Mechanical response of \(^4\)He films adsorbed on single-crystalline graphite

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Abstract. We carried out quartz crystal microbalance (QCM) experiments of a 32 kHz quartz tuning fork for \(^4\)He films adsorbed on exfoliated single-crystalline graphite, and measured the temperature dependence of the resonance frequency and \(Q\) value for various areal densities. To study the sliding direction dependence, two quartz tuning forks oscillating in the different crystal directions were prepared. It was found that the resonance frequency does not decrease greatly in the monolayer region, and the decoupling from the oscillating substrate is enhanced compared with Grafoil. We observed, however, a weak decrease in frequency around the commensurate phase of monolayer, and this decrease shows the sliding direction dependence.

1. Introduction
An atomically thin film on a solid substrate exhibits various unique properties, and has provided a number of attractive topics in physics for many years. Recently, nanotribology has been joined to the subjects of study on this film.

We have already reported quartz crystal microbalance (QCM) experiments with a 32 kHz quartz tuning fork for \(^4\)He films adsorbed on Grafoil.[1] In 32 kHz quartz tuning fork experiments, an external force acting on films was drastically reduced from 5 MHz AT-cut QCM experiments.[2] It was found that the decoupling from the oscillating substrate becomes much weaker than that of 5 MHz AT-cut QCM experiments, although the frequency does not decrease exactly in proportion to the areal density. Under the condition of such a weak decoupling, it is interesting to study the sliding direction dependence. Thus motivated, we have started 32 kHz quartz tuning fork experiments for single-crystalline graphite. In this proceeding, we report the decoupling of \(^4\)He films, comparing with the previous experiments for Grafoil.

2. Experimental Procedure
In QCM experiments using a quartz tuning fork, the change in frequency is related to the coupled mass of film with the oscillating tuning fork. When the film on the top of the tuning fork moves in concert with the substrate, the resonance frequency decreases from that of no films is expressed as

\[
\frac{\Delta f_R}{f_R} \sim -2 \frac{m}{M},
\]

(1)
where \( m \) is the coupled mass of film and \( M \) is the mass of the arms of tuning fork. When the film is decoupled with the oscillating tuning fork due to superfluidity or slippage, the coupled mass decreases and the resonance frequency increases.

In the present experiments, exfoliated single-crystalline graphite was used as the substrate. Single-crystalline graphite was obtained from a kind of mineral called Franklin Marble, by dissolving it with hydrochloric acid. To exfoliate graphite, pieces of graphite were immersed in the mixture of concentrated sulfuric acid and nitric acid for 96 h. Exfoliation of graphite allows us accurate measurements of the coupled mass of film because of a large specific surface area. After neutralization and dehydration, the interlayer space was expanded by heating in furnace at 1050 °C for 15 s. For cleaning, the exfoliated graphite was heat-treated in a vacuum at 900 °C for 4 h. Laue photograph of the obtained exfoliated graphite is shown in Fig. 1, which demonstrates that the graphite remains the crystal orientation in the \( a-b \) plane after exfoliation. The atomically flat region of the graphite was about 100 nm in diameter.[3] It was larger than that of Grafoil.[4]

The graphite was bonded on a 32 kHz quartz tuning fork. The experimental procedure is similar to the previous experiments for Grafoil.[1, 2] To prepare a tuning fork with graphite, 2000 Å-thick Ag film was deposited on the graphite. Then, the tuning fork and Ag-plated graphite were diffusively bonded by heating in hydrogen atmosphere at 300 °C for 1 h. In the process of bonding, the crystal direction of graphite was controlled by adjusting the angle between the oscillation direction and the \( a \)-axis of graphite. One of the tuning fork oscillated in parallel to the \( a \)-axis of graphite, while the other in the direction inclined at 30° with respect to the \( a \)-axis. After bonding, any excess graphite was carefully removed. Finally, the \( Q \) value of the tuning fork was better than 5×10^4 at room temperature in a vacuum. One of the obtained tuning fork with graphite is shown in Fig. 2.

Two tuning forks were mounted in the same sample cell. In addition to the tuning forks, baked Grafoil disks were put in the cell in order to control the areal density of \(^4\)He film precisely. After heating in \( 7 \times 10^{-5} \) Pa at 80 °C for 8 h to remove water, the cell was cooled down to 4.2 K. The resonance frequency and \( Q \) value were measured using a transmission circuit. In the circuit, the tuning fork was placed in series with a coaxial line connecting a 50 Ω cw signal generator and a 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 was the resonance amplitude, and the change in $Q$ value was calculated from this amplitude.

3. Results and Discussion

The QCM experiments carried out for various $^4$He areal densities down to 0.08 K. Figure 3 shows the areal density dependence of the change in frequency at 1.0 K, together with that of Grafoil. In the present experiments, the oscillation amplitude is about 10 nm, and the oscillation direction is inclined at 30° with respect to the $a$-axis of graphite. For Grafoil, the oscillation amplitude was 140 nm, and no clear oscillation amplitude dependence was observed. The vertical axis is normalized by the mass loading at the first layer completion. The estimated mass loading is $0.003 \text{ Hz/(atoms-nm}^{-2} \text{)}$ for single-crystalline graphite and $0.022 \text{ Hz/(atoms-nm}^{-2} \text{)}$ for Grafoil, respectively. The vertical dotted line corresponds to the first layer completion.

![Figure 3. Comparison of the areal density dependence. Red circles correspond to the present experiments, while blue squares to Grafoil. The vertical axis is normalized by the mass loading at the first layer completion.](image)

In the present experiments, the resonance frequency does not decrease in proportion to the areal density; the frequency remains constant up to 8.5 atoms-nm$^{-2}$ in the monolayer region, and decreases rapidly above this areal density. Then, it reaches the estimated value of mass loading at around the first layer completion of 12.0 atoms-nm$^{-2}$. These observations demonstrate that the film undergoes almost decoupling in the monolayer region and then is locked to the oscillation at around the second layer promotion. The above-mentioned areal density dependence is significantly different from Grafoil. For Grafoil, the frequency starts to decrease at around 4 atoms-nm$^{-2}$ and decreases gradually until the first layer completion. The difference may be attributed to the platelet size, and the pinning of film does not take place in the present substrate.

Finally, we shortly comment on the sliding direction dependence. In both the directions parallel to the $a$-axis and inclined at 30°, the resonance frequency shows the similar areal density dependence at 1.0 K; the frequency remains constant up to 8.5 atoms-nm$^{-2}$ and then decreases rapidly. However, there appears a difference at low temperature. At 0.1 K, the frequency for the oscillation direction inclined at 30° with respect to the $a$-axis shows a small decrease at 4-6 atoms-nm$^{-2}$. In contrast, the other does not show the difference between 1.0 and 0.1 K. This areal density corresponds to the commensurate phase. Precise experiments are in progress.
4. Summary
We carried out QCM experiments of a 32 kHz quartz tuning fork for $^4$He films adsorbed on exfoliated single-crystalline graphite, and measured the temperature dependence of the resonance frequency and $Q$ value. From the present experiments, it was found that the decrease in frequency is suppressed in the monolayer region compared with that of Grafoil, which means that the films undergo almost decoupling from the present substrate. This suggests that the platelet size plays an important role in decoupling. Furthermore, we observed a weak sliding direction dependence at 4-6 atoms-nm$^{-2}$ at low temperature.

References
[1] Nihei F, Ideura K, Kobayashi H, Taniguchi J, and Suzuki M 2011 J. Low Temp Phys. 162 559
[2] Hosomi H, and Suzuki M, 2008 Phys. Rev. B 77 024501
[3] Kobayashi K, Taniguchi J, Suzuki M, Miura K, and Arakawa I 2010 J. Phys. Soc. Jpn. 79 014602
[4] Niimi Y, Matsui T, Kambara H, Tagami K, Tsukada M, and Fukuyama H 2006 Phys. Rev. B 73 085421