Anomalous suppression of superfluidity for $^4$He in Gelsil glass

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Abstract.

We have studied the superfluid behavior of liquid $^4$He confined in three samples of Gelsil glass, whose pore size distribution was examined by measuring $N_2$ pressure isotherm. It was found that the superfluid transition temperature under the saturation vapor pressure is 1.37 K for one of the samples whose pore size ranges up to around 6 nm, while it is 0.83 K for another one in which the distribution of pore size is small above 4 nm. Both of the samples have a bottleneck structure of the same size. These results demonstrate that the suppression of superfluidity is related to the pore size distribution rather than the size of the neck.

1. Introduction

Superfluidity of $^4$He in nanometer-size pores is one of the interesting subjects. Especially, the influence of the pore size has attracted the attention of many researchers. Recently, Yamamoto et al. performed torsional oscillator measurements for $^4$He filled in a nanoporous glass (Gelsil glass) containing pores 2.5 nm in nominal pore size. [1] The superfluid transition temperature drops down to 1.4 K at the saturation vapor pressure (SVP), and then decreases monotonically with increasing pressure. From theoretical study, it was found that the random potential induces a localization of Bose-Einstein condensation and strongly suppresses the transition temperature. [2]

The pore size distribution is thought as a possible origin of the random potential. It is interesting to study how the superfluid transition is altered by the distribution. Thus motivated, we have studied the superfluid behavior of three samples of Gelsil glass, and have investigated the relation with the pore size distribution. It was found that the superfluid transition temperature is suppressed strongly for the sample in which the distribution of large pores is small.

2. Experiments

Gelsil glass is produced by the sol-gel technique, and its structure is characterized by a three-dimensional (3D) random network of nano-pores. In the present experiments, we used a cylindrical specimen of Gelsil glass ($φ$9 mm×L18 mm) whose nominal pore size was 2.5 nm. The specimen was cut into three samples. From the end of the specimen, we refer to them as sample A (t2.19 mm), B (t3.66 mm), and C (t3.45 mm). All samples were dried at 150°C in
vacuum for 3 h, before setting into a Be-Cu torsional oscillator head. N$_2$ pressure isotherm was measured by cooling the torsional oscillator down to 77 K for each sample. From Brunauer-Emmett-Teller fitting, the surface area was determined as 90, 134 and 140 m$^2$, for sample A, B and C, respectively.

The torsional oscillator measurements were performed using a phase-locked-loop composed of a lock-in amplifier and a signal generator. The output of the lock-in amplifier was used to control the frequency of the signal generator in order to keep the in-phase output zero. Then, the frequency was locked to the resonance of the torsional oscillator. The resonance frequency in vacuum was 1919.5, 2105.0 and 2002.6 Hz for sample A, B and C, respectively. The $Q$ factor of the torsional oscillator was better than 5$\times$10$^5$ for all samples.

3. Pore size distribution

![Figure 1. N$_2$ adsorption (solid symbols) and desorption (open symbols) isotherms for sample A, B and C. The horizontal axis is the normalized pressure divided by the saturation vapor pressure at 77 K.](image)

Figure 1 shows the adsorption and the desorption isotherms as a function of the pressure ($P$) divided by SVP at 77 K ($P_0$). In the adsorption process, after the first layer completion of 10.3 $\mu$mol/m$^2$, N$_2$ areal density $n$ increases gradually at a slope depending on sample. Finally, it approaches a constant value $n_{\text{full}}$ above a certain pressure, where the pores are filled. On the other hand, in the desorption process, all isotherms show a clear step at around $P/P_0 = 0.48\pm0.02$ MPa. The constant value of $n$ means that the gas-liquid interface is locked at the neck. The results indicate that all samples have a bottle-neck structure. The value of $n_{\text{full}}$ in the N$_2$ pressure isotherm corresponds to the pore volume per unit surface area ($V$). From this value, on the assumption of a uniform cylindrical pore, the average pore diameter ($2r$) can be deduced as 3.2, 3.0 and 2.7 nm, for sample A, B and C, respectively.

To know the detailed distribution of pore volume with respect to the pore diameter, we calculated the pore size distribution ($\Delta V/\Delta r$) by the Dollimore-Heal method. [3] The obtained distribution from the adsorption isotherms is summarized as a function of pore diameter in the upper panel of Fig. 2. For sample A, the distribution shows a peak at around 2.5 nm and has a large value until around 6 nm. On the other hand, the distribution of both sample B and C has a peak at around 2.1 nm, and decreases faster than that of sample A with increasing pore diameter. The difference from sample A becomes apparent in the pore diameter region between about 4 and 6 nm. In this large pore diameter region, the value of sample B is large compared with that of the sample C. Regarding the peak around 2 nm, the difference between the samples is insignificant due to the ambiguity of about 0.4 nm for the diameter.

On the other hand, $\Delta V/\Delta r$ obtained from the desorption isotherms gives the information about the neck size of the bottle-neck structure. For all samples, it shows a sharp peak at 4.2$\pm0.1$ nm as shown in the lower panel of Fig. 2. In spite of the difference in the pore size distribution, the neck size of all samples is almost in agreement with each other.
4. Superfluidity
Torsional oscillator measurements of $^4$He confined in these samples were performed near SVP between 0.05 and 2.5 K. Figure 3 shows the variation of the resonance frequency ($F$) from that of the transition temperature of bulk $^4$He ($F(T_\lambda)$) under its corresponding pressure.

For sample A, the resonance frequency starts to increase slowly at $T_\lambda = 2.16$ K due to the superfluid transition of bulk $^4$He in an open space of the oscillator head. Further decreasing temperature, it increases at 1.37 K, which corresponds to the superfluid transition temperature $T_C$ in the pore. Then, it approaches a constant value at around the lowest temperature. The change in frequency below $T_C$, whose main contribution comes from the superfluid in the pore, reaches about 0.040 Hz at the lowest temperature.

Figure 3 (b) shows the variation of sample B under 0.13 MPa. With decreasing temperature, the resonance frequency bends upward at $T_\lambda = 2.16$ K. With decreasing temperature, it has an upward break at 0.83 K and shows a large oscillatory behavior. And then, it rises at 0.71 K from the resonance frequency which is smaller than that at 0.83 K. Further decreasing temperature, it exhibits another oscillation and then approaches a certain frequency. Although the observed behavior is significantly different from the conventional decoupling of superfluid in porous media, the change in frequency below the temperature of break, 0.83 K, cannot be explained only by the contribution of the bulk superfluid. Thus, we conclude the temperature of break as $T_C$ in the pore. The difference of the resonance frequency between $T_C$ the lowest temperature is about 0.017 Hz, which is about half as large as that of the sample A.

The variation of sample C under 0.01 MPa is shown in Fig. 3 (c). Although the resonance frequency starts to increase at $T_\lambda$ of 2.16 K in the same manner as other samples, it turns to a decrease at around 1 K. Then, it shows a downward break at 0.60 K and has the first oscillation, and then the second one at around 0.24 K. These oscillations are a common feature with sample B. When we take the temperature of break as $T_C$ along with sample B, $T_C$ for sample C is as low as 0.60 K. However, the decrease in frequency at around 1 K, which cannot be explained only by the bulk contribution, may be associated with a superfluid-like behavior in the pore. For sample C, an ambiguity is left for the determination of the superfluid transition temperature.

Next, we focus sample A and B where a clear superfluid transition is observed. $T_C$ is 1.37 K
for sample A, whose pore size ranges from 1.7 to about 6 nm. On the other hand, it is 0.83 K for sample B, whose pore size distribution has a small value above about 4 nm. The results suggest that the suppression of the superfluid transition is enhanced for the porous media where the amount of large pores is small.

Finally, we note the comparison of the present results with that of the different sample of Gelsil 2.5 nm in nominal pore size measured by Yamamoto et al. [1] Both of the transition temperature and the temperature dependence of the superfluid near SVP are in good agreement with those of sample A. It indicates that the pore size distribution of sample A is similar to that of the sample used by Yamamoto et al.

5. Summary
We have studied the superfluid behavior of $^4$He confined in three samples of Gelsil glass, A, B and C, whose pore size distribution was examined by measuring the N$_2$ pressure isotherm. For sample B in which the pore size distribution is small above around 4 nm, the superfluid transition temperature near SVP is as low as 0.83 K and an oscillatory behavior is observed below $T_C$. It gives contrast to $T_C$ of 1.37 K and the conventional rise of the superfluid observed for Sample A in which the distribution remains large up to about 6 nm. Both samples have a bottle-neck structure whose size is 4.2 nm. The suppression of superfluidity in Gelsil glass is related to the pore size distribution rather than the size of the neck.

References
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