Nuclear magnetic resonance on $^3$He confined in 2.8-nm channel of FSM16

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Abstract.
We have performed continuous-wave (cw) nuclear magnetic resonance measurements for $^3$He confined in 2.8-nm channel of FSM16. The magnetization shows a reduction from the Curie law at low temperature for the high areal density where $^3$He in the channel consists of three portions; a solid first-layer, an amorphous-solid overlayer, and a fluid inside. By an analysis for the magnetization and the available heat capacity data, the reduction can be attributed to an interaction of spins in amorphous solid and a degenerate of fluid.

1. Introduction
One-dimensional (1D) quantum many-body systems have been attractive due to large quantum fluctuations. Trials to realize the 1D Fermi system by confining $^3$He into nanometer-size channels are in progress.\[1, 2, 3\] In these cases, the attention has been paid mainly to a dilute limit, where $^3$He atom is expected to behave like a non-interacting gas. At low areal densities, heat capacity measurements for $^3$He in a 2.8-nm channel show that the crossover from a 2D classical gas to a 1D degenerate state occurs.\[1\] Very recently, a nuclear magnetic resonance (NMR) signal of 1D diffusion has been reported for a little narrower channel.\[2\]

On the other hand, $^4$He confined in nanometer-size channels has been also studied to search the features of 1D system.\[4\] Recently, it was made clear that the superfluidity of pressurized liquid $^4$He confined in a 2.8-nm channel has a large frequency dependence. It has attracted attention as a possible signature of the bosonic Tomonaga-Luttinger (TL) liquid.\[5\] In analogy with the $^3$He system, a dense liquid $^3$He in the channel possibly shows a feature of the TL liquid. Thus motivated, we started NMR measurements of dense liquid $^3$He confined in the channel of the same size.

In this paper, we report the preliminary results of NMR measurements for $^3$He confined in a 2.8-nm channel. At high areal densities, the reduction of magnetization ($M$) from the Curie law is observed. By taking the previous heat capacity ($C$) data into consideration\[6, 7\], it is made clear that the reduction can be attributed to the interaction of spins in the amorphous solid and the degenerate of fluid.

2. Experimental
The porous material we used is FSM (Folded Sheets Mesoporous materials) 16, whose 1D channel is 2.8 nm in diameter, and 0.2-0.5 $\mu$m in length.\[8\] The FSM powder was baked at 220°C for 6 hours. For a thermal contact, a mixture of FSM and 70 $\mu$m silver powders was...
compressed, together with a brush of fine silver wires, and sintered at 200°C for 3 hours. The sintered sample was put into a Stycast 1266 sample cell. The surface area of the sample was determined as 100±5 m² from a N₂ adsorption isotherm. According to the previous heat capacity measurements for ³He, the areal densities of the first layer completion and the complete filling of channel are estimated to be 8.5±0.7 and 17.7±0.7 atoms/nm², respectively.[6, 7]

Continuous-wave NMR measurements were performed by sweeping a magnetic field. A measuring frequency was 2.089 MHz. The rf coil wound around the sample cell made a resonant circuit with a capacitance of a coaxial line. The Q factor of the circuit was as high as 85. To avoid a saturation of NMR signal, a small amplitude of the rf field was used. It was 10 mVpp below 0.10 K.

The absorption line was averaged more than 20 times to improve the signal noise ratio. It was slightly broader than a Lorentzian function, due to a magnetic homogeneity of about 4×10⁻⁴. M was obtained from a numerical integration of the absorption line using a convolution of a Lorentzian and a Gaussian function. The uncertainty of M was mainly due to the uncertainty of subtraction of the base line and was about 5% at 0.08 K.

The sample cell and the magnet for the static field were anchored separately to a mixing chamber of the dilution refrigerator. The temperature (T) of the sample cell was measured by means of a RuO₂ thermometer. Measurements were performed in the temperature range between 0.055 and 1.0 K.

3. Results and Discussion
The magnetization was measured for several areal densities between 10 and 18 atoms/nm². Above about 0.4 K, M increases inversely proportional to T, which means that it obeys the Curie law. The Curie constant obtained from the data above 0.4 K increases in proportion to the areal density.

Regarding the low temperature behavior, M at 10 atoms/nm² obeys the Curie law down to the lowest temperature of 0.055 K. As the areal density is increased, M becomes smaller than the Curie law. For 16 atoms/nm², the reduction starts at around 0.2 K (T⁻¹ = 5 K⁻¹), as shown in Fig. 1.

To search the origin of the reduction, we examine the states of ³He confined in the channel, on the basis of previous heat capacity measurements. In the Refs. 6 and 7, it is pointed out that ³He in the channel is divided into three portions, a solid first-layer, an amorphous solid overlayer, and a fluid inside. In this paper, we discuss the temperature dependence of C for each portion. Then, we show that the heat capacity and the reduction of the magnetization can be explained by these contributions.

In the first-layer solid, the heat capacity is quite small at high temperature, and shows an upturn with decreasing temperature below about 40 mK. The temperature dependence of the upturn is T⁻², which is a typical one of the spin heat capacity with an exchange interaction energy (J). The interaction energy at the areal density of the first layer completion is evaluated as J₁/k_B ~1.7 mK.

When the amorphous-solid layer grows (the corresponding areal density ranges from 8.5 to 13 atoms/nm²), at high temperature, a T-linear heat capacity grows, which is characteristic to an amorphous solid.[9, 10] On the other hand, the temperature dependence at low temperature becomes weaker than T⁻²-one, and approaches nearly a constant value, with increasing areal density. A similar temperature dependence is observed for an amorphous solid formed in a Vycor glass, which has a heterogeneity.[11] Golov et al. attributed the temperature dependence to the distribution of exchange interaction, which is caused by a range of local areal densities. Although FSM16 has a uniform structure, the local areal density of ³He adsorbed on the first layer may have a distribution due to the restricted size of the channel. Therefore, in the same manner as the Vycor glass, we assume a uniform distribution of the local density. Due to the
Figure 1. Magnetization as a function of the inverse of temperature. The area density is 16 atoms/nm$^2$. Solid line corresponds to the Curie law. Solid curve shows the calculated magnetization, which is the sum of the contributions from the first-layer solid (dash line), the amorphous solid (dotted curve) and the fluid (dashed-dotted curve). (see text.)

exponential dependence of $J$ on the density, it yields a roughly uniform distribution over $\ln J$,

$$\frac{dN_{aS}(J)}{d(\ln J)} = \text{const},$$  \hspace{1cm} (1)

where $dN_{aS}(J)$ is the number of $^3$He having the exchange interaction energy between $J$ and $J + dJ$. At around the temperature of $J/k_B$, the heat capacity of $^3$He shows a Schottky-like peak. Sum of the Schottky-like heat capacity for the range of $J$ yields the weak temperature dependence.

In fitting the heat capacity of amorphous-solid layer, we use fitting parameters of a lower ($J_l$) and a higher ($J_h$) cut-off of interaction energy, and the effective areal density of amorphous solid ($n_{aS}$). As a lower cut-off, we adopt $J_l/k_B = 1.7$ mK. As a higher cut-off, we use the temperature at around which the upturn due to the spin heat capacity appears. As the areal density is increased, $J_h/k_B$ becomes high, and reaches 120 mK at 13 atoms/nm$^2$, where the amorphous solid is completed. We found that $n_{aS}$ is smaller than the direct experimental values by up to 1.4 atom/nm$^2$. It may come from the increase in the density of the first layer, which is compressed by the overlayer.

When a fluid appears (the corresponding areal density is above 13 atoms/nm$^2$), there is no more growth of the upturn. On the other hand, above about 0.35 K, the increment of heat capacity by the introduction of $^3$He is independent of temperature, and its amount is about 1.4 J·K$^{-1}$·mol$^{-1}$. With decreasing temperature, the increment of heat capacity decreases at around 150 mK, and seems to approach the $T$-linear dependence. The temperature dependence is quite analogous to that of 3D Fermi gas, although its value is about eighth part of the one of the Fermi gas. The quantitative inconsistency also appears in the case of bulk liquid $^3$He, and may come from the complicated interaction peculiar to a dense liquid.[12] Here, assuming that the fluid in the channel behaves as a non-interacting 3D Fermi gas, the temperature dependence is qualitatively reproduced, when the degenerate temperature is $\sim 300$ mK.
Now, we calculate the magnetization assuming the same states as those for the analysis of heat capacity. $J_1/k_B$ (1.7 mK) is smaller more than one order than the measured temperature region. Thus, the first layer contribution is expected to increase inversely proportional to temperature down to the lowest temperature, as shown as a dashed line in Fig. 1. In the amorphous-solid layer, the effective exchange interaction energy ranges from 1.7 to 120 mK. Since the magnetization of the amorphous-solid layer is smaller than the Curie law, we adopt the temperature dependence of magnetization with antiferromagnetic coupling.[13] By summing up each contribution over the distribution range of $J$, a weaker temperature dependence of magnetization $M_{aS}$ than the Curie law is obtained as shown as a dotted curve in Fig. 1. For the fluid, we calculated the magnetization, assuming a non-interacting 3D Fermi gas using the degenerate temperature of 300 mK. It is shown as a dashed-dotted curve in Fig. 1. Finally, we sum these three contributions, as shown as a solid curve in Fig. 1. Within the experimental error, the observed temperature dependence is in agreement with the calculated curve.

The results show that the temperature dependences of both the heat capacity and the magnetization are explained by the contributions of the first-layer solid, the amorphous solid and the fluid. It is of significance that both data support the degenerate of fluid in the channel. In order to study the feature of the degenerate fluid in detail, it would be better to perform the measurements for the system where the first-layer and the amorphous solids are replaced by $^4$He. In addition, the feature of 1D Tomonaga-Luttinger liquid is expected to become pronounced in the dynamical property, such as a relaxation time and a diffusion constant. We think that measurements of spin-lattice and spin-spin relaxation times ($T_1$ and $T_2$) by means of pulse NMR is useful to search the feature of degenerate fluid state in the channel.

4. Conclusion
We have performed cw NMR measurements for $^3$He confined in 2.8-nm channel of FSM16. The magnetization shows a reduction from the Curie law at low temperature above 13 atoms/nm$^2$. The reduction can be attributed to an interaction of spins in the amorphous solid and the degenerate of the fluid.

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