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# Supplementary Materials for

## What is the origin of macroscopic friction?

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**This PDF file includes:**

**Supplementary Text**

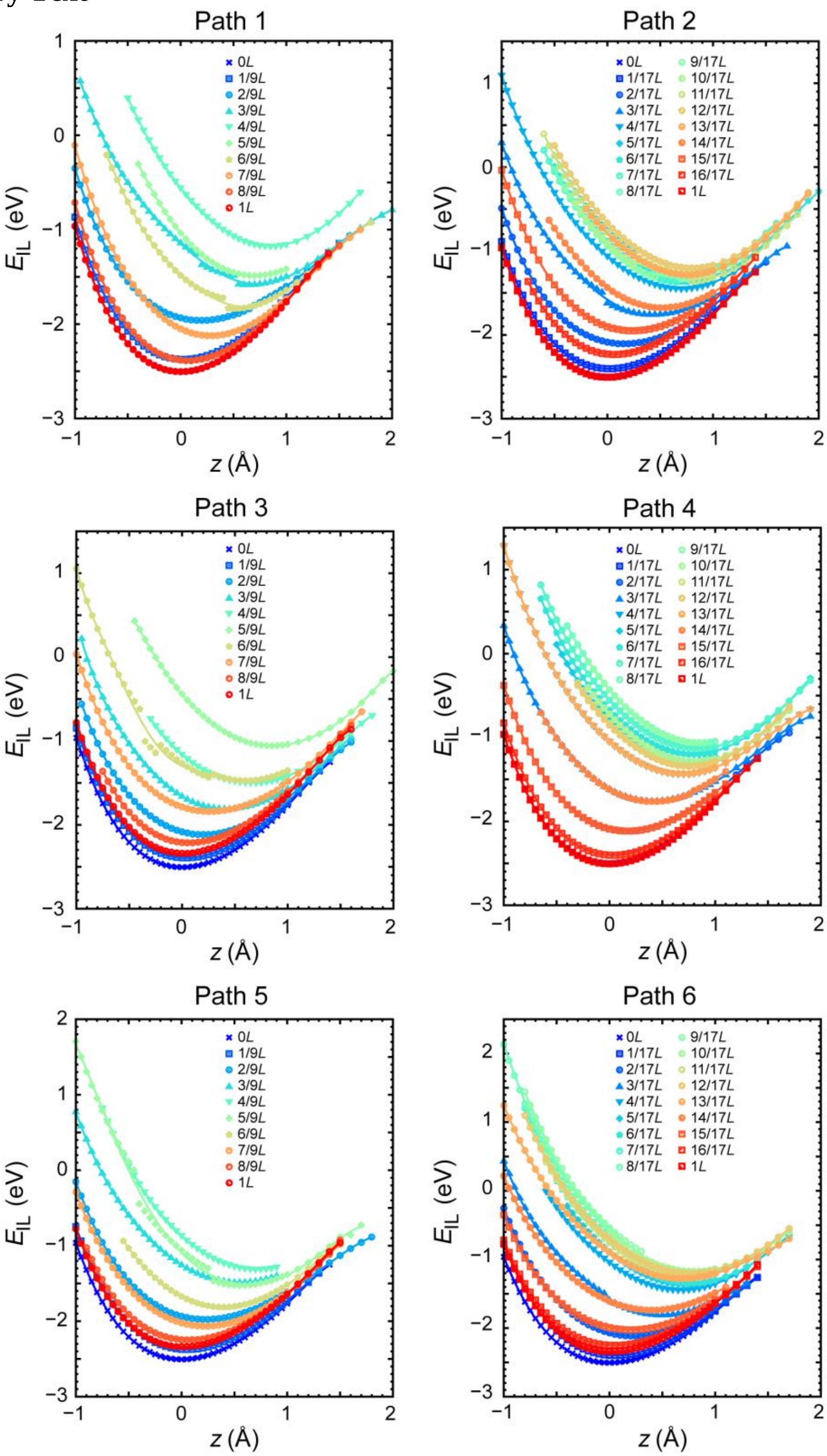


**Fig. S1. Interlayer potential energy $E_{IL}$ along six sliding paths.** The interlayer distance $z = 0$ Å is defined for the distance in the equilibrium interlayer distance of mica. The $E_{IL}$ profiles were calculated at intervals of 0.53 Å along the paths of length $L$. The solid lines indicate the best-fit polynomial equations.

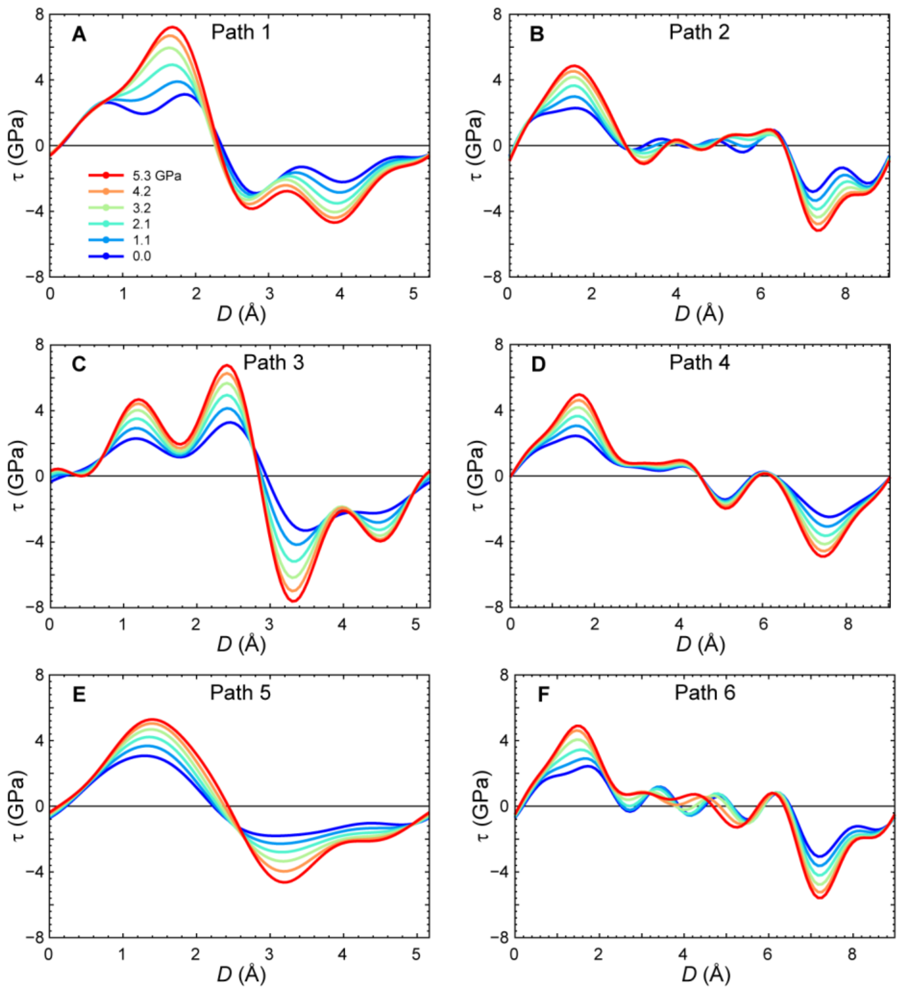


**Fig. S2. Shear stresses (τ) along six sliding paths obtained from the derivative with respect to distance of the potential energy curves shown in Fig. 1.** The positive shear stresses were used for calculating the average shear stresses.

List of Best-fit Parameters of the Shear Stress versus Normal Stress

**Table S1. The list of best-fit parameters of the average shear stress.**

| Sliding path | μ | $\tau_0$ (GPa) |
|---|---|---|
| 1 | 0.141 | 0.893 |
| 3 | 0.139 | 0.970 |
| 5 | 0.118 | 0.799 |
| | | |
| 2 | 0.085 | 0.524 |
| 4 | 0.081 | 0.562 |
| 6 | 0.085 | 0.531 |

Effects of Viscosity and Temperature

The viscous force $F_v$ is proportional to the sliding velocity $F_v = -bv$; the coefficient $b$ was estimated to be ~0.1 Ns/m for mica at the contact area of $1.0 \times 10^{-8}$ $m^2$ (*26*). The shear stress arising from this viscous force was ~30 Pa at a sliding velocity of 3.0 μm $s^{-1}$, which is much smaller than the theoretical shear stress on the scale of gigapascals. Thermal noise effects (*27*) on the friction can be discussed by comparing the roughness of the PES and $k_B T$, where $k_B$ is the Boltzmann constant and $T$ is the absolute temperature: at 298.15 K, $k_B T = 0.03$ eV. Assuming that vibrations of the surface oxygen and interlayer potassium ions arise because of the thermal random force, the possible maximum energy was estimated to be 0.36 eV (=0.03×14 ions), and the average value should be much smaller than this value. The maximum energy of thermal fluctuation is much smaller than the maximum height of the PES (2–2.5 eV). Based on this analysis, we conclude that a major contribution to the friction of mica at room temperature is the roughness of the PES.

Results of the Nanoindentation Test

An estimate of the real area of contact $A_{real}$ is required to predict the macroscopic friction from the theoretical molecular friction. The real area of contact $A_{real}$ should be smaller than the apparent contact area $A$ and can be estimated from the nanoindentation hardness $H$ and the applied load $L$ as $A_{real} = L/H$ *(28)*. The nanoindentation test was conducted for a cleaved muscovite surface. The load-displacement curve showed little hysteresis in this small load regime (**Fig. S3**), indicating that the deformation was elastic *(24)*. In our samples, the hardness was measured to be $H = 6.27 \pm 0.26$ GPa.

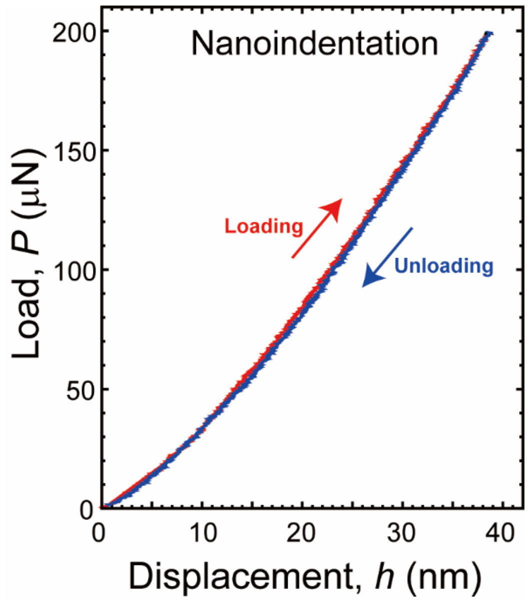

**Fig. S3. A representative nanoindentation profile of a mica cleavage plane.**