Superfluid-Insulator Transition in ^{4}He Films Adsorbed in Vycor Glass

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We have measured the heat capacity of ^{4}He films adsorbed in Vycor glass for temperatures down to 10 mK and coverages close to the critical coverage for the onset of superfluidity at $T = 0$. The data show that the critical coverage corresponds to a mobility edge in the chemical potential, below which the ^{4}He film is an insulator with a gap. We find no evidence for the existence of a gapless Bose glass phase.

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Increasing attention has been focused in recent years on the role disorder plays in destroying superfluidity in Bose systems. This has been evident in the theoretical and experimental efforts devoted to understanding flux pinning in high-T_c superconductors [1] as well as transport in thin superconducting films [2]. Much of the interest in these problems lies in identifying the nature of the insulating phases that appear in these systems as the disorder is increased. This line of inquiry draws heavily on the analogy with the localization problem for fermions, and the concept of a superfluid-insulator transition tuned by the chemical potential or external field has now received considerable theoretical attention [3,4]. Fisher *et al.* [4] have proposed that the onset of superfluidity in ^{4}He films adsorbed on disordered substrates as the ^{4}He coverage is increased is an example of a superfluid-insulator transition. Furthermore, they argue that the insulating phase is a "Bose glass" with a gapless excitation spectrum. This approach is different from the interpretation usually adopted for superflow measurements, in which the film is considered as two parts: a solid layer of density n_c and an overlying superfluid film [5]. Although this "inert-layer" model is adequate for all but the thinnest superfluid films [5,6], it does not address questions regarding the nature of ^{4}He films below onset. Heat-capacity [7,8] and ultrasound [9] measurements indicate that the excitation spectrum of nonsuperfluid films is considerably richer than that of a solid, and a model such as that proposed by Fisher *et al.* may be more appropriate. None of these measurements, however, have been able to confirm whether or not the low-energy ($E/k_B < 100$ mK) excitation spectrum of nonsuperfluid films is gapless as the onset coverage is approached from below.

In this Letter, we report on a detailed high-precision study of the heat capacity of ^{4}He films adsorbed on Vycor [10] for reduced coverages $|n/n_c - 1|$ down to 0.04 and for temperatures down to 10 mK [11]. We find a mobility edge at $n = n_c$ below which all states occupied at $T = 0$ are localized, and we have established that the characteristic energy separating localized and extended states vanishes continuously as the onset transition is approached from below. These results are qualitatively consistent with the existence of a superfluid-insulator transition. We find, however, that the form of the heat capacity of nonsuperfluid films suggests a gap in the excitation spectrum at low energies, in disagreement with the prediction of Fisher *et al.* [4] that the insulating phase should be gapless.

The measurements reported here were conducted in a calorimeter designed to satisfy the requirements of high temperature resolution, low background heat capacity, and rapid thermal response for temperatures down to 10 mK. The substrate comprised 14 Vycor disks of thickness 0.3–0.6 mm. Each disk was bonded to two 0.025 mm thick Ag foils, which were diffusion welded to the wall of the Ag calorimeter. A tin heat switch controlled the thermal contact to the calorimeter. Two different thermometers were used: a 1 kΩ RuO_2 resistor and a miniature CMN magnetization thermometer [12]. The measurements were conducted using the adiabatic calorimetry technique.

The low-temperature heat-capacity data, divided by the temperature T, are shown in Fig. 1. Each curve in the lower panel shows a well-defined cusp, which is known to coincide with the superfluid transition temperature T_c [13]. From these data, we find a line of T_c's which follows that found in earlier superfluid density measurements [5]. The coverage is set by assuming that the onset of superfluidity, determined by extrapolation of the curve of T_c's, occurs at 27 μmole/m^2. The superfluid T_c is shown as a function of coverage in Fig. 2. Some points from the superfluid density study of Crooker *et al.* [5] are included for comparison.

The curves in the upper panel of Fig. 1, which were obtained for lower coverages, show no superfluid transition, but C/T drops in each case from a nearly constant value at high temperatures to a much smaller value as $T \to 0$. This drop is much stronger than that seen for the superfluid films in the regime $T < T_c$, where C/T is nearly linear. We have characterized the crossover for the nonsuperfluid films by defining a break temperature T_B at which C/T is half of its high-temperature value. T_B is shown as a function of coverage in Fig. 2.

Figure 2 shows the central result of this paper, namely, the existence of a characteristic temperature T_B, which vanishes as the onset of superfluidity is approached from

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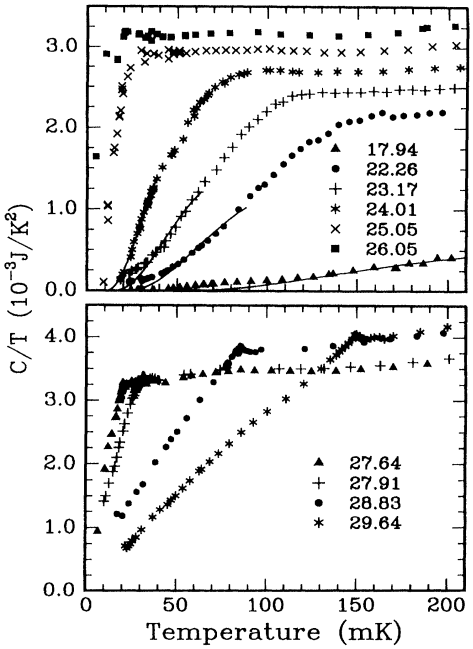

FIG. 1. The heat capacity, divided by the temperature T, is shown for nonsuperfluid (upper panel) and superfluid (lower panel) films. The coverages in μmole/m^2 are indicated in the legend. The solid curves are fits for the density of states discussed in the text.

below. Although the definition of T_B adopted here is arbitrary, any definition of a characteristic temperature for the drop in C/T in the upper panel of Fig. 1 would show a similar coverage dependence. For $T > 2T_B$, the heat capacity for nonsuperfluid films depends linearly on temperature, as it does for superfluid films above T_c. In fact, it is difficult to distinguish a film of density $n > n_c$ at a temperature $T > T_c$ from a film of density $n < n_c$ at $T > 2T_B$. This is emphasized by the existence of the wedge-shaped plateau region in the three-dimensional representation of C/T shown in Fig. 3. Qualitatively, these observations are similar to those made by Tait and Reppy [7] and Finotello *et al.* [8] for the same system at higher temperatures and for coverages farther from the onset of superfluidity. The new measurements establish an upper bound of 10 mK for the lowest value of T_B, corresponding to a coverage within 4% of n_c. We emphasize that these values of reduced coverage and T_B are on the order of the reduced coverage and T_c for the thinnest *superfluid* ^{4}He films studied to date [5].

Figures 1 and 3 show that the excitation spectrum of the nonsuperfluid film undergoes a significant change as the coverage is increased. We now consider this observation in the context of the superfluid-insulator transition introduced above. The insulating phase in the presence of strong disorder is believed to be a Bose glass, which is characterized by the absence of a gap for particle-hole excitations and a correspondingly nonzero compressibility. The transition from a Bose glass to a superfluid as the chemical potential is increased has been studied in detail by Fisher *et al.* [4], who emphasize the importance of quantum fluctuations, which assume the role played by thermal fluctuations at an ordinary second-order phase transition.

Assuming the existence of a compressible Bose glass phase below onset, Fisher *et al.* have derived a scaling theory for the onset of superfluidity. An important component of this theory is the statement that the temperature dependence of all thermodynamic quantities should scale with a characteristic energy $\Omega(n)$ set by the frequency scale for quantum fluctuations. The heat capacity in the limit $\mu \rightarrow \mu_c$ (μ_c is the critical value of the chemical potential and corresponds to the critical density n_c) and $T \rightarrow 0$ is predicted to vary linearly with T. A simi-

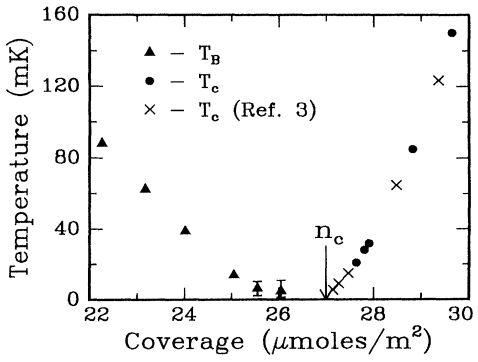

FIG. 2. The break temperature T_B defined in the text is shown as a function of coverage using triangles. The superfluid transition temperature for superfluid films is shown using circles (current work) and crosses (Crooker *et al.* [5]). The onset coverage n_c is indicated with an arrow.

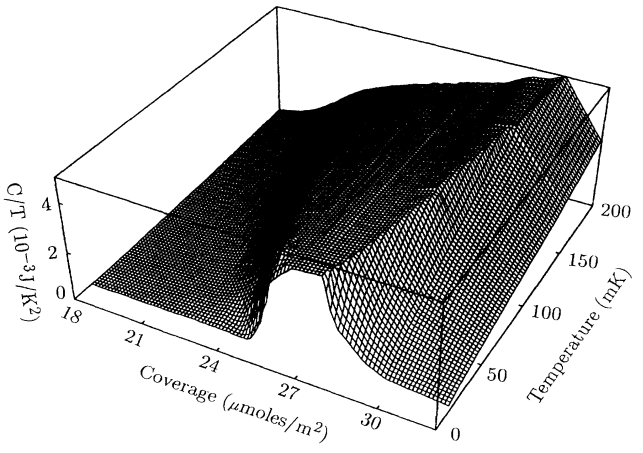

FIG. 3. C/T is shown as a function of temperature and coverage.

lar temperature dependence is expected in the Bose glass phase for $n < n_c$. Moreover, the Bose glass contribution should dominate for temperatures $T \ll \Omega(n)/k_B$ and should disappear entirely only at $n = n_c$, where $\Omega = 0$. This model thus predicts two different regimes of linear behavior for coverages $n < n_c$, with the low-temperature Bose glass regime shrinking as $n \rightarrow n_c$.

The data of Fig. 1 show only a single linear regime for all coverages $n < n_c$, in disagreement with the predictions of the Bose glass model. Even if the Bose glass contribution is for some reason immeasurably small, it is unlikely that the observed linear contribution is due to quantum fluctuations. Previous measurements [7,8] have shown that the linear contribution persists up to temperatures of 1 K, while superfluid density measurements suggest an upper limit of approximately 200 mK for the regime in which quantum fluctuations might be relevant [6].

The rapid drop of C/T in the vicinity of T_B for the nonsuperfluid coverages in Fig. 1 suggests a gap in the density of states, in contrast to the predictions of the Bose glass model. This possibility, noted originally by Tait and Reppy [7], led them to consider a density of states consisting of two parts. The first of these is a low-energy band of localized states, which are each singly occupied at $T = 0$ because of the strongly repulsive He-He interactions. The second part of the spectrum is a band of extended states, unoccupied at $T = 0$, separated from the localized states by a gap Δ. These states are assumed to obey Bose statistics. The density of states within each band is presumed to be a constant. This model leads to a heat capacity of the form

$$C = D(\Delta/2k_BT + 2)e^{-\Delta/2k_BT} \quad (1)$$

for $k_BT \ll \Delta$. The prefactor $D = \sqrt{N_lN_u}\Delta k_B$, where N_l and N_u are the densities of states in the lower and upper bands. We have fitted our data by Eq. (1) for coverages n up to 24 μmole/m^2 and temperatures $T < T_B(n)$. The fits are shown as solid curves in the upper panel of Fig. 1. In each case, we find a gap of order 5–6 times T_B. Like T_B, the gap decreases as the onset transition is approached from below. In contrast, the prefactor D is constant to within 25% over the same coverage range.

Although these fits reproduce the drop in C/T, the actual data are consistently above the fitted curves at the lowest temperatures. It is therefore reasonable to ask if there is a picture intermediate between a glass, in which the density of states is constant and the heat capacity is linear in T, and the model of the previous paragraph. One possibility is a system with a range of activation energies. We believe that the ^{4}He-Vycor system is likely to include such a distribution, but the sharp dropoffs seen in the upper panel of Fig. 1 suggest that the activation energies will be spread over less than an order of magnitude. The density of states will still vanish at low energies, distinguishing the system from a glass.

One of the strengths of the thermal activation model, whatever the detailed structure of the density of states, is that it provides a natural explanation of the linear behavior at high temperatures on *both* sides of the onset transition. For nonsuperfluid films in the temperature regime $T \gg T_B$, the band of extended states, which are occupied by thermally activated particles, makes the dominant contribution to the heat capacity. A linear heat capacity follows from the assumption of a constant density of states. The only difference for films of density $n > n_c$ is that the extended states are occupied at zero temperature. The temperature dependence of the heat capacity at high temperatures, $T > T_c$, should therefore be the same as for films below onset. As shown in Fig. 4, the linear coefficient A in a fit of the high-temperature heat capacity by the form $C(T) = AT + BT^2$, $T < 400$ mK, is almost unchanged at the onset transition, in agreement with this argument. Interestingly, the coefficient B increases rapidly at the onset of superfluidity, although the quadratic contribution remains at least a factor of 2 smaller than the linear heat capacity for temperatures less than 400 mK.

The critical density n_c in the thermal activation model thus corresponds to a mobility edge μ_c in the chemical potential. Extended states are first occupied at $T = 0$ for films of density n_c. The gap Δ at lower coverages is due to inhomogeneities in the adsorption potential. The magnitude of the gap, however, poses an interesting problem. The scale for Δ is set by $\sim 5T_B$, which varies from 500 to 25 mK as the coverage increases from 22 to 27 μmole/m^2. Assuming a van der Waals constant α of (26 K)(layer)^3 for ^{4}He on glass, and a monolayer coverage of 13 μmole/m^2 [14], the chemical potential $\mu(n) = -\alpha/n^3$ increases by 2–3 K over the same coverage range. The gap $\Delta(n)$ is therefore much smaller than $\mu_c - \mu(n)$ over the range of our experiment, and we conclude that the bottom of the band of extended states does not remain at a fixed energy as the coverage changes. This observation indicates the limits of our single-particle picture. Because of the strong ^{4}He-^{4}He

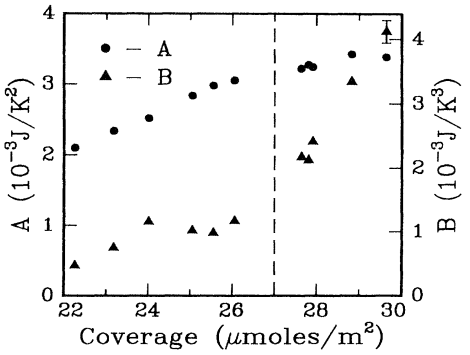

FIG. 4. The coefficients A (circles) and B (triangles) in a fit of the high-temperature heat capacity by the form $C(T) = AT + BT^2$. The dashed line indicates the critical coverage.

interactions, we cannot think of the atoms independently filling states of a disordered adsorption potential in the way that weakly interacting electrons fill the states of an Anderson insulator. The disorder in the ^{4}He case is strongly screened by the adsorbed film. As a result, the gap $\Delta(n)$ changes less rapidly with coverage than the chemical potential.

Another question of interest is the nature of the extended states. For example, are they two dimensional (2D) or three dimensional (3D) in character? This question is of importance in determining the effective dimensionality for the onset transition, a prerequisite for comparing the results of the superfluid density measurements [5,6] with the predictions of the scaling theory of Ref. [4]. Furthermore, the dimensionality itself may give some indication as to whether the van der Waals potential of the substrate plays a role beyond that of determining the critical coverage n_c. In the 3D case, for example, Fisher *et al.* assume a linear expansion of the density difference $n - n_c$ in the chemical potential difference $\mu - \mu_c$. The rapid change of the van der Waals potential with coverage, however, introduces large corrections which make the asymptotic regime of their scaling theory inaccessible experimentally.

One means of addressing the question of dimensionality is to consider the heat capacity for superfluid films in the region $T < T_c$. If the elementary excitation spectrum of thin superfluid films is dominated by phonons, the T^2 dependence of the heat capacity in the lower panel of Fig. 1 indicates that the thin superfluid films are effectively 2D. The existence of the singularity at T_c, however, is consistent with 3D behavior [8,13]. Although a crossover from 2D to 3D as $T \rightarrow T_c$ is accounted for in several theories of ^{4}He films in porous media [15], we emphasize that the data in the superfluid regime are not understood. The mechanism by which the small fluctuation peak at high coverages becomes a simple cusp at very small values of $n - n_c$ is not known. One would expect to see only a nonsingular background once the fluctuation peak becomes too small to resolve experimentally. It is possible that the observed behavior reflects a slow crossover to a weakly interacting Bose gas regime such as that implied by superfluid density measurements [5,16], but measurements at lower temperatures and reduced coverages $n - n_c$ will be necessary to resolve this question.

In conclusion, we have demonstrated the existence of a mobility edge in the ^{4}He-Vycor system. For films less than the critical density n_c, a gap separates the localized states occupied at $T = 0$ from a band of extended states at higher energies. For densities $n > n_c$, the extended states are occupied at $T = 0$, allowing for superfluidity. Although the measurements presented here lend qualitative support to the concept of a superfluid-insulator transition, the data are not in agreement with the Bose glass model of Fisher *et al.*, in which the nonsuperfluid phase is expected to be gapless.

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[1] D. R. Nelson and V. M. Vinokur, Phys. Rev. Lett. **68**, 2398 (1992); Phys. Rev. B **48**, 13 060 (1993); C. J. van der Beek *et al.*, Phys. Rev. Lett. **74**, 1214 (1995).
[2] Y. Liu *et al.*, Phys. Rev. Lett. **67**, 2068 (1991); M. A. Paalanen, A. F. Hebard, and R. R. Ruel, *ibid.* **69**, 1604 (1992).
[3] J. L. Hertz, L. Fleishman, and P. W. Anderson, Phys. Rev. Lett. **43**, 942 (1979); M. Ma, B. I. Halperin, and P. A. Lee, Phys. Rev. B **34**, 3136 (1986); D. K. K. Lee and J. M. F. Gunn, J. Phys. Condens. Matter **2**, 7753 (1990); W. Krauth, N. Trivedi, and D. Ceperley, Phys. Rev. Lett. **67**, 2307 (1991); K. G. Singh and D. S. Rokshar, Phys. Rev. B **46**, 3002 (1992).
[4] M. P. A. Fisher, P. B. Weichman, G. Grinstein, and D. S. Fisher, Phys. Rev. B **40**, 546 (1989).
[5] B. C. Crooker *et al.*, Phys. Rev. Lett. **51**, 666 (1983).
[6] P. A. Crowell, M. Y. Lanzerotti, and J. D. Reppy, J. Low Temp. Phys. **89**, 629 (1992).
[7] R. H. Tait and J. D. Reppy, Phys. Rev. B **20**, 997 (1979).
[8] D. Finotello, K. A. Gillis, A. Wong, and M. H. W. Chan, Phys. Rev. Lett. **61**, 1954 (1988); M. H. W. Chan *et al.*, in *Excitations in Two-Dimensional and Three-Dimensional Quantum Fluids,* edited by A. F. G. Wyatt and H. J. Lauter (Plenum, New York, 1991), p. 301.
[9] N. Mulders, E. Molz, and J. R. Beamish, Phys. Rev. B **48**, 6293 (1993).
[10] Vycor as discussed in this paper is Corning Glass 7930, a precursor to the product sold comercially as Vycor.
[11] A preliminary report on some of our results has been published previously: F. W. Van Keuls, P. A. Crowell, and J. D. Reppy, Physica (Amsterdam) **194-196B**, 623 (1994).
[12] D. S. Greywall and P. Busch, Rev. Sci. Instrum. **60**, 471 (1989); T. C. P. Chui and J. A. Lipa, in *Proceedings of LT-17,* edited by U. Eckern *et al.* (North-Holland, Amsterdam, 1984), p. 931.
[13] S. Q. Murphy and J. D. Reppy, Physica (Amsterdam) **165&166B**, 547 (1990).
[14] E. S. Sabisky and C. H. Anderson, Phys. Rev. Lett. **30**, 1122 (1973).
[15] T. Minoguchi and Y. Nagaoka, Prog. Theor. Phys. **80**, 397 (1988); J. Machta and R. A. Guyer, Phys. Rev. Lett. **60**, 2054 (1988).
[16] P. B. Weichman, M. Rasolt, M. E. Fisher, and M. J. Stephen, Phys. Rev. B **33**, 4632 (1986); P. B. Weichman, *ibid.* **38**, 8739 (1988); P. B. Weichman and K. Kim, *ibid.* **49**, 813 (1989).