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Phase diagram of ^{3}He-^{4}He mixture films

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Superfluid density and dissipation have been measured as functions of temperature in ^{3}He-^{4}He mixture films using a high-Q torsional oscillator. Transition temperatures between 1.3 and 1.9 K and ^{3}He concentrations between 0.5% and 56% were investigated. In these films the superfluid transition temperature decreases monotonically with ^{3}He concentration. This decrease is linear for smaller concentrations but becomes more rapid at higher concentrations. There were no clear signs of a transition to a phase-separated state observed. All the mixture films exhibit the characteristic universal jump in superfluid density at the transition. The results are compared with the bulk phase diagram and also with a simple phenomenological model.

I. INTRODUCTION

It has long been recognized that ^{3}He-^{4}He mixtures form a very interesting, albeit complex system. The properties and structure of bulk mixtures are well understood. ^{3}He atoms in these mixtures contribute only to the normal component of the fluid. This contribution is characterized by an effective mass which is larger than the ^{3}He atom bare mass. As a result, the superfluid density is significantly reduced by the addition of the ^{3}He and the superfluid transition temperature is depressed. In very dilute solutions the ^{3}He atoms prefer to stay near the surface, in what is called a "surface state". Beyond a certain concentration they start to dissolve into the bulk. Below 0.9 K the phenomenon of phase separation is observed, in which a ^{3}He-rich normal layer is formed above the ^{4}He-rich superfluid bulk.

In the form of a film, a ^{3}He-^{3}He mixture is an even richer and more complex system. In addition to the ^{3}He concentration, the film thickness and the interaction between the substrate and ^{3}He and ^{4}He atoms also play a crucial role in determining the properties of the film.

Mixture films, like their bulk counterpart, become superfluid at lower temperatures than pure films having the same amount of ^{3}He. In general, the transition temperature decreases as ^{3}He is added to a film. Wang[1] has seen an exception to this behavior. Superfluid transitions in mixture films are, qualitatively and quantitatively, similar to a Kosterlitz-Thouless transition.[2,3] It has been shown that in films at low enough temperatures, ^{3}He atoms reside in specific discrete states.[4] At higher temperatures there is a tendency towards a more homogeneous mixture.[5]

Two types of phase separation have been discussed for mixture films. One model suggests a bulk-type layered separation into ^{3}He and ^{3}He-rich phases.[6] Another scenario is a lateral phase separation in which ^{3}He-rich islands exit in a ^{4}He-rich sea.[7] There have been a number of experimental reports which present evidence either for a layered[8-10] or for a lateral[10,11] phase separation at low temperatures. Other experiments fail to detect any signs of a phase separation at all.[2,5,12,13] The overall picture in this respect is not very clear, although it seems that at very low temperatures phase separation in some form is likely.

In the present work we set out to confirm, and do a systematic study of, the positive shift in transition temperature with ^{3}He concentration reported by Wang. In addition, we set out to study the phase diagram for mixture films varying concentration, temperature, superfluid density, and transition temperature in the temperature range 1.3 to 1.9 K.

In the films studied, we find only a monotonic decrease of transition temperatures as a function of ^{3}He concentration. All the films display the characteristic universal jump in superfluid mass at the transition. There is no clear evidence for any type of phase separation in these films. In the thickest films, broadening of the dissipation peaks is seen at high ^{3}He concentrations. We also find that some high-concentration films remain superfluid at temperatures above which a bulk mixture of the same concentration would already be normal.

This paper is organized in the following manner. In Sec. II we briefly review previous experimental work on ^{3}He-^{4}He mixture films. Section III describes the experimental techniques utilized in the present work. Finally, in Sec. IV we discuss and analyze our results.

II. EXPERIMENTAL BACKGROUND

Over the past ten years considerable attention has been focused on ^{3}He-^{4}He solution films. These films exhibit a Kosterlitz-Thouless (KT) type of superfluid transition. The precise nature of the transition can be studied as a function of ^{4}He thickness and/or ^{3}He concentration. Properties of these films are significantly influenced by the interaction between the substrate and the ^{3}He and ^{4}He atoms.

Webster *et al.*,[2,12] using a quartz microbalance adsorptometer, measured the superfluid densities of ^{3}He-^{4}He films. They studied films of various thicknesses and ^{3}He concentrations up to 30% in the temperature range 1.3 to 2.1 K. The superfluid transition observed in these films was similar to the Kosterlitz-Thouless transition in pure

^{4}He films. The jump in the superfluid density maintained its universal character and was independent of ^{3}He concentration. The data failed to show any signs of phase separation in these films.

Similar behavior was seen in films 4 to 5 atomic layers thick and with a ^{3}He concentration of up to 50% by Laheurte *et al.*[13] They measured third-sound velocity in these films covering a temperature range of 0.6 to 1.3 K. They also observed a linear decrease of transition temperature with ^{3}He concentration.

Ellis *et al.*[8] measured third-sound velocities down to 0.4 K. Their data shows evidence of layered phase separation up to 0.5 K for a ^{4}He coverage of 5.7 atomic layers. Romagnan and Noiray,[5] also measuring third-sound velocities, failed to see any such evidence. Their data covered a similar temperature range for thinner films. Subsequently, Noiray *et al.*[9] scanned a larger range of ^{4}He coverage at a fixed temperature of 0.4 K. Their measurements of third-sound velocity indicate a layered phase separation for films having a more than 4.5 ^{4}He layers. Films with lesser amounts of ^{4}He behaved as a homogeneous mixture. This result seemed to be independent of ^{3}He content. Bhattacharyya *et al.*[4] carried out heat-capacity experiments on mixture films with ^{4}He thicknesses between 10 and 45 Å, and with 0.03 to 1 atomic layers of ^{3}He. They measured the contribution of ^{3}He to the heat capacity of various films as a function of temperature up to about 0.5 K. The authors proposed a model similar to Andreev's model[14] for bulk mixtures to explain their data. Bhattacharyya *et al.* suggested that, just as in the bulk case, the ground state for ^{3}He atoms in a mixture film is a surface state and has an energy given by

$$E=-\varepsilon_0+p^2/2m_1 \,, \tag{2.1}$$

where ε_0 and m_1 are the binding energy to the surface relative to the vacuum above it and the effective mass, respectively. The values of ε_0 and m_1 for the film are different from that of the bulk case. The excited states are discrete and the energy of the ith state is

$$E_i=-\varepsilon_i+p^2/2m_i \,. \tag{2.2}$$

The number of available states depends on the film thickness. For the thinnest film there is only one excited state. As thickness is increased, the number of states also increases until a three-dimensional (3D) continuum is reached marking the crossover to the bulk regime. In addition to the available states in the film there is also a 3D continuum of free-particle states in the vacuum above the film which are occupied by ^{3}He atoms which have evaporated out of the film.

Bhattacharyya and Gasparini[11] reported some evidence for a lateral phase separation for very thin films (10–12 Å) at low ^{3}He coverage (0.009–0.2 layers). They plot heat capacity of the films as a function of temperature below 150 mK and find a deviation from the expected straight line for a two-dimensional (2D) Fermi system. The slope of the line for such a system is presumably proportional to the surface area. The authors interpret the observed decrease in slope as sign of a phase separation in which "puddles" of high ^{3}He density appear in the system resulting in a decrease of the covered area. There are some discrepancies in the experimental data as pointed out by the authors which tend to undermine this interpretation. First, the existence of a low ^{3}He density phase is not indicated by the low-temperature behavior of the heat capacity. Second, the phase separation is not accompanied by an expected jump in heat capacity. Finally, there seems to be significant amount of entropy missing at low temperatures.

Finotello *et al.*[15] measured thermal conductance of mixture films between 12 and 16 Å thick, with less than 3% ^{3}He. They see a sharp increase in conductance as the films are cooled through the superfluid transitions. At a fixed temperature slightly above T_C, the value of the conductance decreases as the ^{3}He concentration increases. Conductance in this region is known to be proportional to the square of the 2D correlation length. Thus in contrast with the bulk case, ^{3}He tends to reduce the correlation length in films.

Finotello *et al.* also reported that in the superfluid region the maximum value of conductance for a film was lower for higher ^{3}He concentrations. This effect was explained by a presence of free vortices associated with the ^{3}He atoms even at $T<T_c$ for mixture films since the number of free vortices is inversely proportional to the conductance. This explanation at this point seems rather tentative in the absence of other evidence.

Agnolet *et al.*[3] used a torsional oscillator to study films with 0.24 atomic layers of ^{4}He and various ^{3}He concentrations. The superfluid transition temperature of the pure film was 220 mK. Upon adding ^{3}He, the transition shifted down linearly up to a concentration of 40% after which the drop in T_c was more rapid. The superfluid density for various films behaved in accordance with the prediction by Nelson and Kosterlitz[16] of the universal jump at the transition. In addition, films with more than 20% ^{3}He show an excess superfluid mass when extrapolated to zero temperature. This may be read as a sign of phase separation since the superfluid mass in a phase separated film would be larger because of the ^{3}He atoms expelled from the ^{4}He-rich regions.

Wang and Gasparini[10] have reported observations of two superfluid transitions. Using a torsional oscillator they have investigated films with about 1 atomic layer of ^{4}He. For small amount of ^{3}He impurity, two distinct superfluid transitions are seen. The magnitude of the jumps in the superfluid density at the two transitions are not equal. The authors explain these observations in terms of two different models of phase separation.

The first model is due to Mon and Saam.[6] They consider a double layered lattice whose sites can be occupied by either ^{3}He or ^{4}He atoms. A Hamiltonian of this system is written in terms of the interatomic coupling constants. Based on this Hamiltonian the phase diagrams of the system are sketched. It is shown that for certain values of the coupling constants, temperature, and concentration it is possible to have two superfluid transitions for a phase separated configuration, i.e, when one of the layers is ^{3}He-rich the other is ^{4}He-rich.

The second model was proposed by Guyer.[7] Using energy considerations, Guyer has shown that under certain conditions of temperature and concentration a stable configuration for mixture films is one in which patches of larger film thickness and ^{3}He concentration appear spontaneously in the system. According to Wang and Gasparini, two different film thicknesses and concentrations give rise to the two observed transitions of different strengths.

Clearly, we are far from having a coherent picture of the detailed nature of mixture films. Mixture films undergo a KT-like transition accompanied by a universal jump in the superfluid density. The presence of ^{3}He tends to reduce the superfluid density of the film—thereby, in general, decreasing the transition temperature. There are clear signs of some form of phase separation at low temperatures. However, the necessary conditions for this separation and its nature are not well understood.

III. EXPERIMENTAL METHOD

A. The torsional oscillator

A torsional oscillator was first used by Andronikashvili[17] to measure superfluid density in bulk helium. The motivation for this technique came from the two-fluid model. Andronikashvili's oscillator consisted of a stack of metal disks placed close to each other inside a cylindrical container. This cell was suspended by a torsion thread so that it could oscillate in liquid helium. The superfluid did not participate in this motion. The interdisk spacing was less than the viscous penetration depth of the normal fluid so that all of it was dragged along with the disks. Hence, the effective moment of inertia of the system could be used to deduce the superfluid density. This simple technique has been modified over the years for wider application.

The torsional oscillator and the electronic that we have used for our experiment was used earlier by Adams and Glaberson to study vortex dynamics in helium films and has been described in detail in their work.[18] We will outline the experimental method here.

The oscillator is similar to the high-Q oscillator of Bishop and Reppy.[19] A high value of Q enabled us to detect small changes in internal dissipation in the helium film. The resonant frequency of the oscillator was 500 Hz. This frequency was stable enough for us to detect a change of 10^{-10} sec in the period of oscillation which corresponds to less than one hundreth of one atomic layer of helium.

The experimental setup is shown in Fig. 1. It consisted of the cell suspended by a torsion rod, magnetic drive to set the cell in motion, and a set of capacitor pickups to monitor the motion. The cell contained a stack of about 9100 Mylar disks with an average gap of about 1200 Å between adjacent disks. The disks in the cell formed the substrate for the helium films with an effective surface area of 8.0×10^4 cm^2.

The oscillation of the cell gave rise to a fluctuating interplate gap in the pickup capacitors which resulted in a sinusoidal current signal. The current signal was transmitted to the drive coil via a positive feedback circuit. This arrangement drove the oscillator at its resonant frequency.

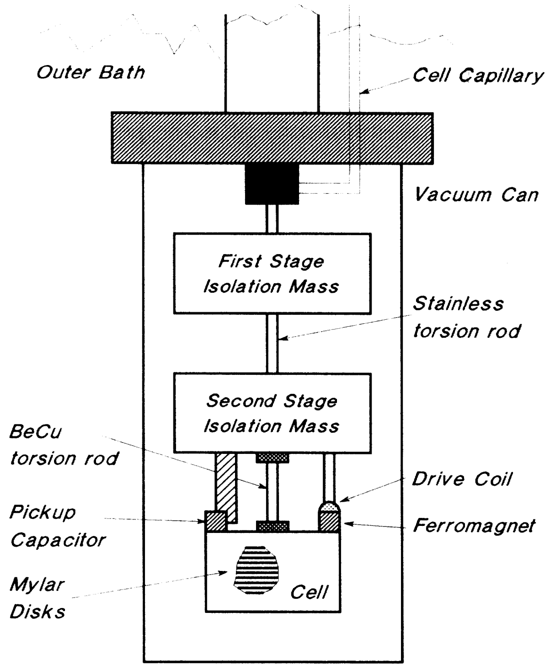

FIG. 1. The experimental setup.

The two isolation masses along with the stainless-steel torsion member connecting them acted as a mechanical filter which insulated the system from external mechanical noise and minimized possible energy losses thereby enhancing the signal to noise ratio and improving the Q of the system. This assembly of the oscillator and the isolation masses was sealed inside a vacuum can which was immersed in a helium bath.

B. Torsional oscillator and superfluid behavior

Changes in the period and amplitude of the oscillator can be used to determine the film thickness, superfluid density, and dissipation for helium films in the cell. In the following analysis of these relations, we express the moment of inertia and the torsion constant in units of $1/2R_{\text{cell}}^2$, where R_{cell} is the radius of the cell. At resonance, the amplitude of oscillation is given by

$$A = f_0 Q / k_0 \,, \tag{3.1}$$

where f_0 is the driving force, Q is the quality factor, and k_0 is the torsion constant.

The viscous penetration depth for this system is defined as the thickness of the fluid which effectively attaches itself rigidly to the substrate. For the normal fluid this is equal to $(2\eta_n/\omega\rho_n)^{1/2}$, where η_n is the coefficient of viscosity of the normal fluid and ω is the frequency of oscillation. In our situation this is about 20 μm, which is much larger than the thickness of our thickest films.

It follows that if the driving force and the external damping are kept fixed, then

$$\Delta A^{-1} \propto \Delta Q^{-1} \,, \tag{3.2}$$

where the change in Q is entirely due to the change in superfluid dissipation. Thus, by monitoring the amplitude of oscillation, we can determine the superfluid dissipation in the system.

The other quantity of interest is the period of oscillation which is given by

$$P=2\pi\sqrt{M'/k_0}\ , \tag{3.3}$$

where M' is the net effective mass of the oscillator. Since the superfluid does not couple to the substrate, M' can be expressed as

$$M'=M-M_s\ , \tag{3.4}$$

where M is the total mass of the cell (the substrate and the helium in the cell) and M_s is the superfluid mass. If P is the period of the cell above T_{KT} when all the helium in the cell is normal and A_s is the surface area of the substrate, when since $M_s/M \ll 1$, we can write

$$2\Delta P/P=\sigma_s A_s/M\ . \tag{3.5}$$

Here ΔP is the change in period relative to P and σ_s is the areal superfluid density. For our system $k_0=5\times10^8$ erg/cm and $A_s=8$ m^2.

Equations (3.2) and (3.5) provide the means to determine the dissipation and superfluid density from a knowledge of the two observables of the oscillator.

C. Determining the film thickness

The helium film thickness was determined by monitoring the change in period while the gas was allowed to enter the cell at a fixed low temperature T. To incorporate the mass loading of the oscillator due to the condensed liquid and the vapor between the mylar disks which are viscously clamped to the substrate, Eq. (3.5) can be rewritten as

$$\frac{2\Delta PM}{PA_s}=\rho_1 d+\rho_g D\ , \tag{3.6}$$

where ρ_1 and ρ_g are the bulk liquid and gas densities, respectively, d is the film thickness, and D is the cumulative gap between the disks occupied by the helium vapor.

Assuming that the helium vapor behaves like an ideal gas, we have

$$p=NRT/V\ , \tag{3.7}$$

where p and V are the pressure and the volume of the gas, respectively. If m_g is the mass of the gas and M_0 its molecular weight, then this equation can be written as

$$p=m_gRT/M_0V=\rho_gRT/M_0\ , \tag{3.8}$$

or

$$\rho_g=M_0p/RT\ . \tag{3.9}$$

Equations (3.6) and (3.9) can be combined to obtain

$$p=\left[\frac{2\Delta PM}{PA_s}-\rho_1 d\right]\frac{RT}{M_0(h-2d)}\ . \tag{3.10}$$

In writing this equation we have substituted $h-2d$ for D, where h is the interdisk gap. We can eliminate the pressure from this equation by incorporating the van der Waals interaction between the substrate and the helium atoms.

We start with the chemical potential for an ideal gas which is given by

$$\mu=(RT/M_0)[\ln(p)+\Phi]\ , \tag{3.11}$$

where Φ is a function of temperature only. Assuming that the chemical potential of a film is that of the bulk, with a contribution from the van der Waals potential, we can write

$$\mu_f=\mu_0-\alpha/d^3\ . \tag{3.12}$$

Here, μ_0 is the bulk chemical potential and α is the van der Waals constant. In equilibrium the chemical potentials of the liquid and vapor phases are the same. Hence, we can replace μ_0 and μ_f by the corresponding ideal gas expressions. This leads to the equation

$$\alpha/d^3=(RT/M_0)\ln(p_0/p)\ , \tag{3.13}$$

or

$$p=p_0\exp(-M_0\alpha/RTd^3)\ , \tag{3.14}$$

where p_0 is the saturated vapor pressure of helium.

From Eqs. (3.10) and (3.14) we obtain

$$\left[\frac{2\Delta PM}{PA_s}-\rho_1 d\right]\frac{RT}{M_0(h-2d)}=p_0\exp(-M_0\alpha/RTd^3)\ . \tag{3.15}$$

For a known ΔP at a given temperature, this equation can be iteratively solved to obtain the film thickness. The value of α was chosen to be 5×10^{-14} erg/cm^{3}g^{-1}.

IV. RESULTS

A. Superfluid density

We measured the superfluid density as a function of temperature in mixture films with ten different ^{4}He coverages ranging from 14 to 34 Å. The corresponding transition temperatures were between 1.32 and 1.86 K. We investigated a broad range of ^{3}He concentrations in these films. For the thinnest films we focused on the low-concentration end, whereas for thicker films ^{3}He content was incremented in larger steps and ranged up to 0.56.

Unless otherwise mentioned, we define the ^{3}He concentration as

$$X_3=\frac{m_3}{m_3+m_4}\ , \tag{4.1}$$

where m_3 is the total mass of ^{3}He in the film and m_4 is the total active mass of ^{4}He. The thickness of the active ^{4}He layer is obtained by subtracting the thickness of the inert layer from the total thickness. An inert ^{4}He layer exists near the substrate as a result of the strong van der Waals force present in the region. We have taken the thickness of this layer to be 7.2 Å (~2 atomic layers). It should also be noted that all the thicknesses and concentrations mentioned are the values at the corresponding transitions temperatures.

For each set of films we started with a pure ^{4}He run. This was followed by mixture film runs with increasing amounts of ^{3}He. Figure 2 shows a typical behavior of the superfluid density as a function of temperature. In each set of films the superfluid density at a fixed temperature decreases with increasing ^{3}He concentration. Thus the presence of ^{3}He atoms tends to decrease the superfluid component in the film.

The solid straight line (slope$=3.49\times10^{-9}$ gm/cm^2 K) drawn on this plot is the theoretical prediction of Nelson and Kosterlitz[16] for a universal jump in superfluid density at T_c. Although this prediction was made for pure ^{4}He films, it was later extended to the case of homogeneous ^{3}He-^{4}He mixture films by Berker and Nelson.[20] It is clear that the experimental jump is in reasonable agreement with the theory. Similar agreement has been reported by Webster *et al.*[12] who used a quartz-crystal adsorptometer, and at much lower temperatures by Agnolet *et al.*[3]

Berker and Nelson[20] have also shown that a lateral phase separation transition in the films would be observed as a kink in the σ_3 versus T curve accompanied by a diminished σ_s jump at T_c. Neither of these characteristics are seen in our data. Thus, we see no evidence for a lateral phase separation transition in the temperature, film thickness, and concentration range that we have studied.

B. Superfluid transition

The superfluid transition temperature T_c is plotted as a function of X_3 in Figs. 3 and 4. In all of the the ^{4}He coverages studied, T_c decreases monotonically with ^{3}He concentration. This is not a surprising result since, as mentioned above, ^{3}He atoms reduce the superfluid density. The change in T_c is linear for small concentrations of ^{3}He. As the concentration increases the rate of this change becomes more rapid. We observe quite a bit of scatter in the data for the thickest films.

The open circles shown in Fig. 4 trace the phase diagram for a bulk mixture. The region to the left of the circles is the superfluid region, and to the right all of the liquid is normal. This superposition of the bulk phase diagram reveals an interesting property of the films. For the same ^{3}He concentration a film can remains superfluid at temperatures much higher than the temperature where the bulk ceases to be superfluid. For instance, at a concentration of 0.4 the bulk becomes normal at about 1.38 K, whereas the film with 34 Å ^{4}He and the same ^{3}He concentration stays superfluid up to about 1.62 K. Since the films considered here are quite thick, this result is not significantly affected even if we include all the ^{4}He mass in the calculation of X_3 and not just the active mass. The physics behind this effect is not very obvious. It might, in part, be a result of the ^{3}He atoms being effectively pushed away from the walls due to the van der Walls force thus leading to a relatively rich ^{4}He layer near the walls. This scenario is consistent with the observations of Wang and Gasparini[10] and Romagnan *et al.*[21] where a tendency to-

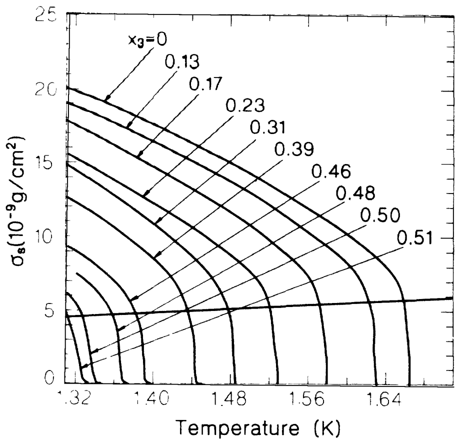

FIG. 2. Areal superfluid density vs temperature in mixture films starting with 23 Å of ^{4}He for the various indicated ^{3}He concentrations. The solid line represents the Nelson-Kosterlitz relation.

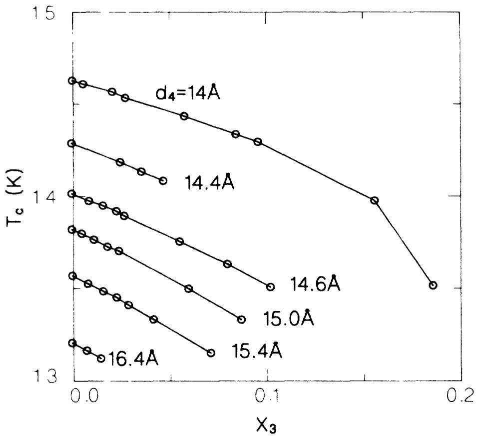

FIG. 3. Transition temperature vs ^{3}He concentration for thinner films.

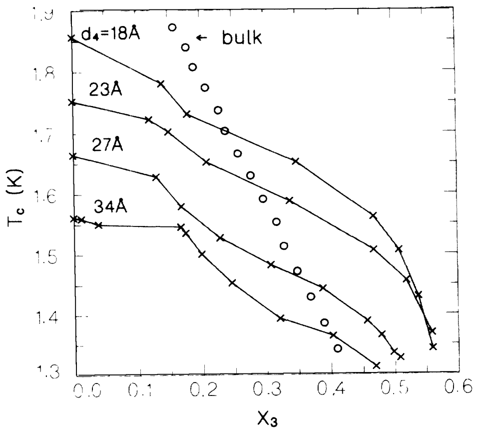

FIG. 4. Transition temperature vs ^{3}He concentration for thicker films and theoretical phase diagram for bulk mixture (open circles).

wards saturation for the change in T_c at large ^{3}He concentrations is seen.

Another possible cause involves the different effective mass of ^{3}He atoms in the two systems. In the bulk, the hydrodynamic backflow associated with the ^{3}He atoms resembles the effect due to a sphere moving in a fluid. The ratio of the effective mass to the actual mass for a ^{3}He atom in the surface state is about 1.5. If this atom is deeper in the bulk, however, the ratio increases to about 2.3.[22] The environment seen by a ^{3}He atom in the film is likely to be closer to that of the surface state than that of the bulk. The effective mass of a ^{3}He atom in the film is presumably smaller than its value in the bulk and is therefore less effective in reducing the superfluid density and transition temperature. This interference is corroborated by other work. Wang[1] has reported empirical values of the mass ratio in the films to be between 1.2 and 1.9—the ratio, in general, increasing with increasing ^{4}He coverage. Similar values and tendencies have also been seen in heat-capacity measurements by Bhattacharyya and Gasparini[11] and DiPirro and Gasparini.[23]

C. Comparison with other work

The monotonic decrease of T_c that we see for all the films is inconsistent with an observation by Wang.[1] He reported a single incidence of positive shift of T_c upon adding a small amount of ^{3}He to a pure ^{4}He film. Figure 5 shows Wang's data along with the behavior of two of our comparable films. The concentration in this plot is defined as $Y_3 = d_3/(d_3+d_4)$, where d_3 is the thickness associated with the ^{3}He atoms and d_4 is the thickness corresponding to the active ^{4}He atoms present. The reason for this discrepancy is not apparent. A possible cause could be a smaller ^{4}He coverage in the pure film run in Wang's experiment as compared to the three mixture runs. This could happen if there was a vapor-liquid disequilibrium in the cell during the first run, or if some ^{4}He leaked into the cell before the other runs.

As mentioned above, Wang and Gasparini[1,10] have reported two superfluid transitions, *A* and *B*, in very thin mixture films. They interpret this as an evidence of a layered phase separation in accordance with the theory of Mon and Saam.[6] We do not see this effect in any of our films in the somewhat higher-temperature range of our experiments.

Figure 6 shows the behavior of one of the films from Ref. 1. The transition temperature is that of the KT-like "*A*" transition. Also shown is the data for some of our thinner films. The slope of our data in the linear region is comparable to that of Wang's data. Although, their data also shows an increased slope beyond a concentration of 0.4, the change is not as drastic as that in our plots and it appears at higher concentrations. This difference is perhaps an indication of the crucial roles that thickness and temperature play in the behavior of mixture films.

This point is further demonstrated by some other reports[10,21] of experiments on even thinner films at lower temperatures. In these works the transition temperature, after falling initially, levels off beyond a certain ^{3}He coverage. This has been interpreted as a sign of layered phase separation which causes all additional ^{3}He atoms to end up in the ^{3}He-rich normal layer without altering the superfluid density of the ^{4}He-rich layer.

The final comparison that we make is with the data presented by Lahuerte *et al.*[13] Results of their third-sound experiment are shown in Fig. 7 along with our data. As before, the transition temperature is plotted as a function of concentration. This plot differs from the previous ones in two respects: first, the ^{3}He concentration is calculated by taking all of the condensed ^{4}He into account (not just the active part); second, the lines in the graph connect points which correspond to a constant total coverage and not constant ^{4}He coverage. In our experiment we had very few runs with the same total coverage since in each series of runs we had a fixed amount of ^{4}He and varying amounts of ^{3}He.

As seen in the graph, Laheurte *et al.*'s data show a linear behavior. On the basis of the small number of our data points we are not in a position to contradict or confirm this behavior. To the extent that our data does show a linear dependence, the slope seems to be smaller than that of Laheurte *et al.*

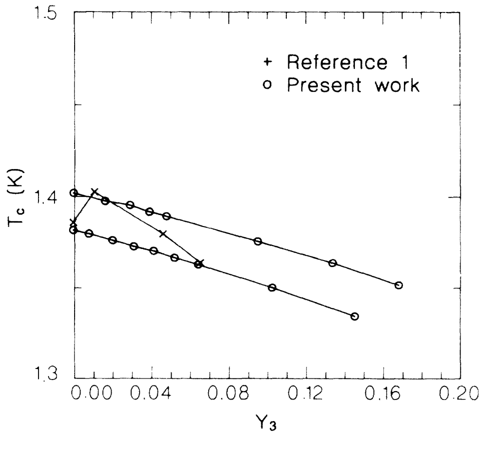

FIG. 5. T_c vs Y_3 for two films from this work and the anomalous data from Ref. 1.

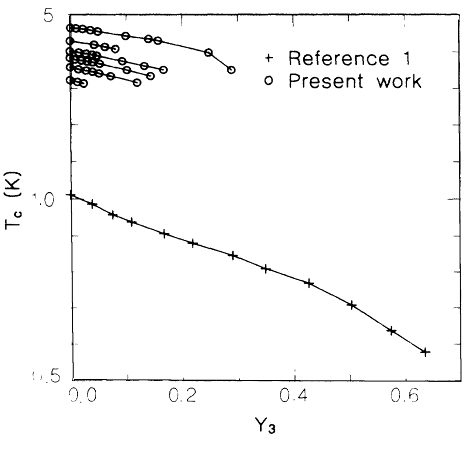

FIG. 6. T_c vs Y_3 for some of our thinner films and one of the films from Ref. 1.

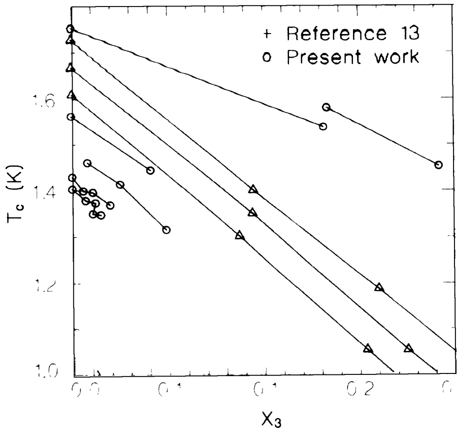

FIG. 7. T_c vs X_3 for films with constant total coverage along with the data from Ref. 13.

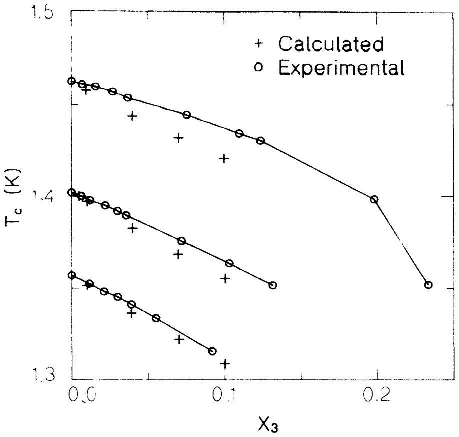

FIG. 8. Calculated and experimental T_c vs X_3 the ^{3}He molar concentration for films with 14.4, 15.0, and 16.4 Å of ^{4}He.

D. A simple model

It is quite clear from the discussion so far that in the temperature range of our experiment there is no empirical evidence for a phase separated mixture film. This suggests that at such high temperatures we have a fairly homogeneous mixture. This is in agreement with what Webster *et al.*[2] see in the same temperature region. They have plotted the areal superfluid density versus the total coverage at a fixed temperature and concentration and find a linear behavior away from the transition—the slope of the line being equal to the bulk superfluid density at that temperature and concentration. Romagnan and Noiray[5] have used this finding to calculate theoretical curves for Tc versus X_3.

We use a somewhat similar procedure to calculate the transition temperatures as a function of concentration. Instead of looking at the superfluid density away from the transition we focus at the transition itself. As we have seen, Nelson's universal jump is manifested by mixture films also. Thus, we can write

$$T_c(X_3)=\sigma_3(T_c)3.49\times10^{-9}\ , \tag{4.2}$$

where $T_c(X_3)$ is the transition temperature of a film with concentration X_3. In our experimental data T_c was fixed as the temperature at which the dissipation peak occurred. We can write the areal superfluid density in terms of the bulk density ρ_s at temperature T_c and concentration X_3 and the film thickness d at T_c

$$\sigma_s(T_c)=f\rho_s d\ . \tag{4.3}$$

Here f is a factor which takes into account the renormalization of the superfluid density at transition. f was determined by observing the transition in the film with the same amount of ^{4}He but no ^{3}He. If T_0 is the transition temperature of the pure ^{4}He film and ρ_{S0} and d_0 are the bulk ^{4}He density and film thickness at T_0, respectively, then

$$f=(3.49\times10^{-9})T_0/\rho_{s0}d_0\ . \tag{4.4}$$

Having obtained the value of f we could solve Eqs. 4.2 and 4.3 iteratively since we have already calculated the film thickness as a function of temperature. The values of ρ_s for various concentrations were obtained from the work of Sobolev and Esel'son[24] and of Dash and Taylor.[25]

Figures 8 and 9 show the calculated and the experimental behaviors for three of the thinner films and one of the thicker films. The agreement between the calculated and the experimental values is best for the thinnest film and progressively worse for thicker films. Since our model assumes bulklike behavior for the films, we would have expected the agreement to be better for thicker films. Obviously, a more rigorous and sophisticated model is needed to explain the behavior of these mixture films. Such a model would have to take into account the hydrodynamics of ^{3}He atoms in a film. As noted earlier, there is also likely to be a concentration gradient perpendicular to the film owing to the fact that the ^{3}He atoms prefer to stay away from the substrate. This would further complicate the hydrodynamic of the system and influence the effective mass of the ^{3}He atoms.

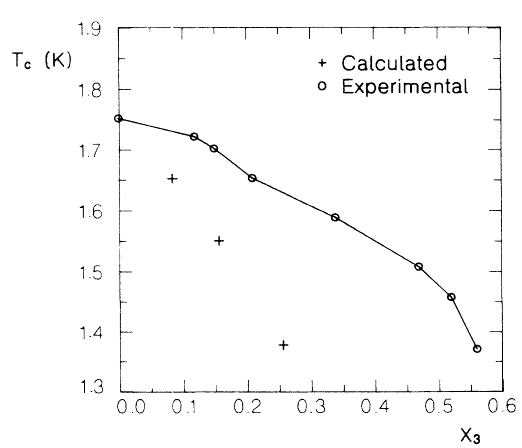

FIG. 9. Calculated and experimental T_c vs X_3 for a film with 27 Å of ^{4}He.

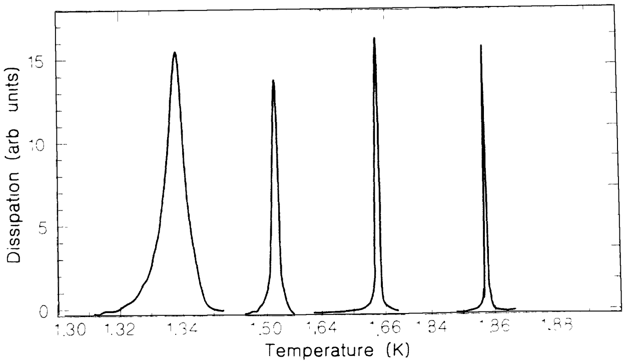

FIG. 10. Dissipation peaks for $d_4 = 34$ Å and various values of d_3: 0, 34, 66, and 86 Å.

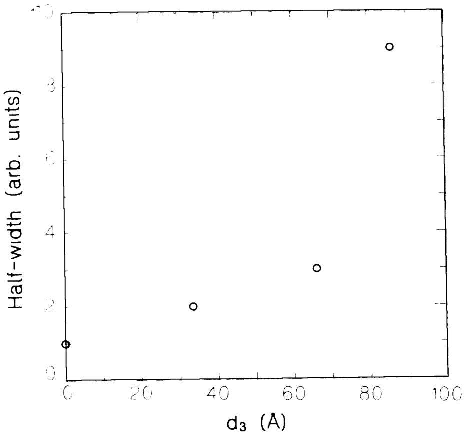

FIG. 11. Half-width of dissipation peaks vs ^{3}He coverage for $d_4 = 34$ Å.

E. Dissipation in mixture films

Excess dissipation in the superfluid films is extracted by subtracting the value of the inverse of the amplitude at some desired temperature from its value above the transition. Equation (3.2) can be used to obtain the dissipation peak at the transition.

In thicker films there seems to be a tendency for the peaks to broaden as large amounts of ^{3}He are added. This behavior is shown in Fig. 10. From these plots we measured the half-widths of the peaks and plotted them as a function of ^{3}He concentration in Fig. 11.

The origin of this broadening of the dissipation peaks is not well understood. It could be a sign of some interesting physical effect that the presence of ^{3}He brings about in an intrinsic way. On the other hand, it could merely be an instrumental effect resulting, for instance, from a possible inhomogeneity in the distribution of ^{3}He or a temperature gradient in the cell. In either of the latter cases, different parts of the liquid would undergo transition at different temperatures resulting in an overall broadening of the transition.

V. SUMMARY

We have investigated ^{3}He-^{4}He mixture films using a torsional oscillator over a wide range of film thicknesses and ^{3}He concentrations. In these films superfluid density and dissipation was measured as a function of temperature. This data was used to extract the superfluid transition temperature for the films.

The superfluid density at a fixed temperature was seen to be a decreasing function of ^{3}He concentration. In all of the films studied the transition temperatures decreased monotonically with the addition of ^{3}He. The jump in the superfluid density was in accordance with the universality prediction for KT-like transitions. We see no signs of any kind of phase separation transition in the entire temperature, thickness, and concentration range scanned.

The principal focus of this work was on the low-concentration end. In the thinnest films T_c decreases linearly with ^{3}He concentration. We do not see a positive shift in T_c, contrary to what has been reported by Wang. At larger concentrations the drop in T_c is more rapid.

The dependence of T_c on concentration in thicker films is similar to what is seen in the thinner films. The data, however, seems to be more scattered in this case. It can be seen that some high-concentration films remain superfluid at temperatures where a bulk solution of the same concentration would already be normal. This is an interesting phenomenon in whose cause is not fully understood at present although it is likely that ^{3}He atoms are expelled from the vicinity of the substrate leading to an effectively lower concentration there.

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