Observation of Two-Dimensional Phase Separation in ^{3}He-^{4}He Films

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New measurements of heat capacity for ^{3}He adsorbed on ^{4}He films are presented. It is found that for the thinnest ^{4}He film used, and starting at a ^{3}He coverage of 0.058 layer, 3.7×10^{-4} Å^{-2}, the data show a very sudden onset of degeneracy. It is found that these data are consistent with phase separation in the two-dimensional layer of ^{3}He at the surface of the ^{4}He film.

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When ^{3}He is added to liquid ^{4}He, the lowest state for the ^{3}He is at the liquid-vapor interface. This state results as a compromise between the helium-helium van der Waals attraction and the zero-point energy.[1] One can achieve in this way a rather ideal two-dimensional Fermi system, the properties of which can be studied in its Boltzmann as well as its degenerate regime. The density can be varied from the smallest detectable level up to about a monolayer, 0.0645 Å^{-2}. Beyond this coverage any additional ^{3}He becomes dissolved in the ^{4}He "substrate."

The ^{3}He at the surface of bulk helium behaves very much like a noninteracting ideal gas of Fermi quasiparticles with an effective mass $m = 1.45m_3$, and a binding energy[1] $\epsilon = 2.22$ K relative to a state within bulk ^{4}He. For ^{3}He in a film of ^{4}He the situation is rather different. Measurements of heat capacity[2] yield a binding energy of promotion from the surface to the first excited state which is lower than for the bulk system. The effective mass tends to be somewhat higher, $\sim 1.7m_3$ at the lowest coverages. More importantly, however, in the case of films one finds an attractive ^{3}He-^{3}He quasiparticle interaction, which depends on the thickness of the ^{4}He. The ability to affect this interaction via the ^{4}He thickness makes the ^{3}He-^{4}He film system very interesting both theoretically and experimentally.

One may regard, especially in the case of thin films, the surface ^{3}He and the ^{4}He as a two-dimensional (2D) mixture. There have been a number of calculations of the phase diagram of such 2D mixtures.[3-7] Common features of these calculations are the presence of a phase-separated region, and a superfluid transition extending from pure ^{4}He to phase separation. The work of Mon and Saam[7] differs somewhat from the others in that they treat a "layered" 2D system which is much closer to the situation realized for ^{3}He in ^{4}He films. The layering refers to the fact that for thin enough ^{4}He films the first excited state of the ^{3}He, the second layer, is also 2D in character, but is localized mostly within the body of the film.[2] Because of a layer-layer coupling this system is richer in possibilities than a strictly 2D mixture. Mon and Saam[7] find for instance that, for certain values of binding energies, one can have two regions of phase separation, and two superfluid transition lines.

On the experimental side there has been no evidence of 2D phase separation.[8] This includes measurements of specific heat for conventional physisorbed ^{3}He-^{4}He mixtures,[9] specific-heat measurements of ^{3}He in films of ^{4}He thicker than 18.8 Å,[2] measurements of surface sound and surface tension for the case of ^{3}He on the surface of bulk ^{4}He,[10,11] and third sound in films.[12,13] We present in this Letter new measurements of specific heat which suggest that we have observed 2D phase separation. These measurements were made with the same calorimeter as in Ref. 2 with modifications to improve thermometry, to lower the background heat capacity, and to facilitate the condensing of small amounts of ^{3}He. A basic feature of the calorimeter is the presence of Nuclepore filters which provide a surface area of 10^6 cm^2, mostly in cylindrical pores of 2000 Å diam. The properties of these filters, and the formation of superfluid films on them, have been studied previously.[14]

The heat capacity of the ^{3}He is obtained by subtracting from the measurement the contribution of the calorimeter. We have established that the ^{4}He film contributes negligibly in the region of interest as long as the thickness exceeds ~ 10 Å above a solid, 3-Å first layer. In Fig. 1 we show the data for 0.03 layer of ^{3}He (1.93×10^{-4} Å^{-2} and Fermi temperature of 0.074 K) on 12.2-Å ^{4}He film. The horizontal line through the data represents the expected 2D Boltzmann value, $N_3 k_B$. This involves no adjustable parameters, just the

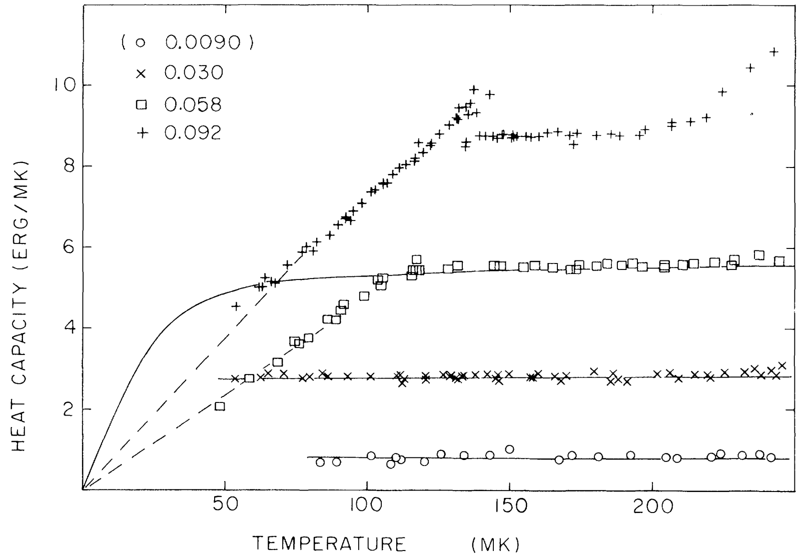

FIG. 1. The heat capacity of ^{3}He on a 12.2-Å ^{4}He film. The coverages are given as fractions of a layer. The lowest coverages are consistent with a homogeneous spread of the ^{3}He. The higher coverages have a sudden onset of degeneracy consistent with 2D phase separation. The solid lines are the expected behavior for a strictly 2D Fermi system. The dashed lines are drawn to guide the eye.

known amount of condensed atoms. The agreement of the data with this expected value is excellent. The slight rise in these data which is visible near 250 mK is due to promotion of the ^{3}He into its first excited state. It is from this aspect of the data, which we have measured up to ~ 800 mK, that the "spectroscopy" of available states can be obtained. We will not discuss these results at present. Further evidence that we are able to measure a very homogeneous and dilute amount of ^{3}He is given by the open circles in Fig. 1. These are data for which we do not know the exact amount of ^{3}He condensed, but, from the constant Boltzmann value of 0.83 erg/mK, we deduce the coverage to be 0.0090 layer (5.75×10^{-5} Å^{-2}). These data represent a small contribution, ~ 5% at 250 mK, above a background which varies by a factor of 3 over the region shown. These results indicate that we are measuring an intrinsic property of the ^{3}He, not influenced by imperfections in the substrate, or affected by inhomogeneities in the ^{4}He film. This is markedly different from the situation of ^{3}He adsorbed on Grafoil where at low temperature the substrate dominates the behavior of the ^{3}He.[15]

The next coverage shown in Fig. 1 is for 0.058 layer. Something very dramatic happens with these data. Down to 110 mK they follow very closely the solid curve which is the expected behavior for a homogeneous 2D Fermi system with $m \simeq 1.73 m_3$. Below this temperature, however, the data show a sudden onset of degeneracy. In the degenerate region the heat capacity is given by

$$C = (k_B \pi / 3\hbar^2) m A T , \tag{1}$$

where A is the area occupied by the ^{3}He. Since m does not vary substantially at low ^{3}He coverage, the low-temperature slope should be the same for *all homogeneous coverages*, with A being the geometrical area. This is the slope of the low-temperature part of the solid curve. The much smaller slope indicated by the dashed line through the squares suggests that the ^{3}He has separated into about 26% of the total area, thus at a concentration of 0.22 layer. Its Fermi temperature is now 0.42 K; hence the degenerate behavior. The dilute phase, which should be in equilibrium with the concentrated phase, seems to contribute very little to the data. We estimate the dilute phase to be $\lesssim 0.0016$ layer.

The next higher coverage shown in Fig. 1 indicates a very similar behavior. Only now we have a jump in the data near 140 mK before the onset of degenerate behavior. The slope in the low-temperature region is consistent with phase separation into an area which is proportionately larger than in the case of the previous coverage. Hence it represents a dense phase at very nearly the same density. We note as well that data from

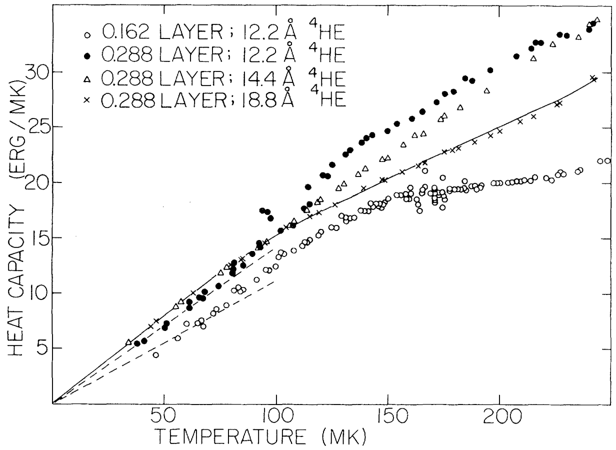

FIG. 2. The heat capacity of ^{3}He at higher coverages. The solid line is a fit of the data for a homogeneous 2D Fermi system; see Ref. 2. The dashed lines are drawn to guide the eye.

several runs near the region of the jump indicate hysteresis, as well as a greater amount of scatter. Although this behavior is definitely associated with the transition, we have observed it even after cooling the calorimeter for several days before taking data. Thus it is not completely understood.

In Fig. 2 we show data at higher coverages, and for two thicker films of ^{4}He. The open circles, 0.162 layer, show an increase in the scatter of the data near 165 mK, and a low-temperature slope which is again less than expected. This indicates again the presence of a denser phase on a subset of the total calorimeter area. There is now in addition an enhancement of the data above the dashed line indicating perhaps a contribution from a dilute phase. The next coverage, 0.288 layer, shows a very similar behavior. There is some structure in the data near 100 mK, and again at low temperatures a slope which is close to but definitely less than that expected for a homogeneous coverage.

In previous work[2] done on ^{4}He films of 18.8 Å and thicker no evidence of 2D phase separation was seen. We thus increased the ^{4}He thickness to check with these earlier data. Upon addition of 2.2 Å of ^{4}He (see Fig. 2) the structure near 100 mK disappears, and the low-temperature slope increases to the expected value, the solid line. Upon adding more ^{4}He, to the thickness of 18.8 Å, the low-temperature data are *unaffected*, supporting the conclusion that the ^{3}He is now homogeneously spread out. The solid line through the data is a fit to the model of a 2D Fermi system discussed in Ref. 2. The fit yields $\epsilon = 1.55 \pm 0.02$ K, and $m = (1.64 \pm 0.04)m_3$. This shows excellent agreement with $\epsilon = 1.57 \pm 0.03$ and $m = (1.65 \pm 0.03)m_3$ as obtained in Ref. 2. We can say then that in a region of the ^{3}He-^{4}He film phase diagram previously unexplored we have seen new behavior, and further, that when we overlap with previous data we obtain excellent agreement.

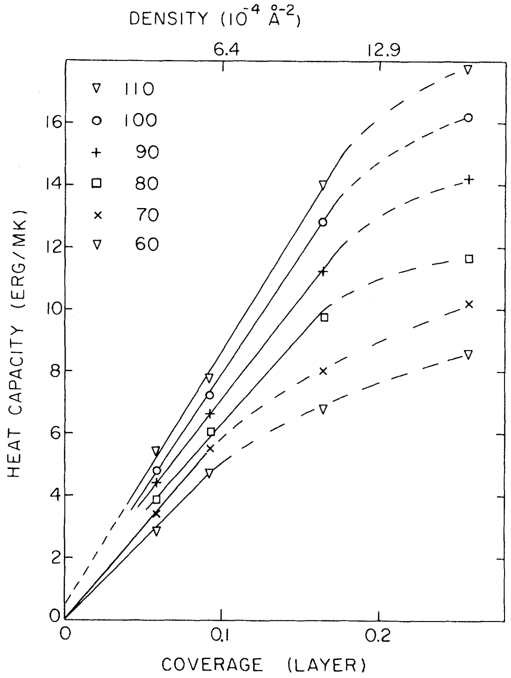

FIG. 3. The heat capacity at fixed temperature, in millikelvins, as a function of total amount of ^{3}He condensed. A phase-separated system at coexistence should yield straight lines. The lines for the lowest temperatures are drawn through the origin and are consistent with a negligible contribution from a dilute phase.

In the case of phase separation we can write the total heat capacity as a sum of a dilute, d, and concentrated, c, phase,

$$C = \sigma_d A_d C_d + \sigma_c A_c C_c , \qquad (2)$$

where the σ's are the number of atoms of ^{3}He per unit area. Since the total area is constant, $A = A_d + A_c$, then

$$C = \sigma_d A C_d + A_c(\sigma_c C_c - \sigma_d C_d). \qquad (3)$$

Thus at a *fixed temperature* the heat capacity depends linearly on the area occupied by the concentrated phase, i.e., the number of atoms of ^{3}He condensed. If, as we suggested previously, σ_d

is very small, then Eq. (3) should yield straight lines through the origin. Figure 3 shows the heat capacity at fixed temperatures as a function of the amount of ^{3}He condensed. We can see that the first three coverages are consistent with this picture down to 80 mK. The lowest coverages are consistent at low temperature with complete phase separation, i.e., the lines go through the origin. The deviation at higher coverages is perhaps not surprising. In the calculation of Mon and Saam it is shown that the phase diagram is determined by the difference in the chemical potential between the surface state of ^{3}He and its excited state. This difference changes as a function of coverage. Thus, at each coverage one is dealing with a slightly different phase diagram. In this manner one violates the condition under which Eq. (3) is strictly valid.

We have presented new data for dilute coverage of ^{3}He on ^{4}He films. We have shown that we are able to measure intrinsic properties of ^{3}He down to very low coverages. For certain coverages we observe a very sudden onset of degeneracy which is consistent with 2D phase separation. This represents the first observation of this phase separation, which had been predicted for 2D mixture and for ^{3}He-^{4}He films.

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