

Time Strengthening of Crystal Nanocontacts - Supplemental Material

UHV Friction Measurements

The analysis of the contact stiffness during stick-slip for different sample temperatures was based on experiments performed in [1](stick-slip on NaCl) and [2](stick-slip on HOPG). In short, the friction experiments were performed under clean ultrahigh vacuum conditions with an atomic force microscope as a function of sample temperature at cryogenic temperatures from about 100 K to room temperature. All measurements were done at a base pressure of $p < 5 \times 10^{-10}$ mbar. The pull-off force was measured at all temperatures to verify that no sudden changes of the tip-sample adhesion occurred, which would indicate significant alterations of the tip geometry.

On one hand the sample surface was a NaCl(001) single crystal, which was freshly cleaved in air and then quickly inserted into the vacuum chamber. Subsequently the sample was annealed in UHV at around 520 K for about 1 hour. This procedure results in atomically flat and clean terraces on which the friction was measured, avoiding any step edges. The second analyzed sample was HOPG, which was cleaved inside the UHV system shortly before the experiments.

As force sensors single crystalline, rectangular silicon cantilevers (LFMR, Nanosensors, Germany) were used, with a nominal tip radius of 20 nm. The procedure described in [3] was used to determine the normal and torsional spring constants of the cantilevers. The lateral forces were calibrated after the method described by Bilas et al. [4].

The lateral force maps were measured on a small area of $3 \times 3 \text{ nm}^2$ of the NaCl(001) and HOPG surfaces. The normal forces during the friction measurements were 3 nN (NaCl) and 30 nN (HOPG). After the measurements were completed at one temperature, the tip-sample adhesion was determined by measuring the pull-off forces. Experiments were performed consecutively with the same cantilever from low to high temperatures.

The contact stiffness during the stick phases of the stick slip measurements on NaCl and HOPG were evaluated according to the protocol described in the main text. Additionally to the results on NaCl in the main text, here Fig. 1 shows the resulting contact stiffness k on HOPG as a function of sliding velocity for temperatures 109K, 155K, 202K, 256K and 295K. In contrast to the measurements on NaCl, at each temperature k remains constant for

all sliding velocities and there are no apparent ageing effects. This characteristic distinction between the two systems is consistent with our conclusion of ageing being an effect driven by atomic attrition. While ageing is possible for a tip covered by NaCl (cf. main text), scanning on the wear resistant HOPG does not generate tip conditions where mobile atoms can facilitate the ageing process.

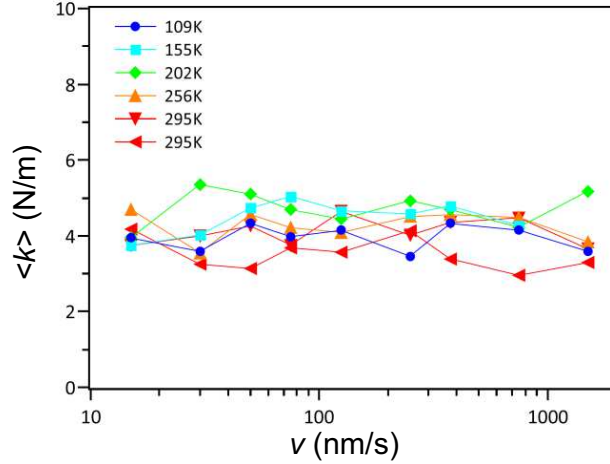


FIG. 1: Average lateral stiffness $\langle k \rangle$ measured for a contact formed by an AFM tip sliding on HOPG as a function of velocity for different sample temperatures (lateral stiffness derived using data from [2])

Numerical Simulation of Stick-Slip with Contact Ageing based on the Langevin Equation

In order to numerically study the dynamics of the ageing contact at finite temperature, we start from the Langevin equation

$$m \frac{d^2 x}{dt^2} + m\gamma \frac{dx}{dt} + \frac{\partial U(R, x, t)}{\partial x} = \xi(t).$$

Here, x and $R = vt$ represent the tip apex and support position, respectively, m is the tip mass and γ is a damping coefficient. $\xi(t)$ accounts for the thermal noise with $\langle \xi(t)\xi(t') \rangle = 2m\gamma k_B T \delta(t - t')$. $U(R, x, t)$ is the total effective potential:

$$U(R, x, t) = -U_{\text{int}}(t) \cos \frac{2\pi x}{a} + k(t) \frac{(vt - x)^2}{2}.$$

Contact ageing is introduced in the model assuming that U_{int} and k depend on time as

$$U_{\text{int}} = U_0(1 + \alpha f), \quad k(t) = k_0 \sqrt{1 + \alpha f},$$

with

$$f(t) = 1 - \exp\left(-\frac{t}{\tau}\right), \quad \tau = \tau_0 \exp\left(\frac{U_{\text{attr}}}{k_B T}\right).$$

The numerical values used in our simulation are given in the caption of Fig. 4.

The integration algorithm that we used is a version of the Runge-Kutta algorithm adapted to stochastic differential equations as described in Refs. 5 and 6.

From the simulations, we can obtain $F(t) = k(vt - x)$ and t_c , defined as the time when the slip event occurs. The latter is easily estimated when a sudden displacement of the tip position is detected. In this moment the potential amplitude and the spring constant are reset to the initial values U_0 and k_0 respectively. The static friction force $\langle F_s \rangle$ is estimated as $\langle F(t_c) \rangle$. The kinetic friction force F_k is estimated as $\langle F(t) \rangle$. The average values are computed over 1000 runs of the simulations. We also observe that during stick $F(t) \approx kvt\eta/(1 + \eta)$ (see Fig. 6).

Analytical Estimation of the First Slip Time at any Velocity and Temperature

In this section, we present the derivation of a theoretical approximation for the average contact time $\langle t_c \rangle$ when the first slip occurs at arbitrary v and T values. At $T = 0$ the contact time is approximately given by (see Ref. 34 in the manuscript for the derivation)

$$t_c \approx \frac{2\pi U_0}{k_0 a v} \sqrt{1 + \alpha f(t_c)} + \frac{a}{4v}.$$

At finite temperature, similar to the friction force, $\langle t_c \rangle$ is considerably reduced by thermal fluctuations. As discussed in the manuscript, the average friction force is given by

$$\langle F_s(v, T) \rangle \approx F_s^0 - F_s^0 \left(\frac{k_B T}{2\sqrt{2}U_{\text{int}}} \log \frac{1.78\eta k_B T}{3a\gamma m v} \right), \quad (1)$$

where F_s^0 , U_{int} and η are evaluated at $\langle t_c \rangle$. To estimate $\langle t_c \rangle$ the known approximation of the force vs. time dependence during the stick-phase can be used (see caption of Fig. 6):

$$\langle F_s \rangle \approx F(\langle t_c \rangle) = k(\langle t_c \rangle) \frac{\eta(\langle t_c \rangle)}{1 + \eta(\langle t_c \rangle)} v \langle t_c \rangle. \quad (2)$$

The system of two equations (1) and (2) with the two unknowns $\langle F_s \rangle$ and $\langle t_c \rangle$ can be solved for any values of parameters. In Figure 2 the values of $v\langle t_c \rangle$ (lines) obtained in this way are

compared to the values computed numerically by solving the Langevin equation (points). A good agreement is found at all velocities and temperatures.

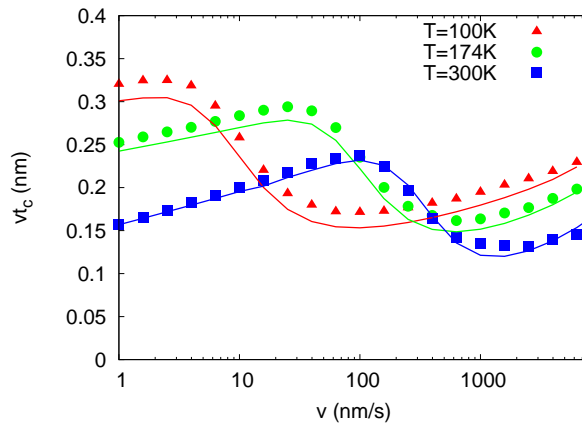


FIG. 2: Comparison between the average contact time numerically estimated by the Langevin equation (points) and the analytical approximation (lines). The parameter values are the same used in Figure 4 in the main text.

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