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INTRODUCTION

Earthquakes are known to result from the stick-slip motion of Earth’s crust, a mechanical instability that is driven by the reduction of friction at the onset of sliding motion (1–3). This difference between static and dynamic friction can be observed in virtually all frictional systems and is captured by the “state” term in rate and state friction theory: The static friction force increases as the logarithm of the time both surfaces have spent in contact (4–10). While rate and state friction theory successfully describes the history and velocity dependence of the friction force in many systems, the microscopic mechanisms that drive this empirical model remain elusive. Adhesive friction originates at—and is presumed to be proportional to—the area of real contact, the molecular area over which surface roughness peaks touch. This area of real contact is often described as a collection of discrete contact points, also known as single-asperity contacts (11–16). At a multisasperity interface, the contact force deforms the surface roughness, leading to elastic deformations at various length scales and local stresses that can approach the yield stress of the materials involved (17–19). Under the influence of these high stresses, contacting materials undergo creep deformation, resulting in growth of the area of real contact with time (4, 20, 21). This geometrical aging is thought to strongly contribute to the logarithmic growth of the static friction force with rest time observed in many friction experiments (4–10). However, direct observation of the area of real contact at multisasperity interfaces is experimentally challenging (4, 10, 22–25) because the frictional interface is buried between two bulk phases and the length scales involved are small.

Here, we show that the transition from static to dynamic friction is not caused by a reduction in the number of touching asperities but by slip weakening of those same asperities. At rest, the asperities harden again: This is the aging that leads to a higher static friction coefficient. This provides a novel explanation for stick-slip phenomena in, e.g., earthquakes.

The fluorescence intensity of the probe molecules is stronger when the local free volume is smaller, enabling us to show that the PS interface is compact at rest (higher σ) and dilates (lower σ) during slip, causing the slip weakening. The difference between static and dynamic friction is therefore not controlled by the size of the area of real contact but by its interfacial shear stress. Since glassy interfaces are ubiquitous (e.g., in earthquakes), this has far-reaching consequences for our understanding of frictional slip.

RESULTS

In the friction experiments, we bring a 600-μm-diameter rough PS sphere into contact with a very flat and smooth float glass coverslip coated with a monolayer of pressure-sensitive molecules (Fig. 1). Contact mechanics models usually map the situation of two rough surfaces in contact onto that of only a single rough surface on a smooth one, which corresponds to the rough-on-smooth situation we investigate here. The sphere is glued off-center to the bottom of a rheometer plate and is lowered into contact with the coverslip (Fig. 1B). Through rotation of the tool, the rheometer can move the sphere tangentially, while simultaneously measuring both normal (load) and tangential (frictional) forces on the contact. In a single friction test, we rotate the rheometer tool such that a speed of 1 μm/s is imposed on the sphere for a total time of 13 s. Initially, however, this deformation is elastically accommodated by the measurement system, and the sphere does not actually slide (linear buildup in Fig. 1A). Once the static friction force is exceeded, macroscopic slip occurs (peak force in Fig. 1A). Static friction now evolves into dynamic friction, and the plateau value of the friction force is the force required to keep the frictional motion going at a constant sliding speed. We repeat the friction measurements consecutively for the same contact and continue to apply a constant normal force of 400 mN between friction measurements. The observation is that not only the static friction but also the dynamic friction increases roughly as the logarithm of the time that has elapsed since the contact was formed (Fig. 1).

As already mentioned, the increase in static friction in time (4–9) is commonly believed to be due to the growth of the area of real contact, attributed to creep deformation of asperities in contact. To probe the contact area, a monolayer of stress-sensitive fluorescent molecules is covalently bonded to the glass substrate (26, 27), and the PS sphere-on-glass contact is illuminated from below by confocal laser scanning microscopy (Fig. 1B). Only molecules that are confined by real contact between sphere and glass emit an increased fluorescence intensity (18, 26) that is captured by the inverted microscope (Fig. 1). The fluorescence image of the contact taken with the microscope therefore

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reveals the area of real contact measured with molecular (nanometer) resolution along the normal (z direction) and diffraction-limited resolution in the in-plane directions. Considerable surface roughness—and therefore contact area structure—can, in principle, exist at lateral scales that are not resolved by the microscope (28). To investigate the presence of such unresolved contact area structure, we complement the contact area observations with contact calculations. As input, the contact calculations use atomic force microscopy (AFM) topographs recorded on the PS sphere surface at the exact location that touches the smooth glass substrate in the visualization experiments. We find that the contact calculation only matches the experimental observation of the area of real contact when both elastic and plastic deformation of the surface roughness are included (Fig. 2). The difference between AFM topographs recorded before and after the contact experiment confirms that there is substantial plastic deformation of the PS surface (Fig. 2B). We previously showed (18) that the nature of this plasticity is strain hardening as the plastic deformation of the PS surface builds up, it hardens. We mimic the limited resolution of the microscope by convoluting the calculated contact areas with Gaussian point spread functions (PSFs) of varying width (Fig. 2C). The important observation is that because of the plastic flattening of the asperities, the contact area does not contain small-scale structure and is therefore fully resolved by the microscope, making the PS-on-glass system ideal for disentangling the relative contributions of geometric and structural aging to the evolution of the friction force.

We track the growth of the area of real contact with time using the fluorescence images. As the sphere is pressed onto the substrate with a constant normal force of 400 mN for a total time of 50 min, we observe a linear increase in contact area with the logarithm of time (Fig. 3A). We now repeat this experiment but additionally impose frictional slip right after the recording of each of the fluorescence images in the time series. Like before, in each of these slip events, a sliding speed of 1 μm/s is imposed on the sphere for 13 s. We continue to measure the age of the contact relative to the moment the contact was formed, rather than relative to the moment when sliding stopped in the previous slip event. The sliding results in an accelerated growth of the area of real contact, particularly for the first slip event that the contact undergoes (just after 32 s). However, the logarithmic growth rate observed in the static experiment also applies to the slide-hold-slide experiment after the first two slip events (Fig. 3A). To visualize the spatial distribution of the contact area growth, we align and subtract the contact areas measured at the beginning (36 s) and end (2992 s) of the static aging experiment (black circles in Fig. 3A). As the normal stress is maximal at the center of a sphere-on-flat contact, substantial growth of the contact area through creep flattening of the roughness can be expected in this region. Unexpectedly, we observe the opposite: Most of the contact area growth occurs at the perimeter of the contact area through flattening of existing contacts (Fig. 3B). We now apply the exact same analysis to the contact areas that were measured before (32 s) and after (78 s) the first slip event in the slide-hold-slide experiment. The growth in contact area in this 46-s time window is roughly equal to the growth observed over 50 min during the static aging experiment. The spatial distribution of this accelerated contact area growth is identical to the one found in the static aging experiment, suggesting that the mechanism that drives the growth in contact area is the same in both cases (Fig. 3B). Furthermore, the direction in which the friction force was applied does not seem to influence the contact area growth; apparently, this growth is not a consequence of shear forces acting on the interface. Instead, we argue that the growth of the area of contact under the influence of externally applied normal and tangential forces is a consequence of the creep deformation of the bulk sphere material: The external forces cause the sphere to flatten in time, thereby creating contacts at the perimeter of the contact area. The central question is then whether the time dependence of static friction (4–9) is caused by this growth of the area of real contact. In Fig. 1B, we plot the static friction force together with the area of real contact measured right before each friction test performed in the slide-hold-slide series. The important observation is that the static friction increases faster than the area of real contact: This means that the friction per unit contact area increases. It increases here from σ = 28 MPa after 30 s to σ = 33 MPa after more than an hour of aging. Both values are within 10% of the shear strength of...
PS (≈30 MPa) so that plastic yielding of the PS seems to be the pertinent physical process. In agreement with this hypothesis, we find that the friction behavior is unchanged when we remove the probe molecules: The friction is controlled by the PS, and the friction mechanism is plastic yielding of a thin PS surface layer (fig. S5) (21, 29, 30).

The physical process that drives the time evolution of $\sigma$ is directly revealed by the probe molecules at the interface. The fluorescence intensity of each pixel within the area of real contact is a local measure of the free volume experienced by the probe molecules (fig. S3). Plotting the total fluorescence intensity emitted by the probe molecules right before each friction test in Fig. 1B, we find that the total fluorescence intensity increases in the same manner as the static friction force; in the area of real contact, the fluorescence becomes brighter. This signals a decrease in the free volume experienced by the interfacial probe molecules; the glassy PS surface becomes more compact during aging. This compaction directly affects $\sigma$; a more compact interface is mechanically stronger and therefore supports a larger static friction force. A detailed study of the contact mechanics, where the static contact area is measured as a function of the normal force and compared with simulations, also shows that, to quantitatively explain the contact mechanics, strain hardening of the asperities needs to be taken into account (18).

The behavior of the friction force is generic for slip events: It increases up to the static limit at which the contact breaks and the sphere starts to slide over the substrate with a smaller friction force (Fig. 4). The transition from static to dynamic friction does not happen over a molecular length scale but gradually occurs as the sphere slides over a distance of several micrometers. To elucidate the physics behind this transition, we consider the area of real contact during interfacial slip. At a rate of 3 frames/s, we record fluorescence images of the contact during a typical slow friction measurement. From these images, we extract the area of real contact, the total fluorescence intensity, and the position of the contact. These quantities are then plotted alongside the simultaneously measured friction force (Fig. 4). As the tangential force builds up toward its peak value, we observe a sudden decrease in contact area, followed by the onset of macroscopic sliding and the

Fig. 2. The area of real contact. (A) Half of the fluorescence image (left) and the mirrored elastoplastic contact calculation (right) of the rough PS on smooth glass contact at a normal force of 392 mN. The plasticity is modeled by increasing the gap at which contact is defined in the purely elastic calculation from 0.1 to 170 nm: This is the only adjustable parameter, and the plain strain modulus is independently measured (18): $E^* = 3.7$ GPa. (B) The AFM topograph of the PS sphere before (left) and after (right; mirrored, areas outside AFM range shown in green) contact with glass shows plastic deformation of the same order. Before the contact experiment, the root mean square roughness of the sphere is 650 nm, and that of the substrate is 2 nm (fig. S4). (C) Calculated contact area as a function of the full width half maximum of the Gaussian PSF with which the contact area is convolved. The AFM and contact calculations both have a pixel size of 32 nm, roughly an order of magnitude lower than the microscopy PSF. The sphere curvature was subtracted from the AFM topographs shown in (B) to highlight the surface roughness. To define the experimental or calculated and convolved area of real contact, we set an intensity threshold (38) and simply multiply the pixel area with the total number of pixels with intensities above the threshold (fig. S1); for all experiments and calculations, the pixel size is smaller than the PSF.

Fig. 3. Contact area growth in time. (A) Contact area as a function of the time the sphere has spent in contact with the glass at $N = 400$ mN (black circles). The contact area growth is accelerated when slip is imposed right after each contact area measurement (red triangles, time measured relative to the moment of contact formation, not to the end of each slip event) but saturates at the same logarithmic growth rate after the first few slip events (black solid lines). Inset images show contact areas (black areas) corresponding to the first data points. (B) Difference images highlighting contact area growth (left) after 50 min of contact (without macroscopic slip) and (right) after the first slip event.

gradual decrease in the friction force from its static to its dynamic value. We first consider the reduction in contact area that is observed before macroscopic slip can be detected. To visualize this change in contact area, we align and subtract the binarized contact images recorded just before and just after the contact area decreases (panel 2-1 in Fig. 5). The difference image reveals two distinct changes: (i) At the perimeter of the overall contact area, trailing edge contacts disappear (red edges on the left in panel 2-1 of Fig. 5) while leading edge contacts grow (blue edges on the right in panel 2-1 of Fig. 5). (ii) Contact patches that are not positioned along the perimeter of the overall contact area all shrink under the influence of the tangential force. Both (i) and (ii) are reversed when the externally applied tangential force is removed: Panel 4-3 in Fig. 5 shows that trailing edge contacts return while leading edge contacts are broken and all other contacts grow (blue edges). Because the changes in contact area at the leading and trailing edge roughly cancel each other out, the drop (rise) in contact area observed before (after) macroscopic slip is a consequence of the shrinking of individual contact patches under the influence of the tangential force. Shear-induced shrinking of the area of contact was, so far, only observed for softer materials in which the effect is much stronger and found to result from a combination of the contacts peeling off at their perimeter (mode I fracture) and contact compression in the tangential loading direction through microslip (25, 31).

The shear-induced reduction in contact area is accompanied by an overall increase in the fluorescence intensity (Fig. 4). As the tangential force builds up, we first observe this increased fluorescence intensity at the perimeter of the contact area from which it propagates toward the center of the contact. To quantify this, we construct radially averaged intensity profiles for the contact images that were recorded before the onset of macroscopic slip and subtract subsequent profiles (Fig. 6). Earlier experiments (32) and simulations (33) have demonstrated the nucleation and propagation of slow rupture fronts along frictional interfaces right before the onset of macroscopic motion. These are driven by slow slip, separate microslipping regions of the interface from sticking regions, and can move at velocities orders of magnitude smaller than the material’s shear wave speed. We interpret the presented intensity waves in this context since microslip can be expected to initiate at the perimeter of the sphere-on-flat contact, where the shear stresses are maximal and the normal stresses are minimal. The increased fluorescence intensity measured at those contact patches that have been subject to microslip indicates that

The stick-slip transition. Normalized contact area (A), total fluorescence intensity (I), contact displacement (D), and friction force (F) measured during the 78-s slip event also shown in Fig. 1. The arrows and numbers indicate which data points were used for the difference images in Fig. 5. Contact displacement is measured by optimally overlapping subsequent fluorescence images to the first fluorescence image in the series. Inset shows shear stress $\sigma$—defined as the ratio of friction force to real contact area—corresponding to the data in the main figure.

Fig. 4. The stick-slip transition.

Fig. 5. Difference images corresponding to Fig. 4. Top: Newly formed contacts are shown in blue, and broken contacts are shown in red. Bottom: Fluorescence intensity changes. Before sliding starts (2-1), existing contacts shrink under the influence of the tangential force (contact edges turn red), while a slight rolling motion of the sphere causes trailing edge contacts (left) to break and leading edge contacts (right) to form. During macroscopic slip (3-2), leading edge contacts continue to grow, while trailing edge contacts disappear. As sliding stops and the tangential force is removed from the contact (4-3), contacts grow again at the asperity level (contact edges turn blue), while the sphere rotates back to restore the trailing edge contacts that were broken and to remove the leading edge contacts that were formed.
and increases at rest. This process is reversed when a sufficient tangential force is applied and the contact slips. The observation that the static friction force is larger than the dynamic one is quite general, the detailed mechanism that causes the difference may not be universal; AFM experiments have shown that the formation of chemical bonds across a frictional silica-silica interface can cause a logarithmic increase in static friction with stop time (8, 34); generally, chemical or physical adhesion may be time dependent and change the static and dynamic friction force (35). We have shown here that $\sigma$, the friction per unit contact area, increases at rest and decreases during slip because of the compaction and dilation, respectively, of amorphous material at the frictional interface. This behavior can be expected at any interface involving contact pressures that approach the material yield stress. Earthquakes, in particular, satisfy this condition as evidenced by the molten rock found along fault lines (36, 37).

**MATERIALS AND METHODS**

The setup consists of a ZEISS Axiovert 200M LSM 5 Pascal inverted laser scanning confocal microscope on top of which an Anton Paar DSR 301 rheometer is mounted using a custom-made frame. Microbeads TS-500 general-purpose PS spheres of 600 $\mu$m in diameter are first roughened by shaking in a container with sandpaper walls (18) and then glued to the rheometer tool using epoxy glue. The distance between the rotation axis of the rheometer and the center of the sphere is measured by microscopy. Using this distance, torque and rotation angle values directly measured by the rheometer can be converted to friction forces and distances, respectively.

We covalently attached rigidochromic probe molecules to glass coverslips. The latter were functionalized with N-(2-aminoethyl)-3aminopropyl-trimethoxysilane, and the probes were attached using an amide bond (18, 26).

To image the real contact area (18), we first aligned the focal plane of the microscope with the frictional interface. Fluorescence images were then point-scanned using 488-nm excitation light. Contacts were immersed in formamide to avoid strong light scattering at the interface and improve image quality. The area of real contact were measured by applying an Otsu (38) threshold to the fluorescence images and multiplying the number of high-intensity pixels with the pixel area (fig. S1). To calculate the total fluorescence intensity $I$, we first measured the average background fluorescence, defined as the average intensity of pixels in the fluorescence image that are not part of the area of real contact. We then multiplied this average background pixel intensity with the number of pixels in the image and subtracted this from the total image intensity to obtain $I$.

In fig. S2, we display the simultaneously measured fluorescence and reflection image of a PS-on-glass contact. At those locations where the gap between the sphere surface and the glass surface is smaller than approximately 80 nm, reflections are suppressed; this is the gap at which the first Newton ring around the contact can be observed (18). The reflection image displays a contact area that is similar but somewhat larger than that observed using the fluorescent molecules, as can be expected. More details on the comparison of the fluorescence and reflection observations can be found in (18).

**DISCUSSION**

In conclusion, the time dependence of friction is controlled by both the size and the mechanical strength of the area of real contact. In our rough-on-smooth system, this contact area is initially formed by elastoplastic deformation of the sphere roughness (18) and can subsequently change because of creep deformation of the bulk material. The friction per unit contact area is set by the density of the interfacial PS
square root of $P$ (fig. S3), indicating that a larger contact pressure results in stronger confinement of the rigidochromic molecules at the interface. This is confirmed by comparison of the contact data with fluorescence intensities measured while the rigidochromic molecules are immersed in an organic solvent under varying hydrostatic pressure; in solution, the molecules give an identical response. We therefore concluded that the fluorescence intensity reflects the local free volume, which is reduced when the pressure is increased (39). As evidenced by Fig. 4, the local free volume is affected not only by normal stresses but also by tangential stresses.

Through AFM, we imaged the surface topography of the glass coverslips that were used as substrates as well as the PS spheres. The coverslips with and without a monolayer of probe molecules covalently attached are very smooth compared to the PS spheres (fig. S4).

We performed friction measurements following the protocol reported above on glass surfaces with and without a monolayer coating of probe molecules and found identical frictional behavior (fig. S5). Furthermore, we experimentally validated that the presence of a low-viscosity immersion fluid does not affect the frictional behavior (fig. S5C).

The frictional shear stress $\sigma = F/A$ corresponding to the measurements shown in Fig. 1 is plotted in fig. S6. We observed that the dynamic shear stress is constant while the static shear stress increases as the contact ages.

In fig. S7, we plotted the normal force measured during the slip event shown in Fig. 4. We found that the normal force remains constant during the transition from static to dynamic friction. The contact calculations included here were performed using the Tribology Simulator that is publicly available at www.tribonet.org.

REFERENCES AND NOTES

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