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One-Dimensional Phonon State of ^{4}He Films Adsorbed in Straight Nanopores

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Abstract We have studied heat capacities of ^{4}He adsorbed in straight nanopores 1.8, 2.2, and 2.8 nm in diameter. Heat capacities of the ^{4}He fluid film on the solid layer at 0.08–0.4 K show power laws close to T in 1.8 nm pores, close to T^2 in 2.8 nm pores, and a crossover from T to T^2 with increasing temperature in 2.2 nm pores. These heat capacities are explained by a model assuming a phonon dispersion with continuous one-dimensional (1D) states in the axial direction and discrete energy levels in the azimuthal direction. By fitting experimental data to the model, the phonon velocity along the pore axis and the energy gap for propagation in the cross section are derived. At temperatures sufficiently lower than the energy gap, where the thermal wave length of phonons is much longer than the effective pore diameter, the ^{4}He fluid films show a T-linear heat capacity of 1D phonons propagating only along the pore axis. At higher temperatures, a 1D-2D crossover of phonons occurs.

Keywords Low-dimensional quantum fluid · Helium 4 · Phonon · Bose-Einstein statistics · Nanopores

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1 Introduction

Recently helium adsorbed on the substrate FSM [1] with straight nanopores appears to be a subject for realization of one-dimensional (1D) quantum fluid [2, 3]. Helium adsorbed on the pore walls forms thin film up to certain coverage n_f close to fullpore [4]. At a coverage n_c near the first layer completion, the low-temperature heat-capacity isotherm of ^{4}He has a peak. Above n_c, the isotherm differs from that of ^{3}He, indicating formation of ^{4}He Bose fluid on localized ^{4}He atoms [2]. In the Bose fluid region, the ^{4}He heat capacity shows the power law on T, which implies that gapless excitations like phonon exist. In 1.8 nm straight pores, the temperature dependence of the heat capacity is dominated by the T-linear term at low temperatures. It is considered to be heat capacity due to 1D phonons which propagate only along the axis of straight pores [2, 5]. For 2.2 nm pores, the heat capacity is analyzed by a phonon model assuming discrete dispersion relation for the cross sectional direction, and explained by a 1D-2D crossover of phonons [6]. In the model, the fluid has a 1D phonon heat capacity proportional to temperature T at low temperatures. At higher T, the heat capacity approaches 2D phonon heat capacity proportional to T^2.

In this article, we attempt to apply the phonon model used in the analysis for 2.2 nm pore [6] to heat capacity data of ^{4}He in the 1.8 nm and the 2.8 nm pore [2, 7], and investigate the pore-size dependence of the phonon excitation of ^{4}He and their 1D-2D crossovers.

2 Heat Capacity of ^{4}He Film Adsorbed in Straight Nanopores

FSM substrates [1] used in measurements have three pore diameters, 1.8, 2.2 and 2.8 nm. These diameters seem to have an ambiguity of about 0.2 nm. The surface areas S of sample substrates are 195 m^2, 137 m^2, and 85 m^2 for 1.8 nm, 2.2 nm, and 2.8 nm pores, respectively. The ^{4}He adsorption properties of these substrates are shown in [3] in detail. ^{4}He atoms are adsorbed on the straight nanopore wall as a cylindrical film, up to the coverage n_f for each pore.

The low-temperature heat capacity isotherm of the ^{4}He [2, 6, 7] has a peak at $n_\mathrm{c}(< n_\mathrm{f})$ dependent on T, which is shown in Fig. 1a. Above n_c, the temperature dependence of the heat capacity shows a power law. It indicates that, in this region, the thermal excitation is phonon-like. Temperature dependences of ^{4}He heat capacity above n_c in 1.8 nm pore [2] and 2.8 nm pore [7] are shown in Fig. 2. The heat capacity is close to linear on T in 1.8 nm. On the other hand, in 2.8 nm, the power of the heat capacity is close to T^2. In intermediate 2.2 nm pore, a crossover from T to T^2 with increasing temperature is observed [6].

3 Phonon Model

We assume that $n - n_{\mathrm{c}0}$ of the ^{4}He atoms have a phonon-like dispersion relation at coverages $n > n_\mathrm{c}$, where $n_{\mathrm{c}0}$ is n_c at $T = 0$. ^{4}He adatoms form 2D film on the nanopore surface. However, when the thermal wave length of phonons is

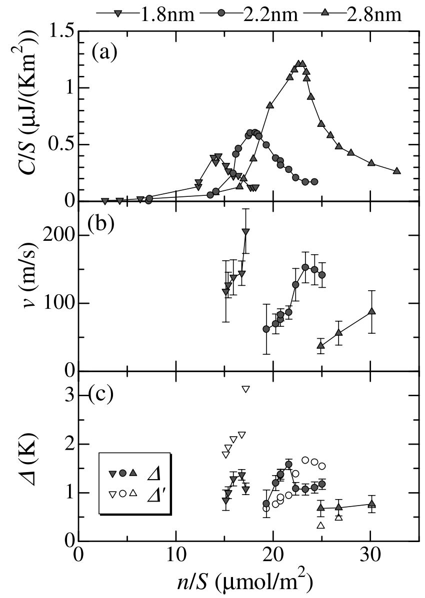

Fig. 1 **a** Heat capacity isotherms at 0.1 K of ^{4}He adsorbed in straight nanopores 1.8, 2.2, and 2.8 nm in diameter. **b** 1D phonon velocity along the *pore axis* derived by the fitting to the phonon model described in Sect. 3. **c** Gap energy to the first excited state in the cross-section. *Solid symbols* are derived by the fitting to the phonon model. Values plotted by *open symbols* are estimated from v along the *pore axis* (see text)

comparable to the pore diameter, discrete wave numbers in the azimuthal direction should be considered. To calculate the total energy, we used a dispersion relation $\varepsilon_{k,l}^2 = (\hbar v k)^2 + (l k_{\mathrm{B}} \Delta)^2$, where k is the wave number of the phonon along the axis of the cylindrical pore, and then v is the 1D phonon velocity. Δ is the gap energy for the first excited state in the cross section, and the label l is assumed to be integer as in the case of 2.2 nm pore [6]. This means that the dispersion relation in the cross section is assumed to be the same as that along the axis, but independent on v, since the number of atoms in the cross section may be so small to consider simple phonon excitations. The heat capacity is calculated by the Debye model using this dispersion relation of phonons. To simplify the model, we ignore temperature dependences of v and Δ, and neglect a contribution of the heat capacity from the solid layer.

At $T \ll \Delta$, occupations of phonon states with $l \neq 0$ are negligible. The contribution of $l \neq 0$ states to the total heat capacity is estimated to be about 1% at $T = 0.1\Delta$ [6]. Thus, at low temperatures, a 1D phonon system is realized, where the excitation has a 1D dispersion relation $\varepsilon_k = \hbar v |k|$, resulting the heat capacity proportional to T. At $T > 0.1\Delta$, the heat capacity gradually deviates from that of the 1D phonon.

4 Results

The experimental data of each pore are fitted to the calculated heat capacities with two fitting parameters v and Δ. The fitting region, where the heat capacity indicates the power law, was taken to be $T < 0.5$ K and $C < 0.05(n - n_{\mathrm{c0}})R$ as in the case of the 2.2 nm pore [6]. The fitting lines are shown by solid lines in Fig. 2 with heat capacity data for 1.8 and 2.8 nm.

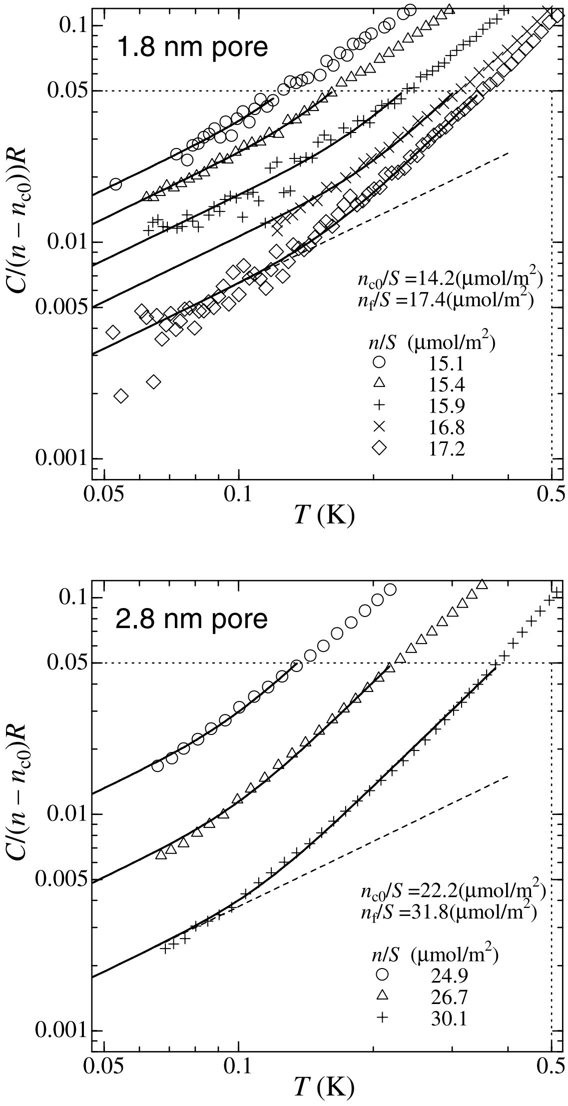

Fig. 2 Heat capacities of ^{4}He fluid film in 1.8 nm and 2.8 nm pores. The heat capacities are divided by the amount of ^{4}He fluid $n - n_{c0}$ and the gas constant R. *Solid lines* are fitting results to the phonon model in the region indicated by *dotted lines*. Ideal 1D phonon heat capacities are shown by *dashed lines* for each pore

The obtained parameters, v and Δ, are shown in Fig. 1b, c, respectively. In each pore, the phonon velocity increases with the coverage, corresponding to the decrease of the 2D compressibility in the second layer of adatoms. The 1D phonon velocity in 1.8 nm pore quantitatively agrees with the compressibility derived from the vapor pressure [5]. The phonon velocity around 100 m/sec agrees with results of the other substrates.

The energy gap Δ in the cross section decreases as the pore diameter increases, as shown by solid symbols in Fig. 1c. To see a relation among v, Δ and the pore diameter, we estimated an energy gap Δ' when the "phonon" velocity in the cross section is assumed to be the same as the 1D phonon velocity v. Effective pore diameters d

are estimated to be 1.0, 1.4, and 1.8 nm for 1.8, 2.2, and 2.8 nm pores, respectively, by subtracting the film thickness at n_{c0} derived by the vapor pressure [4]. Δ' is calculated to be $\hbar v k_1 = \hbar v(2\pi/\pi d)$, where k_1 is the wave number of the first excited state in the azimuthal direction. At $T = \Delta'$, the thermal wave length of 1D phonons equals to πd. Estimated Δ' is shown by open symbols in Fig. 1c. Considering an error of the effective pore diameter and the fitting errors, Δ and Δ' are almost the same. Thus, we can consider that the phonon propagates in the cross section similarly as in the pore axis direction. Above temperatures around $\Delta \sim \Delta'$, the phonons propagating along the pore axis starts to propagate also in the azimuthal direction. It means a 1D-2D crossover of phonons.

In concluding, the 1D phonon regions appears at temperatures sufficiently below Δ shown in Fig. 1c. As seen in Sect. 3, at $T = 0.1\Delta$, deviation of the heat capacity from the T-linear dependence is within 1% [6]. As seen in Fig. 2, the T-linear heat capacities are clearly observed for the 1.8 nm pore but not obvious for the 2.8 nm pore. It agrees that the T-linear regions should appear typically below 0.1Δ.

5 Conclusion

We studied the heat capacity of ^{4}He adsorbed in straight pores 1.8 nm, 2.2 nm, and 2.8 nm in diameter. The excitation of the fluid layer was analyzed by a phonon model with discrete dispersion relation in the cross section, which demonstrates a 1D-2D crossover of phonons. At temperatures sufficiently below the energy gap Δ for azimuthal propagation, where the thermal wave length of phonon is much longer than the pore diameter, the 1D phonon state with the T-linear heat capacity has been realized. It is actually observed especially in measurements down to 70 mK in 1.8 nm pores.

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