

Supporting information for article

“Superlubric sliding in a multi-contact fullerene-molybdenum disulfide heterostructure”

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1. Temperature effect on friction

In the dense, crystallized state, C_{60} fullerenes show a dynamic transition from orientational ordering (with only eventual orientational jumps) to free diffusional rotation above a temperature of around 260 K [1]. This transition could have important implications for the lubrication property of the nanoparticles investigated in our work. We therefore investigated the temperature dependence of the friction behavior to check for any effect of thermal activation. The friction force as a function of normal force for three different temperatures, 5 K, 300 K and 500 K, is shown in Fig. S1. No systematic difference between the temperatures is observed (besides at the very highest force $F_N=600\text{eV}/\text{\AA}$, which could indicate a thermally activated process under this strongly confining pressure). All temperatures seem to indicate a linear relation above some normal force, which allows determination of the friction coefficient. The lack of a temperature dependence indicates that at this low coverage, there is no sudden dynamical transition in the rotational motion, unlike for C_{60} in the dense, solid state. Furthermore, the compression of the fullerenes between the MoS_2 slabs slows down their rotation upon increasing of normal force.

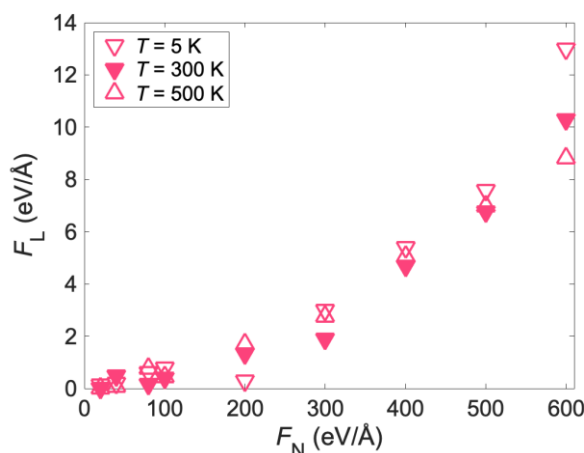


Figure S1 Effect of temperature on friction force

The simulations are under the condition of low number coverage. $N=40$ mol, $V=0.01$ ps/\AA.

2. Mean-square displacement

To elucidate the translational motion of the fullerenes in between the two sliding MoS_2 planes, we first look at their mean-square displacement (MSD). To do so, we subtract any drift motion of the fullerene layer from the individual particles' displacement, and plot the resulting MSD for two different temperatures in Fig. S2. At low temperature, the MSD shows load-dependent regimes (Fig. S2a): Under low applied normal load, the fullerenes slide collectively, leading to very small MSD, which stabilizes into steady state quickly. At medium load, the particles appear to wiggle, indicating some stick-slip behavior. At high load, the applied shear introduces significant diffusion of the fullerenes, as reflected in the increasing and saturating MSD. The distinction of these three regimes is diminished at room temperature, where thermal energy always leads to some diffusion of the particles. (see Fig. S2b).

3. Rotational map at room temperature

In the manuscript, we show the rotational motion of individual fullerenes at low temperature, $T=5$ K. We observe transitions

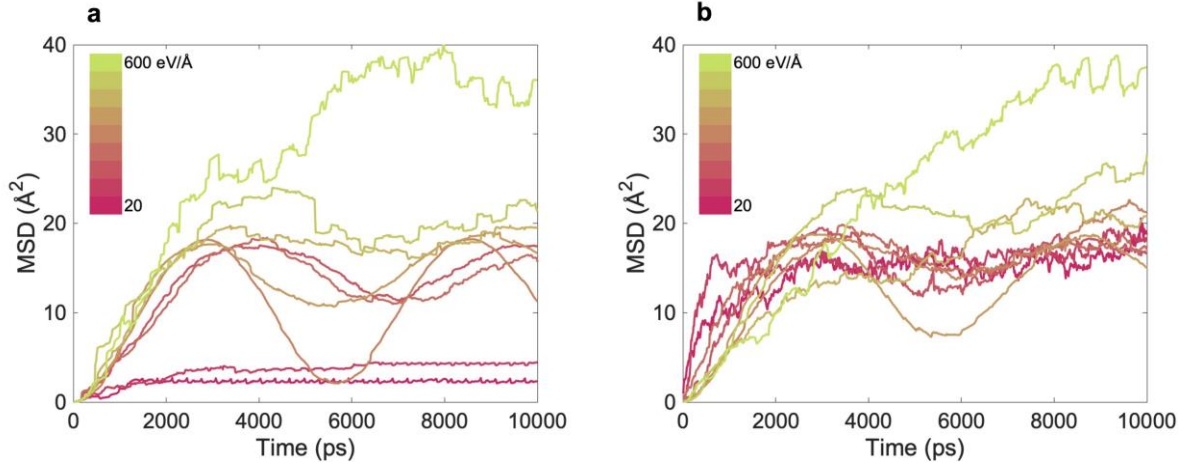


Figure S2 Effect of temperature on transverse locomotion

The mean-square displacement of nanoparticles at the temperature (a) $T=5\text{K}$ and (b) $T=300\text{K}$. $N=40$ mol, $V=0.01\text{ps}/\text{\AA}$.

from wiggling to rotation to chaotic rotation with increasing normal force. Here, we look at the rotational map at room temperature, $T=300\text{K}$, see Fig. S3. While a similar transition towards rolling is observed, clearly, rotational diffusion governs the low normal-force regime. At these low loads, the fullerene is not sufficiently confined by the MoS_2 slabs, and thermal energy is large enough to induce rotational diffusion. Rolling occurs in a medium range of the applied normal force, as shown in Fig. S3(c-f). At high load, the rolling is exceedingly suppressed due to flattening of the fullerene (Fig. S3(g, h)).

4. Rotational autocorrelation function

The rotational autocorrelation function related to rotation of the fullerenes has been calculated and plotted for $T=5\text{K}$ in the main text. To check for thermal effects at room temperature, we plot the autocorrelation function for $T=300\text{K}$ in Fig. S4. Similar to Fig. 4 in the main text, we observe a transition from sliding to rolling back to sliding with increasing load, which we associate with the formation of an adhesion contact (transition towards rolling) and flattening-induced suppression of rolling (transition back to sliding) with increasing load. Yet, in contrast to $T=5\text{K}$, significant decorrelation is observed even at low load, indicating the thermally-induced rotational diffusion. At higher loads, the behavior is similar to the one observed at low temperature showing that the applied confinement suppresses any strong rotational diffusion. The dependence on the fullerene coverage is depicted for the higher loads in Fig. S5. Clearly, the loss of sinusoidal behavior indicating transition from rolling to sliding is no longer observed; the higher fullerene density supports the increasing load and prevents fullerene deformation, thus keeping rotation intact. However, at even higher coverage, the rotation is obviously lost (Fig. S5b), indicating that now fullerene-fullerene

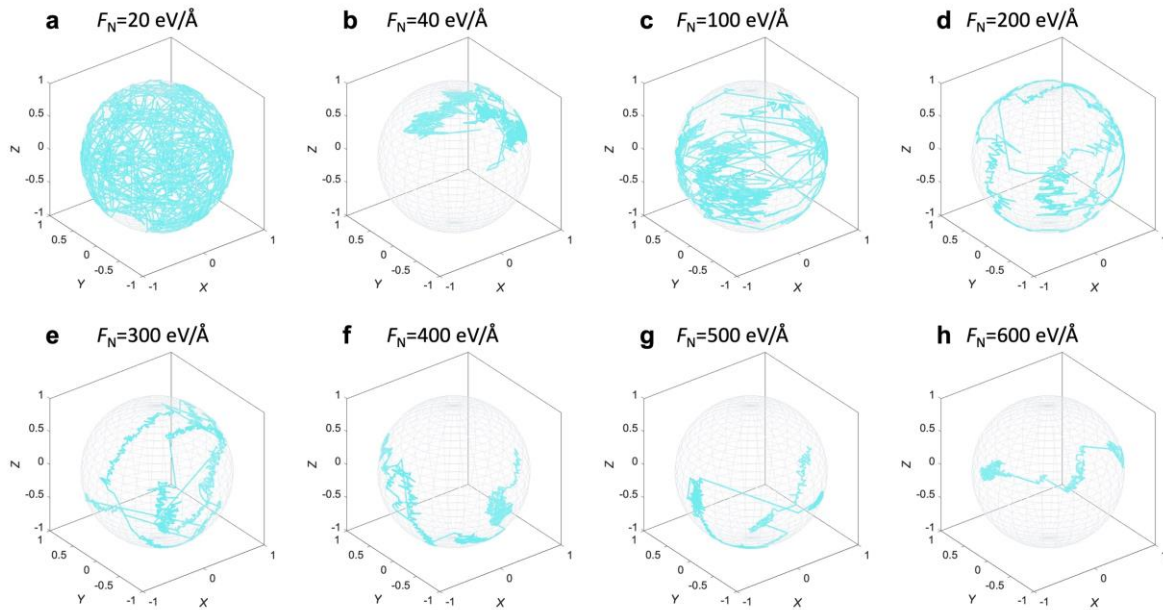


Figure S3 Representative rolling trajectory on the unit sphere surface

(a-h) The trajectory map of a point on the single fullerene sphere changes with the normal force. The shearing process lasts 10 ns with applied different normal loads to the sandwich model. $T=300\text{K}$. $N=40$ mol, $V=0.01\text{ps}/\text{\AA}$.

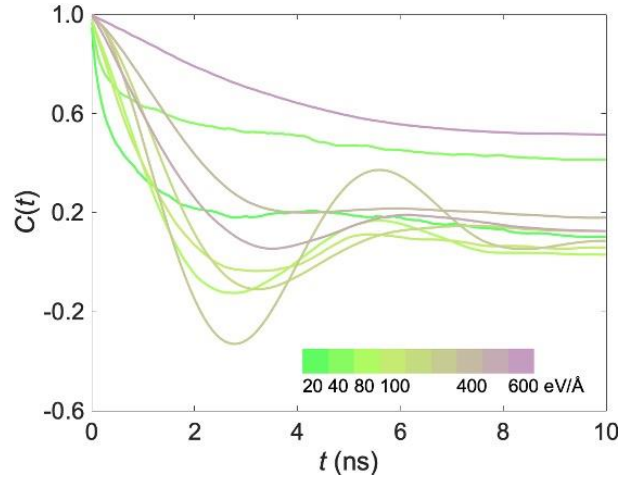


Figure S4 Rotational autocorrelation function as a function of time delay at $T=300\text{K}$ for $N=40$ mol

The evolution of the rotational autocorrelation function is plotted for different normal forces (see color bar).

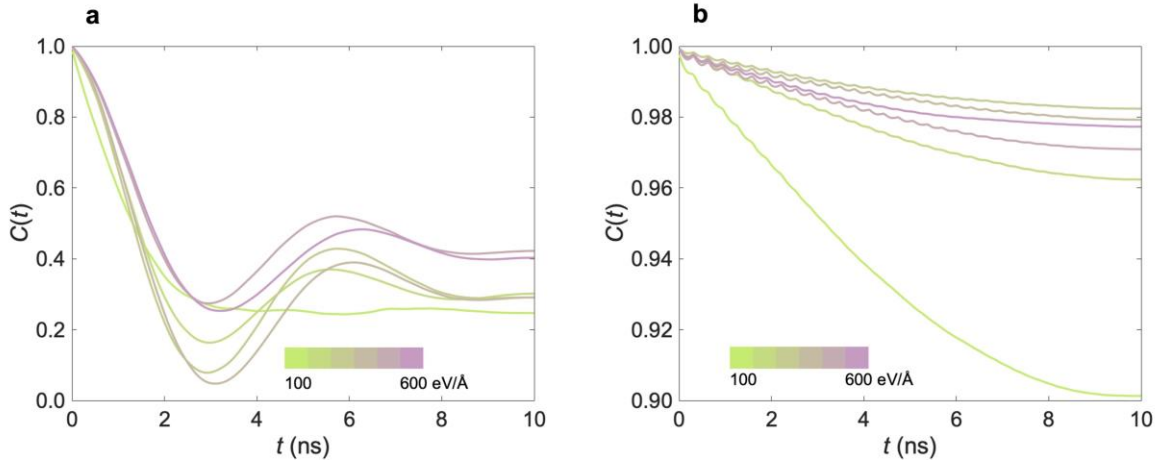


Figure S5 Rotational autocorrelation function as a function of time delay at $T=300\text{ K}$ for $N=60$ and $N=80$ mol

Evolution of the rotational autocorrelation function is plotted for different normal forces (see color bar), for coverages $N=60$ mol (a) and $N=80$ mol (b).

interactions become dominant, which block their mutual rotation. This blocking of rotation reminds of the dynamic transition from orientational freezing to orientational diffusion observed at $T \sim 260\text{ K}$ in dense C_{60} solids.

5. Ensemble-averaged rotation angle

The above observations suggest the occurrence of rolling, and its suppression at high normal load and high fullerene coverage. To most directly test this interpretation, we quantify the overall rolling of the fullerenes in the sheared system by calculating the average rotation angle of all particles as

$$\theta(t) = \frac{1}{N} \sum_{i=1}^N \theta_i(t) \quad (\text{S1})$$

where $\theta(t)$ represents the average rotation angle of the fullerenes in space during shearing time (t), θ_i is the absolute rotation angle of fullerene i , and N is the total number of unbroken nanoparticles in the system.

The ensemble-averaged rotation angle evolves linearly with time, giving evidence of rolling, see Fig. S6. The slope clearly decreases with applied load, suggesting that the resulting squeezing of the fullerenes attenuates their rolling. The slope is also reduced for high surface coverage (Fig. S6c), indicating that particle-particle interactions suppress rolling at high fullerene density. The fluctuations around the average shown in Fig. S6(d-f) reveals similar trends, indicating that at these loads above $100\text{ eV/\AA}$, rotational diffusion is largely coupled to their shear-induced rolling. Specifically, under medium normal force, the large fluctuation value reflects the large contribution of rolling, which yet seems not correlated among the particles. When the normal force and the coverage number increase, the mean-square rotation of the fullerenes gets suppressed similar to their overall rotation amplitude.

6. Structural profile and velocity distribution

To get insight into the atomic contact formation and corresponding ceasing of atomic sliding, we plot the height distribution of the individual carbon atoms of the fullerenes together with their velocity profile in Fig. S7(a-c), which is an extended version of Fig. 5 in the main text. Two spikes occurring under low fullerene coverage indicate the formation of an adhesion contact

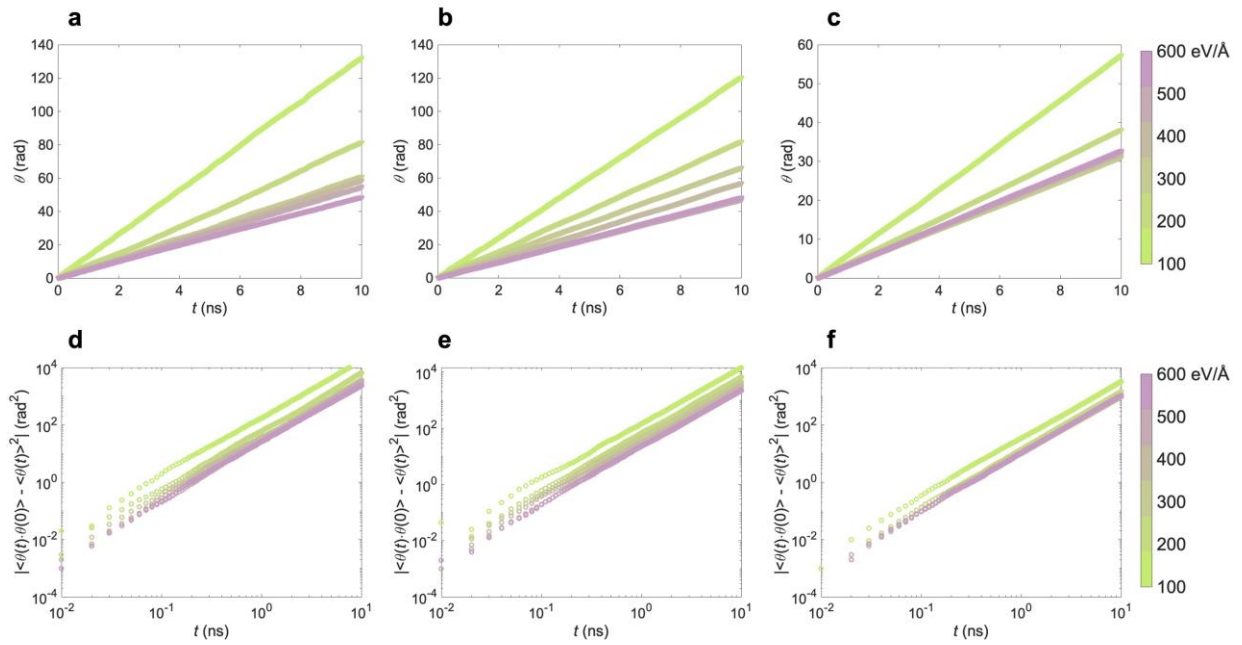


Figure S6 Rotational angle and fluctuation from average

The averaged rotational angle θ changes with time and normal force under the nanoparticles coverage number (a) $N=40$ mol; (b) $N=60$ mol; and (c) $N=80$ mol. (d-f) The fluctuation from the average is calculated by $\langle \theta(t) \cdot \theta(0) \rangle - \langle \theta(t) \rangle^2$ corresponding to (a-c).

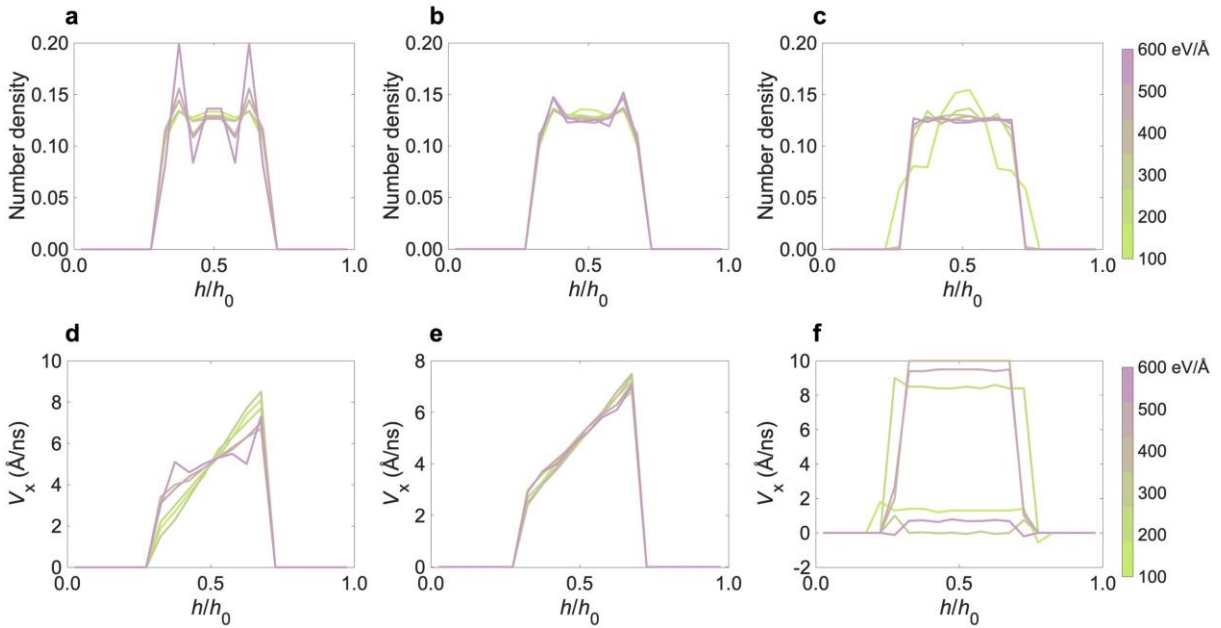


Figure S7 Structural profile and velocity distribution

Time averaged structural profile of nanoparticles in the sandwiched structure. (a-c) Number density of carbon atoms changes with normalized height (h/h_0), where h is the z coordination of each chunk, and h_0 is total height of model. Under different coverage numbers (a) $N=40$ mol, (b) $N=60$ mol and (c) $N=80$ mol. (d-f) Velocity distribution in shearing direction (V_x) corresponding to the structural profiles (a-c).

between the fullerene and the MoS₂ in Fig. S7a. These “contact spikes” sharpen with increasing applied load, indicating the fullerene flattening and intensification of the contact formation. Concomitantly, the velocity profile flattens slightly, indicating a partial suppression of rolling. The contact spikes reduce for higher fullerene coverage that better supports the applied load and hence reduces the flattening. The spikes disappear completely at the highest fullerene coverage, indicating that fullerene-fullerene interactions now dominate and no adhesion contact is formed with the MoS₂. The corresponding velocity profiles also flatten completely, clearly evidencing the suppression of rolling and transition to sliding as shown in Fig. S7f.

7. Friction force versus time

To check the applicability of our shearing protocol, we looked at the raw force data. The typical time-dependent friction force is shown in Fig. S8. The data reveals that the friction force mostly fluctuates around a constant, normal-force dependent value indicating continuous sliding.

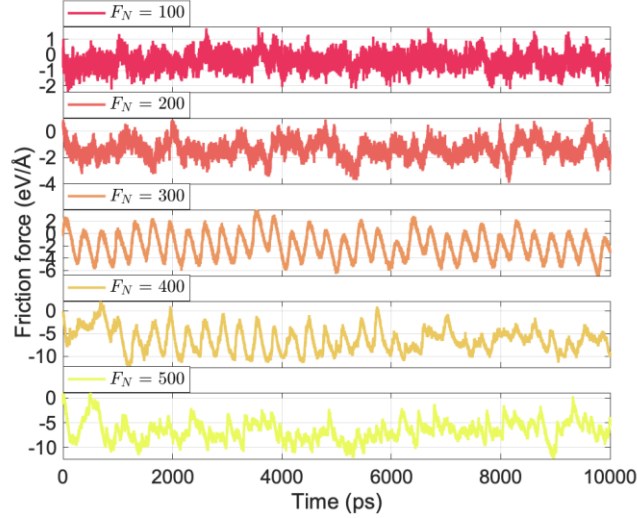


Figure S8 Time-dependent friction force under different normal forces for the case $N=40$, $V=1\text{m/s}$, and $T=300\text{ K}$. To calculate the mean friction force, data from the initial 1000 ps were excluded to avoid transient effects.

8. Influence of simulation box size

To investigate the effect of box size and higher normal force on the friction behavior, we built a larger simulation model with lateral dimensions $W \times L \sim 126.64\text{Å} \times 137.09\text{Å}$ and the same areal density of C_{60} , as shown in Fig. S9a. The areal density of C_{60} in the main text is: $\rho = 40/(7.3\text{nm} \times 7.1\text{nm}) = 0.77\text{nm}^{-2}$ for the $N = 40$ fullerene system, and $\rho = 80/(7.3\text{nm} \times 7.1\text{nm}) = 1.54\text{nm}^{-2}$ for the $N = 80$ fullerene system. The comparison of the larger system with that of the smaller one presented in the manuscript is shown in Fig. S9b. Except for small deviations, the two systems show the same friction behavior, i.e. very similar friction force versus normal-force behavior. To keep simulation costs manageable, in the manuscript, we therefore limit our studies to the smaller simulation system. In addition, we note that if the thickness of the MoS_2 was changed, the friction between the C_{60} and MoS_2 is expected to change, as the friction of 2D materials is known to depend on the number of layers, decreasing to the bulk value for thick slabs [2-4]. Nevertheless, the main mechanisms that we observe and investigate in this work are mediated by the interactions between the C_{60} molecules, not the MoS_2 . Thus, while thicker layers would likely lead to some quantitative change in the friction between the C_{60} and MoS_2 , we would not expect it to affect our results qualitatively.

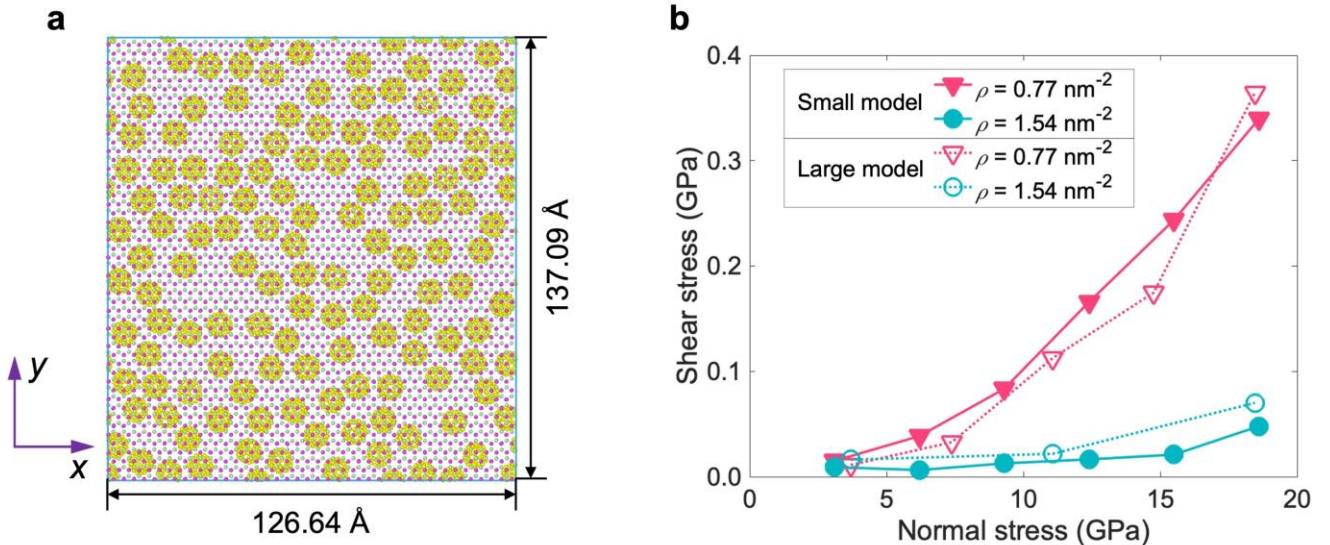


Figure S9 Friction behavior in a large simulation model

(a) Representation of the larger simulation model with lateral size of $126.64\text{Å} \times 137.09\text{Å}$. (b) Friction force per area versus normal force per area for sliding velocity of $v=1\text{m/s}$ at $T=300\text{ K}$.

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