Mechanical response of noble gas films to an oscillating substrate

Hajime Kobayashi\textsuperscript{a}, Junko Taniguchi\textsuperscript{a}, Masaru Suzuki\textsuperscript{a}, Kouji Miura\textsuperscript{b}, and Ichiro Arakawa\textsuperscript{c}

\textsuperscript{a} Department of Engineering Science, University of Electro-Communications, Chofu, Tokyo 182-8585, Japan.
\textsuperscript{b} Department of Physics, Aichi University of Education, Kariya, Aichi 448-8542, Japan.
\textsuperscript{c} Department of Physics, Gakushuin University, Toshima, Tokyo 171-8588, Japan

E-mail: hajime@phys.uec.ac.jp

Abstract. We carried out quartz-crystal microbalance (QCM) experiments for Xe films adsorbed on an exfoliated single-crystalline graphite substrate (Xe/Gr) and Kr films adsorbed on a synthetic mica substrate (Kr/mica) around LN\textsubscript{2} temperature. For Xe/Gr, it was found that the resonance frequency decreases greatly around the first layer completion, while it does not decrease at low coverages. The observed behavior is similar to that of Kr films on a graphite substrate (Kr/Gr). This demonstrates that the layer completion strongly affects the sliding motion of noble gas films on graphite.

1. Introduction

Sliding motion of physisorbed films on a lateral oscillating substrate is an interesting topic of nanotribology. It is well known that noble gas films adsorbed on graphite grow in layers at low temperature. In addition, the films take various structures, e.g., the commensurate (C) and the incommensurate (IC) phases for monolayer films.[1, 2] They enable us to make clear the relation between the sliding motion and the structure of film.

We have reported two types of quartz-crystal microbalance (QCM) experiments for Kr films adsorbed on a single-crystalline graphite (Kr/Gr) : one was a 5 MHz AT-cut quartz-crystal, and the other a 32 kHz quartz crystal tuning fork.[3, 4] For both experiments, it was found that the sliding motion of Kr/Gr starts to be suppressed in the C phase of the monolayer film.

It is of interest to confirm whether the C phase plays an important role in the suppression of sliding motion. Xe films adsorbed on graphite do not take the C phase above 80 K.[2] Thus, we carried out experiments for Xe films adsorbed on a single-crystalline graphite (Xe/Gr). In this paper, we show the coverage dependence of sliding motion for Xe/Gr, and compare it with that for Kr/Gr. We also report preliminary experiments for Kr films adsorbed on a mica substrate (Kr/mica).

2. Experiment

2.1. QCM technique

The sliding motion of noble gas films with a lateral oscillating substrate was measured by the QCM technique. In our experiments, two types of the quartz crystal were used. One is an
AT-cut quartz crystal, which oscillates in a shear mode with a resonance frequency of 5 MHz. The other was a quartz crystal tuning fork with a resonance frequency of 32 kHz, which has a flexural oscillation perpendicular to the arms.

First, we explain the QCM technique for a 5 MHz AT-cut quartz crystal. When the film adsorbed on the substrate moves in concert with the oscillating substrate, the resonance frequency decreases from that of no film is expressed as

$$
\frac{\Delta f}{f} = -\frac{\sigma}{M},
$$

where $f$ is the resonance frequency, $\sigma$ and $M$ are the areal mass densities of the film and the crystal. In case when the film slides uniformly on the oscillating substrate and the frictional force is proportional to the sliding velocity $v$ as $F = -\sigma \frac{1}{\tau} \cdot v$, the changes in resonance frequency and $Q$ value are related to the slip time $\tau$ as

$$
\frac{\Delta f}{f} = -\frac{\sigma}{M} \frac{1}{1 + (\omega \tau)^2},
$$

$$
\Delta \left( \frac{1}{Q} \right) = 2\frac{\sigma \omega \tau}{M} \frac{1}{1 + (\omega \tau)^2},
$$

where $\omega$ is the angular frequency of oscillation.[5] From these changes, the effective coupled mass is obtained. For the QCM of a 32 kHz quartz crystal tuning fork, Eq. (1) is replaced from $\sigma/M$ to $2m/w$, where $m$ is the mass of film, and $w$ is the mass of the tuning fork arms.

The resonance frequency was measured using a transmission circuit. The quartz crystal was placed in series with a coaxial line connecting to a 50 Ω cw signal generator and an RF lock-in amplifier. The frequency of the signal generator was controlled in order to keep the in-phase output zero and was locked to the resonance frequency. The quadrature output at this frequency is the resonance amplitude.

2.2. Sample preparation and the setup

We have already reported the sample preparation for an exfoliated single-crystalline graphite substrate.[3, 4] Here, we mainly describe the sample preparation for a 5 MHz AT-cut quartz crystal with mica substrate. A synthetic single-crystalline mica was used, which was commercially available.[6] To prepare the crystal with mica, 30 nm-thick Ag-Au alloy film was sputtered onto the mica. The Ag-Au plated mica and the crystal were pressed together and were heated in hydrogen atmosphere at 350°C for 1 h. Then, the mica was bonded on both sides of the gold electrodes of the crystal. After bonding, any excess mica was carefully cleaved to obtain a clean surface. After these processes, the $Q$ value of the crystal remained higher than $2 \times 10^4$ in a vacuum at LN$_2$ temperature.

The crystal was mounted in a sample cell in a LN$_2$ cryostat. Noble gas atoms were physically adsorbed on the substrate by means of the introduction from room temperature. The fractional coverage of films was calculated from the amount of introduced noble gas. With the aim of the minimization of the desorption and the control of the coverage of films, a pellet of synthetic mica powder was prepared. The pellet was formed by mixing mica powder with 70 μm silver powder and sintering at 200°C for 3 h. It was set on the bottom of cell in addition to the crystal. The total surface area of the cell with the pellet increased up to 3.8 m$^2$. After setting, the cell was cooled down to LN$_2$ temperature.

3. Results and discussion

3.1. Comparison Xe/Gr with Kr/Gr

We carried out QCM experiments of a 32 kHz quartz crystal tuning fork for Xe/Gr at 110 K. The oscillation amplitude was about 1.4 nm. Figure 1 shows the variation of the resonance frequency
as a function of fractional coverage, together with that of Kr/Gr at 80 K. The coverage is divided by that of the first layer completion, which was determined from the adsorption isotherm. The dashed line corresponds to the first layer completion. For Xe/Gr, the resonance frequency does not change greatly in monolayer. At 0.9 monolayer, it starts to decrease slightly, and the decrease is enhanced around 1.0 monolayer with increasing coverage. Above 1.2 monolayer, it is suppressed. In contrast, the resonance frequency for Kr/Gr starts to decrease at 0.85 monolayer. The decrease becomes drastically large around 1.0 monolayer, and continues until 1.4 monolayer.

It is interesting to compare these behaviors to the phase diagram.\cite{1, 2} In contrast to Kr/Gr, Xe/Gr do not take the C phase at 110 K. The frequency for Xe/Gr starts to decrease at the IC phase, while for Kr/Gr at the C phase. Without respect to the difference, the decrease is enhanced around the first layer completion in both cases. These results indicate that the effect of the C phase for the decrease in frequency is small, i.e., the effective coupled mass of noble gas films adsorbed on graphite does not depend on the commensurability in monolayer. We found that the layer promotion plays an important role in the effective coupled mass.

We comment on the coverage dependence at low coverages in monolayer. As mentioned above, Xe/Gr and Kr/Gr are decoupled from a lateral oscillating substrate at the coverages, which is a common feature for noble gas films adsorbed on graphite substrate.\cite{7, 8} It is, however, reported that the resonance frequency decreases at low coverages in monolayer for Xe films adsorbed on graphen grown epitaxially on top of a Ni(111) (Xe/graphen/Ni).\cite{9} There is a possibility that the difference between our results and those for Xe/graphen/Ni comes from roughness or heterogeneity of substrate, but the origin is an open question.

### 3.2. Kr film on mica substrate

We carried out a 5 MHz AT-cut QCM experiments for Kr/mica at 82 K. The oscillation amplitude was about 2.0 nm. Figure 2 shows the variation of the resonance frequency, the $Q$ value and the slip time as a function of fractional coverage. The coverage is divided by that of the first layer completion, which was determined from the adsorption isotherm. It was found that the resonance frequency decreases along $\Delta f_{\text{max}}$ until the coverage is around 0.2 monolayer. Between 0.2 and 0.8 monolayer, it does not decrease greatly with increasing coverage. Above 0.8 monolayer, the decrease is enhanced, and continues after the first layer completion. These changes may be attributed to the liquid-solid phase transition of Kr films.

Finally, we shortly comment on a comparison of the slip time between for several substrates. Table I shows the slip time of Kr film and the electric conductivity. It was found that conductivity...
Figure 2. (a) Variation of the resonance frequency \(\Delta f\) and the \(Q\) value \(\Delta (1/Q)\) at 82 K for Kr/mica as a function of coverage. The vertical axis shows the change from 4.973868 MHz. The solid line in (a) corresponds to the estimated sensitivity for the mass loading. The horizontal axis shows the fractional coverage divided by the value of the first layer completion determined from the adsorption isotherm. (b) Variation of the slip time as a function of coverage.

does not depend greatly on the slip time. This suggests that the electronic friction does not contribute to the slip time significantly for the system studied here.

Table 1. Slip time and conductivity for various substrates

| Substrate | Slip time for Kr monolayer films (ns) | Conductivity (\(\Omega\)-m) |
|-----------|---------------------------------------|-----------------------------|
| Graphite  | 13-25 \(^a\)                          | \(1 \times 10^{-7}\)        |
| Gold      | 5-10 \(^b\)                           | \(0.5 \times 10^{-8}\)      |
| Mica      | 2-3                                   | \(1 \times 10^{13}\)        |

\(^a\) From Ref. [3].
\(^b\) From Ref. [10].

4. Summary

We carried out QCM experiments for Xe films adsorbed on an exfoliated single-crystalline graphite at 110 K, and Kr films adsorbed on synthetic mica at 82 K, respectively. It was found that effective coupled mass of noble gas films on graphite does not depend on the commensurability in monolayer. In contrast, there is possibility that the effective coupled mass for Kr films adsorbed on mica depends on the film structure in monolayer.

References

[1] Butler D M, Litzinger J A, Stewart G A, and Griffiths R B 1979 *Phys. Rev. Lett.* **42** 1289; Butler D M, Litzinger J A, and Stewart G A 1980 *ibid.* **44** 466
[2] Hawoong Hong, Peters C J, Mak A, Birgeneau R J, Horn P M, and Suematsu H 1989 *Phys. Rev. B* **40** 4797
[3] Kobayashi H, Taniguchi J, Suzuki M, Miura K, and Arakawa I 2010 *J. Phys. Soc. Jpn.* **79** 014602
[4] Kobayashi H, Taniguchi J, Suzuki M, Miura K, and Arakawa I 2010 *J. Phys.: Conf. Series* **258** 012018
[5] Krim J and Widom A 1988 *Phys. Rev. B* **38** 12184
[6] We purchased synthetic single-crystalline mica from Crystal Base Co., Ltd.
[7] Hosomi N and Suzuki M 2008 Phys. Rev. B 77 024501
[8] Nihei F, Ideura K, Kobayashi H, Taniguchi J, and Suzuki M 2011 J. Low Temp. Phys. 162 559
[9] Coffey T and Krim J 2005 Phys. Rev. Lett. 95 076101
[10] Coffey T and Krim J 2005 Phys. Rev. B 72 235414