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Experiments on superfluidity in ^{3}He-^{4}He mixture films

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The results of our experiments on superfluidity in thin films of ^{3}He-^{4}He mixtures are reported. The measurements span the temperature range from 1.3 to 2.1 K. All concentrations of ^{3}He were investigated for films ranging up to 100 Å in thickness. We present detailed curves showing the superfluid content of mixture films as a function of the total amount of film present. For the first time, the dependence of these curves upon ^{3}He concentration is exhibited. Characteristic of the superfluid onset process we observed are distinctive distances which have an interpretation as measuring the "healing length." These are presented as functions of temperature and ^{3}He concentration.

I. INTRODUCTION

Extensive data[1-14] have appeared in the literature for a number of years on the superfluidity of ^{4}He in constrained geometries. Many experiments of this kind, performed by a variety of ingenious methods, were motivated by the extensive theoretical literature on healing lengths[15-18] and superfluidity in two-dimensional systems.[19-23] This reflects the general interest in the quantum phenomena made manifest only in those systems in which spatial dimension constitutes an experimental variable.

To date, little information has appeared in the literature which describes the effects of adding ^{3}He to superfluid ^{4}He in constrained geometries. From the experiments[10,24-27] reported it is clear that the onset temperature for superfluidity is dramatically affected by the presence of ^{3}He. However, the data have been limited to a sparse scattering of points and no details of the onset phenomenon as a function of the film's extent and its ^{3}He concentration. In the present study we present a detailed and systematic survey over all concentrations and film thicknesses of the superfluidity manifested within the temperature range explored—1.3 to 2.1 K.

Since our initiation of this work, several theoretical investigations have appeared[28-31] which are directly pertinent to our experiments. These relate to the recent remarkable predictions that there is a quantum zero point of superfluid areal mass density which a two-dimensional system may support. Our experiments confirm the prediction that this precipitous onset persists even in the presence of ^{3}He. In fact, our experimental findings show this effect to persist even beyond the concentrations of ^{3}He believed to limit the effect.[28,29]

Our results, however, go beyond confirming this prediction. We have gathered data on several other aspects of superfluidity in ^{3}He-^{4}He mixture thin films which we present here. In particular, we have measured the superfluid areal mass density as a function of film "thickness" thus giving details of the inception of superfluidity. These measurements have been performed over temperatures ranging from 1.3 to 2.1 K and over a selection of ^{3}He concentrations spanning the range which exhibits thin-film superfluidity (~ 30 at.% ^{3}He). The data reveal some significant characteristic lengths. These are presented as functions of temperature and of liquid film surface ^{3}He concentration. Hence we report here the first measurements pertaining to the healing length in ^{3}He-^{4}He mixtures as a function of concentration.

II. NATURE OF THE EXPERIMENTS

The essentials of the experiment are as follows (cf. Ref. 32). A mixture of ^{3}He and ^{4}He gas is admitted into an experimental chamber. The molar concentration c of ^{3}He in the gas mixture is an experimental variable as are the pressure P of this gas in the chamber and the temperature T at which the chamber is held and at which measurements are made. By the nature of the system an adsorbed film is formed on all surfaces within the chamber. In particular, a helium mixture film is formed on the surface of our quartz-crystal adsorptometer. This instrument directly measures the adsorbed film mass per unit area σ_m which loads the crystal. That portion of the film which is superfluid does not couple to the transverse motion of the adsorptometer surface. The superfluid is detected as a crystal loading deficit. If we denote the σ the total mass per unit area in the adsorbed film then the superfluid mass per unit area σ_s is deduced from

$$\sigma_s = \sigma - \sigma_m \quad . \tag{1}$$

The mass per unit area loading the crystal σ_m is a directly measured quantity. The deviation in the frequency of resonance Δf of our quartz-crystal adsorptometer is proportional to σ_m. A detailed discussion of the operation of this instrument and of the experimental configuration employed with it is given in Ref. 32. We note here that, at the unloaded operating resonance frequency of $f_0 = 24$ MHz, the relationship between the measured frequency change Δf and the mass loading σ_m is given simply by

$$\sigma_m/\Delta f = -0.385 \text{ ng/cm}^2\,\text{Hz} \ . \tag{2}$$

The total mass adsorbed per unit area σ is not generally a quantity directly measured by our instrumentation. However, we are able to deduce σ from our data by virtue of our extensive study of films in the nonsuperfluid regime. This derives from the following: Our measurements on films exhibiting no superfluidity ($\sigma_s = 0$) yield $\sigma_m = \sigma(T,P,c)$. The dependence upon each of the three measured variables T, P, and c is quite strong. However, by reorganizing our data we have been able to demonstrate the existence of a set of universal curves of adsorption.[32] By a "universal curve" we mean one for which, because of the proper choice of new variables (functions of T, P, and c), the explicit dependence of $\sigma = \sigma(T,P,c)$ on many variables is suppressed in favor of an implicit dependence contained in fewer new variables. In our case we reduced the three variables T, P, and c, to two. These are $T\ln[P/P_d(c,T)]$ and $\beta = \beta(c,T)$. In the first variable the quantity $P_d(c,T)$ is the dew (condensation) pressure for a gas mixture at concentration c and temperature T. This is a tabulated quantity found in the literature.[33] Our own measurements complemented the tabulated ones where published values were lacking. The second variable β represents the ^{3}He concentration of bulk liquid which is in thermodynamic equilibrium with a gas of concentration c at temperature T. In a ^{3}He-^{4}He phase diagram of pressure versus concentration, β is simply determined by

$$P_b(\beta,T) = P_d(c,T) \ , \tag{3}$$

where P_b represents the boiling (vaporization) pressure of a liquid mixture at concentration β and temperature T. Values of $\beta = \beta(c,T)$ are available in the literature.[33] Both β and c are molar concentrations in this exposition.

The motivation for employing these variables and their significance is discussed at length in Refs. 30 and 32. Here we merely note that $T\ln(P/P_d)$ measures the van der Waals's energy binding the adsorbed atoms to the substrate. And β represents the liquid ^{3}He concentration in the film. More precisely, β represents the film liquid ^{3}He concentration at its interface with the gas. The liquid concentration β differs markedly from c, the gas concentration prepared for an experimental run.

Using the new variables outlined above greatly condenses and clarifies our experimental data. Figure 1 presents some of our nonsuperfluid regime data plotted so as to exhibit the universality. The abscissa represents a thickness estimate constructed by dividing our measured values of adsorbed areal mass density by $\rho = \rho(\beta,T)$. The bulk density ρ represents the mass per unit volume for a liquid mixture at temperature T and ^{3}He concentration β.[34] Hence σ/ρ, although not the actual thickness of the film, represents a close estimate of it (cf. Ref. 32).

The central and crucial element of the foregoing is that the data reorganization described allows us to determine σ from the measurables T, P, and c in those regimes where the presence of superfluid precludes a direct measurement of σ. This becomes clear in Fig. 2. Each element of the universal curve contains points from a wide range of pressures and temperatures for the given concentration β. Hence the entire universal curve is completely defined by data points taken only in the nonsuperfluid regime. Thus the curve is available even for those special and limited combinations of T, P, and β for which superfluidity exists and for which our adsorptometer

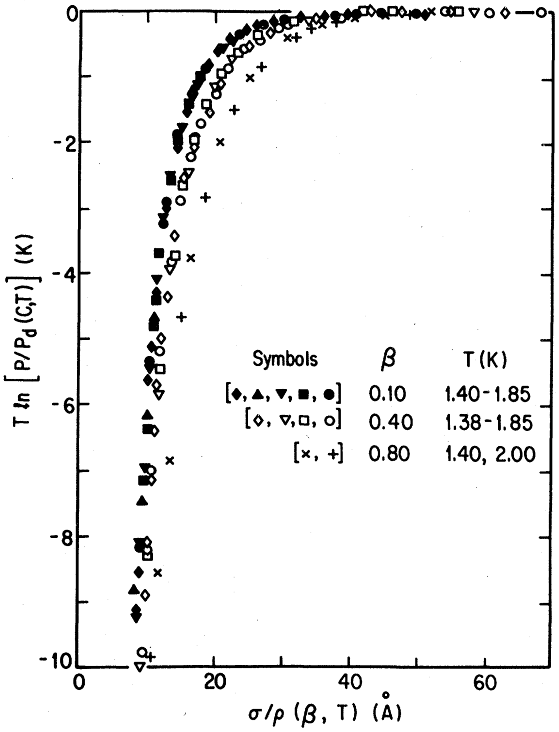

FIG. 1. Three of the adsorption universal curves displayed for the values of β listed. Data on adsorbed mass, measured at all pressures, temperatures, and gas concentrations subject only to the constraint that β be constant, fall on a single curve.

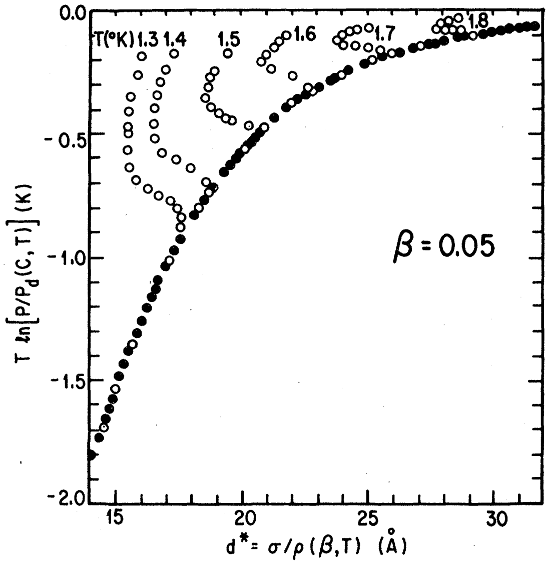

FIG. 2. Adsorbed mass measurements (abscissa) in the presence of superfluidity. The deviation from the universal curve (dark circles) represents the loading mass deficit attributable to superfluidity at each of the temperatures listed.

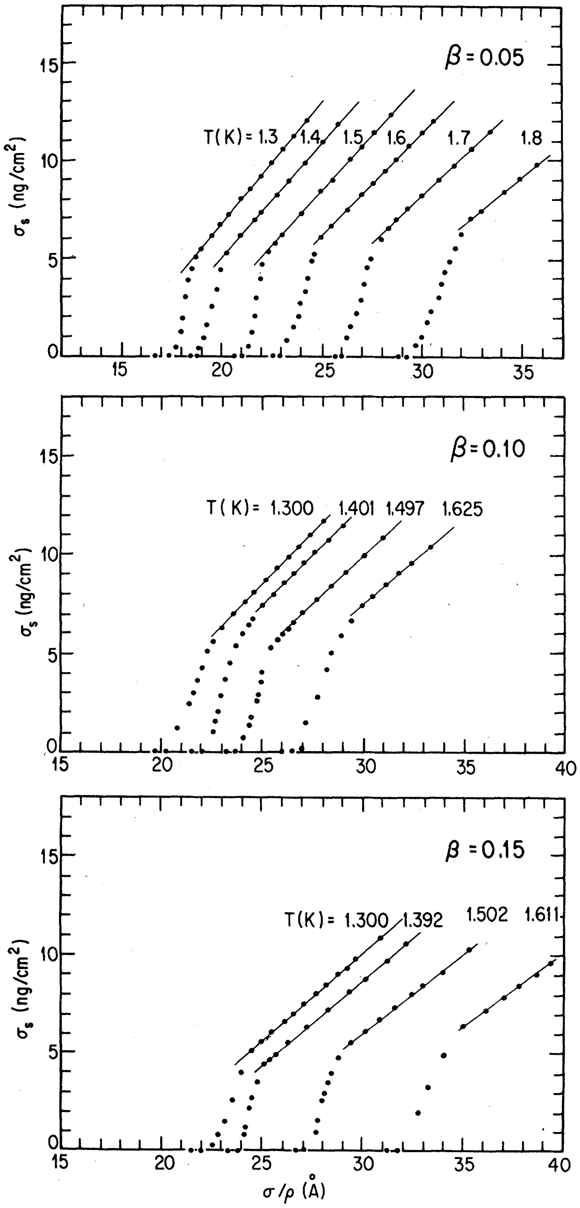

FIG. 3. The superfluid content as a function of the total amount of film present. The black circles are our experimental data points. The straight lines drawn through them have slope $\rho_s(\beta,T)$; that of the superfluid density characterizing a bulk mixture of ^{3}He concentration β at temperature T.

records σ_m instead of σ. Figure 2 shows the data when superfluidity is present together with the universal curve characterizing the particular β explored. The abscissa represents σ/ρ or σ_m/ρ depending upon which curve is being read. The difference between the two—the horizontal separation in the figure—represents σ_s/ρ by virtue of Eq. (1).

III. EXPERIMENTAL RESULTS

The abscissa differences of Fig. 2 may be plotted against the universal curve abscissas themselves for any of the curves characterizing a particular β. Although the universal curve is not a function of temperature, the loading mass σ_m, in the presence of superfluidity does depend upon temperature. Hence the plots mentioned present us with the content of superfluid as a function of the amount of film present at any temperature and concentration β.

In Figs. 3 and 4 we offer our experimental results for the six different values of β explored. Concentrations higher than 30% exhibited no superfluidity in our accessible temperature range. Each curve details the mass per unit area of superfluid present, σ_s, as a function of the total amount of film present, for the particular temperature and ^{3}He concentration labeling the curve. The abscissa in each of these curves derives from our measurements of the mass of film present. We offer this measure of film extent because it issues directly from our instrumentation. However, we have also deduced from our measurements the magnitude of the van der Waals's thickness to which each of our mass estimate thicknesses corresponds. Our curves relating these two, together with an exposition on how we measured the relationship, is described in Ref. 32.

A central feature of the data obtained and

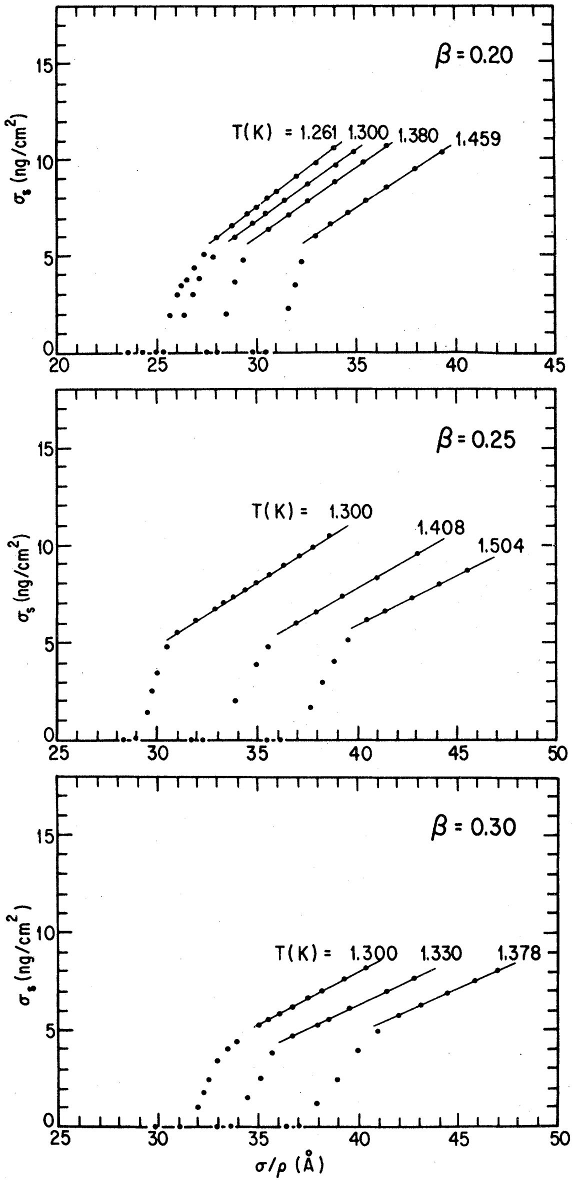

FIG. 4. The superfluid content in mixture films at the higher ^{3}He concentrations shown. The straight lines drawn each have slope $\rho_s(\beta, T)$.

displayed in Figs. 3 and 4 is that the superfluid data points tend very quickly to fall along a straight line. The lines that have been drawn through the data points each have slope $\rho_s(\beta, T)$. That is, the slope of each curve quickly tends toward a value just equal to the superfluid density corresponding to that for bulk helium, at the temperature T and concentration β explored in the experiment. We therefore conclude from the data that almost immediately after onset the differential superfluid fraction in the film $d\sigma_s/d\sigma$ corresponds to bulk. We define the "intercept areal mass density" σ_I by denoting the place on the abscissa at which the extrapolated straight-line region of the curve intercepts it as σ_I/ρ.

As is evident, each of the curves shows the relatively precipitous onset predicted by Kosterlitz, Thouless, and Nelson (KTN).[19,20] As reported earlier,[35] the height of the onset does not appear to depend upon β. It depends upon temperature in quite good agreement with the theoretical predictions[28,29]

$$\sigma_s^0 = (8\pi m^2 k/h^2)T \ , \tag{4}$$

where m is the mass of a ^{4}He atom and k and h are Boltzmann's and Planck's constants, respectively.

Both the onset areal mass density σ_0 and the intercept areal mass density σ_I relate to the microscopic length characterizing superfluidity in the mixture system. If one defines as the onset that place on the curve where the extrapolated straight line intersects the line $\sigma_s = \sigma_s^0$, then it follows from simple geometry that

$$\frac{\sigma_0}{\rho} = \frac{\sigma_I}{\rho} + \frac{\sigma_s^0}{\rho_s} \ . \tag{5}$$

Figure 5 is a graphical compilation of our characteristic length results. The figure displays our data points on the intercept "thickness" for each temperature and ^{3}He concentration explored. This "thickness" is not a literal one; rather it is our thickness estimator σ_I/ρ derived from the amount of mass in the film.

There is another commonly accepted thickness estimator for thin films which is much employed in the experimental literature on pure ^{4}He. To make contact with the wealth of data[2-7] using the van der Waals's film thickness estimates we offer this brief discussion on the relationship between the two film

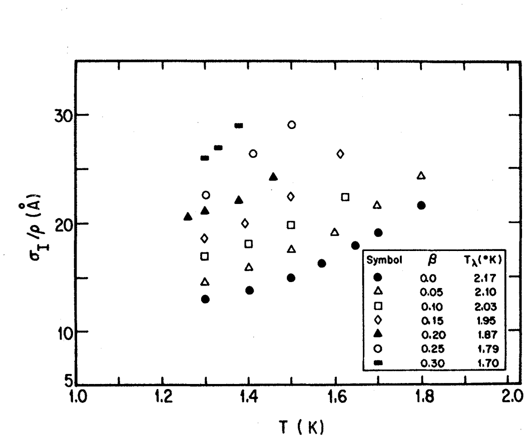

FIG. 5. The characteristic length for superfluidity as deduced from our measurements. The lengths shown are direct measures of the mass of film which fails to exhibit superfluidity even as the film grows infinitely thick.

thickness estimators. The thickness of an adsorbed film as estimated from van der Waals considerations is given in terms of pressure, temperature, and gas concentration as

$$d = D_0\{\Theta/T\ \ln[P_d(c,T)/P]\}^{1/3} \ , \tag{6}$$

where $D_0 = 3.6$ Å and our measured value of the "strength" of the van der Waals attraction Θ is $\Theta = 25$ K($\pm 7\%$). By virtue of our previous extensive measurements on the mixture system when it shows no superfluidity[32] and by reason of the fact that the intercept quantities, although representing microscopic information, are obtained by extrapolation from the straight-line thick film regime, the relationship between d_I and σ_I is simply given by

$$\sigma_I = \rho d_I + s \ , \tag{7}$$

where s is our experimentally determined "mass excess." Experimentally s seems to be a function of β only. Our measured values of s are displayed in Fig. 6. By contrast the relationship between d and σ is neither simple nor analytic. This relationship is given graphically in Ref. 32, together with the experimental derivation of s.

As in all pure ^{4}He data on superfluid characteristic lengths, there is some uncertainty as to how to extract the healing length, a fundamental parameter of superfluidity, from the lengths measured experimentally. By the "healing length" is usually meant that length over which the superfluid order parameter drops to zero at a boundary. It is generally believed that some amount, representing that part of the film—say the solid—which does not participate in the superfluid process, must be subtracted from σ_I (or σ_0) in order to determine a healing length. The amount to be subtracted is not unequivocal. For pure ^{4}He, values subtracted from d_I in the past range from one to two atomic layers, i.e., from 3.6 to 7.2 Å. The extensive measurements available on pure ^{4}He suggest that at sufficiently low temperatures the characteristic length d_I approaches a constant value of about 5.4 Å (one and one-half atomic layers) independent of temperature.[2,6] A good argument can be made that this low-temperature length limit characterizes the amount of inactive helium, due to its being compacted at high pressure and solidified at the substrate. Hence this may be the amount to be subtracted from d_I to yield the healing length.

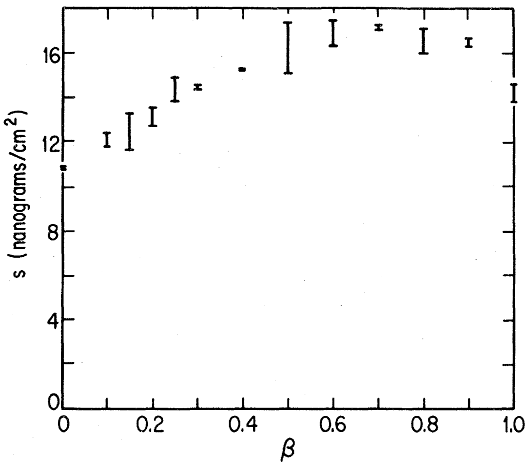

FIG. 6. Adsorbed mixture-film measured mass excess as a function of ^{3}He concentrations. This figure is reproduced from the J. Low Temp. Phys., Ref. 32.

Our view is that the significant quantity to consider is σ_I—that obtained from the intercept—not σ_0. The length d_I represents that thickness of film which fails to exhibit superfluidity even as the film grows infinitely thick. Similarily the intercept mass σ_I is precisely that mass of film which fails to exhibit superfluidity even as the total film mass approaches infinity. Since our measurements, reported here, are the only such extant on mixtures and these range down in temperature only to 1.3 K, where the intercept thickness still appears to descend with decreasing temperature, we have no evidence on a possible assymptotic value of σ_I. And because the ^{3}He content affects what might be attributable to an inactive (subtraction) thickness in the film, we offer our direct experimental results as given in Fig. 5 so as to be as free as possible from any weakly founded prejudicial notions leading to misinterpretations of the data. We hope, in a forthcoming publication, to demonstrate that all of the data extant—on pure ^{4}He and on mixtures—can be fitted with the results of a single simple idea: That the characteristic length effect observed in all films is attributable to interface (2D surface) excitations characterized by an energy gap of the order of 5 K.

Independent of any further theoretical analysis, certain qualitative features of our data have immediate implications.

(i) With regard to the possibility that our adsorbed films were significantly laterally inhomogeneous either due to surface-roughness-induced puddles[36,37] or to spontaneous droplet[38–41] formation.

The nearly exact universality of the onset jump height argues strongly against this. In any coalescence-based model of the precipitous onset phenomenon, the film thickness and surface tension play dominant roles in determining the onset point. Increasing thickness produces larger droplets and greater probability of a percolation onset. Decreasing surface tension also produces greater "wetting," resulting in a more uniform film. And the addition of ^{3}He produces both these effects. Mixtures of ^{3}He and ^{4}He are known to possess reduced free-surface tension[42–44] as compared to ^{4}He. The substrate-film interfacial tension is less altered because the fluid adjoining the substrate is nearly pure ^{4}He.[9,45–49] Our measurements show that the actual coverage (or thickness) at onset is much greater—almost twice as great, in some cases—for mixtures, as compared to pure ^{4}He. Only by an improbable coincidence could the coalescence of puddles or droplets ever be supposed to mimic the predicted KTN precipitous onset as a function of two variables—T and β. But even such a coincidence is difficult to propose when the basic driving forces seem to point in the opposite direction.

We feel that this is an important result of our work. Prior to this, even the remarkable experimental confirmation of the KTN-like predicted excess dissipation[22,23] performed by Bishop and Reppy[8] could have been explained as due to droplet or puddle coalescence mechanisms, leaving grave doubts as to the reliability of all thin-film measurements on imperfect substrates. But, as we interpret it, our mixture film evidence shows conclusively that fairly uniform films, yielding useful experimental results (at least in the multilayer region) are formed on materials other than specially prepared graphite.

(ii) With regard to the true thickness of the excess normal fluid region. Almost immediately after onset begins, and certainly within 3.6 Å, the differential superfluid fraction, $d\sigma_s/d\sigma$, assumes its bulk value of ρ_s/ρ. This indicates that the mechanism responsible for the excess normal fluid (superfluid deficit) in the film does not have a long "tail." Hence if the excess normal fluid were produced by surface excitations then their wave functions are sharply confined and do not have a long exponential decay into the fluid. This is in contradiction to one of the recent neutron scattering conclusions for ^{4}He films adsorbed on graphite.[50] They suggested that the surface rotons extended some 4 atomic layers (≈ 15 Å) into the fluid. Of course, both the strength and form of the van der Waals's potential in the liquid portion of films adsorbed on graphite is considerably different from that on our substrate (oxidized, vacuum-deposited aluminum), because of the differing substrate materials and solid layer thicknesses. This may well account for the difference observed. If so, it has important implications for theoretical models of those surface excitations. They must incorporate the density structure of the film.

IV. CONCLUSION

We have presented our experimental results on the details of onset and on the lengths characterizing the inception of superfluidity in films of mixtures of ^{3}He and ^{4}He. As expected, the qualitative effect of the presence of ^{3}He is roughly the same as that of increasing temperature; they both require a greater extent of film for superfluidity to appear, i.e., greater d_l. The essential data which relate to the healing length in mixtures are presented in Fig. 5. We have seen no evidence of the 2D phase transaction recently predicted theoretically.

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