Superfluidity of Atomically Layered ^{4}He Films

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The superfluidity of ^{4}He films adsorbed on the atomically flat surface of graphite, preplated with HD to tune the surface binding potential, has been studied using a torsional oscillator. The superfluidity of a single uniform fluid layer of ^{4}He shows an intrinsic coverage dependent suppression, while the fluid bilayer is fully superfluid at $T = 0$. The contribution of nonvortex excitations in the film to the normal density shows a strong dependence on coverage, arising from the atomic layering of the film. [S0031-9007(98)06517-X]

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A thin ^{4}He film on a planar substrate is a paradigm two dimensional Bose system. It is predicted to undergo a superfluid phase transition due to the unbinding of vortex-antivortex pairs [1], with a discontinuous jump in the superfluid density obeying a universal scaling relation with the transition temperature [2]; $\rho_s(T_c)/T_c = 2m^2k_B/\pi\hbar^2$. This was first verified by torsional oscillator studies of films adsorbed on a Mylar sheet [3].

Since such an atomically rough substrate provides a heterogeneous binding potential for the adsorbed ^{4}He atoms, there is a threshold coverage, referred to as the "inert" layer, before superfluidity is observed. The simplest picture is that an amorphous solid ^{4}He coating of the surface is required in order to screen the disordered substrate potential, before subsequent ^{4}He atoms are delocalized and can undergo a superfluid transition [4].

More recently there has been renewed interest in films adsorbed on the atomically flat surface of graphite, which provides an essentially uniform binding potential, resulting in a ^{4}He film that, by contrast, displays distinct atomic layering. Evidence for such layering comes from vapor pressure isotherms [5], heat capacity [6], and third sound measurements [5], as well as first principles calculations of the film structure [7]. This layered structure influences the development of superfluidity in the film, as first shown by Crowell and Reppy [8].

This Letter discusses (i) the superfluidity of a fluid monolayer, (ii) the properties of a superfluid bilayer, and (iii) the dependence of the nonvortex excitations in the film on its structure. We have made a systematic investigation of the effect of tuning the substrate potential by preplating the graphite surface with hydrogen deuteride (HD) on the superfluid response. The number of solid ^{4}He layers that can form is reduced to one for a bilayer [5] or trilayer preplating, and zero for the thick preplating film we have investigated. By contrast, two ^{4}He layers will solidify on bare graphite. In these systems we are able to study the superfluid transition for a single fluid layer, which for the thick preplating corresponds to "submonolayer superfluidity." For all three preplatings we find 2D condensation for fluid coverages <3.5 nm^{-2} and clear evidence at higher densities for a coverage dependent suppression of superfluidity in the uniform fluid layer. This latter effect is quite distinct from the "inert layer" found on heterogeneous substrates [3,5] and appears to be an intrinsic property of the fluid. A fluid bilayer participates fully in superfluidity; in this case the inert layer is simply the integral number of solid layers and the transition is consistent with Kosterlitz-Thouless universality. We also find that the layering of the film strongly influences the nature of the nonvortex excitations, and, hence, the superfluid transition temperature.

We have used the torsional oscillator technique, which is best suited both to the investigation of very thin films and for measurements near the superfluid transition temperature, where the attenuation of third sound is high. The oscillator operates at 1056 Hz and its motion is driven and detected capacitatively. Further experimental details and a preliminary account of some of the results are given elsewhere [9].

The bilayer and trilayer HD preplating films are defined by vapor pressure isotherms at 12 and 10 K. Two HD layers correspond to 46.05 STP cm^3 and three layers to 66.22 STP cm^3 for our substrate, while point B of a ^{4}He vapor pressure isotherm at 4.2 K corresponds to 27.4 STP cm^3. Since these data scale very well with neutron scattering measurements of the densities of solid helium and hydrogen films [10], we are confident that the chosen preplating coverages are close to exactly two and three layers, providing a well characterized surface for adsorption of ^{4}He. The thick preplating film was grown by first depositing a bilayer of neon, followed by five layers of HD. Although an isotherm at 10 K showed evidence for some intermixing of the neon and HD [11], we expect the resulting film to be of reasonable quality.

For the bilayer and trilayer preplating, the first ^{4}He layer solidifies and superfluidity is first detected in the subsequent layer, which we will henceforth refer to as the first fluid layer. A typical set of observed superfluid signatures is shown in Fig. 1. The period shift arises from the superfluid film decoupling from the torsional oscillator and reducing its effective moment of inertia and is a

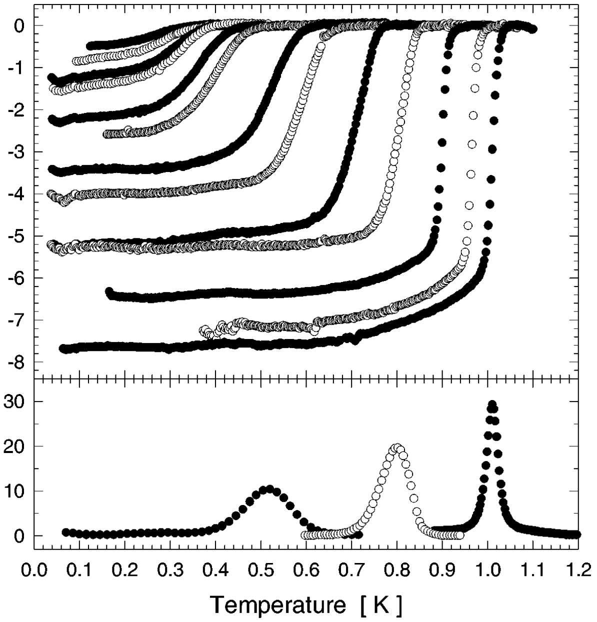

FIG. 1. Period shift (trilayer preplating) for ^{4}He coverages: 7.02, 7.50, 8.15, 8.47, 9.11, 9.35, 9.61, 9.74, 10.16, 10.60, 11.71, 12.58, and 13.11 nm^{-2}. Dissipation peak at superfluid transition at coverages 9.61, 10.60, and 13.11 nm^{-2} is also shown.

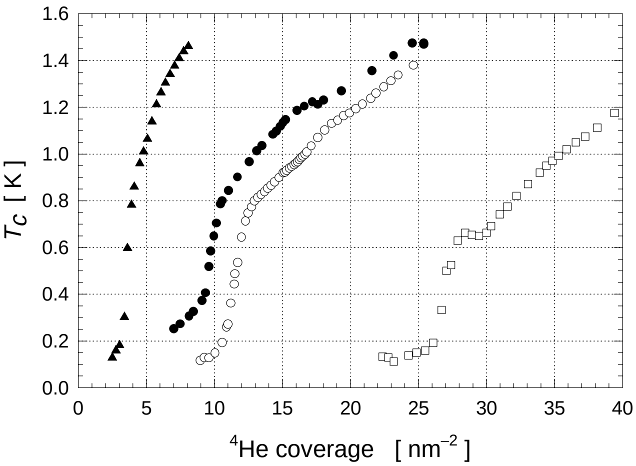

FIG. 2. Superfluid transition temperature vs ^{4}He coverage for (○) bilayer, (●) trilayer, (▲) thick preplating, and (□) data of [8].

measure of the superfluid density in the film. The period shift is corrected for film desorption, using an *in situ* pressure gauge. The dissipation peak is characteristic of a vortex unbinding transition on a planar substrate. In order to investigate the systematics of the evolution of superfluidity with coverage, we define the critical temperature of the transition T_c as the temperature of the dissipation maximum, Fig. 2, and for each coverage determine $\Delta P(0)$, the total period shift in the limit $T \rightarrow 0$, Fig. 3. $\Delta P(0)$ determines the total superfluid mass [12]. The large shifts in the coverage dependence of T_c apparent in Fig. 2 arise from the difference in the number of solid layers for different preplating conditions. It is clear that, with the thick HD preplating film, the first ^{4}He layer does not solidify, as predicted for ^{4}He on the surface of bulk hydrogen [13].

For the bilayer and trilayer preplatings, between second and third layer promotion (regime I), the ^{4}He film consists of a fluid layer atop a solid first layer. The fluid coverage is estimated by subtracting the coverage at second layer promotion from the total coverage. At lower fluid coverages, $n \lesssim 3.5$ nm^{-2}, the fluid layer appears to be condensed into 2D liquid puddles, as predicted theoretically [13–15]. This conclusion follows from the following observations: (i) There is a clear break in the coverage dependence of both T_c and $\Delta P(0)$ which is attributable to the coverage, indicated by the vertical arrow in Fig. 3, at which the line of superfluid transitions emerges from a liquid-gas coexistence region. The possibility of such behavior has been suggested previously [8,16,17]. (ii) At the lowest four coverages below the break in Fig. 1, the temperature at which a period shift is first discernible is coverage independent. This behavior is consistent with a superfluid transition occurring in patches of constant density. Heat capacity measurements also suggest a two-phase coexistence region [18].

Thus above the break we believe the fluid layer to be uniform. It is striking that both T_c and $\Delta P(0)$ increase rapidly with coverage up to promotion. Indeed the rate of increase of T_c with coverage exceeds the slope of the KT line by a factor of more than 2 [19]. If the observed line of period shifts (in Fig. 3 between the arrow and third layer promotion) is extrapolated to zero, this determines the fluid density, n_0, at which the onset of superfluidity would be expected for a uniform fluid layer. The results for the submonolayer superfluid on the "thick" preplating film are very similar and for all three preplatings we find the critical fluid density to be $n_0 \sim 3$ nm^{-2}. This common behavior is also apparent when we plot T_c vs fluid coverage for all three preplatings (Fig. 4). This

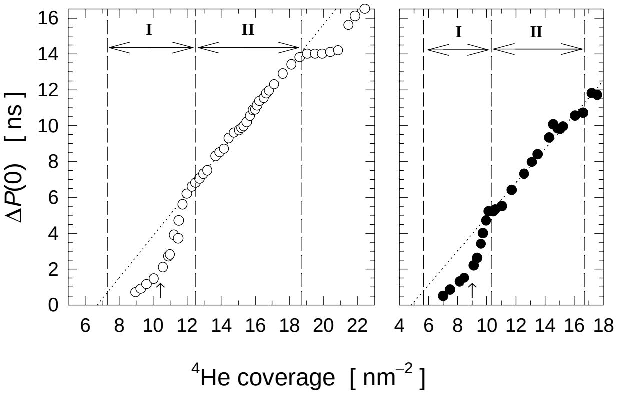

FIG. 3. Total period shift vs coverage for bilayer (○) and trilayer (●) preplatings. Vertical dashed lines show layer promotions, as determined by compressibility minima obtained from vapor pressure isotherms [9].

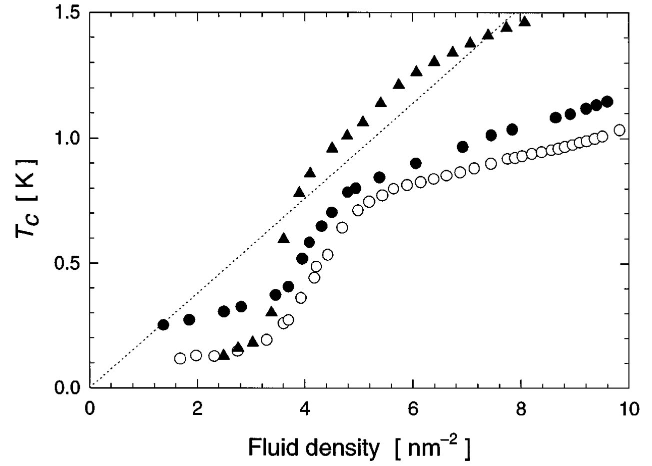

FIG. 4. Superfluid transition temperature vs fluid coverage for (○) bilayer, (●) trilayer, and (▲) thick preplating to show common fluid inert layer of density ~3 nm^{-2}. The dotted line shows the KT line [19].

agreement for different substrate potentials suggests that the complete suppression of superfluidity below n_0 (not directly observable because of the intervention of 2D liquid-gas condensation) is not due to residual substrate heterogeneity, but is an intrinsic property of the single fluid layer [20].

This conclusion is reinforced by the behavior observed for a helium film comprising two fluid layers, corresponding to coverages between third and fourth layer promotion (regime II) for bilayer and trilayer preplating. It is clear from Figs. 2 and 3 that there is a sharp break in the coverage dependence of both T_c and $\Delta P(0)$, which occurs quite precisely at third layer promotion, where a second fluid layer forms. After this break the period shift data are linear with coverage and extrapolate to zero at a coverage close to that of second layer promotion, clearly showing that both fluid layers are superfluid in the low temperature limit [21].

This behavior is quite distinct from that on strongly heterogeneous substrates, where the inert layer is independent of coverage. Here we find evidence for suppression of the superfluidity of a single uniform fluid film, with a "nontrivial" fluid inert layer common to all three preplatings studied, while for the fluid bilayer the inert layer is simply the first solid layer [22]. This novel suppression of superfluidity in a single layer could arise from the periodic substrate potential to which the ^{4}He layer is exposed. On the other hand, a new instability of a uniform fluid monolayer has recently been found theoretically [23], in which vortex-antivortex bound pairs are spontaneously created at densities <3.7 nm^{-2}. Above this coverage the vortex mass is predicted to decrease rapidly with coverage. The present experimental results may be evidence of this phenomenon.

One predicted feature of the evolving structure of such layered films is a sequence of "layering transitions" [14] in which each newly formed layer is initially self-condensed before evolving into a layer of uniform density. Although self-condensation clearly occurs in the first liquid layer, there is no obvious confirmation for this phenomenon in subsequent layers in the present data. The single plateau we observe with bilayer preplating just above fourth layer promotion we ascribe to a reconstruction of the first solid layer.

However, it is clear from the temperature dependence of the period shift below the transition region that the dispersion of nonvortex excitations in the film is strongly influenced by the layering of the film. Period shift data, scaled by T_c, are shown in Fig. 5 for coverages above that of the completed first fluid layer for the bilayer preplating. Two features are apparent: (i) The period shift at T_c is consistent with the predicted universal jump in superfluid density. (ii) The temperature dependence of the period shift (superfluid density) below T_c becomes more marked with increasing coverage, due to the appearance of nonvortex excitations.

The strong coverage dependence of ρ_n/ρ is illustrated in Fig. 5 (inset). At 12.74 nm^{-2} the second fluid layer has just begun to form, while at 17.62 nm^{-2} there are two uniform fluid layers. At 17.62 nm^{-2} the normal density increases with temperature in a manner consistent with the T^3 dependence expected from excitations with a linear dispersion relation, as found in recent first principles calculations [24]. Identifying this mode with third sound we infer a velocity of 52 ms^{-1} [25], approximately a factor of 2 larger than that reported from direct measurements at this ^{4}He coverage on graphite plated with a bilayer of hydrogen [5], possibly attributable to the substrate of that third sound resonator having an

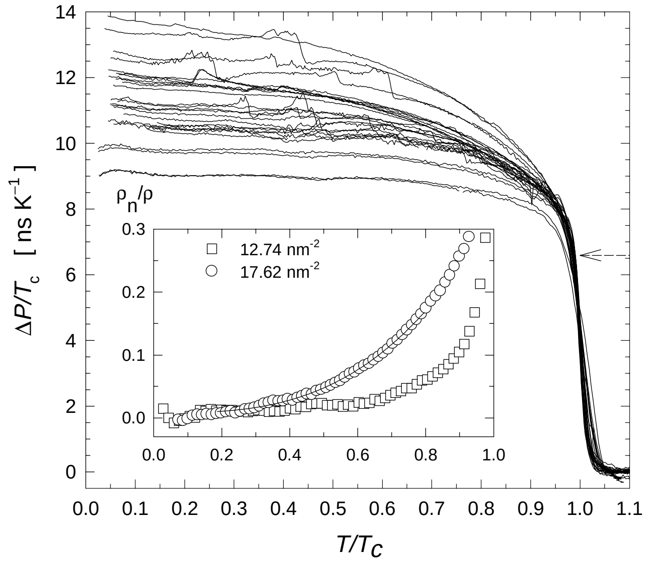

FIG. 5. Scaled period shift for ^{4}He coverages above 12 nm^{-2} (bilayer preplating). Arrow shows expected jump [2], calculated from measured χ factor [12]. Noise is attributable to third sound resonances. Inset: Inferred normal fraction. The solid line shows fit to T^3.

index of refraction of order 2. We find such a T^3 dependence only in the uniform film region. At coverages for which layering transitions are expected the normal density depends more strongly on temperature.

We now turn to the evolution of the superfluid transition temperature with coverage. As we have seen, there is a sharp drop in the rate of increase of T_c observed on formation of a second fluid layer, for both bilayer and trilayer preplatings. A more rounded feature is also apparent in earlier data on the heterogeneous substrate Mylar [26]. This behavior can be understood as a consequence of the increase in normal density arising from the appearance of layered nonvortex excitations [24], which, together with the required universal value of the superfluid density at T_c, largely determine T_c [27]. For the thick preplating film there is a regime, not seen under other preplating conditions, where T_c is proportional to coverage close to the KT slope. This probably arises from the suppression of nonvortex excitations by the expected higher binding energy of the superfluid ^{4}He layer to the substrate.

These experiments have demonstrated novel effects in the superfluidity of a fluid monolayer of ^{4}He adsorbed on an atomically flat surface and in fluid bilayers, which appear robust to changes in the surface binding potential that has been tuned using preplating techniques. Thick films of hydrogen on graphite appear to provide a convenient means of realizing a well characterized weak binding substrate. We find that the superfluidity in a uniform fluid monolayer is suppressed. The origin of this effect and the nature of the vortex excitations in these highly layered films remain open questions. This system also allows detailed investigation of influence of atomic layering on the nonvortex excitations in helium films.

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