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1992 J. Phys.: Condens. Matter 4 9525

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^4He superfluidity on hydrogen and C_{60}

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Received 29 July 1992, in final form 6 October 1992

Abstract. We report measurements of the real ^4He superfluid density, σ_s , as a function of temperature and coverage on hydrogen and C_{60} substrates using a high- Q Al oscillator. Sub-kelvin measurements of the helium mass loading of a 780 nm thick C_{60} film deposited on the active surface of the oscillator show that helium does not significantly permeate the interstitial sites of the C_{60} matrix, in either its superfluid or normal phase. In fact, the Kosterlitz–Thouless superfluid transition is observed on the C_{60} at very modest mass loadings. The superfluid behaviour on C_{60} is found to be very similar to what we have observed for helium on solid hydrogen, namely a suppression of $\sigma_s(T_c)$ in transitions near 1 K and excess vapour dissipation in the normal phase.

1. Introduction

Over the past three years there has grown a significant interest in the role of helium–substrate interaction in the wetting [1, 2] and superfluid behaviour of ultra-thin helium films [3, 4]. It is now known that the strength of the helium–substrate interaction can have a profound influence on wetting with wetting being completely suppressed on the very weakly binding surface of caesium [5–7]. Similarly, the interactions of a helium film with its interfaces should have significant bearing on its superfluid characteristics [8]. In the experiments described below the apparent superfluid behaviour is modified by both the film–substrate interface and the film–vapour interface.

2. Experimental method

We have made superfluid density and dissipation measurements in thin helium films on two rather weakly binding substrates, solid H_2 and poly-crystalline C_{60} , using an ultra-high Q (quality factor) torsional oscillator. This is one of the most sensitive and direct techniques available for probing thin film superfluidity and we were able to discern changes in superfluid density corresponding to less than $\frac{1}{30}$ of a monolayer of helium. The details of our oscillator design have been described elsewhere [4, 9]. For the purposes of the present discussion it should be noted that the oscillator active surface was a single 0.025 cm thick and 1.27 cm diameter disc operated in a cell with 130 cm³ open volume. Typically at 1 K the intrinsic Q of the oscillator was 4×10^6 at a resonant period $P_0 = 4.1 \times 10^{-4}$ s. The oscillator amplitude was held constant via proportional-integral feedback on the drive and the period was measured to a part in

10^9 . The first measurements described below were done after coating the oscillator with H_2 by introducing hydrogen into the cell at 15 K and then slowly cooling to 4 K. Sufficient hydrogen was put into the cell so as to form a saturated film. The measurements on C_{60} were made after thermally evaporating a total of 780 nm of C_{60} on both sides of the active oscillator surface at room temperature. The C_{60} film was believed to be in the form of a disordered FCC crystal.

3. Results and discussion

Since the oscillator was operated in an open geometry the film thickness decreased with increasing temperature due to vaporization. This complicated our analysis in that the liquid density on the oscillator surface was not simply proportional to the amount of helium added to the cell, N . Furthermore, helium vapour produces mass loading and excess dissipation via its viscous coupling to the oscillator [4, 10]. Nevertheless, one can still obtain important information about the transition in an open geometry. Shown in figure 1(A) and figure 1(B) are the reduced period and excess dissipation of the hydrogen coated oscillator as a function of temperature with various amounts of helium in the cell. The large upward shifts in ΔQ^{-1} with increasing temperature are due to vapour. Similar shifts in reduced period would also be seen if there were no film evaporation from the oscillator surface. In curve (g) of figure 1(A), for instance, helium leaving the surface causes the decrease in period between 0.7 K and 0.9 K and increasing vapour pressure produces the increase in period between 0.9 K and the superfluid transition at 1.05 K. Notwithstanding these complications, the 2D superfluid transitions are clearly evident on the vapour backgrounds. The steps in reduced period represent the superfluid density at the transition. The superfluid density at the transition is finite in 2D and is given by the Kosterlitz–Thouless relation [11, 12]

$$\sigma_s(T_c) = \frac{2k_B m^2}{\pi \hbar^2} T_c \quad (1)$$

where m is the helium atom mass and T_c is the transition temperature. There is also an excess dissipation associated with the transition [12, 13] which can be seen as the small dissipation peaks in figure 1(B).

The low-temperature tails of the reduced periods in figure 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 consequence of surface disorder and binding strength. In helium studies on Mylar [14], superfluidity is only observed in coverages above about 1.5 layers. In this figure $2\Delta P/P_0 = 1.25 \times 10^{-7}$ corresponds to one layer of non-superfluid on the surface. Note that the *apparent* dead layer on hydrogen is a function of coverage. We found that even well into the superfluid phase only about $\frac{2}{3}$ of an incremental dose of helium would actually decouple from the oscillator. This may have been a consequence of both a coverage dependent localized helium layer [15] and inertia associated with superfluid flow around surface irregularities. Nevertheless, extrapolation of the data in figure 1(A) to $T_c = 0$ gives a minimum dead layer on hydrogen of about $\frac{1}{2}$ monolayer. This is in reasonable agreement with third sound measurements of the dead layer on hydrogen coated sintered copper [3].

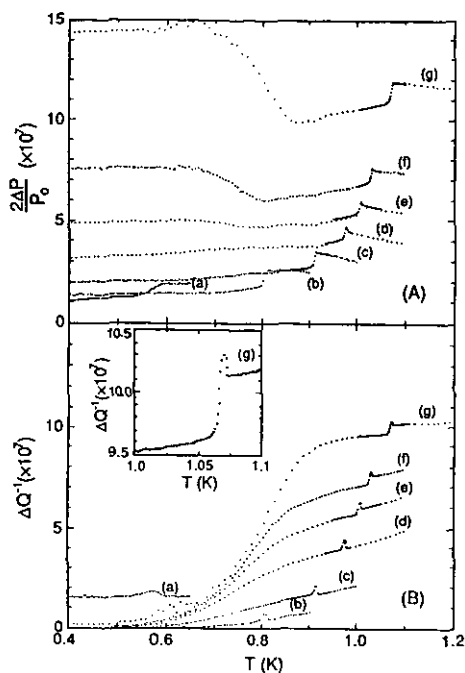


Figure 1. (A) Reduced period of the hydrogen cell coated oscillator as a function of temperature with various amounts of helium in the cell. The helium doses in μ moles are (a) 6.4, (b) 8.4, (c) 14.1, (d) 26.0, (e) 38.0, (f) 50.0, (g) 81.7. The steps in the reduced period represent the 2D superfluid 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. The inset is a magnification of the dissipation peak in curve (g). The break in the background in the inset is due to enhanced vapour slip on the superfluid side of the peak.

Two other characteristics of the data in figure 1 are of interest. The first is the appearance of excess vapour dissipation on the hot side of the transition as shown in the inset of figure 1(B). This we believe is due to excess gas atom slip in the superfluid phase [4]. The second unusual aspect of the transitions is the apparent suppression of $\sigma_s(T_c)$ in transitions near 1 K. This latter anomaly may again be a consequence of film–vapour coupling or it may have something to do with the layering behaviour of helium on hydrogen. Shown in figure 2 are the reduced period jumps at T_c as a function of T_c . The dashed line is the prediction of equation (1). We believe the suppression was occurring near second monolayer completion where presumably the helium–helium correlations were greatly enhanced. We were not, however, able to get low to enough temperatures to see the effect of first monolayer completion on the transition. Therefore we could not rule out the possibility that the suppression in $\sigma_s(T_c)$ near 1 K is related to superfluid–vapour coupling and, in fact, had nothing to do with layering.

In an effort to further probe the relative roles of film–substrate and film–vapour interactions on the superfluid dynamics we extended our studies to helium films on C₆₀. We began these measurements fully expecting that superfluidity would not be observed on the surface of C₆₀ until the interstitial sites between the spherical

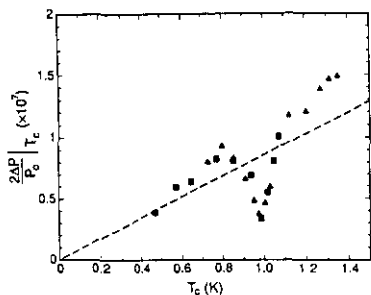


Figure 2. The magnitude of the reduced period steps at the transition as a function of transition temperature for helium on hydrogen. The dashed line is the prediction of equation (1). The two symbols represent measurements on different hydrogen crystals. In this figure $2\Delta P/P_0 = 1.25 \times 10^{-7}$ corresponds to the mass loading of one layer of normal phase helium.

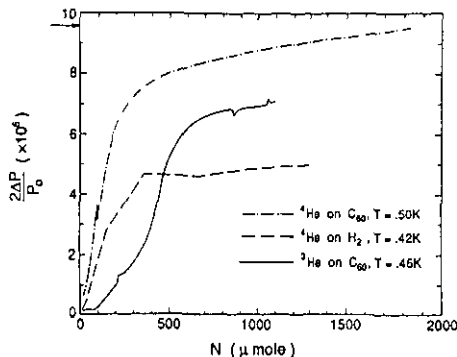


Figure 3. The low-temperature mass loading of the oscillator with coatings of hydrogen and C_{60} . The arrow marks the expected period shift for the absorption of one ^4He atom per C_{60} molecule.

molecules were filled with helium. Helium cannot penetrate a C_{60} molecule due to a barrier of several eV [16] therefore, the only available sites in the crystal are two vacant tetrahedral and one octahedral interstitial site per molecule [17]. These sites are large enough to accommodate spheres of radius 0.112 nm and 0.206 nm respectively. Given that the covalent radius of a helium atom is 0.093 nm it is not unreasonable to expect a rather large mass loading associated with absorption of helium into the C_{60} film. This, however, is not the case as we now argue. The arrow in figure 3 represents the expected reduced period shift for the absorption of one helium atom per C_{60} molecule. The dash-dot line is the mass loading for ^4He on C_{60} at 0.5 K. The reduced period appears to saturate at a value suggesting helium is going into the octahedral sites. However, the oscillator technique cannot distinguish between mass on the surface and mass absorbed into the crystal. In fact, the ^4He mass loading of C_{60} in figure 3 is only about a factor of 2 larger than that obtained on solid hydrogen (dashed line). The large mass loadings on hydrogen were probably due to the hydrodynamic inertia of superflow around large scale surface structure. Therefore, the data in figure 3 indicate that at most only about $\frac{1}{6}$ of the interstitial sites were occupied at large coverages. Similar results have been obtained via quartz microbalance measurements [18]. Also shown in figure 3 is the mass loading of ^3He . These data were taken to rule out the somewhat unlikely possibility that helium was, in fact, permeating the crystal as a superfluid. If this were the case, then helium might not couple and thus appear to not soak the C_{60} crystal. However, it is clear from the data that ^3He does not load the crystal significantly.

More direct evidence for non-absorption came from the observation of superfluidity on the surface of C_{60} at relatively small mass loadings. Shown in figure 4 are the 2D superfluid transitions on C_{60} in which the vapour background has been subtracted from the data for clarity. The $T_c \sim 0.5$ K transition was observed after a mass loading of only $2\Delta P/P_0 \sim 5 \times 10^{-7}$. This is a factor of 20 less than the mass loading associated with one helium atom per C_{60} . In fact, if all of the helium was

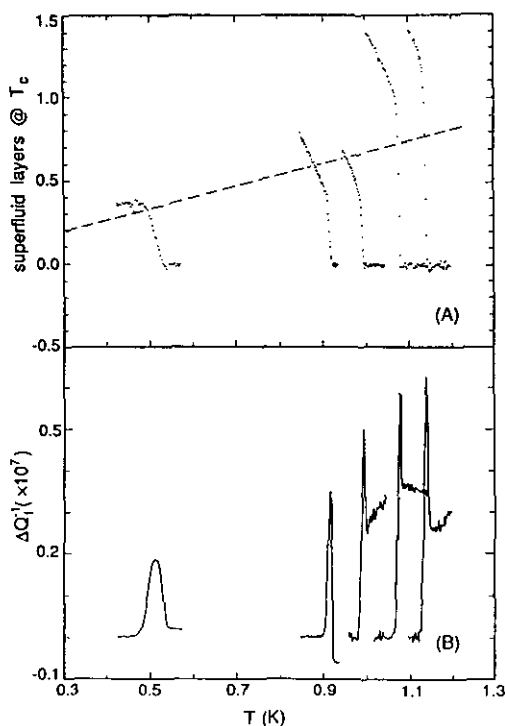


Figure 4. Superfluid transitions on C_{60} . The helium vapour backgrounds have been subtracted from the data for clarity. In part (A) the superfluid is expressed in units of layers of helium at normal liquid density. The dashed line is the predicted superfluid thickness from equation (1).

on the surface of the carbon film then the lowest transition suggests a dead layer of about 3–4 nominal monolayers on C_{60} . This seems reasonable given the corrugations in the surface of C_{60} .

Our data suggests that the interaction between helium and the interstitial sites is repulsive. Another possibility is that the interaction is only very weakly attractive and is, therefore, unable to accommodate the zero-point energy of the helium atoms. A very rough estimate of the zero-point energy, $E_{zp} \approx \hbar^2 / (mr^2)$, where r is the site radius, gives $E_{zp}/k_B \sim 3$ K for the octahedral site. This energy is significant since our estimate is probably low due to the finite size of a helium atom.

We will now turn our attention to the superfluid transitions on the surface of C_{60} . Note that the dissipation peaks in figure 4(B) develop an asymmetry in transitions occurring at significant vapour pressure. As the transition temperature is increased the difference in dissipation across the transition first becomes negative with less dissipation on the hot side. It then quickly changes sign becoming significantly positive. Since this phenomenon is so clearly associated with the superfluid transition it is almost certainly a direct probe of the interaction between the vapour and underlying superfluid in a regime where there is some finite compliance between the two. The asymmetry disappears at higher transition temperatures where presumably the vapour is completely clamped into a no-slip boundary condition by normal fluid excitations in the film. The fact that this same behaviour was also manifest

on hydrogen indicates that we were indeed probing the film–vapour interaction. Unfortunately, to our knowledge, there is no quantitative description of the interface between a superfluid film and its vapour to which we can compare our data.

The superfluid density on C_{60} near the transition is shown in figure 4(A). The dashed line is the prediction of equation (1). Note that the magnitudes of the transitions are not monotonic in T_c . There is a noticeable attenuation of the transition at $T_c = 1$ K. This is the same anomalous behaviour that is displayed in figures 1(A) and 2 for transitions on hydrogen. Since these two substrates are very different, our data suggests that the anomaly is of the same origin as the dissipation asymmetry, namely, that a rapid change in gas coupling associated with the superfluid transition is producing a period shift of the opposite sign to that due to the film in transitions near 1 K. At this temperature the mean-free-path and the viscous penetration depth of the vapour are comparable [19]. Therefore the magnitude of the slip length and the viscous penetration depth are probably also comparable at 1 K. Perhaps it is only in this regime that a change in slip length at T_c produces a significant signature in the data.

Acknowledgments

The authors would like to thank D Browne, R Hallock and R Kalia for helpful discussions. We would also like to thank R C Haddon and A V Makhija for supplying us with purified C_{60} . 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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