## Size-Dependent Kosterlitz-Thouless Superfluid Transition in Thin <sup>4</sup>He Films Adsorbed on Porous Glasses

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We report the first quantitative study of the size-dependent superfluid transition in <sup>4</sup>He films adsorbed on porous glasses with pore diameters d from 50 to 10000 Å.  $T_c$  for the submonolayer films decreases as d increases up to d = 2900 Å, whereas  $T_c$  for the full-pore liquid increases with d. The d dependence of  $T_c$  and the dissipation at the transition for the films reveal that the vortex-pair unbinding on length scales smaller than the pore sizes determines the superfluid transition. We obtain the diameter of the vortex core  $a_0 = 25 \pm 12$  Å from the fit by letting the 2D coherence length  $\xi_2^+(T_c) = \pi d/2$ .

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There has been a great deal of interest in the Kosterlitz-Thouless (KT) transition<sup>1</sup> of two-dimensional (2D) systems, such as 2D XY magnets, liquid-crystal films, and superfluid or superconducting films. Superfluid <sup>4</sup>He films are the most favorable examples of the KT transition. Many experiments<sup>2,3</sup> have confirmed the theoretical prediction of the universal jump and the dynamical properties.<sup>4</sup> At low temperatures, thermally excited vortex pairs are bound and <sup>4</sup>He films exhibit superfluidity. The superfluid transition temperature  $T_{\rm KT}$  is defined as the temperature where the vortex pairs of infinite separation unbind. As temperature T increases above  $T_{\rm KT}$ , the vortex pairs of longer separation unbind. The maximum separation of the bound vortex pairs is equal to the 2D phase coherence length,<sup>4,5</sup>

$$\xi_{2}^{+}(T) = a_{0} \exp\left[\frac{2\pi}{b} \left(\frac{T}{T_{\rm KT}} - 1\right)^{-1/2}\right], \qquad (1)$$

where  $a_0$  is the diameter of the vortex core, which was introduced as a cutoff length to avoid the divergence of flow fields at small distances, and b is a constant. The most striking prediction from Eq. (1) is that the <sup>4</sup>He films exhibit superfluidity on length scales smaller than  $\xi_2^+$ , even above  $T_{\rm KT}$ , and the superfluid transition temperature  $T_c$  shifts upwards from  $T_{\rm KT}$ .<sup>6</sup> The system size plays a crucial role in the KT transition, yet there has not been any experiment reported for a systematic study of size dependence.

In a torsional-oscillator experiment for flat <sup>4</sup>He films, the diffusion length of the vortices during a period of the oscillation,  $r_D = (14D/\omega)^{1/2}$ , corresponds to the characteristic size to determine  $T_c$ ,<sup>2-4</sup> where D is the vortex diffusivity and  $\omega$  is the angular frequency of oscillation. The  $\omega$  dependence of the KT transition in 2D <sup>4</sup>He films, however, has not yet been studied except for the ultrasonic experiment for <sup>4</sup>He films adsorbed on porous Vycor glass at MHz frequencies by Mulders and Beamish.<sup>7</sup>

<sup>4</sup>He films adsorbed on porous materials offer a good example for the study of the size-dependent KT transition, because the film is 2D on length scales smaller than the pore sizes, whereas the film has three-dimensional (3D) connectivity on length scales much larger than the pore sizes. There are two aspects of the dimensionality of the superfluid transition in such films. Bishop et al.<sup>8</sup> studied the influence of 3D connectivity on the superfluid transition of <sup>4</sup>He films adsorbed on porous Vycor glass with a 60-Å pore diameter. Kotsubo and Williams<sup>6</sup> found that the velocity of the third sound in <sup>4</sup>He films on  $Al_2O_3$  powder depends on the powder size. Shirahama et al.<sup>9</sup> made torsional-oscillator experiments for <sup>4</sup>He films on Pt packed powder with diameters from 60 to 500 Å. They found a powder-size-dependent behavior of the superfluid fraction  $\rho_s/\rho$ , where  $\rho = \rho_s(T=0 \text{ K})$ , and the dissipation peaks at the transition expected for the 2D vortex-pair unbinding. Then Minoguchi and Nagaoka<sup>5</sup> theoretically clarified that the unbinding of the vortex pairs whose separations are smaller than the pore size determines  $T_c$ .

In this Letter we report the first quantitative study of the size-dependent KT transition in thin <sup>4</sup>He films adsorbed on porous glasses with pore diameters from 50 to 10000 Å. A preliminary report was presented in Ref. 10.

In order to study quantitatively the pore-size dependence of the superfluid transition, we must compare the data of <sup>4</sup>He films at a constant film thickness for several different pore-size materials, because the properties of the films, in particular  $T_c$ , depend on the film thickness as well. A determination of the film thickness by a surface-area measurement would cause a large systematic error.<sup>9</sup> We use two sets of torsional oscillators containing different pore-size glasses with a common <sup>4</sup>He gas inlet. This setup realizes an equivalent <sup>4</sup>He film thickness in both porous glasses without the need of the surface-area measurement.

We employed seven porous glasses with different pore

diameters, each of which has a much sharper pore-size distribution than that of the packed powder.<sup>9</sup> These glasses were produced especially for this work by acid leaching of phase-separated soda borosilicate glasses,<sup>11</sup> which is a similar process as that for the well-known porous Vycor glass produced by Corning Glass Works. The same kinds of porous glasses as the ones used in the present work have been employed in studies of the solidification and nuclear relaxation phenomena of <sup>3</sup>He in pores.<sup>12</sup> The pore diameters d are 50, 260, 410, 1100, 2900, 7000, and 10000 Å. We regard the peak of each pore-diameter distribution curve as d, which is determined by the N<sub>2</sub> desorption isotherm for the 50-Å glass and the Hg injection for the others. The full width at half maximum of the peaks is smaller than 0.1d for all of the glasses. The porosity is about 50% for each glass sample. The surface area is, for example,  $105 \text{ m}^2/\text{g}$  for the 50-Å glass. A detailed description about the porous glasses will be given elsewhere.<sup>13</sup>

Each of the torsional oscillators<sup>13</sup> consists of a hollow Be-Cu torsion rod and a brass cell containing stacks of the different pore-size glass disks with typical diameter 11 mm and thickness 0.5 mm. Two oscillators are mounted on a massive Cu platform. The <sup>4</sup>He gas inlet line is connected to each cell through the platform and



FIG. 1. The  $\rho_s/\rho$  data for typical coverages of <sup>4</sup>He of six combinations of two different pore-size glasses as a function of temperature. The vertical arrows indicate different  $T_c$ . (a) Inset: Schematic illustration of our experiment.

the torsion rods. The schematic illustration of the setup is shown in Fig. 1(a). The resonant frequencies f at 4.2 K are 1049, 1350, 385, 271, 361, 295, and 475 Hz for the 50-, 260-, 410-, 1100-, 2900-, 7000-, and 10000-Å cells, respectively. The amplitude of the oscillation is estimated to be about 10 Å at the edge of the cell. Qvalues are greater than 10<sup>6</sup> at 4.2 K. The whole setup is mounted under the mixing chamber of a dilution refrigerator. The torsional oscillators are operated at the self-resonant mode by a feedback circuit.<sup>13</sup> We measure the shift of the resonant frequency  $\Delta f$  due to the decoupling of the superfluid component, which is proportional to  $\rho_s$ , and the amplitude of the oscillation, which is proportional to the Q value, as a function of temperature. The ratio of  $\Delta f(T)$  to the value of  $\Delta f$  extrapolated to T=0 K,  $\Delta f(0)$ , gives  $\rho_s/\rho$ . The hydrodynamic disturbance due to the tortuous structure of the pores has no influence on such analyses. We made measurements for six combinations of two different pore-size glasses: 50 and 260 Å, 260 and 410 Å, 410 and 1100 Å, 1100 and 2900 Å, 7000 and 10000 Å, and 410 and 10000 Å.

In Fig. 1, we show  $\rho_s/\rho$  for typical coverages of the six combinations of glasses. In 50-2900-Å porous glasses,  $T_c$  decreases with increasing d. The  $T_c$  of the 10000-Å films is slightly higher than that of the 7000-Å films as shown in Fig. 1(e).

We made measurements for many different amounts of adsorbed <sup>4</sup>He *n*. In Figs. 2(a) and 2(b), we show the *n* dependence of  $T_c$  for two typical experiments. For all porous glasses,  $T_c$  rises linearly with increasing *n* in the range of 0.1 K <  $T_c$  < 0.7 K. Note that the "superfluid" coverage is less than one monolayer in this  $T_c$  range. Nonzero intercepts of  $T_c$ -*n* lines in Fig. 2 are found to coincide within 0.1%. This shows that the thickness of the "nonsuperfluid" layers is the same for all of the porous glasses.

In our experimental situation, the chemical potentials of the <sup>4</sup>He films are equivalent for both of the cells.



FIG. 2.  $T_c$  vs the total adsorption amount of <sup>4</sup>He *n* in two cells for (a) 50-260-Å and (b) 260-410-Å combinations (squares: 50 Å; circles: 260 Å; triangles: 410 Å). (c) Inset: Relative variation of the  $T_c$ -*n* slopes as a function of the pore diameter, where the slope for 2900-Å films is arbitrarily set equal to 1.000.

Cohen et al.<sup>14</sup> pointed out that the superfluid film in a smaller pore is thicker than that in a larger pore. The  $T_c$  difference due to the film-thickness difference is estimated to be of the order of 1  $\mu$ K for all of our glass combinations, which is 4 orders of magnitude smaller than the observed  $T_c$  difference. Therefore the equivalent thickness is realized for both of the cells within the experimental precision. We use the slopes of the  $T_c$ -n lines as a measure of the relative variation of  $T_c$  for a fixed film thickness, which is shown in Fig. 2(c). Our " $T_c$ " is not dependent on the coverage, but determined uniquely for each pore size.

We have also measured  $\rho_s$  for liquid <sup>4</sup>He which completely fill the pores, the "full-pore" case. The full-pore  $T_c$ 's are 1.825, 2.082, and 2.111 K for the 50-, 260-, and 410-Å glasses, respectively. The shifts of  $T_c$  of the fullpore <sup>4</sup>He can be understood in terms of a 3D finite-size scaling.<sup>15</sup> The pore-size dependence of  $T_c$  seen for the 50-2900-Å films has exactly the opposite trend to that of the full-pore <sup>4</sup>He. This fact strongly suggests that the mechanisms of the transition in films is quite different from that of the full-pore <sup>4</sup>He.

In Fig. 3, we show the excess dissipation  $\Delta Q^{-1}$  together with  $\rho_s/\rho$  at the transition as a function of temperature. Sharp dissipation peaks are found at the transition. Note that the maxima of these dissipation curves are located just below  $T_c$ . Similar peaks observed in flat films on Mylar are located at the temperature where  $\rho_s(T) \sim \rho_s(0)/2$ .<sup>2,3</sup> The dissipation peaks are distinguished from the ones due to third-sound resonances, which would cause cusplike behavior in  $\rho_s/\rho$ .

The dissipation peaks clearly show that the vortex pairs unbind at the superfluid transition. Minoguchi and Nagaoka<sup>5</sup> showed that the vortex-unbinding temperature  $T_c$  in films on a multiply connected surface is equal to that on a single cylindrical pore of the same pore diameter, because of the quantization of azimuthal circulation induced by the vortex pairs whose separation r is larger than half of the pore circumference  $\pi d/2$ . The pairs which satisfy  $r > \pi d/2$  do not unbind until  $\xi_2^+$  becomes equal to  $\pi d/2$  and hence they do not contribute to the



FIG. 3. The excess dissipation  $\Delta Q^{-1}$  and  $\rho_s/\rho$  near the transition for (a) 410-Å and (b) 10000-Å films as a function of temperature.

transition. The pairs which satisfy  $r < \pi d/2$  do unbind, and play a crucial role in the superfluid transition. From Eq. (1), the pair unbinding occurs at the temperature where

$$\xi_2^+(T_c) = \pi d/2 \tag{2}$$

is satisfied. From Eq. (2), we see that  $T_c$  increases with decreasing *d*. This is consistent with the observed poresize dependence of  $T_c$  for the 50-2900-Å glasses.

We attempt to fit our data by Eq. (2). In the fit, the 7000- and 10000-Å data are excluded because the frequency effect is considered to be dominant in these pore sizes, as discussed later.  $T_{\rm KT}$  is chosen in the range which satisfies  $T_{\rm KT}/T_c(2900 \text{ Å}) < 1.0$  as the only fitting parameter. We plot  $(T_c/T_{\rm KT}-1)^{-1/2}$  as a logarithmic function of d for the various  $T_{\rm KT}$  values. The  $(T_c/T_{\rm KT}-1)^{-1/2}$  data are found to collapse onto a straight line for the parameter range  $0.965 < T_{\rm KT}/$  $T_c(2900 \text{ Å}) < 0.980$ . The linearity of the line is not sensitive to the  $T_{\rm KT}$  value in this range. In Fig. 4, we show the result with  $T_{\rm KT}/T_{\rm c}$  (2900 Å) = 0.975 as an example. The linear relation seen on the graph for 50-2900 Å shows a good agreement with the theoretical prediction. A detailed discussion of the fitting will be given in a subsequent paper.<sup>13</sup>

From the fit, we can obtain the vortex-core diameter  $a_0$  and the constant b. The nonzero intercept and the slope of the straight line in Fig. 4 give  $a_0 = 25 \pm 3$  Å and  $b = 7.7 \pm 0.1$ , respectively. The choice of the fitting parameter  $T_{\rm KT}/T_c(2900 \text{ Å})$  gives an ambiguity in the  $a_0$  and b values. Yet, using the  $T_{\rm KT}$  value in the range  $0.965 < T_{\rm KT}/T_c(2900 \text{ Å}) < 0.980$ , we eventually obtain  $a_0 = 25 \pm 12$  Å and  $b = 7.3 \pm 1.4$  from the fit. The b value obtained is comparable to that of the flat-film experiments.<sup>16</sup> The  $a_0$  value is much larger than that of bulk <sup>4</sup>He, which is about 2.6 Å.<sup>17</sup> Recent experiments for <sup>4</sup>He films on porous materials have also suggested a large vortex-core size.<sup>6,18</sup>



FIG. 4.  $(T_c/T_{\rm KT}-1)^{-1/2}$  and the averaged dissipation peak height as a logarithmic function of the pore diameter. The straight line for 50-2900 Å is a fit by Eq. (2) with  $T_{\rm KT}/T_c$  (2900 Å) =0.975. Other lines are guides for the eye.

It should be noted that the pore diameter 50 Å is near the limit to form a vortex pair of core diameter 25 Å which satisfies  $r < \pi d/2$  in the pores. Yet half of the pore circumference  $50\pi/2 \sim 80$  Å is still large enough to compare with the obtained  $2a_0$  value. In order to clarify the limit in which the Kosterlitz-Thouless picture remains valid, a further experiment employing smaller pore-size glasses will be very useful, and is under preparation by us.

Williams<sup>19</sup> argued that, in submonolayer films,  $a_0$ should scale as the mean interatomic distance, i.e.,  $a_0 \propto (\rho_s)^{-1/2}$ . However, our  $a_0$  is uniquely determined for  $T_c$  ranging from 0.1 to 0.7 K. The large  $a_0$  suggests that the thin <sup>4</sup>He film is a new type of superfluid, which has a very long "healing" length, and may be suited for the study of quantum interference effects.

In the 2900-10000-Å range of d,  $T_c$  hardly depends on the pore diameter, as shown in Fig. 4. This strongly suggests a crossover from a regime where  $T_c$  is determined by  $\pi d/2$  to one where  $T_c$  is determined by the oscillating frequency. In the films on porous glasses, the pair-unbinding mechanism is equivalent to that of the flat film<sup>4</sup> when  $r_D < \pi d/2$ . The crossover is expected to occur where  $r_D = \pi d/2$  is realized. In the region  $r_D$  $> \pi d/2$ ,  $r_D$  is meaningless in determining  $T_c$  and the dissipation peak is expected to be suppressed. In Fig. 4, we show the dissipation peak heights normalized by the masses of the cells and the temperature as a function of d. The fact that the dissipation peak height decreases as d decreases supports our picture of the crossover.

In our experiment,  $T_c(2900 \text{ Å})$  is equal to that of the 7000-Å glass at  $f(7000 \text{ Å}) \sim 295$  Hz, so that  $r_D$  is estimated to be  $2900\pi/2 \sim 4560$  Å at 295 Hz, and the vortex diffusivity D is  $2.8 \times 10^{-7}$  cm<sup>2</sup>sec<sup>-1</sup> and  $D/a_0^2$  is  $4.4 \times 10^6$  sec<sup>-1</sup>. A difference in the observed  $T_c$  for the 7000- and 10000-Å films is due to the different oscillating frequency,  $f(10000 \text{ Å}) \sim 475$  Hz. It should be noted that for the glasses with d < 2900 Å the frequency is considered not to play a major role in determining  $T_c$ . Our  $D/a_0^2$  is close to the value  $5.6 \times 10^7$  sec<sup>-1</sup> obtained by Agnolet *et al.* for a thin <sup>4</sup>He film on Mylar,<sup>3</sup> among very scattered higher values obtained by other experiments.<sup>16</sup>

Our experiment has revealed that the 2D phasecoherence length determines the superfluid transition of the thin <sup>4</sup>He films on porous glasses. When  $\pi d/2$  is smaller than  $r_D$ ,  $\pi d/2$  determines  $T_c$ , and, otherwise,  $r_D$ does. As to the 3D problem, our experiment does not seem to reveal very much, though, below  $T_c$ , 3D behavior is theoretically expected to appear as the phase coherence grows macroscopically.<sup>5,20</sup> The relation between the 2D properties obtained in the present work and the 3D connectivity of the films is open to further investigation.

We have measured  $\xi_2^+$  by intentionally changing the pore sizes, and have obtained  $a_0 = 25 \pm 12$  Å, and also  $r_D$ and D. The porous glasses have turned out to be a useful tool for studies on microscopic and mesoscopic properties of <sup>4</sup>He films.

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- <sup>1</sup>J. M. Kosterlitz and D. J. Thouless, J. Phys. C 6, 1181 (1973).
- <sup>2</sup>D. J. Bishop and J. D. Reppy, Phys. Rev. B 22, 5171 (1980).

<sup>3</sup>G. Agnolet et al., Phys. Rev. B 39, 8934 (1989).

<sup>4</sup>V. Ambegaokar et al., Phys. Rev. B 21, 1806 (1980).

 ${}^{5}$ T. Minoguchi and Y. Nagaoka, Prog. Theor. Phys. 80, 397 (1988).

<sup>6</sup>V. Kotsubo and G. A. Williams, Phys. Rev. B 33, 6106 (1986).

<sup>7</sup>N. Mulders and J. R. Beamish, Phys. Rev. Lett. **62**, 438 (1989).

<sup>8</sup>D. J. Bishop et al., Phys. Rev. B 24, 5047 (1981).

<sup>9</sup>K. Shirahama *et al.*, Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 293 (1987).

<sup>10</sup>K. Shirahama et al., in Quantum Fluids and Solids-1989,

edited by G. G. Ihas and Y. Takano, AIP Conference Proceedings No. 194 (American Institute of Physics, New York, 1989), p. 195.

<sup>11</sup>H. Tanaka et al., J. Non-Cryst. Solids 65, 301 (1984).

<sup>12</sup>M. Shimoda *et al.*, J. Low Temp. Phys. **64**, 285 (1986); Y. Kondo *et al.*, *ibid.* **75**, 289 (1989).

 $^{13}$ K. Shirahama, M. Kubota, S. Ogawa, N. Wada, and T. Watanabe (to be published).

<sup>14</sup>S. M. Cohen et al., Phys. Rev. B 33, 4664 (1986).

<sup>15</sup>F. M. Gasparini *et al.*, Phys. Rev. B 29, 138 (1984).

<sup>16</sup>J. Maps and R. B. Hallock, Phys. Rev. B 27, 5491 (1983).

 $^{17}$ See, for example, G. W. Rayfield and F. Reif, Phys. Rev. **136**, A1194 (1964).

<sup>18</sup>D. F. McQueeney, Ph.D. thesis, Cornell University, 1988 (unpublished); G. M. Godshalk and R. B. Hallock (to be published).

<sup>19</sup>G. A. Williams, Jpn. J. Appl. Phys. 26, Suppl. 26-3, 305 (1987).

<sup>20</sup>J. Machta and R. A. Guyer, Phys. Rev. Lett. **60**, 2054 (1988); F. Gallet and G. A. Williams, Phys. Rev. B **39**, 4673 (1989).