Heat Capacity and Superfluid Density of Thin ^{4}He Films on Porous Gold and on H_2

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We report simultaneous measurement of the superfluid density and heat capacity of thin ^{4}He films on porous gold and on gold preplated with H_2. The superfluid density shows a crossover from 2D-like to 3D critical behavior for temperatures close to T_c, the transition temperature. On both substrates we observe an unexpected heat capacity signature associated with the superfluid layer well below T_c. [S0031-9007(98)06076-1]

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The superfluid transition of thin helium films adsorbed on a two dimensional surface has been shown [1,2] to be an excellent realization of the Kosterlitz-Thouless (KT) theory [3]. In particular, the superfluid density ρ_s vanishes abruptly at T_{KT}. At temperatures just below the transition, ρ_s is directly proportional to T_{KT}:

$$\rho_s(T_{\mathrm{KT}}^-) = \frac{2k_B m^2}{\pi\hbar^2}\, T_{\mathrm{KT}}\,. \tag{1}$$

Here k_B is the Boltzmann constant, m the mass of the ^{4}He atom, and $\hbar$ the Planck constant. The heat capacity of the film is predicted not to show a peak at T_{KT}, but to exhibit a broad maximum at a temperature above T_{KT} [4]. The height of the peak is predicted to be of the order of one k_B per atom. Heat capacity peaks were found for ^{4}He films adsorbed on Millipore and Anopore filter papers [5] with pore diameters of 5000 and 2000 Å, respectively. Since these experiments measured only the heat capacity, the location of the observed peaks could not be correlated with the superfluid transition temperature.

For atomically thin films adsorbed on a porous substrate one expects the KT behavior to be modified by the finite curvature of the substrate and the interconnectivity of the pores. Torsional oscillator [6] measurements of ^{4}He films adsorbed on porous Vycor glass (with a porosity of 30% and characteristic pore diameter of 70 Å), in fact, showed that the superfluid density exhibits power law dependence of the form $\rho_s = \rho_{s0}t^{\zeta}$ for $t < 0.1$, where $t = 1 - T/T_c$ is the reduced temperature. The exponent ζ was found to be 0.635 ± 0.050, which within the uncertainty is indistinguishable from the bulk ^{4}He value of 0.6705 ± 0.0006 [7]. In addition, a heat capacity peak is found precisely at T_c [8]. These results led to the interpretation that the transition is a 3D critical phase transition. A 3D phase transition is thought to be possible since the correlation length $\xi(t)$ of the film for $t < 0.1$ deduced from Josephson's relation [9]

$$\xi(t) = \xi_0 t^{-\zeta} = \frac{k_B T_c m^2}{\hbar^2}\,\frac{1}{\rho_s(t)} \tag{2}$$

exceeds the characteristic length of the porous structure in Vycor.

The contrasting 2D versus 3D interpretation of a thin helium film on a porous substrate is reconciled in a number of recent papers [10]. The authors model the porous substrate as interconnected cylinders on a square lattice. They find that far below T_c, ρ_s exhibits planar KT-like behavior. As the temperature is brought close to T_c, a 3D XY critical region is found. The results of Shirahama *et al.* [11] on Vycor-like porous glass of varying pore diameter and Cho *et al.* [12] on slip-cast alumina powder are qualitatively consistent with the above picture.

In this Letter we present simultaneous torsional oscillator and heat capacity measurements of thin helium films adsorbed on porous gold and H_2 preplated porous gold. The superfluid density results on both substrates show a 2D to 3D crossover as temperature is brought towards T_c and, to our surprise, we find a heat capacity anomaly related to the superfluidity of the film that is clearly below the superfluid onset temperature.

The experimental cell used in this study is the same as that used in a recent torsional oscillator experiment of filled pore ^{4}He near the superfluid transition [13]. It has been modified to simultaneously perform heat capacity measurements. The resonant frequency of the oscillator is 578 Hz with a quality factor of 8×10^5 at low temperature. An ac calorimetric technique with 1 Hz heating frequency is used. The resolution and reproducibility of the heat capacity results are found to be $\pm 0.3\%$.

The substrate is a disk of porous gold 1 cm in diameter and 0.5 mm in height. It is made by selectively leaching silver out of a gold-silver alloy with 70 at.% silver, and, as a consequence, the gold has a porosity of 70% [13,14]. Scanning electron microscope pictures show that the substrate can be characterized as interconnected gold strands of a uniform diameter. The distance between the connections of these strands is 1 to 3 times their diameter. The remaining open space forms an interconnected pore structure. Our methane vapor pressure isotherm reveals a total surface area of 0.82 m^2. If we assume a cylindrical model for the gold strands, their diameter is calculated to be 610 Å. On the other hand, if we assume, less realistically, a model where the pore volume is an array

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of uniform cylinders, then the pore diameter is 1300 Å. This pore diameter is larger than that given in Ref. [13] due to heating of the cell which leads to coarsening of the porous structure [14].

In Fig. 1 we show ρ_s versus T at several surface coverages of ^{4}He on bare Au and of ^{4}He on Au preplated with 4.2 layers of H_2, which will be referred to as the H_2 surface. The superfluid density data on both substrates show KT behavior on a coarse temperature scale as indicated by the location of the KT line. However, very close to the onset temperature, ρ_s shows a power law dependence. This is shown in Fig. 2 together with the dissipation and the heat capacity. The dashed lines in this figure are fits to the data between $10^{-2.5} < t < 10^{-1.5}$ of the form $\rho_s = \rho_{s0} t^{\zeta}$, keeping the exponent ζ fixed at 0.67 and having T_c and ρ_{s0} as fitting parameters. Analysis of the data by treating ζ, as well as T_c and ρ_{s0}, as a free parameter is difficult since the results of the fit are highly dependent on the range of data included.

Figure 3 shows ρ_s versus t in a log-log scale for various ^{4}He coverages on H_2. Although the data of these five ^{4}He coverages deviate from the power law fit at different values of t, the correlation length given by Eq. (2) at these different crossover temperatures is approximately the same, 4200 Å for all coverages. A reasonable explanation is that the superfluid data indicate a crossover from a KT-like to a 3D critical behavior as T is increased towards T_c. As ξ exceeds 4200 Å, which is approximately 7 times the diameter of the gold strands, the interconnected porous structure appears homogeneous to the superfluid, and the adsorbed film becomes a three dimensional system. In this interpretation deviations from the power law for t less than 10^{-3}, where ξ exceeds 2 μm, are the result of macroscopic inhomogeneity of the porous gold. To our knowledge, porous gold is the only substrate other than Vycor [6] on which 3D critical power law behavior of superfluid films have been observed.

Our ρ_s data on bare gold and H_2 are similar, except for the minimum ^{4}He coverage required for superfluidity or the inert layer n_0. On gold n_0 is found to be 26.0 μmol/m^2, which corresponds to 2.2 monolayers at bulk liquid density. The inert layer on H_2 is found to be 6.1 μmol/m^2, which is in reasonable agreement with that found in other recent studies [15]. Another difference

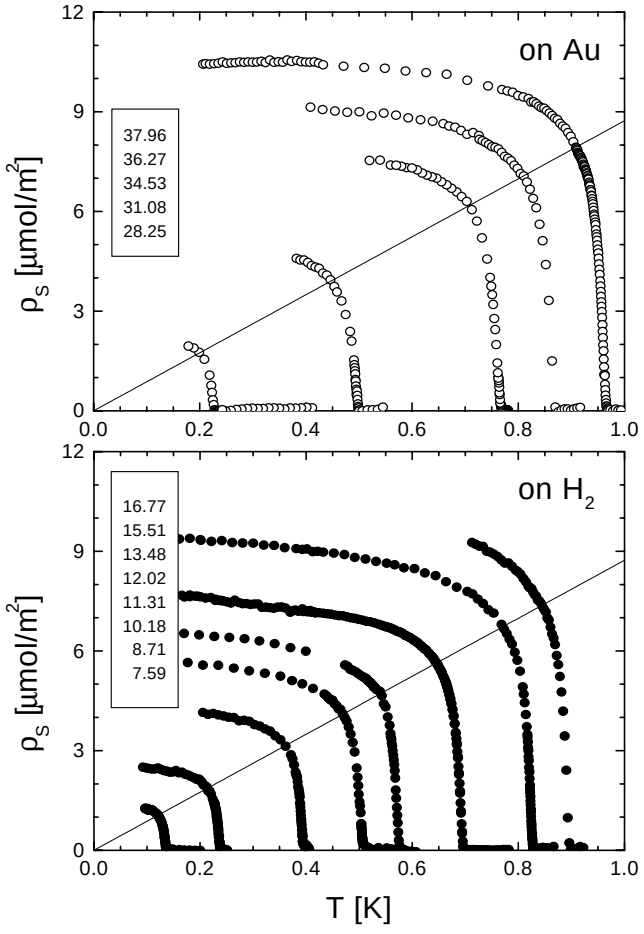

FIG. 1. Superfluid density versus temperature of helium films on porous gold (upper plot) and gold preplated with H_2 (lower plot). Coverages are in μmol/m^2. The KT prediction is shown as a solid line.

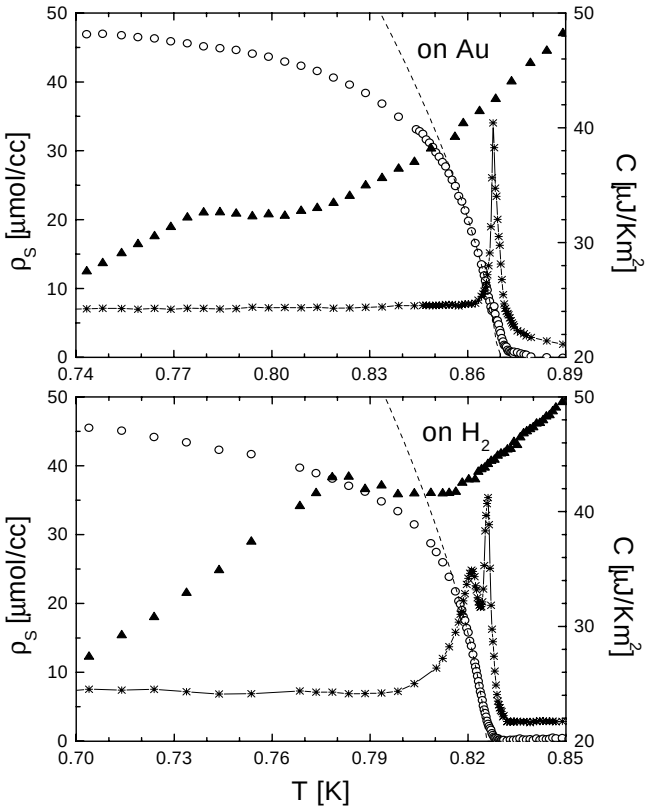

FIG. 2. An expanded view of the transition region for the film of 36.25 μmol/m^2 on porous gold (upper plot) and 15.51 μmol/m^2 on the H_2 surface (lower plot). We used open circles for the superfluid density, stars for the dissipation in the film, and filled triangles for the heat capacity. The dissipation is shown in relative units.

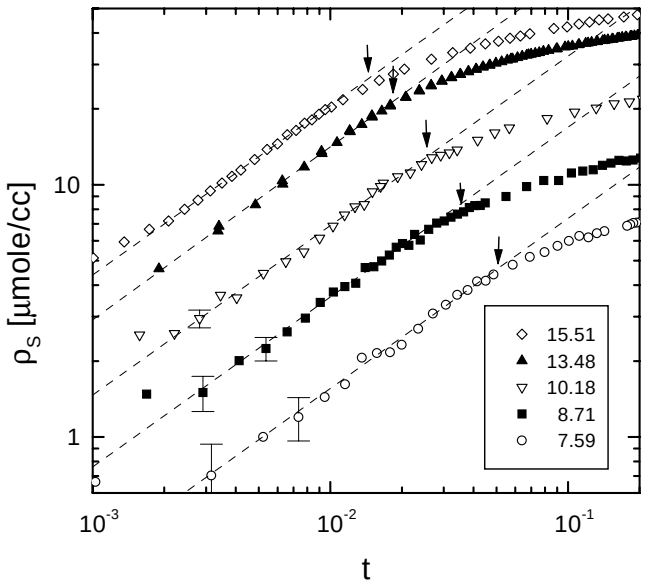

FIG. 3. Superfluid density versus reduced temperature for several films on H_2. The dashed lines are power law fits to data with the exponent fixed at 0.67. Coverages are in μmol/m^2. Arrows are placed where the correlation length of the film is 4200 Å.

is the dissipation near the onset temperature. Whereas on gold a single dissipation peak is found very close to the transition temperature, similar to that found in porous glass by Shirahama *et al.* [11], on H_2 two distinct peaks are present.

In contrast to ^{4}He films in Vycor [8], there is no evidence of any heat capacity peak at T_c. The explanation lies in the hyperuniversality hypothesis [16]. For different systems in the same universality class the product of the singular part of the heat capacity and ξ_0^3 is a constant. Because of the larger values of ξ_0, the expected heat capacity peak for the ^{4}He film in porous gold is about 3×10^{-7} times that of bulk ^{4}He and almost 75 times smaller than that of ^{4}He film in Vycor. Such a small peak is beyond the resolution of our measurements.

While we do not see any heat capacity peak at T_c, Fig. 2 shows a heat capacity peak at a temperature clearly below T_c and outside the 3D critical region. The position of this peak is inconsistent with the KT prediction [4]. Heat capacity versus T/T_c for a few film thicknesses is shown in Fig. 4 for Au and H_2 substrates. The maxima of the heat capacity peaks are situated around $T = 0.90T_c$ for ^{4}He on Au and near $T = 0.95T_c$ for ^{4}He on H_2. No anomaly in the heat capacity is seen for films with $T_c < 0.6$ K (see insets of Fig. 4). For $T_c > 0.6$ K, the magnitude of the peak first increases, shows a maximum for films with T_c close to 0.8 K, then decreases with increasing onset temperature. The size of the peak is similar on bare gold and on H_2 for comparable values of T_c, suggesting that it scales not with the total amount of ^{4}He but with the superfluid coverage (i.e., total coverage

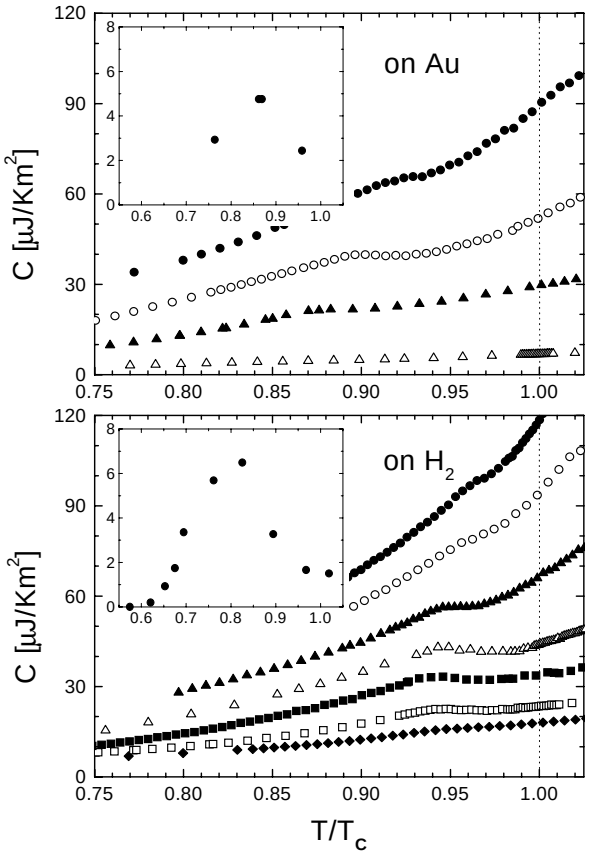

FIG. 4. Heat capacity of various ^{4}He films on porous gold (upper plot) and on H_2 preplated gold (lower plot). T_cs are 497, 764, 869, and 962 mK on Au and 653, 695, 761, 826, 894, 968, and 1019 mK on H_2. Insets indicate the height of the heat capacity anomalies (in μJ/Km^2) versus T_c of the corresponding film after a linear background has been subtracted.

minus n_0). For films with T_c of 0.8 K, where the peak is the highest, the thickness of the superfluid layer is close to that of one single monolayer. The height of the peaks is less than $0.1k_B$ per atom when normalized by the number of atoms in the superfluid layer, which is an order of magnitude less than the predicted value for a KT system [4]. The heat capacity peak is also present on other substrates (HD, D_2, Ne, and Ar) we have studied. The height of the heat capacity peaks found in Anopore and Millipore [5] shows a similar trend. However, these peaks [5] are 50% wider in temperature, and their maximum height is about 3 times larger when compared in units k_B per atom of the superfluid layer.

For multilayer ^{4}He films on graphite, a heat capacity peak related to the melting of the underlying solid layer has been seen [17]. The temperature of such a peak also tends to increase with total ^{4}He coverage. In order to determine whether the heat capacity peaks in Figs. 2 and 4 have such an origin, we have introduced ^{3}He into the film. Figure 5 shows the effect of such a dilution on a ^{4}He film on Au. These data show that T_c is shifted towards lower temperatures with increasing ^{3}He concentration. The

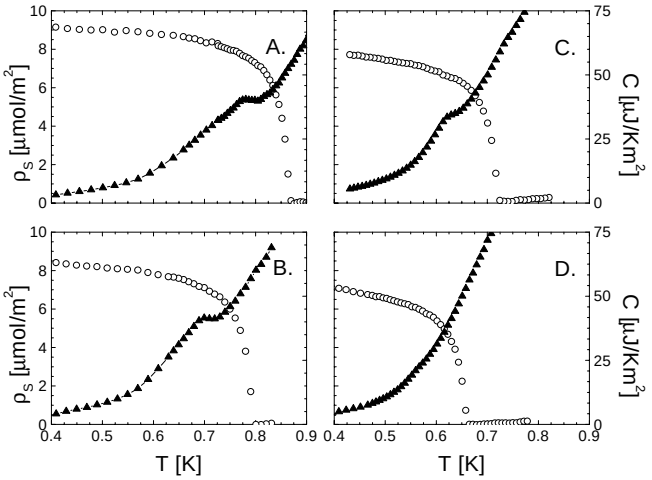

FIG. 5. The effect of ^{3}He dilution on ρ_s (open symbols) and heat capacity (filled symbols) of ^{4}He on Au. The ^{4}He coverage is 36.27 μmol/m^2 and the amounts of added ^{3}He are 0, 0.91, 1.89, and 2.86 μmol/m^2 for panels (A), (B), (C), and (D), respectively.

shift may be a result of ^{3}He dissolving into the ^{4}He film or of ^{3}He phase separating on top of the ^{4}He film [18]. There is a concomitant decrease in the temperature of the heat capacity peak. The separation between this peak temperature and T_c remains approximately the same. When the transition temperature is depressed below 0.6 K the heat capacity peak, as in the case of pure ^{4}He, is no longer observable. We have seen a similar behavior of ^{4}He films diluted by ^{3}He on HD. This tracking of the peak position with the *decrease* in T_c (Fig. 5) supports the idea that the heat capacity peak is related to the superfluidity of the film.

In an attempt to explain the two third sound modes in the superfluid phase found by Chen *et al.* [19] on H_2 and D_2 substrates, recent theories propose the existence of hexatic and vortex lattice phases for planar superfluid films on a weak substrate in addition to the standard bound vortex-antivortex superfluid phase [20]. It is tempting to associate the heat capacity anomalies we have observed with such an additional phase transition in the superfluid film. However, Fig. 2 shows no signature whatsoever of ρ_s or dissipation of the film in the vicinity of the heat capacity peak. The heat capacity peaks found seem not to be related to the extra resonances seen in Ref. [19], and they cannot be accounted for by the theories [20] mentioned above. The origin of the heat capacity peaks remains to be explained.

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