Friction Fluctuations and Friction Memory in Stick-Slip Motion

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We demonstrate that friction is not always an exact number. In a simple lubricated system (atomically smooth mica separated by molecularly thin films of squalane), sinusoidal shear forces were applied for millions of cycles. The kinetic friction increased steadily over repetitive cycles and collapsed intermittently to the average value. Individual slip cycles also consisted of a cascade of smaller slip events. The distribution of fluctuation size and duration followed a power law. [S0031-9007(96)01696-1]

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We tend to take for granted that friction is an exact number. The textbooks state that solid surfaces, when impelled to slide over one another, remain at rest until a friction force is overcome; thereafter velocity determines the force that resists sliding [1]. Various "friction-velocity" models have been considered recently in the literature [2–4]. Parallel to this dogma is the experience, familiar to all who make measurements, that fluctuation and irregularity are endemic whenever friction is actually measured [5]. It is often easy to explain irregularity as reflecting some kind of sample resonance or inhomogeneity. This is why it is common to dismiss all observed irregularity, in friction measurements, as having no fundamental origin [1,5,6]. In this Letter we describe experiments, in lubricated sliding, that call renewed attention to the problem.

There are two impediments to performing controlled experiments. The first is the practical difficulty of minimizing heterogeneity of the sliding surfaces; to this end, we employed step-free single crystals that were buffered from one another by a molecularly thin film of chemically pure fluid. The second difficulty is to apply friction forces in a controlled way. The usual solution is to apply a given sliding force at a given velocity. But it is not so simple: Because the force that promotes sliding also acts on additional degrees of freedom within the friction machine [1], actual sliding motion often involves repetitive periods of rest, acceleration, and deceleration. Recently an experimental strategy was suggested to deal with both difficulties: Oscillatory sliding forces were applied whose amplitudes were sufficient to pass from rest to sliding, but small compared to the dimensions of the contact zone [7]. Then the residual unavoidable surface heterogeneity remained constant because the same area of contact was probed throughout the experiment. But only time-averaged results were reported, leaving open the question of fluctuations around the mean.

The apparatus, a modified surface force apparatus, was described in detail previously [8–10]. Briefly, a droplet of the sample fluid was confined between two automatically smooth crystals of muscovite mica. Sinusoidal shear forces were applied to one piezoelectric bimorph and the resulting sinusoidal displacement was monitored by a second piezoelectric bimorph. A lock-in amplifier was used to decompose the output into one component in phase with the drive and a second component out of phase with it. A digital oscilloscope was used to inspect single output wave forms. The apparatus compliance was calibrated separately [10]. The temperature was 25 ± 1 °C. A desiccant, P_2O_5, was kept inside the sample chamber for hours to days before each experiment. Squalane $C_{30}H_{62}$ (purim grade, purchased from Fluka) was chosen as the lubricant fluid because experiments could be conducted at a fixed film thickness (18 Å). Therefore the startup of sliding and the transition from a solid-like state to the sliding state could be examined with oscillatory displacements whose amplitude was much less than the diameter, ≈45 μm, of the contact.

An illustrative experiment with time-averaged data is shown in Fig. 1. Oscillatory forces were applied. Confinement-induced slow relaxation [7–15] caused the surfaces to be pinned essentially at rest for small deflections; the deflection amplitude increased linearly with force and elastic energy was stored. At a critical force the deflection jumped discontinuously to a state where the displacement was predominantly 90° out of phase with the drive [7]. This, as discussed below, we identify with kinetic friction. Figure 1 is tantamount to a force-velocity curve since the product of displacement amplitude and oscillatory frequency defines the effective velocity of this experiment [7]. The experiment in oscillatory deformation, rather than in steady sliding, minimized dynamic instabilities [16]. Figure 1 is similar to friction-velocity models in the literature [2,3].

We now turn to fluctuations: first during a *single* cycle of stick-slip. The calibrated contribution of device compliance [10] and piezoelectric *RC* decay [9] were separated from measured voltage wave forms monitored with a digital oscilloscope. In Fig. 2, displacement of the sliding surface is plotted against time during a representative cycle at 1 Hz. One observes that the overall stick-slip process (top panel) was in fact proceeded by jagged microscopic stick-slip events (magnified in the

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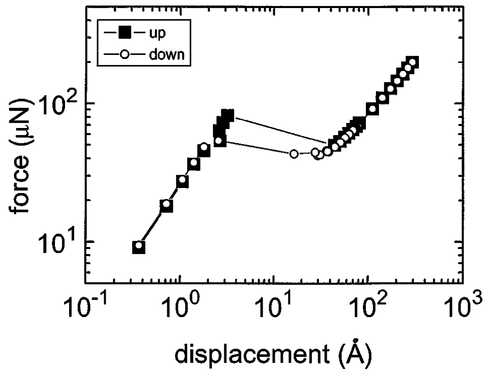

FIG. 1. Transition from static to kinetic friction. Squalane was confined between step-free, atomically-smooth mica to a thickness of 18 Å. Oscillatory shear forces were applied, $f(t) = f_0 \sin \omega_1 t$ (t indicates time, ω_1 indicates radian frequency, and f_0 indicates force amplitude), and resulting displacement was analyzed at this same frequency. Data were taken at 256 Hz in the direction of increasing (squares) or decreasing (circles) displacement amplitude. Static friction held when f_0 was small. At a critical f_0 the deflection amplitude jumped discontinuously to a state of kinetic friction.

bottom panel). The sliding surface moved on the order of nanometers, paused, then moved again. Even over a single cycle, sliding was neither smooth nor strictly periodic. But these data are noisy.

To improve the statistics, we analyzed stick-slip during millions of cycles. To limit the quantity of data to a manageable amount we turned to analyzing the displacement that was 90° out of phase with the drive at the same frequency. This we identify with the kinetic friction because, owing to cancellation of higher harmonic contributions [17], only the fundamental frequency of a sinusoidally periodic experiment determines the energy dissipated during a cycle of oscillation. In Fig. 3, the kinetic friction force following slip is plotted against the number of cycles for 2×10^6 cycles (at 256 Hz). The friction was irregular over short times but followed a statistical pattern: It tended to increase steadily at a well-defined rate, over many cycles, and then to collapse abruptly to the average value.

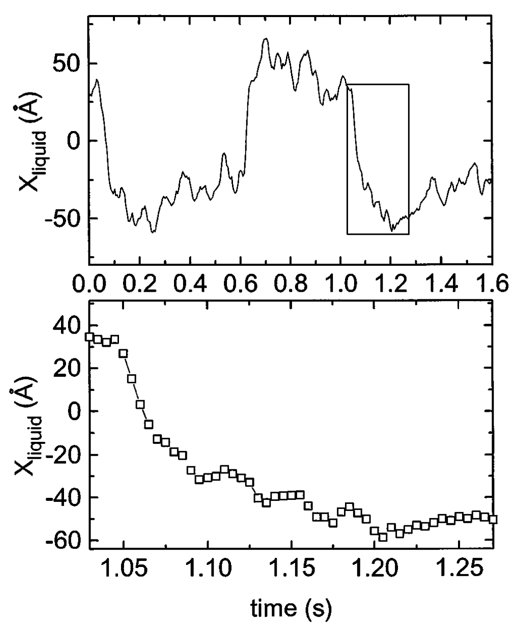

FIG. 2. A representative stick-slip cycle measured at 1 Hz. Displacement of the sliding surface, X_{liquid}, is plotted against time. The overall stick-slip process (top panel) proceeded by jagged microscopic stick-slip events (magnified view of box in top panel is presented in bottom panel).

A pattern of small triangles nested within larger triangles, indicating self-affine behavior, appears when the data are examined over successively fewer cycles (successively from top to bottom panel, Fig. 3). Friction was correlated over up to a hundred thousand successive stick-slip cycles. Also, with increasing drive amplitude, we observed increased intermittency of the fluctuations [18]. Eventually, for high enough drive amplitude (high enough velocity), they became nothing more than bursts of irregularity separated by long intervals of flat baseline [18].

For quantitative analysis, first the power spectra were calculated. In Fig. 4 the power spectrum is plotted against frequency (f) on log-log scales. The observed

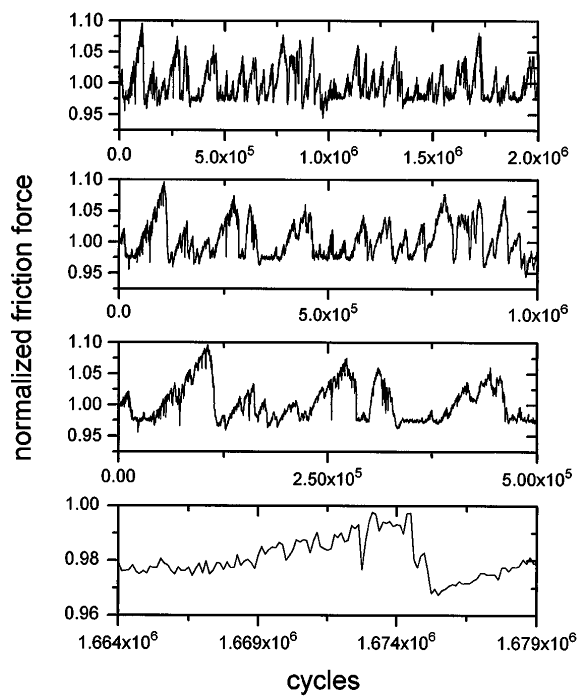

FIG. 3. Fluctuations of kinetic friction, normalized to the mean, during millions of repetitive shear cycles at 256 Hz with amplitude just sufficient to pass from rest to sliding. The mean dissipative force was 150 μN. The contact area was 6×10^{-9} m^2. The data were sampled at 4 Hz using a lock-in amplifier with a time constant of 30 ms. Viscous heating was negligible (<0.5 °C) owing to the great surface-to-volume ratio of the narrow gap. The traces from top to bottom, over successively smaller time intervals, suggest self-affine triangular-shaped structure.

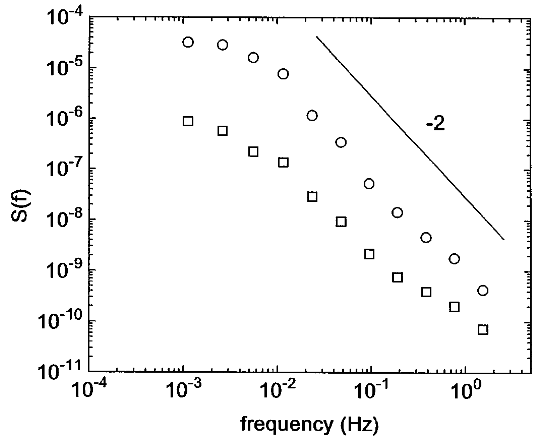

FIG. 4. Power spectra. Squared amplitudes of the Fourier components of the data, $S(f)$, are plotted against frequency (f) on log-log scales. The power spectra were octave-averaged for smoothing. Note that the slope of power-law decay approached -2 when drive forces were just sufficient to pass from rest to sliding. Circles: data from Fig. 3. Squares: data at higher displacement amplitude. The mean peak amplitudes were $d_0 =$ 41 and 58 Å, respectively.

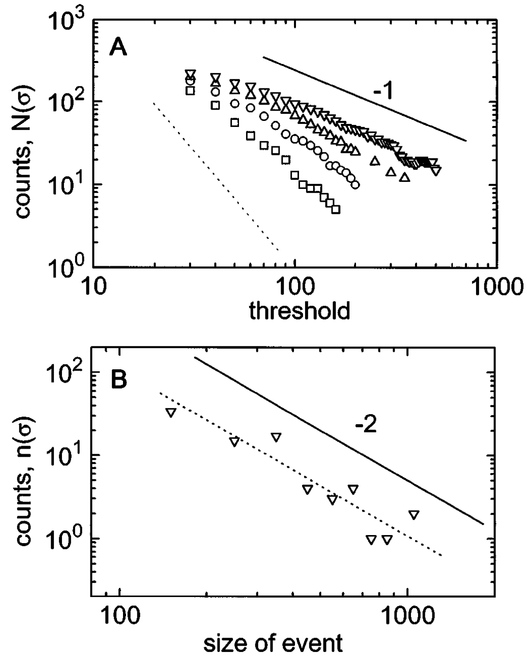

FIG. 5. (a) Cumulative number of fluctuations larger than a threshold σ, $N(\sigma)$, plotted against σ on log-log scales. The units of σ are 1 part in 10^4 of the mean kinetic friction. The drive amplitude was $d_0 = 41$ Å (triangles down), 47 Å (triangles up), 52 Å (circles), and 58 Å (squares). Empirically, the data indicate power law dependence. The dotted line shows the estimated level of Gaussian electronic noise. The data empirically suggest power law dependence, except when electronic and digitization noise confounded the measurements for the smallest σ. The power approached the limiting value -1 when drive forces were just sufficient to pass from rest to sliding. (b) Number of events of given amplitude $n(\sigma)$ plotted against event size for data in Fig. 3. Bin size was 100 counts and $\sigma > 100$. The data are consistent with the power-law slope of -2 expected, by Ref. [21], from the slope -1 in Fig. 5(a).

$1/f^2$ dependence at high frequency follows the $1/f^2$ dependence expected for regular triangles. It is also predicted, for different reasons, by a recent minimalistic theoretical model [19], as well as observed for dry friction [20]. In this case, as the present fluctuations became intermittent with larger drive amplitude, $1/f^\alpha$ behavior was observed with $\alpha < 2$. Figure 4 also shows that the power spectrum became independent of f at frequencies below 0.005 Hz. The significance is to quantify the correlation time: ≈200 sec, or ≈10^5 cycles of stick-slip. To anticipate discussion that follows, we tentatively attribute this time to correlations within the sheared fluid film during interfacial sliding.

For further quantification, the number of fluctuation events was counted. It was necessary to specify a threshold (σ) to distinguish a fluctuation event from random noise. How is this done without being arbitrary? The cumulative distribution of events whose magnitude exceeded a given (but variable) threshold was determined. In Fig. 5(a) this cumulative distribution $N(\sigma)$ is plotted against σ on log-log scales. The data are empirically consistent with a power law $N(\sigma) \sim \sigma^\beta$ (except for fluctuations of least amplitude, in which case electronic and digitization noise confounded the measurements). In particular, $\beta \rightarrow -1$ as the stick-slip transition was approached.

Therefore the number of events shown in Fig. 3 was in strict inverse proportion to the yardstick used to measure them. As a corollary, the derivative of this distribution implied the event size distribution at fixed threshold [21]. This distribution, shown in Fig. 5(b) from direct counting, confirms the slope -2, implied from the slope in Fig. 5(a) [22]. We do not understand the significance of the power-law slopes in Fig. 5. The simple integers comprise a challenge for theoretical explanation.

This friction problem may belong to a broader class of instabilities in other dynamical systems. The prediction of complex behavior comes from models as diverse as self-organized criticality [23], the Burridge-Knopoff model of earthquake fault dynamics [2], the sliding of randomly pinned charge density waves [24], sandpiles [25], and chaos in tape peeling [26]. The source of heterogeneity in this system seems not to lie within the solid surfaces themselves (deformation of the solid surfaces during sliding was negligible because they were so stiff) and not in frictional heating (negligible because heat transfer to the bulk was very efficient, owing to the large surface-to-volume ratio of the narrow gap between the sliding surfaces). It is reasonable to expect this behavior only over some window of spring constants within the larger dynamical system [27]; however, we have varied the spring constants without large effect [28]. By default, we identify the main contribution as degrees of freedom within the interfacial lubricant film itself.

We cannot be certain whether this interfacial inhomogeneity was spatial as well as temporal, but the data, especially in Fig. 2, certainly suggest this. This is also consistent with the view that static friction of confined fluids reflects a glassy response [12,14,29] or other kinds of domain structure [11,13]. In fact it is astonishing that the friction fluctuations did not average to a constant value over the large contact area of the experiment. This implies that some kind of coherent dissipative structures extended over lateral distances huge compared with molecular dimensions. On physical grounds, we expect these shear-induced structures to reflect fluctuations of the yield point between rest and sliding, and the subsequent slip amplitudes after yield.

The significance of these findings is to demonstrate complexity in lubricated sliding. In recent literature it has been proposed repeatedly that the onset of slip in lubricated sliding involves phase changes of the lubricated film (from solid-like to fluid-like). But it can happen that the confined fluid molecules have insufficient time, owing to confinement-induced slow relaxation, to adjust to confinement and sliding conditions. The resulting dynamics are seductively complex.

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