Torsional Oscillator Measurements of Liquid $^4$He Confined in 2.5-nm Channel of FSM

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Abstract. We measured the superfluid response of liquid $^4$He confined in 2.5-nm channel under low pressure by means of the torsional oscillator to clarify how the superfluid response varies with decreasing channel diameter. In the previous measurements, in the 2.8-nm channel under 0.13 MPa, the superfluid fraction appears at 1.8 K, and then shows a sharp rise at 0.9 K. On the other hand, in the 2.2-nm channel, no sharp rise is observed although the superfluid fraction appears. In the present measurements for 2.5-nm channel, the superfluid appears at around 1.5 K, and then shows a sharp rise at 0.13 K. By comparison, with decreasing channel diameter, the temperature of superfluid appearance is suppressed with gentle acceleration, which is attributable to the suppression of falling into a BEC-like state. In contrast, the temperature of the rise in the superfluid is suppressed drastically compared with that of superfluid appearance, by decreasing the channel diameter from 2.8 to 2.5 nm.

1. Introduction
One-dimensional (1D) quantum many-body systems have attracted many researchers’ interest due to large quantum fluctuations. In particular, Liquid $^4$He confined in 1D nanometer-size channel is a promising system to study the superfluidity specific to 1D Bosonic Tomonaga-Luttinger (TL) liquid. [1]

We have experimentally studied the superfluid response of pressurized liquid $^4$He confined in a nanoporous medium called FSM, using the torsional oscillator technique. FSM has a uniform straight channel whose diameter ranges from 2 to 5 nm and can be precisely controlled.[2] In the channel 2.8 nm in diameter, we found that a sharp rise in superfluid at 0.9 K under 0.13 MPa is accompanied by a broad dissipation peak at a resonance (observation) frequency of 2 kHz.[3] When the observation frequency is lowered from 2 to 0.5 kHz, both of the sharp rise and the dissipation peak were shifted to the low temperature by 40 mK.[4] The observed frequency dependence is qualitatively explained by the theory based on the bosonic TL liquid model.[5]

Another important property of superfluid response in the 2.8-nm channel is a two-stage growth of the superfluid; under 0.13 MPa the superfluid fraction already appears at 1.8 K, much higher than the temperature of sharp rise, 0.9 K, and keeps gradual growth between these two temperatures.[6] Such a two-stage growth is not observed in the system of either 3D bulk liquid or 2D film, suggesting a property specific to $^4$He in 1D channel.

Regarding the two-stage growth, there have been two theoretical studies by means of a quantum Monte Carlo method. Kelchyskyy et al. demonstrated that the superfluid response to the rotational motion of walls of the channel survives to the high temperature, compared with
that to the longitudinal one.[7] Kiriyama et al. also reported the same tendency and showed that the superfluid response to the rotational motion appears at the same temperature as BEC takes place.[8] Both groups pointed out that the superfluid response is strongly suppressed by decreasing channel diameter.

Up to the present, we have studied the superfluid response in the channels whose diameters are 4.7, 2.8, and 2.2 nm.[3, 9] Between 2.8 and 2.2 nm, the superfluid response changes drastically. In the 2.2-nm channel, no sharp rise was not observed and only gradual growth was observed below about 1.2 K under 0.1 MPa. To clarify how the two-stage growth of superfluid varies between 2.8- and 2.2-nm channels, we started the torsional oscillator measurements of $^4$He confined in FSM with 2.5-nm channel. In this paper, we report the superfluid response in the 2.5-nm channel under low pressure, in comparison with the responses in the 2.8- and the 2.2-nm channel, and then discuss the channel diameter dependence of the superfluid response.

2. Experimental details
The used substrate is one of FSM-N series. Here, N is the number of alkyl chain in the alkyltrimethylammonium used as a template in synthesis. The channel diameter of FSM is controlled by this number. In the present measurements, we used FSM-14, whose nominal channel diameter is 2.5 nm.[10] The 2.8- and the 2.2-nm channels correspond to FSM-16 and -12, respectively.

When certain kinds of adatoms are introduced into such a narrow channel, a capillary condensation occurs, causing a plateau in the adsorption isotherm. The pressure of the plateau lowers monotonously with decreasing channel diameter. In order to examine how clearly the channel diameter of FSM-14 is different from FSM-12 and -16, we measured the $N_2$ adsorption isotherm. The measurements were performed using a pellet which was sintered after mixing FSM-14 powder with silver one for thermal contact.

Figure 1 shows the adsorption isotherm of FSM-14 at 77 K as a function of $N_2$ areal density, accompanied by those of FSM-12 and FSM-16. Every isotherm has a plateau, whose center...
3. Results and Discussion

Figure 2. Change in the resonance frequency from $T_\lambda$ for 2.5-nm channel filled with liquid $^4$He under 0.05 MPa is plotted against the temperature divided by $T_\lambda$. The data for 2.8-nm (0.01 MPa) and 2.2-nm (0.1 MPa) channels are from Ref. 3. The frequency change is divided by the contribution of bulk superfluid outside the FSM powder at absolute zero. The solid curve is the contribution of bulk superfluid. Inset is the enlarged figure of the low-temperature range for 2.5-nm channel. The dotted line is the extrapolation from the high-temperature side. Upward and downward arrows point $T_1$ and $T_2$ in the text, respectively.

As the temperature is decreased, the resonance frequency for 2.8-nm starts to deviate upward from the bulk contribution at 0.83 $T_\lambda$ (1.8 K), due to the appearance of superfluid in the channel. This deviation temperature ($T_1$) is suppressed with decreasing channel diameter, as 0.71 $T_\lambda$ pressure is clearly separated. The values of center pressure indicate that the difference of channel diameter between the neighboring samples is 0.3–0.4 nm. The plateau of FSM-16 ranges the widest areal density region, corresponding to the largest channel volume. As the channel diameter decreases, the areal density region of the plateau gets narrow, indicating that the ratio of the channel volume to the surface area becomes small.

We put this pellet into a torsional oscillator to measure the superfluid response. The total surface area of the pellet is evaluated as 77 m$^2$ by fitting the $N_2$ adsorption isotherm at 77 K to the Brunauer–Emmett–Teller equation in the pressure range of 0.05 $\leq$ P/SVP $\leq$ 0.15. Here SVP is the saturation vapor pressure. The torsional oscillator is made of Be-Cu, and oscillates at a resonance frequency $f$ of 1774.977 Hz, with a quality factor $Q$ of $7.3\times10^5$, at 0.1 K under the condition that the channel is filled with liquid $^4$He under 0.05 MPa.
(1.5 K) for 2.5-nm and 0.56 $T_\lambda$ (1.2 K) for 2.2-nm. Further decreasing temperature, a rise of frequency appears at 0.41 $T_\lambda$ for 2.8-nm, corresponding to the second rapid growth of superfluid. In the case of 2.5-nm channel, as shown in the inset of Fig. 2, a small rise appears at around $0.06T_\lambda$ (0.13 K). For 2.2-nm, no rise was observed at least down to 0.16 K, the lowest temperature we measured.

First, we discuss the appearance of superfluid at $T_1$. In the case of 2.8-nm channel, it was made clear that liquid $^4$He in the channel falls into a “BEC-like” state \cite{12} at almost the same temperature as $T_1$, from the heat capacity measurements. \cite{6} On the other hand, in the quantum Monte Carlo calculation by Kiriyama et al., the BEC temperature is about 1.7 K for 2.8-nm channel, and suppressed with decreasing channel diameter. \cite{8} In the present experiments, $T_1$ is suppressed with gentle acceleration, and the obtained values of $T_1$ are close to the calculated BEC temperature. This coincidence supports that in the channel 2~3 nm in diameter the superfluid appears at the same time as the “BEC-like” state appears.

Next, we focus on the sharp rise in superfluid at $T_2$. $T_2$ is suppressed drastically by decreasing the channel diameter from 2.8 to 2.5 nm. It may be related to a change in the number of liquid layers in the channel. In the channel, inside the two-atomic inert solid layer tubes, liquid layers are formed. According to the calculation of Kulchytskyy et al., there exist the third and the fourth liquid layer tubes in the 2.8-nm channel. As the channel diameter is decreased from 2.6 to 2.4 nm, the fourth layer tube shrinks into a line, and then disappears at 2.2 nm. In 2.0-nm channel, only the third liquid layer tube exists. In order to detect the superfluid along the channel, the superfluid coherence has to be established throughout the channel length. The decrease in the number of liquid layers can make this superfluid coherence fragile drastically, since it drops the number of candidates for particle exchange discretely. In fact, in the calculation, the superfluid response also varies discretely between 2.6 and 2.4 nm.

Another point to be noted is that the observed $T_2$ (0.13 K) is significantly lower than the temperature of the superfluid rise, ~0.4 K, for a thick film observed by Matsushita et al. \cite{13} In the case of 2.8 nm, $T_2$ is suppressed by about 0.2 K by increasing $^4$He amount from 20.4 atoms/nm$^2$ to the liquid under 0.13 MPa. \cite{14} This suppression is attributable to the increase of density in the channel. Although the reason why the suppression is stronger for 2.5-nm channel than for 2.8-nm, this tendency implies that the second rise of liquid $^4$He in the 2.2-nm channel is further suppressed.

In 2.8-nm channel, the second rise has a strong observation frequency dependence, and is thought as a signature of 1D TL liquid behavior. \cite{4} It is a future issue to study whether the $^4$He in the 2.5-nm channel also has a strong frequency dependence.

4. Summary
We measured the superfluid response of liquid $^4$He confined in 2.5 nm channel under low pressure and compared with those for 2.8- and 2.2-nm channels. We found that the temperature where the superfluid appears in the channel, is suppressed with gentle acceleration, which is attributable to the suppression of BEC. On the other hand, the temperature of the sharp rise in the superfluid is suppressed much faster than that of superfluid appearance, by decreasing the channel diameter from 2.8 to 2.5 nm.

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References
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[10] When the sample was supplied to us, the channel diameter was evaluated as 2.5 nm. However this value seems to have an ambiguity of about 0.2 nm, because the precise estimation method has not been established yet. The used sample is the same batch of that used in the work by Matsushita et al.[13], where the channel diameter is displayed as 2.4 nm.
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[12] At present, it is not clear whether the BEC state and a bosonic TL liquid state can coexist. From the heat capacity measurements, the existence of a low-entropy state is confirmed, which we call a “BEC-like” state.
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