Study of the Superfluid Transition in Two-Dimensional ⁴He Films

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We have studied the superfluid transition of a thin two-dimensional helium film adsorbed on an oscillating substrate. The superfluid mass and dissipation when analyzed in terms of the dynamic theory of Ambegaokar, Halperin, Nelson, and Siggia support the Kosterlitz-Thouless picture of the phase transition in a two-dimensional superfluid. The value for the jump in the superfluid density at the transition given by Kosterlitz and Thouless, $\rho_s(T_c^-) = 8\pi k_{\rm B} (m/h)^2 T_c$, is in good agreement with estimates from experiment.

In this Letter we report a study of the superfluid transition in two-dimensional ⁴He films using the Andronikashvili torsional-oscillator technique.¹ We have made precise measurements of both the superfluid mass and dissipation near the transition temperature for films with transitions ranging from 0.3 to 1.6 K. Preliminary reports on this work have been given earlier.² Our observations correspond closely to the predictions of the Kosterlitz-Thouless^{3,4} theory of two-dimensional superfluids and its recent extension to finite frequency and velocity.^{5,6}

In the Kosterlitz-Thouless theory,^{3,4} the twodimensional superfluid at low temperatures is populated by a system of bound vortex-antivortex pairs. At sufficiently high temperature a dissociation of a finite fraction of the vortex pairs takes place, and superfluidity is destroyed. Kosterlitz,⁴ in a renormalization-group calculation, has shown that, for the static case, the superfluid mass drops discontinuously to zero at the transition. Further, as has been emphasized by Nelson and Kosterlitz,⁷ the ratio of the superfluid mass per unit area, to the transition temperature, is a universal quantity, independent of film thickness, given by $\rho_s(T_c^{-})/T_c = 8\pi k_B (m/h)^2$, where m is the mass of the ⁴He atom, h is Planck's constant, and $k_{\rm B}$ is Boltzmann's constant.

Present techniques for the determination of the superfluid mass in thin helium films do not test the static theory directly, since they all require measurements at a nonzero frequency and superfluid velocity. As a result, one does not expect to see the discontinuous jump given in the static theory, but to find a continuous variation of the superfluid mass with temperature at the transition. In addition, one expects to find considerable dissipation associated with the vortex motion induced by the superflow required for the superfluid mass measurements. The experimental method employed in the present investigation is similar to that used in the previous determinations of the superfluid density for films adsorbed on porous Vycor glass.¹ In the experiment reported here, helium films are adsorbed on a Mylar⁸ film substrate. A strip of the plastic Mylar film, 6×10^{-4} cm thick, 1.0 cm wide, and about 21 m long, is wound as a spiral on the axis of a torsional oscillator. The oscillator is driven at its resonant frequency of about 2.5 kHz. After a helium film of the desired thickness has been adsorbed on the Mylar substrate, a series of period and Q measurements are made to determine the temperature dependence of the superfluid mass and dissipation.

When the oscillator is cooled below the transition temperature the superfluid decouples from the torsion pendulum and the period of the oscillator decreases. In Fig. 1, we have plotted the shift in period, ΔP , due to the formation of superfluid for a film with a transition temperature of near 1.22 K. It is seen that the variation in the superfluid signal is rather abrupt near the transition, but nevertheless is continuous when examined with sufficiently high temperature resolution.

In Fig. 1 we have also plotted, as open circles, the excess dissipation Q^{-1} , due to the superfluid. A pronounced narrow peak is seen in the superfluid dissipation in the region of the transition and the dissipation remains at a nonzero level as the temperature is lowered well below the transition temperature. The width of the transition region where the period changes rapidly and the peak in dissipation occurs is less than 1% of the transition temperature.

These features in the dissipation are unique to the two-dimensional superfluid. Neither the Andronikashvili experiments in bulk helium nor the experiments performed for films adsorbed on the three-dimensionally connected substrate,



FIG. 1. The shift in period, ΔP , and dissipation Q^{-1} are shown as a function of temperature at the super-fluid transition.

porous Vycor glass,¹ exhibit any excess dissipation associated with the superfluid transition. The peak in dissipation in the present experiment points to a fundamental difference between onset phenomena in two- and three-dimensional superfluids.

The behavior in the two-dimensional fluid as seen in our experiment can be understood in terms of the dynamic theory of Ambegaokar, Halperin, Nelson, and Siggia (AHNS).⁶ In their theory, as well as in the calculation of Huberman, Myerson, and Doniach,⁵ the dissipation is associated with the diffusive motion of two-dimensional vortices driven by the oscillating superflow.

The dynamic theory given by AHNS (Ref. 6) is directly applicable to the data in the high-frequency regime of the present experiment.

In these experiments we have varied the oscillator amplitude by a factor greater than 100. At low amplitudes, where the superfluid velocity is less than 10^{-3} cm/sec, we find that the period and Q^{-1} are amplitude independent, while at larger velocities nonlinear effects set in, the transition region and dissipation peak are broadened.

In Fig. 2, we display, on an expanded temperature scale, a set of low-amplitude data obtained in the neighborhood of the transition. The solid curves drawn through the data represent a fit⁹ of the AHNS theory to these data. The gross features of the curves are controlled by the choice for the transition temperature, T_c , and the value for the jump in the superfluid mass per unit area,



FIG. 2. The reduced period shift, $2\Delta P/P$, and dissipation Q^{-1} are shown for a superfluid transition temperature of 1.215 K. The solid lines are fits using the dynamic theory of AHNS (Ref. 6) and the dashed curve is the result of the static theory.

 $\rho_s(T_c^{-})$, at T_c . These quantities appear in the expression for the superfluid density near the transition given by Kosterlitz and Thouless for the static film:

$$\rho_s(T) = \rho_s(T_c^{-}) [1 + b(1 - T/T_c)^{1/2}].$$
(1)

The quantity, b, in Eq. (1) determines the strength of the square-root cusp in the static theory. The curves marked dynamic theory in Fig. 2 are based on the linear-response calculation described in Ref. 6. In brief, the reduced shift in period, $2\Delta P/P$, and the superfluid dissipation Q^{-1} are related to a frequency-dependent dielectric constant ϵ by

$$2\Delta P/P = (A/M)\rho_s(T_c^{-1}) \operatorname{Re}(\epsilon^{-1})$$

and

$$Q^{-1} = (A/M)\rho_s(T_c^{-1}) \operatorname{Im}(-\epsilon^{-1}).$$

The real part of ϵ is taken as due to bound pairs according to Eq. (9a) of AHNS. It is calculated by a numerical integration of the Kosterlitz recursion relations.⁴ For the imaginary part of ϵ contributions due to bound pairs, free vortices, and a constant background (to account for the dissipation remaining well below the transition) are added together. In addition to the three param-

eters of the static theory, three new parameters appear in this fit of the dynamical theory: a dimensionless parameter $\ln(2D/\omega a^2)$ related to the vortex diffusion constant D, the vortex-core radius a, and the frequency ω of the oscillator; the coefficient of the free vortex dissipation; and a background term, referred to above, which plays almost no role in the transition region itself. In the ratio A/M, A is the area of the Mylar substrate and M is the effective mass of the pendulum bob when the pendulum is treated as a linear oscillator. This ratio is obtained from a knowledge of the area of the substrate and a measurement of the sensitivity of the oscillator period to changes in the mass per unit area of the adsorbed helium.

In the analysis of our data taken for different coverages of adsorbed helium, we allow the value of the Kosterlitz-Thouless jump in ρ_s to be a free parameter to be determined by an optimization of the fit through a nonlinear least-squares routine.¹⁰ As an example the value for $\rho_s(T_c^-)$ obtained for the fit shown in Fig. 2 is 0.96 times the exact theoretical value, $8\pi k_{\rm B}(m/h)^2 T_c$, given by Kosterlitz and Thouless.

For the calibration measurement, we hold the temperature of the system constant and observe the period of the oscillator as the mass per unit area of adsorbed helium is increased. When the coverage of helium is less than a critical amount, which depends on the temperature at which the observations are made, the adsorbed helium is entirely locked to the substrate and contributes its entire moment of inertia to the pendulum bob. This serves to calibrate the torsion oscillator. The surface area is taken to be the geometric area of the Mylar film.

Since 1968 there has been evidence that the superfluid density in two-dimensional helium films might be nonzero at the superfluid onset. This was shown most clearly in the third-sound experiments of Rudnick *et al.*¹¹ and also by persistent-current measurements on helium films,¹² which indicated that the superfluid critical velocity was becoming zero while the superfluid density was still finite. However, neither thirdsound nor persistent-current measurements can be used to pursue the question of the superfluid density behavior in the transition region, since the third-sound signals become heavily damped and do not propagate, and persistent currents decay away.

The first experiments to show that the dynamic superfluid density goes continuously to zero were

the quartz-microbalance experiments of Chester and Yang.¹³ These experiments, which are very similar in concept to the present work, were performed in the MHz frequency range and showed, as would now be expected on the basis of the dynamic theory,⁶ considerable broadening of the transition region.

These older experiments can now be analyzed in terms of our present understanding to obtain estimates of the Kosterlitz-Thouless jump in the superfluid mass per unit area at the two-dimensional phase transition. 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. If the third-sound signal disappears at this point, then as can be seen in Fig. 2 the value of the superfluid mass is still up on the shoulder of the curve and a reasonably good estimate for the static value of the Kosterlitz-Thouless jump can be obtained. Recently, Rudnick¹⁴ has reanalyzed his third-sound data using his latest estimate of the van der Waals constant, and has obtained values for $\rho_s(T_c^{-})$ which are in good agreement with the value predicted by the Kosterlitz-Thouless theory.^{3, 4, 6}

In Fig. 3 we have plotted as a function of the transition temperature, the values for the static jump in the superfluid density obtained from the analysis of our data using the dynamic theory. We have also included in Fig. 3 the estimates obtained by Rudnick¹⁴ and additional values provided by Mochel and Hallock from their third-sound



FIG. 3. Results of all of our data, in addition to previous third-sound results for the discontinuous superfluid density jump $\rho_s(T_c^-)$ as a function of temperature. The solid line is the Kosterlitz-Thouless (Refs. 3 and 4) static theory.

work.¹⁵ The solid line drawn in Fig. 3 is the theoretical prediction given by Kosterlitz and Thouless. It is clear that the data from all the different experiments are in good general agreement with the theoretical prediction and therefore provide strong support for the Kosterlitz-Thouless picture of the phase transition in the two-dimensional superfluid.

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Metallic Glasses of Oxidized Rubidium and Cesium

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Metallic glasses of the alkali metals Rb and Cs containing 13-20 at.% oxygen have been obtained for the first time by rapid cooling (~ 10^2 K/s) of liquid samples. An investigation of the electronic and structural properties leads to the conclusion that a chemical bonding model is more appropriate for these glasses than a free-electron model. Strongly bonded ionic clusters are responsible for the enhanced tendency for glass formation.

A large number of metallic glasses have recently been obtained by quenching of the liquid state.¹ The enhanced glass formation has been explained with three different models on the basis of (i) packing of hard spheres,² (ii) chemical bonding effects,³ and (iii) the influence of conduction electrons.⁴ The general validity of models (i) and (ii) has been questioned,^{3,5,6} but so far, there seems to be no experimental evidence which rules out model (iii) in any special system. The nonexistence of alkali metal glasses has been accepted as an additional proof for the validity of model (iii).⁷ It should be noted that the proposed models are not contradictory. The range of applicability of each model can be explored only by investigating new metallic glass systems.

We report on the first metallic glasses of alkali metals stabilized by oxygen. Rubidium and cesium can be obtained in the amorphous state by addition of ~16 at.% and 14-20 at.% oxygen, re-