Very Thin Films of ^{3}He: A New Phase?

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We report the first measurements of the superfluid density of thin ^{3}He films for a broad range of thicknesses. In contrast to previous experiments, we find good agreement with the predictions of the general theory for the suppression of the superfluid transition temperature T_c. In addition, we find evidence for a new phase of the superfluid in films thinner than 2750 Å.

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Superfluid ^{3}He has long been a testing ground for our understanding of systems in which the Cooper pairs have a nonzero orbital angular momentum, including perhaps heavy-fermion and high-T_c superconductors. An area of great current interest involves the effects of confining such systems to lengths approaching the coherence length $\xi(T)$. In this case surface scattering becomes important. It has long been known that $l>0$ Cooper pairs are broken by any type of diffuse scattering,[1] leading to complete suppression of the transition temperature as the dimensions approach $\xi(T)$.[2]

A second effect which is predicted to arise due to walls is the stabilization of phases of the superfluid different from those present in the bulk.[3-7] For the slab geometry in the weak-coupling limit, it is predicted that the A and planar phases are degenerate for film thicknesses t such that the reduced film thickness, $w=t/\xi(T)$, is less than 7 (Refs. 6 and 7) and that the B phase will be stable for thicker films. We use the same definitions and values as appear in Ref. 7. A number of possible two-dimensional phases have also been suggested for sufficiently thin films.[8-10]

Several experimental studies on confined ^{3}He have been reported in the relatively simple slab or pore geometries which are most easily compared to theory.[11-17] Measurements to date on the suppression of T_c are generally in poor quantitative agreement with the theoretical predictions of Kjäldman, Kurkijärvi, and Rainer. Only two experiments on flow through relatively large pores have found good agreement with the predicted suppressions.[11,12] While quantitative agreement is generally lacking, a number of groups[13-16] have found qualitative agreement with the general prediction that the transition should be strongly suppressed as the film thickness is reduced.

The only definite identification of the phase of superfluid ^{3}He in a thin film comes from the work of Freeman *et al.*[17] They found the A phase in their 0.3-μm slab for pressures between 1.5 and 22 bars at all temperatures. They attributed the absence of a phase transition from the A phase into the B phase to supercooling effects. Evidence for a phase transition in 0-bar films was also presented by Harrison *et al.*[16] in thicker films based on a change in the critical current.

In order to further study these effects we have developed a new technique to grow metastable films of varying thickness on the copper disk of a torsional oscillator.[18] Our technique has a number of advantages. First, each film studied has a single relatively uniform thickness. Second, we obtain a direct determination of the average film thickness during a given measurement by monitoring the frequency of the oscillator just above the superfluid transition. Third, the shift in frequency below the transition is directly proportional to the superfluid density ρ_s. The measured frequency stability allows us to determine changes equivalent to a 10-Å-thick layer of film completely decoupling from the oscillator.

The experimental cell has been described elsewhere in detail.[19] Briefly, the cell, shown in the inset to Fig. 1, consists of a thin copper disk on which the film is grown. The disk hangs by a copper torsion rod from the top of a brass can and is independently thermally anchored to our copper nuclear-demagnetization stage. The brass can is mounted on a second torsion rod attached to the copper stage. The cell is driven by electrodes on the brass can in the mode in which the can and copper disk counterro-

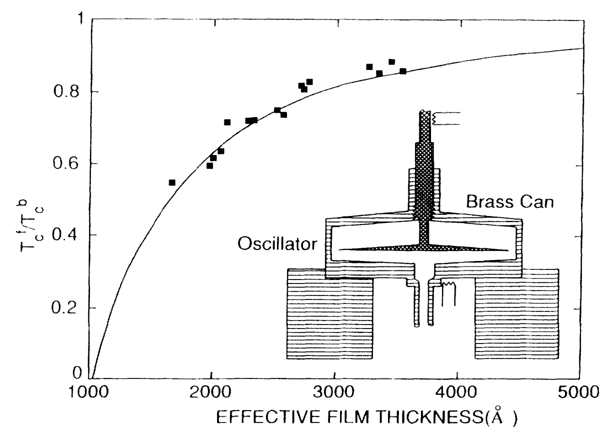

FIG. 1. The reduced transition temperature (T_c^f/T_c^b) for several effective film thicknesses. The solid line is the calculation of Kjäldman, Kurkijärvi, and Rainer for diffusely scattering walls. Inset: The experimental cell.

tate. This mode is a factor of 25 times more sensitive to liquid on the disk due to the ratio of the moments of inertia. No signature of the superfluid transition in the bulk liquid in the can was visible in this mode.

The cell was cooled by a copper nuclear-demagnetization stage to temperatures below 300 μK. Thermometry was provided by a standard lanthanum-doped cerium magnesium nitrate (LCMN) susceptibility thermometer calibrated against a melting-curve thermometer using the Greywall scale.[20] Typical time constants between the LCMN thermometer and the experimental cell were less than 3 min, indicating relatively good contact.

Films are grown at $\sim$250 mK by applying heat to a small well at the base of the brass can. This heats liquid in the well above the temperature of the copper disk causing it to transfer through the vapor to the copper disk. Once the desired film thickness is reached, as measured by the frequency shift of the oscillator, the cell is cooled down, trapping the film on the oscillator with a nonequilibrium thickness. Since the films are grown and annealed over the course of several hours while in equilibrium with the vapor, we believe them to be relatively uniform.

We found that our films would slowly thin if held well below the superfluid transition. Typically the film thickness would change on the order of 100 Å in a day. The rate was higher for thicker films and lower temperatures. The rate was also found to depend on how carefully the film was annealed when grown. The observed behavior leads us to believe that the liquid was leaving the oscillator by flowing up the torsion rod. With reasonable assumptions about the film thickness at higher points in our cell and about the superfluid fraction we can account for the observed rates of thinning.

For all films reported the drift amounted to less than a 1% change in the thickness while data were being taken. This drift was actually an advantage since it allowed us to study several thicknesses in a single demagnetization. The film thickness for the final film in a series was determined by the frequency shift observed as the film was removed at 100 mK. All other film thicknesses were determined by comparison of the frequencies just above the transition, giving a consistent set of thicknesses.

The surfaces of the copper disk were mechanically polished. Electron micrographs of similarly prepared pieces showed a generally smooth surface at the 500-Å level with approximately 5% of the surface covered with a network of 3000–5000-Å scratches. We believe these scratches simply filled with liquid due to surface-tension effects and had little effect on the films. We obtained an upper limit on the amount of liquid in these scratches by allowing the film to grow or thin to its equilibrium value with the bulk liquid a known distance below the oscillator. The observed equilibrium thickness of 400 Å was found to be very close to that expected assuming no liquid in the scratches. Allowing for the uncertainty in predictions of film thickness based on height above the free surface we conclude that less than the equivalent of 100 Å of film is contained in the scratches. This would lead to a possible systematic overestimation of our film thickness by that amount.

A smooth, featureless, temperature-dependent frequency background similar to that seen in other oscillators was measured with a 600-Å film present and subtracted from the data. Such a film is below the critical thickness and should not contribute to the background. On the other hand, this procedure should remove the small effects of any transition in the scratches.

After subtraction of the background, the frequency of the oscillator should be constant. The onset of superfluidity is indicated by a shift to higher frequency as liquid decouples from the oscillator. Shown in Fig. 1 are representative data from our cell of the superfluid transition temperature in the film T_c^f normalized to the bulk transition temperature as a function of the effective film thickness t which is twice the measured film thickness to account for the expected specularity of the free surface.[14] The solid line is from the theoretical work of Kjäldman, Kurkijärvi, and Rainer which should be valid for all temperatures and thicknesses. As can be seen, our results are the first which find good quantitative agreement with the theoretical predictions over a broad range of film thicknesses. We believe this is due to our having a single uniform thickness, the value of which is directly determined during the measurement. We stress that no adjustable parameters enter in this comparison.

We show in Fig. 2 measurements of the reduced superfluid density ρ_s/ρ for several different film thicknesses. As expected, ρ_s is strongly suppressed for our films. The value of ρ_s/ρ is determined from the frequency shift of the oscillator below T_c, $\Delta f(T)$, normalized to the total shift due to the film, Δf_{tot},

$$\frac{\rho_s}{\rho}=\frac{\Delta f(T)}{\Delta f_{\text{tot}}}\frac{1}{1-\chi}\,.$$

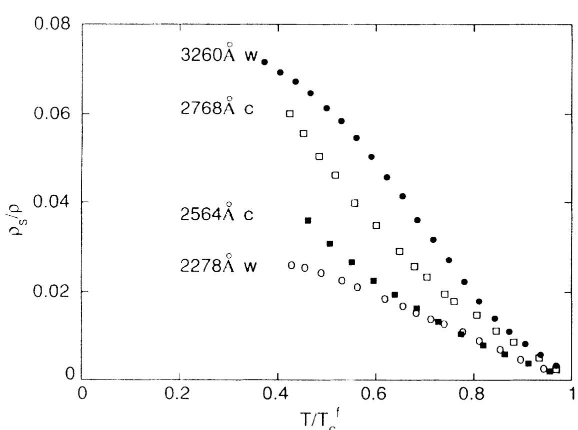

FIG. 2. The measured superfluid density normalized to the total ^{3}He density as a function of the reduced temperature for the effective film thicknesses listed.

The factor of χ accounts for any obstructions in the flow path.[21]

By studying ^{4}He in the same cell it is possible to obtain a value for χ. It is not clear to us, however, that this technique is correct due to the large difference in coherence lengths. An obstruction with a physical width of 0.1 μm, for example, would have an effective width of 0.2 μm for a ^{3}He film at $T=p=0$. As noted in Ref. 17 this will also cause χ to be temperature and pressure dependent. Because of this uncertainty, we choose to present our data with $\chi=0$. We note that a χ of 0.5 ± 0.1 would bring our data into reasonable agreement with the 1.5-bar data of Ref. 17.

At present the only theoretical predictions for the suppression of the superfluid density are done in the Ginzburg-Landau regime.[6,7] Unfortunately, none of our films are in this regime. Our thickest film at its T_c has $w=2.6$ whereas theory would predict a value of π (Refs. 6 and 7), making comparison impossible.

In order to detect possible phase transitions in our data, we plot in Fig. 3 $\rho_s/\rho_{s,\text{bulk}}$ as a function of w for the representative film thicknesses shown. Such a plot is expected to be universal in the Ginzburg-Landau regime. Surprisingly, as shown in Fig. 3, the data collapse roughly onto two curves, determined by whether the film thickness is above or below about 2750 Å. We stress that the normalization procedure is only weakly dependent on our choice of T_c. We have checked that reasonable variations in the choice of T_c do not affect the character of Fig. 3. We believe our data indicate that a transition to a new state of ^{3}He occurs at this critical film thickness.

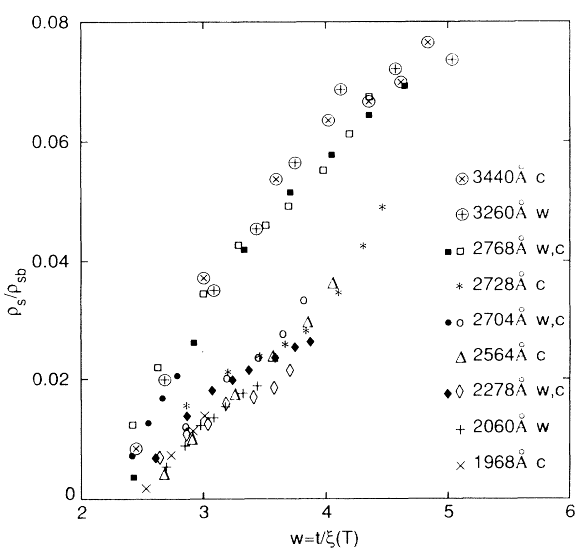

FIG. 3. The measured superfluid density normalized to the bulk superfluid density as a function of the reduced film thickness w. For each effective film thickness shown we have indicated whether the data were taken while warming w or cooling c.

Data for the 2768- and 2728-Å films were taken a few hours apart during the same demagnetization. The two sets of data clearly fall on different curves, indicating that this transition occurs over a very narrow range of thicknesses. A second interesting data set is from the 2704-Å film. In this case the film was cooled through the transition and left cold overnight during which the film thinned from $\sim$2900 to 2704 Å. The data taken on warming show the thick-film behavior even though the film at that point is thinner than the critical thickness. The same film was immediately cooled again and showed the typical behavior for a film below the critical thickness. This suggests that superheating and supercooling effects are important and may explain the absence of a transition between the curves.

In examining the literature we find possible supporting evidence for this transition in the data of Davis *et al.*[15] Their thinnest film has $T_c^f/T_c^b=0.82$ which is between the values for our 2768- and 2728-Å films. They found a significantly smaller slope for the critical current in that film compared to their thicker films. For the range of w covered in their data this result can be explained at least qualitatively by the transition we observe in ρ_s. The transition we observe is not evident in the data of Harrison *et al.*,[16] although that is possibly due to supercooling effects since they always started with very thick films.

The first possible explanation for our data would be a transition from the B phase to either the planar or the A phase, which is predicted to occur for $w<7$. Experimentally, Freeman *et al.*[17] have found the A phase for 3000-Å-thick films at 1.5 bars. Harrison *et al.*[16] observe a flow-rate transition which they take to be the expected B-to-A phase transition. While superthinning effects are important in their experiment, we may take the thickest film for which they observe a transition (6000 Å) as a lower limit on the thickness at which the equilibrium B-to-A phase transition occurs. Both the theoretical and experimental evidence indicates that the B-to-A phase transition occurs for much thicker films suggesting that the transition which we observe is into a new phase. We know of no theoretical work on possible stable phases for films such as ours far from the bulk T_c.

We have also considered the possibility that we are observing one of the several possible two-dimensional phases.[8-10] The relevant length scale is $\xi(T)$ and so we would expect to see a crossover from the lower curve to the upper curve at progressively lower temperatures for thinner films if this interpretation were correct.

Finally, we have considered the possibility that the effects we are observing are due to some form of thickness variation in our cell. If some region of the film were thinner, it might remain normal, cutting off part of the flow channel around the oscillator. We point out, however, that the relevant length is again $\xi(T)$ so we would expect to see this additional channel open up at progressively lower temperatures as the films are made thinner. No evidence for such a series of transitions is present in the data. If instead we had many such channels with different thicknesses, we would expect to see a gradual shift in the curves not a sharp change within a 40-Å range.

If the *A* phase is stable for our film, another possibility might be a domain wall between regions with opposite orientations of the *l* vector. These could be produced if slightly thicker regions first became superfluid independently or if we started in the phase described in Ref. 8. If this domain wall were somehow pinned, flow through it would presumably be reduced leading to a smaller measured superfluid density such as we see. It is hard to see, however, how such a domain wall could go from being unstable to stable over a 40-Å range since we might expect that the pinning force would only vary linearly with thickness.

To summarize, we present the first experimental data which find good agreement with the predicted suppression of the superfluid transition temperature for a broad range of film thicknesses. We also present the first data on the superfluid density as a function of film thickness for films thinner than 3000 Å. Finally, we have found evidence in our superfluid density data for a phase transition to a new state of the superfluid. This transition is found to be a sharp function of the thickness, occurring at 2750 ± 20 Å.

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