Helium-4 Bose Fluids Formed in One-Dimensional 18 Å Diameter Pores

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Bose fluids restricted in one dimension (1D) are realized by adsorbing ^{4}He atoms on the 1D pore walls with a diameter of about 18 Å. The Bose fluid appears above an adsorbed amount after the pore walls are coated with the inert ^{4}He atoms. Heat capacity of the fluid was observed to have a temperature-linear term at low temperatures. This corresponds to the phonon heat capacity of the Bose fluid in the 1D pores. We estimate the phonon velocity and the interaction of the 1D Bose fluid.

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Quantum superfluids of helium 4 and 3 in various dimensions have been studied to test the phase transition mechanisms characteristic of the dimensionality and to understand the relation between the Bose-Einstein condensations (BEC) and the superfluid. In addition to the three-dimensional (3D) studies on the usual bulk superfluids, time-dependent superfluid transition by rapid cooling (4D) has been studied as a realistic simulation of the Kibble-Zurek mechanism [1] which is proposed for the second transition in the early universe [2]. The most typical example of the Kosterlitz-Thouless (KT) superfluid transition [3] has been demonstrated in the 2D ^{4}He liquids adsorbed on flat solid surfaces, where the superfluid occurs at below the BEC temperature. True 2D fluids were made by the vertical motion of the helium adatoms to the surface being in the ground state of the discrete energy levels. A new one-dimensional ^{4}He Bose fluid can be realized by setting one of the two remaining motional freedoms in a ground state. One possible way is to confine the helium fluid films in a narrow tunnel where the motion in the cross section is in the ground state.

On flat solid surfaces, superfluid ^{4}He layers are formed after coating the surfaces with inert ^{4}He layers. Superfluid made on Hectorite substrates [4] shows 2D phonon and a roton heat capacity. The phonon velocity v_I obtained from these increases with the density of the fluid layers, because of the increase in interaction by the hard cores of the ^{4}He atoms. For a liquid film tube adsorbed on the wall inside a 1D pore that has the diameter d and length L, the phonon motion along the circle πd of the cross section has discrete energy levels. At low temperatures where $k_B T$ is much smaller than the gap energy $h v_I / \pi d$, the 1D liquid can be realized when L is sufficiently long; it corresponds to $T \ll 1.8$ K for $d = 18$ Å and $v_I = 200$ m/sec, a typical value of the bulk liquid.

In this Letter, we report the realization of the ^{4}He Bose fluid system adsorbed on 1D pores with a diameter of 18 Å and a length of 3000 Å [5]. The layer formations on the 18-Å pore walls were studied by measuring the pressure of adsorption. The heat capacity of the ^{4}He fluids in the 1D pores show temperature linear dependence that indicates a 1D phonon excitation in the 1D interacting Bose fluids.

The 1D mesopores are formed in a grain about 3000 Å in size which has a honeycomb framework as shown schematically in the inset of Fig. 1(a). The 1D pore diameter of the substrate called FSM-16 can be changed by the template of surfactants with different alkyl-chain lengths [5]. The present 1D pores are made from the chains of $(CH_2)_{10}HN^+(CH_3)_3$. Since a method of pore diameter measurement has not yet been established, the estimated diameter ranges about 4 Å depending on the method used [6]. The Kelvin method for the N_2-pressure isotherm gives 16 Å which is the lowest estimate. We thus determined the nominal pore diameter to be 18 ± 2 Å.

To measure the pressure of adsorption and the heat capacity, we made a sample cell which contains about 0.2 g of the substrate. After dehydrating at 200 °C for about 2 h, the cell was mounted on a cryostat in a helium gas atmosphere and vacuumed for several hours. The adsorption area S was measured by a N_2 pressure isotherm at 78 K by fitting the Brunauer-Emmett-Teller equation in the pressure range between 5 and 15 kPa and using the value of 16.2 Å^2 per one N_2 molecule. The area was estimated to be $S = 195 \pm 10$ m^2. From the grain and the pore sizes, the grain surface area was determined to be less than 1% of the total adsorption area, so the influence of the ^{4}He atoms adsorbed on the grain surfaces could be ignored in the present experiments.

Pressure of the ^{4}He adsorption was measured down to 0.1 Pa using a diaphragm pressure sensor mounted near the cell and an absolute pressure sensor at room temperature. The pressure P of the gas phase equilibrium with the adsorbed helium is a function of the adsorbed amount n and the temperature T. These dependencies show a layer-by-layer adsorption and a full adsorption with increasing n, as described below [4,7,8]. Mounting the same sample cell on a dilution refrigerator, heat capacities of the ^{4}He adatoms were measured down to about 80 mK at

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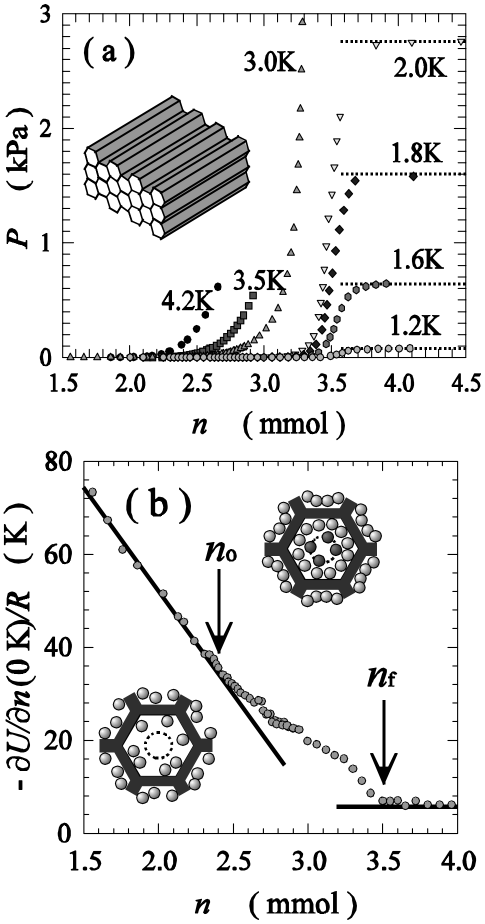

FIG. 1. (a) Pressure isotherms of ^{4}He adsorbed on FSM-16 that has 1D pores 18 Å in diameter as shown in the inset. The abscissa n indicates the amount adsorbed on the present sample cell having the adsorption area of 195 m^2. The dotted lines are the saturated vapor pressures (P_0) at respective temperatures. Much lower pressure than P_0 indicates the adsorption of ^{4}He atoms on the pore walls. (b) Adsorption energy, $-\partial U/\partial n(0\ \mathrm{K})/R$, of the ^{4}He adatoms as a function of n, which indicates layer formations on the pore walls of FSM-16. The ^{4}He atoms are adsorbed on the pore walls up to n_o to form the first layer. The second layer is promoted above n_o with the energy about 30 K. The energy becomes equal to the latent heat (7 K) of the bulk liquid above n_f.

several coverages by the usual adiabatic heat pulse method [4]. To identify the quantum fluids of the adatoms by the isotope effect, we also measured the ^{3}He heat capacities.

The pressure isotherms of ^{4}He are shown in Fig. 1(a). The dotted line shows the saturated vapor pressure P_0 at each temperature. The isotherms show that P becomes P_0 above 3.7 mmol and that it is much smaller than P_0 at $n < 3.4$ mmol. In many pores of the diameter 20–40 Å, ^{4}He atoms are adsorbed on the pore walls up to the pressure about $0.3P_0$. Then, the P isotherms [Fig. 1(a)] indicate that the ^{4}He atoms are adsorbed on the pore walls up to $n_f \sim 3.4$ mmol, or a slightly larger amount below 3.7 mmol.

From the temperature dependence of the pressure P at a constant amount n, one can obtain an isosteric heat of sorption q_{st} by $q_{st} = -R[\partial \ln P/\partial(1/T)]_{n=\mathrm{const}}$, where R is the gas constant [9]. We obtained q_{st} down to the temperature where P becomes 0.1 Pa and at $T > 1.2$ K. Extension of $q_{st}(T)$ to $T = 0$ K was done from the heat capacity C data using the following relation:

$$q_{st}(T) = \frac{5}{2}RT - \frac{\partial U(0\ \mathrm{K})}{\partial n} - \int_0^T \frac{\partial C(T')}{\partial n}\, dT', \quad (1)$$

where $-\partial U/\partial n(0\ \mathrm{K})$, the adsorption energy, was set to agree with the q_{st} result obtained from the pressure measurement [4]. Figure 1(b) shows $-\partial U/\partial n(0\ \mathrm{K})/R$ as a function of n. With increasing n, it decreases monotonously due to the increase in interaction between the adatoms. Above n_o ($\sim$2.4 mmol), the dependence deviates upward. This stepwise change shows the promotion of a new layer. Above n_f ($\sim$3.5 mmol), the adsorption energy becomes equal to the latent heat (7 K) of the bulk ^{4}He liquid. From these results, we conclude that the second layer is promoted above n_o as shown by the right inset of Fig. 1(b) and that it is completed at n_f.

The same conclusion was deduced from the criterion by the compressibility κ_T defined by $\kappa_T = S/(n^2RT)[\partial \ln P/\partial n]^{-1}_{T=\mathrm{const}}$ [7,8]. Similar to the case of ^{4}He adsorbed on graphite [7], we observed a minimum of κ_T at $n \sim 2.4$ mmol, which indicates formation of a new layer above that amount [10].

Heat capacity isotherms of $T = 0.1$ and 0.2 K are shown in Figs. 2(a) and 2(b) for the ^{3}He and ^{4}He adatoms,

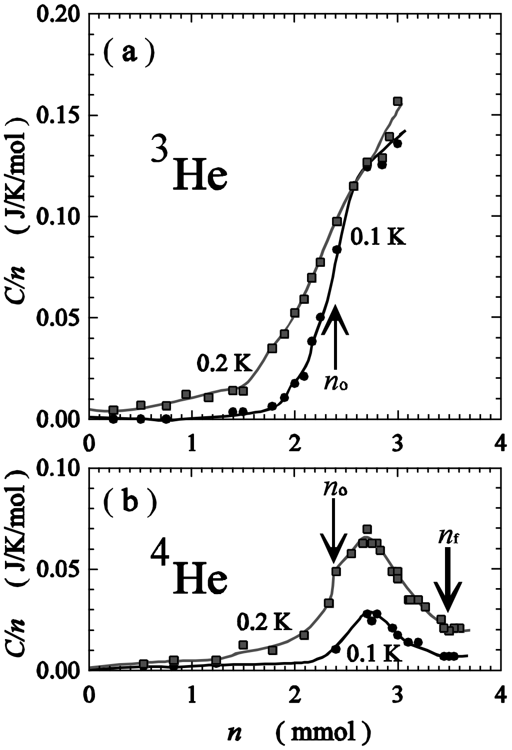

FIG. 2. Heat capacity isotherms of ^{3}He (a) and ^{4}He (b). Qualitatively different dependencies above about n_o indicate that the second layer adatoms are Fermi and Bose fluids, respectively.

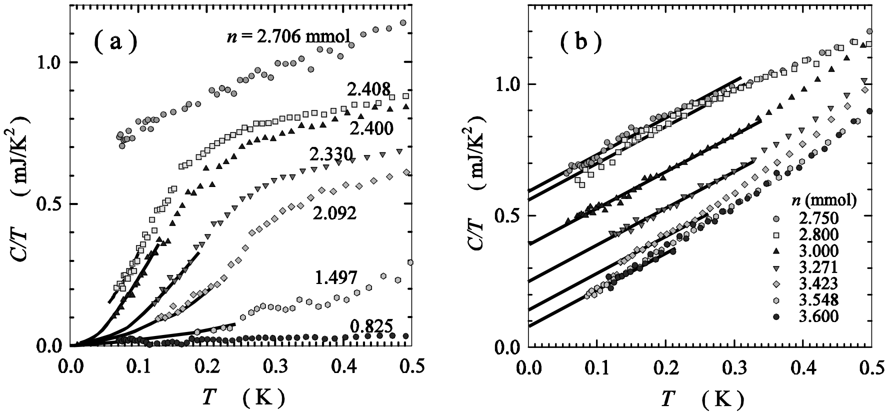

FIG. 3. C/T against T of the ^{4}He adatoms as a function of n. (a) C/T is close to zero at the lowest temperature, for $n < 2.4$ mmol. (b) Low temperature dependencies can be described as $C/T = \alpha(n) + bT$ for $n > 2.7$ mmol, as shown by the solid lines. The first term $\alpha(n)$ strongly depends on n and is assigned to the fluid layer. The coefficient b is constant at 1.4 mJ/K^3 due to the inert layer.

respectively. When ^{3}He and ^{4}He are adsorbed, both heat capacities are small and almost the same. Above about n_o (~2.4 mmol), the ^{3}He heat capacity for $T = 0.1$ K becomes much larger than that of ^{4}He. The large ^{3}He heat capacity is due to the nuclear heat capacity caused by the quantum atomic exchanges which are as large as the bulk and the 2D ^{3}He Fermi liquids [11]. The superfluid of the ^{4}He adatoms has been observed for the present 18 Å pores by a torsional oscillator [12]. Normalizing the adsorption area of the torsional cell to that for the present measurements, the superfluid appears above $n = 2.4 \pm 0.1$ mmol which agrees with n_o of the second layer onset.

The heat capacity above the coverage n_o consists of two terms of the first inert and the second fluid layers. To obtain the fluid heat capacity, temperature dependencies of the ^{4}He heat capacity C are plotted as C/T vs T in Figs. 3(a) and 3(b). At the coverages lower than n_o, C/T is close to zero at the lowest temperature. However, above $n \sim 2.7$ mmol which is a little larger than the second layer onset n_o (~2.4 mmol), C/T is likely to go to a finite value at $T = 0$ K. As shown by the solid lines in Fig. 3(b), the low temperature dependencies below about 0.3 or 0.2 K are simply described as $C/T = \alpha(n) + bT$, using the constant of $b = 1.4$ mJ/K^3. The first term $\alpha(n)$ is strongly dependent on n as shown in Fig. 4, and it becomes almost zero at n_f. Since the property of the fluid strongly depends on the density, $\alpha(n)$ is likely to be of the second fluid layer. The bT term that does not depend on n is of the first inert layer. Then, the heat capacity of the fluid formed in the 1D pores becomes $C_{\mathrm{liq}} = \alpha(n)T$.

To show the influence of the dimensionality of the substrate, we compare the present results with those on the 2D substrate of Hectorite [4]. In Fig. 2(b), the ^{4}He isotherm above n_o becomes maximum at $n_{\max} \sim 2.7$ mmol and minimum at n_f. Quite a similar isotherm was observed for ^{4}He adsorbed on Hectorite [4]. The corresponding coverages are $n_o = 24.3$ μmol/m^2, $n_{\max} \sim 26$ μmol/m^2, and $n_f \sim 37$ μmol/m^2. The coverage dependence between $n_{\max}$ and n_f is caused by $C_{\mathrm{liq}} = \beta(n)T^2$. This is the phonon heat capacity of the 2D Bose liquid. The correspondence indicates that $C_{\mathrm{liq}} = \alpha(n)T$ and $\beta(n)T^2$ are the dimensional properties of the fluids formed on the 1D and 2D substrates, respectively.

In the 1D 18 Å pores, $C_{\mathrm{liq}} = \alpha(n)T$ of the second fluid layer is understood to be the 1D phonon heat capacity, because the transverse motion to the tunnel should be in the ground state. From $\alpha(n)$, the phonon velocity v_l can be obtained by

$$\alpha = \frac{2\pi^2}{3}\frac{L}{hv_l}k_B^2, \qquad (2)$$

where L is the total 1D length. Here, we have to consider the thickness of the inert first layer on the diameter d and the area S of the cylindrical fluid films from which L is given by $L = S/\pi d$. Assuming the thickness of the first layer is 3.5 Å, these are estimated to be $d = [(18-3.5) \times 2]$ Å and $S = (11/18) \times 195$ m^2, respectively. Thus, the velocity v_l was obtained as shown in Fig. 4.

The excitation spectrum of an interacting Bose fluid has the phonon dispersion at small wave vectors whose wave lengths are larger than the mean distance. We estimate the strength of the interaction in the present ^{4}He Bose fluid in terms of Lieb's solution for a 1D Bose system [13],

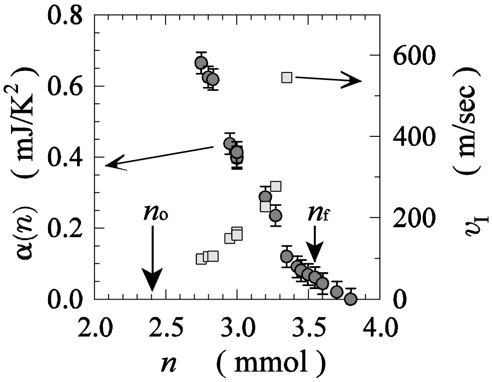

FIG. 4. Coverage dependence of $\alpha(n)$ (closed circles). From $C_{\mathrm{liq}} = \alpha(n)T$, the 1D phonon velocity v_l was estimated, as shown by squares.

where the interaction between two particles with mass m and at positions x_i and x_j along the 1D axis is described as $(\hbar^2/m)c\delta(x_i - x_j)$. The phonon velocity at the particle density ρ in 1D is given by

$$v_I = \frac{\hbar}{m}\rho\left[\frac{c}{\rho} - \frac{1}{2\pi}\left(\frac{c}{\rho}\right)^{3/2}\right]^{1/2}. \quad (3)$$

In the case of the ^{4}He fluid film tube, ρ is calculated from the coverage n by $\rho = N_A(n - n_o)/(S/\pi d)$ where N_A is Avogadro's number. At $n = 2.75$ mmol, we obtained the interaction as $c \sim 0.7$ Å^{-1} from $v_I \sim 100$ m/sec (Fig. 4).

Around the second layer completion $n \sim n_f$, $\alpha(n)$ becomes small by a large phonon velocity (Fig. 4). The phonon velocity and the spectrum at large wave vectors are influenced by a finite size of the ^{4}He hard core. When the coverage n is increased from n_o to n_f, the mobile area for the ^{4}He atoms of the second layer will be reduced from S to zero, from which we estimated the hard core area to be $A_0 = (4.24$ Å$)^2$. A model calculation that takes into account the finite hard core size suggests that a rotonlike spectrum appears at high densities [14]. In Fig. 3(b), the upward deviations of C/T from the solid lines at $n \sim n_f$ seem to be caused by the roton excitations, similar to the case for the 2D ^{4}He Bose liquids on Hectorite [4] where the roton heat capacity was observed above about 0.2 K.

In contrast to BEC's in 2D and 3D, 1D Bose fluid with the phonon dispersion as well as the 1D free Bose particles cause no BEC down to $T = 0$ K, when the 1D scale length is infinite. However, calculation shows there is a case of BEC at a finite temperature if the length of the 1D free Bose gas is finite [15]. The condition of BEC has to be studied using the present ^{4}He Bose fluids formed in the 1D pores.

The superfluid transition of the ^{4}He films on the 2D flat solid surfaces is the most typical KT transition [16], where the phase coherence is destroyed above T_{KT} by unbinding the vortex pairs. In terms of the vortex mechanism, Minoguchi and Nagaoka [17] explained the superfluid onset of the ^{4}He fluid films formed on pores with a diameter larger than 50 Å [18]. Their theory also suggests that the vortex mechanism will break down for the pores whose diameter is equal to or smaller than the vortex core diameter. Since the vortex core diameter was estimated to be 25 ± 12 Å in the experiment [18], the present ^{4}He fluid film tube formed on the 18 Å pore is a possible case where the tube diameter is smaller than the vortex diameter. We note characteristic properties of the superfluid onset observed by the torsional oscillator [12]. The onset temperature T_o in the 18 Å pores increases linearly with n above n_o up to 2.55 mmol, and T_o is about 9 times higher than T_{KT} expected for the same film density. Just below T_o, the superfluid density changes steeply, and this change divided by T_o is 1 order smaller than the universal jump $8\pi k_B(m/\hbar)^2$ of the KT transition.

In conclusion, the layer formation up to the second layer was observed for the ^{4}He atoms adsorbed on the 1D 18 Å pore walls by the pressure of adsorption. The heat capacity and the superfluid observation indicate the Bose fluid of the second layer. The fluid film is a tube with a diameter on the order of 10 Å, which is reduced by the inert first layer. The ^{4}He Bose fluids show the T-linear heat capacity which is understood to be the 1D phonon heat capacity. Interaction in the 1D ^{4}He Bose fluids was estimated by comparison with Lieb's solution, The ^{4}He Bose fluid in the 1D 18-Å pores provides an actual example with which we study the influence of one-dimensionality on BEC and the superfluid transition.

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