Size Dependence of Superfluidity for $^4$He Confined in FSM16

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Abstract. The superfluidity for liquid $^4$He confined in one-dimensional channels has been studied by means of torsional oscillator. In the present experiments, FSM16 with the channel of 4.7-nm in diameter was used, together with preplating monolayer N$_2$ film on the channel (the effective diameter of 4.1 nm). As the channel is preplated, the superfluid transition temperature at 0.1 MPa shifts to a lower temperature from 1.81 K for 4.7-nm channel to 1.69 K for N$_2$ preplated one. The results show that the superfluidity is suppressed strongly with decreasing diameter.

1. Introduction
The Superfluidity of $^4$He in confined geometry has attracted the attention of many researchers for several decades. A large amount of experimental work has been performed using substrates with pores larger than about 10 nm in size [1]. It suggests that the reduction of the superfluid transition temperature obeys a power law with respect to the typical size of pores, with an exponent equal to the critical index of the bulk superfluid density.

Recently, torsional oscillator measurements for $^4$He confined in Gelsil glass with three-dimensionally (3D) connected pores of 2.5 nm in diameter have been performed. It was reported that the transition temperature is suppressed significantly at zero pressure and is decreased rapidly with increasing pressure. In addition, a quantum phase transition from the superfluid to nonsuperfluid phases takes place at absolute zero at a critical pressure [2]. This behavior is different from the conventional pressure dependence almost in parallel to $\lambda$ line of bulk $^4$He, which was reported for $^3$He confined in larger pores [3, 4, 5, 6]. More recently, torsional oscillator measurements for $^4$He confined in FSM16 with one-dimensional (1D) channels of 2.2 nm in diameter have been performed, and the superfluid transition was not observed clearly [7]. These results suggest that the superfluidity is suppressed strongly in pores several nanometers in diameter.

It is interesting to investigate how the superfluid property evolves in nanometer-size pores, varying pore size systematically under the condition that the pore structure is fixed. Thus motivated, we have started a study for the superfluidity in 1D nanometer-size channels, varying the diameter. In this paper, we report the superfluidity of $^4$He confined in 1D channels of 4.7 nm in diameter and the channels narrowed by monolayer N$_2$ adsorption. A clear reduction of the superfluid transition temperature was observed.
2. Experimental

The substrate we used is FSM16, which has a straight nanometer-size channel with uniform size. In the present experiments, we used 0.143 g of FSM16 with 4.7-nm channel as a sample. It is in the form of powder with a particle size of about 0.3 μm, and the total surface area of the sample $S$ is estimated to be 101 m$^2$ by fitting the $N_2$ adsorption isotherm at 78 K in the pressure range of 5-15 kPa.

In order to realize the channel with the diameter smaller than 4.7 nm, we preplated a monolayer $N_2$ film on 4.7-nm channel. The diameter of $N_2$ preplated channel can be evaluated by the amount of $^4$He necessary to fill the channel, as follows. We measured the vapor pressure of $^4$He, $P$, introduced into both of bare 4.7-nm and $N_2$ preplated channel samples. As shown in Fig. 1. (a), the vapor pressure isotherm for the $N_2$ preplated channel is shifted to the lower amount, as compared with that for the bare channel. From the vapor pressure, we calculated the two-dimensional (2D) isothermal compressibility $\kappa_T$, described as

$$\kappa_T = \frac{S}{n^2 k_B T} \frac{\partial n}{\partial \ln P},$$

(1)

where $n$ is the introduced amount of $^4$He, $k_B$ is the Boltzmann constant, and $T$ is the temperature. It is well known that for $^4$He adsorbed on nanometer-size channels, $\kappa_T$ has two minima at the coverage of the first layer completion and the filling of channel [8]. The obtained compressibility is shown in Fig. 1. (b) as a function of $n$. The second minimum is observed at 4.73 and 3.74 mmol for bare and $N_2$ preplated channels, respectively. Since the amount of $^4$He necessary to fill the channel is proportional to the cross section of the channel, the diameter of the $N_2$ preplated channel is evaluated to be 4.1 nm. It agrees with the diameter estimated using the van der Waals radius of nitrogen of 0.15 nm.

To study the superfluidity of $^4$He in the channel, we employed a torsional oscillator, which consists of a Be-Cu sample cell. The cell contains a sample pellet. It was prepared by mixing FSM16 with silver powders in a 2:1 mass ratio and dehydrating the mixture in a vacuum at 200 °C for two hours. The porosity in the bare channel and of the outer space of the powders are 49 and 27%, respectively. And, for the $N_2$ preplated channel, the porosity in the channel and
of the outer space are evaluated as 38 and 26%, respectively. For both bare and N₂ preplated channel samples, the cell oscillated with a high quality factor of \( Q = 1.8 \times 10^6 \) in a vacuum.

### 3. Results and Discussion

Figure 2 shows the change in the resonance frequency from 2.16 K, which is the superfluid transition temperature \( T_\lambda \) of bulk \( ^4\)He outside powders, for bare 4.7-nm and N₂ preplated channels filled with liquid \( ^4\)He at 0.1 MPa. For the 4.7-nm channel sample, the resonance frequency rises at \( T_\lambda \), and then at a certain temperature, shows another smooth rise. These two rises are due to the superfluid transition of bulk \( ^4\)He in the outer space of the powders and of \( ^4\)He in the channel.

The two superfluid transition should cause two jumps in the temperature derivative of the resonance frequency, \( df/dT \). In the inset of Fig. 2, \( df/dT \) is shown as a function of temperature. It has a clear jump at \( T_\lambda \) due to the superfluid transition of bulk \( ^4\)He in the outer space of grains, and then at the lower temperature another broad decrease appears ranging from 1.72 K to 1.90 K. This decrease is attributed to the superfluid transition in the channel. The broadening of the transition may be caused by the connectivity of the channel with outer space or the distribution of the channel diameter. Thus, we can conclude that the superfluid in the channel grows rapidly in the temperature region of decrease in \( df/dT \), and obtain the transition temperature \( T_c \) to be \( 1.81\pm0.09 \) K for the bare 4.7-nm channel.

The results for the N₂ preplated channel sample are also shown in Fig. 2 and its inset. The superfluid transition in the channel is also observed, and the decrease of \( df/dT \) appears ranging from 1.61 K to 1.77 K. Therefore, \( T_c \) for the N₂ preplated channel is \( 1.69\pm0.08 \) K, which is suppressed as compared with that of the 4.7-nm channel.

It is well known that the reduction of \( T_c \) by confinement becomes large with decreasing pore size, \( d \). The pore size dependence of the reduction of \( T_c \) from \( T_\lambda \) for various pores larger than about 10 nm are well explained by a power law of \( (1 - T_c/T_\lambda) = (d/d_0)^{-1/\nu} \), where \( d_0 \) is a constant. In the vicinity of \( T_\lambda \), the reduction has been discussed in terms of the bulk correlation length, \( \xi \). When the temperature is raised, \( \xi \) increases as \( \xi \propto (1 - T/T_\lambda)^{-\nu} \). Eventually near
\( T_\lambda \) becomes comparable to the pore size, resulting in the vanishment of superfluidity in the channel. Thus, the reduction of transition temperature is expected to obey the power law of the pore size with the exponent, \(-1/\nu\). The exponent \( \nu \) is predicted as \( 2/3 \) from the Ginzburg-Pitaevskii-Mamaladze theory \([9]\). Various experimental work reveals that \( \nu \) ranges between 0.65 and 0.67, and \( d_0 \), 0.1-0.2 nm, which approximately agree with the theoretical prediction.

Whether \( T_c \) obtained from FSM16 with 4.7-nm and \( \text{N}_2 \) preplated channels is in range of the power law for the larger pores, is an interesting question. Substituting \( \nu = 0.67 \) and \( d_0 = 0.2 \) nm, the reduction for pores of 4-5 nm in size is estimated to be 0.25-0.29 K, which is smaller than the observed reduction of 0.35-0.47 K. The effective pore size for superfluid \( ^4\text{He} \) is narrowed by the inert layer. The observed reduction is still large in consideration of this effect. Furthermore, the reduction enhancement evaluated as 0.35 K/0.47 K = 0.78 is larger than that calculated by the theory with the ratio of pore size. These results indicate that narrowing the pore size in the range of 4-5 nm enhances the reduction of \( T_c \) as compared with the case of pores larger than about 10 nm.

It is also interesting to compare the change in \( T_c \) by narrowing the pore size with those observed for 3D connected pores with similar pore size, Gelsil. For Gelsil, it was reported that the decrease in the pore size from 5.8 to 4.7 nm by monolayer Kr adsorption lowers the transition temperature from 1.92 to 1.88 K \([10]\). The reduction by narrowing the pore for FSM16 is larger than that for Gelsil. It is still unclear whether the difference comes from the effect of pore size or pore structure. The experiments for \( ^4\text{He} \) in nanometer-size pores varying pore size with fixed pore structure and the distribution of the pore size, will enable us the discussion of the size dependence of the superfluidity in detail.

4. Summary
We have carried out torsional oscillator measurements for \( ^4\text{He} \) confined in FSM16 with bare 4.7-nm and \( \text{N}_2 \) preplated channels. By preplating monolayer \( \text{N}_2 \) film, the diameter becomes to 4.1 nm, and the superfluid transition temperature is reduced by 0.12 K. The reduction of the transition temperature from \( T_\lambda \) is larger than the magnitude predicted from the bulk-correlation length scaling law.

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