

Unexpected Mass Decoupling for ^3He - ^4He Films on a Solid Hydrogen Substrate

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Quartz oscillator measurements reveal new behavior in ^3He - ^4He mixture films on a H_2 substrate. For mixture films of total coverage greater than one monolayer, in addition to the expected Kosterlitz-Thouless transition, a second mass decoupling event is observed, which behaves differently from the Kosterlitz-Thouless transition.

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On strong binding substrates ^4He shows superfluid behavior for film coverages that exceed \sim two monolayers of inert ^4He adjacent to the substrate [1]. On a H_2 substrate, which is a much weaker-binding substrate, ^4He exhibits superfluidity at submonolayer coverages [2–6]. Such a weak-binding surface provides an interesting environment in which to study thin film superfluidity. Here we report the results of quartz crystal microbalance measurements for ^4He and ^3He - ^4He mixtures on a H_2 substrate that show the presence of a Kosterlitz-Thouless transition at T_{KT} and a second mass decoupling feature at a characteristic temperature T_c : $T_c < T_{\text{KT}}$.

The apparatus consisted of two AT cut quartz crystal microbalances (QCM) with Au electrodes in a 169 cm^3 brass sample cell that contained 784 cm^2 of loosely crinkled and rolled copper foil that resulted in a total macroscopic geometric area $\approx 1042\text{ cm}^2$. A feedback circuit [7] tracked the QCM fundamental (5 MHz) or third harmonic (15 MHz) shear-mode frequency so that any change in the adsorbed mass, δm , coupled to the Au electrodes could be detected as a linear change in the frequency, $\delta f \propto -\delta m$. Measurement of the power output of the crystal provided a measure of the dissipation, $\sim Q^{-1}$. The fractional frequency shift of the QCM obeys the relation $\delta f/f = \sigma/(\rho t)$ where σ is the areal mass density coupled to the QCM, ρ is the density of the quartz, and t is an effective thickness of the quartz disk that can be calculated from the fundamental frequency. The measurements presented here were taken in the third harmonic mode, which provided a greater signal to noise ratio than the fundamental, allowing adsorbed mass resolution of $\sim 6 \times 10^{-11}\text{ g/cm}^2$ ($\sim 0.02\text{ Hz}$).

After cooling the cryostat to liquid helium temperatures, $6.01 \pm 0.06\ \mu\text{mol}$ of H_2 was admitted to the cell at 19 K through a double-walled, heated fill tube held at a warmer temperature than the sample cell. The sample cell temperature was slowly reduced to 2 K over a 6 d period while maintaining the cell colder than the fill tube to ensure that the H_2 plated the sample cell.

To the $0.263 \pm 0.007\ \text{\AA}^{-2}$ (3.0 bulk-density layers) of H_2 , room temperature helium gas was added with the sample cell at $T \leq 200\text{ mK}$. The first run (#1) consisted

of data taken at 11 incrementally increased amounts of ^4He up to a total dose of $2.942\ \mu\text{mol}$ (1.65 bulk-density layers). After each dose of ^4He , the sample was annealed for 10 h at $\geq 700\text{ mK}$ before data were taken. For each coverage, the QCM was monitored as the cell slowly cooled to below 35 mK. Data taken while warming show little or no hysteresis. At the completion of run #1, ^4He was removed from the cell while leaving the hydrogen substrate intact by pumping for 15 h at 1.8 K. Run #2 began with several small pure ^4He doses to determine the amount of ^4He left in the cell after pumping (by observing T_{KT}). Mixture films were then studied by incremental additions of ^3He or ^4He . The two QCMs at different locations in the cell showed very similar behavior.

Data taken for pure ^4He films in excess of $\approx 1.5\ \mu\text{mol}$ (0.8 layers) exhibit an abrupt increase in frequency as a function of decreasing temperature corresponding to decoupling of superfluid at the Kosterlitz-Thouless transition temperature, T_{KT} . A corresponding peak in the dissipation [8,9] is observed at T_{KT} . Knowledge of T_{KT} provides us with a measure of the surface area within the cell. The superfluid areal number density jump, n_s , at the transition temperature is proportional [10] to T_{KT} : $n_s = 8\pi m k_B T_{\text{KT}}/h^2$. From the QCM properties mentioned above, the frequency shift observed at T_{KT} , Δf_{KT} , can be converted to a corresponding decoupled areal mass density that is in good accord with predictions of theory [Fig. 1(a)]. The KT transitions vary somewhat in width, an effect seen previously by Agnolet *et al.* [11] for pure ^4He on a Mylar substrate.

In order to determine the effective surface area [12] within the cell, we consider the total number of ^4He atoms in the cell as a function of T_{KT} [Fig. 1(b)]. The relationship is linear except for the highest several pure ^4He coverages. The departure of the highest coverage data points from linearity [Fig. 1(b)] is due to desorption of ^4He from the substrate and allows a measure of the binding energy of the ^4He [6]. Through comparison of superfluid density measurements for ^4He on Mylar, the data of Agnolet *et al.* [11] suggest [3] that $\sigma_s(T_{\text{KT}}) \approx 0.75\sigma_s(0)$, where $\sigma_s(T)$ is the superfluid number density at T . Chen *et al.* [3] used this result combined with KT theory to show that the slope, α ,

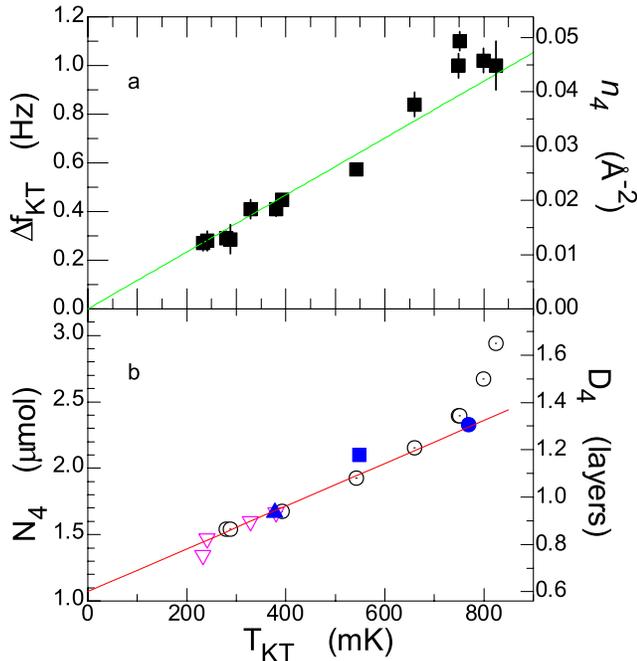


FIG. 1 (color online). (a) Size of the frequency shift observed at T_{KT} versus temperature. The right-hand axis displays the jump in ^4He superfluid density corresponding to the frequency shift. The line is the predicted ^4He superfluid jump at T_{KT} . (b) Total amount of ^4He in the cell versus T_{KT} for pure ^4He films. The open circles and open triangles are taken from pure ^4He data from runs 1 and 2, respectively. The solid triangle, square, and circle are each linear extrapolations of mixture film data to the zero ^3He limit (See the text discussion).

of the linear region is proportional to the surface area within their cell: $A = 0.75\alpha\hbar^2/(8\pi mk_B)$. This procedure applied to our data yields an effective area of $1374 \text{ cm}^2 \pm 10\%$, which is 32% larger than our estimate of the total macroscopic surface area. The uncertainty in this value is due to the quality of the fit to our data ($\sim 4\%$) and the uncertainty in the Agnolet *et al.* [11] result. This area was used to calibrate coverages: $1 \mu\text{mol} = 0.0438 \text{ \AA}^{-2} = 0.56$ bulk-density ^4He layers, or 0.68 bulk-density ^3He layers.

The intercept of the linear fit on Fig. 1(b) shows that the thickness of the zero temperature inert layer is 0.60 bulk-density layers (0.047 \AA^{-2}), consistent with the work of some [2–6] who report submonolayer inert layers, but thinner than the inert coverage ≥ 1 monolayer seen in other studies [13]. We have never seen in our pure ^4He measurements any second KT-like feature such as that reported by Chen *et al.* [3].

To prepare for the addition of ^3He to the cell most of the ^4He was removed and small increments of ^4He were added [inverted triangles, Fig. 1(b)] to confirm the previous KT behavior. At a ^4He coverage of 0.94 layers (0.073 \AA^{-2}), the introduction of 0.071 bulk-density ^3He layers (0.0046 \AA^{-2}) and subsequent additions of ^3He re-

duced T_{KT} (solid triangles, Fig. 2). The reduction of T_{KT} with the addition of ^3He is commonly observed in multilayer films on strong binding substrates [14]. The addition of ^4He to these mixture films raised T_{KT} (\times 's, dotted segments, Fig. 2). Data taken at several fixed ^4He coverages and increasing ^3He coverage show that T_{KT} decreases linearly with ^3He coverage as is shown by the various solid symbols in Fig. 2. Extrapolations of the linear behavior to zero ^3He coverage yield T_{KT} intercepts that fall on the KT line shown in Fig. 1(b) (solid symbols). Tulimieri *et al.* [5] saw, in a torsional oscillator study, that the addition of up to 0.066 layers ^4He to 0.59 layers ^4He on a hydrogen substrate of density 0.358 \AA^{-3} increased T_{KT} . This result may suggest a change in behavior in the vicinity of one ^4He monolayer.

At the higher mixture film thicknesses studied, a new feature appears in the QCM frequency as a function of temperature. At a ^4He coverage near one bulk monolayer (1.01 layers ^4He , 0.30 layers ^3He), this new feature appears as a broad decoupling of mass from the QCM at a temperature, T_c , just below T_{KT} . We illustrate this behavior for a fixed ^4He coverage of 1.18 layers in Fig. 3, which shows that this shift in the QCM frequency increases in size with increasing ^3He coverage with $140 \leq T_c \leq 290$ mK. While a dissipation peak accompanies the KT transition (Fig. 3), no corresponding dissipation feature is resolved at T_c . Figure 2 shows the movement of T_c (open symbols) and T_{KT} (solid symbols) as the ^3He coverage is increased: $T_c < T_{KT}$. The size of the decoupling shift, Δf_c , at T_c is strongly dependent on the ^3He coverage. Shown in Fig. 4 are the values of $\Delta f_{KT}/T_{KT}$ and $\Delta f_c/T_c$ determined from the data. The Kosterlitz-Thouless transitions follow the ex-

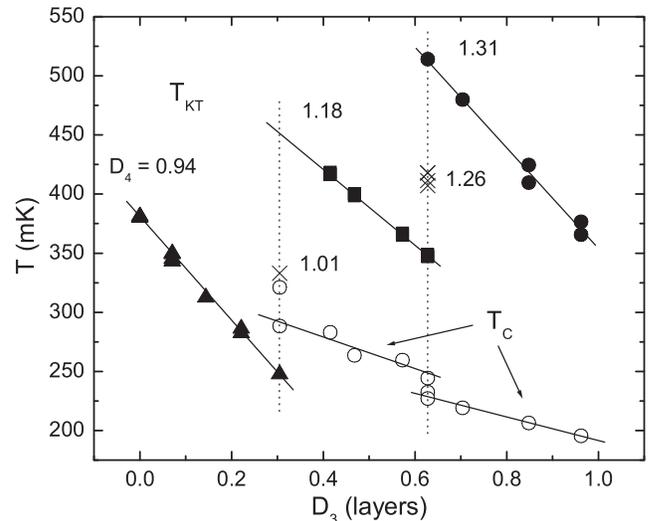


FIG. 2. T_{KT} and T_c versus ^3He coverage. ^4He coverages are given as bulk-density monolayers. Dotted lines indicate a change in ^4He coverage. Solid lines are linear fits. Zero ^3He coverage intercepts of the linear fits to T_{KT} are shown with the corresponding solid symbols in Fig. 1(b).

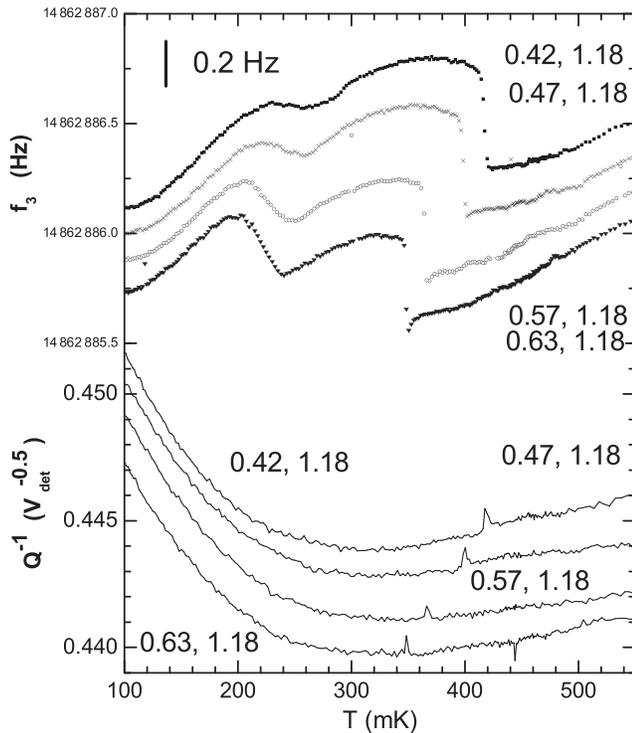


FIG. 3. Third harmonic frequency (near 14.862 MHz, top data sets) and corresponding dissipation (bottom data sets) as a function of temperature for ^4He coverage of 1.18 layers. ^3He coverage is given in bulk-density layers. The frequency shifts for $T > 340$ mK with corresponding dissipation peaks are KT transitions.

pected trend, a behavior similar to that seen previously for mixture films by Webster *et al.* [15] for thicker films at higher temperatures, except for the highest concentrations.

Wang and Gasparini (WG) [16] extended the earlier work of Bishop and Reppy [9] to investigate double KT transitions in mixture films on Mylar (no hydrogen). With a torsional oscillator technique, they observed mass decoupling at two temperatures with a dissipation peak accompanying *each* decoupling transition for *active* ^4He coverages (i.e., coverage above the inert layer) of 1.19 and 1.22 layers and ^3He coverages between 0.04 and 1.51 layers. WG concluded that their results were likely due to regions of differing density and concentration that independently underwent KT transitions. Our results are in a regime of active ^4He coverage lower than studied by WG, and our data at T_c do not display KT-like features. There is no resolved dissipation peak at T_c , Δf_c at T_c grows as T_c is reduced, and $\Delta f_c/T_c$ does not behave as predicted by Kosterlitz-Thouless theory. The work of Chen *et al.* [3] observed two transitions on hydrogen, but for pure ^4He . We have never observed double transitions for pure ^4He on hydrogen.

In an effort to understand the possible cause of this frequency shift at T_c , we consider several models that involve a decoupling of either ^3He atoms or ^4He atoms.

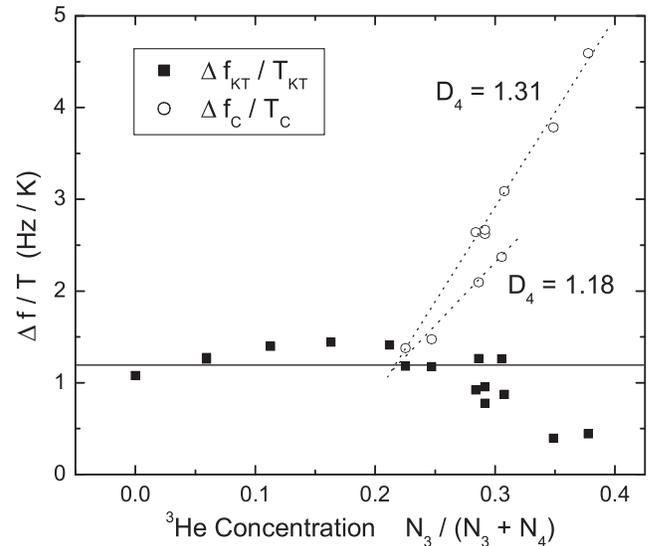


FIG. 4. $\Delta f_{\text{KT}}/T_{\text{KT}}$ and $\Delta f/T_c$ versus ^3He concentration, $N_3/(N_3 + N_4)$. Solid symbols are Kosterlitz-Thouless transitions $\Delta f/T_{\text{KT}}$ as observed; the solid line is the expected $\Delta f_{\text{KT}}/T_{\text{KT}}$. Open symbols are the observed values of $\Delta f_c/T_c$ for two different ^4He coverages where data as a function of concentration are available.

Based on the QCM properties, the dashed line on Fig. 5 is the frequency shift that would be expected if *all* of the ^3He decoupled from the QCM. The solid line is a zero-intercept linear fit to the data. The data are consistent with nearly all of the ^3He decoupling from the hydrogen substrate. NMR studies of mixture films of comparable ^3He coverage on Nuclepore [17] show activation to the first excited state for $T \geq 250$ mK. For films with thickness of several total layers this excited state is predicted [18] to reside within the ^4He film. Thus, a transition from an excited state within the film to the ground state at the ^4He free surface as the temperature is reduced might decouple some ^3He to the free surface where it should be more mobile. A temperature dependent occupation of an excited state in the film might result in a broadening of the transition, T_c . The plausibility of this scenario is enhanced by the fact that NMR experiments have shown that an excited state for the ^3He persists to coverages thin enough so that only the inert ^4He is present [19]. Perhaps the ^3He phase separates or otherwise displaces normal-state ^4He from the substrate, thus allowing more ^4He to participate in the superfluidity. This could happen if the ground state for the ^3He were adjacent to the hydrogen with the first excited state at the ^4He surface. Theoretical work [20] predicts a ^3He substrate state for mixture films on H_2 . If this is the ground state here, the linear fit to the data in Fig. 5 indicates that 1.5 ^3He atoms are required to displace each ^4He atom from the H_2 substrate, a number that seems large. In response to these data, Saslow [21] has recently suggested that our observations may be explained by a proximity effect by which the ^4He layer closest to the substrate undergoes a Kosterlitz-

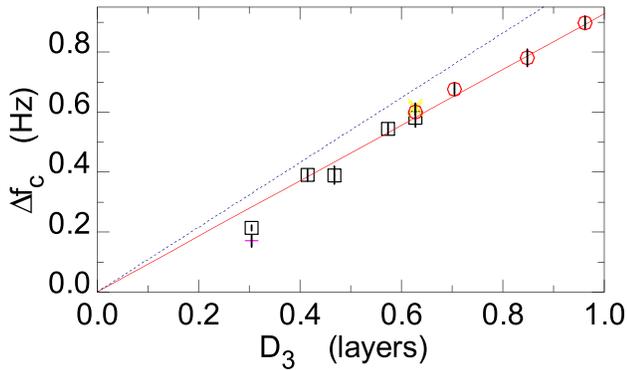


FIG. 5 (color online). Frequency shift at T_c versus ^3He coverage. The dashed line corresponds to complete decoupling of the ^3He . The solid line is a zero-intercept linear fit to the data. The symbols represent ^4He coverage as indicated in the legend of Fig. 2.

Thouless transition and for higher coverages additional ^4He joins the superfluid state. He supports his suggestion with a Ginsberg-Landau theory. A superfluid transition for 2D ^3He has been predicted for mixture films for $T \leq 10$ mK or in the presence of high magnetic fields [22], conditions not met by the present experiments. It is possible that some change may take place in the H_2 substrate, but since decoupling at T_c does not occur in the case of pure ^4He , we doubt that a change in the H_2 substrate dynamics [23] is involved.

In conclusion, we have studied ^4He and ^3He - ^4He mixture films with a zero temperature inert layer of 0.60 monolayers. A mass decoupling of the mixture film from the substrate occurs in addition to the KT transition, with $T_c < T_{KT}$. Curiously, the size of this feature is consistent with all of the ^3He decoupling from the substrate. At present we have no clear explanation for this unexpected decoupling.

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 [21] W. Saslow (private communication). This work makes three hypotheses: (1) In the presence of ^3He , there are two distinct but coupled ^4He order parameters, one for a layer near the substrate and one above it, and a coupling that permits vortices to coexist in both layers. (2) The approach of T_c to T_{KT} as $D_4 \geq 1$ suggests a proximity effect coupling between the layers. (3) When the upper layer softens enough to signal the KT transition, the common vortices nevertheless have the lower layer's relative large stiffness, thus preventing the vortex-unbinding occurring at a KT transition.
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