Critical Behavior of the Thermal Conductance of ^{3}He-^{4}He Mixture Films at the Kosterlitz-Thouless Transition

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We report measurements of the convective conductance of ^{3}He-^{4}He films near the superfluid transition. This thermal response is tested for the critical behavior observed in pure ^{4}He films. We find that this is preserved for mixtures. The parameters b and D/a^2, however, show a strong renormalization, and an unusual ^{3}He dependence. The conductance at fixed $T-T_c$ decreases upon addition of ^{3}He implying a decrease of the 2D correlation length. We observe a decrease in the maximum value of conductance which we attribute to a residual free-vortex density, even for $T<T_c$.

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The superfluid transition of ^{4}He films when physisorbed on a two-dimensional (2D) substrate can be described by the vortex-antivortex unbinding theory of Kosterlitz and Thouless.[1] Measurements which yield the superfluid density—third sound,[2–5] quartz microbalance,[6] and torsional oscillator[3,7]—as well as experiments on thermal transport,[8] persistent currents,[9] and vortex diffusivity[10] have provided tests for the static and dynamic aspects of this theory. A first series of measurements on aspects of 2D to 3D crossover and the universal character of the transition as a function of film thickness have also been reported.[11]

There is, in addition, in the case of liquid helium, the possibility to study the universal character of the transition in the presence of ^{3}He. Mixture films are, of course, the 2D analog of 3D mixtures; but, in fact, they are substantially richer in possibilities because of the important roles of the liquid-solid and liquid-vapor interfaces, and the arrangement of the ^{3}He relative to and along these interfaces. As regards the critical behavior, mixture films have been studied near the superfluid transition in several realizations: a submonolayer of ^{4}He and ^{3}He [12]; multilayer films of ^{4}He with a more substantial amount of ^{3}He, concentrations up to 50%[13,14]; and a near monolayer of ^{4}He with a large amount of ^{3}He.[7] All of these realizations are of interest because, among other effects, the boundary conditions on the superfluid are quite different. In the first case one is dealing with a monolayer mixture bounded by a solid substrate on one side and effectively vacuum on the other. In the second case, larger amounts of ^{3}He and ^{4}He, one is dealing with what one may call a "slab" of a 3D mixture with the twist that the concentration is not quite uniform because of the underlying van der Waals field. In the last case one has a submonolayer mixture "sandwiched" between the solid substrate and a 3D-like layer of ^{3}He. There are aspects of these realizations having to do with location of the ^{3}He and ^{4}He, and possible mass rearrangement as the temperature is changed. Many of these questions are still not settled, and the extent to which they affect the critical behavior at the superfluid transition is not clear.

The existing measurements with mixtures—mostly of the superfluid density—yield a picture of the transition which is qualitatively very similar to that of pure ^{4}He. The transition has the universal jump[13,14] in the superfluid density predicted for pure films by Nelson and Kosterlitz,[15] and an increase in dissipation due to the vortex-antivortex unbinding. Theoretical phase diagrams[16] are not reproduced particularly well,[14] but the general trend is that of a lower T_c with increased ^{3}He concentration. Torsional-oscillator measurements in particular show a less pronounced dissipation compared with pure films of the same T_c, and a higher residual dissipation for $T<T_c$.

We report in this Letter the first measurements of thermal transport for mixture films to probe more quantitatively the universal character of the superfluid transition. We have chosen for this study to look at rather dilute mixtures, $\lesssim 3\%$ of ^{3}He, or equivalently 0.1 layer of ^{3}He. In this limit, and with the thickness of the ^{4}He in the 12–16-Å range, the ^{3}He behaves as an impurity bound in 2D subbands defined by the ^{4}He film.[17] This low-temperature behavior, which is determined from measurements of specific heat, could very well be modified near the superfluid transition by the presence of vortices and the possible trapping of the ^{3}He into the vortex cores. The thermal transport near the transition takes place via the convective flow of liquid and refluxing gas. The effective conductance K has been shown[18] to be inversely proportional to the number of free vortices per unit area. It is a measure of the square of the 2D correlation length. Specifically, one may write

$$K=h^{-1}\frac{f(T)}{D/a^2}\exp\left(\frac{4\pi}{b}t^{-1/2}\right), \tag{1}$$

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where $t = T/T_c - 1$, h is the film thickness, $f(T)$ is a regular function of temperature involving the latent heat, D is the diffusion constant, a is the vortex core parameter, and b is a nonuniversal constant.

Measurements of thermal conductivity are quite informative. In the zero-power limit they yield most directly values of D/a^2 and b via Eq. (1). A study of these parameters as a function of ^{4}He thickness has been reported.[11] The measurements we report here have been done with the same experimental cell. This consists of two copper plates linked with a 3 m$\times$2.52 μm$\times$2.52 cm coiled ribbon of Mylar. The conductance is measured with a differential thermometer arrangement and a regulation thermometer at the top of the cell. With this design, we have demonstrated that the largest conductivity that one can measure is determined by the Kapitza resistance between the helium film and the copper. This limits the exponential divergence of the film's conductance. The smallest conductance, on the other hand, is due to the diffusive conduction through the vapor.

We show in Fig. 1 the measured conductance for two films of ^{4}He, 12.1 and 16 Å in thickness, along with a number of mixtures. The data are labeled by the percentage of ^{3}He in the liquid film. This number represents the portion of the ^{3}He which is not in the vapor. We measure this via an *in situ* gauge which registers the incremental pressure when ^{3}He is added. We note that if the ^{3}He were at the free surface (such as at $T=0$), then a concentration of 1% for the 12.1-Å film would correspond to a ^{3}He coverage of 1.41 μmole/m^2 or 0.044 atomic layer.[17] We point out several aspects of the data shown in Fig. 1: the rise associated with the transition, the shift in T_c, and the surprising decrease in the upper bound with increasing concentration.

The film's convective conduction, the critical behavior, can be extracted from these data by allowing for the limiting series and parallel conductances, the upper and lower bounds. This conduction, if universal, should obey Eq. (1). To test this we plot in Fig. 2 the conductance normalized by $f(T)h^{-1}$ against $t^{-1/2}$ on a semilog scale. It seems clear from the straight lines in this figure that the mixture data are consistent with this equation. The solid lines are least-squares fits of these data with D/a^2, b, and T_c as variational parameters. This, we believe, is the first test of mixture films to this equation. We note, in particular, the following behavior. For either of the ^{4}He films the conductance at fixed t tends to drop upon the addition of ^{3}He. This is true for both the quantity $Kh/f(T)$ plotted in Fig. 2, and K itself. Since we have that $K \sim \xi_{2D}^2$, this implies that the 2D correlation *decreases* upon addition of ^{3}He. This is the opposite of what is observed in 3D mixtures.[19]

In Fig. 3, we have plotted the parameters D/a^2 and b as functions of concentration. To emphasize the similarity of these parameters in their concentration dependence, we have shifted the values of b, and rescaled the values of D/a^2 for the 16-Å film to match the higher-concentration region of the 12.1-Å film.[20]

We see from Fig. 3 that both of these parameters undergo a rapid initial variation, within a concentration region of ~ 0.002, and then a somewhat less rapid variation beyond this. In order to understand this, we calculate the number of free vortices, n_f, present in a

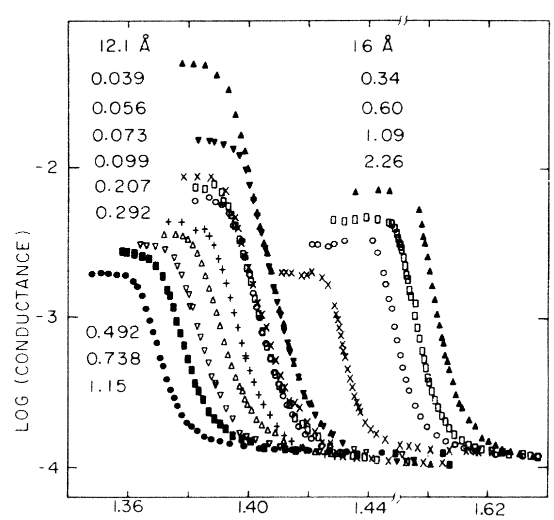

FIG. 1. The measured conductance in watts per kelvin for two films of ^{4}He. The numbers for each film indicate the percentage of ^{3}He for each successive concentration, with the highest curve for 12.1 Å being for pure ^{4}He.

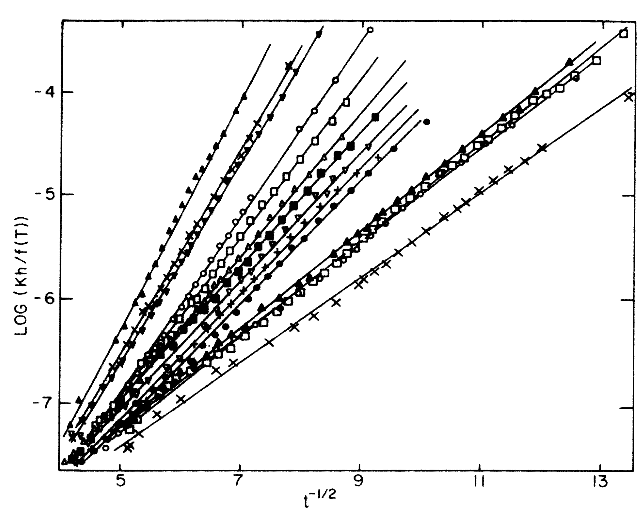

FIG. 2. The normalized conductance, $Kh/f(T)$, plotted vs $t^{-1/2}$ to test Eq. (1). The solid lines are least-squares fits.

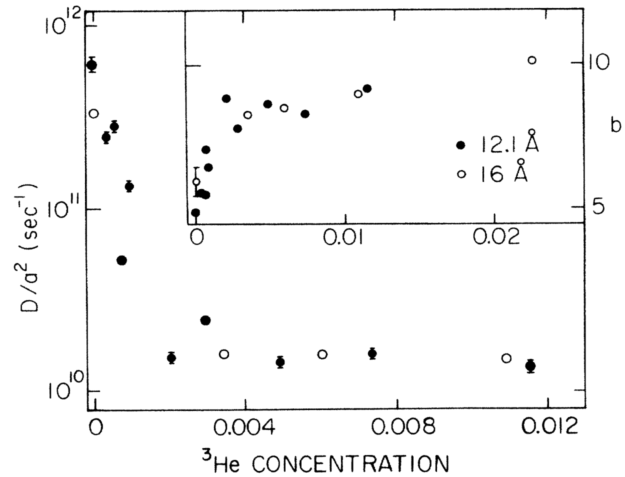

FIG. 3. Dependence of D/a^2 and b on ^{3}He concentration. The results for the 16-Å film have been rescaled (see Ref. 20).

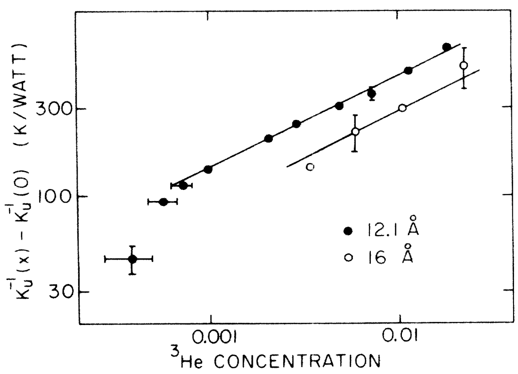

FIG. 4. The dependence of $K_u^{-1}(x)$ on concentration. See text. The solid lines have a slope of $\frac{1}{2}$.

pure ^{4}He film for $T > T_c$. This given by[21]

$$n_f = 2LlS_g k_B T m^2 / K W h^2 D, \tag{2}$$

where l and W are the length and width of the flowing film, L is the latent heat, and S_g is the entropy of the vapor. Using Eq. (2), a value of $a^2 = 2.2$ Å^2, and the experimental value of D/a^2, we calculate $n_f \sim 10^9$–10^{13} cm^{-2} in the region in which our data are analyzed. By comparison, the number density of ^{3}He in the range of concentrations studied is between 10^{12} and 3×10^{13} cm^{-2}. Thus, as we progress from low to high concentration the number of ^{3}He goes from being within the range of the native free-vortex density of the pure film to a value exceeding this. This crossover takes place between the data at $x = 0.0029$ and 0.0049. It is perhaps this crossover which is responsible for the two regions in the behavior of D/a^2 and b in Fig. 3.

There are two ways in which we see the ^{3}He affecting the behavior of the film. As a normal impurity it must affect the hydrodynamic flow associated with the vortices and hence the vortex-antivortex interaction. Further, it is known that ^{3}He—at least in bulk ^{4}He—resides preferentially at the vortex core.[22] This dressing up of the vortex should lower the diffusion constant and increase the core radius. This is consistent with our observation of a decrease in D/a^2. If we take the ^{3}He binding energy to a vortex as 3 K, the value appropriate for *bulk* vortices,[22] then we estimate that all but $\sim 6\%$ of the ^{3}He is trapped on vortex cores.[23]

Another result of our measurements is the lowering of the upper value of the conductance. We suggest that this is due to the *presence of free vortices even for* $T < T_c$. To see this, we note from Eq. (2) that $n_f D \propto K^{-1}$. For pure films n_f vanishes at T_c and K would diverge but for the limit imposed by the Kapitza resistance. In the case of mixtures, K is bounded by a value much lower than in pure films, $K_u(x)$. We interpret this as indicating that $n_f \neq 0$ even for $T < T_c$. To show the concentration dependence of $K_u^{-1}(x)$, we have plotted in Fig. 4 $K_u^{-1}(x) - K_u^{-1}(0)$ versus concentration on a log-log scale. By subtracting $K_u^{-1}(0)$, we are taking away the effect of Kapitza resistance. The solid lines in this figure describe these data well in the higher-concentration region. These lines have a slope of $\frac{1}{2}$; thus, $n_f D \sim x^{1/2}$. Since D is expected to *decrease* with the addition of ^{3}He, then n_f must increase even more rapidly than $x^{1/2}$. This He^3-induced free-vortex density applies to T below and near T_c.[24]

This suggestion, that the presence of ^{3}He results in a free-vortex density, is consistent with experiments on the decay of persistent currents. These data are taken for T much below T_c.[9] It is found that the addition of ^{3}He greatly enhances the decay. This is consistent with a greater density of free vortices in addition to those induced by finite velocity or perhaps depinning or other effects.[25,26]

Lastly, we mention our results of the shift in T_c with ^{3}He. We find that T_c shifts nonlinearly with the addition of ^{3}He, particularly in the low-concentration region. Indeed, it appears that T_c at first might *increase.* This has been observed previously by Smith *et al.*[12] and is something that we are exploring further. We do note, however, that the shift in T_c that we observe is at higher concentrations is not in disagreement with the results of Laheurte *et al.*[14] which are for concentrations greater than 10%. The result of these workers shows a more canonical, nearly linear shift.

In summary, we have reported new data for the thermal conductance of helium films which for the first time, in the case of mixtures, test the universal character of this response near the superfluid transition. We find that the sharp divergence observed for pure films is retained but with a strong renormalization of parameters within a relatively narrow region of concentration. We observe a lowering of the upper value of conductance which we interpret as due to the presence of free vortices even for $T < T_c$. We attribute this behavior to the presence of ^{3}He.

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[23]In making this estimate, we have assumed that the ^{3}He can move as a 2D gas (see Ref. 17) within the film, either trapped on a vortex or not. We have taken the ratio of the ^{3}He effective masses in these two states as 1.5.

[24]Note that a change in the superfluid density cannot account for the changes that we observe in the upper bound. This density changes by about 2% while the upper bound changes by a factor of ~ 40.

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