High Speed Microtribology with Quartz Crystal Resonators

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Quartz resonators in contact with metal-covered spheres have been used to study the tribology of micron-sized metal-metal contacts at high speed (1 m/sec) and high frequency (12 MHz), while maintaining a shear amplitude in the nanometer range. The data acquisition is based on ring-down experiments, where the electrical excitation is periodically interrupted and the free decay of the oscillation is analyzed. At contact, an amplitude-dependent frequency and decay rate are found, indicative of an underlying nonlinear equation of motion. Using perturbation theory and the two-timing approximation, a nonlinear spring constant $\kappa_i(x)$ and a nonlinear drag coefficient $\xi_i(x)$ are explicitly derived. We find a local slip-to-stick transition at a shear amplitude of 0.5 nm. Coating the gold surfaces with a self-assembled thiol monolayer removes the stick.

Friction, although an everyday phenomenon, is not well understood on the microscopic scale [1]. On macroscopic surfaces, mechanical contact usually occurs across a large number of small asperities. The size of these asperities adjusts itself through plastic deformation until the local stress is just below the yield stress. As a consequence, the local stress usually is in a range where nonlinear stress-strain relations and mechanical instabilities have a strong influence. What on the macroscopic scale appears as sliding or rolling, is a superposition of local stick-slip and bonding-debonding mechanisms. In order to address these phenomena at a fundamental level, single-asperity contacts with molecularly well-defined geometry are needed. Investigations of this kind have been termed "nanotribology." The most common instruments in nanotribology are the atomic force microscope (AFM) [2,3] and the surface forces apparatus (SFA) [4,5]. A positioning accuracy below 1 nm is easily achieved with these instruments, but it is difficult to reach lateral speeds above a few microns per second [6]. Speeds of many meters per second are often encountered in technical environments. Quartz crystal resonators can fill this gap because of their high frequency of oscillation [7–13]. Typical amplitudes of lateral displacement are a few tens of nanometers. With a frequency of 12 MHz this translates to a speed of a few m/s.

Figure 1 shows a sketch of the setup. A silicon nitride sphere (5 mm diameter, rms roughness < 3 nm) is approached to a quartz crystal thickness shear resonator. Both surfaces are coated with thermally evaporated gold layers. The experiments are performed under dry nitrogen atmosphere at room temperature. The resonator is driven by an ac voltage (0.2 V) at its 3rd harmonic at about 12 MHz. Out of contact, the bandwidth is 40–100 Hz. The excitation is periodically interrupted at a frequency of 40 Hz and the free decay of the quartz oscillation in the quiet period ("ring-down") is analyzed [8,11]. The resonance frequency $f$ and decay rate $1/\tau = 2\pi \Gamma$ with $\Gamma$ being the half-band-half (HBH) width are determined by fitting the function

$$y(t) = A \cdot e^{-2\pi \Gamma t} \cdot \cos(2\pi ft + \phi_0)$$

(1)

to the data trace, where $A$ is the amplitude and $\phi_0$ is a phase. When the surfaces are not in contact, Eq. (1) provides an excellent fit to the data. Upon contact both the resonance frequency and the bandwidth increase (Fig. 2) [7]. In our experiments, typical contact radii are 10–50 $\mu$m [14]. When contact is just barely established, one often observes a peak in the bandwidth which is related to interfacial friction. The peak is strong on clean metal and polymer surfaces while it is absent on ceramic surfaces. By replacing the sphere with a glass rod through which the contact area can be imaged one can assure that the contact region is free of dust [8]. The data in Figs. 2 and 3 are from such a clean contact. When contact is just being made, the quality of the fit with Eq. (1) decreases. The main source of discrepancy is a variation of frequency $f$ ("chirp") and decay rate $2\pi \Gamma$ over the duration of the decay. In order to quantify the chirp and the nonlinear decay rate we have divided the data trace into 10–40 subsets and performed fits with Eq. (1) on each subset separately. The results are sets of frequency shifts $\delta f_i(A_i)$ and HBH widths $\Gamma_i(A_i)$ where $A_i$ is the instantaneous amplitude. The fact that frequency and bandwidth depend

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**FIG. 1.** Sketch of the experimental setup: A sphere is approached vertically to a laterally oscillating quartz resonator.
FIG. 2. Frequency $f$ and half-band-half width $\Gamma$ when approaching a gold-covered sphere to the gold electrode of a quartz resonator. $f$ and $\Gamma$ were derived by fitting Eq. (1) to the raw data and assuming that they are independent of time. In contact, the fit quality decreases due to nonlinear interactions and chirp. A fit with a single set of parameters still yields a unique result. Since the data were not statistically weighted, the high-amplitude portion of the decay trace dominates the outcome of the fit.

on the amplitude is a consequence of nonlinear interactions. Quantitative analysis of these nonlinearities is possible because the sphere-plate interaction is a weak perturbation to the plate’s intrinsic elasticity. The analysis builds on the fact that frequencies and decay rates can be determined with very good precision. An overtone analysis, for instance, is more difficult [18].

The formalism is based on the “two-timing approximation” [8,19]. We start out from a nonlinear oscillator equation of the form

$$m \cdot \ddot{x} + \left[ \xi_0 + \xi_1(\dot{x}) \right] \cdot \dot{x} + [\kappa_0 + \kappa_1(x)] \cdot x = 0. \quad (2)$$

The parameter $m$ is the mass of the active portion of the quartz plate. $\xi_0$ and $\kappa_0$ are the drag coefficient and the spring constant of the free resonator, $\xi_1(x)$ and $\kappa_1(x)$ are a nonlinear drag coefficient and nonlinear spring constant describing the interaction between the sphere and the plate. Equation (2) assumes that $\kappa_1$ and $\xi_1$ have no microscopic history dependence. This approximation is reasonable because the period of oscillation is short. There is little time for “stick” to evolve. In addition, Eq. (2) excludes cross terms of displacement $x$ and velocity $\dot{x}$ such as $\kappa_1(x, \dot{x})$ and $\xi_1(x, \dot{x})$. In the following, we explicitly derive $\xi_1(x)$ and $\kappa_1(x)$ from the experimental data.

We first render Eq. (2) dimensionless by introducing a dimensionless time $\theta = \omega_0 \cdot t = \sqrt{\kappa_0 / mt}$. Drag coefficients and spring constants are converted to frequencies by writing $\xi_0 = \gamma_0 m$, $\xi_1 = \gamma_1 m$, and $\kappa_1 = \omega_1^2 m$. With these conventions Eq. (2) can be rewritten as

$$\ddot{x} + x + \epsilon h(x, \dot{x}) = 0,$$

$$\ddot{x} + x + \frac{\gamma_0}{\omega_0} \left( 1 + \frac{\gamma_1(x)}{\gamma_0} \right) \dot{x} + \frac{\omega_1^2(x)}{\gamma_0 \omega_0} x = 0. \quad (3)$$

Here the primes denote differentiation with respect to $\theta$. $\epsilon h(x, \dot{x})$ is a small perturbation to the oscillator equation $\ddot{x} + x = 0$, where $\epsilon = \gamma_0 / \omega_0$ is the perturbation parameter. The two-timing approximation starts out from the ansatz

$$x_0(t) = A(T) \cdot \cos[\omega_{01} t + \Phi(T)], \quad (4)$$

where $T = \gamma_0 t = (\gamma_0 / \omega_0) \theta$ is a second, slow time scale. $A(T)$ and $\Phi(T)$ are a slowly varying amplitude and phase, respectively. Applying perturbation theory and demanding that the first order solution is free of resonant forcing, one arrives at the following time-averaged equations [19]:

$$\frac{dA}{dT} = \frac{\langle h \sin(\theta) \rangle_0}{2\pi}; \quad A \frac{d\Phi}{dT} = \frac{\langle h \cos(\theta) \rangle_0}{2\pi}. \quad (5)$$

Inserting $h(x, \dot{x})$ from Eq. (3) and the zeroth order solution from Eq. (4), we find

$$2\pi \Gamma(A) = -\frac{A}{A} = \frac{\gamma_0}{2\pi \omega_0} \int_0^{2\pi} \left[ 1 + \frac{\gamma_1(x)}{\gamma_0} \right] \sin^2(\theta) d\theta, \quad (6a)$$

$$2\pi \delta f(A) = \Phi = \frac{1}{2\pi \omega_0} \int_0^{2\pi} \omega_1^2(x) \cos^2(\theta) d\theta. \quad (6b)$$

Since $x(\theta)$ and $\dot{x}(\theta)$ are piecewise monotonic, one can change the variable of integration from $d\theta$ to $dx$ or $d\dot{x}$

$$\Gamma(A) = \frac{\gamma_0}{4\pi} + \frac{1}{2\pi \omega_0 \omega_1} \int_0^{\omega_1 A} \left[ \gamma_1(x) \left( \frac{\dot{x}}{\omega_0 A} \right)^2 d\theta \right] dx, \quad (7a)$$

$$\delta f(A) = \frac{1}{2\pi \omega_0 \omega_1} \int_0^{\omega_1 A} \left[ \omega_1^2(x) \left( \frac{\dot{x}}{A} \right)^2 d\dot{x} \right] dx, \quad (7b)$$

where $A$ is the maximum displacement and $\omega_0 A$ the maximum speed. After discretization, Eqs. (7a) and (7b) provide a matrix relating the measured quantities $\delta f(A_i)$ and $\Gamma(A_i)$ to the discrete values of the nonlinear spring constant $\kappa_1(x_i)$ and drag coefficient $\xi_1(x_i)$. The matrix is well conditioned and can be inverted numerically. The friction force is calculated as $F(x, \dot{x}) = \xi_1(x) \cdot \dot{x}$ and the elastic force as $F(x, \dot{x}) = \kappa_1(x) \cdot \dot{x}$. A detailed derivation of Eqs. (5)–(7) is given in Ref. [8]. Figure 3 illustrates the data analysis for a gold-gold contact at three different positions, which are “out-of-contact,” “top of bandwidth peak,” and “deep in contact” (arrows in Fig. 2). Figures 3(a) and 3(b) show the amplitude-dependent shifts of frequency, $\delta f$, and HBH width, $\delta\Gamma$. The accuracy is about 1 Hz for both $\delta f$ and $\delta\Gamma$. Out of contact, $f$ and $\Gamma$ are constant and Eq. (1) provides a good fit to the data. Deep in contact, the amplitude dependence is small but noticeable. Deep in contact, most of the energy loss occurs via radiation of sound waves [7], not by interfacial friction. Right at contact, there is a rather strong chirp of more than 20 Hz.

Figures 3(c) and 3(d) show the nonlinear spring constant $\kappa_1(x)$ and the nonlinear drag coefficient $\xi_1(x)$ as functions of displacement and speed, respectively. On bare gold surfaces, one finds a threshold behavior with a sharp increase of the drag coefficient at a critical amplitude of 0.5 nm [Fig. 3(b)], corresponding to a speed of
3 cm/s [Fig. 3(f)]. At about the same amplitude the spring constant drops [arrow in Fig. 3(c)]. Comparing the frequencies in Figs. 3(a) and 3(c), one sees that the jump in the elastic interaction is about equal and opposite to the jump in dissipation. This suggests that when the amplitude of motion drops below 0.5 nm, an interaction that was dissipative turns into an elastic interaction. This statement holds regardless of the perturbation analysis: the step in the bandwidth is also seen in Fig. 3(b), which does not rely on the conversion to $\xi_1$. One sees a local slip-to-stick transition. The slip is confined to the rim of the contact area (see below).

Figure 4 shows a second data set displaying the same behavior. The filled circles represent an experiment where the gold surfaces were covered with a self-assembled thiol monolayer (16-mercapto hexadecanoic acid). Evidently, in our experiments the thiol layer acts as a lubricant and removes the stick.

The sublinear increase of the elastic force with displacement [Fig. 3(e)] is explained by the microslip model [15,17]. When a sphere touching a surface experiences an oscillating lateral stress, there is partial slip inside a circular annulus next to the contact line. The width of this slip region increases with amplitude and the spring constant decreases accordingly. The lateral elastic force $F_{\text{elastic}}$ transmitted through the part of the contact which does not slip is given by

$$F_{\text{elastic}} = F_{\| \max}(1 - (1 - \delta x/x_{\max})^{3/2}).$$

(8)

At a maximum displacement of $x_{\max}$ the annulus of slip covers the entire contact and the partial slip turns into total slip. The dashed line in Fig. 3(e) is a fit of Eq. (8) to our data, where $x_{\max}$ and $F_{\| \max}$ have been used as fit parameters. The agreement is very good.

In order to predict the dissipative interaction within the microslip model one needs relations connecting the local sliding speed to the local frictional stress. The standard microslip model employs Coulomb friction ($F_{\text{fric}} = \mu F_n$). Under this assumption, the energy dissipation per cycle $\Delta w$ is derived as [16]

$$\Delta w = \frac{9(2 - \nu)(\mu F_n)^2}{10 K r_c} \left[ 1 - \left(1 - \frac{F_{\| \max}}{\mu F_n}\right)^{5/3} - \frac{5F_{\| \max}}{6\mu F_n} \right] \times \left[ 1 + \left(1 - \frac{F_{\| \max}}{\mu F_n}\right)^{2/3}\right],$$

(9)

with $\mu$ the friction coefficient, $\nu$ the Poisson number, $F_n$ the normal force, $K$ the modulus, $r_c$ the contact radius,
and $F_{\parallel,\text{max}}$ the maximum lateral force [15–17]. This prediction is shown as a dashed line in Fig. 3(b).

As Fig. 3(b) shows, the prediction does not match the measured friction forces at all. Coulomb friction is today believed to be inapplicable on the microscopic scale. It clearly is not the microscopic friction law encountered in oscillatory shear between gold surfaces. Note that the microslip model also failed to describe the friction in the macroscopic experiments by Johnson [20] (while it yielded excellent agreement for the elastic force, just as in our experiments). The bandwidth was independent of amplitude at low stress and increased sharply at high stress. Johnson attributed the dissipation to elastic hysteresis in the deformation of asperities. At large stresses the deformation becomes plastic, which explains the sharp increase in dissipation.

Our findings differ from the macroscopic experiments in two ways. First, we do see an increase in the damping at low amplitude, although it is not linear (as predicted by the microslip model) but displays a step at an amplitude of 0.5 nm. We attribute this step to a slip-to-stick transition on the local scale. This slip-to-stick transition disappears, when the gold surface is covered with an organic self-assembled monolayer. Second, we do not see the sharp increase at high stresses although we do reach the transition from partial to total slip at low normal forces. The absence of the sharp increase can naturally be explained by the time scales involved. If the time required for plastic deformation exceeds the period of oscillation (100 ns) plastic deformation is eliminated as a mechanism for dissipation.

In conclusion, we have demonstrated that a nonlinear interaction between an oscillating quartz resonator and a sphere touching its surface can be quantitatively probed via the chirp and the nonexponential decay in ring-down experiments. The friction force in metal-metal contacts increases about linearly with speed. However, there is a minimum critical amplitude below which stick yields a much decreased friction coefficient and a correspondingly increased spring constant.

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