Anomaly in the Heat Capacity of Fluid Monolayers of ^{3}He at Low Millikelvin Temperatures

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(Received 5 March 1990)

Two-dimensional Fermi-fluid heat-capacity results are presented for the first and second layers of ^{3}He adsorbed on graphite. The data exhibit anomalous behavior extending up to 50 mK and a sharp feature which is located at 3.2 mK independent of the areal density.

PACS numbers: 67.70.+n, 65.20.+w, 67.50.−b

The first monolayer of ^{3}He adsorbed on graphite, at low areal densities, is generally believed to constitute a two-dimensional (2D) Fermi fluid.[1] This is also true for the second layer formed at much higher total coverages, when the underlying compressed ^{3}He layer exists as an incommensurate solid. The identification of each of these low-density layers as a fluid is based, in part, on thermodynamic measurements: Above a few kelvin the heat capacity[2] is temperature independent with a magnitude close to the classical value of k_B per atom, while below 100 mK the Fermi system is degenerate, and the measured heat capacity[3] is nearly proportional to the temperature.

In this Letter we report heat-capacity measurements for the fluid phases of both the first and second layers of ^{3}He on graphite which extend down in temperature to 2 mK and which show anomalous behavior. Although the heat capacity depends linearly on the temperature between roughly 3 and 50 mK, it does not extrapolate to zero heat capacity at zero temperature, but instead to a small positive value. Moreover, there is a sharp break in the trend of the data at 3.2 mK. Below this temperature the heat capacity decreases more rapidly with decreasing temperature.

In a recent paper[3] we noted the excess heat capacity for the second-layer fluid above roughly 4 mK and speculated then that the anomaly was due to a classical contribution to the heat capacity from some unknown excitation which was activated when atoms were promoted into the second layer. We find now that the anomaly should not be associated with layer promotion and should be attributed to the 2D fluid itself.

The heat-capacity measurements were made on pure ^{3}He samples (7.5 ppm ^{4}He) in the temperature range between 2 and 200 mK using the standard heat-pulse technique and the apparatus described in Ref. 3. The only modification to the calorimeter was the replacement of the working thermometer with a faster thermal response device. The cerium magnesium nitrate susceptibility thermometer was calibrated against a melting-curve thermometer which was in turn calibrated using the temperature scale of Ref. 4. A smooth susceptibility-versus-temperature relationship was obtained with no unusual features in the vicinity of 3 mK. Checks against possible shifts in calibration were made for each run by comparison with a germanium thermometer above 100 mK. All of the data were obtained with the nuclear-demagnetization-refrigerator magnet swept to zero field.

The submonolayer samples were annealed at 12 K for several hours and then allowed to cool at a rate of roughly 1 K/h. Because of the much higher vapor pressure, the second-layer samples were warmed only to 2 K for a few hours. The coverage scale used in this work is based on a total graphite substrate surface area of 198 m^2. This value is 2.5% smaller than the area we used previously for the same substrate. The adjustment was made in order to place $\sqrt{3}$ registry for the first ^{3}He layer at the expected coverage of 0.064 atom/Å^2.

Figure 1 shows first- and second-layer fluid heat capacities at several different areal densities for temperatures between 5 and 50 mK. The data plotted for the second-layer fluid are actually total-sample heat-capacity values; however, the contribution from the compressed incommensurate solid first layer is negligibly small. The second-layer areal densities are estimates obtained by subtracting from the total areal density ρ the first-layer density at layer promotion, namely, 0.112 atom/Å^2. Each of the sets of data exhibits a linear temperature dependence, which extrapolates at zero temperature to a small positive intercept β. These intercepts show a weak coverage dependence and are slightly larger for the second layer.

The heat capacity of an ideal degenerate 2D fluid is independent of the density and is given by $\pi k_B^2 m_3 A T / 3\hbar^2$, where A is the total surface area. In the presence of ^{3}He-^{3}He interactions Landau theory predicts that this expression should be altered only by the replacement of the bare mass with a density-dependent effective mass. There is no accompanying shift in the heat capacity or the temperature which would be required to describe the experimental results. If the heat-capacity offsets are ignored, the slopes of the data imply similar effective masses for both layers. At densities near 0.01 atom/Å^2, m_3^*/m_3 for each layer is close to unity,[5] as would be expected in the low-density limit. Although the heat-capacity offsets are relatively most significant for the lowest-temperature data, it should be noted that the

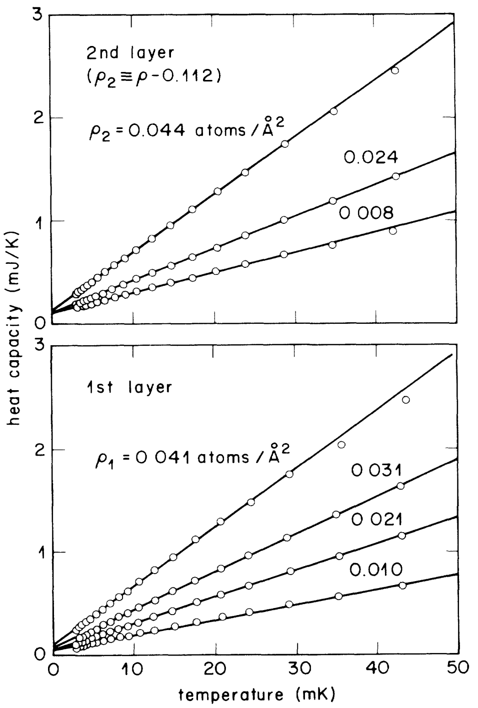

FIG. 1. Low-temperature heat capacities for the first and second fluid layers of ^{3}He adsorbed on graphite. Note that the straight lines drawn do not pass through the origin.

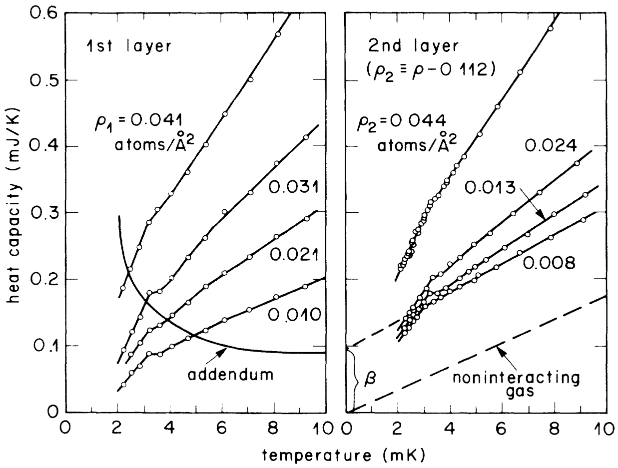

FIG. 2. First- and second-layer fluid heat capacities in the vicinity of a sharp feature at 3.2 mK. Also indicated are the addendum and the heat capacity of an ideal degenerate 2D Fermi gas.

anomalous behavior extends out to temperatures of the order of tens of mK.

The data obtained below 10 mK are shown on expanded scales in Fig. 2. Each of the curves, for both the first and second layers, shows a well defined kink corresponding to an abrupt change in temperature dependence. The kinks are located at $T_k = 3.2$ mK, independent of the sample density. Although the feature can be described as a cusp and the smooth curves in Fig. 2 have been drawn with this assumption, a small discontinuity at T_k is also consistent with the data. Moreover, there is a precursor to the feature starting 1 or 2 mK above T_k where the heat capacity begins to smoothly deviate below the extrapolation of the higher-temperature data.

Between 2 mK and T_k the data are not accurate enough to determine a particular temperature dependence. The experimental difficulty is due to the rapidly increasing addendum contribution to the total heat capacity near 2 mK (see Fig. 2). The magnetic contribution in the background heat capacity is from the paramagnetic salt in the susceptibility thermometer. Taken at face value, the lower-density first-layer data suggest a limiting low-temperature power-law dependence for the heat capacity which is greater than linear. However, the larger, and therefore the more reliable, heat-capacity data for both the first and second layers indicate that the heat capacity may be proportional to temperature as $T \to 0$.

Although the background heat capacity becomes relatively significant at the lowest temperatures, it cannot be responsible for the anomalous behavior attributed to the fluid. In support of this statement we note the following: (1) The addendum heat capacity has a smooth temperature dependence and shows no special features near 3 mK. (2) The β values are density dependent and so cannot be eliminated by a single adjustment of the addendum. (3) All of the fluid samples exhibit an excess heat capacity which appears to persist up to temperatures of the order of tens of mK, where the addendum is only a small fraction of the total heat capacity. (4) The measured heat capacity of the compressed incommensurate solid first layer is, as expected, small compared to the magnitude of β.

Item 4 also rules out significant excess contributions to the heat capacity which are independent of the physical state of the film. Such contributions might have originated from, for example, pockets of bulk ^{3}He in capillary-condensed regions on the substrate, from ^{3}He atoms adsorbed on the edge plane surfaces of the graphite crystallites, or from atomic excitations perpendicular to the substrate. This last possibility is also ruled out by the fact that the calculated first excited state normal to the basal plane is more than 50 K above the ground state.[6]

It is interesting that viewing the anomaly as a constant excess heat capacity above 3 mK leads to the number of classical entities contributing k_B to the heat capacity being comparable to the expected number of homogeneous regions (100 Å diam) on the entire substrate. This line of reasoning could perhaps lead to an explanation for the density independence of T_k. On the other hand, the number of independent homogeneous regions should decrease considerably with the buildup of several layers and yet the excess appears to scale with the number of fluid layers. Moreover, the homogeneous regions must have a large distribution of sizes which contradicts with the sharpness of the feature at T_k. This also rules out other types of finite-size effects.

The statement that the excess heat capacity scales with the number of fluid layers is based on Fig. 3 which shows data[3] taken at a total coverage of 0.37 atom/Å^2. At this coverage the ^{3}He film, which consists of two solid and four fluid layers, exhibits an offset heat capacity roughly 4 times larger than the monolayer β. The β value was extracted from these multilayer data by fitting the results between 3 and 20 mK using the function $a/T^2+\beta+\gamma T$. The first term accounts for the exchange contribution to the heat capacity from the solid layers. The T^2 phonon contribution from these solid layers is negligible.

It also seems unlikely that the small number of ^{4}He impurities (7.5 ppm) could be, in some way, responsible for the anomalous behavior. Even if each of the ^{4}He atoms contributed k_B to the total heat capacity, this would amount to only 0.1% of β. Furthermore, because the ^{4}He atoms are bound more strongly to the substrate than the ^{3}He atoms, the second-layer fluid should be purified, yet it exhibits an anomaly at least as big as that of the first layer.

In recent heat-capacity measurements[7] made on dilute ^{3}He-^{4}He films adsorbed on a plastic substrate, anomalous behavior was observed which in several ways is similar to our findings. The ^{3}He in these films is believed to form a monolayer which floats on the surface of the thicker superfluid ^{4}He film. For ^{3}He coverages of less than 0.01 atom/Å^2 and for ^{4}He films roughly 10 Å thick, the temperature dependence of the heat capacity abruptly changes at $T_{k'}$ and becomes proportional to T at lower temperatures. $T_{k'}$ varies little with ^{3}He density. There is also evidence for a small discontinuity in the heat capacity at this temperature. On the other hand, in these mixture measurements $T_{k'}$ is of the order of 100 mK, and so this anomaly occurs when the ^{3}He layer is nondegenerate. Although this feature has been interpreted as indicating a transition from a dilute to a dense phase, there are unexplained aspects of the data which leave open other interpretations. Certainly it is possible that the anomaly in the mixture films is related to the lower-temperature anomaly we find for the pure ^{3}He system.

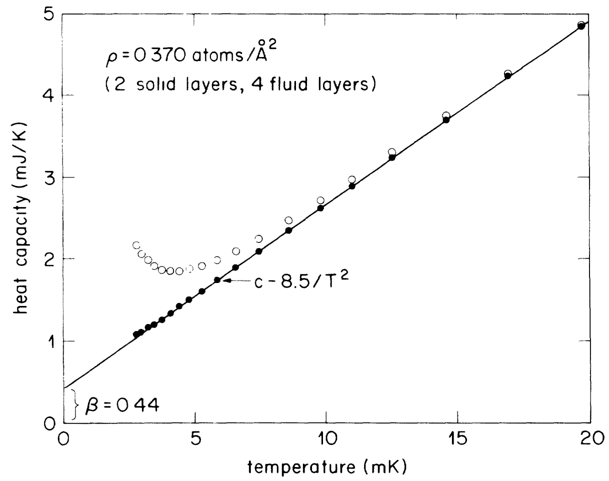

FIG. 3. Heat capacity of a sample consisting of two solid and four fluid layers. The linear extrapolation of the higher-temperature data yields an intercept β which is about 4 times the monolayer value.

Assuming now that our anomaly is reflecting an intrinsic property of the 2D Fermi fluid phase of ^{3}He adsorbed on graphite, can the anomaly correspond to a gas-liquid phase transition with a very small latent heat? The possibility of a gas-liquid coexistence below T_k and a pure gas phase above T_k can be immediately dismissed for two reasons. First, the heat capacity would then be expected to be proportional to temperature above T_k; and second, a two-phase region at low temperature would be identified by low-temperature isotherms which are linear functions of the density. This is not what is observed. Moreover, theoretical calculations[8] indicate that 2D ^{3}He is not self-bound and should therefore not liquify.

Another possibility is for some type of pure phase below T_k and a two-phase coexistence at higher temperature. The temperature dependence of the heat capacity above T_k is generally consistent with this proposal; however, the density dependence of the high-temperature isotherms is again not linear in ρ. It should also be noted that moving into a two-phase region on warming should result in a positive discontinuous jump in the heat capacity, due to the latent heat and the conversion of phases. The data suggest the possibility of a small *negative* step.

The lack of evidence for two-phase coexistence both above and below T_k suggests a second-order transition or a first-order transition which does not involve a change in density. The latter possibility would also imply, however, that the high-temperature phase does not possess a heat capacity which is proportional to T. The absence of the heat capacity being proportional to T for a second-order transition might be explained by fluctuating effects which extend well above T_k.

It seems natural to consider the possibility of a Kosterlitz-Thouless[9] superfluid transition. Even though this theory predicts only an unobservable essential singularity for the heat capacity at T_c, somewhat above the transition there should be a maximum due to the entropy liberated by an increasing number of unbound vortices.[10] This maximum might be occurring at T_k. Because this contribution to the heat capacity is nonuniversal, the density independence of T_k would not necessarily imply density independence for T_c. On the other hand, T_k/T_F

is small, implying that fluctuation effects should be negligible and that the transition should be mean-field-like.

Another possibility is some type of magnetic transition in the 2D liquid layer.[11,12] For bulk ^{3}He, quasiparticle interactions enhance the $T=0$ magnetic susceptibility over the value for the noninteracting system by an order of magnitude, meaning that the system behaves as a nearly magnetic Fermi liquid. In two dimensions the interaction effects are even stronger,[3] as evidenced by an effective mass which is a factor of 2 larger at the same nearest-neighbor distance, and this may be sufficient to drive an actual magnetic transition.

Certainly, there are other possible explanations for the heat-capacity results which are also intriguing.[13] But to distinguish between the various proposals it will be necessary to have results from other types of measurements, in particular NMR measurements extending below 1 mK. The present evidence indicates that the anomalous behavior observed for the fluid phase of adsorbed ^{3}He is not spurious and actually represents an intrinsic property of the 2D Fermi fluid on graphite.

We gratefully acknowledge the advice of O. E. Vilches concerning the annealing of the lowest-density samples and the helpful comments of F. M. Gasparini.

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