Frictional Dissipation in Stick-Slip Sliding

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The time variation of the frictional force between two surfaces, undergoing stick-slip sliding across a molecularly thin film of a confined model liquid, was examined at high time and force resolution, showing clearly that dissipation of energy occurs both during the slip, and at the instant of stick (via transfer of residual momentum). Detailed analysis indicates that, in marked contrast to earlier suggestions, of order 90% or more of the dissipation occurs by viscous heating of the confined shear-melted film during the slip, and only a small fraction of the energy is dissipated at the instant of stick.

A crucial issue in friction is to understand its dissipative nature. This was recognized as early as 1804, when Leslie pointed out that Coulomb's neat explanation of the da Vinci–Amonton classic laws of friction would result in no energy dissipation [1,2]. A large number of studies have since addressed this issue both experimentally, theoretically and via computer simulations [2–5]. A particularly intriguing case arises when friction occurs without either wear or plastic deformation of the rubbing surfaces, as in that case the origin of the frictional dissipation is more subtle, and several theoretical models have been proposed to account for this [2,3,6–11]. A characteristic case of such wearless friction occurs in stick-slip sliding of surfaces across molecularly thin films of liquids. This has been studied in a controlled manner by several groups in recent years using the mica surface force balance (SFB), in which a single, well-defined contact region is examined, within which the liquid is trapped between the parallel, molecularly smooth solid surfaces, and undergoes layering between them [12–17]. The SFB experiments, supported by detailed molecular dynamics simulations, have revealed that simple organic liquids confined to films that are thinner than ca. 5–8 monolayers, become solidlike in the sense that shear of the film is associated with a yield stress [18–21], and may then proceed via a stick-slip mechanism. Here I examine in detail the modes of energy dissipation for such motion in a model system, showing—in contrast to earlier suggestions—that most of the dissipation occurs during the slip itself.

A typical configuration of the surfaces corresponding to the SFB experiments [17,22] is shown schematically in Fig. 1, together with characteristic stick-slip friction-force traces for the model organic liquid octamethylcyclotetrasiloxane (OMCTS) confined between two sliding mica surfaces. The top mica surface, on a mounting of total mass M, is subject to a lateral force via a spring of constant K whose end is pulled in the $\vec{x}$ direction at a constant velocity v_s. The top and bottom surfaces are in contact over an area A across a thin film of thickness D. As the tension in the spring rises, at some extension of the spring

x_0 (tension Kx_0) a yield point y is reached (yield stress = Kx_0/A), the confined film abruptly liquefies [18,20,23,24], and the top surface slips past the bottom one by an amount Δx_0 to the point s where the film solidifies again. The process then repeats itself as the end of the spring keeps moving at a steady v_s. We denote the time per stick-slip cycle as τ_{ss}, and the time over which slip occurs, between y and s, as τ_s.

Here we analyze in detail such stick-slip motion to determine how the energy is dissipated during a cycle. We note at once that there is no frictional energy dissipation during the stick part of the cycle, from the solidification point s to the next yield point y; this is because there is no relative motion of the surfaces during the stick (any microscopic, shear-induced strain in either the surfaces or the confined film during the "stick" is elastic and reversible). The stored elastic energy at the point y must therefore be dissipated during the slip (y to s), via shear of the confined, shear-liquefied film, and also at the point of solidification itself when any residual momentum of the sliding top surface is lost.

Figures 2(a)–2(c) show, at high shear-force resolution and at increasing time resolution, the shear force over the course of a single stick-slip cycle measured in the SFB across a confined OMCTS film of thickness $D = 3.5 \pm 0.3$ nm (4 monolayers of OMCTS). Unlike the traces in Fig. 1 for a similar film, measured using a mechanical recorder with a time response of ca. 0.1 sec (to display a larger range of the data), the traces in Fig. 2 are measured with a recording oscilloscope, with a far higher time resolution ($<10^{-3}$ sec). The trace shows the extent of bending of the shear spring (as in Fig. 1), including any contribution resulting from the strain of the confined film: in region a, Figs. 2(b) and 2(c), the surfaces are coupled in adhesive contact, and the spring bends to its maximum extent x_0 before the yield point at y. During the slip (region b) the surfaces slide past each other across the confined liquid film, till they abruptly stick together again at the solidification point s. At this point, the high time resolution enables us to see clearly that the transfer of the residual
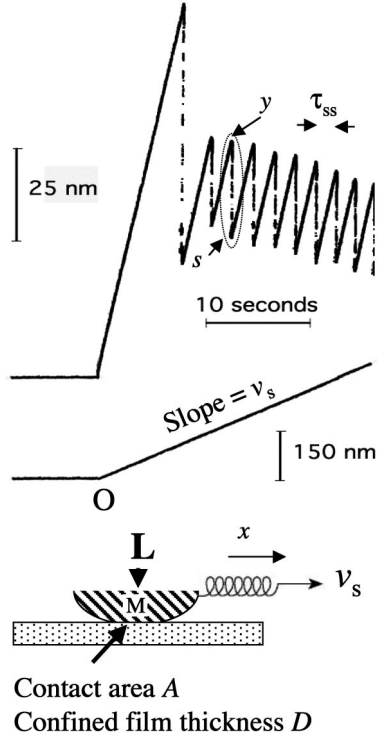
FIG. 1.   Friction-force traces between mica surfaces sliding across a film of OMCTS of thickness $D = 35 \pm 2$ Å ($n = 4$ molecular layers), under a load $\mathbf{L} = 16$ μN (for details of OMCTS preparation see Ref. [14]). The lower and upper traces, taken directly from the $\mathbf{X}$-t chart recorder, show, respectively, the applied uniform motion (at velocity v_s) of the end of the shear spring, and the extension of the spring in response to stick-slip motion between the upper and lower surfaces. The cartoon illustrates schematically the geometry in the SFB (for details see Ref. [22]). Motion of the spring end (lower trace) commences at the point O, and the initial stick spike (upper trace) is characteristically larger than subsequent stick spikes (the slight downward trend of the stick-slip cycles is due to thermal drift). The points y and s, indicated for clarity only for one (circled) cycle, are the yield and solidification points of the confined film.
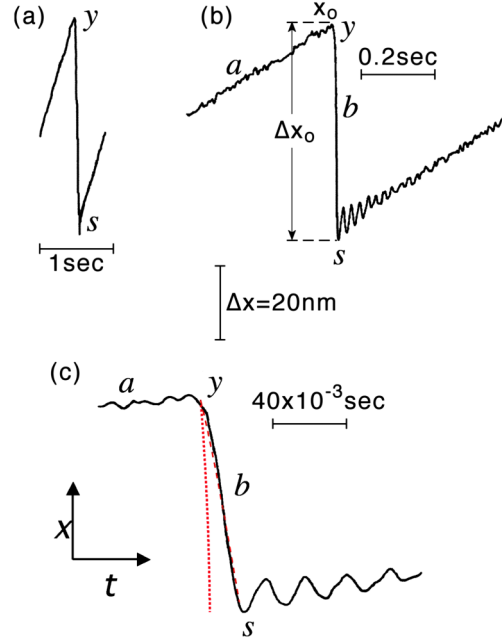

FIG. 2 (color online).   (a) A single typical stick-slip cycle (region similar to the marked cycle in Fig. 1, but from a different experiment recorded via a LeCroy 9310M recording oscilloscope) for friction between mica surfaces across a 35 ± 3 Å film of OMCTS ($n = 4$ monolayers) under a load of 42 μN. The contact area A at this load, evaluated from Johnson-Kendall-Roberts contact mechanics expression [14], is $A = (4 \pm 0.4) \times 10^{-10}$ m^2. Traces (b) and (c) focus on the slip region of the same cycle at magnified time scales, where a and b are, respectively, stick and slip regions, x_0 and Δx_0 are, respectively, the spring bending at the yield point y and the extent of slip from y to s. The SFB shear-spring constant $K = 97$ N/m, while the mass of the moving surface and its mount is $M = 1.47$ g. The dashed curve (red online) is the predicted variation $x(t)$ given by Eq. (2) in the text, with a value of B corresponding to an effective viscosity of the OMCTS film given by $\eta_{\mathrm{eff}} = 27$ Pa s. The dotted curve (red online) corresponds to the predicted variation of $x(t)$ with the viscosity of the OMCTS film given by its bulk value $\eta_{\mathrm{bulk}} = 2.5 \times 10^{-3}$ Pa s.

momentum of the top surface (and its mount), excites vibrations, or "ringing" in the overall system (top and bottom surfaces adhered together). These decay (with a characteristic time of ca. 80 ms for the trace shown), and their energy is dissipated as phonons (heat) in the SFB apparatus.

From Fig. 2 we can therefore clearly identify two modes of energy dissipation of the stick-slip cycle: during the slip the shear-melted liquid film undergoes viscous heating, while at the solidification point the residual momentum of the top surface is lost in the ringing vibrations of the system (which then rapidly decay). To proceed we write down an equation of motion of the top surface from the instant of yield at y, where $x(t)$ is the extent of sliding of the top surface at time t after slip has commenced at y ($t = 0$, $x = 0$ at y). Since $\tau_s \ll \tau_{ss}$ (for the data of Fig. 2, $\tau_s \approx 25 \times 10^{-3}$ sec, while $\tau_{ss} \approx 1$ sec), we can, to a good

approximation, take the extension of the shear spring to be $[x_0 - x(t)]$ throughout the slip regime b. Thus we write

$$M(d^2x/dt^2) + B(dx/dt) = K(x_0 - x) \qquad (1)$$

valid in the regime $0 \le x \le \Delta x_0$ (the total extent of the slip from y to s), and subject to boundary conditions $x = (dx/dt) = 0$ at $t = 0$. $B(dx/dt)$ is the damping term due to shear of the confined film during the slip. Equation (1) is not exact because it ignores additional contributions to the potential energy, such as the energy ΔE_f stored in the confined film prior to slip, as pointed out by Luan and Robbins [11]. Indeed, it is readily shown that, whatever the form of the damping term B, the energy $\Delta E_{\mathrm{spring}}$ released by the shear spring over the slip from y to s is exactly dissipated by the damping [25]: thus Eq. (1) fails to predict the residual energy ΔE_{osc} responsible for the ringing os-

cillations. However, as we show below, $\Delta E_f \ll \Delta E_{\mathrm{spring}}$ (and likewise ΔE_{osc} is small compared to the viscous dissipation), so that we may proceed with Eq. (1), bearing these limitations in mind. If we assume the shear-melted film behaves in a Newtonian fashion, with effective viscosity η_{eff} throughout the slip (implying the strong assumption of shear-rate and D-independent viscosity), and the standard zero-fluid-velocity boundary conditions at each surface [26], then the damping term has the newtonian form $B(dx/dt) = (A\eta_{\mathrm{eff}}/D)(dx/dt)$. The solution to Eq. (1) subject to the boundary conditions is

$$x(t) = x_0[1 - e^{-t/\tau_0}\{\cosh(\omega t/\tau_0) + (1/\omega)\sinh(\omega t/\tau_0)\}],\tag{2}$$

where $\tau_0 = 2M/B$ and $\omega = [1 - (4MK/B^2)]^{1/2}$.

Fitting this solution (dashed curve) to the actual slip variation $x(t)$ in Fig. 2(c), we find $\eta_{\mathrm{eff}} = 27 \pm 4$ Pa s. This is larger by a factor of some 10^4 than the bulk viscosity of OMCTS, $\eta_{\mathrm{bulk}} = 2.5 \times 10^{-3}$ Pa s at room temperature [14]. It is of interest that, were the viscosity of the shear-melted film during slip to revert to its bulk value—as some simulations suggest [4,18]—the damping term would be negligible and the motion determined by inertial effects alone, yielding the variation shown as the dotted curve in Fig. 2(c). If such an effective viscosity applies [27], the viscous heating $\Delta E_{\mathrm{viscous}}$ due to shear of the film during the slip is given by

$$\Delta E_{\mathrm{viscous}} = \int_{x=0}^{x=\Delta x_0} \eta_{\mathrm{eff}}\frac{(dx/dt)}{D}A\,dx.\tag{3}$$

From Eq. (1), as noted, we expect $\Delta E_{\mathrm{viscous}}$ to equal the elastic energy increment $\Delta E_{\mathrm{spring}}$ in increasing the spring extension from $(x_0 - \Delta x_0)$ to x_0, on going from s to y [25]: $\Delta E_{\mathrm{viscous}} = \Delta E_{\mathrm{spring}} = K(x_0 - \frac{\Delta x_0}{2})\Delta x_0 \approx 4 \times 10^{-13}$ J for the conditions corresponding to the stick-slip cycle in Fig. 2 (see caption to Fig. 2). Explicit evaluation of Eq. (3) [approximating (dx/dt) by its mean value between y and s] indeed shows $\Delta E_{\mathrm{viscous}}$ to be closely similar to $\Delta E_{\mathrm{spring}}$.

We now examine the energy loss via residual momentum transfer at the solidification point s, seen as the ringing excited at this point [Fig. 2(c)]. The decaying oscillations seen, at a frequency $\nu_c \approx 45$ Hz, represent the lateral motion component of that surface attached to the spring. The associated energy is $\Delta E_{\mathrm{osc}} \approx (M_{\mathrm{eff}}\nu_0^2/2)$, where ν_0 is the maximal velocity of the adhered surfaces (following the solidification point s) due to the oscillations (prior to their decay) and M_{eff} is their effective mass. We may take $\nu_0 \approx 2a_0\nu_c$, where, from trace 2c, $a_0 \approx 12$ nm is the maximal amplitude of oscillations prior to their decay, and estimate the combined effective mass of the adhered surfaces and their mounts as $M_{\mathrm{eff}} \approx 15$ g. Thus the energy transferred in exciting these oscillations is $\Delta E_{\mathrm{osc}} \approx 1 \times 10^{-14}$ J. This is only some 2%–3% of the viscous dissipation $\Delta E_{\mathrm{viscous}}$ evaluated from Eq. (3). though other excited

mechanical modes (such as tilt), not revealed in the shear-force traces, could increase this value somewhat. This magnitude of ΔE_{osc} is of interest: In Eq. (1) we ignored contributions to the stored elastic energy during the stick part of the cycle other than that in the shear-spring bending, such as the potential energy ΔE_f stored in the confined film and in the surface layers of the confining mica surfaces [11] prior to the yield. ΔE_f may be estimated [28] as the maximum force just prior to slip, Kx_0, acting over a microscopic dimension δ comparable to the surface lattice spacing, $\delta \approx 1$ nm (since motion over δ brings the film into an equivalent state with respect to the confining surfaces). That is, $\Delta E_f \approx Kx_0\delta \approx 10^{-14}$ J. It is suggestive that this magnitude of ΔE_f, which was omitted from Eq. (1), is indeed close to the residual ringing energy ΔE_{osc}, which is not accounted for by Eq. (1). We emphasize that ΔE_f is very much smaller than the energy ΔE_{el} stored in the shear spring: this self-consistently justifies the use of Eq. (1) to extract the effective viscosity.

The overall picture therefore is one where, during the stick, elastic energy is stored both in the shear spring as well as in the confined solid film and confining surfaces, with the former accounting for the bulk (of order 90% or more) of the stored energy. During the slip, the major part of the stored energy (again, of order 90% or more) is dissipated as viscous heating of the shear-melted film, while the rest is dissipated as mechanical oscillations due to momentum transfer to the external system at the instant of stick.

Examination of other similar stick-slip traces indicates that this conclusion—that the major part of the frictional energy loss in stick-slip is expended in viscous heating of the confined film during the slip cycle [29]—is quite general, at least for this model liquid. We note that this conclusion differs diametrically from earlier suggestions that most of the dissipation during stick-slip occurs at the point of solidification, and that only a small part of it is due to viscous heating during slip [17,23]: the main reason is that in the earlier work [17,23] there was no detailed comparison with the actual stick-slip traces.

In summary, using traces of the frictional response in stick-slip sliding of solid surfaces across a molecularly thin film of a model organic liquid, with exceptionally high time and spatial resolution, we have been able to analyze the resulting motion in detail. The traces demonstrate directly that energy is dissipated both by momentum transfer to the external system at the instant of stick (exciting mechanical oscillations which decay rapidly), and by viscous heating during the slip of the shear-melted film. The analysis shows that, in contrast to earlier suggestions, the dissipative loss over a stick-slip cycle is primarily [O(90%) or more] via the viscous-heating mechanism during slip, with the residual kinetic energy transferred to the system at the point of stick accounting for the remaining, very much smaller part.

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