Third-sound velocity measurements in layered mixture films

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We present third-sound velocity measurements in layered mixture films. The analysis of our data, with use of independent superfluid mass measurements by torsional-oscillator techniques, constitutes a direct check of the validity of the hydrodynamic relations in layered films, and provides some information on the interfacial region between the superfluid and normal phases.

I. INTRODUCTION

^{3}He-^{4}He mixture films constitute an experimental system of increasing interest as they afford the opportunity to study both the superfluid transition and phase separation. The superfluid transition in mixture films has been observed[1-3] and obeys the universal Kosterlitz-Thouless[4] prediction for superfluid mass jump. On the other hand, the phase separation in mixture films is a much more complex transition.[5,6] For mixture films that are very dilute[7,8] in ^{3}He ($d_3 < 0.2$ atomic layers), heat-capacity measurements[9] show evidence of lateral phase separation occurring at the surface of the film. In the opposite limit of mixture films which contain such a large amount of ^{3}He that superfluidity cannot exist if the film is in the homogeneous state, a layered phase separation has been observed.[2] The intermediate region, where both the homogeneous and the stratified film states can exhibit superfluidity, is expected to show a rich range of phenomena,[10,11] and has been extensively studied by third-sound velocity measurements. Ellis *et al.*[12] have found that for a ^{4}He coverage equal to 5.7 atomic layers, mixture films exhibit a nearly complete layered phase separation. The present authors[13] have observed a continuous crossover from the homogeneous state to the stratified one as the ^{4}He coverage was increased from 3 to 6 atomic layers. At this point it is important to notice that both these third-sound experiments were analyzed assuming that the superfluid and the normal phases were, respectively, pure ^{4}He and pure ^{3}He in the layered state. Now, in layered mixture films, direct measurements of the superfluid mass per unit area by torsional-oscillator techniques[2] show a strong temperature dependence for σ_s, which led McQueeney *et al.*[2] to the conclusion that the two phases in a layered film structure are not just pure ^{4}He and pure ^{3}He. Then the previous third-sound velocity analysis must be reconsidered. In order to clarify this situation, we present a new set of third-sound velocity measurements in mixture films. For large ^{3}He coverages, the analysis of these data using independent areal superfluid density measurements of McQueeney *et al.*[2] constitutes a direct check of the validity of the hydrodynamic relations in layered films, and provides some information on the interfacial region between the superfluid and normal phases.

II. EXPERIMENTAL RESULTS

Our experimental cell contains 2000 Nucleopore filters[14] whose typical pore size is 0.2 μm. The third-sound velocity is measured by the usual time-of-flight technique.[15] Two strips of Ag and Al, which are, respectively, the third-sound emitter and the receiver thermometer, are evaporated onto the surface of a virgin polycarbonate Nucleopore material membrane.

To scale the amounts of gas condensed in our cell to the coverages used by McQueeney *et al.*[2] to characterize their experiments, preliminary superfluid onset temperature determinations on pure ^{4}He films were made. Although it is not possible to follow a third-sound signal through the transition region,[16] it can be followed to the point where dissipation begins to rise rapidly. So the highest temperature for which a third sound is detected is slightly lower than the temperature for which the dissipation peak is observed: however, in our experiments, the difference between these two temperatures corresponds to a relative uncertainty of about 1% and can be neglected. For convenience in this paper, for each amount of gas condensed in our experimental cell, we give the equivalent total thickness of the film, obtained from the ^{4}He adsorption isotherm using $\Theta = 50$ K (layer)^3.[13]

We first study the superfluid onset temperatures T_0 of mixture films obtained by adding measured volumes of ^{3}He to a constant amount of ^{4}He. Our experimental measurements are presented in Fig. 1 for $d_4 = 2.6$ atomic layers (solid squares) and $d_4 = 3.0$ atomic layers (solid triangles). We see that, as d_3 increases, T_0 decreases[17] asymptotically towards a constant value. This behavior, also observed by Wang *et al.*[10] for a lower coverage of ^{4}He, means that for a given amount of ^{4}He, the onset superfluid temperature T_0 of a mixture film (i.e., from the Kosterlitz-Thouless theory, its areal superfluid density σ_s) remains nearly constant as more ^{3}He is added beyond a critical coverage. Although no systematic study has

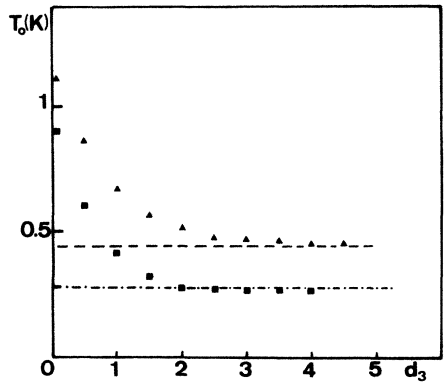

FIG. 1. Superfluidity onset temperatures T_0 of mixture films at various ^{3}He coverages (in atomic layers) for two constant amounts of ^{4}He. Solid triangles: $d_4=3$ atomic layers. Solid squares: $d_4=2.6$ atomic layers. As d_3 increases, T_0 decreases asymptotically towards a constant value which is identical, within experimental uncertainty, to the onset superfluid temperature measured by McQueeney *et al.* (Ref. 2) for the same amount of ^{4}He, but loaded with 12 atomic layers of ^{3}He (dashed line and dot-dashed line, respectively). This means that addition of ^{3}He beyond 4 atomic layers does not affect the areal superfluid density σ_s.

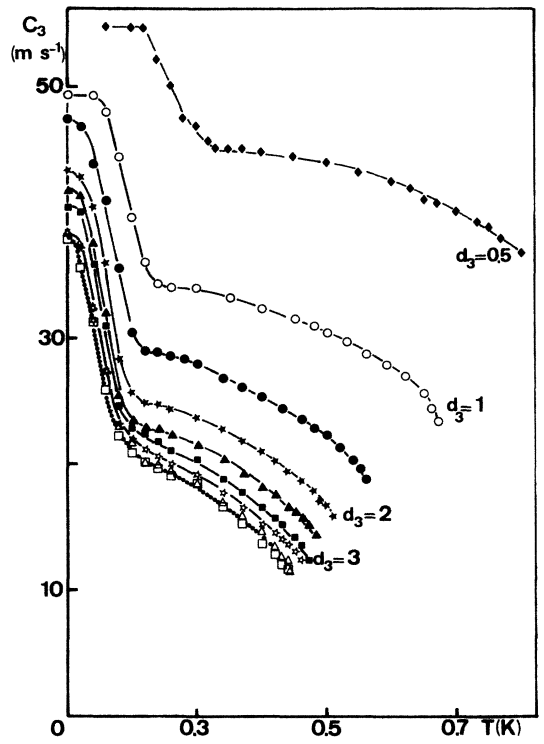

FIG. 2. Temperature dependence of the third-sound velocity in mixture films for a constant amount of ^{4}He ($d_4=3$ atomic layers). As d_3 is increased (by 0.5-atomic-layer steps), the data coalesce to a single curve. The third-sound velocity measurements for $d_3=4$ and $d_3=4.5$ are found identical within experimental uncertainty.

been carried out in the present work, it appears (Fig. 1) that this critical ^{3}He coverage becomes larger as the ^{4}He amount is increased.

For the constant amount of ^{4}He ($d_4=3.0$ atomic layers) which corresponds to the highest coverage studied by McQueeney *et al.*,[2] we have measured the temperature dependence of the third-sound velocity as d_3 was varied. The experimental curves $C_3(T)$ are presented in Fig. 2. As d_3 is increased, the experimental data coalesce to a single curve (dotted line in Fig. 2): the third-sound velocity measurements for $d_3=4$ atomic layers and $d_3=4.5$ atomic layers are found identical within experimental uncertainty. This means that for $d_4=3$ atomic layers, both σ_s and d_s (the superfluid phase thickness) remain constant as more ^{3}He is added beyond 4 atomic layers. Of course, a homogeneous state of the mixture film must be ruled out, and the mixture film configuration with nonuniform thickness, proposed by Wang *et al.*[10] to account for their observation of two superfluid transitions, cannot be invoked at present as these two transitions are only observed for d_3 lower than 1.8 atomic layers. So we consider the experimental asymptotic behaviors observed in Figs. 1 and 2 as the signature of the stratified state of the mixture films for d_3 larger than 4 atomic layers. At low temperature ($T=0.2$ K) we observe a rapid increase of the third-sound velocity. This point will be discussed in the next section.

It must be pointed out in Fig. 1 that T_0 decreases asymptotically towards constant values which are identical, within experimental uncertainty, to the superfluid onset temperatures measured by McQueeney *et al.*[2] for the same amounts of ^{4}He, but loaded by 12 atomic layers of ^{3}He (dashed line and dot-dashed line in Fig. 1). This weak ^{3}He coverage dependence of T_0 indicates that in a layered mixture film the properties of the superfluid phase are only weakly affected by the "evaporation" of the ^{4}He atoms in the normal phase, and leads to the conclusion that it is the "dissolution" of the ^{3}He atoms in the superfluid phase which essentially determines the value of its superfluid density σ_s. The good agreement between our asymptotic values of T_0 and the measurements of McQueeney *et al.*[2] gives us great confidence in their superfluid mass measurements to analyze our third-sound velocity measurements for $d_3>4$ atomic layers.

III. ANALYSIS OF THIRD-SOUND VELOCITY MEASUREMENTS

The third-sound velocity[18] value in a superfluid film is essentially governed by the areal superfluid density σ_s and the restoring force due to the van der Waals interaction with the substrate:

$$C_3^2 \propto \sigma_s / d_s^4 , \tag{1}$$

where d_s is the superfluid phase thickness.

But in a layered film, the superfluid phase is loaded by the normal phase of thickness d_n. To account for this effect, Ellis *et al.*[12] proposed a corrective factor which we note: $f(d_n/d_s)$.

Assuming that the superfluid phase is pure ^{4}He one gets

$$C_3^2 = C_{30}^2 f \ , \tag{2}$$

where C_{30} is the measured third-sound velocity in a pure ^{4}He film containing the same amount of ^{4}He as the mixture film considered. Obviously expression (2) cannot fit our velocity measurements for the mixture films $d_4 = 3$ atomic layers and $d_3 = 4$ atomic layers and 4.5 atomic layers: We get a constant, too-large value (31.9 m s^{-1}), presented as a solid horizontal line in Fig. 3.

Now, we know that the areal superfluid density of the superfluid phase is different from that of a pure ^{4}He film. Assuming that our mixture film displays the same restoring force as a pure ^{4}He film, we get

$$C_3^2 = C_{30}^2 f \sigma_s(T)/\sigma_4(T) \ . \tag{3}$$

The ratio $\sigma_s(T)/\sigma_4(T)$ is given by the experimental work of McQueeney *et al.*[2] Relation (3), represented as a solid curved line in Fig. 3, still gives values of the third-sound velocity that are too large, but accounts very well for the temperature dependence that we observe for $T > 0.2$ K.

At this point, if we want to keep the corrective factor f, the only way to obtain our measured values of the third-sound velocity above 0.2 K is to assume that the restoring force is weaker for the superfluid phase of the layered mixture film than for a pure ^{4}He film containing the same amount of ^{4}He:

$$C_3^2 = C_{30}^2 f \sigma_s(T)/\sigma_4(T)(d_s/d_4)^4 \ , \tag{4}$$

where d_s governs the restoring force for the superfluid phase of the mixture film. Using $d_s = 3.4$ atomic layers (while $d_4 = 3$ atomic layers), from relation (4) we get a good fit of our experimental data for $T > 0.2$ K (dashed line in Fig. 3). This value of d_s, larger than d_4, does not seem, however, unreasonable as we qualitatively expect that the ^{3}He atoms, which belong to the superfluid phase and weaken its superfluid density, also increase its thickness. The important point we want to stress again is that we have analyzed third-sound velocity measurements in mixture films, using independent determinations[2] of their superfluid density σ_s. Between 0.2 K and the superfluid onset temperature T_0, while σ_s is typically varying by a factor of 3, we find that the ratio C_3^2/σ_s is temperature independent, which means that the intrinsic restoring force for third-sound propagation is temperature independent above 0.2 K.

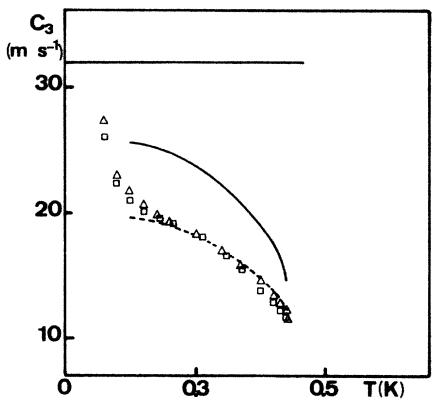

FIG. 3. Measured third-sound velocity in mixture films ($d_4 = 3$ atomic layers) for $d_3 = 4$ (open triangles) and $d_3 = 4.5$ (open squares). Assuming that the superfluid phase is pure ^{4}He, relation (2) gives a constant, too-large value (solid horizontal line). Using independent superfluid mass measurements (Ref. 2), we obtain from relation (3) the solid curved line which still gives too-large values of C_3, but accounts very well for the temperature dependence we observe for $T > 0.2$ K. To obtain a good fit of our measured values above 0.2 K (dashed line), we must use a weaker restoring force.

Below $T = 0.2$ K we observe a sudden rise of the third-sound velocity (Fig. 2). This rapid variation of C_3 is quite reminiscent of the rapid variation of surface tension and surface sound velocity observed by Eckard *et al.*[19] in very dilute bulk ^{3}He-^{4}He mixtures. The latter, however, are characterized by an exponential T dependence, since it is basically a "single-^{3}He" effect. Here, on the other hand, for "macroscopic" ^{3}He coverages, we expect ^{3}He evaporation to be Fermi-like as for bulk: AT^2 (though with a d_4-independent prefactor A, see next section). This, as we have seen, correctly describes the situation above 0.2 K (dashed line in Fig. 3).

The rapid variation of C_3 at about 0.2 K (a characteristic temperature, which like T_0 is largely d_3 independent for $d_3 > 3$ atomic layers) seems to require a separate explanation. We observe a sudden increase of the third-sound velocity while the measured areal superfluid density σ_s remains nearly constant:[2] this spectacular effect would suggest an increase of the restoring force occurring at that temperature. A quantitative evaluation of the restoring force in a layered mixture film is out of the scope of the present paper. A few remarks, however, could be made. It is not merely coincidental that the jumps we observe in C_3 occur at 0.2 K, which is the temperature at which evaporation of ^{4}He in ^{3}He starts (Fig. 3 of Ref. 2). Without going into detail, we want to suggest here that the onset of ^{4}He evaporation, while too modest an effect to appreciably alter σ_s, might cooperatively destabilize the interfacial zone, increasing its fuzziness so to speak, and leading to a weakening of the restoring force, and therefore, of the third-sound velocity itself as the temperature is increased. This effect is expected to be more spectacular the thinner d_4, in good agreement with experimental observations.[20]

Therefore, above 0.2 K, the transfers of He atoms are free and the restoring force must be evaluated at fixed chemical potential. Below 0.2 K these transfers are blocked and the third sound propagates at fixed concentration. In both regimes C_3^2/σ_s should indeed be temperature independent. We plan to come back to this point later on, but obviously it would be interesting to look for a possible frequency dependence of the crossover temperature.

IV. SUPERFLUID PHASE OF A LAYERED MIXTURE FILM

The question arises now of how we should describe the ^{3}He atoms present in the superfluid phase. McQueeney

et al.[2] give a good account for their observed temperature dependence of σ_s by a solubility model using the functional forms of both the concentrated and dilute sides of the bulk phase separation diagram. Two mechanisms weaken σ_s as the temperature is increased. The first is the "evaporation" of the ^{4}He into the normal phase, accounted for by the term $BT^{3/2}e^{-C/T}$, where B is a constant proportional, as expected, to the amount of ^{3}He.[21] The second mechanism is the dissolution of ^{3}He in the superfluid phase. It provides the largest contribution AT^2, where A is a constant found to be independent of d_4 for their set of experiments.[2] This is a surprising result, consistent with an *interface effect* rather than with a dissolution mechanism in the "bulk" of the superfluid phase. The existence of an interface effect is suggested by two other experimental observations. The first one is the analysis of σ_s at zero temperature:[2] the "proximity" of the normal phase decreases the areal superfluid density by a constant value [corresponding to about 0.5 atomic layers[22]] independently of the ^{4}He thickness (Fig. 4 of Ref. 2). The second one is the good agreement observed between third-sound velocity measurements in mixture films for $d_4=6$ atomic layers,[12,13] and the calculated values from relation (2), assuming that the superfluid phase is pure ^{4}He. This agreement is hardly explainable in term of an "equilibrium bulk concentration," as the important discrepancy observed for $d_4=3$ atomic layers (see Fig. 3) should persist independently of the superfluid phase thickness. An interface effect is expected to become relatively less important as the superfluid phase thickness increases. This is supported by the continuous evolution of C_3 towards the curve calculated from relation (2) as d_4 increases (Fig. 2 of Ref. 13).

So we suggest that ^{3}He "evaporates" in ^{4}He according to a Fermi-like law AT^2, but stays close to the interface. This we understand on the basis of Refs. 23 and 24, which provide a variational theory for the binding of a single ^{3}He atom to a ^{4}He film. The spectrum of excited states is a discrete set of two-dimensional (2D) states whose number and energy depend on the thicknesses of the ^{4}He film. It is only for thick ^{4}He films ($d_4>50$ Å) that the "bulk" states energies merge into the true bulk state energy (-2.8 K) which corresponds to the ordinary 3D phase equilibrium with ^{3}He concentration 0.063.

These single-particle theories[23,24] have served as a model for the analysis of specific-heat measurements of Bhattacharyya *et al.*[25] for ^{3}He coverages less than one monolayer. They found that for ^{4}He thicknesses up to 25 Å, a single excited state within the film, localized relatively close to the interface, satisfactorily describes their data.

The analysis of both the results of McQueeney *et al.*[2] and our data leads us to the conclusion that this description of the "bulk" ^{3}He excited states should remain valid for thicker ^{3}He coverages. In these stratified structures, the superfluid phase thickness is still the important parameter as it determines the 2D character of the ^{3}He excited states, which, due to the Van der Waals field, are localized close to the interface.

V. CONCLUSION

We have presented systematic measurements of third-sound velocity C_3 in mixture films, for a given amount of ^{4}He ($d_4=3$ atomic layers). From the d_3 dependence of both C_3 and superfluidity onset temperature T_0, it appears that for $d_3=4$ atomic layers the state of the mixture film is one of a layered phase separation. Moreover, addition of ^{3}He beyond $d_3=4$ atomic layers does not affect the properties of the superfluid phase. The temperature dependence of C_3 is in good agreement with independent superfluid mass measurements by torsional-oscillator techniques, supporting the idea that the superfluid phase is not pure ^{4}He. However, we suggest that addition of ^{3}He affects the underlying ^{4}He film essentially via an interfacial effect rather than in the "bulk." The analysis of third-sound velocity values using independent superfluid mass measurements provides information on the restoring force acting on the superfluid phase surface. We identify a characteristic temperature (0.2 K), largely independent of d_3, above which we find that the restoring force is weaker than expected. This could be lated to a fuzziness of the interface.[26] We suggest that the interface gets sharper below 0.2 K, this "superstratification" instability resulting in an increase of the third-sound velocity. A NMR experiment, measuring both susceptibility and relaxation times,[27,28] although clearly delicate in this two-dimensional context, would be very helpful in assessing the whole picture in more microscopic detail. Finally, let us note that our picture of the structural instability at 0.2 K, triggered by the onset of ^{4}He evaporation into ^{3}He, might prove useful for nonquantum thin films as well.

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