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Phase diagram for some ^{3}He-^{4}He mixture films from third-sound experiments

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Third-sound velocity measurements in ^{3}He-^{4}He mixture films are used to obtain the phase diagram for different coverages. The characterization of these films is discussed, and a correct parameters choice is proposed. Using a rough estimate of the superfluid density, we find that at the superfluid transition temperature the same universal feature as in ^{4}He film is observed. The general behavior of our films is, however, not similar to theoretical predictions for two-dimensional mixtures, and the phase separation is not detected. New experiments are suggested by our results in order to clarify the behavior of mixture films.

I. INTRODUCTION

The study of ^{3}He-^{4}He mixture films is the subject of a growing interest both theoretically[1,2] and experimentally.[3] These films should show, as in the case of bulk mixtures, both superfluidity and phase separation. So they represent one of the best materials to study transitions in two dimensions (2D). Starting from a spin-1 Ising model, originally used by Blume, Emery, and Griffiths[4] to simulate the thermodynamics of bulk ^{3}He-^{4}He mixtures, recent theoretical calculations[1,2] have considered a generalization of this model and apply renormalization procedures for 2D mixtures. They obtain the phase diagrams of 2D, ^{3}He-^{4}He mixtures which include of course the phase separation and superfluid transition. The general features of the diagrams depend on the ratio K/J, which is associated with experimental conditions such as substrate type and film thickness. Depending on this ratio different phase diagrams have been predicted. None have been observed yet. The location of the end points of the various curves and the relative extension of the different phases in the temperature-concentration space $(T-x)$ is strongly affected by the value of K/J. The superfluid transition itself is described with the picture of superfluidity due to Kosterlitz and Thouless[5] which is very successful for ^{4}He films. One of the striking consequences of this model is that, as long as no phase separation occurs, the superfluid density jumps discontinuously at the superfluid transition temperature T_s in a universal way[6]

$$\lim_{T\to T_s} \frac{d\langle\rho_s(T)\rangle}{T} = \frac{2m^2k_B}{\pi\hbar^2} = 3.49\times10^{-9}(\mathrm{g\,cm^{-2}\,K^{-1}}) \quad . \tag{1}$$

It was mentioned that these models can be applied to real ^{3}He-^{4}He films of several atomic layers, as long as average quantities over the film thickness are used. There is, of course, strong motivation for experimental determination of phase diagrams to check these theories and the universal character of the superfluid transition.

However there is not yet to our knowledge any precise experimental determination of such diagrams for ^{3}He-^{4}He films. This fact is related to the great difficulty of characterization of the parameters in mixture films. The determination of the thickness and concentration of the film requires great care. Effectively it is well known[7,8] that the concentration and pressure are not uniform in the vicinity of a solid wall. So, for instance, the concentration at the free surface of the film is different from its average concentration. The effect of both the concentration and dimensionality has already been experimentally observed.[9] However the lack of rigorous characterization of the film parameters make these observations essentially qualitative. Also specific-heat measurements of monolayer films adsorbed on grafoil have been done.[10] The results were inconclusive about the existence of phase separation in this monolayer. More recently, Webster *et al.*[3] have studied the superfluid transition in mixture films using a microbalance technique above $T=1.3$ K. Their results confirm the universal character of the superfluid density jump[6] predicted by the Kosterlitz-Thouless theory, which does not depend on concentration. In this experiment the film was indirectly characterized through the knowledge of pressure and concentration of the vapor phase. This method is not easy to use at lower temperature and also does not give directly the average concentration useful for a phase diagram.

In this work we present the results of third-sound velocity measurements used to give the onset superfluid temperature for several mixture films. Our characterization of the thin films is directly the average concentration $\bar{x}$ and n the number of atoms adsorbed per unit surface. The range of measurements extends from 0.5 to 1.8 K for three different coverages and average concentration going up to 0.4.

In the second section we describe a rigorous way to characterize the mixture film parameters. The third section is devoted to the experimental technic description. In Sec. IV we present our results and the analysis. In the last section we summarize and give some concluding remarks.

II. CHARACTERIZATION OF THE ADSORBED FILM MIXTURE

A. General description

In order to get with good accuracy the film average concentration and the number of atoms adsorbed by unit area we use a porous material (P) with a large adsorption surface S. The test chamber contains a porous material (P), and a glass substrate (G) on which third-sound experiment is performed. Because of the large value of S, the number of atoms adsorbed on the glass substrate and on the chamber walls is negligible with respect to the total number of atoms adsorbed on the porous material, and if we call N_{3a} and N_{4a} the number of atoms, respectively, of ^{3}He and ^{4}He adsorbed in the experiment (N_{3a} and N_{4a} are easily determined as we shall see in Sec. III) we can define within a very good approximation: $n_P=(N_{3a}+N_{4a})/S$, the number of atoms adsorbed by unit area on the porous material; and $\bar{x}_P=N_{3a}/(N_{3a}+N_{4a})$, the average concentration of the film adsorbed on the porous material. As the experiment does not study the film adsorbed on the porous material but the one adsorbed on the glass substrate, this last one must be characterized by two similar parameters n_G and $\bar{x}_G$.

As the van der Waals interaction between He atoms and the two substrates (G) and (P) are different we do expect n_P and n_G to be different. On the other hand, the relation between $\bar{x}_P$ and $\bar{x}_G$ is not obvious as we know that the concentration changes spatially in the film. The determination of n_G and $\bar{x}_G$ is however very easy as soon as we use a continuum model[7] to describe the film. In the experimental chamber for a given temperature T, the pressure P and gas concentration C are fixed. Equilibrium conditions imply that the concentration and the pressure at the free surface of the films adsorbed on the two different materials are equal:

$$x_G(d_G)=x_P(d_P)=\beta \ , \tag{2}$$

$$P_G(d_G)=P_P(d_P)=P_0(C,T) \ , \tag{3}$$

where β is the equilibrium liquid concentration corresponding to the gas concentration C at the temperature T; $P_0(C,T)$ is the liquefaction pressure of the gas which must be equal to the liquid's vaporization pressure

$$[P_0(C,T)]_{\text{dew}}=[P_0(\beta,T)]_{\text{boil}} \ . \tag{4}$$

As long as no capillary condensation occurs in the porous material, the films are uniform and the thicknesses of the films adsorbed on glass, d_G, and on the porous material, d_P, are related[11,12] by

$$\Theta_G d_G^{-3}=\Theta_P d_P^{-3} \ , \tag{5}$$

with Θ_G and Θ_P the van der Waals interaction constants between an He atom and each substrate.

We can then use the thermodynamic theory of thin-film ^{3}He-^{4}He solutions[7] to deduce a useful property of scaling in the films. Effectively it is easy to show by integrating Eqs. (14) and (13) of Ref. 7 that

$$x_i=F(P_i,P_0,\beta,T) \ , \tag{6}$$

$$\frac{\Theta_i}{Z_i^3}-\frac{\Theta_i}{d_i^3}=H(P_i(Z_i),P_0,\beta,T) \ , \tag{7}$$

where the subscript i is used for the two materials (P) and (G), Z_i being the distance of an He atom from the wall substrate.

The last relation implies that for given β, T, and P_0, which is the case for the two films adsorbed on (P) and (G) in the same chamber, we find in the two films the same pressure P_i and so with Eq. (6) the same concentration x_i, when

$$\frac{\Theta_P}{Z_P^3}-\frac{\Theta_P}{d_P^3}=\frac{\Theta_G}{Z_G^3}-\frac{\Theta_G}{d_G^3} \ . \tag{8}$$

Then using Eq. (5) we find that the profile in the film adsorbed on material (G) can be deduced from the profile in the film adsorbed on material (P) by simple scaling factor $(\Theta_G/\Theta_P)^{1/3}$ along the Z axis

$$P_P(Z_P)=P_G((\Theta_G/\Theta_P)^{1/3}Z_P) \ , \tag{9}$$

$$x_P(Z_P)=x_G((\Theta_G/\Theta_P)^{1/3}Z_P) \ . \tag{10}$$

The calculation of n_G is then easy. If we define the density number of He atoms on glass $n_G(Z_G)$, we can write

$$n_G=\int_0^{d_G} n_G(Z_G)\,dZ_G \ . \tag{11}$$

Using the properties of scaling obtained for our thermodynamic variables, $n_G(Z_G)=n_P((\Theta_P/\Theta_G)^{1/3}Z_G)$, and then changing our variable $Z_P=(\Theta_P/\Theta_G)^{1/3}Z_G$ in the integral (11) we get

$$n_G=(\Theta_G/\Theta_P)^{1/3}n_P \ . \tag{12}$$

In the same way it is easy to show that

$$\bar{x}_G=\bar{x}_P \ . \tag{13}$$

So in conclusion the mixture film adsorbed on the glass substrate is characterized by a coverage

$$n_G = (\Theta_G/\Theta_P)^{1/3} \frac{(N_{3a}+N_{4a})}{S} \tag{14}$$

and average concentration

$$\bar{x}_G = \frac{N_{3a}}{(N_{3a}+N_{4a})} \ . \tag{15}$$

B. Porous material

We use as porous material 75 millipores filters.[13] The average pore diameter is 500 Å. The surface adsorption area is measured by the classical Brunauer, Emmett, and Teller (BET) (Ref. 14) method with nitrogen adsorption isotherm. We find $S = 37$ m^2. The dead volume of the porous material is 2.26 cm^3 while its porosity is 0.77. For a good understanding of He film adsorption on this material, we have first measured the adsorption isotherm of pure ^{4}He at 2.2 K. Knowing the saturated vapor pressure P_0 and measuring both the pressure P in the chamber and the number of adsorbed atoms N_a, we can represent our results (Fig. 1) as $[T \ln(P_0/P)]^{-1/3}$ vs N_a.

The experimental points show two domains. First of all we observe a linear variation. As $(T \ln P_0/P)^{-1/3}$ is proportional to the film thickness in the Frenkel-Halsey-Hill[15] description, it means that the film thickness increases proportionally to the number of adsorbed atoms N_a. This fact is easily understandable, as above the two first atomic layers the density remains constant in the film and equal to bulk liquid density. The thickness change for a given increase ΔN_a should be $\Delta N_a v_4/S$, where v_4 is the atomic volume of bulk liquid ^{4}He. Then the slope of the straight line we observe in Fig. 1 is $v_4 \Theta_P^{-1/3}/S$. We then get an estimation of the van der Waals interaction constant between an He atom and the porous material $\Theta_P = 63$ K (atomic layers)^3.

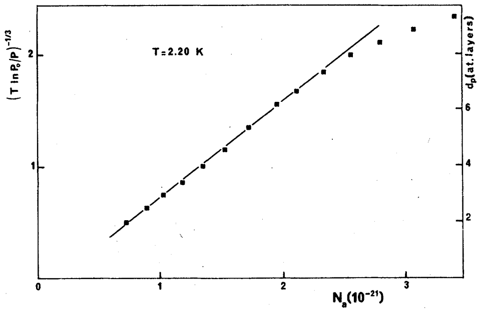

FIG. 1. ^{4}He isotherm adsorption on the porous material. The experimental results, plotted as $(T \ln P_0/P)^{-1/3}$ vs the number N_a of adsorbed atoms, show two domains. From the slope of the linear region we deduce an estimation of the van der Waals interaction constant for the porous material $\Theta_P = 63$ K (atomic layers).^{3} This value is used to obtain the thickness d_P of the adsorbed film on the porous material (right vertical axis). For $N_a \simeq 2.3 \times 10^{21}$ we observe a departure from the linear behavior, characteristic of the onset of the capillary condensation.

From this value we can easily calibrate the vertical axis in thickness d_P (Fig. 1). We also notice that the straight line does not go through the axis origin. This comes from the fact that the two first atomic layers have a larger density than the bulk one. We find that we need 1.27 more atoms to build the two first layers than the two next ones. This figure is in good agreement with the results of other experiments.[16] Above a critical value $N_{ac} = 2.3 \times 10^{21}$ (which corresponds to $n_{Pc} = 0.62$ at./Å^2) we observe a departure from the linear behavior when introducing more He atoms. The pressure increases very slowly with N_a. It is a characteristic feature of capillary condensation which is now well known to occur in porous material with large surface adsorption area.[17] The thickness growth of the film is *no more uniform* as added He atoms condense at contact points. These effects have been recently studied by sound velocity measurements in powders.[18] As mentioned in Sec. II A, a good characterization of our films by Eqs. (14) and (15) is only valid when the films are uniform, i.e., when $n_p < n_{Pc}$. In order to study dimensionality effects we must change the film thicknesses in a sufficient range; so we need materials for which capillary condensation occurs for a large value n_{Pc} of the number of atoms adsorbed by unit area. Many materials have been tested[19] to find the best compromise between large S and n_{Pc}. For millipore filters that we finally use, capillary condensation will only occur with ^{4}He for a critical coverage $n_{Pc} = 0.620$ at./Å^2.

As the surface tension of ^{4}He is greater than the one of ^{3}He we are sure that mixture critical coverage will be larger than n_{Pc}. In this work we always use coverage smaller than 0.62 at. Å^{-2} to prevent any capillary condensation. The three different coverages used are $n_{P1} = 0.46$ at. Å^{-2}, $n_{P2} = 0.50$ at. Å^{-2}, and $n_{P3} = 0.53$ at. Å^{-2}.

C. Glass substrate

The superfluid onset temperature in mixture films are obtained from third sound velocity measurements in films covering the glass substrate. Characterization of this film is possible using relations (14) and (15). N_{3a} and N_{4a} are obtained from gas mixture introduction (see Sec. III), all other parameters except Θ_G are known [$S = 37$ m^2, $\Theta_P = 63$ K (atomic layers)^3]. We use for Θ_G the value 27 K (atomic layers)^3 which is in the literature[20] and also generally used by other authors[21] working with glass. However

TABLE I. Superfluid onset (on glass substrate) of pure ^{4}He films for different coverages n_p of the porous material. The thickness d_{G4} of the film adsorbed on the glass substrate is obtained from pressure measurements using $\Theta_G = 27$ K (atomic layer)^3.

n_p (at. Å^{-2})	0.456	0.497	0.531
n_G (at. Å^{-2})	0.344	0.375	0.400
d_{G4} (atomic layer)	3.96	4.37	4.69
T_s (K)	1.61	1.67	1.73
$10^9 d_{G4}\langle\rho_s\rangle/T_s$ (g cm^{-2} K^{-1})	3.24	3.45	3.57

to check this value in our experiment we have measured the superfluid onset temperature T_S for three different ^{4}He films covering the glass substrate. The film thickness is obtained from pressure measurements using relation

$$d_{G4} = \left[\frac{T}{\Theta_G} \ln \frac{P_0}{P} \right]^{-1/3} \tag{16}$$

with $\Theta_G = 27$ K (atomic layers)^3. The value of $d_{G4}\langle\rho_s\rangle/T_s$ for this film obtained as usual from third-sound velocity[22] is also evaluated. These results are presented in Table I.

The good agreement between the theoretical value[6] 3.49×10^{-9} g cm^{-2} K^{-1} and the experimental ones can be used as a test of the correctness of $\Theta_G = 27$ K (atomic layers)^3.

So in the following the three different coverages of the films on glass that we study are: $n_{G1} = 0.34$ at. Å^{-2}; $n_{G2} = 0.37$ at. Å^{-2}; and $n_{G3} = 0.40$ at. Å^{-2}.

III. EXPERIMENT

The preparation of gas mixture is made at room temperature. The total number of ^{3}He atoms, N_3, and ^{4}He atoms, N_4, is obtained from pressure measurements in calibrated volume. This mixture is then condensed at low temperature in the experimental chamber through a stainless-steel filling capillary (0.6 mm i.d.). The pressure in the experimental chamber is measured with a digital pressure gauge.[23] Thermomolecular corrections are calculated from McConville results.[24] The number of ^{3}He and ^{4}He atoms in the vapor phase at the onset temperature T_s, respectively, N_{3v} and N_{4v}, are evaluated from the measured pressure, knowing the free volume and the gas concentration C. The free volume at low temperature in the experimental chamber is 4.06 cm^3; at room temperature it is 25 cm^3. This means that in our experiments we always have more than 97% of the gas atoms in the experimental chamber. The gas concentration C is evaluated from numerical calculations in the frame of the thermodynamic theory of thin-film mixtures.[7] In the experiments that we present in this paper the value of C holds between 0.85 and 1. The numbers of adsorbed atoms N_{3a} and N_{4a}, respectively, of ^{3}He and ^{4}He are

$$N_{3a} = N_3 - N_{3v} \ , \quad N_{4a} = N_4 - N_{4v} \ . \tag{17}$$

From these values we deduce

$$n_P = (N_{3a} + N_{4a})/S$$

and

$$\bar{x}_P = N_{3a}/(N_{3a} + N_{4a}) \tag{18}$$

and of course we characterize our film covering the glass by

$$\bar{x}_G = \bar{x}_P \tag{19}$$

and

$$n_G = n_P \left(\tfrac{27}{63}\right)^{1/3} \ .$$

We must outline that these values are obtained for each experiment at the onset temperature T_s.

The uncertainty on the average concentration $\bar{x}_G$ is due to errors in the evaluation of N_v and essentially on estimation of C. However, using a porous material with a large surface area S, makes the number of adsorbed atoms larger than the number of atoms in the vapor phase and improves the accuracy on $\bar{x}_G$. Of course the lower the pressure is (i.e., the temperature) the more accurate becomes the determination of $\bar{x}_G$ (see Table II): the relative error on $\bar{x}_G$, which is about 11% at 1.3 K, is only 5% at 1 K and becomes less than 1% below 0.8 K.

Of all our experiments, we have chosen to present here those for which we find nearly the same coverage n_G of the mixture film at its onset temperature T_s. We then present three sets of experimental results characterized by a mean value of n_G. The scatter in n_G within a set of experiments is always lower than 0.6% (see Table II).

The third-sound velocity is measured by the usual technique.[25] Two aluminum strips are evaporated on the glass substrate at a distance of 1.6 cm. They are used, respectively, as emitter and detector and polarized with the help of an external magnetic field near

TABLE II. Experimental results: superfluid onset temperatures of mixture films.

n_G (at. Å^{-2})	$\bar{x}$	T_s (K)	d_G (atomic layer)	$10^9 d_G \langle \rho_s \rangle / T_s$ (g cm^{-2} K^{-1})
0.345 ± 0.002	0.113 ± 0.011	1.30	4.06	2.80
	0.195 ± 0.008	1.05	4.17	2.74
	0.323 ± 0.001	0.640	4.30	3.79
	0.360 ± 0.001	0.570(*D*)	4.37	3.55
0.375 ± 0.002	0.121 ± 0.013	1.35	4.51	2.82
	0.221 ± 0.01	1.05	4.63	3.49
	0.303 ± 0.004	0.815	4.75	3.67
	0.357 ± 0.001	0.679(*B*)	4.82	3.98
0.401 ± 0.003	0.123 ± 0.014	1.40	4.84	2.95
	0.202 ± 0.012	1.185	4.94	3.08
	0.281 ± 0.007	0.950	5.05	3.95
	0.366 ± 0.002	0.753(*A*)	5.18	4.38
	0.414 ± 0.001	0.607(*C*)	5.24	4.70

the superconducting transition. The signal coming from the detector is averaged in a Tracor-Northern[26] multichannel analyzer.

The correct determination of the superfluid onset temperature T_s has been for a long time a difficult question as the signal is strongly attenuated and its shape modified near T_s. The work of Bishop and Reppy[27] has clarified this point. Although it is not possible to follow a third-sound signal through the transition region, it can be followed to the point where the dissipation begins to rise rapidly. The value of the superfluid mass at this point gives a reasonably good estimate for the static value of the Kosterlitz-Thouless jump. This has been checked recently by Rudnick.[28] We have seen (Sec. II C) that our determinations in the case of pure ^{4}He are in good agreement with results obtained by Rudnick *et al.*[21,28]

IV. RESULTS AND DISCUSSION

We present our experimental results of superfluid transition temperature deduced from third-sound velocity measurements in Fig. 2 and Table II. For the three different coverages chosen we observe that the superfluid transition temperature in mixture films decreases almost linearly with increasing ^{3}He concentration for a given coverage and that the superfluid transition temperature is reduced when the coverage decreases.

A complete discussion of the superfluid transition should of course rely on the Kosterlitz-Thouless picture[5] and the value of $d_G \langle \rho_s \rangle / T_s$ at the transition. Such value is only indirectly obtained by third-sound velocity C_3 measurements, as $\langle \rho_s \rangle$ is evaluated from C_3 using a hydrodynamic theory. This theory has recently been proposed in the case of mixture films.[29] The detailed analysis requires a very good knowledge

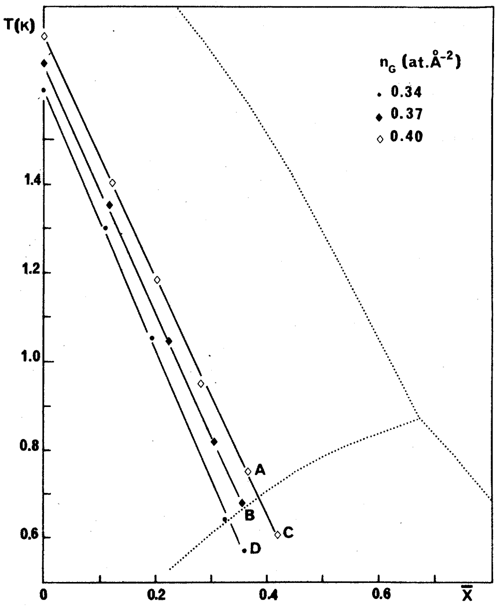

FIG. 2. Superfluid transition temperatures of thin film mixture for three different coverages. Experiments *A*, *B*, *C*, *D* refer to the text and Table II. The bulk phase diagram is represented in dotted line.

of some thermodynamic quantities and derivatives which implies profile calculations in the film. We delay such analysis for a future work. However it is already possible to get an approximate estimation of $\langle\rho_s\rangle$ by writing[25]

$$\frac{\langle\rho_s\rangle}{\langle\rho\rangle C_3^2}\frac{3\Theta_G}{d_G^3}=1 \ , \tag{20}$$

where C_3 is the measured third-sound velocity. For each coverage n_G, the mixture film thickness d_G is obtained from the measurement of the pure ^{4}He film thickness d_{G4} at the same coverage (see Sec. II C and Table I). The mixture film thickness d_G is defined by Eq. (11) which also can read

$$n_G=\int_0^{d_G}\frac{1}{v}dz_G \ , \tag{21}$$

where v is the atomic volume in the mixture at distance z_G from the glass substrate.

Using the fact that the atomic volumes are quantities weakly dependent on z compared to the concentration x, it can be shown from Eq. (21) that a good estimate of d_G is given by

$$d_G=d_{G4}v(\bar{x})/v_4 \ , \tag{22}$$

where $v(\bar{x})$ is the atomic volume of a mixture with concentration $\bar{x}$.[30] The values of d_G obtained by this way are reported Table II. From these values we get a rough estimate of $d_G\langle\rho_s\rangle/T_s$, at the transition temperature T_s, in our mixture film. We notice that the values (Table II) are gathered around $(3.53\pm0.63)\times10^{-9}$ g cm^{-2} K^{-1} for all our data. This value is not far from the universal value of the onset jump predicted by the theory,[6] and so confirms its validity for mixtures in an unexplored temperature range. The theoretical model of Berker and Nelson,[1] analyzing 2D mixture properties, predicts that a two-dimensional phase separation should occur for $\bar{x}>0.12$, resulting in values of the onset jump smaller than the universal one predicted by Nelson and Kosterlitz.[6] We do not observe significant deviation in our results in agreement with the direct and accurate determinations of the onset jump reported by Webster *et al.*[3]: they do not see any evidence for two-dimensional phase separation and diminution of the onset jump for concentrations beyond 0.12. They suggest that surface inhomogeneities possibly inhibit the phase separation. Relative to the question whether the thickness range explored in these experiments and ours is suited or not to observe 2D behavior, the Fig. 3 appears very instructive: we have plotted our experimental superfluid transition lines with both bulk mixtures[31,32] and theoretical 2D mixtures[1] diagrams. Clearly the slopes of the experimental transition lines are lower than the slope of the 2D diagram, but we observe that as the coverage is reduced the experimental slope increases towards the predicted 2D one. This would suggest that our films are still too thick to strictly behave as 2D films, or that the existing concentration and pressure gradients perpendicular to the films, not considered in the theory, are of crucial importance. Further experiments with thinner mixture films at low temperature are planned in order to clarify this point. Then it appears interesting to discuss, in the frame of a continuum model,[7] the possibility of a phase separation perpendicular to the surface at larger concentrations.

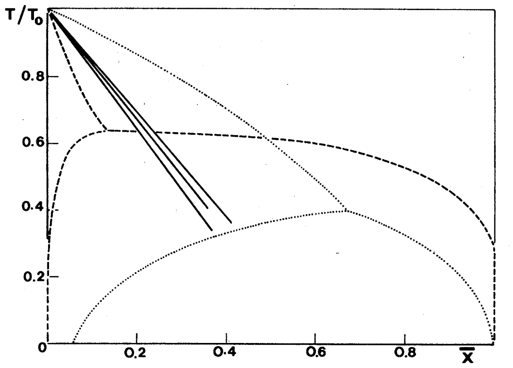

FIG. 3. Representations of the bulk phase diagram (dotted line), of the 2D phase diagram ($K=0$) from Ref. 1 (dashed line), and of our superfluid transition temperature determinations (full line). In each case the temperature is normalized to the transition temperature T_0 at $\bar{x}=0$.

It is obvious (see Fig. 2) that some of our points are inside the unstable domain of bulk mixtures. This means that in some of our films we could expect a phase separation. Yet we do not observe any experimental evidence related to the occurrence of phase separation: the general shape of our transition lines is regular (see Fig. 2) and do not present any accident. This is a puzzling question for which we cannot propose a satisfactory explanation. If we use the continuum model,[7] we know that the concentration varies spatially and that its value at the free surface of the adsorbed film is larger than the average concentration $\bar{x}$. So it is clear that, at least for experiments C and D (see Fig. 2), a region should exist near the free surface where bulk conditions for phase separation are satisfied. Of course, we know that in a thin mixture film, the growth of a new separated phase is governed by a supplementary condition, the conservation of the total number of atoms in the film. Taking it into account we can get an estimate of the ^{4}He-rich phase thickness. For instance in experiment C, at the superfluid onset temperature we find that the ^{4}He-rich phase thickness is only 0.8 of the initial film thickness. So the thickness and average concentration of the ^{4}He-rich phase are smaller than the initial ones. In such conditions the fact that the points C and D remain on the lines (Fig. 2)

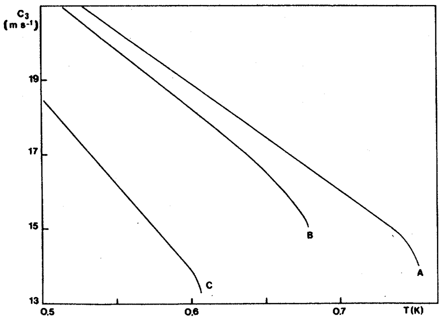

FIG. 4. Temperature dependence of the third-sound velocity below the superfluid transition temperature. Experiments *A*, *B*, and *C* refer to the text, Fig. 2 and Table II.

should appear as a fortuitous coincidence if we believe in the idea of phase separation in these films. Clearly a full description and analysis require the knowledge of the set of transition curves at lower temperature.

For three experiments (referred as *A*, *B*, *C*, in Fig. 2 and Table II), we have measured the third-sound velocity variations (see Fig. 4), from the superfluid onset temperature down to 0.5 K (Ref. 33) (the lowest temperature accessible with our present apparatus): here again we observe a regular variation without any detectable accident which could indicate that phase separation occurs. This last observation and our superfluid onset temperature measurements do not show any special feature which could prove without ambiguity the occurrence of phase separation in mixture films.

V. SUMMARY AND CONCLUSION

In this work we have presented the experimental results of third-sound velocity measurements in mixture films. First of all we show the importance of a good characterization of the film parameters. Using porous material to ensure a large surface adsorption area we get easily the useful parameters characterizing a film covering any substrate in the same experimental chamber. We then present the results of the superfluid onset temperature T_s in films for three different coverages (corresponding to estimated thicknesses from 4 to 5 atomic layers) and in a range of concentration $0 < \bar{x} < 40\%$. The experimental transition lines are regular with a reduction of T_s when the film thickness decreases or when the average concentration increases. A rough estimation of $d\langle\rho_s\rangle/T_s$ gives values that are consistent with the universal one obtained in the Kosterlitz-Thouless theory.[5,6] No significant effects on the transition lines and on third-sound velocity were observed which could prove without ambiguity the occurrence of phase separation in mixture films.

In conclusion new experiments are needed, and are planned in two main directions: first to extend the range of study to lower temperature in order to clarify the occurrence or not of phase separation; and secondly to study thinner films to bring the experiments closer to theoretical 2D mixture description.

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