [b]	$\delta(0.8\ \mu\text{m})$
0	$\sim$10	...	...
0.3		...	17.0
0.5		...	18.2
1.0		19.9	22.2
1.9			25.6
2.0	$\sim$29	30.5	...
2.4		40.7	34.0
2.7		49.8	36.5

[a]Derived values from Ref. [9].
[b]From our last study [8].

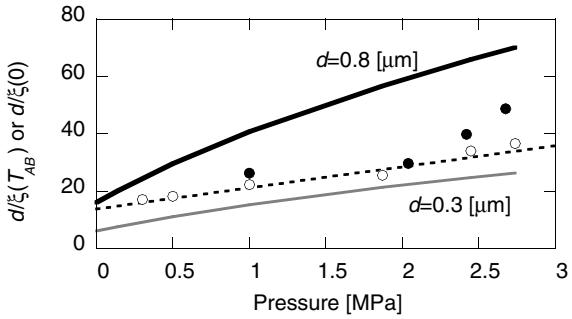

FIG. 4. Critical thickness $\delta = d/\xi(T_{AB})$ as a function of pressure. Solid and open circles are critical thicknesses in 0.8 μm and 1.1 μm, respectively. Black and gray curves are the ratios of $d/\xi(0)$ of 0.8 μm and 0.3 μm. The dotted straight line is Eq. (1). For details, see in the text.

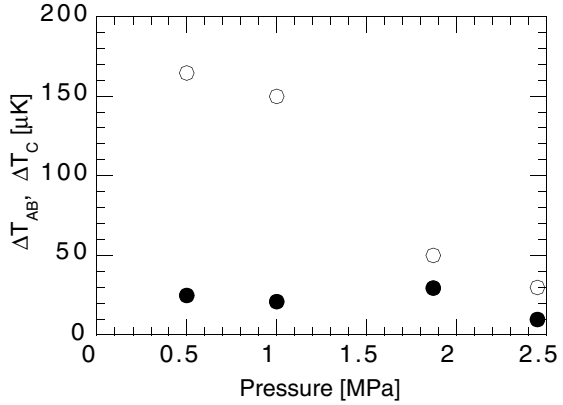

FIG. 5. Increase of A-B transition temperatures ΔT_{AB} and superfluid transition temperature ΔT_C with 2.5 ^{4}He layers. Solid and open circles are ΔT_C and ΔT_{AB}, respectively.

cess of ^{3}He quasiparticles scattering at the boundary to the more specular scattering than that without the superfluid ^{4}He film [17]. Putting 2.5 ^{4}He layers on the sheet, the negative frequencies in A phase were similar to those before ^{4}He coating. We could not see the B phase signal below the temperatures which were a bit lower in temperature than T_{AB}. This behavior was attributed to both the small magnetization without the surface solid ^{3}He and the broadening of NMR spectra in the B phase due to the lack of the rapid exchange between the surface solid ^{3}He and the liquid ^{3}He. We observed the increase of both T_{AB} and the superfluid transition temperature T_C. They are plotted as a function of pressure in Fig. 5. ΔT_C is 30 μK at the lowest pressure and decreases almost linearly to 10 μK at 2.47 MPa. ΔT_{AB} also changes almost linearly from 150 μK at 0.5 MPa to 30 μK at 2.47 MPa. T_C with 2.5 ^{4}He layers was still suppressed from the bulk liquid T_C. This behavior is consistent with that of the partly specular scattering in the fourth sound experiment [18]. ΔT_{AB} at 2.47 MPa is similar to that observed in 1.1 μm spacing at 2.4 MPa [8]. The increase of A-B transition temperature is opposite to the theoretical expectation for the specular scattering [3] where T_{AB} is expected to decrease with a specular scattering. It is thought that the spacing between parallel plates becomes a bit wide effectively by the specular scattering. It is interesting that both increases of ΔT_C and ΔT_{AB} changing linearly with respect to pressures is similar to the specularity behavior in Fig. 5 in Ref. [18] measured by the fourth sound experiment with 2.6 ^{4}He layers.

In summary, we studied superfluid ^{3}He in 0.8 μm slab geometry and observed A-B transition above 0.3 MPa. The obtained critical thickness is in agreement with the theoretical calculation. We also observed the suppression of superfluid transition temperature. The pressure dependence on frequencies at the lowest temperatures is interesting. This newly found phenomenon may involve interaction between liquid and surface solid and the exchange interaction in the surface solid phase itself under pressure. Increases of T_C and T_{AB} by 2.5 ^{4}He layers were understood by increasing the specularity of ^{3}He quasiparticle scattering.

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