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Probing and tuning frictional aging at the nanoscale

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Time-dependent increase of frictional strength, or frictional aging, is a widely observed phenomenon both at macro and nanoscales. The frictional aging at the nanoscale may result from nucleation of capillary bridges and strengthening of chemical bonding, and it imposes serious constraints and limitations on the performance and lifetime of micro- and nanomachines. Here, by analytical model and numerical simulations, we investigate the effect of inplane oscillations on friction in nanoscale contacts which exhibit aging. We demonstrate that adding a low amplitude oscillatory component to the pulling force, when applied at the right frequency, can significantly suppress aging processes and thereby reduce friction. The results obtained show that frictional measurements performed in this mode can provide significant information on the mechanism of frictional aging and stiffness of interfacial contacts.

The ability to control and manipulate friction during sliding is extremely important for a large variety of applications. Development of novel efficient methods to control friction requires understanding microscopic mechanisms of frictional phenomena. One of the main difficulties in understanding and predicting frictional response is the intrinsic complexity of highly non-equilibrium processes going on in any tribological contact, which include detachment and reattachment of multiple microscopic junctions (bonds) between the surfaces in relative motion[1–8]. Even for an apparently sharp AFM tip sliding on a crystalline surface, the actual interface consists of an ensemble of individual atomic junctions[9,10]. On larger scales the multicontact picture becomes even more obvious. Friction is not simply the sum of single-junction responses, but is influenced by temporal and spatial dynamics across the entire ensemble of junctions that form the frictional interface. The way how individual junctions can be averaged to yield friction response has been the focus of intense research in the past decades[7,8,11–16]. One of the highly debated topics in this field is the origin of time-dependent increase of frictional strength, or frictional aging[15,17–20]. Frictional aging has been observed for both macroscopic and nanoscale contacts[17–19], and typically, in these systems the static friction grows logarithmically with time when the surfaces are held in stationary contact. It is generally assumed that frictional aging can be caused by increase of contact area due to plastic deformation and creep as well as by time-dependent strengthening of bonding at asperity contacts. The latter mechanism may involve nucleation of capillary bridges[21–25] or formation of hydrogen or covalent bonding[15,19].

One unique path to controlling and ultimately manipulating the friction forces between material interfaces is through externally imposed oscillations of small amplitude and energy. Validity of this approach has been demonstrated experimentally at nano[26–28] and macroscales[29–31] and numerically with minimal models[32,33] and molecular dynamics simulations[34,35]. In spite of these promising experimental and numerical contributions, we still lack understanding of the most basic questions: (1) What are the fundamental mechanisms behind these phenomena? (2) In what types of systems should these methods be applicable? (3) What are the relations between optimal values of the control parameters and the physical properties of the tribological system?

In this Report we focus on the effect of inplane oscillations on friction in nanoscale contacts which exhibit aging. We demonstrate that adding a low amplitude oscillatory component to the pulling force, when applied at the right frequency, can significantly suppress the formation of multiple junctions and thereby reduce friction. The results obtained show that frictional measurements performed in this configuration can provide significant information on the kinetics of junction formation and stiffness of interfacial contacts.

Results

In order to mimic the AFM measurements, we consider a rigid tip with mass M and center-of-mass coordinate X that is pulled along the surface, through a spring of spring constant, K_d. The tip interacts with the underlying surface through an array of microscopic junctions representing molecular bonds or capillary bridges, as shown in Fig. 1 (See Methods for equations of motion and details of the model and simulations). This picture has emerged from recent simulations[9] and experiments[10] which proposed that even apparently smooth nanoscale contacts can be viewed as consisting of discrete atomic junctions.

Formation of molecular bonds and capillary bridges is a thermally activated process, and its rate, k_{on}, is given by the equation:

$$k_{on}^{(i)} = \omega_{on} \exp\left[-\Delta E_{on}^{(i)} \big/ k_B T\right] \tag{1}$$

where ω_{on} and $\Delta E_{on}^{(i)}$ are the attempting frequency and the barrier height for the formation of i-th junction. The systems exhibiting frictional aging are characterized by a broad distribution of the barrier heights[15,19,21–25]. Here, for simplicity we assume a uniform distribution of barrier heights above a bottom threshold, ΔE_{on}^{min}. Then a fraction of junctions with $\Delta E_{on}^{(i)} < \Delta E_{on}^{*}$ is equal to $s_E\left(\Delta E_{on}^{*} - \Delta E_{on}^{min}\right)$, with s_E being the density of the distribution. Qualitative conclusions of this work are not sensitive to a particular choice of the distribution. The rupture of junctions can be also considered as a thermally activated process with an activation barrier, ΔE_{off}, which is force dependent and diminishes as the force acting on the junction, f_i, increases[36]. The simulations discussed below have been performed for the values of microscopic parameters which are typical for friction phenomena dominated by formation of capillary bridges[22–25].

A broad distribution of barrier heights $\Delta E_{on}^{(i)}$ leads to a long-scale dependence of number of bound junctions on the time of contact between the tip and the substrate, and thereby to the effect of frictional aging. The average number of junctions, $N_{mf}(t)$, which are formed during the time t, can be estimated using the mean-field approach that gives[24]:

$$N_{mf}(t) = N_0 \begin{cases} t/\tau, & t < \tau \\ ln(t/\tau) + \gamma, & t > \tau \end{cases} \tag{2}$$

where $N_0 = s_E k_B T$, $1/\tau = \omega_{on} \exp\left[-\Delta E_{on}^{min} / k_B T\right]$ and $\gamma \simeq 0.5572$ is the Euler-Mascheroni constant. The number of junctions as a function of time shows a linear behavior at short times and logarithmic one at longer times.

In order to study the effect of inplane oscillatory modulation on friction, we add a low amplitude oscillatory component to the ramped forces. Then the displacement of the stage reads as $X_d = V_0 t + A\cos(2\pi\nu t)$, where V_0 is a constant velocity, ν and A are the frequency and amplitude of oscillations. When the tip is pulled with constant velocity, time series of the spring force exhibit stick-slip behavior corresponding to collective rupture and reattachment of microscopic junctions (see Figs. 2a, b). Once small harmonic perturbations are introduced to the ramped loading, this picture changes significantly. Figures 1b and 2a show that for frequencies of oscillations above a threshold one, ν_{th}, the force traces represent a set of alternating segments of stick-slip oscillation and low friction sliding, which are marked as t_{s-s} and t_{lag}, respectively. For high frequencies (green curve in Fig. 2a) the stick-slip oscillations are completely suppressed and the spring force remains low over the entire time of simulations shown in Fig. 2a. In order to elucidate the mechanism of reduction of friction we show in Fig. 2b the effect of oscillations on the time-dependent number of bound junctions, $N(t)$. While in the absence of oscillations $N(t)$ starts to grow monotonically directly after the slip event, application of small-amplitude oscillations suppresses the formation of junctions during finite time-intervals, t_{lag}. This effect results in a low friction regime of motion corresponding to uncorrelated rupture of small clusters of junctions (See Supplemental material for effect of oscillations on rupture). Because of stochastic nature of junction formation, the regime of low friction motion persists only for a finite time, t_{lag}, until the number of junctions formed during a half-period of oscillations becomes larger than a critical value N_c that cannot be ruptured by the oscillatory component of force, as shown by dashed-dotted line in Fig. 2b. Then, oscillatory modulations become inefficient, and the number of bound junctions grows until the ramped component of the loading force causes the collective rupture of junction similar to what happens in the absence of modulations. The value of loading force corresponding to the collective rupture in the presence of modulation is only slightly below the maximal force for constant velocity pulling (see Fig. 2a) and it depends weakly on ν. However, the length of the time-intervals of low friction, t_{lag}, increases rapidly with ν and as a result the average friction force decreases.

Figure 2c shows the ratio $\langle t_{lag}\rangle/\langle t_{s-s}\rangle$ as a function of ν for two amplitudes of oscillation, where $\langle t_{lag}\rangle$ and $\langle t_{s-s}\rangle$ are the mean values of t_{lag} and t_{s-s} which have been calculated by averaging over a large number of realizations. Inplane oscillations induce nonzero time-intervals of low friction, t_{lag}, only for frequencies exceeding a threshold value, ν_{th}, which decreases with the amplitude of oscillations.

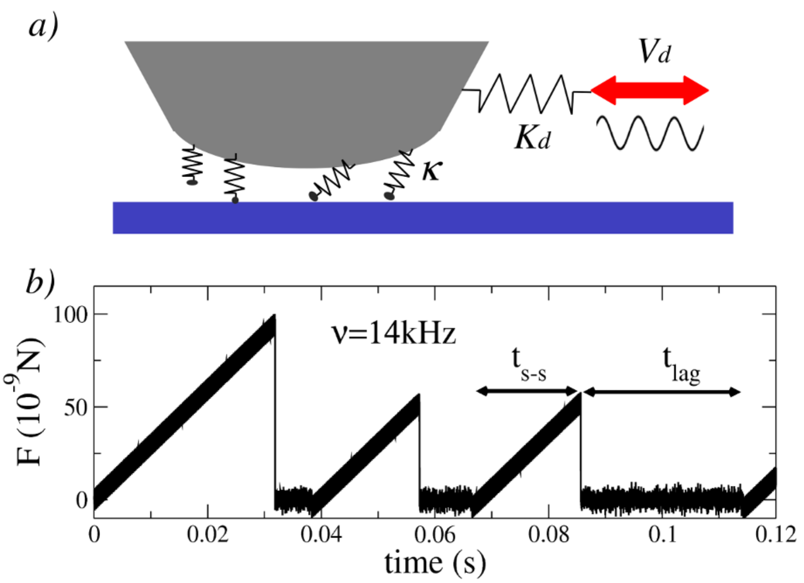

Figure 1 | (a) Sketch of the model used to simulate the effect of inplane oscillatory modulations on nanoscopic friction. The tip is pulled by the force that includes a low amplitude oscillatory component. (b) Force trace corresponding to lateral modulations with the amplitude $A = 1$ nm and $\nu = 14$ kHz. The trace shows alternating segments of stick-slip and low friction motion with durations t_{s-s} and t_{lag}, respectively.

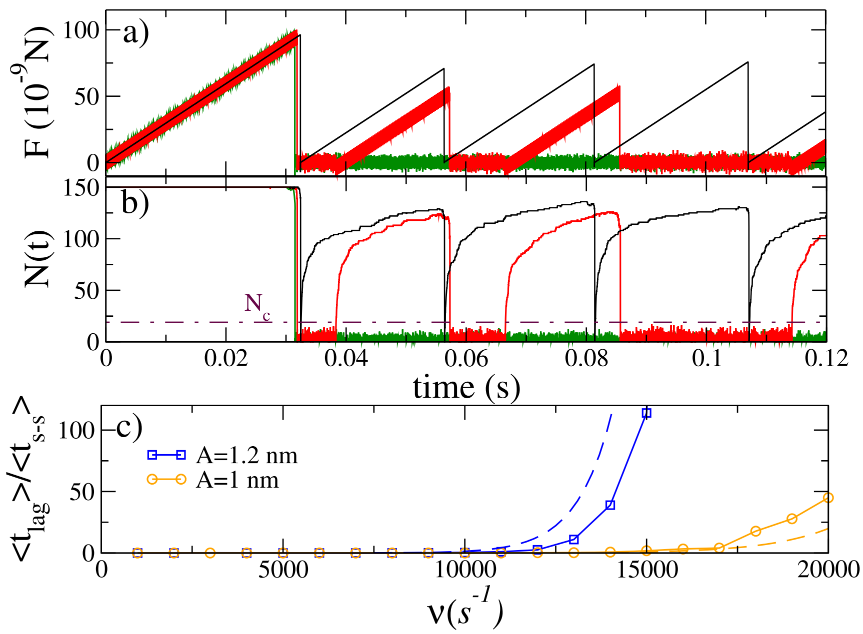

Figure 2 | (a) Force traces and (b) time-dependent number of bound junctions, $N(t)$, calculated in the absence of oscillations (black curves) and including the oscillatory component of the force with $A = 1$ nm and $\nu = 14$ kHz (red curves) and $\nu = 19.6$ kHz (green curves). Dashed-dotted line in the panel (b) indicates the maximal number of junctions, N_c, which can be ruptured by the oscillatory modulations with the amplitude $A = 1$ nm estimated using Eq.(4). (c) Ratio $\langle t_{lag}\rangle/\langle t_{s-s}\rangle$ as a function of ν for two amplitudes of oscillations which are indicated in the figure. Solid and dashed curves present results of simulations and analytical theory, respectively.

Above the threshold frequency, $\langle t_{lag}\rangle/\langle t_{s-s}\rangle$ increases sharply with ν and the average friction force is reduced. It should be noted that over the entire range of data presented in this Report the amplitude of the applied force oscillations was lower than the tenth of the force needed to initiate a slip of the tip in the case of constant velocity pulling. Thus, despite strong effects on stick-slip dynamics the perturbations are decidedly small.

Figure 3 presents the average friction force, $\langle F\rangle$, and the average number of bound junctions, $\langle N(t)\rangle$, as functions of frequency and amplitude of oscillations. One can see that for a given amplitude $\langle N(t)\rangle$ decreases steeply above a threshold frequency, $\nu_{th}(A)$, and the friction force follows this behavior. Figures 3b,d demonstrate a similar reduction of $\langle N(t)\rangle$ and $\langle F\rangle$ with increase of the amplitude of oscillations for a given frequency. Thus, application of small amplitude oscillations with frequencies of few kHz allows to reduce the friction force by more than one order of magnitude.

The mechanism of reduction of friction discussed here differs significantly from those suggested in previous works[27–35] on the

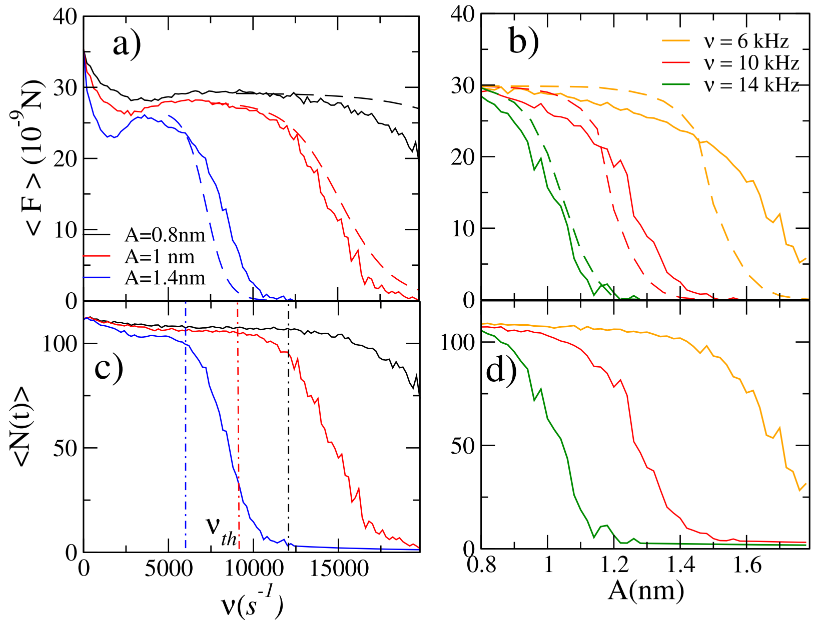

Figure 3 | Average friction force, $\langle F\rangle$, as a function of frequency (a) and amplitude (b) of oscillations, and the corresponding variations of average number of bound junctions, $\langle N(t)\rangle$, reported in panels (c) and (d). Solid and dashed curves in (a) and (b) show results of simulations and analytical calculations according to Eq.(8), respectively. Vertical dashed-dotted lines in (c) present analytical estimations of the threshold frequency, ν_{th}, corresponding to different values of the amplitude A.

effects of oscillatory modulations on friction. It can operate only in tribological contacts exhibiting aging where times for junction formation are widely distributed or there is a long time-scale strengthening of junctions. In these systems a frequency of force modulations can be chosen in a way that only a small number of junctions is formed during the half-period of oscillations, and these "fresh" junctions can be ruptured by the oscillatory component of the loading force. Thus, small inplane oscillations are able to prevent the formation of multiple junctions and reduce friction. In order to achieve similar reduction of friction in contacts characterized by a narrow distribution of times for junction formation (in the absence of aging) much higher amplitudes or/and frequencies of modulation are required.

The main features of numerical results presented above can be reproduced by an analytical model based on a mean field description of ensemble of junctions. When the tip is pulled with the velocity $\dot{X}_d$, the force, f_i, acting on a junction grows with a time-dependent rate $K_{eff}\dot{X}_d/N(t)$, where $K_{eff}(t)=\dfrac{N(t)\kappa K_d}{N(t)\kappa+K_d}$ is the effective stiffness of the system that includes the pulling spring, K_d, and the molecular junctions with total stiffness $N(t)\kappa$. Assuming that all junctions are ruptured simultaneously at time t, when the force acting on one of them approaches f_c, the condition for a collective rupture can be written as

$$\int_0^t \frac{\kappa K_d}{K_d+N(t)\kappa}\dot{X}_d dt = f_c \tag{3}$$

This equation allows to estimate the key parameters which define the effect of oscillations on friction, such as: the maximal number of junctions, N_c, which can be ruptured by the oscillatory modulations, and the threshold frequency, ν_{th}, above which the force oscillations produce low friction segments of motion. Considering that rupture occurs at a time corresponding to the maximum of the oscillatory force, $t = T_\nu = 1/(2\nu)$, and using Eq.2 for $N(t)$ we get the following approximate equation for $N_c = N(T_\nu)$:

$$N_c\frac{\kappa}{K_d}\left(1-\frac{f_c}{4AK_d}N_c\right)=\ln\left(1+\frac{\kappa}{K_d}N_c\right) \tag{4}$$

Then, for a linear regime of growth of $N(t)$ in Eq.(2) the threshold frequency can be estimated as $\nu_{th}\simeq\frac{N_0}{2\tau N_c}$. Estimations of N_c and $\nu_{th}(A)$ reported in Figs. 2b and 3c, respectively, show a good agreement with the results of simulations.

In order to describe the effect of oscillations on friction we have to consider a stochastic nature of junction formation. The number of junctions formed during the half-period of oscillations fluctuates around an average value $N(T_\nu)$ given by Eq.(2). While $N(t)$ is below the critical value, N_c, the spring force, $F(t)$, remains low but if during one of the oscillations $N(T_\nu)$ exceeds N_c the force oscillations become inefficient and $F(t)$ grows. The probability, $P(m, t)$, of formation of m junctions in time t is given by a sum of probabilities of formation of all possible clusters of junctions of size m. In the case of identical junctions, $P(m, t)$ can be easily calculated taking into account that there are $\dfrac{N_s!}{(N_s-m)!m!}$ clusters of size m, where N_s is a total number of available junctions. However, in systems exhibiting aging, different junctions have different barrier heights $\Delta E_{on}^{(i)}$, and correspondingly different probabilities of formation which can be calculated as, $P_i(t)=1-\exp\left[-k_{on}^{(i)}t\right]$, where $k_{on}^{(i)}$ is the rate of formation of i-th junction given by Eq.(1). Then, the probability $P(m, t)$ can be found using the recursive equation[37]

$$P(m,t)=\begin{cases}\prod\limits_{i=1}^{N_s}(1-P_i(t)), & m=0\\ \frac{1}{m}\sum\limits_{i=1}^{N_s}(-1)^{i-1}P(m-i,t)Q_i(t), & m>0\end{cases} \tag{5}$$

where $Q_i(t)=\sum_{j=1}^{N_s}\left(\frac{P_j(t)}{1-P_j(t)}\right)^i$. The average length of the time-interval, $\langle t_{lag}\rangle$, during which the number of connected junctions is below N_c, and the spring force remains low, can be calculated as

$$\langle t_{lag}\rangle=\frac{T_\nu}{\tilde{P}(N_c,T_\nu)} \tag{6}$$

where $\tilde{P}(N_c,T_\nu)=\sum_{i=N_c}^{+\infty}P(i,T_\nu)$ is a probability that no less than N_c junctions are formed during the time T_ν. As the frequency ν increases, the probability $\tilde{P}(N_c,t_\nu)$ decreases rapidly and $\langle t_{lag}\rangle$ increases. Dashed curves in Fig. 2c present results of analytical calculations of $\langle t_{lag}\rangle$ in Eq.(6), which agree qualitatively with the results of numerical simulations discussed above.

Considering that in the presence of oscillatory modulations the force series represent the set of alternating segments of stick-slip oscillations and low friction sliding (see Figs. 1–2), the average friction force can be estimated as

$$\langle F\rangle=\frac{\langle F_{s-s}\rangle\langle t_{s-s}\rangle}{\langle t_{s-s}\rangle+\langle t_{lag}\rangle} \tag{7}$$

where $\langle t_{s-s}\rangle/(\langle t_{s-s}\rangle + \langle t_{lag}\rangle)$ is a fraction of time corresponding to the stick-slip state of motion, $\langle F_{s-s}\rangle$ is the average force experienced by the tip in that state, and the contribution of the low friction sliding was neglected. Approximating by $\langle F_{s-s}\rangle\simeq\frac{K_dV_d\langle t_{s-s}\rangle}{2}$, we get the following equation for the average friction force

$$\langle F\rangle(\nu,N_c)=\frac{2\langle F_{s-s}\rangle^2}{2\langle F_{s-s}\rangle+K_dV\langle t_{lag}\rangle} \tag{8}$$

With increase of A or/and ν the length of low friction segments $\langle t_{lag}\rangle$ grows and the friction force decreases rapidly. Dashed curves in Figs. 3a, b show $\langle F\rangle$ as functions of frequency and amplitude of modulations which have been calculated according to the Eq.(8). The analytical results are in qualitative agreement with numerical simulations. It should be noted that the mean field description given by Eqs.(3)–(8) assumes that all bound junctions are ruptured simultaneously under the action of the pulling force and $\langle F_{s-s}\rangle$ is independent on A. These assumptions are inaccurate for high amplitudes and frequencies of oscillation, and in this range of parameters the analytical results deviate essentially from the numerical ones (see Figs. 3a, b).

Discussion

Our simulations suggest that applying small-amplitude inplane oscillations to the stage of AFM one can give important information on the kinetics of frictional aging and the stiffness of molecular junctions. In order to do this we propose to bring the oscillating tip in contact with the surface and to follow the time variation of the amplitude of the tip oscillations. Figure 4 shows the tip motion and the kinetics of junction formation for the tip that is driven at the velocity $V_d = -2\pi\nu A\sin(2\pi\nu t)$ and brought in the contact at $t = 0$. The tip exhibits high amplitude oscillations for the time interval $t < t_{lag}$ during which the formation of junctions is suppressed by oscillations. In this regime the amplitude of tip oscillations, A_{tip}, is only slightly below the driving amplitude $A = 1$ nm, as shown in Fig. 4a. When the number of junctions formed during a half-period of oscillations exceeds the critical one, N_c, (inset in Fig. 4b) the cluster of bound junctions starts to grow and the amplitude, A_{tip}, is greatly reduced as shown in the inset to Fig. 4a. Considering the balance of forces acting on the tip, $A_{tip}(t)N(t)\kappa = (A - A_{tip}(t))K_d$, the time-variation of the amplitude of tip oscillations can be related to the time-dependent stiffness of the cluster of bound junctions,

$$N(t)\kappa=K_d\left(\frac{A}{A_{tip}(t)}-1\right) \tag{9}$$

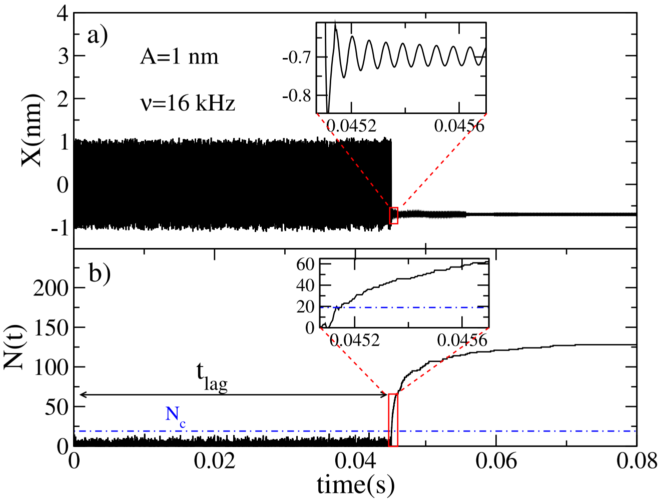

Figure 4 | (a) Displacement of the tip, $X(t)$, as a function of time in response to harmonic driving. The tip is brought in contact with a substrate at t = 0. (b) Dynamics of junction formation, $N(t)$. Dashed dotted line in the panel (b) indicates the maximal number of junctions, N_c, which can be ruptured by the oscillatory modulations with the amplitude A = 1.0 nm. The insets zoom in the region corresponding to the transition from the low to high friction state.

Thus the proposed measurements can provide direct information on the kinetics of frictional aging and the stiffness of molecular junctions. Additional information on the distribution of heights of barriers for junction formation can be obtained comparing the measured values of t_{lag} with the results of calculations according to Eq.(6). This comparison allows to estimate the main parameters of the distribution of the barrier heights, such as the minimal barrier height, ΔE_{on}^{min}, and the density, s_E.

In conclusion, we have investigated the effect of inplane oscillations on friction in nanoscale contacts exhibiting aging. The time-dependent strength of the contacts has been described by introducing a broad distribution of barrier heights for formation of microscopic junctions. Using an analytical model and numerical simulations we demonstrated that adding a low amplitude oscillatory component to the pulling force, when applied at the right frequency, can significantly suppress formation of multiple junctions and thereby reduce friction. Our simulations suggest that applying small-amplitude inplane oscillations to the stage of AFM one can get direct information on the kinetics of frictional aging and the stiffness of molecular junctions.

Methods

The motion of the driven tip is described by the equation:

$$M\ddot{X} + \eta\dot{X} - F_{bond} + K_d(X - X_d(t)) = 0 \qquad (10)$$

where X_d is the position of the microscope support stage, F_{bond} is the force produced by the junctions and η is a damping coefficient. In a wide range of parameters the results of calculations are independent of the value of η. The instantaneous lateral spring force, which is the main observable in friction experiments, reads as, $F = K_d(X_d(t) - X)$, and its time average is equal to the friction force $\langle F \rangle$.

As long as a junction is intact it responds elastically to the applied force, and we model it as an elastic spring with stiffness, κ[14,24]. Under the action of a pulling force, the junctions are stretched in the lateral direction with a velocity equal to the velocity of the tip, and therefore the tip experiences the force, $F_{bond} = -\Sigma_i f_i$, where $f_i = \kappa l_i(t)$, and $l_i(t)$ is the time-dependent junction length. In the case of capillary bridges and covalent bonding[15,24] the height of potential barriers for rupture is much higher than thermal energy, and the rupture occurs preferentially when the force f_i is close to a threshold value, f_c, for which the potential barrier, $\Delta E_{off}(f_c)$ vanishes. Here we consider the systems which satisfy this condition. The existence of the threshold rupture force, f_c, introduces a typical length scale f_c/κ, that is a maximal elongation of the junction.

Computer simulations of the system have been performed using a Velocity-Verlet algorithm. In simulations the nanoscopic tip with mass $M = 5 \cdot 10^{-11}$ kg is pulled by a spring with a spring constant $K_d = 6$ N/m, moving at velocity $V_0 = 5 \cdot 10^{-7}$ m/s. The tip kinetic energy is dissipated through a damping coefficient $\eta = 2 \cdot 10^{-5}$ kg/s. Energy barriers for formation of microscopic junctions are distributed with a constant density, $s_E = 6.6 \cdot 10^2$ eV^{-1}, above a bottom threshold, $\Delta E_{on}^{min} = 0.2eV$. Microscopic junctions have a stiffness $\kappa = 4$ N/m and are formed with an attempting frequency $\omega_{on} = 10^7$ s^{-1}. A threshold force for rupture of junctions has been chosen to be $f_c = 1$ nN. Temperature has been fixed at $T = 370$ K.

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Author contributions

R.C. performed the calculations. All three authors contributed to the analysis of the results. R.C. and M.U. wrote the manuscript.

Additional information

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