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Dynamical frictional force of nanoscale sliding

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We measured the dynamical frictional force acting on highly oriented pyrolytic graphite and C_{60} substrates by a sliding Si_3N_4 tip as a function of sliding distance using the probe-tip–quartz-crystal-resonator technique. It was found that the dynamical frictional force undergoes a drastic change when the oscillation amplitude is approximately the lattice constant of each substrate: For a small case, it is directly proportional to the amplitude, while for a large case, it does not depend on the amplitude. The observed behavior is qualitatively understood by a simple one-dimensional Tomlinson model.

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I. INTRODUCTION

In nano- or microscale systems the surface effects, such as the frictional and stictional behaviors, are of great importance because of their large surface-to-volume ratio. Even in macroscopic bodies, their friction and stiction are governed by the real area of contact, which is much smaller than the apparent area of contact. To control the surface effects, understanding of small contacting asperities has an industrial significance.[1,2]

By using a probe-tip–quartz-crystal resonator, it is possible to study the frictional behavior of small contacting asperities.[3–5] In 1999, Laschitsch and Johannsmann studied this by contacting a small sphere with a quartz surface and reported a positive frequency shift and a decrease in the Q factor.[4] This observation was explained as being caused by the emanation of a spherical sound wave from the point of contact into the sphere. They also reported an additional decrease in the Q factor for high-friction interfaces of a metal-metal contact, which may be attributable to frictional processes in the contact area. Thereafter, Borovsky *et al.* measured nanomechanical properties using a depth-sensing nanoindenter probe and a quartz-crystal microbalance (QCM).[5]

The atomic force microscope (AFM) is a powerful tool to investigate the friction on a nanometer scale.[6–8] In 1987, Mate *et al.* first observed atomic-scale features on the lateral force acting on a tungsten wire tip sliding on a graphite surface.[6] Its lateral force showed an atomic-scale stick-slip behavior during sliding. In 2004, Socoliuc *et al.* measured the lateral force acting on a NaCl surface.[8] For a normal load of 4.7 nN, the force clearly showed two opposite sawtooth profiles when scanning forwards and backwards. Although the sawtooth profile can be attributed to a corrugation potential with a periodicity of the lattice, it can be observed using something other than a single-atom tip; in fact, Miura *et al.* observed the atomic-scale sawtooth profile in the case of a graphite flake sliding on a graphite surface.[9]

It is believed that the dynamical frictional force is connected to the hysteresis for the sawtooth profile observed by AFM. We can expect that the force of a nanoscale contact drastically changes when the sliding distance becomes less than the lattice constant. Thus, we directly measured the force acting on highly oriented pyrolytic graphite (HOPG) and C_{60} substrates by a sliding Si_3N_4 tip as a function of sliding distance using the QCM technique. In this paper, we report our observation and a comparison with the calculation of a simple Tomlinson model.

II. EXPERIMENTAL SETUP

To study the dynamical frictional force, or the energy dissipation due to this force, by changing the sliding distance, we combined an AFM cantilever with an AT-cut quartz crystal resonator. Figure 1 gives a sketch of the experimental setup. A resonator with a HOPG or C_{60} substrate was mounted on a piezo-scanner base and was set facing an AFM cantilever as a force sensor. In the present experiments, a Si_3N_4 self-detective cantilever with a spring constant of 2.2 N/m (NPX1CTP003, SII) was used, and the typical radius of the tip was 20 nm.

The normal load acting on a Si_3N_4 tip was controlled by driving the piezo-scanner base. The cantilever was connected to the dc bridge circuit with an applied voltage of 1.0 V as bias. The out-of-balance signal corresponded to the normal load, and its sensitivity was better than 1 nN.

To clarify the effect of the periodicity of corrugation potential, we prepared two kinds of substrates: HOPG and C_{60}. For the HOPG substrate, an AT-cut quartz crystal with a fundamental resonance frequency of 3.26 MHz (SMD-49, Daishinku Corporation) was used as the resonator, and a HOPG flake of 1 $mm^2 \times 5$ μm was pasted with varnish. After heating at 130 °C for 1 h, the flake was cleaved to prepare a clean surface. The Q factor of the resonator remained higher than 2.0×10^4 in air. For the C_{60} substrate, an AT-cut quartz crystal with a fundamental resonance frequency of 4.99 MHz (SEN-5P, TAMADEVICE Co., Ltd) was used. C_{60} was thermally deposited on the Au electrode at 0.3 ML/min with a boat temperature of about 420 °C. The average thickness of C_{60} film was 90 ML, and the grain size was about 200 nm. After deposition, the Q factor was higher than 1.6×10^4.

The decrease in Q factor is connected to the energy dissipation, and the change in resonance frequency is related to the *effective* spring constant as follows:

$$\Delta\left(\frac{1}{Q}\right) = \frac{\Delta E}{2\pi E}, \qquad \frac{\Delta f_R}{f_R} = \frac{1}{\omega_R^2 M_C}\kappa, \qquad (1)$$

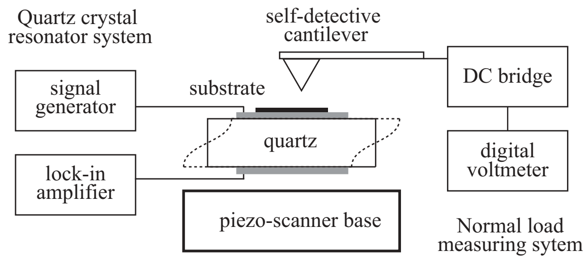

FIG. 1. Schematic diagram of the present apparatus. An AFM cantilever is combined with an AT-cut quartz-crystal resonator.

where ΔE is the energy dissipated per cycle, E is the energy stored in the system, M_C is the mass of the oscillating area, and κ is the *effective* spring constant.[4,5]

The resonator was placed in a transmission circuit, in which a 50 Ω cw signal generator and a rf lock-in amplifier were connected in series. The signal transmitted through the resonator was detected by the lock-in amplifier, and the frequency of the signal generator was controlled in order to keep the in-phase signal zero. The frequency was then locked to the resonance frequency. The quadrature signal at this frequency was the resonance amplitude, and this decrease was converted to the decrease in Q factor. The sliding distance corresponded to the oscillation amplitude of the resonator and was controlled by the output signal of the signal generator. In the present experiments, we obtained the oscillation amplitude from the current transmitted through the resonator.[11,12]

III. RESULTS AND DISCUSSION

A. Experiment

We measured the resonance frequency and the Q factor while advancing and retracting the tip under the condition of the resonator oscillating at a constant amplitude. The amplitude was controlled in the range of 0.03–3 nm for the HOPG substrate, which corresponds to the maximum substrate velocity of 0.6–60 mm/s. The experiments were carried out at atmospheric pressure with less than 40% relative humidity.

Figures 2(a)–2(c) show the variation of the normal load N, the frequency shift $\Delta f_R/f_R$, and the energy dissipation $\Delta(1/Q)$ for the HOPG substrate as a function of piezo travel. These sets of data were taken when the tip was advancing at 6 nm/s. The crystal orientation of HOPG substrate was not determined. It was found, however, that different positions and substrates showed qualitatively the same behavior and further that it did not depend strongly on the angle between the oscillation and the cantilever.

As shown in Fig. 2, a clear jump-in was observed at the contact, and N increased almost linearly. At the jump-in, both $\Delta f_R/f_R$ and $\Delta(1/Q)$ increased rapidly and showed a clear oscillation amplitude dependence. As the amplitude increased, the rapid increase of $\Delta f_R/f_R$ decreased monotonously; in contrast, that of $\Delta(1/Q)$ took a maximum value at a certain amplitude. The normal load dependence of $\Delta f_R/f_R$ and $\Delta(1/Q)$ was small after the rapid increase, although $\Delta f_R/f_R$ increased gradually at a small amplitude.

For the tip–quartz-crystal-resonator experiments of Laschitsch and Johannsmann,[4] the maximum value of $\Delta f_R/f_R$

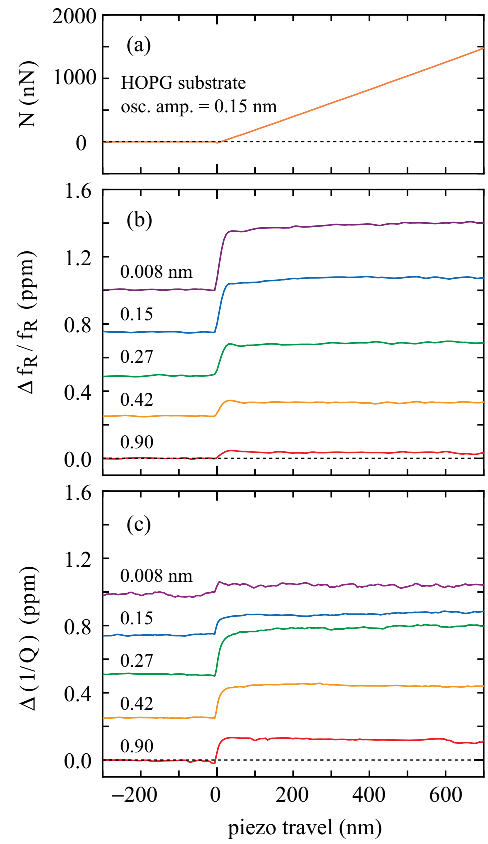

FIG. 2. (Color online) Variation of (a) the normal load N, (b) the frequency shift $\Delta f_R/f_R$, and (c) the energy dissipation $\Delta(1/Q)$ for the HOPG substrate. These sets of data were taken when the tip was advancing, and the horizontal axis is the piezo travel of the stage. Different colors correspond to different oscillation amplitudes. The data in (b) and (c) are shifted vertically.

was as high as 25 ppm at about 1 N for an Au-plated sphere of 3.5-mm radius. For those of Borovsky *et al.*,[5] the value also reached 11 ppm at 450 μN for a silica sphere of 0.5-mm radius. Their value is two orders of magnitude larger than the present experiments. Although their experiments showed that $\Delta f_R/f_R$ increases gradually with increasing N, the different behavior may be attributable to the experimental conditions. Here, we estimate the contact area A_c of the present experiments from the Johnson-Kendall-Roberts (JKR) equation to be about 90 nm^2 at the normal load of 400 nN.[10,13] Compared with their experiments, A_c is expected to be remarkably small, although it is difficult to study it quantitatively. For the C_{60} substrate, the variation of $\Delta f_R/f_R$ and $\Delta(1/Q)$ was found to be similar to that in the HOPG substrate, although the maximum value of $\Delta(1/Q)$ is shifted to a larger amplitude and the normal load dependence of $\Delta f_R/f_R$ is slightly enhanced.

Figures 3(a) and 3(b) show $\Delta f_R/f_R$ and $\Delta(1/Q)$ for HOPG and C_{60} substrates as a function of oscillation amplitude for the normal load of 400 nN; the behavior is qualitatively similar in the two substrates. They remained constant at a small amplitude. As the amplitude increased, $\Delta(1/Q)$ increased rapidly and took the maximum value, then decreased in inverse proportion to the amplitude. On the other hand, $\Delta f_R/f_R$ decreased after $\Delta(1/Q)$ took the maximum value. One of the

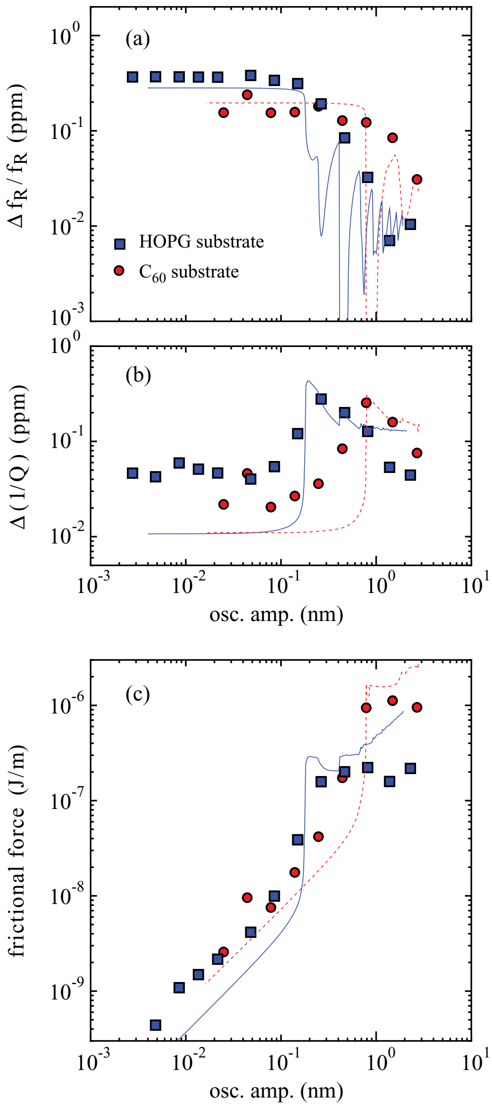

FIG. 3. (Color online) Oscillation amplitude dependence of (a) $\Delta f_R/f_R$, (b) $\Delta(1/Q)$, and (c) the frictional force at the normal load of 400 nN. The squares correspond to the HOPG substrate, while the circles correspond to the C_{60} substrate. The solid and dash lines are calculated numerically from Eq. (2).

differences between substrates appears to be the amplitude at the maximum value of $\Delta(1/Q)$. For the HOPG substrate it was about 0.3 nm, while for the C_{60} substrate it was about 0.8 nm.

The decrease in Q factor is connected to the energy dissipated per cycle as Eq. (1) and is converted to the energy dissipation per unit distance, i.e., the *average* dynamical frictional force, using the oscillation amplitude.[14] Figure 3(c) shows the amplitude dependence of this force. For the HOPG substrate, the force is directly proportional at a small amplitude. The transition occurs around 0.1 nm, and it becomes almost constant above 0.3 nm. For the C_{60} substrate, the force is directly proportional to the amplitude below 0.3 nm, and the transition occurs around 0.4 nm. It should be noted that the amplitude when the transition occurs depends on the substrate and is close to its lattice constant.

From these observations, we found the nanoscale contact has a common feature. When the oscillation amplitude is sufficiently smaller than the lattice constant, the dynamical frictional force is proportional to the amplitude. As the amplitude reaches around the lattice constant, it undergoes a transition. Following the transition, it does not depend on amplitude, i.e., the force does not depend on sliding velocity, or the Amontons-Coulomb-like behavior, although it does not increase proportionally with normal load.[16] This feature can be understood qualitatively as follows: For a small amplitude, the deformation due to the contact is proportional to the amplitude, and it is expected that the energy dissipation is proportional to the deformation in the case of the first-order perturbations. For a large amplitude, the energy dissipation is roughly proportional to the number of corrugations which the contact passes through.

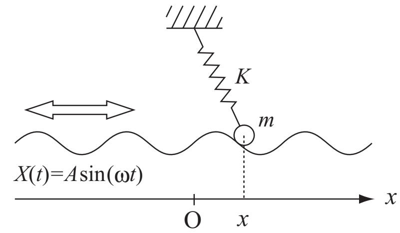

FIG. 4. One-dimensional Tomlinson model.

B. Model calculation

We compare the observed behavior with the calculation of a simple one-dimensional Tomlinson model.[15] Figure 4 shows the present model. Here, the substrate is replaced with a sinusoidal potential, and the tip is replaced with a point mass with a spring. The equation of motion is expressed as

$$m\frac{d^2x}{dt^2} = -Kx - \eta\left(\frac{dx}{dt} - \frac{dX(t)}{dt}\right) - \frac{2\pi}{a}U_0\cos\left\{\frac{2\pi}{a}[x - X(t)]\right\}, \qquad (2)$$

where the first term on the right-hand side is the restoring force, the second one is the viscous force, and the third one is the force from the effective corrugation potential. For the HOPG substrate, the periodicity of the sinusoidal potential is chosen to be $a = 0.246$ nm of the lattice constant of graphite and oscillates as $X(t) = A\sin(\omega t)$, where $\omega = 2\pi \times 3.26 \times 10^6\ \mathrm{s}^{-1}$. On the other hand, for the C_{60} substrate, the periodicity is chosen to be $a = 1.00$ nm from the (111) plane of C_{60} crystal, and $\omega = 2\pi \times 5.00 \times 10^6\ \mathrm{s}^{-1}$.

To compare with the experiments, the changes in resonance frequency and Q factor are calculated from the in-phase and quadrature-phase components of the force acting on the substrate.[17] It was found that $\Delta(1/Q)$ in the calculation shows a rapid increase at a certain oscillation amplitude under the stick-slip condition of $2\pi^2 U_0/(Ka^2) > 1$. As U_0 decreases from the stick-slip condition, the increase becomes small and finally disappears. The amplitude at the increase is close to a. However, it also depends on K and shifts to a larger value with increasing K. In addition, it was found that $\Delta f_R/f_R$ oscillates vigorously at a large amplitude. This is caused by enhancement of high-harmonic generation under the condition when the point mass climbs over the sinusoidal potential. The solid lines in Fig. 3 are the calculated curves with parameters $m = 3.1 \times 10^{-13}$ kg, $K = 3.2 \times 10^3$ N/m, $U_0 = 1.2 \times 10^{-17}$ J, and $\eta = 2.5 \times 10^{-5}$ Ns/m, while the dashed lines have parameters $m = 4.8 \times 10^{-13}$ kg, $K = 5.1 \times 10^3$ N/m, $U_0 = 3.1 \times 10^{-16}$ J, and $\eta = 3.9 \times 10^{-5}$ Ns/m.

We found that the model calculation reproduces the experimental data. The calculation shows that $\Delta f_R/f_R$ and $\Delta(1/Q)$ are constant at a small amplitude. As the amplitude increases, $\Delta(1/Q)$ increases rapidly and takes the maximum value around when the amplitude is close to half of the periodicity of sinusoidal potential; thereafter, it decreases in inverse proportion to amplitude. The drastic change in $\Delta(1/Q)$ is caused by the change in sliding manner. For a large amplitude, a rapid motion of the point mass due to high-harmonic oscillations increases the energy dissipation. According to this behavior, the frictional force is proportional to the amplitude for a small case, while it remains a large value for a large case.

The model calculation can qualitatively explain the observed frictional force. It should be emphasized that only the parameter η for energy dissipation in the calculation determines the whole frictional behavior. Thus, we can conclude that the principal mechanism of energy dissipation in the present experiments does not depend on sliding distance, although the frictional force drastically changes whether or not the sliding distance is smaller than the lattice constant.

Here, we make a short comment on Berg and Johannsmann's experiments in 2003.[18] They studied the tribology of micron-sized Au-Au contacts based on the probe-tip–quartz-crystal ring-down technique. They found that the frictional force remains small below the velocity amplitude of 0.4 m/s and explained that a local slip-to-stick transition occurs at the oscillation amplitude of about 0.5 nm. It is of interest that the frictional behavior is altered at a small sliding distance even by a micron-sized contact.

IV. SUMMARY

We measured the dynamical frictional force of a nanoscale contact as a function of sliding distance using the probe-tip–quartz-crystal-resonator technique. For a small oscillation amplitude, the force is directly proportional to amplitude. When the amplitude is about that of the lattice constant, it undergoes a transition to the Amontons-Coulomb-like behavior. This is qualitatively understood by a simple one-dimensional Tomlinson model.

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[12] The oscillation amplitude A_m can be calculated from the current I_m transmitted the resonator as $A_m = \frac{d}{2Se_{26}} I_m$, where d is the thickness of the crystal, S is the effective electrode area, and $e_{26} = 9.65 \times 10^{-2}$ C/m^2 is the piezoelectric stress coefficient.

[13] According to the the JKR model, the contact area for a sphere-on-plane situation is expressed as $S = \{\frac{4R}{3E^*}(N + 3\pi\gamma R + \sqrt{6\pi\gamma RN + (3\pi\gamma R)^2})\}^{2/3}$, where R is the radius of sphere, E^* is the effective Young's modulus, and γ is the surface tension. Here, we adopt $E^* = 9.4 \times 10^{11}$ Pa and $\gamma = 0.11$ J/m^2 estimated from physical quantities of Si_3N_4 and graphite.

[14] The *average* dynamical frictional force is calculated from the energy dissipation per cycle divided by the sliding distance as $\langle F\rangle_{\text{ave}} = \frac{\pi E}{2A_m}\Delta(\frac{1}{Q})$, where A_m is the oscillation amplitude

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[16] The normal load dependence is considerably different from the Amontons-Coulomb law. The force jumped up at the contact and remained almost constant. Because of this dependence, the coefficient of friction was considerably large at the normal load of 400 nN. This is an open question at present. One possibility is that only part of the contact area sticks and contributes the energy dissipation and the effective spring constant.

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