Surface Superfluidity in Dilute ^{4}He-^{3}He Mixtures

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Quantitative measurements have been made of the superfluid mass per unit area for the interfacial superfluid phase present at the boundary wall of low-concentration mixtures of ^{4}He in liquid ^{3}He. We find a rapid jump in the superfluid mass at the transition temperature in agreement with the prediction of the Kosterlitz-Thouless theory of the superfluid transition in a two-dimensional system.

Since the successful application of the Kosterlitz-Thouless[1,2] (KT) theory of vortex pair unbinding to the understanding of the superfluid transition in thin ^{4}He films, a wide search has been made for other systems in which these ideas might be applicable. Extension of the theoretical ideas by Halperin and Nelson[3] and Young[4] to the case of two-dimensional (2D) melting has very much increased the variety of possible systems in which one might look for a realization of a vortex (dislocation) pair unbinding transition. In this Letter we report the study of a 2D superfluid system of a type quite different in structure from the usual adsorbed film of ^{4}He. In the present work, we have studied the properties of a "superfluid sandwich" formed at the container boundaries in a dilute ^{4}He-^{3}He mixture.

It has been known since the early 1960's that at sufficiently low temperatures dilute mixtures of ^{4}He in liquid ^{3}He display an interesting state of "surface superfluidity." Since its discovery by Brewer and Keyston,[5] this phenomenon has been investigated extensively by Laheurte and co-workers[6-8] and more recently by Ellis *et al.*[9] From such investigations it is clear that ^{4}He atoms are preferentially adsorbed on the walls of the container as a result of the relatively smaller molar volume of ^{4}He as compared to that of ^{3}He and the strong van der Waals force induced by the wall. For dilute concentrations of ^{4}He in bulk ^{3}He, the ^{4}He atoms are almost exclusively confined at low temperatures to the interfacial region at the boundaries of the system. The surface superfluid phase which occurs at the interfacial region of enhanced ^{4}He concentration is a natural candidate for 2D phase transition studies. We present here the first quantitative measurements of the superfluid properties of this system over a broad range of temperatures and concentrations.

For experimental convenience, we have chosen to work with thick films of ^{3}He with a low concentration of ^{4}He. At high temperatures a certain amount of the ^{4}He is dissolved in the ^{3}He overlayer somewhat depleting the ^{4}He-rich layer near the

wall. It is desirable to work with a ^{3}He layer sufficiently thick so as to have essentially bulk properties and yet sufficiently small in total volume so that the superlfuid transition is not dominated by solution effects.

Torsional oscillator techniques similar to those employed by Bishop and Reppy[10] and Agnolet, Teitel, and Reppy[11] in studies of the Kosterlitz-Thouless transition in pure ^{4}He films are used to measure the superfluid response. The helium film is adsorbed on a Mylar substrate which constitutes the moment of inertia of the torsional oscillator. The geometric surface area of the Mylar is 1.95 m^2 and the total open volume accessible to helium at low temperatures is 1.6 cm^3. Under conditions of constant temperature, the period of oscillation is stable to 1 part in 10^9 which corresponds to a resolution of 10^{-4} atomic layer of ^{4}He. A ^{3}He-melting-curve thermometer of the type described by Greywall and Busch[12] was used as the reference thermometer.

Preliminary to the mixture measurements, a series of pure ^{4}He films was examined. The lowest coverage (29 μmol/m^2) exhibited a transition temperature, T_c, below 100 mK. Changes in the period, $\Delta P(T)$, and amplitude of the oscillator measure changes in the superfluid mass and dissipation of the film, respectively. Data taken as a function of temperature for a sequence of coverages are shown in Fig. 1. The transition region for each coverage is indicated by a rapid change in period and an associated peak in the superfluid dissipation. It is important to note that although the superfluid response of these films corresponds to much less than one completed monolayer, the general character of the transition is similar to that of thicker films.[10,11] The solid line drawn through the period data indicates the predicted value of $\Delta P(T_c)$ obtained from the KT theory and the calibration constants of our cell. Since the KT line passes through the pronounced corners in the $\Delta P(T)$ curves, we conclude that the KT theory remains valid over this
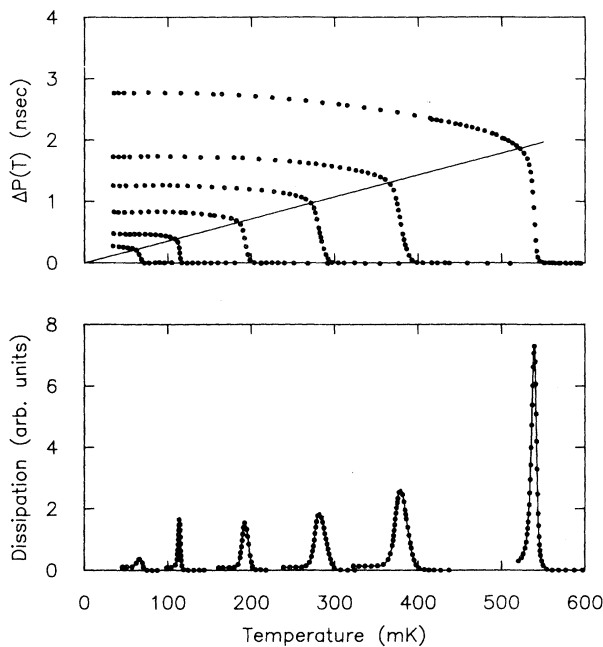
FIG. 1. Period shift $[\Delta P(T)]$ and dissipation of the torsional oscillator for a series of pure ^{4}He coverages with T_c ranging from 100 mK to 1 K. The solid line on the $\Delta P(T)$ graph is the Kosterlitz-Thouless mass-jump prediction for our cell. The unsystematic variation in the dissipation peak heights and widths will be discussed elsewhere (Ref. 13).

FIG. 2. Period shift and dissipation for a series of mixtures with ~ 12 layers of ^{3}He. The solid straight line on the $\Delta P(T)$ vs T plot is the KT prediction for our cell. The dashed curve is a fit to the mixture data based on a phase-separation model.

range of coverages and transition temperatures. An analysis of the data in terms of the dynamic theory[14] has proven to be difficult and will be discussed elsewhere.[13]

After the pure ^{4}He data were obtained, the ^{4}He was removed and 136 μmol/m^2 of ^{3}He were introduced to the cell. Measured amounts of ^{4}He were then added until a superfluid response was seen at the lowest temperatures. We find that 5.9 μmol/m^2 of additional ^{4}He are required over what was sufficient in the pure films for the first indications of superfluidity to be seen. A series of mixture films with increasing amounts of ^{4}He and a fixed coverage of 136 μmol/m^2 of ^{3}He is shown in Fig. 2 along with the predicted KT jump (indicated by the straight solid line).

All of the mixture films display the KT signature of a jump in the oscillator period and an associated dissipation peak. The superfluid mass jump at the transition is less conspicuous than in the pure films since it is superimposed on a large temperature-dependent background. This evidence demonstrates that the interfacial layer behaves as a 2D Kosterlitz-Thouless superfluid. It should be noted that the mixture dissipation data show a larger value
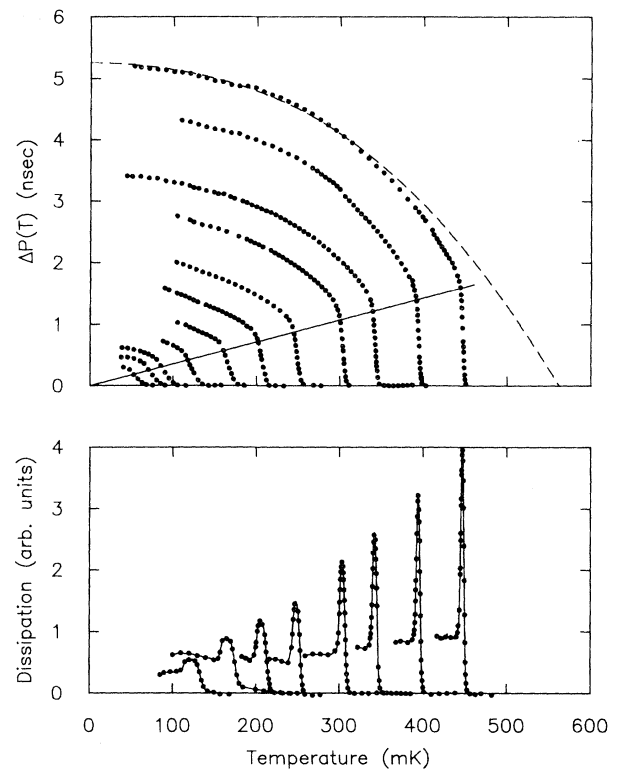
below the transition region. The origin of this additional dissipation is not understood at present.

The strong temperature dependence seen in the $\Delta P(T)$ data of Fig. 2 at temperatures below the KT transition may be due to several mechanisms. Most prominent will be effects which are analogous to those seen in the case of bulk ^{3}He-^{4}He phase separation.[6-8] As the temperature is increased from the lowest values, ^{3}He is expected to dissolve in the ^{4}He-rich layer giving a reduction in the superfluid signal proportional to T^2. At higher temperatures "evaporation" of ^{4}He into the overlayer of ^{3}He following an exponential law is expected. Following these ideas,[6-8] we find that a function of the form

$$\Delta P(T) = \Delta P(0)(1 - AT^2 - BT^{3/2}e^{-C/T}) \qquad (1)$$

gives a reasonable fit to our mixture data for temperatures sufficiently below the KT transition. An example of such a fit to mixture data is shown in Fig. 2 for the data set (coverage 41) with the highest transition temperature.

The size of the exponential term in Eq. (1) can be estimated by comparing the superfluid response for two films containing the same amount of ^{4}He but with different coverages of ^{3}He. After measuring $\Delta P_{41}(T)$ for film coverage 41 (the highest T_c mixture film shown in Fig. 2) we increased the ^{3}He coverage from 136 to 264 μmol/m^2 while holding the ^{4}He coverage fixed at 42.5 μmol/m^2. The addition of ^{3}He reduces the T_c of the film; however, for temperatures below 150 mK where "evaporation" effects are minimal both films have the same superfluid signal. The difference between the two signals, $\Delta P_{41}(T) - \Delta P_{42}(T)$, is plotted in Fig. 3 as a function of temperature. We have fitted the data with the function

$$\Delta P_{41}(T) - \Delta P_{42}(T) = \Delta P(0) T^{3/2} B' e^{-C/T}. \quad (2)$$

A nonlinear least-squares method was used to obtain the constants $\Delta P(0)$, B', and C. The quantity B used in the fit shown in Fig. 2 to coverage 41 is taken to be $(136 \ \mu\text{mol/m}^2)/(264 - 136 \ \mu\text{mol/m}^2)$ times B'.

We have proceeded to fit the remaining mixture data sets with the temperature-dependent function given by Eq. (1). Satisfactory fits are obtained with values of $A = 2.08 \pm 0.2$ K^{-2}, $B = 2.31 \pm 0.04$ K$^{-3/2}$, and $C = 0.92 \pm 0.08$ K, allowing only $\Delta P(0)$ to change with ^{4}He coverage.

Although this solution model describes the basic temperature dependence of the mixture films, we do not wish to rule out the possibility that there are excitations present that are unique to the mixture system. Our estimates for the normal-fluid contribution from surface excitations, i.e., capillary waves and third-sound modes, are an order of magnitude smaller than the AT^2 effect observed. Therefore, such a contribution would be difficult to extract from our present data.

In Fig. 4 we have plotted the zero-temperature signal, $\Delta P(0)$, obtained from our fits mentioned above as a function of total ^{4}He coverage. We also show the estimated values for $\Delta P(0)$ obtained by extrapolation of the pure ^{4}He data.

Our result for the pure ^{4}He films (open circles) is similar to that obtained previously by other experimenters. Initially, a large amount of ^{4}He must first be adsorbed before superfluidity can be observed. Once this critical amount (28.7 μmol/m^2 in the present experiment) is reached, the magnitude of the zero-temperature superfluid signal increases linearly with additional ^{4}He. The interesting result is that an essentially identical behavior is observed for the interfacial superfluid. The only effect of the overburden of 12 layers of ^{3}He is to require an additional 5.9 μmol/m^2 (approximately $\frac{1}{2}$ layer) of ^{4}He before superfluidity can occur. Subsequent additions of ^{4}He produce the same increase in the superfluid mass as in the pure films.
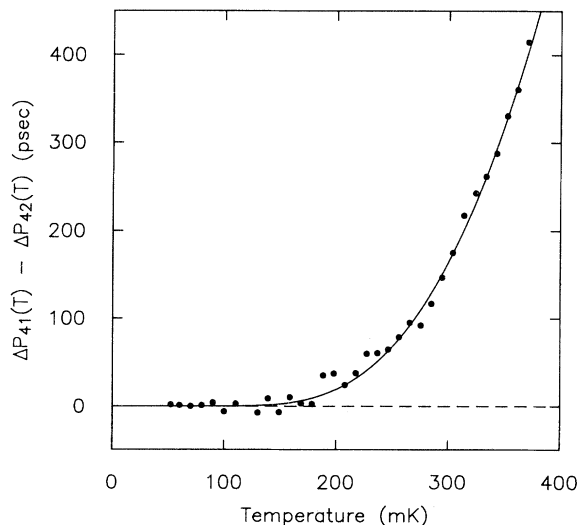
The slope of the $\Delta P(0)$ vs ^{4}He coverage lines

FIG. 3. The period difference between two films, coverage 41 and 42, where the ^{4}He content has been held constant and the ^{3}He coverage has been increased from 136 μmol/m^2 for coverage 41 to 264 μmol/m^2 for coverage 42. The curve through the data is a fit of the form $\Delta P(0) B' T^{3/2} \exp(-0.92/T)$.
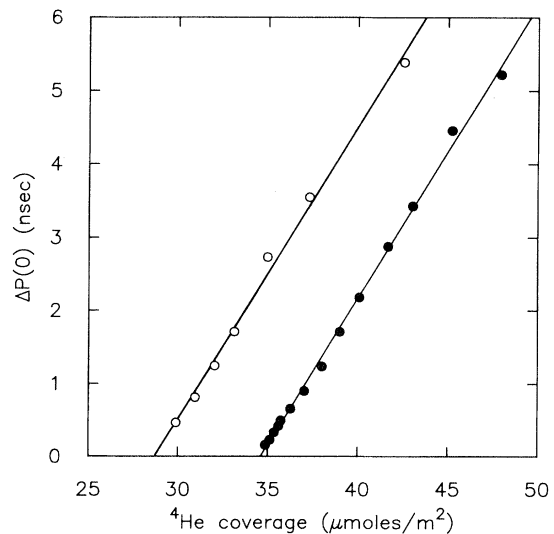

FIG. 4. Period shift extrapolated to $T = 0$ for pure ^{4}He (open circles) and mixtures (closed circles) plotted against ^{4}He coverage. The straight lines are fits to the data, and the slopes are identical within the uncertainty of the fits.

plotted in Fig. 4 measures the fraction of ^{4}He that becomes superfluid at $T = 0$. Since these lines are parallel, it is evident that adding ^{3}He to the system does not disturb the zero-temperature properties of the superfluid layer except near the ^{3}He interface. In particular, if the ^{3}He concentration in the superfluid film were greater than $\sim 5\%$, we would be able to resolve the difference in slope due to the effective mass of the normal ^{3}He in the ^{4}He layer.

In conclusion, we have demonstrated that a wide range of pure and ^{4}He-^{3}He mixture films obey the Kosterlitz-Thouless prediction for superfluid mass jump, providing a significant extension of the range of conditions under which a ^{4}He film behaves as a 2D Kosterlitz-Thouless superfluid. The addition of ^{3}He to the system depresses the KT transition temperature, and alters the superfluid signal below T_c.

In the mixture films, the extra temperature dependence can be accounted for by a solution model. The functional forms of both the concentrated and dilute sides of the phase separation diagram are needed to describe the behavior of the system. This provides a contrast to third-sound measurements made by Ellis et al.[9] where the data were modeled by a layered phase separation, with the ^{4}He and ^{3}He layers essentially pure. Our measurements support the picture of Laheurte and co-workers[6-8] where solution effects are crucial in determining the properties of the superfluid component in the ^{4}He-^{3}He mixture films.

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