

## Superfluid Transition in $^4\text{He}$ Films on Hydrogen and Its Effect on the Film-Vapor Coupling

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(Received 12 December 1991; revised manuscript received 24 February 1992)

We have measured the 2D  $^4\text{He}$  superfluid density  $\sigma_s$  on hydrogen using a high- $Q$  Al oscillator. We find that only a  $\frac{1}{2}$  layer of helium remains normal well below the transition on hydrogen as opposed to 4 layers on bare Al. For transition temperatures  $T_c$  below 0.9 K we find that  $\sigma_s(T_c) \propto T_c$  as predicted by Kosterlitz-Thouless theory. For transitions above 1 K, where there is significant vapor damping, we see a striking suppression of  $\sigma_s(T_c)$  and find clear evidence for the appearance of excess gas-atom slip in the superfluid phase.

PACS numbers: 67.40.Rp, 67.70.+n

Recent investigations of the interaction of  $^4\text{He}$  with weak binding substrates [1] (i.e., alkali metals, noble gases, solid  $\text{H}_2$ ) have led to the discovery of nonwetting of cesium [2,3] and the probable existence of prewetting phases [4,5]. Naturally, this has spurred widespread interest in the nature of thin-film superfluidity on inert surfaces. Recent measurements of superfluid density on solid  $\text{H}_2$  using third sound have been particularly compelling, with reports of submonolayer superfluidity [6], coverage-induced third-sound velocity modulations [6,7], and the appearance of a possible new superfluid phase in submonolayer films [8]. To this date, however, there have been no mechanical oscillator measurements of superfluidity on weak binding substrates. The high- $Q$  torsional oscillator technique, in particular, provides an extremely sensitive and direct measure of the superfluid density through the transition. In this Letter we present the first torsional oscillator study of the 2D superfluid transition on crystalline hydrogen and find strikingly different behavior than has been previously observed on stronger binding and perhaps more disordered substrates, such as Mylar [9] and quartz [10].

We have measured the superfluid density and dissipation of thin helium films using an Al alloy 5056 torsional oscillator [11]. The active oscillator surface is a single 0.025-cm-thick and 1.27-cm-radius disk with a geometrical surface area of 10.1  $\text{cm}^2$ . In the experiments described below the oscillator was operated in a cell with an open volume of 130  $\text{cm}^3$ . A stack of Al foil disks was used as an area ballast, giving a geometrical cell surface area of 3700  $\text{cm}^2$ . The typical low-temperature quality factor of our oscillator was  $Q \sim 5 \times 10^6$  at a resonant frequency of 2450 Hz. The oscillator was operated at a constant amplitude via a proportion-integral gain arrangement on the drive to circumvent possible amplitude-induced period shifts. In a typical experiment,  $\text{H}_2$  was introduced to the cell near 15 K and the oscillator was preferentially cooled to 4 K, with a 1-K temperature gradient, over a period of several hours. We put in enough  $\text{H}_2$  to form liquid at the bottom of the cell which, assuming a  $\text{H}_2$ -Al van der Waals constant of  $\sim 150$  Klayer [3], resulted in  $\text{H}_2$  coatings of at least 100 atomic layers thick [12]. After growing the hydrogen, the background quality factor  $Q_0$  and period  $P_0$  were measured as a function

of temperature and later subtracted from the data. Measured amounts of helium were then introduced to the cell and the temperature dependence of the period and  $Q$  were studied between 0.4 and 1.4 K.

Our oscillator sensitivity is such that 1 layer of normal helium liquid, taken to be 0.36 nm thick, results in a relative period change  $2\Delta P/P_0 = 1.25 \times 10^{-7}$ . With a period stability of 1 part in  $10^9$  we are able to resolve changes in superfluid density corresponding to less than  $\frac{1}{30}$  of a monolayer. In an open geometry, the period and dissipation shifts from the oscillator's intrinsic values depend on both the film on the surface [9] and the helium vapor [13]:

$$\frac{2\Delta P}{P_0} = \frac{\pi(\sigma - \sigma_s)R^4}{I_0} + \frac{\alpha\pi R^4}{I_0} \left( \frac{\rho_g \eta}{2\omega} \right)^{1/2}, \quad (1)$$

$$\Delta Q^{-1} = \Delta Q_f^{-1} + \frac{\alpha\pi R^4}{I_0} \left( \frac{\rho_g \eta}{2\omega} \right)^{1/2}, \quad (2)$$

where  $\sigma$  is the total 2D mass density of helium on the surface,  $\sigma_s$  is the superfluid density,  $R=1.27$  cm is the substrate radius,  $I_0=0.34$   $\text{g cm}^2$  is the oscillator moment of inertia,  $\omega=2\pi/P_0$  is the resonant frequency,  $\rho_g$  is the vapor mass density, and  $\eta$  is the viscosity of the vapor. The second terms in Eqs. (1) and (2) are due to the vapor and we have assumed that the helium gas can be treated hydrodynamically. We have included a dimensionless parameter  $\alpha$  in these terms to account for vapor coupling to the oscillator detection and drive vanes [11]. For a simple disk,  $\alpha=1$ . We calibrated the period and  $Q$  shifts at 18 K using hydrogen gas and obtained  $\alpha=1.9$ .

The dynamics of the superfluid transition [14] are determined by  $\sigma_s(T)$  and  $\Delta Q_f^{-1}(T)$  in Eqs. (1) and (2). For our purposes, the only two characteristics of the theory that are important are that  $\Delta Q_f^{-1}(T)$  is not significant except for temperatures very near the transition temperature  $T_c$ , where it produces a dissipation peak, and that the superfluid density at the transition is simply proportional to the transition temperature [9,14]:

$$\sigma_s(T_c) = (2k_B m^2 / \pi \hbar^2) T_c, \quad (3)$$

where  $m$  is the helium atom mass. The above relation is the hallmark of the 2D superfluid transition in helium which is commonly known as the Kosterlitz-Thouless

(KT) transition [15]. Equation (3) has been verified in torsional oscillator [9], conductance [16], and third-sound [17] measurements of superfluid density on strong binding substrates.

In our system,  $\sigma$  decreases with increasing temperature due to vaporization. This complicates our analysis in that the liquid density on the oscillator surface is not simply proportional to the amount of helium added to the cell,  $N$ . Fortunately, however, we were able to systematically push the transitions up in temperature starting from below 0.5 K where  $\rho_g$  is negligible and then use Eq. (3) to estimate the film thickness at the higher temperature transitions. Shown in Figs. 1(a) and 1(b) are the reduced period and excess dissipation of the hydrogen-

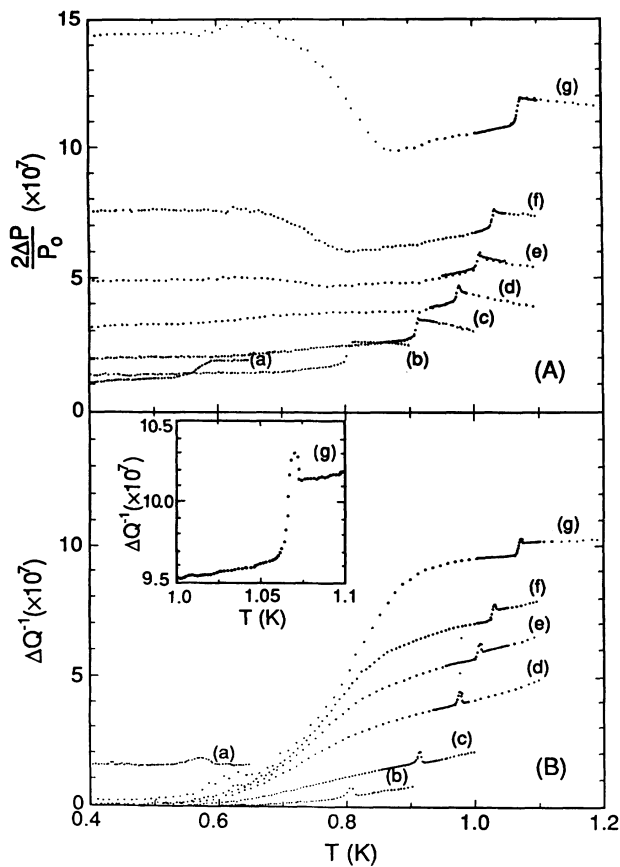


FIG. 1. (a) Reduced period of the hydrogen-coated oscillator as a function of temperature with various amounts of helium in the cell. The helium doses in  $\mu\text{moles}$  are (curve *a*) 6.4, (*b*) 8.4, (*c*) 14.1, (*d*) 26.0, (*e*) 38.0, (*f*) 50.0, (*g*) 81.7. The steps in the reduced period represent the 2D superfluid transition. (b) The change in inverse  $Q$  of the oscillator as a function of temperature. The small dissipation peaks correspond to the steps in (a). Curve *a* has been shifted upward for clarity. Inset: A magnification of the dissipation peak in curve *g*. The break in the background in the inset is due to enhanced vapor slip on the superfluid side of the peak.

coated oscillator as a function of temperature with various amounts of helium in the cell. Figure 1 contains a representative subset of all the data taken. The steps in the reduced period and corresponding dissipation peaks represent the KT superfluid transition. The transitions are superimposed on a vapor background [i.e., second terms in Eqs. (1) and (2)] and are therefore shifted upward in the higher coverage curves. The dramatic structure below 0.9 K in curves *f* and *g* of Fig. 1(a) results from the competition between film condensation on the oscillator, which shifts the period up (see "dead layer" discussion below), and decreasing vapor density, as the oscillator is cooled from 1 to 0.4 K.

The low-temperature tails of the reduced periods in Fig. 1(a) are a measure of the portion of the films that presumably will not decouple from the oscillator, even at  $T=0$ . This remnant of normal fluid far below the transition is commonly known as the dead layer and is probably a function of both surface disorder and binding energy. The minimum dead layer on Mylar [9] is about 1.5 layers. Shown in Fig. 2 are the extrapolated  $T=0$  period shifts as a function of the helium dose for bare "as-machined" Al and  $\text{H}_2$ -coated Al. The dashed line in Fig. 2 represents the expected period shift for a purely nonsuperfluid film and all of the data should fall on or below this line. In this figure, 4.8  $\mu\text{moles}$  of helium corresponds to 1 monolayer coverage. Note that  $\text{H}_2$  decreases the dead layer below that on bare Al by about a factor of 4, and that in both cases the *apparent* dead layer is a function of coverage. Similar behavior is observed on Mylar where, in coverages greater than the dead layer,

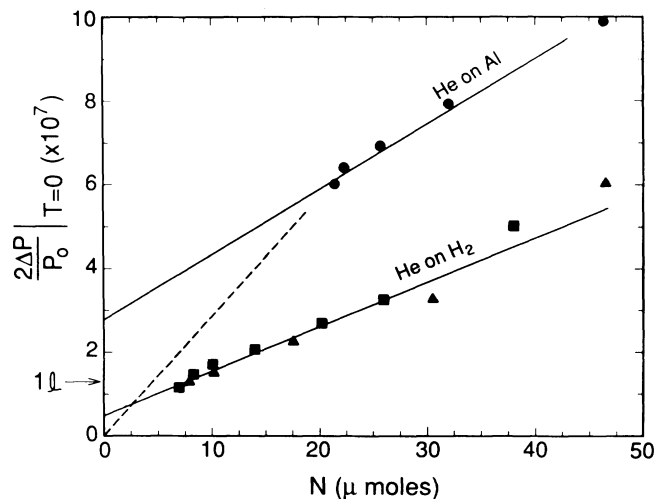


FIG. 2. The extrapolated  $T=0$  period shifts as a function of helium dose. These data represent the dead layer on  $\text{H}_2$  and Al. The triangles and squares are data from two different hydrogen crystals. The solid lines are provided as a guide to the eye. The dashed line is the expected period shift for nonsuperfluid helium. The arrow is the period shift due to 1 layer of normal helium on the surface at liquid density.

about 15% of a film remains coupled at temperatures well below the transition [9]. We suspect that this may, in part, be a consequence of inertia associated with inviscid flow around surface irregularities. The oscillator technique cannot distinguish between dynamic and static inertia.

There have been several recent measurements of the dead layer of helium on hydrogen using third sound in which widely varying values have been reported. Shirron and Mochel [6] observed a dead layer of about  $\frac{1}{3}$  monolayer in very thin films with transition temperatures near 0.2 K. Zimmerli, Mistura, and Chan [7] have reported superfluidity only above 1 layer in thicker films with transition temperatures between 0.65 and 1 K. Finally, Brisson, Mester, and Silvera [18] have observed superfluidity only above 2 helium layers in films thicker than 2.5 layers. We believe that all of these results may be consistent with the coverage dependence of the *apparent* H<sub>2</sub> dead layer shown in Fig. 2. The solid lines in Fig. 2 are provided as a guide to the eye and we interpret the intersection of the H<sub>2</sub> line with the oscillator sensitivity line (dashed line) as an upper limit to the actual localized fraction of the film. This gives a static dead layer on hydrogen of about  $\frac{1}{2}$  layer thick, in reasonable agreement with that reported by Shirron and Mochel [6].

Shown in Fig. 3 are the reduced period jumps at the transition as a function of transition temperature. By the KT relation in Eq. (3), these jumps, which measure the 2D superfluid density at the transition, should simply be proportional to the transition temperature. Note the sharp suppression of the superfluid density for transitions near 1 K. A similar analysis of transitions on bare Al revealed a rather weak remnant of the behavior in Fig. 3. Presumably this was a result of the relative roughness of the as-machined Al.

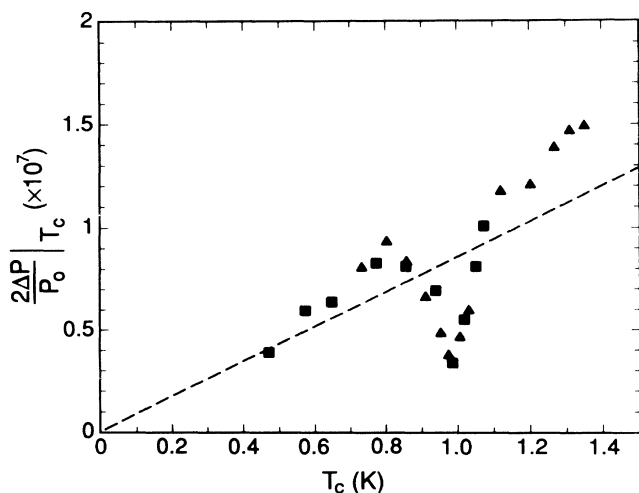


FIG. 3. The height of the reduced period steps at the transition as a function of transition temperature. The dashed line is the prediction of Eq. (3). The two symbols represent measurements on different hydrogen crystals.

From our measurements of the dead layer we are reasonably sure that the dip in Fig. 3 is occurring near the completion of the second monolayer. As can be seen in Fig. 1(a), a curious overshoot, which was independent of drive amplitude, also appears in the reduced period in this same region. We do not understand this apparent breakdown of KT behavior, but speculate that if it is a layering effect [19], it may be related to the third-sound velocity modulations observed in helium films on hydrogen and graphite [6,7]. Another possibility, which we discuss in some detail below, is that the film-vapor coupling is changing abruptly at these transitions and therefore the period shifts are not simply proportional to  $\sigma_s$ .

We have, for the first time, measured the change in viscous drag at a superfluid-vapor interface through the superfluid transition. Though there have been previous reports of excess vapor slip on superfluid films [20,21], there have not been any experiments that simultaneously measure superfluid density and vapor dissipation directly across the KT transition. This is absolutely necessary in order that the effects of superfluidity be unambiguously identified. The dissipation peaks in Fig. 1(b) are on a helium vapor background given by the second term in Eq. (2). Note that for the higher coverage curves, *e-g*, the dissipation backgrounds are shifted upward on the hot side of the transition. This effect, which is most clearly evident in the inset of Fig. 1(b), is independent of drive amplitude. Curves *e-g* are very unusual in that one typically observes slightly more dissipation on the *cold* side of the peak than on the hot side [9]. We interpret the break in these curves as a disappearance of superfluid-induced gas-atom slip as one warms through the transition.

The change in gas-atom coupling across the transition was never larger than about 5% of the total gas-induced dissipation. In Fig. 4 we have plotted the change in  $Q^{-1}$  across the transition as function of transition temperature. For transition temperatures below 0.9 K, dissipa-

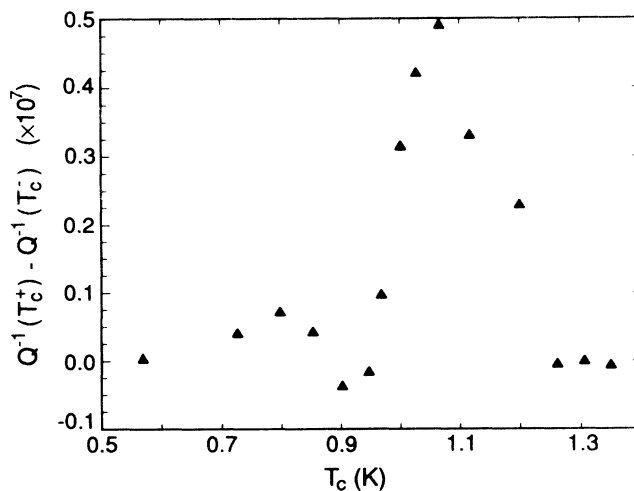


FIG. 4. The difference in inverse  $Q$ , as measured across the transition, as a function of transition temperature.

tion tails in the superfluid phase [9] may have masked the effect, but, clearly, we got significant positive differences for transition temperatures near 1.1 K. Interestingly, the differences completely disappeared for transition temperatures above 1.3 K. We do not understand this, but the data imply that there is significant coupling between the gas and excitations in the superfluid (i.e., third sound, rotons, phonons, vortex pairs) and that gas-atom slip is not simply proportional to  $\sigma - \sigma_s$ . In fact, a cursory inspection of Figs. 3 and 4 suggests that we are measuring the real and imaginary components of a resonant coupling phenomenon.

Measurements of the interaction between vapor and counterflowing bulk superfluid helium between 2 and 1.3 K by Osborne [22] showed that vapor does not couple to the superfluid component of the liquid. Later measurements of persistent currents in films with finite vapor pressure [23] conclusively demonstrated that vapor does not transfer momentum to a superfluid. Nevertheless, vapor does not necessarily decouple from a superfluid film. In fact, on thermodynamic grounds it is expected that the relative velocity between the normal-fluid component and the vapor is zero at the interface [24], and this is, in fact, what Osborne observed in bulk helium down to 1.3 K and is consistent with what we observe in films with  $T_c > 1.3$  K. Obviously, though, this no-slip boundary condition should break down at lower temperatures where the normal-fluid density is rapidly approaching zero, thereby giving one a probe of the microscopics of the vapor-film coupling. Thus, the data in Figs. 3 and 4 may represent the crossover between an essentially no-slip regime to that of significant slip. This picture is strongly supported by recent evidence that superfluidity induces specularly in  $^3\text{He}$  quasiparticle scattering on  $^4\text{He}$  films [25].

The authors would like to thank Dr. W. Johnson, Dr. D. Browne, Dr. R. Hallock, Dr. J. Parpia, and Dr. S. Putterman for helpful discussions. This work was supported by the National Science Foundation under Grant No. DMR-89-11233 and Louisiana Education Quality Support Fund Grant No. LEQSF(1989-92)-RD-A-1.

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