# Effect of $^3$He on Submonolayer Superfluidity

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We have studied the superfluid response of $^3$He-$^4$He mixture films adsorbed onto porous gold for a wide range of $^3$He and $^4$He coverages, focusing on submonolayer superfluidity. At $T = 0$, $^3$He appears to float on top of $^4$He and can be viewed as a second substrate that induces its own inert layer. Depending on the $^4$He content, the zero-temperature superfluid mass and the superfluid onset temperature either saturate or vanish with the addition of $^3$He. The $T = 0$ superfluid-insulator phase boundary, which can be described by a simple function, is found in the $^3$He-$^4$He coverage plane.

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The superfluid transition in pure $^4$He films adsorbed onto amorphous substrates is a subject of considerable theoretical [1] and experimental [2–6] interest. There have also been a number of experiments exploring the nature of superfluidity and ordering of $^3$He-$^4$He mixture films on such substrates [7–19]. In most of these studies, $^3$He ranging from a small fraction of a monolayer to multilayers was added to pure $^4$He films with superfluid coverage on the order of and exceeding 1 monolayer (the monolayer coverage is 12.9 $\mu$mol/m$^2$ for $^4$He and 10.6 $\mu$mol/m$^2$ for $^3$He). It was found that superfluidity is suppressed [7,8] with the addition of $^3$He and that in the $T = 0$ limit $^3$He tends to reside near the free surface of the film [10–14]. For multilayer $^3$He this configuration has been termed the "superfluid sandwich" [11].

An interesting question is how does $^3$He affect superfluidity of a $^4$He film when the total superfluid coverage is only a small fraction of a monolayer. In this Letter we report a systematic study on the suppression of superfluidity by up to 8.5 monolayers of $^3$He in $^4$He films with superfluid coverage ranging from 4% of a layer to 1 monolayer. For films with superfluid coverage of more than 0.40 monolayers of $^4$He we found that the superfluid fraction decays exponentially with the addition of $^3$He and superfluidity persists in the low temperature limit no matter how much $^3$He is added to the film. The measurements allow us to deduce quantitatively the $T = 0$ phase diagram.

The experimental cell is the same as the one used in Ref. [5]. The substrate is porous gold of 70% porosity consisting of multiply interconnected gold strands of 0.06 $\mu$m in diameter. After each new $^3$He and/or $^4$He dose, the adsorbed film is annealed at a temperature at which there is considerable vapor pressure (>0.1 torr), then it is cooled to the superfluid onset temperature $T_c$. This procedure yields consistent results [19]; by repeating this procedure several times without adding any new helium to the film, $T_c$ reproduces to within 0.5 mK.

The 12 different experimental runs were performed by keeping the $^4$He coverage $n_4$ constant and incrementally adding $^3$He to the mixture. Figure 1 shows a data set for $n_4 = 29.35$ $\mu$mol/m$^2$ with $n_{4s}$, the superfluid coverage in the absence of $^3$He, of 0.32 layers. The superfluid coverage, $n_{4s}$, is $n_4$ less the minimum coverage necessary for superfluidity $n_0 = 25.25$ $\mu$mol/m$^2$, often called the inert layer coverage. The areal superfluid density $\rho_s$, measured by torsional oscillator technique, has been corrected for the tortuosity [20] of the porous gold sample. The curve with the highest $T_c$ corresponds to the pure $^4$He film. One can determine $T_c$ by either extrapolating the superfluid density to zero or by locating the dissipation peak that accompanies the onset of superfluidity. For films with $T_c > 30$ mK, the values obtained by the two different methods are always within 6% of each other. For films with $T_c < 15$ mK we cannot reliably determine $T_c$ from the superfluid density data. Therefore we determine $T_c$ for all films from the dissipation signal.

Figure 2 shows the dependence of $T_c$ on the $^3$He coverage $n_3$. For films with $n_{4s} \geq 0.42$ monolayers, $T_c$ decreases rapidly then saturates with the addition of $^3$He. Such a behavior has been observed in earlier measurements [8–10]. Films showing this behavior are marked by filled symbols in Fig. 2. Films with lower starting $^4$He coverages, however, show a vanishing $T_c$ at a finite $n_3$,

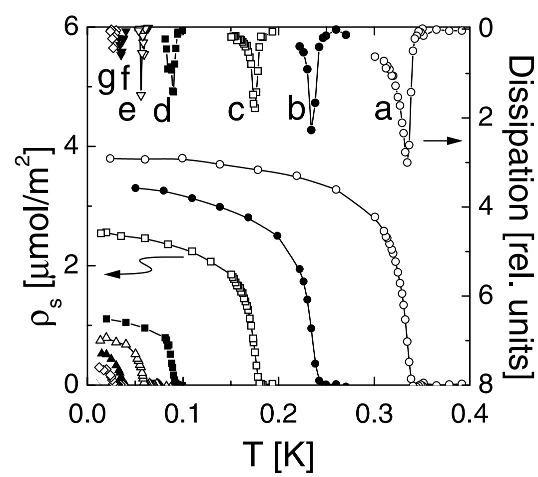


FIG. 1. Superfluid density and dissipation versus temperature for several mixture films having $n_4 = 29.35$ $\mu$mol/m$^2$. The $^3$He coverages for curves a–g are 0, 1.91, 3.83, 7.34, 9.15, 12.55, and 14.91 $\mu$mol/m$^2$. For clarity, the magnitude of dissipation for curves d–g has been doubled.

  

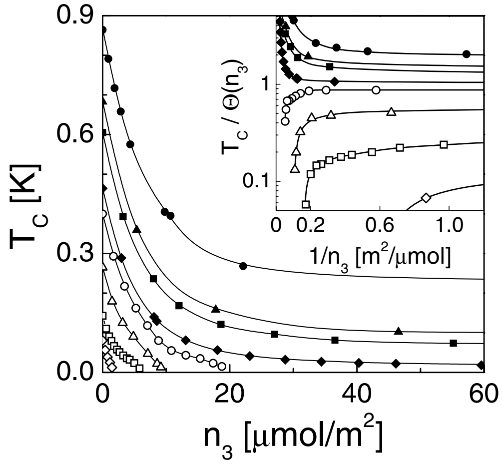


FIG. 2. Superfluid onset temperature versus $n_3$ for eight mixture films of different $n_4$. The starting superfluid coverages $n_{4s}$ at $n_3 = 0$ are 0.09, 0.16, 0.26, 0.37, 0.42, 0.55, 0.63, and 0.85 monolayers. Lines are guides to the eye and are anchored to data points of higher $^3$He coverages. The inset shows the same data renormalized by the critical curve $\Theta(n_3)$.

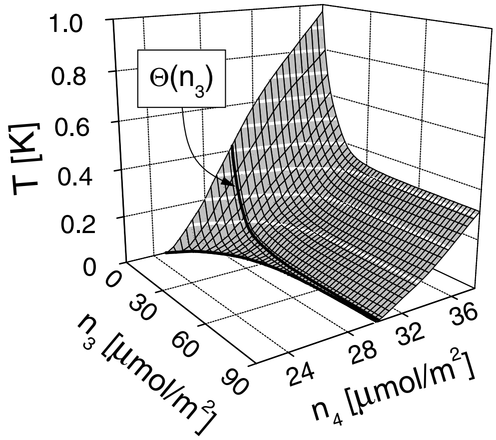


FIG. 3. The phase diagram in the $n_3$, $n_4$, $T$ space of $^3$He-$^4$He mixture films adsorbed onto porous gold. The onset sheet separates the superfluid (below) and the insulating (above) region. Note the disparity of scales; the range of $n_3$ is almost 10 times that of $n_4$. The $T = 0$ phase boundary and the critical curve $\Theta(n_3)$ are shown as thick lines.

as shown by curves with open symbols. For example, for the curve with $n_{4s} = 0.09$ and 0.26 layers, superfluidity is completely suppressed when $n_3$ exceeds 0.17 and 0.94 layers, respectively. A critical curve $\Theta(n_3)$ with $n_{4s}$ that is between 0.37 and 0.42 monolayers, which separates the two different classes of behavior, should exist. This curve will smoothly decrease with $n_3$ and extrapolate to $T_c = 0$ in the limit of a very large $n_3$. Since the curves with $n_{4s} = 0.37$ and 0.42 monolayers collapse quite well when vertically shifted, this critical curve $\Theta(n_3)$ can be approximated by subtracting from the $n_{4s} = 0.42$ layer curve 18 mK, the asymptotic $T_c$ value at large $n_3$. The critical curve has a total $^4$He coverage $n_4^{\text{crit}} = 30.4 \pm 0.2$ $\mu$mol/m$^2$ and a starting superfluid onset temperature $T_c = 430 \pm 20$ mK.

The qualitatively different behaviors at large $n_3$ are accentuated if we divide all curves of Fig. 2 by the critical curve $\Theta(n_3)$. The result, shown in the inset in Fig. 2, bears an interesting resemblance with that for amorphous superconducting Bi films [21]. This comparison illustrates that the ground state at $T = 0$ of mixtures with large $^3$He content and that of Bi films changes from insulating to superfluid with just a very small change in the parameter favoring superfluidity (i.e., $^4$He and Bi surface coverages).

Replotting our data in the $n_3$, $n_4$, $T$ parameter space, the complete phase diagram can be constructed. The onset of superfluidity at different $n_3$ and $n_4$ appears as a sheet in Fig. 3. This onset sheet separates the superfluid and insulating phases, the superfluid being stable below the sheet. The curve in the $n_3 = 0$ plane is the phase boundary for pure $^4$He films and it has been explored earlier [6]. For $T_c$ between 0.2 and 0.6 K, this phase boundary is linear. Below 0.2 K, however, there is a significant deviation toward a smaller $n_4$ from this linear behavior [2,6]. While the intercept based on data with $T_c \leq 0.12$ K of this curve with the $n_4$ axis is $n_0 = 25.25 \pm 0.1$ $\mu$mol/m$^2$, the linear region with $T_c > 0.2$ K extrapolates to zero at an apparent inert coverage $n_0^*$ that is larger than $n_0$ by a few percent of a monolayer.

The top panel in Fig. 4 shows the $T = 0$ superfluid density, $\rho_{s0} = \rho_s(T = 0)$, as a function of $n_3$. $\rho_{s0}$ is deduced by fitting $\rho_s(T)$ measured in the temperature range of $T < 0.6T_c$ to the form $\rho_s(T) = \rho_{s0} - aT^2$ [3]. A quantitative analysis of the result shows that the functional dependence of $\rho_{s0}$ on $n_3$ is identical for *all* data sets that have $n_{4s} \geq 0.42$ monolayers. The fits to these data sets (filled symbols) are given by

$$\rho_{s0}(n_3, n_4) = \rho_{s0}(n_3 = 0, n_4) - A[1 - \exp(-n_3/B)], \tag{1}$$

with $A = 5.1 \pm 0.2$ $\mu$mol/m$^2$, $B = 9.5 \pm 0.5$ $\mu$mol/m$^2$. Because of the limited range of $n_3$, it is not possible to get meaningful fits to vanishing data sets, i.e., for $n_{4s} \leq 0.37$ monolayers, shown with open symbols. Nevertheless Eq. (1), with identical parameters $A$ and $B$, appears to give a good description of these data sets. It is therefore reasonable to conclude that Eq. (1) provides a "universal" description of the depletion of superfluidity with $^3$He in the $T = 0$ limit. We note that while curves of Fig. 2 resemble those of Fig. 4, there is no simple exponential dependence of $T_c$ on $n_3$ that fits all data. This is the case because the solubility of $^3$He in $^4$He and $^4$He in $^3$He is highly temperature dependent [10,11].

The inset in the top panel in Fig. 4 shows that the $\rho_{s0}$ curves at different $n_3$, obtained by interpolating curves of Fig. 4, increase linearly with $n_4$. Previous studies [2–4] have established that for pure $^4$He films this dependence is linear when $n_{4s}$ exceeds a tenth of a layer. $\rho_{s0}$ was also found to be linear with $n_4$ for mixture films with $n_3 = 12$ layers [11]. The slope of this linear function is

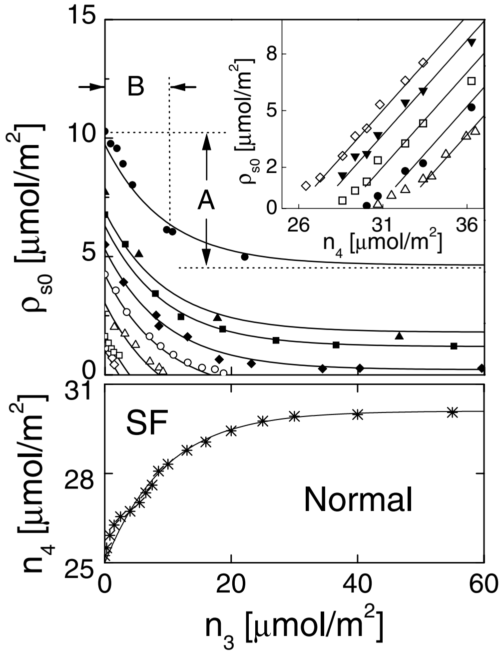


FIG. 4. The top panel shows the evolution with $n_3$ of $\rho_{s0}$ for mixture films of different $^4$He content. $^4$He coverages are the same as in Fig. 2. Solid lines have the same functional form given by Eq. (1). The inset shows $\rho_{s0}$ as a function of $n_4$ for $^3$He coverages, from left to right, of 0, 0.28, 0.75, 1.7, and 8.5 layers. The lower panel depicts the $T = 0$ phase boundary. The extrapolated data (stars) are well approximated by an exponential function (solid line), particularly for $n_3$ more than a monolayer.

the same as that for pure $^4$He films [11]. The inset in Fig. 4 shows that the same linear dependence of $\rho_{s0}$ versus $n_4$ with the same slope is found for all, including submonolayer, $^3$He coverages. This is a direct consequence of Eq. (1). Therefore, for submonolayer superfluid films, as in thicker films [10–12], there is no evidence of $^3$He dissolving into $^4$He at $T = 0$.

The parameter $A$ is the height of the exponential decays in Fig. 4 and its physical interpretation can be obtained by letting $n_3 \rightarrow \infty$. We find

$$A = \rho_{s0}(n_3 = 0) - \rho_{s0}(n_3 \rightarrow \infty), \tag{2}$$

which means that $A$ is the superfluid mass that will turn normal upon addition of a large $^3$He dose to a pure $^4$He film thicker than $n_4^{\mathrm{crit}}$. The depletion of superfluidity, i.e., the increase of the localized $^4$He induced by the $^3$He, is governed by the exponential function $\exp(-n_3/B)$. $B$ is nearly a monolayer of $^3$He.

The intersection of the superfluid onset sheet with the $T = 0$ plane separates the superfluid and insulating phases in the $n_3$-$n_4$ plane. Stars in the lower panel in Fig. 4 show this phase boundary as obtained by interpolating the $T_c$ versus $n_3$ curves in Fig. 2 and then extrapolating the results to $T = 0$ for all $n_3$. For large $^3$He doses this phase boundary approaches $n_4^{\mathrm{crit}}$ asymptotically. To quantitatively obtain the $T = 0$ phase boundary one should solve either $T_c(n_3, n_4) = 0$ or $\rho_{s0}(n_3, n_4) = 0$. Using Eq. (1), the solution for the latter equation can be obtained only if we know the explicit dependence of $\rho_{s0}(n_3 = 0, n_4)$ on $n_4$. Our measurement cannot resolve the deviation of $\rho_{s0}(n_3 = 0, n_4)$ from linearity described above. We adopt the simplest (linear) approximation $\rho_{s0}(n_3 = 0, n_4) \simeq n_4 - n_0^*$. With this approximation, Eq. (1) yields

$$n_4 \simeq n_0^* + A[1 - \exp(-n_3/B)]. \tag{3}$$

The approximation used for $\rho_{s0}$ overestimates the inert coverage. We can correct for this by replacing $n_0^*$ with $n_0$ in Eq. (3), which is equivalent to a 0.25 $\mu$mol/m$^2$ shift of the $n_4$ scale. Such a shift is within the uncertainty of our measurements. The resulting curve is shown in the lower panel in Fig. 4 as a solid line. The important conclusion is that the $T = 0$ phase boundary mirrors the exponential decay of $\rho_{s0}$ from Eq. (1). While the shifted Eq. (3) describes well the extrapolated data for large $n_3$, at low $n_3$ there is a significant discrepancy. The phase boundary shows a step near $n_3 = 6$ $\mu$mol/m$^2$. It is not clear if this feature is related to those seen in the damping of third sound [15], magnetization [16], NMR relaxation times [17], and heat capacity [18] of mixture films at higher $^4$He coverages.

We found that the amount of localized $^4$He increases from 25.25 $\mu$mol/m$^2$ with no $^3$He in the film and saturates exponentially at 30.4 $\mu$mol/m$^2$ when a very large amount of $^3$He, up to 8.5 layers, is added to the film. The reason for the saturation is that $^3$He atoms farther (i.e., beyond the first monolayer) from the $^3$He-$^4$He interface are expected to have a diminishing effect in localizing the superfluid. The difference of 5.15 $\mu$mol/m$^2$ is identical within error to the value of $A$ obtained from Eq. (1). This number is within 13% of the previously measured value [11].

What is the microscopic configuration of a $^3$He-$^4$He mixture film in the $T = 0$ limit? Complete phase separation is found theoretically for an ideal two dimensional mixture film [22]. On a real substrate, however, where vertical displacement is allowed, the different zero point energies of $^3$He and $^4$He will separate the two isotopes in the van der Waals field perpendicular to the surface. Such a mechanism is indeed responsible for the superfluid sandwich model and it appears to be valid, according to the results summarized in Fig. 4, irrespective to the surface coverages of $^3$He and $^4$He.

In a recent study [6] of superfluid $^4$He films absorbed onto various substrates, no correlation was found between the inert coverage and the strength of the long range van der Waals tail of the $^4$He-substrate interaction potential. The inert coverages of 6.1, 8.7, 10.3, 19.4, 22.5, and 25.3 $\mu$mol/m$^2$ on, respectively, $H_2$, HD, $D_2$, Ne, Ar, and Au substrates, however, were found to scale with the well depths of the potentials. This suggests that the short range $^4$He-substrate forces play the crucial role in determining $n_0$. We think the $^3$He layer that resides at the free surface of the superfluid film can be viewed as a substrate which induces another inert layer that saturates at 5.1 $\mu$mol/m$^2$. If we approximate the well depth of the interaction of one $^4$He atom with 8.5 layers of $^3$He to be 8.7 K, the value calculated for one $^4$He atom and a semi-infinite space of $^4$He [23], then the inert coverage of 5.1 $\mu$mol/m$^2$ on $^3$He follows the trend found for the aforementioned substrates. This suggests that the mechanism of inducing the non-superfluid or inert $^4$He close to the $^3$He overlayer at $T = 0$ is similar to that on an amorphous solid substrate. We note that an inert layer that is less than the monolayer coverage, like that on $H_2$ and $^3$He, is clearly a phenomenological concept. It is a measure of the localization effect of the substrate on a superfluid film.

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