

Electronic Supplementary Material

Atomic alignment and friction behavior of dense fullerene packings revealed by molecular dynamics simulations

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1 Potentials for fullerene intermolecular interactions

In this section, we investigate a range of carbon atomic potential models to model the C60 fullerene molecules. Originally, the interaction of covalently bonded carbon atoms within carbon molecules was modelled by the reactive empirical bond order (REBO) potential by Brenner [1], which is well established. Yet, for fullerenes, the inter-molecular interactions are dependent on the rotation and transverse motion of the nanoparticles, and previous research has shown [2] that these interactions between fullerene molecules have significant effect on the local stacking mode of neighbors and the resulting friction properties. To account for these effects, the adaptive intermolecular reactive empirical bond order (AIREBO) potential has been developed by Stuart et al. [3], adding the truncated Lennard-Jones (6-12 LJ) potential for nonbonded interactions, and torsion interactions. As the LJ potential depends only on the interatomic distances between atom pairs, however, AIREBO fails to describe the energy corrugation during the transverse sliding and rotational displacements from the equilibrium configuration. To handle the problem of transverse sliding, Kolmogorov and Crespi [4] developed the register-dependent KC potential. Still, the KC potential cannot correctly predict the force in the out-of-layer direction when one layer is rotated relative to the other layer. Thus, while it is much more accurate than LJ potential to describe the corrugation of graphene interlayers [5], it cannot distinguish forced from the AA/AB stacking mode between graphene layers [6, 7]. To further improve the accuracy of the interlayer interactions, Wen [7] introduced a dihedral-angle-corrected registry-dependent interlayer potential (DRIP) to describe the interlayer transverse sliding and rotation.

Since both KC and DRIP are not chemical reactive models to

describe bond interactions, and are both related to an atomic layer or molecule [8], the simulations will be interrupted when the atomic coordination gets lost upon bond breaking under high pressure. AIREBO overcomes this problem by allowing dynamic bond breaking and formation in carbon-based materials, which makes it a kind of chemical reactive potential. Furthermore, as the LJ potential rises too steeply at short distance, a modified Morse type of AIREBO potential (AIREBO-M) was introduced, which is suitable for extreme pressure [9]. On the other hand, based on the REBO potential [1], Los and Fasolino [10] proposed a long-range carbon bond-order potential (LCBOP) that includes both angular and coordinate-dependent interactions. In this potential, both short-range and long-range (weak) interactions are employed, which turn out to be suitable for modeling structural properties of carbon materials.

To investigate the robustness of the results presented in the main text, we compare here the results of three different potentials, AIREBO-M, LCBOP, and DRIP. In particular, we study their influence on the stacking orientation of the nearest neighbor particles, as well as the friction properties along a plane of fullerenes to test the robustness of the results presented for the AIREBO potential in the main text.

Influence of atomic potential on fullerene stacking

We compare projection maps under different normal loads in Figs. S1-S3, computed with AIREBO-M, LCBOP, and DRIP potential, respectively. From Fig. S1, we conclude that the stacking probability obtained with the AIREBO-M potential is comparable to that of the AIREBO presented in the main article. When it comes to LCBOP, the stacking probability deviates: it is equally distributed along the edges of the polygons, suggesting no distinction between AA and TA stacking as shown in Fig. S2. Concerning the DRIP potential (Fig. S3), the stacking probability reproduces the consistent results of AIREBO and the derived AIREBO-M potential under lower normal force.

But due to higher computation cost associated with the DRIP potential, it works not very well under higher pressure, leading to breaking of fullerene bonds. We thus cannot access the high normal load regime and are limited to normal forces of about 96 nN. The pair potentials between two fullerene molecules predicted by the different potential models are plotted in Fig. S4. Here, we distinguish the two stacking configurations, AA and TA, which

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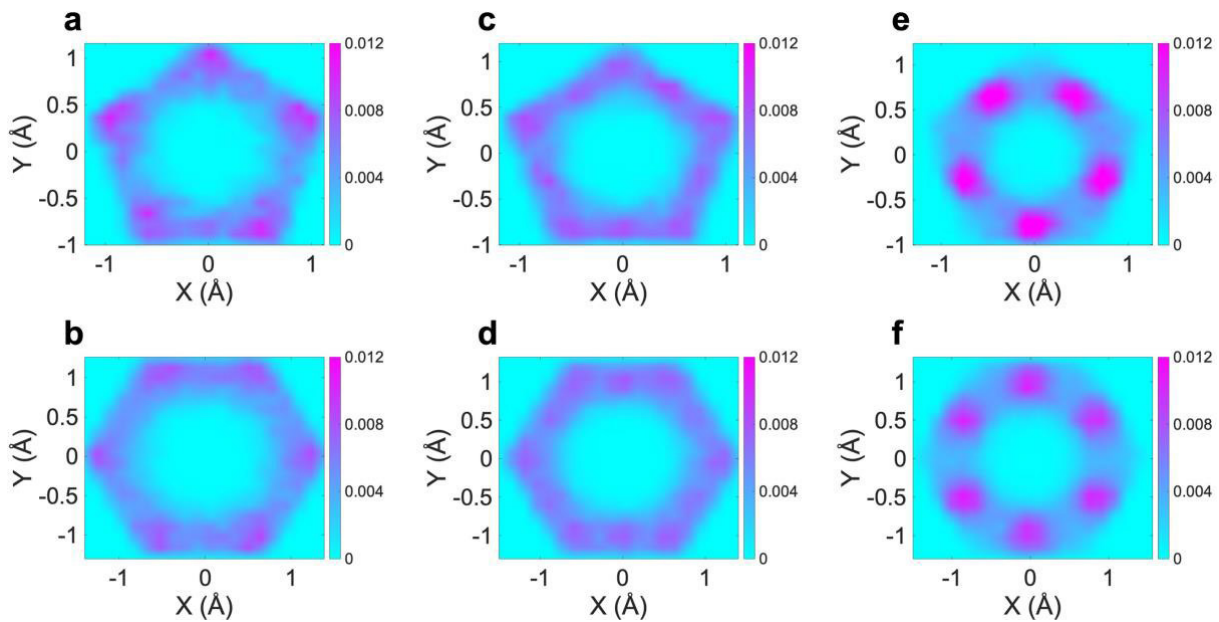


Fig. S1 Atomic stacking for AIREBO-M potential.

Probability distribution of vertex projection into C5 (top row) and C6 (bottom row). Normal force varies from (a–b) 3.2 nN, (c–d) 32 nN, to (e–f) 320 nN. Under half roughness model, $V = 1$ m/s, $T = 300$ K.

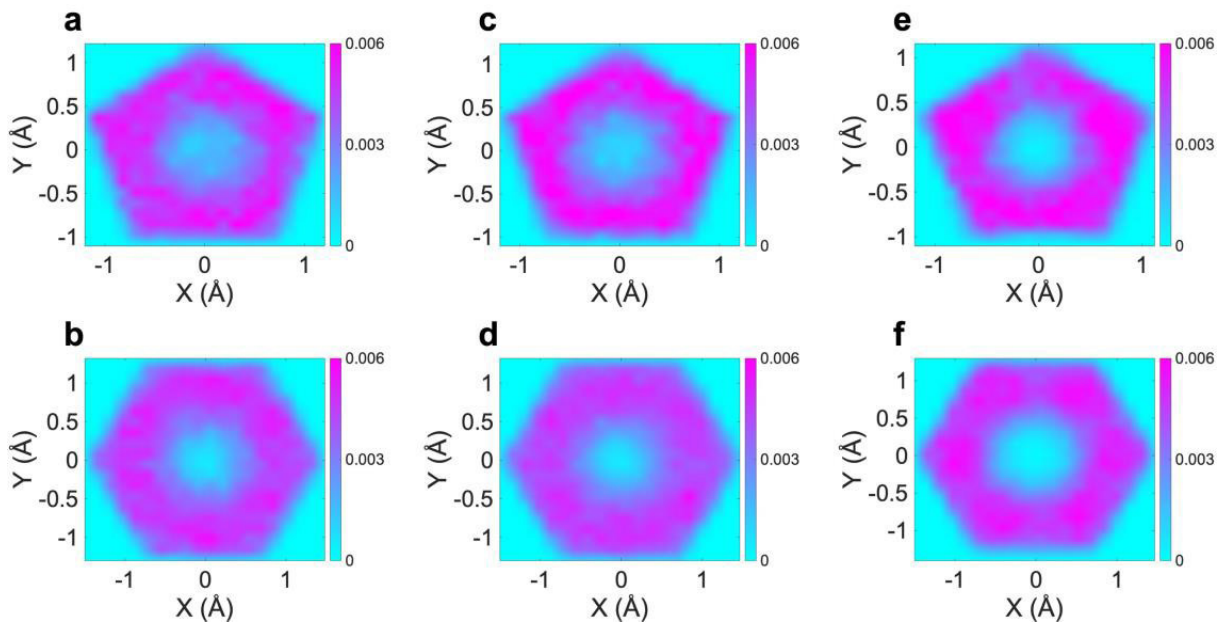


Fig. S2 Atomic stacking for LCBOP potential.

Probability distribution of vertex projection into C5 (top row) and C6 (bottom row). Normal force varies from (a–b) 3.2 nN, (c–d) 32 nN, to (e–f) 320 nN. Under half roughness model, $V = 1$ m/s, $T = 300$ K.

have slightly different potentials. Notably, both AIREBO and DRIP potentials can reproduce the transition from AA to TA stacking: with decreasing fullerene distance, the TA stacking develops a deeper minimum, indicating transition to this stacking configuration under increasing load. The LCBOP potential, in contrast, does not show this distinction between the stackings, and hence would not predict the transition with increasing applied load. Recently, potentials have also been trained by machine learning, a famous representative being the hNN-Grx potential [8]. The latter shows a slightly deeper potential for the AA configuration, opposite to the DRIP and AIREBO potentials, hence similarly failing to identify the stacking transition between fullerenes in equilibrium. Since our DRIP and AIREBO results are

in agreement with DFT calculations for graphene [7], we believe they provide a reasonable approach for our study. Furthermore, since there is consensus that lower interaction energy is more favorable for stacking [11], we conclude that the DRIP and AIREBO potentials and simulation results are reasonable to predict the stacking transition between fullerene molecules.

Influence of atomic potential on friction properties

As described in the main text for the AIREBO potential, we computed the friction force for a wider range of potentials and plot the results as a function of normal force in Fig. S5. Here, rough boundary conditions have been used, where boundary sliding is prevented. Again, nearly proportional functions are observed, the slope of which indicates the friction coefficient. We

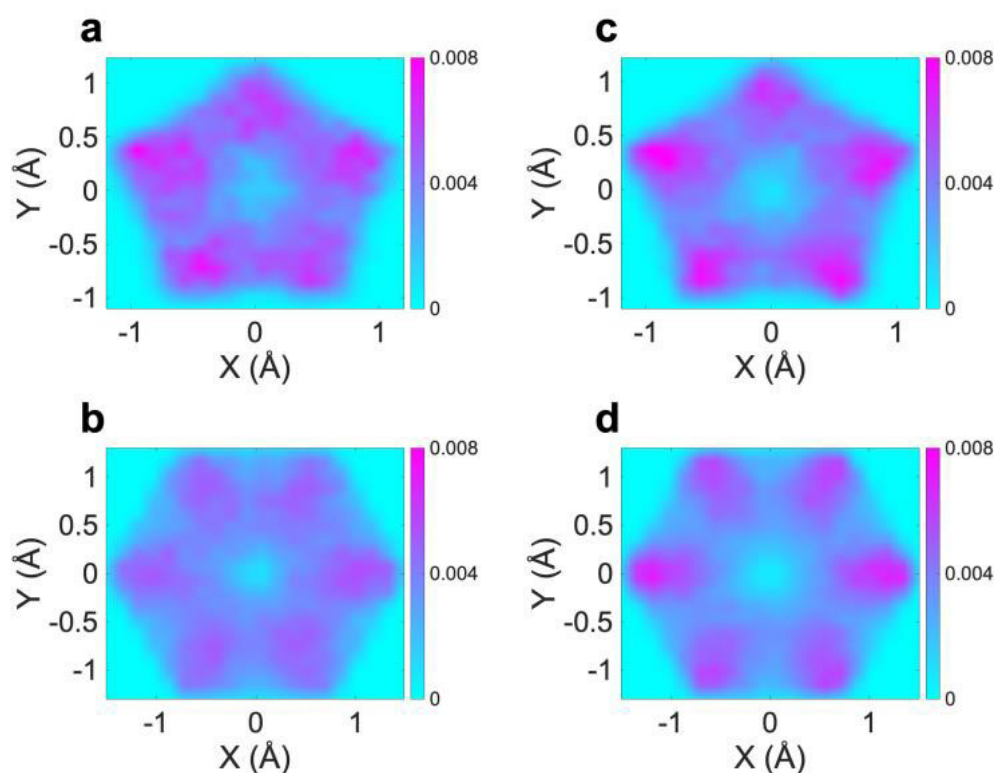


Fig. S3 Atomic stacking for DRIP potential.

Probability distribution of vertex projection into C5 (top row) and C6 (bottom row). Normal force varies from (a–b) 32 nN, to (c–d) 96 nN. Under half roughness model, $V = 1$ m/s, $T = 300$ K.

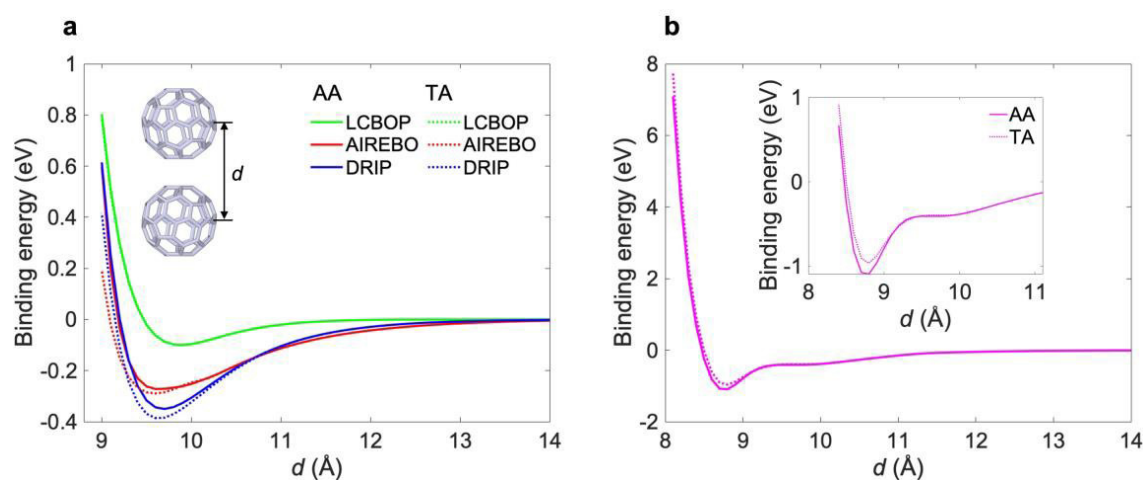


Fig. S4 Binding energy versus distance between two C60 molecules under AA/TA stacking.

(a) The LCBOP, AIREBO, DRIP and (b) hNN-Grx potentials. The solid line is used for AA stacking, and the dotted line represents TA stacking.

observe reasonably consistent values, ranging from 0.15 to 0.23. Furthermore, the lowest value in the amorphous C60 system is approaching the experimentally measured friction coefficient of about 0.15 [12, 13]. The variation in the applied intermolecular potentials has therefore only a minor effect on the friction properties. Considering the constant properties of intermolecular contacts and mechanical behavior, eventually, we choose the AIREBO potential for the simulations presented in the main text.

2 Deformation of fullerenes

To evaluate the contribution of fullerene deformation to the observed friction behavior, we monitored their deformation

during the shearing process. Firstly, based on the inertia of the spherical shell, we calculate the effective radius of the deformed C60 molecules. As can be seen in Fig. S6a, the radius of the fullerenes decreases linearly with increasing normal force almost independent of the boundary roughness. In our simulations, the maximum normal force is 320 nN, resulting in a deformation of the fullerenes of less than 4%. We also quantify the fullerenes' deformation by calculating the aspect ratio of their vertical versus lateral extension, as measured by the two radii in these directions. As can be seen in Fig. S6b, the aspect ratio decreases down to 0.93 at the highest applied normal force of 320 nN, indicating a small squeezing of the fullerenes in the vertical direction.

($\epsilon = c/a$) of C60 under shearing process. $V = 1$ m/s, $T = 300$ K.

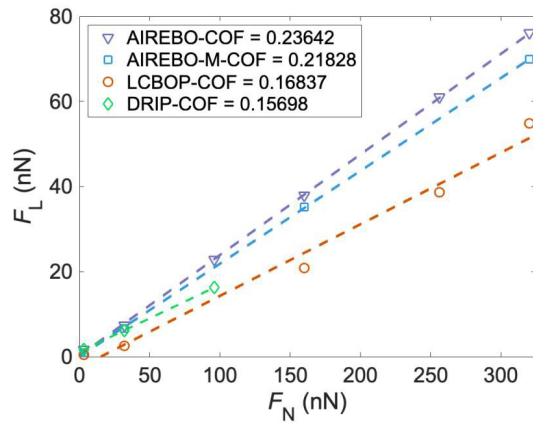


Fig. S5 Atomic potential influence on friction. Friction force versus normal force under rough boundary condition for four different atomic potentials: AIREBO, AIREBO-M, LCBOP, and DRIP. Dashed vertical fitting lines indicate some difference in the friction coefficient obtained for the different potentials.

3 Structure of the fullerene packing under shear

To check for any ordering of the fullerene packing, we analyzed the radial distribution function $g(r)$ of the particles, indicating the probability of occurrence of a particle as a function of distance from a reference particle relative to the ideal gas distribution. The $g(r)$ of the centers of the particles before and after shearing is shown in Fig. S7. All samples before shearing exhibit amorphous order, as shown in Fig. S7, top row. With increasing normal load, the position of the first peak of $g(r)$ decreases, reflecting a slight compression of the system, until it approaches 9.25 Å, at the highest load. After shearing, there is some signature of crystallization, which appears to become more pronounced as the boundary roughness increases. For the roughest boundary, we observe a transition from disordered to a (partially) ordered structure at high normal forces, after shearing for about 10 ns. This likely reflects the action of the bulk shear that occurs for the rough boundaries: the suppression of slip, and transition towards

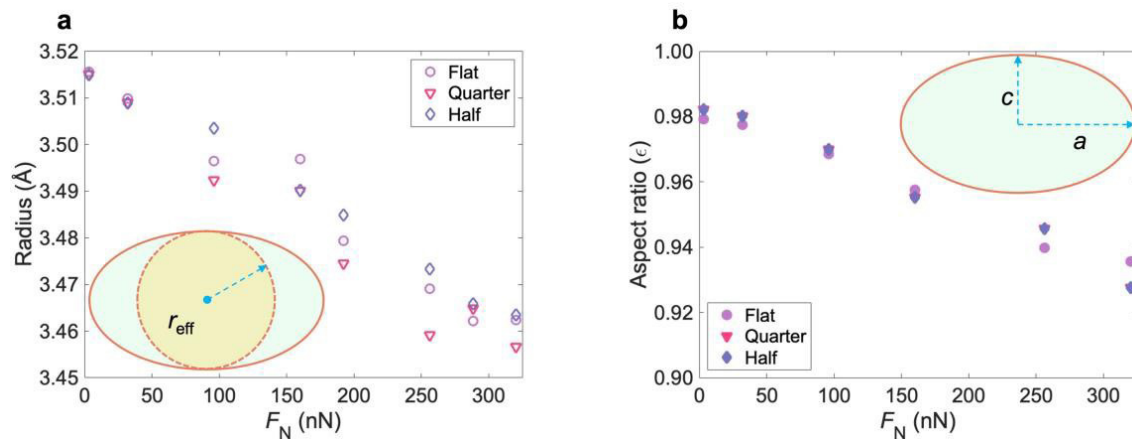


Fig. S6 Compression of the fullerenes. (a) The effective radius r_{eff} and (b) the aspect ratio

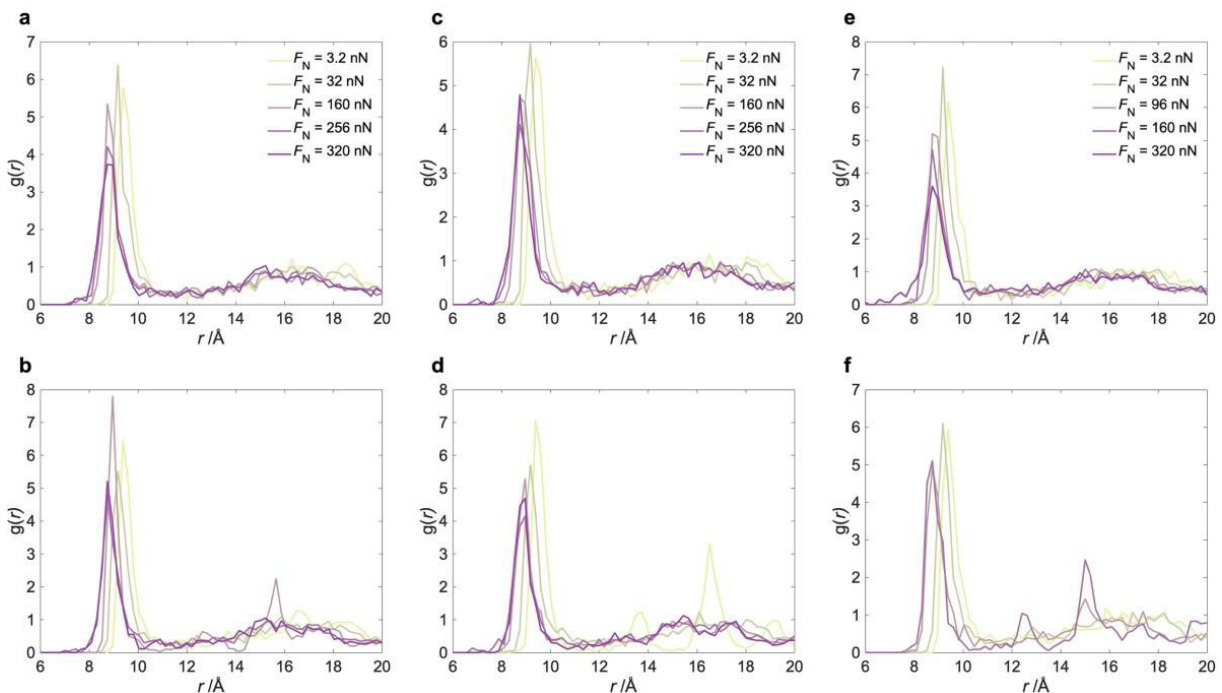


Fig. S7 Particle structure. Radial distribution function $g(r)$ for different normal force before (top row) and after shearing for 10 ns (bottom row) under the flat (a, b), quarter-displaced (c, d), and half-displaced boundary model (e, f), respectively.

bulk shear for rough boundaries, provides extra energy (shear-induced noise) for rearrangement in the C60 packing, allowing to reach a higher degree of ordering. This tendency is less observed for flat boundaries, that exhibit slip, but almost no bulk shear of the packing.

The tendency towards ordering is also reflected in the height distribution of the particles. We show particle density profiles in the compression direction in Fig. S8. In qualitative agreement with the pair correlation function in Fig. S7, we observe flat profiles, and only some appearance of peaks for flat surfaces, which however become more pronounced for higher roughness, reflecting increasing tendency towards ordering. Specifically, for low normal forces on flat and quarter-displaced boundaries; peaks start to appear, yet become more pronounced for the roughest (half-displaced) boundary under higher normal forces, indicating a tendency for layering, similar to the propagation of peaks in Fig. S7.

4 Contact model at the nanoscale

The Hertzian contact model for calculating the contact area between a spherical surface and a plane exhibits nonlinear behavior. Using this contact model, one can show that the friction force F_L as a function of normal force (F_N) can be described by the power-law relation [14–16]

$$F_L = \alpha(F_N - \beta)^{2/3} \quad (S1)$$

where the parameters α and β are constants.

Alternatively, for the adhesive contact in the JKR [17] and DMT model, the friction versus load was fitted by the intermediate regime of Maugis-Dugdale model [18] which is approximately quantified by the Carpick, Ogletree, and Salmeron (COS) equation [19]. The COS model yields

$$F_L = F_{L0} \left(\frac{\alpha_T + \sqrt{1 - F_N/F_a}}{1 + \alpha_T} \right)^{4/3} \quad (S2)$$

where F_{L0} represents the friction force at zero load, and F_a is the pull-off force or adhesion force.

The transition parameter α_T is related to the Tabor's parameter [20].

While increasing the roughness of boundaries from single asperity to multi-asperity contacts, a modified Amonton law was shown to fit the friction versus load relation well. In addition to the applied normal force F_N , there is an internal force due to adhesion between the tip and the contacting sample. In this case, the modified Amonton law can be written [21] as Eq. (S14):

$$F_L = \mu F_N + F_0 \quad (S3)$$

where F_0 is the intercept of lateral force resulting from the adhesion interaction.

The friction force determined from our fullerene simulations is shown together with the fits according to the COS model in Fig. S9. At small roughness, for the flat surface or quarter-displaced surface, the data are well-fitted by the COS model. For rough boundaries, the friction force appears to be better described by a simple linear relation following Amonton's law, except perhaps the small y-axis offset indicating the presence of adhesion [20]. The fitting parameters of the COS model are shown in Table S1; the values $0 \leq \alpha_T \leq 1$ indicate an adhesion model intermediate in the range of DMT to JKR model [22].

5 Variation of simulation parameters

Effect of temperature

In addition to the room-temperature simulations presented in the manuscript, we also performed simulations at low temperature of $T=50$ K, far below the phase transition temperature of 260 K [23]. The resulting friction force as a function of normal load is shown in Fig. S10. Mostly, an offset is observed between the two datasets, indicating a stronger influence of adhesion at the lower temperature, as expected. While the low-temperature data shows larger scatter, the slope (friction coefficient) is the same within error bars. The stacking probability at the lower temperature likewise shows the same trend as at room temperature, as shown in Fig. S11. Even though any thermal activation is much reduced, the results show that a similar stacking transition from AA to TA occurs with increasing normal force. There is only a minor effect of temperature on the stacking probability distribution.

Effect of velocity

To investigate the effect of shear rate and sliding speed on the particles' stacking behavior, we changed the shear velocity of the rough boundary to a higher speed. The velocity is set to $V = 10$ m/s and the results are shown in Fig. S12. Comparison with the distributions in the manuscript shows that the higher velocity barely influences the stacking probability distribution of the nearest neighbors.

Effect of boundary roughness

As described in the main text, we changed the roughness by shifting fullerene particles out of the periodically arranged plane by various amounts, 0, $\frac{1}{4}$ and $\frac{1}{2}$ of their diameter. We showed probability distributions of atomic stacking for rough boundaries, for which bulk shear occurred. Here, in comparison, we show the stacking probability distribution for a flat boundary, for which there almost no bulk shear, and mostly plug flow. As shown in

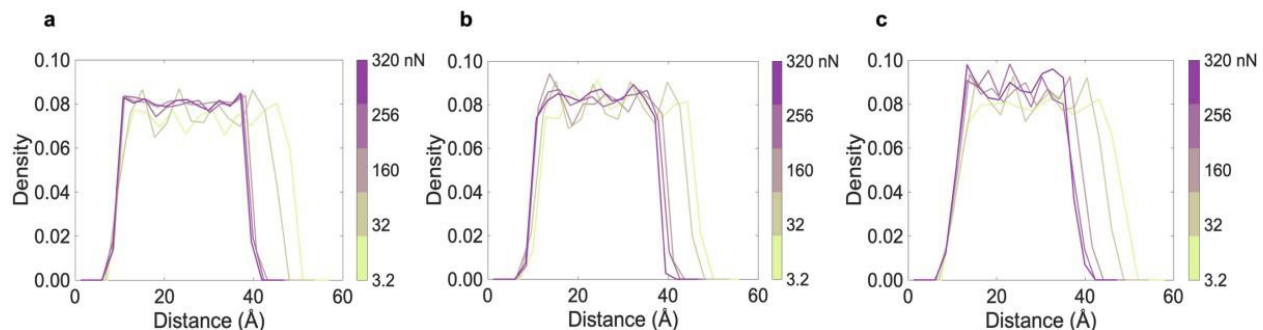


Fig. S8 Density profiles of the sheared fullerenes. Density profiles of C60 after shearing for 10 ns under flat (a), quarter-displaced (b), and half-displaced boundary model (c), respectively. Color bar indicates normal force, increasing from 3.2 to 320 nN.

Fig. S13, the boundary conditions and corresponding lack of bulk shear do not seem to have any significant effect on the atomic stacking of the fullerenes in the bulk.

Effect of simulation box size

To investigate any size effects of the confined layers, we also varied the thickness and lateral size of the sandwiched structure. We increased the length and width of the simulation box to $L \times W = 72 \text{ \AA} \times 72 \text{ \AA}$ as shown in Fig. S14, and correspondingly reduced the height to have the same amount of fullerene molecules (100) in the system. The thickness of the fullerene packing then decreases to three or four layers when the packing is compressed under increasing normal force.

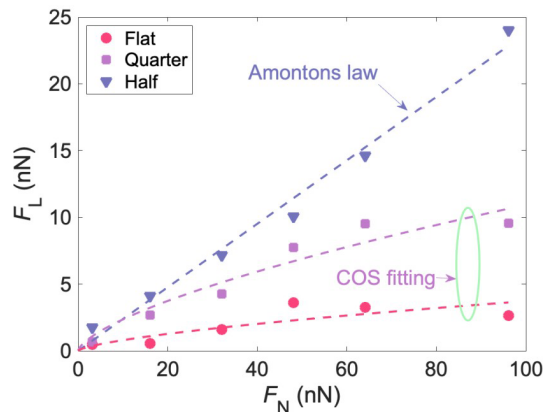


Fig. S9 Friction force with COS fit. Friction force versus normal load for flat, quarter, and half boundaries as in the manuscript, but dashed lines for flat and quarter boundaries correspond to COS fits. The constants F_{L0} , α_T and F_a for COS model are listed in Table S1.

Table S1 Fitted constants for COS equation

Model	Parameters		
	F_{L0}	α_T	F_a
Flat model	0.0019	0.1474	-0.0011
Quarter model	0.0028	0.2287	-0.0003

Again, we explored three boundaries with different roughness, flat, quarter, and half. The corresponding friction behavior is shown in Fig. S15. Consistent with the results in the main article, we again observe closely superlubric behavior for the flat boundary, and an increase in the friction coefficient with increasing roughness. The stacking probability is shown in Fig. S16. Clearly, the stacking orientation again changes from AA to TA under increasing applied load, consistent with the results presented in the main article.

6 Monolayer confined structure of fullerenes

To contextualize the lubrication behavior for flat boundaries, where superlubric boundary slip is observed, we also looked at a single layer of fullerenes between two flat fullerene slabs. We again calculated the mean translational and averaged effective rotational displacement of the fullerenes as explained in the main text to quantify the contribution of rolling and sliding. To specifically address the role of particle-particle interactions in the rolling contribution, we varied the density of fullerenes in the single layer from a lightly to a densely occupied layer. The resulting

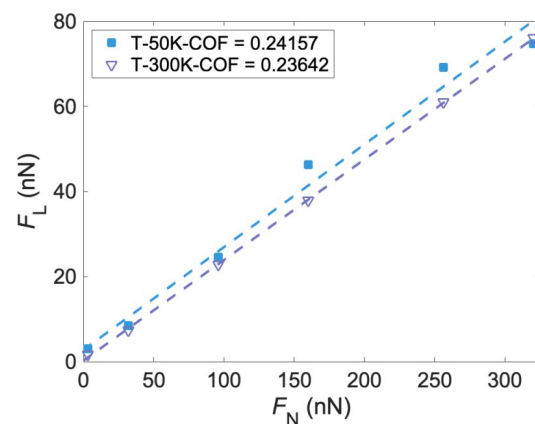


Fig. S10 Effect of temperature on friction force. Friction force versus normal load for the rough ("half") boundary condition computed with AIREBO potential, $V = 1 \text{ m/s}$.

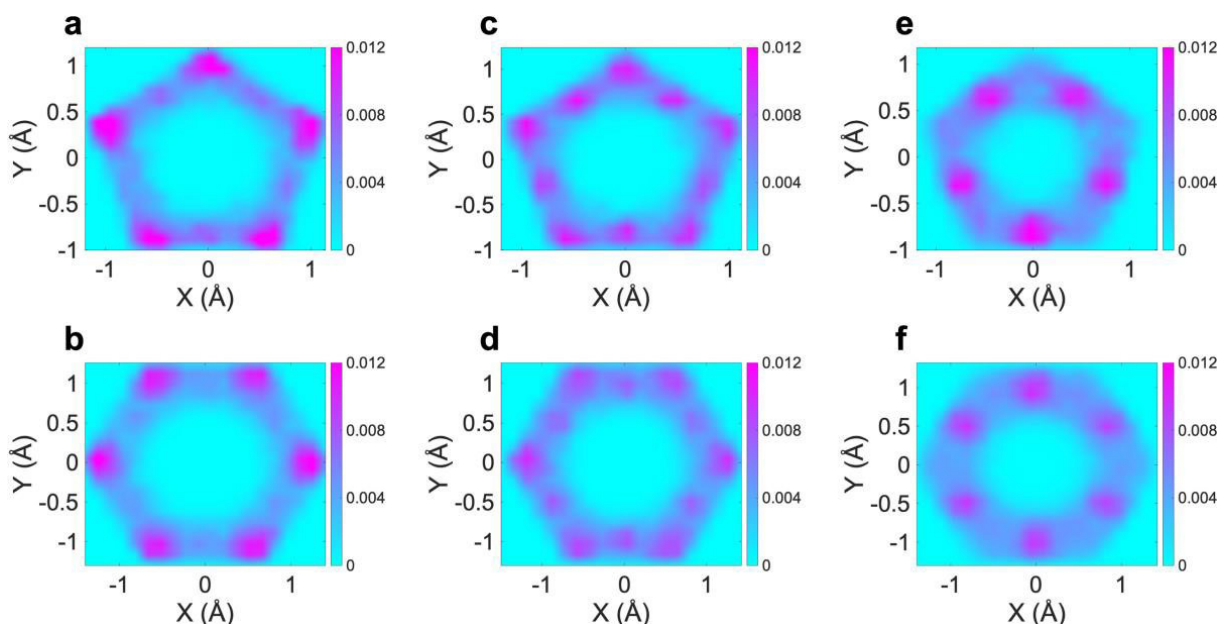


Fig. S11 Atomic stacking for $T=50 \text{ K}$. Probability distribution of vertex projection into C5 (top row) and C6 (bottom row). Normal force varies from (a-b) 3.2 nN, (c-d) 32 nN, to (e-f) 320 nN. Under half roughness model, AIREBO, $V = 1 \text{ m/s}$.

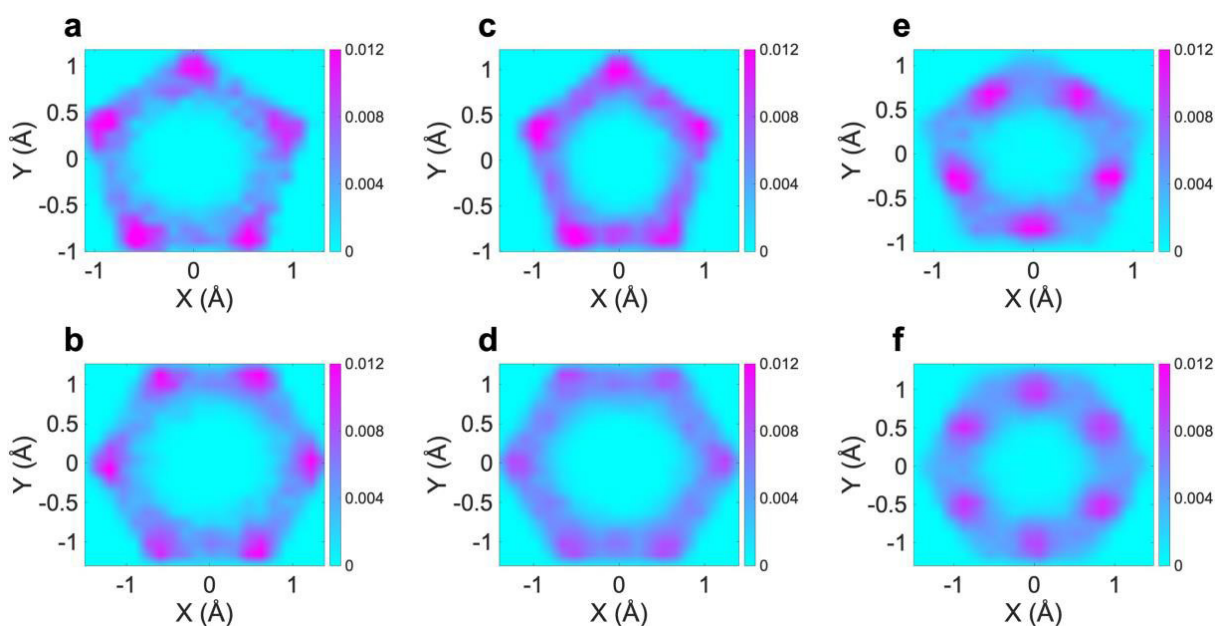


Fig. S12 Atomic stacking for higher shear velocity. Probability distribution of vertex projection into C5 (top row) and C6 (bottom row). Normal force varies from (a-b) 3.2 nN, (c-d) 32 nN, to (e-f) 320 nN. Under half roughness model, AIREBO, $V = 10$ m/s, $T = 300$ K.

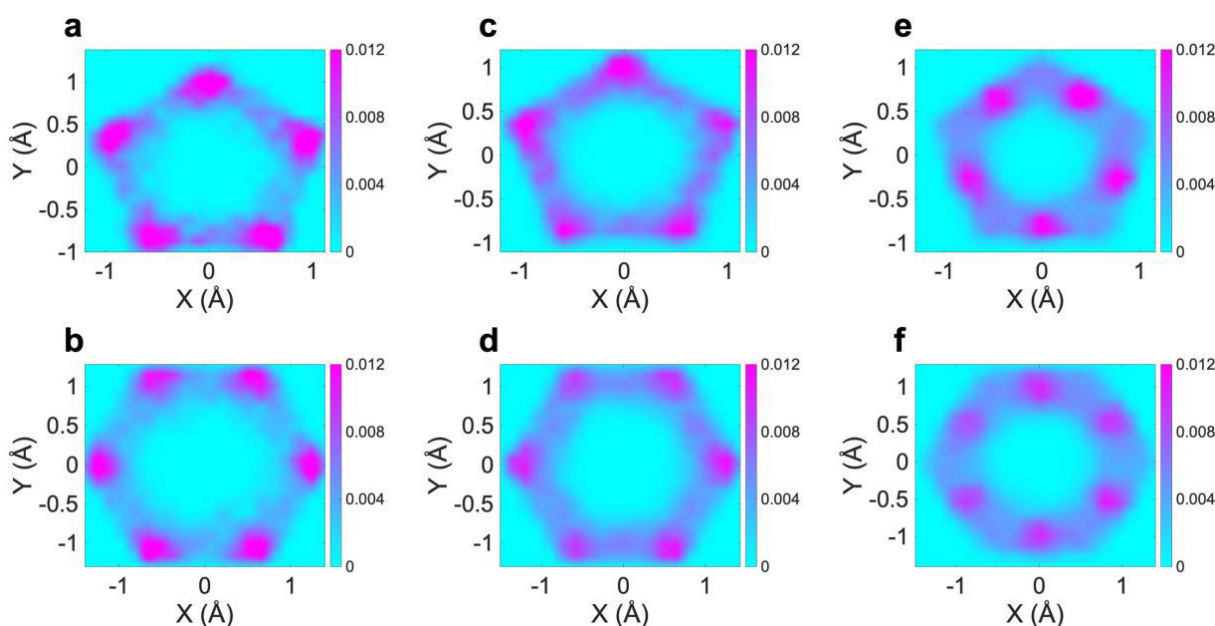


Fig. S13 Atomic stacking for flat boundary. Probability distribution of vertex projection into C5 (top row) and C6 (bottom row). Normal force varies from (a-b) 3.2 nN, (c-d) 32 nN, to (e-f) 320 nN. Under flat boundary model, AIREBO, $V = 1$ m/s, $T = 300$ K.

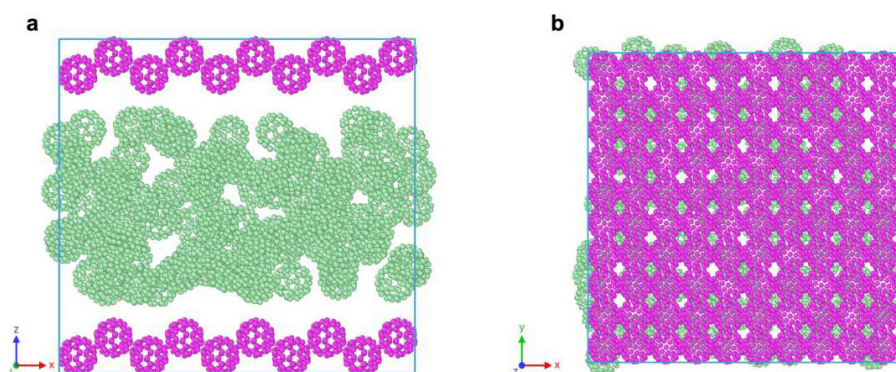


Fig. S14 Wider simulation model. Molecular dynamics simulation model under larger size of 10×10 periodic patterned slabs. (a) Front and (b) top views.

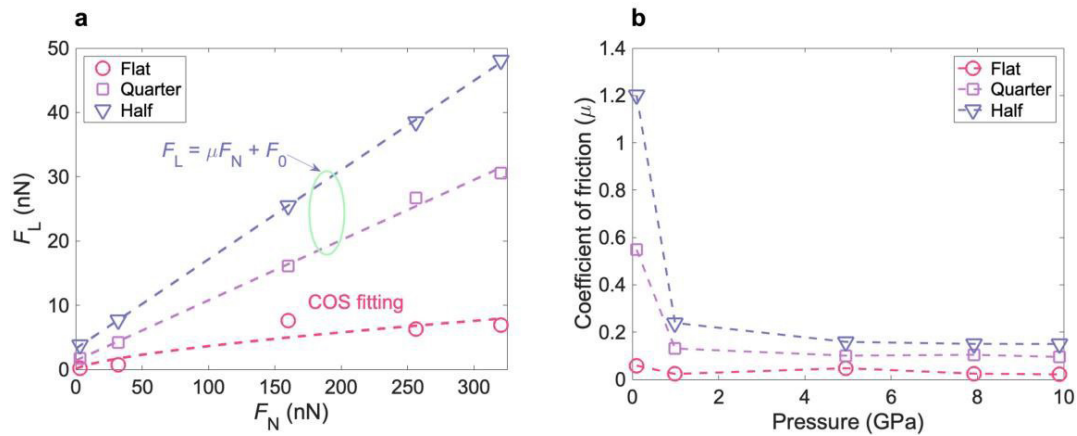


Fig. S15 Friction behavior for wider simulation model. (a) Friction force versus normal force and the transition parameter $\alpha_T = 0.33$ for the fitting of COS model. (b) The coefficient of friction ($\mu = F_L/F_N$) as a function of pressure.

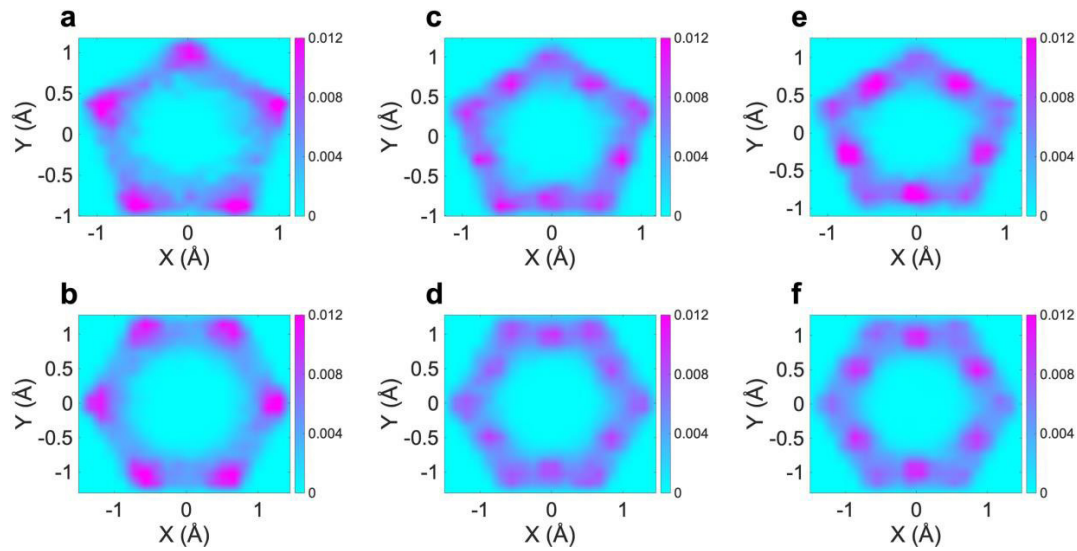


Fig. S16 Atomic stacking for wider simulation model. Probability distribution of vertex projection into C5 (top row) and C6 (bottom row). Normal force varies from (a-b) 32 nN, (c-d) 192 nN, to (e-f) 320 nN. Under big area (10×10) of rough boundary model, AIREBO, $V = 1$ m/s, $T = 300$ K.

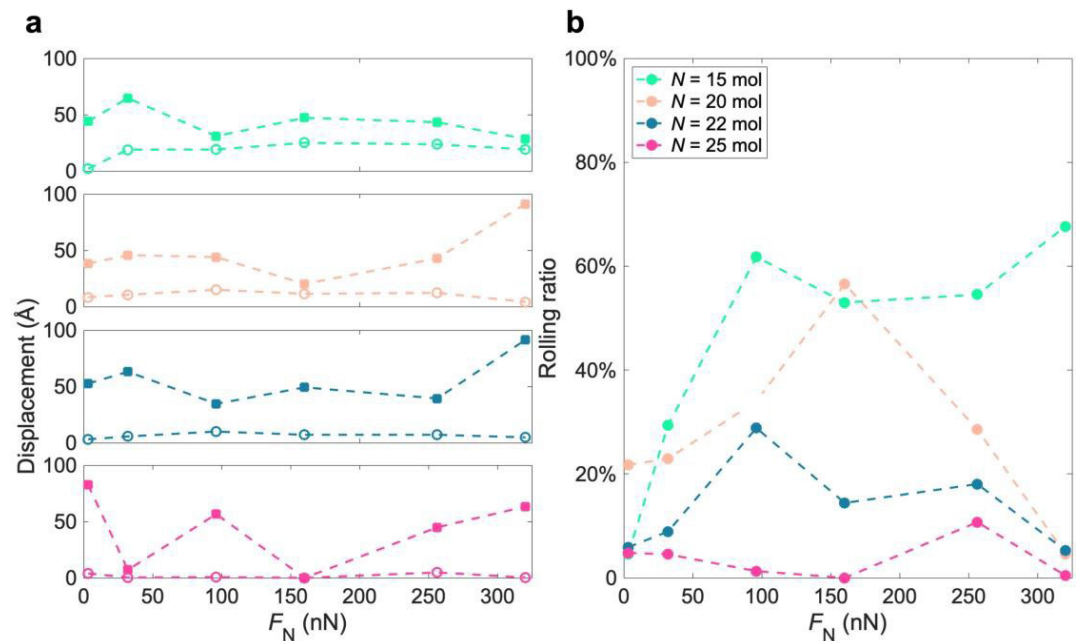


Fig. S17 Rolling contribution in the fullerene monolayer. (a) Rolling displacement (empty circle) and the total displacement (solid square) of particles. (b) Rolling ratio versus normal force. Rolling and translation displacement under flat boundary conditions for different fullerene coverages.

contribution of rolling to the total displacement is shown as a function of normal force in Fig. S17a. Interestingly, the contribution of rolling decreases as the fullerene density is increased, indicating that fullerene-fullerene-interactions block their mutual rotation against each other. This is likewise observed in Fig. S17b, where we show the rolling contribution to the total displacement. At small fullerene density, a high rolling contribution is observed, which almost vanishes for high fullerene density.

References

- [1] Brenner D W, Shenderova O A, Harrison J A, Stuart S J, Ni B, Sinnott S B. A second- generation reactive empirical bond order (REBO) potential energy expression for hydrocarbons. *Journal of Physics: Condensed Matter* **14**(4): 783–802 (2002)
- [2] Legoas S B, Giro R, Galvao D S. Molecular dynamics simulations of C60 nanobearings. *Chemical Physics Letters* **386**(4-6): 425-429 (2004)
- [3] Stuart S J, Tutein A B, Harrison J A. A reactive potential for hydrocarbons with intermolecular interactions. *Journal of Chemical Physics* **112**(14): 6472–6486 (2000)
- [4] Kolmogorov A N, Crespi V H. Registry-dependent interlayer potential for graphitic systems. *Physical Review B* **71**(23): 235415 (2005)
- [5] Reguzzoni M, Fasolino A, Molinari E, Righi M C. Potential energy surface for graphene on graphene: Ab initio derivation, analytical description, and microscopic interpretation. *Physical Review B* **86** (24): 245434 (2012)
- [6] Li H, Kim W K. A comparison study between the Lennard-Jones and DRIP potentials for friction of graphene layers. *Computational Materials Science* **180**: 109723 (2020)
- [7] Wen M, Carr S, Fang S, Kaxiras E, Tadmor E B. Dihedral-angle-corrected registry- dependent interlayer potential for multilayer graphene structures. *Physical Review B* **98**(23): 235404 (2018)
- [8] Wen M, Tadmor E B. Hybrid neural network potential for multilayer graphene. *Physical Review B* **100**(19): 195419 (2019)
- [9] O'Connor T C, Andzelm J, Robbins M O. AIREBO-M: A reactive model for hydrocarbons at extreme pressures. *J Chem Phys* **142**(2): 024903 (2015)
- [10] Los J H, Fasolino A. Intrinsic long-range bond-order potential for carbon: Performance in monte carlo simulations of graphitization. *Physical Review B* **68**(2): 024107 (2003)
- [11] Zhu S, Pochet P, Johnson H T. Controlling rotation of two-dimensional material flakes. *ACS Nano* **13**(6): 6925-6931 (2019)
- [12] Campbell S E, Luengo G, Srdanov V I, Wudl F, Israelachvili J N. Very low viscosity at the solid-liquid interface induced by adsorbed C60 monolayers. *Nature* **382**(6591): 520–522 (1996)
- [13] Liang Q, Tsui O K, Xu Y, Li H, Xiao X. Effect of C60 molecular rotation on nanotribology. *Phys Rev Lett* **90**(14): 146102 (2003)
- [14] Schwarz U D, Allers W, Gensterblum G, Wiesendanger R. Low-load friction behavior of epitaxial C60 monolayers under hertzian contact. *Phys Rev B Condens Matter* **52**(20): 14976–14984 (1995)
- [15] Okita S, Matsumuro A, Miura K. Tribological properties of a C60 monolayer film. *Thin Solid Films* **443**(1-2): 66–70 (2003)
- [16] Miura K, Kamiya S, Sasaki N. C60 molecular bearings. *Physical Review Letters* **90**(5): 055509 (2003)
- [17] Johnson K L, Kendall K, Roberts A D. Surface energy and the contact of elastic solids. *Proceedings of the Royal Society of London. A. Mathematical and Physical Sciences* **324**(1558): 301–313 (1971)
- [18] Maugis D. Adhesion of spheres: The JKR-DMT transition using a dugdale model. *Journal of Colloid and Interface Science* **150**(1): 243–269 (1992)
- [19] Carpick R W, Ogletree D F, Salmeron M. A general equation for fitting contact area and friction vs load measurements. *J Colloid Interface Sci* **211**(2): 395–400 (1999)
- [20] Mo Y, Turner K T, Szlufarska I. Friction laws at the nanoscale. *Nature* **457**(7233): 1116–1119 (2009)
- [21] Liu S W, Wang H P, Xu Q, Ma T B, Yu G, Zhang C, Geng D, Yu Z, Zhang S, Wang W, Hu Y Z, Wang H, Luo J. Robust microscale superlubricity under high contact pressure enabled by graphene-coated microsphere. *Nat Commun* **8**(1): 14029 (2017)
- [22] Prokopovich P, Starov V. Adhesion models: From single to multiple asperity contacts. *Adv Colloid Interface Sci* **168**(1-2): 210–222 (2011)
- [23] Yu R C, Tea N, Salamon M B, Lorents D, Malhotra R. Thermal conductivity of single crystal C60. *Phys Rev Lett* **68**(13): 2050–2053 (1992)