Reentrant Superfluidity in ^{4}He Films Adsorbed on Graphite

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We report on torsional oscillator studies of superfluidity in the second and higher layers of ^{4}He films adsorbed on basal plane graphite. Our measurements reveal several novel features. We observe a reentrant superfluid phase in the second layer of adsorbed helium. Superfluidity disappears before the completion of the second layer and reappears early in the third layer. A well-defined plateau in the transition temperature occurs at the beginning of the fourth layer.

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The study of helium adsorbed on basal plane graphite has been a rich field for two-dimensional (2D) physics. Studies of the heat capacity of the ^{4}He-graphite system have been particularly rewarding [1–3]. The structure of helium adsorbed on graphite is dominated by strong layering effects, with a complex sequence of fluid and crystallographic phases revealed in the first and second layers of adsorbed helium. These structural aspects may also be expected to play a role in the superfluid phenomenology of the first few adsorbed layers. Since experiment has established that both the first and second adsorbed layers are 2D close-packed solids at layer completion [1,3], the observation of superfluidity in either the second or third layers would provide an opportunity for the study of an almost ideal 2D superfluid system: atoms in the superfluid state moving in a single layer above a well-ordered substrate. Heretofore, third sound studies have been the most fruitful for the investigation of the superfluid properties of the ^{4}He-graphite system [4]. Unfortunately, this technique has not been feasible for coverages below about three and a half layers because the third sound signal is attenuated.

In this Letter, we report the first observation of superfluidity in the second and third layers of absorbed ^{4}He on graphite. We have used a torsional oscillator for these measurements, which extend over a range of coverage from 1.5 to 7 atomic layers and temperatures from 20 mK to 1.5 K. The second layer superfluid phase first appears at approximately two-thirds filling of the layer and disappears before layer completion. We next observe superfluidity just above second layer completion. After two-thirds of the third layer has been filled, the magnitudes of the superfluid signal and the transition temperature T_c increase rapidly until the layer is complete. Following third layer completion, plateaus in both T_c and the superfluid signal occur for low coverages in the fourth layer. A modulation of the superfluid signal with coverage is observed through the completion of the seventh layer.

In our experiment, the graphite substrate was provided by Grafoil disks, 12.2 mm in diameter [5]. A helium vapor-pressure isotherm obtained at 0.9 K showed layer-by-layer growth through six layers, indicating a high quality surface. The sample surface area of 12.8 ± 0.3 m^2 was determined from this isotherm using the coverage scale of Zimmerli, Mistura, and Chan [4]. The resonant period and the amplitude of the oscillator were measured using a phase-locked loop operated at a constant drive level. Each coverage was admitted to the cell and annealed at a temperature which varied from 4 K for the lowest coverages down to 0.9 K for coverages above three layers. Each coverage was cooled to low temperature, typically 20 mK, over the course of several hours. The resonant period and amplitude were then measured as a function of temperature over the range of interest.

The resonant period data are analyzed by comparison to the empty-cell background. In the absence of superfluidity, the oscillator period is shifted from the empty cell value by a temperature-independent amount proportional to the mass of the adsorbed helium. When superfluidity is present, a temperature-dependent period shift, $\Delta P(T)$, proportional to the superfluid mass, is observed. The period data for films with transition temperatures above 0.8 K require a correction for the mass lost through desorption. Under constant drive conditions, the amplitude of the torsional oscillator is proportional to the quality factor Q of the system and varies inversely as the dissipation.

In Fig. 1 we have plotted the magnitude of our lowest temperature signals $\Delta P(0)$ as a function of coverage. We cannot determine the zero-temperature signal size for the second layer superfluid phase, so we have taken $\Delta P(0)$ as the value of $\Delta P(T)$ measured at our lowest temperature, typically 20 mK. In Figs. 2(a) and 2(b), the period shift $\Delta P(T)$ and the dissipation Q^{-1} are shown as functions of temperature for a series of representative coverages. The features near 300 and 500 mK evident in the data for some coverages are not associated with superfluidity; similar features also appear in the empty-cell background. The exact location and size of these features vary randomly from coverage to coverage, making reliable corrections impossible.

A peak in the dissipation, associated with the superfluid, first becomes evident for coverages above 22 atoms/

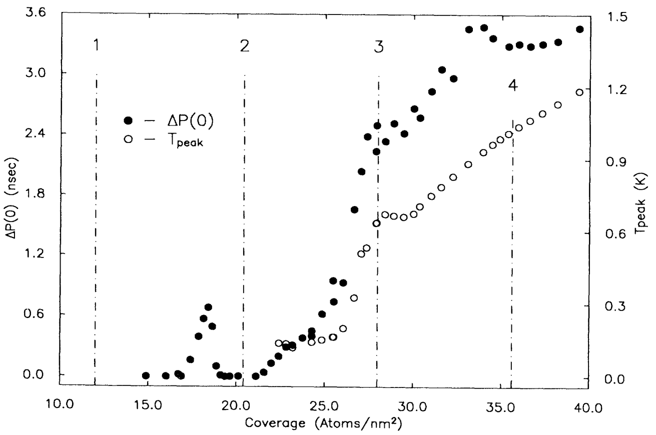

FIG. 1. The low-temperature period shift $\Delta P(0)$ (●) and the temperature $T_{\rm peak}$ (○) of the dissipation maximum associated with the superfluid transition are shown as a function of coverage. The dashed lines indicate layer completion.

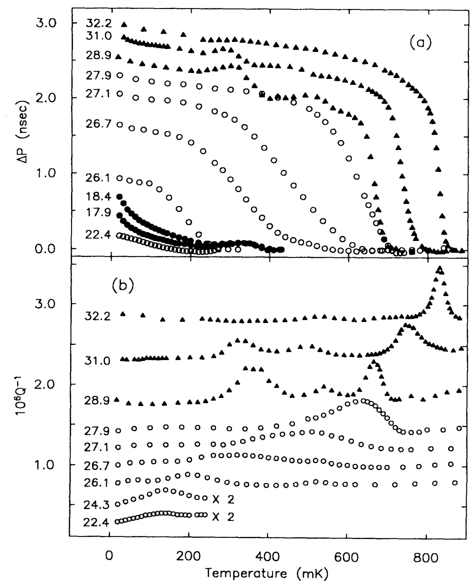

FIG. 2. The period shift data ΔP (a) are shown as a function of temperature for coverages in the second (●), third (○), and fourth (▲) layers. The dissipation data Q^{-1} (b), displaced for clarity, are shown as a function of temperature. The dissipation data for the lowest two coverages have been multiplied by a factor of 2. The coverage (in atoms/nm^2) is indicated at the left for each data set.

nm^2. The temperatures of the dissipation maxima, $T_{\rm peak}$, are indicated in Fig. 1 as open circles. The superfluid transition becomes much sharper for coverages above three layers, and the period and dissipation data agree qualitatively with the form expected for the Kosterlitz-Thouless (KT) transition in a uniform two-dimensional superfluid [6]. Further discussion of the dissipation data will be given elsewhere.

We now turn to a detailed discussion of the data shown in Figs. 1 and 2. The most exceptional feature of these data is the presence of a reentrant superfluid phase in the second layer. This phase corresponds to the peak in Fig. 1 between 17 and 19 atoms/nm^2. The peak, which is slightly asymmetric in character, has a maximum at 18.4 ± 0.3 atoms/nm^2. This is very close to the coverage of 18.7 atoms/nm^2 at which a solid melting peak first appears in the data of Greywall [3]. The small discrepancy is within the limits established by the uncertainty in our surface area and the slight difference between our coverage scale and that used by Greywall. Growth of the solid phase with increasing coverage provides a natural explanation for the suppression of the superfluid signal to the right of the peak. The superfluid signal disappears at approximately 19.1 atoms/nm^2. This value is consistent with the data of Greywall [3], which indicate that full solidification occurs at 19.7 atoms/nm^2. We expect the superfluid signal to disappear before full solidification is achieved since the growing solid destroys the connectivity of the superfluid phase.

The temperature dependence of the superfluid signal for the reentrant phase is remarkable in several respects. First, the $\Delta P(T)$ data, shown in Fig. 2 as closed circles, do not approach an asymptotic value as the temperature is decreased toward zero. We find that the superfluid period shift varies linearly with the logarithm of the temperature above 20 mK. This dependence is emphasized by plotting the data for several coverages on a logarithmic scale in Fig. 3. This figure also demonstrates the difficulty in assigning a well-defined transition temperature for these films. The temperature at which the superfluid signal vanishes does not appear to be simply proportional to the maximum signal at low temperature.

We see no evidence of superfluidity between 19.1 atoms/nm^2 and the completion of the second layer at 20.4 atoms/nm^2. Superfluidity reappears just above second

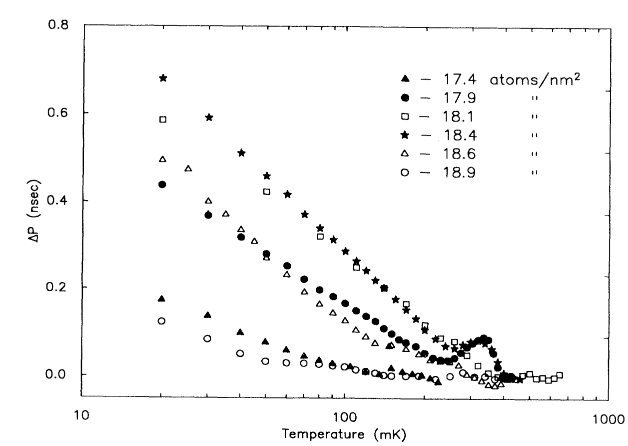

FIG. 3. The period shift data for the second layer superfluid are shown on a logarithmic temperature scale.

layer completion. For coverages between 21 and 26 atoms/nm^2, $\Delta P(0)$ increases linearly with increasing coverage, growing to 0.9 nsec at 26 atoms/nm^2. We observe a dissipation peak associated with the superfluid for coverages above 22 atoms/nm^2. At first this peak is rather broad (see Fig. 2), and the temperature $T_{\rm peak}$ of the dissipation maximum remains fixed at approximately 150 mK until a coverage of 26 atoms/nm^2 is reached. Above this coverage both $\Delta P(0)$ and $T_{\rm peak}$ increase rapidly in magnitude and the dissipation peak narrows. By third layer completion, at 28 atoms/nm^2, both the period shift and the dissipation have evolved into the Kosterlitz-Thouless form [6].

At the beginning of the fourth layer a remarkable feature appears. For coverages between 28 and 30 atoms/nm^2, a plateau in both $T_{\rm peak}$ and $\Delta P(0)$ can be seen in Fig. 1. In the case of the period shift data the plateau is somewhat obscured by noise. A much clearer picture of the dependence of the superfluid signal on coverage can be obtained from an isothermal measurement. In Fig. 4, we show the development of the superfluid signal as the coverage was increased at a fixed temperature of 0.5 K. These data actually show a *decrease* in ΔP between 28 and 30 atoms/nm^2. Plateaus near layer completion can be seen in Fig. 4 up to the completion of the sixth layer. In this figure, the plateaus, which are broader than the feature observed above third layer completion, mirror the modulation seen in the velocity of third sound as a function of coverage [4].

The phenomena reported here stand in sharp contrast to the evolution of superfluidity on heterogeneous substrates such as Mylar, where the superfluid density and transition temperature increase smoothly and monotonically with increasing coverage [7]. The long-range order of the graphite substrate stabilizes a variety of phases that are not observed for ^{4}He films adsorbed on disordered substrates. These phases, which have been studied

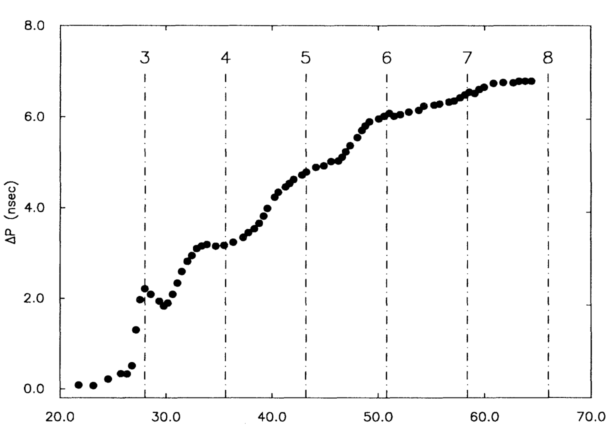

FIG. 4. The period shift ΔP is shown as a function of coverage. This measurement was made by adding ^{4}He to the cell while the temperature was held constant at 0.5 K.

extensively through heat capacity measurements [1–3], could have a significant impact on superfluidity. We will focus here on the second and third layers. The heat capacity data indicate that at low density each of these layers consists of coexisting gas and liquid phases for temperatures below 0.7 K. Greywall suggests a density for the liquid phase of 4 atoms/nm^2 [3]. When the total density is less than this value the liquid is condensed in patches. As the coverage is increased the liquid patches expand and coalesce until the entire layer is a uniform fluid. The second layer solidifies at a density of approximately 7 atoms/nm^2, well before layer completion [3]. The heat capacity data for the third layer are less definitive; however, as discussed below, our data indicate that the third layer is fluid at completion.

Dash has discussed the onset of superfluidity in a ^{4}He film for coverages inside the liquid-gas coexistence region [8]. He argues that the superfluid transition temperature is fixed by the areal density of the liquid phase, but that superfluidity cannot be observed in a flow experiment until the liquid patches percolate to form a continuous network across the sample. This should occur at a coverage that corresponds to approximately 50% of the liquid density. Recently, Greywall and Busch have proposed that the heat capacity peak traditionally associated with the coexistence curve in each of the first three layers is instead a signature of superfluidity [3]. If this hypothesis is correct, then the percolation model proposed by Dash should apply in the low coverage region of the second and third layers.

We find it unlikely that this percolation model can explain the superfluid behavior we observe in the second layer. The onset of superfluidity occurs at 17 atoms/nm^2, which corresponds to a second layer density of 5 atoms/nm^2. This is well above the coverage range where the second layer liquid-gas coexistence region is observed [2,3]. Nonetheless, the lack of a well-defined transition temperature in this regime suggests that the connectivity of the system does play a role in the onset of superfluidity. We would expect a more direct correlation between the size of the superfluid signal and the transition temperature if the observed behavior were due only to changes in the superfluid density. The anomalous temperature dependence shown in Fig. 3 may be due to the temperature-dependent conductivity of weak links in the superfluid. We cannot identify the origin of these weak links, although possibilities include the mosaic structure of the Grafoil as well as macroscopic defects created during the exfoliation process. The conductivity of the weak links should saturate at a sufficiently low temperature. Measurements at lower temperatures will be required to test this hypothesis.

The onset of superfluidity in the third layer occurs just above the completion of the second layer at 20.4 atoms/nm^2. We first resolve a dissipation peak at the superfluid transition for coverages above 22 atoms/nm^2. As shown in Fig. 2(b), this peak evolves continuously into the

Kosterlitz-Thouless peak observed at higher coverages [6]. The plateau in $T_{\rm peak}$ seen in Fig. 1 for coverages between 22 and 26 atoms/nm^2 is reminiscent of the fixed transition temperature expected if the percolation model applies. The broadening of the transition is extreme, however, so that the identification of $T_{\rm peak}$ with the superfluid transition temperature may not be justified. The interpretation of the period shift data poses a greater difficulty since a superfluid signal is observed for coverages just above layer completion, where we do not expect to have a percolated network of liquid patches. An understanding of this observation will require a detailed knowledge of the structure of the film in the proposed coexistence region. The distribution of the liquid phase may be very nonuniform, allowing for the percolation of patches over a macroscopic part of the sample. This may occur even if the total area covered by the liquid is very small.

The superfluid transition becomes increasingly sharp between 26 atoms/nm^2 and the completion of the third layer at 28 atoms/nm^2. At layer completion, the dissipation maximum occurs at 0.67 K and the transition width is only 50 mK. Here $T_{\rm peak}$ is a reasonable approximation to the Kosterlitz-Thouless transition temperature $T_{\rm KT}$. Using the universal relation of Nelson and Kosterlitz [9] for the ratio of superfluid density ρ_s to temperature as the transition is approached from below, $\rho_s(T_c^-)/T_c = 8\pi k_B \times (m_4/h)^2$, we estimate that the superfluid coverage at third layer completion is at least 3.5 atoms/nm^2. Allowing for the reduction of the superfluid density by excitations, we believe that the entire third layer is superfluid at completion.

The plateau in $T_{\rm peak}$ between 28 and 30 atoms/nm^2 poses a challenge since a detailed heat-capacity study of the fourth layer has not yet been undertaken. One interpretation of the feature is that it corresponds to a liquid-gas coexistence region similar to that suggested for the second and third layers [3]. As with the lower layers, we cannot reconcile these data in detail with the percolation model, which predicts that the superfluid signal should increase in this regime while the transition temperature remains fixed. This is not consistent with the period shift data shown in Fig. 4.

This Letter has established some of the basic phenomenology of thin film superfluidity on ordered substrates. While these results indicate that there can be multiple superfluid phases in such a system, several unanswered questions remain. Identifying the origin of the anomalous temperature dependence in the second layer reentrant phase will require measurements at lower temperatures than those used in the present experiments. A more detailed understanding of the phase diagram for the third and fourth layers would be useful in interpreting the data reported here. Finally, the status of the first one and a half layers remains uncertain. We have begun a survey of this region for evidence of superfluidity.

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