Decoupling of Solid $^4$He Layers under the Superfluid Overlayer

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Abstract.
It has been reported that in a large oscillation amplitude, the mass decoupling of multilayer $^4$He films adsorbed on graphite results from the depinning of the second solid atomic layer. This decoupling suddenly vanishes below a certain low temperature $T_D$ due to the cancellation of mass decoupling by the superfluid countercflow of the the overlayer. We studied the relaxation of the depinned state at various temperatures, after reduction of oscillation amplitude below $T_D$. It was found that above the superfluid transition temperature the mass decoupling revives with a relaxation time of several 100 s. It strongly supports that the depinned state of the second solid atomic layer remains underneath the superfluid overlayer.

1. Introduction
An atomically thin films adsorbed on a solid substrate exhibit various unique properties reflecting the geometry of the substrate, and provide a number of attractive topics in physics for many years. Recently, the nanofriction is added to this study. Krim and co-workers performed quartz crystal microbalance (QCM) measurements for several physisorbed films on metal substrates and found that the films undergo partial decoupling from the oscillating substrate.[1] Mistura and co-workers also observed this decoupling on metal substrates, and reported the pinning-depinning transition against the driving force of oscillating substrates.[2] They found that the decoupling state remains down to a lower driving force than the depinning transition. It indicates that the decoupling state is metastable below the depinning transition. It indicates that the decoupling state is metastable below the depinning transition.

$^4$He film on graphite ($^4$He/Gr) is an ideal system for a study on decoupling mechanism, since the interaction between the adatom and the substrate is so small that the decoupling is expected to occur at a small driving force. In addition, $^4$He/Gr shows a layer-by-layer growth up to the five-atomic layers. We have studied multi-layer $^4$He films on graphite using the QCM technique. In $^4$He/Gr, we observed that the two- and three-atom-thick films start to decouple below a certain temperature $T_S$ at an oscillation amplitude larger than $A_S$.[3, 4] Furthermore, we confirmed that the decoupling state is metastable; after keeping an oscillation amplitude larger than $A_S$, we switched a sufficiently lower oscillation amplitude than $A_S$, and found that the decoupling state remains with a long relaxation time, which reaches several thousands seconds at low temperatures.

Figures 1(a) and (b) show our naive picture about the observations. In $^4$He/Gr, the areal density of the second solid atomic layer is smaller than that of the first solid atomic layer, which...
Figure 1. Picture of decoupling in the two- and three-atom-thick films on graphite. The bars show the lattice of the first solid atomic layer. The circles and the colored areas show $^4$He atoms and domains in the second solid atomic layer, respectively. (a) Stable structure where $^4$He atoms in the second solid atomic layer are locked to stable positions at the oscillation amplitude below $A_S$. (b) Disordered structure where $^4$He atoms are depinned from the stable positions at the oscillation amplitude larger than $A_S$. After the oscillation amplitude is switched to that below $A_S$, this remains with a long relaxation time at low temperatures.

causes edge dislocations between the layers. At an oscillation amplitude below $A_S$, $^4$He atoms in the second atomic layer cannot overcome the potential barrier owing to the first solid atomic layer. Thus, $^4$He atoms in the second atomic layer are locked to stable positions, as shown in Fig. 1(a). When the oscillation amplitude is larger than $A_S$, i.e. when the driving force is large enough to overcome the barrier, the $^4$He atoms in the second atomic layer are depinned from the stable positions, and forms a disordered (domain) structure, as shown in Fig. 1(b), resulting in the decoupling of the second solid atomic layer from the oscillation of the first solid atomic layer. After the oscillation amplitude is switched to that below $A_S$, the depinned $^4$He atoms in the second atomic layer relaxes to the stable positions with a certain relaxation time.

We have also performed QCM measurements on four-atom-thick films, where the third- and the four-atomic layers become a superfluid below the transition temperature $T_C$. We found that the decoupling suddenly vanishes below a certain temperature $T_D$, which is lower than $T_C$. This vanishment is explained by the cancellation of mass decoupling of the second atomic layer by the superfluid counterflow of the overlayer.[5] This explanation indicates that the $^4$He atoms in the second solid atomic layer is still in the depinned state, even though the entire mass decoupling vanishes below $T_D$.

It is of interest to clarify whether or not, the depinned state of the second atomic layer remains underneath the superfluid overlayer. Thus motivated, we studied the relaxation of the depinned state of the second solid atomic layer by developing a new procedure. In this paper, we report how we detect the relaxation, and then discuss the relaxation of the depinned state.

2. Experiment

We used the QCM technique to measure the mass decoupling. In the QCM technique, the coupled mass to the oscillating substrate is obtained from the change in the resonance frequency $\Delta f$ as

$$\frac{\Delta f}{f} = -\frac{m}{M}, \tag{1}$$

where $m$ is the coupled mass density of film, $M$ is the areal density of the crystal, and $f$ is the resonance frequency. When the film is decoupled from the oscillation, the coupled mass
Figure 2. Experimental procedure. (a) First, in a large amplitude of 0.25 nm, the film was slowly cooled down from 1.0 K to 0.20 K (red). (b) At 0.20 K, the amplitude was switched to a small amplitude of 0.025 nm. (c) Then, the film was quickly warmed up to $T_m$ and (d) was kept at this temperature to measure the change in resonance frequency (blue).

In the present experiments, the resonator is a 5.0 MHz AT-cut quartz crystal. The crystal was commercially available, and no special treatment was applied to the Ag electrode. At first, Grafoil (exfoliated graphite) was baked in a vacuum at 900°C for 3 h, and a 30-nm-thick film of Ag was deposited onto it. The crystal and Ag-plated Grafoil were pressed together and were heated in a vacuum at 350°C for 2 h. Then, Grafoil was bonded on both sides of the Ag electrode. After bonding, the excess amount of Grafoil was removed. To keep good thermal contact, the crystal was fixed to the metal holder with electrically conductive adhesive. After these processes, the $Q$ value of the crystal remained better than $10^4$, and the areal density of Grafoil was 7.30 g/m$^2$. After being heated in $2 \times 10^{-6}$ Pa at 130°C for 5 h, the crystal was mounted in the sample cell.

The resonance frequency was 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 a RF lock-in amplifier. The frequency of the signal generator was then controlled in order to keep the inphase output zero, and was locked to the resonance frequency. The quadrature output at this frequency is the resonance amplitude.

We prepared the four-atom-thick film of 32.0 atoms/nm$^2$ with $T_C$ of ~ 0.5 K, and carried out relaxation experiments with the following procedure as shown in Fig. 2:

(a) The film was slowly cooled down from 1.0 K to 0.20 K in a large amplitude of 0.25 nm. The resonance frequency increases below $T_S$ of 0.85 K, i.e. the mass decoupling of solid layers occurs. Then, this frequency is suddenly dropped at $T_D$ of 0.23 K, i.e. the entire mass decoupling vanishes.

(b) At 0.20 K (below $T_D$), the amplitude was switched to a small amplitude of 0.025 nm.

(c) The film was quickly warmed up to the setting temperature of $T_m$ at the warming rate of 0.03 K/min.[$6$]

(d) At $T_m$, the change in resonance frequency was measured for 2 h.
Figure 3. Change in the resonance frequency against waiting time at $T_m$. The resonance frequency decreases slowly at the beginning above 0.5 K, while it does not change greatly below 0.4 K.

Then, the film is warmed up to 1.0 K, and the procedure of (a)-(d) was repeated for different $T_m$. If the depinned state of the second solid atomic layer remains underneath the superfluid overlayer, the decoupling is expected to revive at $T_m > T_C$ on the condition that the warming time is sufficiently shorter than the relaxation time of the depinned state.

3. Results and Discussion

Figure 3 shows the change in the resonance frequency for several $T_m$. Below 0.4 K ($< T_C$), the resonance frequency does not change greatly. In contrast, above $T_C \sim 0.5$ K the resonance frequency decreases at the beginning of waiting time, with a relaxation time of about several 100 s.[7] The observations clearly show that the decoupling revives above $T_C$, due to the lack of the cancellation by the superfluid counterflow. It means that the depinned state of the second solid atomic layer remains underneath the superfluid overlayer.

Next we focus on the relaxation time of the depinned state after the temperature becomes constant. In Fig. 3, the relaxation time for $T_m = 0.7$ K is shorter than that for 0.6 K. This tendency is consistent with that observed in the three-atom-thick films.[8] In the three-atom-thick films, the relaxation time obeys the Arrhenius law, indicating that the existence of energy barrier between the stable and the disordered structure in the second solid atomic layer. It remains a future issue to examine the energy barrier for the four-atom-thick films and clarify how the energy barrier varies by increasing the superfluid overlayer.

4. Summary

We measured the mass decoupling after the reduction in amplitude below $T_D$, by developing a new procedure. We found that the mass decoupling revives above $T_C$. It strongly supports that the depinned state in the second layer remains below $T_D$ underneath the superfluid overlayer, even though the entire mass decoupling vanishes by the superfluid counterflow.
References
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[6] The frequency drop with increasing temperature from 0.2 to 0.5 K at the oscillation amplitude of 0.025 nm is not from the change in decoupling amount of $^4$He film, but from the change in background peculiar to this amplitude.
[7] The decoupling amount at the beginning of relaxation measurements (~3 Hz) is about 60% of that at the large oscillation amplitude just above $T_D$. This reduction is attributable to the relaxation during the warming time (800 s for $T_m = 0.6$ K).
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