Effect of $^3$He on Superfluid $^4$He Films Adsorbed on Grafoil

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Abstract. We have studied an effect of $^3$He on superfluid $^4$He films adsorbed on exfoliated graphite (Grafoil) using a quartz-crystal microbalance (QCM) technique. It is known that zero-temperature superfluid mass and superfluid onset temperature of $^4$He films adsorbed on porous gold substrate are suppressed, or even eliminated with the addition of $^3$He. We found that the superfluid onset temperature of $^4$He films on graphite decreases with the addition of $^3$He. However, the superfluid mass does not increase monotonically with decreasing temperature; it has a peak and is suppressed below a certain temperature, which suggests a phase separation of the film at low temperatures.

1. Introduction

The superfluidity of $^4$He films is an interesting subject. In particular, the depletion of the superfluidity by adding $^3$He has been reported for various substrates. $^3$He tends to reside near the free surface of the film at low temperatures. From the specific heat measurement, Bhattacharyya and Gasparini have reported that in low areal densities of $^3$He at low temperatures, $^3$He on thin $^4$He films formed on Nuclerpore undergoes a transition from a dilute, uniform density phase, to a denser, degenerate phase [1]. For thicker $^4$He films and higher $^3$He areal densities, the $^3$He remains homogeneously spread out over the free surface of the film.

Csáthy and Chan have carried out torsional oscillator measurements for $^3$He-$^4$He mixture film formed on porous gold. They reported that the superfluid onset temperature $T_C$ decreases with the addition of $^3$He. The superfluid mass shows the $T^2$-dependence at low temperature and its value at 0 K is decreased smoothly with increasing $^3$He as $\rho_s(n_3, n_4) = \rho_s(n_3 = 0, n_4) - A[1 - \exp(-n_3/B)]$, where $n_3$ and $n_4$ are the areal densities of $^3$He and $^4$He, $A$ and $B$ are some constants which does not depend both on $n_3$ and on $n_4[2]$. For the mixture films formed on Mylar sheet, McQueeney et al. observed the universal jump of superfluid mass at $T_C$, which corresponds to the Kosterlitz-Thouless (KT) signature. In addition, the superfluid mass increases monotonously with decreasing temperature reflecting solubility of the mixture [3].

In this paper, we report a novel depletion of superfluidity of $^4$He film formed on exfoliated graphite (Grafoil) by adding $^3$He. We have carried out quartz-crystal microbalance (QCM) experiments. All the data has been taken for a fixed areal density of $^4$He (four-atom thick) with changing addition of $^3$He from zero to 3.8 atoms/nm$^2$. 

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2. Experimental procedure

The experimental procedure has been reported in detail elsewhere [4]; here, we describe it briefly. In our experiments, the resonator is a 5.0 MHz AT-cut quartz crystal. Grafoil was first baked in 5 × 10⁻⁵ Pa at 900 °C for 3 h, and a 300 Å thick film of Ag was deposited onto it. To bond Grafoil on the Ag electrode, the crystal and Ag-plated Grafoil were pressed together and were heated in 1 × 10⁻⁵ Pa at 350 °C for 2 h. After bonding, the excess amount of Grafoil was removed. To retain good thermal contact, the crystal was fixed to the metal holder with electrically conductive adhesive. The Q value of the crystal was better than 10⁴. After being heated in 2 × 10⁻⁶ Pa at 130 °C for 5 h, the crystal was mounted in the sample cell. During transport it was briefly (one minute or less) exposed to air. Then, the sample cell was evacuated and cooled down to 4.2 K. To minimize the effect of desorption, Grafoil disks were put on the bottom of the cell.

The graphite crystallites of Grafoil are oriented with their basal planes parallel to the lateral oscillation. The full width at half maximum of the c-axis distribution is 12°, as determined by x-ray scattering. From the specific surface area of 15.4 m²/g, and the change in the resonance frequency from bare Ag to Grafoil/Ag electrodes, the sensitivity for the mass loading of ⁴He is estimated to be 4.2 Hz/atoms/nm². The resonance frequency and Q value were measured using a transmission circuit. In the circuit, the quartz crystal was placed in series with a coaxial line connecting a 50 Ω cw signal generator and an RF lock-in amplifier. The frequency of the signal generator was then controlled in order to keep the in-phase signal zero, and was locked to the resonance frequency. The quadrature signal at this frequency is the resonance amplitude, and the change in the Q value is calculated from this amplitude.
3. Results and discussion
The experiment was performed by keeping the areal density of $^4$He constant as 33 atoms/nm$^2$, which corresponds to four-atom thick film. After a dose of $^3$He was admitted to the cell, it was annealed at 1.5 K to ensure that the film was uniform over the entire substrate, and we took 10 h to cool down the cell. Figure 1 shows the changes in the resonance frequency and $Q$-value for several amounts of $^3$He. The oscillation amplitude of the quartz crystal is 0.07 nm. In the case of pure $^4$He, the resonance frequency increases rapidly at around 0.9 K, which is the KT signature. In the present experiments, on the contrary to the torsional oscillator measurements, the dissipation remains large at low temperatures below the increase in resonance frequency. We hence determine $T_C$ from the cusp of the dissipation signal. With the addition of $^3$He, $T_C$ shifts to lower temperature, and reaches 0.5 K for 3.8 atoms/nm$^2$ of $^3$He.

Figure 2 shows $T_C$ as a function of the addition of $^3$He, together with $T_C$ observed for the $^3$He-$^4$He mixture films on porous gold [2]. $T_C$ is suppressed rapidly in small amounts of $^3$He and shows a gentle decrease afterwards. As shown in the figure, the observed depletion of $T_C$ is in good agreement with the mixture films on porous gold. In addition, as shown in Fig. 1, the resonance frequency shows the rapid increase at $T_C$ with the increase in dissipation. The increase

![Figure 2](image-url)  
**Figure 2.** Areal density of $^3$He versus $T_C$. Squares and circles correspond to the present measurements with different oscillation amplitudes for $^3$He/$^4$He/Gr. Triangles to $^3$He-$^4$He mixture films formed on porous gold [2].

![Figure 3](image-url)  
**Figure 3.** Change in resonance frequency normalized by $\Delta f_{res,0}$ as a function of $T/T_C$ for $^3$He/$^4$He/Gr. $\Delta f_{res,0}$ is the frequency shift of KT mass-jump prediction. The areal density of $^4$He is 33 atoms/nm$^2$. The numbers in the figure represent $^3$He areal densities in the unit of atoms/nm$^2$. 

![Diagram](image-url)
in the frequency is proportional to $T_C$ being consistent with the KT mass-jump prediction, except for 3.8 atoms/nm$^2$ of $^3$He. These features of the superfluid onset are very similar to those in the measurements for porous gold and for Mylar sheet, at least below 2.8 atoms/nm$^2$ of $^3$He [2,3]. Then, we can conclude that for the mixture films formed on Grafoil, the mechanism of the depletion of $T_C$ is the same as porous gold and Mylar sheet. This similarity also means that the most amount of $^3$He condenses until the cell is cooled down to $T_C$.

In the low temperature region, however, the temperature dependence of the superfluid mass for Grafoil is different from that of the other samples. Figure 3 shows the change in resonance frequency normalized by $\Delta \text{freq}_S$ as a function of $T/T_C$, where $\Delta \text{freq}_S$ is the frequency shift of KT mass-jump prediction. As mentioned above, the rapid increase of the frequency below $T_C$ coincides with each other, which is the KT signature. For porous gold and Mylar sheet, the frequency increases monotonously as the temperature is lowered. On the other hand, for Grafoil, it has a peak and decreases below a certain temperature. This suppression becomes remarkable with more addition of $^3$He. Above 1.3 atoms/nm$^2$ of $^3$He, it tends to zero at around 0.3 K. The most remarkable case is of 3.8 atoms/nm$^2$; the suppression is strong so that it ruins the KT mass jump.

These observations mean that the mechanism of the suppression of superfluidity in the low temperature region is different from the depletion of $T_C$. We ascribe that suppression in the low temperature region to a phase separation. Excess pressure by $^3$He overlayer on $^4$He films is, however, at most 0.3 MPa, so that it is not enough high to suppress the superfluid density as we observed. The suppression may be explained as the following scenario: It is known that Grafoil is composed of small graphite crystalline platelets of which sizes are typically about $\ell = 10$ nm in diameter. In the present experiments, the connectivity of the platelets for the superfluid is poor and $\chi$ factor is about 0.94. We speculate that the weak connectivity is easily blocked by the separated $^3$He condensed between the platelets, therefore the superfluidity is suppressed in the low temperature region.

4. Summary
We have carried out QCM experiments for $^3$He-$^4$He mixture film formed on Grafoil. It is found that $T_C$ is suppressed with the addition of $^3$He and that the KT signature of the superfluid onset remains at least below 2.8 atoms/nm$^2$. However, the superfluid mass does not increase monotonically with decreasing temperature and tends to zero around 0.3 K above 1.3 atoms/nm$^2$. This suggests a phase separation of the film at low temperatures.

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