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Heat capacity of multilayer ^{3}He adsorbed on graphite

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Heat-capacity measurements in the multilayer region of adsorbed ^{3}He on Grafoil display a new set of transitions. These effects first appear as small peaks in the film heat capacity at low temperature, for coverages greater than second-layer completion, $n = 0.186$ Å^{-2}. With increasing film coverage, the peaks grow in height until they dominate the film heat capacity for the highest coverages measured, up to $n = 0.275$ Å^{-2}. Eight films in this range of coverage have been studied for temperatures between 0.04 and 1.5 K. Two important observations have been made with regard to the data: the incremental low temperature, $T < 0.5$ K, heat-capacity asymptotes to a value similar to bulk liquid ^{3}He, and the signal near the peak is exponential in form with an activation energy close to the latent heat of bulk liquid ^{3}He at the peak temperature. The peaks are assumed to signal evaporation of bulk liquid ^{3}He. These results suggest a distinct transition from two- to three-dimensional behavior in helium films. Stability of the proposed three-dimensional phase versus that of the flat film is discussed in terms of current nucleation theory. Reevaluation of ^{4}He data for the same coverage range is encouraged by these findings.

I. INTRODUCTION

Until the last decade, investigations of helium films attracted to surfaces by van der Waals interactions have concentrated on the superfluid phase of ^{4}He.[1] Measurements of the properties of these films were designed to contribute to the understanding of the λ transition as well as superfluid film flow. In the case of heat-capacity studies,[2,3] a modified λ transition was evident down to films as thin as 3 or 4 atomic layers. For lower coverages no phase transition was observed. More recently, research on adsorbed ^{4}He has intensified. Numerous experiments[4-7] have observed onset of superfluid properties in the multilayer region of adsorbed ^{4}He with variations apparently due to the substrates and experimental probes involved.

Adsorbed helium on graphite has proven to be a unique system for the study of surface phases. The multilayer region of the ^{4}He-graphite system has been investigated in film-flow experiments by Herb and Dash[8] and in heat-capacity measurements by Bretz.[9] The behavior of heat capacity can be divided into two categories. The second layer appears to behave similarly to the submonolayer, at least qualitatively obeying two-dimensional (2D) models. For thicker films, a modified λ transition is first seen between three and four layers, although the shapes of these peaks are significantly different from other thin-film studies. On increasing the coverage, these transitions appeared to evolve continuously toward the bulk form.

Considering the remarkable properties seen in multilayer ^{4}He, a similar study was undertaken on ^{3}He. We consider the behavior of the ^{3}He film which is adsorbed on top of two complete layers. Eight ^{3}He film coverages from 0.187 to 0.275 Å^{-2} have been investigated for temperatures 0.04 to 1.5 K. At low temperatures the underlayers are in the form of 2D solids and can, at least to first order, be separated from the contribution due to further adsorption. For elevated temperatures, layer promotion can no longer be ignored and the separability of individual layers becomes less reliable. As will be discussed, the results obtained are comparable to other systems of multilayer films on various substrates. They also indicate a more direct path to the evolution of thick-film and bulk behavior as the coverage is increased.

II. EXPERIMENT

This paper is intended to be taken in conjunction with a previous article dealing with the second layer of ^{3}He on Grafoil.[10] Experimental procedures are similar to the second-layer study. A few features of the multilayer measurements are unique and will be discussed here. For further details of the experiment the reader is referred to Refs. 10 and 11.

The procedure for administering a sample was significantly different from the previous work. Since the first two layers of helium lower the adsorption energy substantially, the vapor pressure at 4 K was large enough that condensation in the fill line could be a problem. In order to ensure complete adsorption, the sample chamber, cell *B* of Ref. 11, was slowly cooled to 1.5 K for about 12 h before starting the dilution refrigerator. An experimental run was begun the following day when the cell had reached its minimum temperature.

All data were taken as the cell was slowly drifting to higher temperature.

III. RESULTS

Throughout this discussion, n, the total coverage, will be defined as the ratio of the total number of particles to that needed to form the $\sqrt{3}$ structure in ^{4}He divided by the area per particle in that structure (15.71 Å^{-2}). The coverage of the individual layers will be called n_1, n_2, n_3.... The term density will be reserved for when the evidence indicates that the film is uniform and occupies a given layer.

The total coverage to complete two atomic layers of ^{3}He on Grafoil has been estimated to be 0.186 Å^{-2}. This quantity was determined by locating the minimum in the low-temperature heat capacity as a function of density and attributing that to the completed second layer.[10] As for the first layer ^{3}He, the second layer forms a two-dimensional solid at densities near layer completion. The heat capacities of the two coverages just above second-layer completion are plotted in Fig. 1. These results display second-layer melting peaks at about 1 K. In addition, a new low-temperature transition in the heat capacity is evident. The low-temperature part of the data is emphasized by plotting the logarithm of the total heat capacity versus temperature. It is clear from these data that two distinct peaks in the heat capacity are occurring for each coverage, although for $n = 0.187$ Å^{-2}, the low-temperature maximum is below the accessible range of the cryostat. Multiple transitions in a film system can occur from separate layer behavior suggesting that the new effect is occurring in the adsorbed film on top of two solid layers. Furthermore, the location of these new peaks on a two-dimensional phase diagram would be quite different from either of the two previous layers.

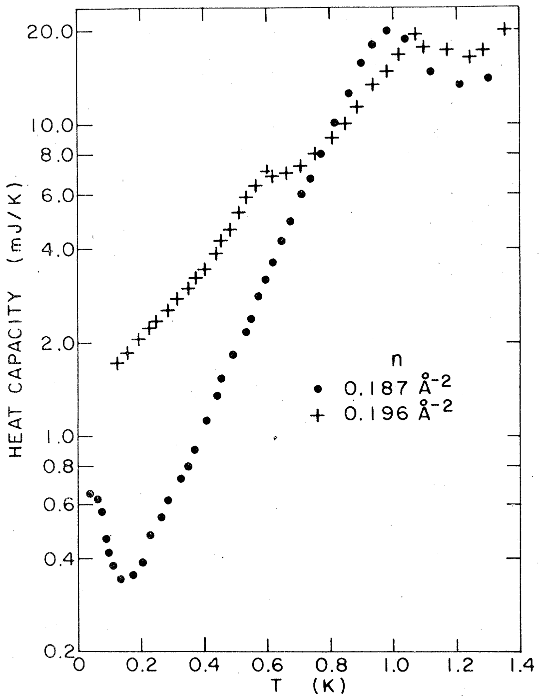

FIG. 1. Heat capacity of ^{3}He films adsorbed on graphite for coverages slightly above second-layer completion.

Upon adding more ^{3}He to a total coverage of 0.203 Å^{-2}, the observed effects change significantly (Fig. 2). The second-layer melting peak, which was narrowing and moving to higher temperature in the previous cases, has nearly vanished. The remnant transition is seen by a broad deviation from the exponential desorption tail at high temperatures. For the low-temperature signal, the new effect has changed substantially from the previous runs with a strong peak now occurring at 0.69 K. Below the peak, the heat capacity is quite large, being of the order of ten times the expected contribution of a 2D solid with melting point 1 K.

The data on the five coverages greater than 0.203 Å^{-2} are plotted in Fig. 3. Only the two extreme cases show the low-temperature signal for the sake of clarity. Several gross features should be noted on these data. The peak height and temperature are both increasing functions of coverage. Also, similar to the submonolayer results, the transition signaled by the peak appears continuous. Above the peaks, the sharp upturn in the data is indicative of desorption. This process probably masks the second-layer melting transi-

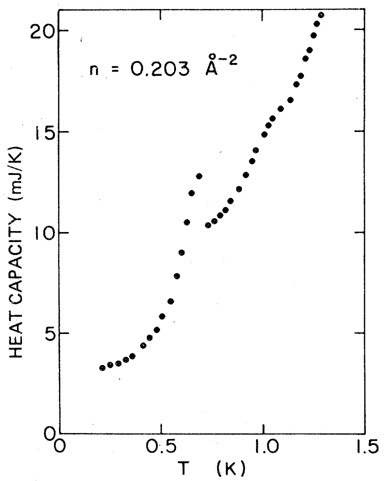

FIG. 2. Heat capacity of ^{3}He film above second-layer completion.

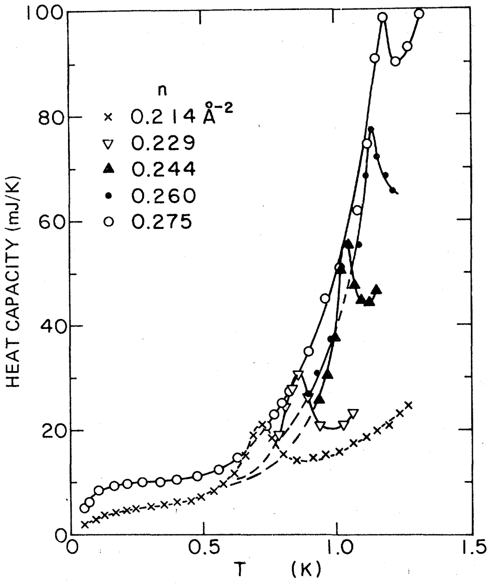

FIG. 3. Heat capacity of five highest multilayer coverages of ^{3}He.

tion which is expected to be very broad by comparison to the previous coverage.

An estimate of the specific heat of the additional film on two complete layers can be made by dividing the low-temperature heat capacity by the third-layer coverage. For this analysis, the coverage to complete two layers was assumed constant at 0.186 Å^{-2}. Results for the five highest coverages are shown on isothermal plots for three temperatures in Fig. 4. These heat capacities per third-layer atom still include the contribution of the second-layer solid for lack of an accurate way of taking it into account.[12]

The feature which should be noted in the data is the apparent convergence to a specific-heat form which is relatively insensitive to film coverage. This behavior is remarkably different from the 2D gas specific heats which show a strong variation with density.[10,11] In addition, the form of this specific heat is not very different from that obtained on various densities of bulk liquid ^{3}He. Shown on the right of the figure are the range of specific heats of bulk liquid ^{3}He as they depend on density.[13] The horizontal scale has been plotted $\rho^{2/3}$ to provide a common scale of comparison. The asymptotic values of each temperature appear to be close to the range of the specific heat of bulk liquid, the best agreement being with that of the high-density limit.

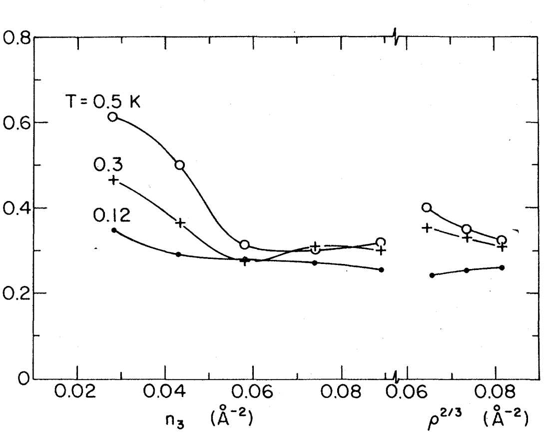

FIG. 4. Heat capacity isotherms for the five highest coverages compared to bulk liquid.

IV. DISCUSSION

The tendency of these low-temperature specific heats to approach that of bulk liquid ^{3}He suggests that in some way the film is developing three-dimensional (3D) liquid character. It is, therefore, reasonable to assume that the transitions in the heat capacity result from the evaporation of the liquid. It should be noted at the outset that the peaks in the heat capacity lack the characteristic first-order shape one would expect from a liquid-gas transition. This feature is most probably an artifact of the system. Two possible explanations are available. As has been suggested for the monolayer films, the peak shape may not appear to be first-order due to substrate effects such as inhomogeneities.[11] Alternatively, the system would not be expected to possess first-order character since there are several coexisting phases present, 3D liquid and 2D and 3D gas.

To test for 3D liquid character, a careful evaluation of the heat capacity near the transition is required. The best approach to this problem is to evaluate the thermodynamic expression for the heat capacity near a phase transition

$$C_N = N_l\, c_l + N_v\, c_v + T\left(\frac{S_v}{N_v} - \frac{S_l}{N_l}\right)\left(\frac{dN_v}{dT}\right)_{\text{eq}}, \qquad (1)$$

where $N = N_l + N_v$, N_l, and N_v are the quantities of ^{3}He atoms in the liquid and vapor phase, and c_l and c_v are the specific heats of the two phases. The last term is the transfer term and incorporates the latent heat of the transition. This expression has been applied to the problem of surface normal desorption by Dash, Peierls, and Stewart.[14] The transfer term is often of activation energy form $\exp(-q/kT)$, where q is the latent heat of the transition. We estimate the heat capacity due to the coexisting phases by assuming that the total excess above two layers, $N_3 = N - N_1 - N_2$, is equal to N_l for all temperatures. The heat capacity of the liquid can be subtracted from the total using the specific heat of bulk liquid ^{3}He. Several approximations were made here. Since specific-heat data are not available for pressurized ^{3}He above 0.6 K, we were forced to further approximate the behavior of the unevaporated film using saturated vapor pressure data.[15] This approximation introduces difficulties in fitting the low-temperature data which were resolved by choosing the effective N_3 necessary to make the magnitude of the heat capacities of the liquid and film agree at 0.4 K. This correction required at most a 20% shift in N_3 and, typically, much less. The assumption that $N_3 = N_l$ for all temperatures introduces additional errors. However, the corrections made by handling this question more carefully are not large since the liquid and vapor specific heats are similar. There are also normal desorption corrections which have been ignored in the analysis. All these simplifications contribute to the error in this calculation, which is sizable for the low densities, but which gets progressively smaller as N_3 increases.

Employing the above method, the excess heat capacity, which should approximate the transfer term, is the difference between the measured value and that due to N_3 atoms of liquid ^{3}He:

$$C_{\mathrm{ex}}(N_3,T) \equiv C_{\mathrm{meas}}(N_3,T) - N_3 c_l(T) . \qquad (2)$$

Excess heat capacities for all five of the highest coverages are plotted on a semilog plot versus inverse temperature in Fig. 5. The straight lines demonstrate the exponential dependence characteristic of a desorption process. Latent heats extracted from this graph are recorded on Table I. Also tabulated is the latent heat of bulk liquid ^{3}He at 1 K,[16] which is very near these transition temperatures. The agreement between the bulk value and the high density limit for the films is remarkable.

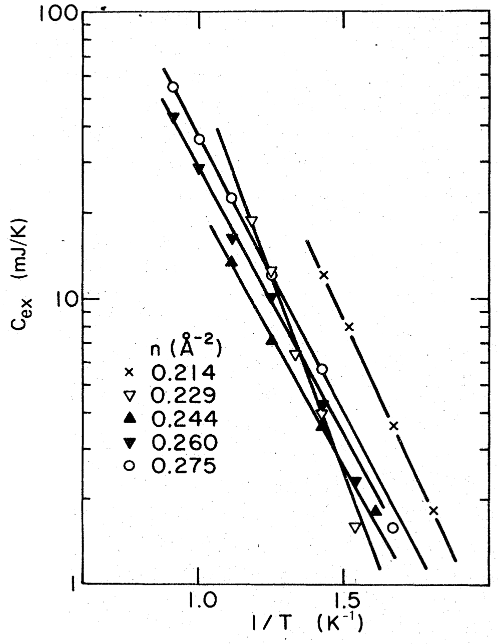

FIG. 5. Excess heat capacity vs inverse temperature. Lines through the data are the best linear fits.

TABLE I. Latent heats of evaporation for various film coverages above second-layer completion compared to bulk liquid at 1 K.

n (Å^{-2})	L (K)
0.214	5.0 ± 1
0.229	6.3 ± 1
0.244	4.2 ± 0.3
0.260	4.3 ± 0.2
0.275	4.4 ± 0.2
Bulk (SVP, 1 K)	4.5 (Ref. 16)

The behavior of the completed second layer during multilayer evaporation is of considerable concern. As we observed in the low-coverage multilayers, i.e., 0.203 Å^{-2}, the effect of increasing the density is to broaden the transition identified as second-layer melting. That observation is in striking contrast with the first-layer data, which noted a strengthening and narrowing of the transition above layer completion.[17,18] The mechanism which produces the broadening of the second-layer transition is not understood, although it may be connected with the weaker binding of the film. It is believed that the behavior of the second layer at low temperatures is basically unchanged by the presence of the additional film, but that the melting may be occurring over a wider range of temperature.

We can investigate the behavior of the second layer in this range of coverage by further consideration of the evaporation signals. Returning to Eq. (1), a point of simplification is at the peak in the heat capacity, where N_l vanishes. The remaining terms must be extensive in N_v. The peak data for all coverages greater than second-layer completion are shown in Fig. 6. All values appear to fall on one of two straight lines. For 0.229 Å^{-2} and below, the peaks form a line with slope $\Delta C/\Delta Nk$ equal to 2. Furthermore, even without the lowest peak at 0.04 K, the best straight-line fit to the peak heat capacities intersects the horizontal axis at 0.186 ± 0.001 Å^{-2}. This value is identical to the density of second-layer completion estimated from the low-temperature signal. For 0.229 Å^{-2} and above, there is a clear break in the slope of the best straight-line fit. The peak heat

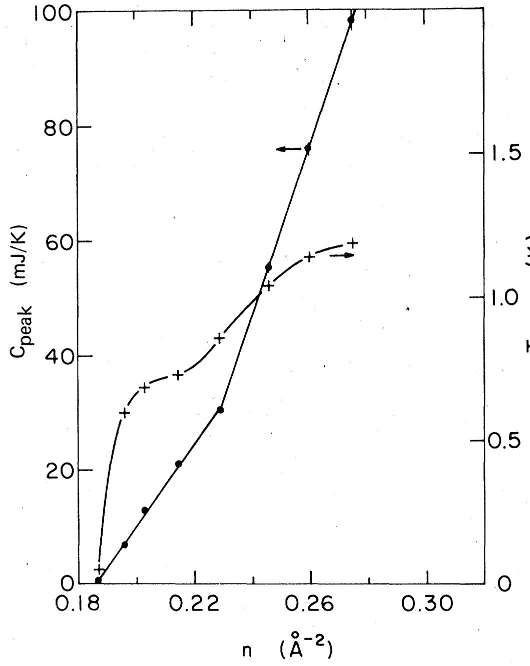

FIG. 6. Peak heat capacity and temperature vs coverage for all runs showing evaporation peaks.

capacities for these higher coverages appear to follow a straight line of approximately twice the slope. Also shown in the figure are the peak temperatures of the new transitions. There is an inflection point in the line drawn through these peak temperatures at nearly the same density.

That both these quantities are transforming as the evaporation peak approaches the second-layer melting temperature is significant. The noted change in slope of both curves indicates a conversion from a region where evaporation of the liquid is completing before the second layer melts to the other extreme, with the second layer melting before the evaporation takes place. Thus, the excess heat capacity and the elevated peak temperatures are a result of the second-layer melting first providing additional liquid for the evaporation process. That this effect occurs over a range of temperatures adds credence to the continuous melting hypothesis for solid monolayers.

A final feature of the data in Fig. 6 is the slope of the peak temperature versus density. Analysis of the four highest coverages indicates that $\Delta T_p/\Delta n$ is a rapidly decreasing function of n with an apparent saturation value ($n \to \infty$) of $T_p = 1.23$ K. This observation will be discussed further in Sec. V along with the theoretical aspects of multilayer adsorption.

V. THEORETICAL CONSIDERATIONS

A variety of experimental evidence presented in Sec. IV was used to defend the hypothesis that these new transitions in the heat capacity result from evaporation of liquid ^{3}He. The basic question related to this observation is how can a film of only three atomic layers behave so similarly to bulk liquid? We begin by presenting a conceptual argument as to how liquid formation could occur. Discussion of the more rigorous theories will follow.

Experiments on monolayer and second-layer films of ^{3}He displayed a number of distinct properties. One of these, regarding the existence of a 2D liquid phase in ^{3}He films, is still an open question. Theoretical analyses by several workers[19,20] have shown that the ^{3}He liquid state is not bound in 2D. These results have basic importance to the distinguishability of 2D and 3D systems. If we adopt the conventional approach to film growth, the multilayer region should develop layer by layer with each layer population being negligible until the previous layer reaches completion. A failure of this model is that it does not account for the difference in phase of each layer. This simplification is quite serious in the case of ^{3}He since the 2D liquid state appears unbound and the 3D liquid must be the limit for very thick films.

These concepts combined with the observation of these new transitions in the film heat capacities suggest an alternative classical view for multilayer film growth. A possible configuration for the film as it is developing the third layer is schematically shown in Fig. 7(a). For temperatures below 1 K,

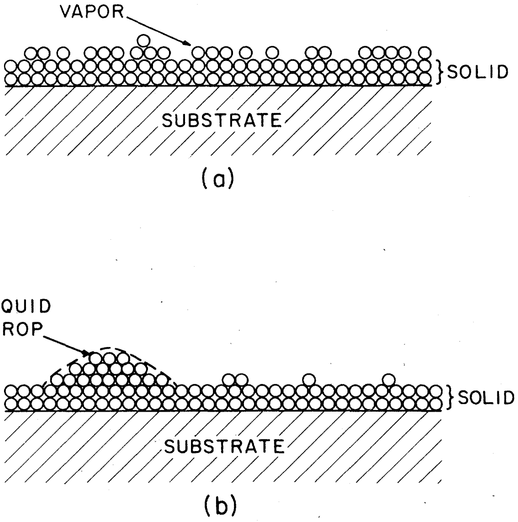

FIG. 7. Two possible configurations for multilayer film on solid substrate.

the first two layers are solids. The third layer is in the 2D vapor phase. If the third layer would solidify at completion, the same development would occur on the fourth layer. Several problems result from this progression. Since the liquid phase is the state of the thick film and the 2D liquid is not bound, the 3D liquid state would have to evolve from the increased coordination number of the underlayers. This mechanism would prevent liquid evaporation from occurring until at least third-layer completion. Perhaps more confusing is that the model would never permit the surface layer to liquefy even in the thick-film limit.

An alternative view of the multilayer film is shown in Fig. 7(b). In this configuration the first two layers are still distinct but the "third layer" prefers to cluster into a 3D liquid drop. The driving force for this tendency is the increased cohesive energy of the 3D cluster. Substrate binding, which tends to keep the film flat, generally decays as the inverse cube of the distance from the substrate. Therefore, at a certain thickness determined by the relative magnitude of the cohesive, substrate, and surface energies, the film would undergo a transition to the clustered state. For progressively thicker films these clusters will grow in size and stability.

Recently, Dash[21] has analyzed this problem by considering film growth with increasing coverage as being controlled by three interfacial tensions and the two-dimensional spreading pressure. Three classes of adsorbate-substrate behavior are defined according to the relative sizes of these quantities. It appears from the analysis that the ^{4}He-graphite system is of "class II," characterized by a uniform adsorption followed by a clustering transition at approximately 2.8 layers. Although the theory is classical and macroscopic, it is believed that the basic elements of the problem have been taken into account.

A possible approach to considering the ^{3}He-graphite system would be to apply the theory directly, predicting critical thickness and equilibrium size and shape of clusters on the substrate. The major difficulty with this procedure is that the various parameters which enter the theory are quite uncertain. It is for this reason that we approach the application of the theory by using the experiment to predict the interfacial tensions of the system.

We believe that the experimental indications for clustering in the ^{3}He-graphite system are quite strong. It appears that the clustered phase occurs after a certain amount of preadsorption. The existence of a clustering transition demands that the interfacial tensions obey the equality at the critical thickness

$$\gamma_{sv}=\gamma_{lv}+\gamma_{sl}\,, \tag{3}$$

where s, l, and v are for solid, liquid, and vapor phases. The interfacial tension between the solid and vapor is affected by adsorption so that

$$\gamma_{sv}=\gamma_{s0}-\phi\,, \tag{4}$$

where 0 refers to the vacuum and ϕ is the two-dimensional spreading pressure. The above condition is limited by the extent to which we can ignore the volume terms in the expression for the thermodynamic potential. Combining Eqs. (3) and (4) we develop an expression for the interfacial tension at clustering onset

$$\gamma_{s0}-\gamma_{sl}=\phi_c+\gamma_{lv}\,, \tag{5}$$

where ϕ_c is the critical spreading pressure.

For the case of ^{3}He-graphite, the right-hand side of Eq. (5) has been determined experimentally. We have observed clustering onset at a total coverage of 0.187 Å^{-2}. Elgin, Goodstein, and Greif[22] have measured the vapor pressure and heat capacity in this range of coverage above 2.5 K. They found ϕ to have a weak temperature dependence which is approximately linear below 4 K. A linear extrapolation to absolute zero of Elgin and Goodstein's results for 0.187 Å^{-2} gives an estimated $\phi_c=16.85$ erg/cm^2. Using the accepted value of 0.16 erg/cm^2 for γ_{lv} of ^{3}He,[23] the right-hand side of Eq. (5) equals 17.01 erg/cm^2. If we further assume the graphite surface tension values of 130 ± 20 erg/cm^2, a value of 113 ± 20 erg/cm^2 is deduced for the critical value of interfacial tension.[24] It should be noted that using the published value for γ_{s0} could be misleading since it was determined by contact angle measurements, where preadsorption may have occurred as it does for He.

Based on the above analysis, we can begin to see why clustering occurs at about two layers, if it occurs at all. We see empirically that actual onset of the clustered phase occurs when the difference $\gamma_{s0}-\gamma_{sl}$ is less than 17 erg/cm^2. However, if we use the data of Elgin *et al.* to estimate the saturated film spreading pressure, we get a value of approximately 20 erg/cm^2. This means that 85% of the spreading pressure is included in the first two layers of film. Consequently, if the interfacial tension difference is not close to critical by second-layer completion, it is unlikely that it could be reached by increasing the coverage further. The above argument is precisely the reason most films that cluster do so in the first few layers where most of the spreading pressure is developed.

It is expected that the clustered phase will be unstable against a rise in temperature since ultimately the vapor is the high-temperature phase. It follows that the maximum possible temperature for which clustering could be present would be the bulk critical temperature. The actual maximum would be somewhat lower, an effect seen in the data where the transition temperatures appear to approach a maximum value of 1.23 K. That the peak temperature is saturating in the thick-film limit is consistent with the observed saturation in the spreading pressure seen by Elgin *et al.*[22] From the standpoint of the clustering transition this temperature represents the maximum for which stable clusters can exist in equilibrium with the vapor on the graphite substrate.

An alternative explanation for the existence of bulk character in these films is that it occurs due to substrate mediated condensation. Cole[25] has argued that a kind of capillary condensation may occur in inhomogeneous regions of the substrate. These condensed regions would dominate the thermal character for films nominally four layers thick. The basis for this interpretation is a theoretical study of condensation in cylindrical pores[26] which showed instabilities between flat films and partially filled pores.

We believe that the two explanations are not in conflict, but rather explain different aspects of the problem. The Dash theory deals with the interaction between bulk liquid, vapor, and substrate. Although the surface tension is somewhat indirect, it contains all the interactions of the system. If one could calculate these interfacial tensions accurately, it would be possible to predict the thermodynamic behavior of the system. Considering the size of the interfacial tensions involved, it is quite clear that the liquid-gas surface tension cannot be the driving mechanism for clustering. This indicates that the theory can be applied to condensation of either positive or negative curvature. In a general sense, the Dash work deals with the question of when bulk liquid can occur on the substrate. This question is significantly different from the Cole arguments which are entirely dependent on the curvature of the condensed liquid. This explanation considers where liquid will form, given its existence. In this way, the two analyses appear supportive.

VI. CONCLUSION

In this paper, experimental evidence is presented which strongly suggests the existence of bulk liquid character in ^{3}He films greater than two atomic layers. A new set of transitions in the heat capacity are quite similar to bulk liquid ^{3}He signals, both in low-temperature specific heat and latent heat of evaporation. These results appear to be the first evidence for bulk liquid formation in helium films as thin as two atomic layers.

We have analyzed a recent model[21] which suggests that the clustering transition in multilayer films is caused by a delicate balance of the interfacial tensions of the system. This places rather stringent requirements on the differences between the surface tension of the bare graphite and the interfacial tension between graphite and liquid ^{3}He.

The ^{3}He-graphite system has proven to be an excellent probe for the multilayer film thermodynamics. A great deal is known about both liquid and gaseous ^{3}He. In addition, we avoid the complicating, although exciting, feature of the superfluid transition in thin films. Provided we can interpret the ^{4}He data in light of this work, it appears that the multilayer helium film region is a very complicated system. The existence of a clustering transition certainly confuses the concept of thin-film superfluidity making the analysis of that system far more difficult than was previously believed.[27]

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