Frequency Dependence of the Superfluid Transition in ^{4}He Films on Porous Vycor Glass

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We have used an ultrasonic technique to measure the superfluid density and the dissipation in ^{4}He films adsorbed on porous Vycor glass. The superfluid transition is quite sharp at the lowest frequency (5 MHz) but broadens and shifts to higher temperatures as the frequency is increased (up to 196 MHz). In contrast to earlier torsional-oscillator measurements on this system, we observe a dissipation peak at the transition. We discuss the information which these results provide about the role of two-dimensional vortices in the superfluid transition in films adsorbed on porous substrates.

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Superfluidity in helium films has been the object of much theoretical and experimental interest. For films on flat substrates, the superfluid transition is of the Kosterlitz-Thouless type,[1] involving the unbinding of two-dimensional (2D) vortices.[2] For helium on porous substrates, however, finite size effects and the complex multiply connected geometry of the surface are important, to the extent that films adsorbed on porous Vycor glass appear to behave three dimensionally in the critical region.[3] In order to determine the role of 2D vortices in such films, information on the dynamics of film motion would be valuable. We report here results of the first measurements of the frequency dependence of the superfluid density and dissipation in helium films on Vycor, obtained by use of an ultrasonic technique. We observe a narrow dissipation peak at the superfluid onset and find that the transition broadens and shifts to higher temperatures as the frequency is increased.

A number of recent experiments have studied superfluidity in ^{4}He films on porous substrates. The results for both alumina[4] and platinum[5] powders were similar to the flat-substrate case, namely, a sudden decrease in the superfluid density at the superfluid transition accompanied by substantial dissipation. These features were interpreted in terms of the Kosterlitz-Thouless theory, with broadening of the transition due to finite size effects in the small powders. On the other hand, torsional-oscillator experiments[3] using Vycor substrates found a relatively sharp superfluid transition near which the superfluid density disappeared with a critical exponent close to that of bulk liquid helium, which was considered evidence that the transition was three dimensional. In these experiments, the dissipation peak characteristic of 2D vortices on flat substrates was not observed. Very recent experiments have used other glass substrates (silica xerogels and/or aerogels) with pore sizes similar to Vycor but with different microstructures. Torsional-oscillator measurements[6] of the superfluid density (for full pores) found sharp transitions but the critical exponent was different for each of the substrates. Specific-heat experiments[7] found sharp peaks at the superfluid transitions for films on both Vycor and xerogel substrates, an unexpected result since earlier measurements had shown only broad features.

Several authors have extended the Kosterlitz-Thouless theory to finite geometries. Initial work[4,8] considered vortices on the surface of isolated spheres and predicted a very broad transition with negligible dissipation in pores as small as those in Vycor. Recently, the theory has been generalized[9,10] to the case of an interconnected network of cylinders. With this model of a porous substrate, both the dependence of the transition temperature on film thickness and the existence of a 3D critical region could be explained. A quite different approach is to consider the substrate as a random potential acting on the helium[11] and it has been suggested[6,12] that it is the long-range structure of this "quenched disorder" that gives the different critical exponents for different substrates.

There is not yet a satisfactory understanding of superfluidity in films on porous substrates nor even agreement on the effective dimensionality of such systems. The frequency dependence of the superfluid transition can provide valuable information about the role of 2D vortices since their response depends on the measurement frequency as well as the pore size. Near the transition, most of the dissipation is due to vortex pairs with separations equal to the diffusion length $r_D = (2D/\omega)^{1/2}$, where D is the vortex diffusivity and ω is the drive frequency. This length is of the order of microns at torsional-oscillator or third-sound frequencies. In porous media, the restriction of vortex motion by the pore geometry is expected[4] to broaden the transition and reduce the associated dissipation. These finite size effects become less important than finite frequency effects when the diffusion length is smaller than the pore size. So far, the only experimental information on the effects of pore size on vortex motion comes from measurements using powders of different sizes.[4,5] However, since the microstructure is not identical in all powders, measurements as a function of frequency would be preferable. In Vycor, the vortex diffusion length would become smaller than pore dimensions at frequencies above about 12 MHz (estimating the diffusivity[13] D as $\hbar/m$ and the characteristic length scale of Vycor[14] as 200 Å). In order to see whether such a crossover occurs in helium films on Vycor, we have used an ultrasonic technique to extend the measurements of the superfluid density and dissipation to 200 MHz.

Our technique is to measure the sound velocity v and attenuation α of shear waves in the Vycor substrate. When the adsorbed film becomes superfluid, only the normal-fluid fraction remains viscously locked to the substrate and so the sound velocity increases by an amount proportional to the decoupled superfluid fraction:

$$\Delta v/v = \tfrac{1}{2}(1-x)\sigma_S S, \tag{1}$$

where σ_S is the areal density of the superfluid component and S is the sample's specific surface area. The fraction x of the superfluid which remains effectively locked to the substrate depends on the tortuousity of the pore surface. Ultrasonic measurements thus provide essentially the same information as does a torsional oscillator since the change in the sound speed is analogous to the oscillator's period shift and the sound attenuation is related to the damping Q^{-1} by

$$Q^{-1} = 2v\alpha/\omega. \tag{2}$$

The samples were made from commercial Vycor (Corning 7930 "Thirsty Glass") and were about 0.3 cm in diameter and 1 cm long. They were cleaned by heating at about 80°C in a 30% hydrogen peroxide solution for 1 h and rinsed in distilled water for several hours. The samples were air dried and then evacuated at room temperature for a few hours. Two 20-MHz $LiNbO_3$ ultrasonic transducers were bonded to the ends using a silicone fluid whose high viscosity prevented it from being drawn into the pores. The sample and transducers were mounted, using spring-loaded pins, into a copper cell (dead volume about 7 cm^3) which was then sealed with an indium O ring and evacuated. We have previously characterized an identically treated Vycor sample from the same batch.[15] The pore volume fraction was 29%, the specific surface area (from a nitrogen Brunauer-Emmett-Teller measurement) was 105 m^2/g, and mercury porosimetry gave a pore radius of 19 Å.

For the ultrasonic measurements a heterodyne technique was used to determine simultaneously the in-phase and quadrature components of the received sound pulse at each frequency. The velocity resolution of the system was $\Delta v/v \approx 10^{-6}$ so that we could detect the decoupling of 4×10^{-4} monolayers of superfluid. The system could also measure 0.005-dB attenuation differences, corresponding at 100 MHz to changes in the damping ΔQ^{-1} of 4×10^{-7}. About 15 min were required to take data at all frequencies before changing the temperature. Measurements were made at low signal amplitudes (peak power $< 10^{-6}$ W, duty cycle 0.1%) where tests showed that there was no sample heating or amplitude dependence of the signal.

When a sample (with an adsorbed film) is cooled below the superfluid transition temperature T_c, we find that the sound velocity increases and that there is an attenuation peak at T_c. In order to separate the changes due to superfluidity in the helium films from those due to the glass itself, a temperature- and frequency-dependent background must be subtracted from the data. This background is due to the interaction of the sound wave with the two-level systems which are characteristic of amorphous materials.[16] The presence of helium in the pores is known[17] to increase the relaxation rates of the two-level system in the glass so we cannot simply subtract the empty-Vycor data. However, measurements with different film thicknesses show that the onset of superfluidity does not significantly change the background, so that we can determine it from the data far from the transition. For the sound velocity, we simply extrapolate the linear temperature dependence observed above the transition, and for the attenuation, we fit the data on both sides of the peak by a low-order polynomial.

Figure 1(a) shows the sound velocity (after back-

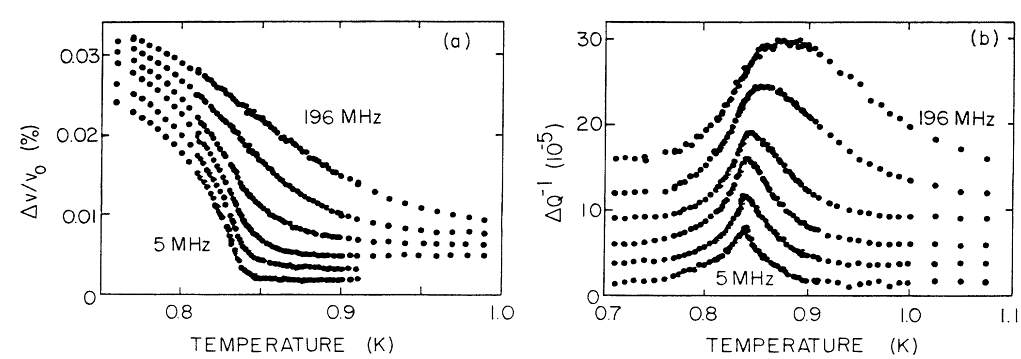

FIG. 1. (a) Velocity and (b) damping of ultrasonic shear waves. Curves for different frequencies have been vertically displaced for clarity. From bottom to top, the sound frequencies are 5, 8, 16, 30, 83, and 196 MHz.

ground subtraction) at frequencies from 5 to 196 MHz for one film thickness. The decoupling of the superfluid fraction is seen as a velocity increase. The transition is quite sharp at the lowest frequencies but becomes progressively more rounded at higher frequencies. At 5 MHz there is a rounded tail region extending about 5 mK above the transition. This is comparable to the rounding seen[3] in torsional-oscillator experiments at 1570 Hz, indicating that there is very little frequency dependence up to 5 MHz. By fitting the low-frequency data close to the transition, we can find a value of the critical exponent for the superfluid density. For all films, the data are consistent with the bulklike exponent of $\frac{2}{3}$ observed in the torsional-oscillator experiments, but this has a considerable uncertainty due to the rounding near the transition.

Figure 1(b) shows the corresponding attenuation changes [converted to ΔQ^{-1} using Eq. (2)]. In contrast to torsional-oscillator experiments, there is a clear dissipation peak. At the lowest frequencies, this peak is sharp and its maximum occurs at the transition temperature indicated by the velocity data. As the frequency increases, the peaks grow and become rounded. They become much wider and the maximum shifts to higher temperature. The broadening is asymmetric, occurring almost totally above the transition temperature.

We made measurements for a number of different helium coverages. Transition temperatures ranged from 0.15 K for the thinnest superfluid film to 1.94 K for full pores. Figure 2 shows the damping peaks for the film data at a single frequency (30 MHz). The peak height scales with the transition temperature and hence with the amount of superfluid present (since both are proportional to the film thickness[3]). The shape of the peaks and their frequency dependence are similar in all of the films.

Figure 3 summarizes the dependence of the damping peak height on frequency and on film thickness. We have divided the peak heights by the transition temperature for each film to illustrate the proportionality of the peak height to film thickness. Below 100 MHz the peak heights decrease with decreasing frequency for all films, although less rapidly for the thickest film. This decrease explains why torsional-oscillator experiments at kHz frequencies did not detect dissipation associated with the transition.

Within the temperature resolution of our data (better than 2 mK), the low-frequency peaks (16 MHz and below) are sharp cusps with maxima at identical temperatures. Together with the velocity data, this implies that the onset of superfluidity in helium films on Vycor is a genuine phase transition with an inherent rounding of at most a few mK.

The dissipation peaks can be compared to those seen in torsional-oscillator measurements on flat substrates. There, finite frequency effects shift the transition to temperatures slightly above its zero-frequency position and give the peak a finite width. The original experiments also observed additional dissipation below the transition,[2] but later experiments[13] indicated that this was not an inherent property of the films. Although these measurements were done at a single frequency, the transition is expected to broaden and shift to higher temperatures with increasing frequency.[4] The behavior which we observe (Fig. 1) is consistent with the predictions of the Kosterlitz-Thouless theory in finite geometries. The motion of "pore vortices" which is considered in recent theories[9,10] occurs on longer length scales and so will be important at much lower frequencies. At high frequencies, vortex pairs with separations smaller than pore dimensions will dominate, and the motion of vortices becomes essentially two dimensional. The characteristic frequency for films on Vycor was estimated above to be about 12 MHz. Our data show just the expected broadening and shift to higher temperatures at frequencies above 16 MHz. The similar frequency dependence in the different films can be taken to indicate that the vortex diffusivity D does not depend strongly on film

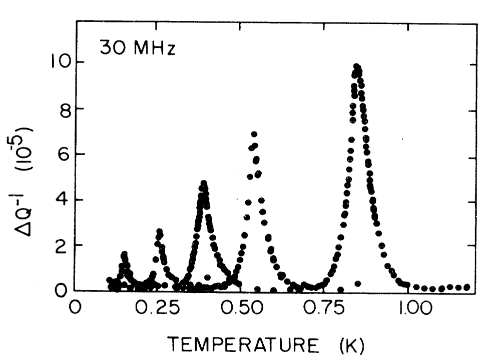

FIG. 2. Damping peaks at 30 MHz for different helium film thicknesses.

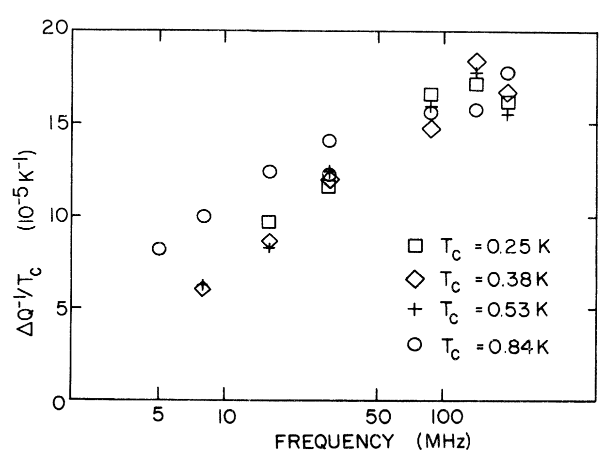

FIG. 3. Damping peak height ΔQ^{-1} divided by transition temperature T_c as a function of frequency and film thickness.

thickness.

Although the dissipation peaks have some resemblance to the critical attenuation seen in liquid helium at the lambda transition, the scaling with frequency is quite different.[18] In bulk helium, the attenuation peak is much narrower and occurs slightly below the transition temperature. As the frequency increases this peak broadens on both sides of the transition and its maximum shifts to lower temperature, in contrast to the behavior shown in Fig. 1(b). We have also observed that, when the Vycor pores are completely filled, the dissipation peaks are comparable or even smaller in size than those in the film with $T_c = 0.84$ K, even though the full pores contain roughly 5 times as much superfluid. This suggests a different dissipation mechanism in full pores. Because of the large dead volume of our sound cell, desorption of helium above 1.2 K prevents us from using thicker films to study this behavior in detail.

Further experiments using other substrates will clarify the relationship between the role of vortices in the superfluid transition and the pore size and structure of the porous material on which the films are adsorbed.

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[1]J. M. Kosterlitz and D. J. Thouless, J. Phys. C **6**, 1181 (1973); V. Ambegaokar, B. I. Halperin, D. Nelson, and E. D. Siggia, Phys. Rev. B **21**, 1806 (1980).

[2]D. J. Bishop and J. D. Reppy, Phys. Rev. B **22**, 5171 (1980).

[3]D. J. Bishop, J. E. Berthold, J. M. Parpia, and J. D. Reppy, Phys. Rev. B **24**, 5047 (1981).

[4]V. Kotsubo and G. A. Williams, Phys. Rev. B **33**, 6106 (1986).

[5]K. Shirihama, N. Wada, Y. Takano, T. Ito, and T. Watanabe, Jpn. J. Appl. Phys. Suppl. **26-3**, 293 (1987).

[6]K. I. Blum, S. Murphy, M. H. W. Chan, D. D. Awschalom, and J. D. Reppy, Jpn. J. Appl. Phys. Suppl. **26-3**, 275 (1987); M. H. W. Chan, K. I. Blum, S. Q. Murphy, G. K. S. Wong, and J. D. Reppy, Phys. Rev. Lett. **61**, 1950 (1988).

[7]D. Finotello, A. Wong, K. A. Gillis, D. D. Awschalom, and M. H. W. Chan, Jpn. J. Appl. Phys. Suppl. **26-3**, 283 (1987); D. Finotello, K. A. Gillis, A. Wong, and M. H. W. Chan, Phys. Rev. Lett. **61**, 1954 (1988).

[8]C.-T. Wang and L. Yu, Phys. Rev. B **33**, 599 (1986).

[9]T. Minoguchi and Y. Nagaoka, Jpn. J. Appl. Phys. Suppl. **26-3**, 327 (1987) and Prog. Theor. Phys. **80**, 397 (1988).

[10]J. Machta and R. A. Guyer, Phys. Rev. Lett. **60**, 2054 (1988).

[11]M. Ma, B. I. Halperin, and P. A. Lee, Phys. Rev. B **34**, 3136 (1986).

[12]P. B. Weichmann and M. E. Fisher, Phys. Rev. B **34**, 7652 (1986).

[13]P. W. Adams and W. I. Glaberson, Phys. Rev. B **35**, 4633 (1987).

[14]P. Wiltzius, F. S. Bates, S. B. Dierker, and G. D. Wignall, Phys. Rev. A **36**, 2991 (1987).

[15]K. L. Warner and J. R. Beamish, J. Appl. Phys. **63**, 4372 (1988).

[16]S. Hunklinger and M. von Schickfus, in *Amorphous Solids; Low Temperature Properties,* edited by W. A. Phillips (Springer-Verlag, Berlin, 1981).

[17]J. R. Beamish, A. Hikata, and C. Elbaum, Phys. Rev. Lett. **52**, 1790 (1984).

[18]J. S. Imai and I. Rudnick, Phys. Rev. Lett. **22**, 694 (1969).