

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

D. Zhou^{a,b}, A.K. Gupta^a, K. Bi^b, A.S. de Wijn^c, P. Schall^{a,*}

^a Institute of Physics, University of Amsterdam, the Netherlands

^b Jiangsu Key Laboratory for Design and Manufacture of Micro-Nano Biomedical Instruments, School of Mechanical Engineering, Southeast University, China

^c Department of Mechanical and Industrial Engineering, Norwegian University of Science and Technology, Norway

ARTICLE INFO

Keywords:

Superlubricity
Fullerenes
Multi-contact heterostructure
Rolling
Sliding
Stacking order

ABSTRACT

Recent research shows that superlubric 2D materials offer great potential for friction reduction, yet their requirements of flatness and small scale hinder real-world applications. Here, we study a multi-contact van der Waals heterostructure, composed of fullerene (C₆₀) molecules sandwiched between molybdenum disulfide (MoS₂) atomic layers by molecular-dynamics simulations. We show that despite the multi-contact geometry, the confined layer of C₆₀ nanoparticles exhibits superlubric behavior at high fullerene density or high applied normal force, where the fullerenes' rotational degrees of freedom are suppressed and the MoS₂ layer slides over the fullerenes. At lower fullerene density or load, fullerene rolling leads to increased frictional dissipation. We demonstrate that the atomic stacking at the fullerene-MoS₂ contact evolves from aligned into misaligned with increasing fullerene coverage, reflecting the increasing fullerene-fullerene interactions suppressing their rolling degrees of freedom. Thus, counterintuitively, in the fullerene-MoS₂ heterojunction system, rolling of the fullerenes is not beneficial for friction reduction, as superlubric slip offers an alternative, lower-friction mechanism. These results on the multi-contact sliding of a heterojunction of fullerene nanoparticles and 2D material are useful for guiding nanoscale superlubric properties into macroscopic scale superlubric applications.

1. Introduction

Energy dissipation and material failure resulting from friction and abrasion pose serious concerns in materials science and engineering, especially in machinery components. Two-dimensional (2D) or van der Waals (vdW) layered materials [1,2], have been shown to exhibit superior lubrication performance in dry contact, due to structural superlubricity. It has been demonstrated that the combination of two different 2D materials, for instance, the stacking of a single layer of graphene and hexagonal boron nitride (h-BN), or layers of transition metal dichalcogenides such as molybdenum or tungsten disulfide (MoS₂/WS₂), possess uniquely low friction coefficients, along with extraordinary thermal and optical properties [3–7]. These extraordinary physical properties of the heterojunction result from the unique combination of their physical characteristics at the interface such as their band alignment and atomic lattice stacking.

Structural superlubricity, which indicates the sliding of two surfaces at vanishing or near-zero friction coefficient due to structural incompatibility [8,9], can be achieved either in an incommensurate

alignment of two identical 2D materials (homojunction) or an arbitrary alignment of different ones (heterojunction). In both cases, the friction and physical properties hugely depend on the bilayer stacking degrees of freedom (DOF) [10,11], which are related to the twist angle in homogenous stacks, and to the intrinsic lattice mismatch in heterostructures. The strongly twist angle dependent friction behavior of two stacked layers of the same vdW material was demonstrated in [12,13]. On the other hand, a good lattice mismatch of two different materials in a vdW heterostructure similarly provides a super-low friction coefficient and even exhibits twist-angle independence [14]. The superlubricity of heterostructures holds notable potential in applications of nanoelectromechanical systems with large surface-to-volume ratios, including nanoelectronics, optoelectronic and electrochemical devices [15,16].

Great progress has been made in the measurement of superlubric behavior, yet the observation of superlubricity is usually rather limited to the micrometer scale [17–20]. There are significant challenges in scaling up superlubricity from micron scale to large-dimensional superlubric systems, precluding the exploitation of superlubricity in

* Corresponding author.

E-mail address: p.schall@uva.nl (P. Schall).

<https://doi.org/10.1016/j.triboint.2025.111317>

Received 2 July 2025; Received in revised form 30 September 2025; Accepted 15 October 2025

Available online 16 October 2025

0301-679X/© 2025 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

macroscopic applications. In practical macroscopic applications, surfaces are always rough [21–23], exhibit contaminations [24–26], defects and wear damage [27–29], precluding the straightforward application of structural superlubricity. To address these obstacles, one possible route is to construct multi-contact junctions that allow superlubric patches or particles to move randomly within the contact interfaces, while supporting the applied load [30–33]. To this end, graphfullerene [34,35], a polymeric fullerene network, is expected to be a superlubric material providing lubrication via multi-contacts at the rubbing interfaces [36]. This synthesized two-dimensional fullerene network, which is connected in a quasi-hexagonal fullerene structure, has been shown to provide good stability and high strength [35,37,38]. However, fixing the fullerenes in a network restricts their rotational DOF in the monolayer, while adaptive rotational and translational DOFs are crucial for enabling the ball bearing effect of the nanoparticles [39,40]. It remains unclear whether the rolling of molecular particles leads to friction reduction at nanoscale. Neither the findings of experiment [41, 42] nor those of simulations [43,44] showed conclusively that the rotational DOF yielded lower friction than sliding at the microscopic level. Depending on the size of the contact and its surface separation, the van der Waals force-related adhesion interactions and interfacial bonds are responsible for frictional dissipation [45]. A simple model of a C_{60} /MoS₂ heterojunction has been experimentally found to exhibit an ultralow coefficient of friction [46], however the lubrication mechanism of nanoparticles remains unclear. Fullerene nanoparticles in a ball-plate heterogeneous interfaces have been rarely studied, and in particular the resulting multi-contact structure at the molecular level, and its load- and friction dependence needs to be elucidated to obtain insight into the ultra-low friction mechanism of these heterojunctions.

Here, we investigate the superlubric properties and friction mechanisms of a C_{60} /MoS₂ heterojunction to obtain insight into the friction-related DOFs of the fullerene particles. We use molecular-dynamics (MD) simulations to study a monolayer of C_{60} fullerenes sandwiched

between two MoS₂ slabs and elucidate their deformation and rotational properties as a function of fullerene density and applied load. We find a transition from superlubric to finite friction behavior: while a high fullerene coverage reduces their load-induced flattening that suppresses rolling, too high of a coverage causes fullerenes to interact with each other, which again suppresses rolling. The resulting slip of the MoS₂ layer in the absence of rolling leads to an ultralow friction coefficient in the superlubric regime. At the atomic scale, the flattening causes alignment of the atomic motifs at the interface, which is lost as fullerene-fullerene interactions dominate at high coverage. These results on the superlubric behavior of a multi-contact heterojunction are useful for designing lubrication systems that work under dry and vacuum environment, especially in nanoelectromechanical systems with large aspect ratios.

2. Computational details

We simulate a layer of fullerene molecules (C_{60}) between two atomic monolayers of 2D molybdenum disulfide (MoS₂), as shown in Fig. 1(a, b). The bulk lattice parameters of MoS₂ along its *x*, *y* and *z* axes are 3.16 Å, 5.48 Å and 18.41 Å, respectively. The system size is $W \times L \times H \sim 73 \text{ Å} \times 71 \text{ Å} \times 25 \text{ Å}$, which we keep constant, while varying the number (*N*) of fullerene particles in between the MoS₂ monolayers, increasing from 40 to 60–80, corresponding to areal number densities of 0.77, 1.16 and 1.54 nm^{-2} . Simulations performed in a significantly larger system show very similar results, see SI; we thus conclude that there are no significant finite size effects. To model the interactions of the fullerenes with each other, we use the force field for fullerene C_{60} molecules as described by the AIREBO potential [47], which is a reactive force field allowing bonds to disassociate and reconnect. The interactions within the MoS₂ substrate are modelled using the Stillinger-Weber (SW) potential [3,48], expressing three-body interactions. Furthermore, to mimic the vdW interactions between the C_{60}

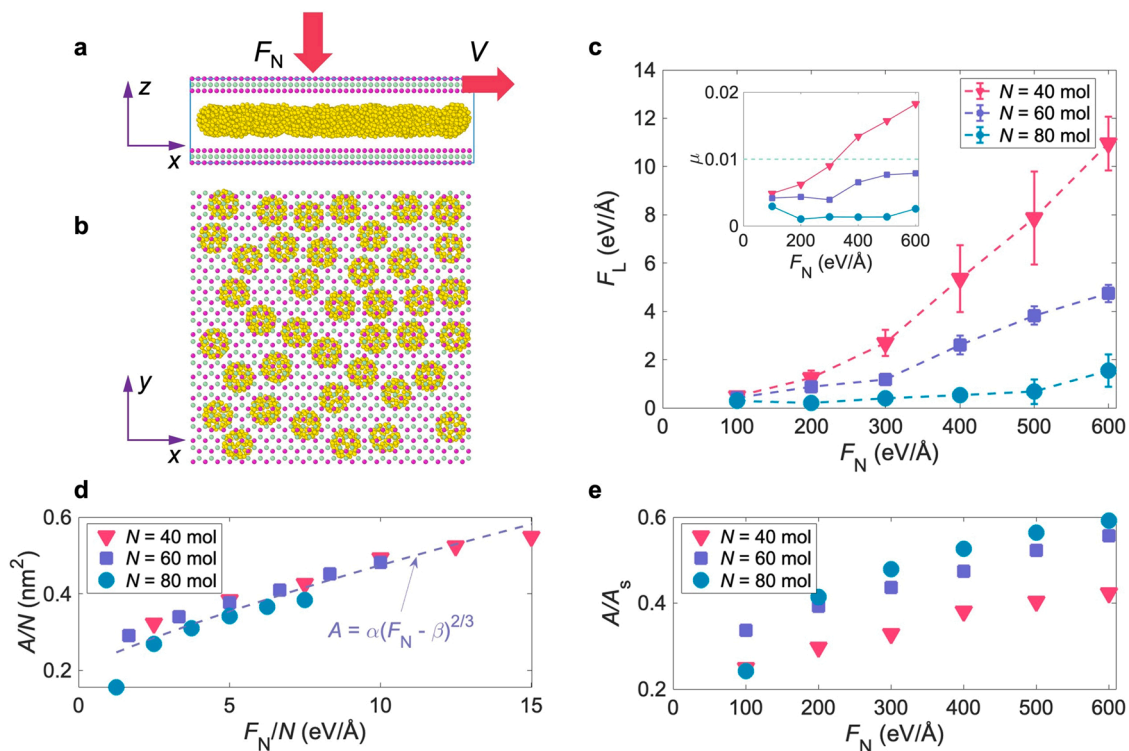


Fig. 1. MD simulation model and friction behavior. (a-b) Front and top views of simulation model. (c) Friction force versus normal force for different number of fullerenes between the slabs (see legend). Error bars are calculated as standard deviation over five independent runs. Inset: resulting friction coefficient determined from the data in the main panel. (d) Average contact area (*A*) per particle as a function of average load per particle. Hertzian behavior is observed, see fit (dashed line). (e) Fraction of the contact area with respect to the total surface area (A_s) as a function of normal load.

and MoS₂, the Lennard-Jones (6–12 LJ) potential is applied according to

$$E_{ij}^{LJ}(r_{ij}) = 4\epsilon_{ij} \left[\left(\frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}} \right)^6 \right] \quad (1)$$

where r_{ij} is the interaction distance between atoms of the two different entities, with i representing carbon (C) atoms, and j representing sulphur (S) or molybdenum (Mo) atoms, and ϵ_{ij} and σ_{ij} are the attractive well depth, and equilibrium distances, respectively. The LJ potential is thus described by the well depth ϵ_{ij} and the equilibrium distance σ_{ij} .

These two LJ parameters are determined separately for the C-S and C-Mo interactions according to the standard Lorentz-Berthelot mixing rules [14,49,50]. Here, the parameters $\epsilon_{C-S} = 2.60342$ meV, $\epsilon_{C-Mo} = 1.2351$ meV, $\sigma_{C-S} = 4.0$ Å and $\sigma_{C-Mo} = 4.025$ Å, are determined based on the interactions of a graphene/MoS₂ heterostructure [49]. We furthermore set the cutoff distance to 10.2 Å for both the LJ and the AIREBO interactions. To lower the computational cost and because they are well-separated during the entire simulation, we neglect the interactions between the opposing MoS₂ slabs.

To simulate the shearing of the sandwich structure under compression, the topmost atomic layer (top S atom layer in the top MoS₂ sheet) is treated as a rigid body and the downmost atomic layer (bottom S atom layer in the bottom MoS₂ sheet) is entirely fixed. Initially, the time step is 1 fs and energy minimization with the conjugate gradient algorithm is used for optimization of the fullerene structure, where the energy and force tolerance are set to 10^{-6} , after which the structure of the system is further relaxed in the canonical ensemble (NVT) for 1 ns to eliminate internal stresses and reach equilibrium. The temperature of the system is controlled by Langevin thermostats with a damping time of 100 fs at the bottom and top Mo layers, acting as a symmetrical heat bath to keep the system temperature at 5 K or 300 K. Then, a normal force F_N in the range of 0–600 eV/Å is applied to the upmost rigid layer of S atoms, and the system is relaxed for 2 ns to reach an equilibrated state under compression. In this normal-force regime, the fullerenes remain intact throughout the experiment, while we observed that higher applied loads could eventually break them. Ellipsoids are fitted to the atomic coordinates of fullerenes using a linear least-squares method solved via singular value decomposition (SVD). The principal axes and their lengths are then obtained by eigenvalue decomposition of the fitted quadratic form matrix. The aspect ratio (b/a) is defined as the ratio of the length of the minor axis to that of the major axis. Finally, non-equilibrium molecular dynamics simulations are performed, moving the upper rigid slab at a constant velocity, while recording the shearing force on the top rigid layer. We checked that the shear force fluctuates around a constant, normal-force dependent value, see SI: due to the elasticity of the C₆₀ nanoparticles as well as their contacts with the MoS₂ layer, the heterosystem is significantly compliant, so that a spring is not necessary for pulling the top layer, and a constant velocity driving method for the shearing process is preferred. The shearing process lasts 10 ns for a total distance of 10 nm, corresponding to a velocity of $v = 0.01$ Å/ps. In the simulations, the time-averaged shear force is output every 1000 fs. To calculate the mean friction force, data from the initial 1 ns is discarded due to transient effects, and the mean friction force is calculated as the average over the remaining 9 ns. The LAMMPS package is used to carry out all the MD simulations [51].

3. Results

The steady-state shear force on the top slab is shown as a function of normal force in Fig. 1c. It depends strongly on the fullerene coverage: counterintuitively, the higher the fullerene coverage, the lower the sliding force that is observed. This is contrary to a homogeneous system in which lubricating fullerene molecules and confining substrates are made of the same molecules, as observed in experiments on C₆₀ fullerenes between graphite substrates [52,53], and supported by simple two-dimensional models of circular fullerenes confined in between

atomic chains [45,54]. The data in Fig. 1c suggest that, above some coverage-dependent normal force, the friction force increases linearly with the normal force; the slope then gives the friction coefficient μ , as shown in the inset. Superlubric behavior ($\mu < 0.01$, dashed line) is observed for high coverages, $N = 60$ and 80. We investigated the sliding at different temperatures and found that temperature has only little effect on the friction force, as shown in Fig. S1 of the supporting information (SI). To investigate the contact formation and dependence on fullerene coverage in more detail, we looked at the contact area between the fullerenes and the MoS₂ plane. For an individual fullerene, the average contact area per fullerene (A/N) shows a Hertzian behavior as a function of applied load (F_N/N) on a single fullerene, as shown in Fig. 1d. Here, the contact area between the fullerene and MoS₂ has been determined from the number of C atoms of the fullerene that are in the range of the equilibrium distance (around 4 Å) from the closest sulphuric layer of the MoS₂. In the multi-contact system, the contact area per particle is significantly reduced, while the total contact area increases with normal load in a way similar to that of the single particle, as shown in Fig. 1e.

To elucidate the squeezing of the fullerenes in more detail, we look at the slab separation as a function of normal force, as shown in Fig. 2a. Generally, the separation decreases with applied load, reflecting the squeezing of the fullerenes. A higher fullerene coverage keeps the slabs further apart, reflecting the increasing support of the applied load. Mostly, the fullerenes are compressed into a single plane; only at the lowest load and highest fullerene density, thermal fluctuations are strong enough to push particles out of plane, as reflected in a sudden increase in the slab separation at low F_N . The nearing of the slabs causes (i) enhanced interactions of the fullerenes and slab due to shorter mutual distances, and (ii) elastic squeezing of the fullerenes causing them to flatten. The latter is clearly reflected in the aspect ratio of the loaded fullerenes, which we calculate from the two major axes of a fitted spheroid, as shown in Fig. 2b. As the normal force increases, the spherical fullerenes flatten into ellipsoids with aspect ratio down to ~ 0.7 , extending their surface interaction from point contact to finite contact area. The elastic squeezing of the fullerenes is counteracted by higher surface coverage: as the normal force is spread over more molecules, this reduces the normal load per fullerene, and hence its deformation. The latter is always below the elastic limit of a single fullerene, which has been determined to be about 2.3 Å for the minor semi axis [55]; hence, the fullerenes stay intact and do not fracture at the loads studied here. Nevertheless, the flattening of the fullerenes affects their rolling ability under applied sliding.

To elucidate this effect and obtain insight into the rolling versus sliding motion, we look at the rolling of individual particles. We first do this at $T = 5$ K, where the trend is most clear as thermal fluctuations are small. Examples of rolling trajectories, which we obtain by tracking a single atom on the fullerene shell, are shown in Fig. 3. Rolling occurs in a medium range of the applied normal forces, as shown in Fig. 3(c–f). At lower loads, in contrast, the particle only wiggles and appears to slide along the MoS₂ slabs (Fig. 3a, b); while at the highest loads a more chaotic motion involving rolling is observed (Fig. 3g, h). At higher temperature, a similar trend is observed, yet rotational diffusion adds to the rolling motion, see Fig. S3 in the SI. This is most pronounced at low applied normal load, where the particle exhibits nearly free rotational diffusion, causing the emergence of rolling to be less distinct.

This trend is confirmed and quantified when we look at the rotational autocorrelation of the fullerenes in between the MoS₂ slabs. We compute time correlations in the rotation of the fullerenes according to

$$C(t) = \left\langle \frac{1}{N} \sum_{i=1}^N U_i(0) \cdot U_i(t) \right\rangle \quad (2)$$

where U_i is the unit vector from the center of fullerene i to a point on its surface, N is the number of fullerenes in the system, and angular brackets denote the canonical ensemble average. Eq. (2) computes the rotational autocorrelation function related to the motion of the

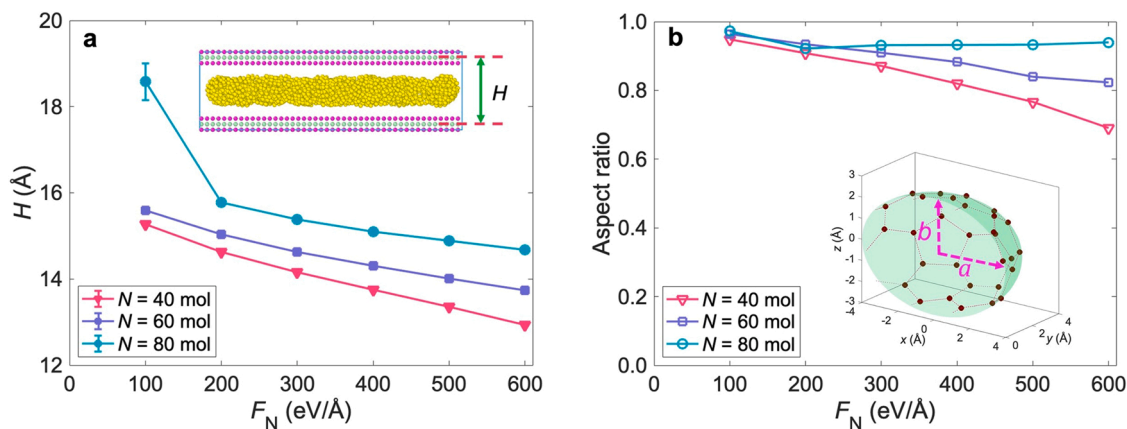


Fig. 2. Structural deformation. (a) Height of contact interface between the center of mass of top and bottom slabs, see simulation snapshot in the inset. (b) Aspect ratio (b/a) of fullerenes computed from the semi major minor axis of a fitted ellipsoid, see inset as an example for the case of $N = 40$ and $F_N = 600$ eV/Å.

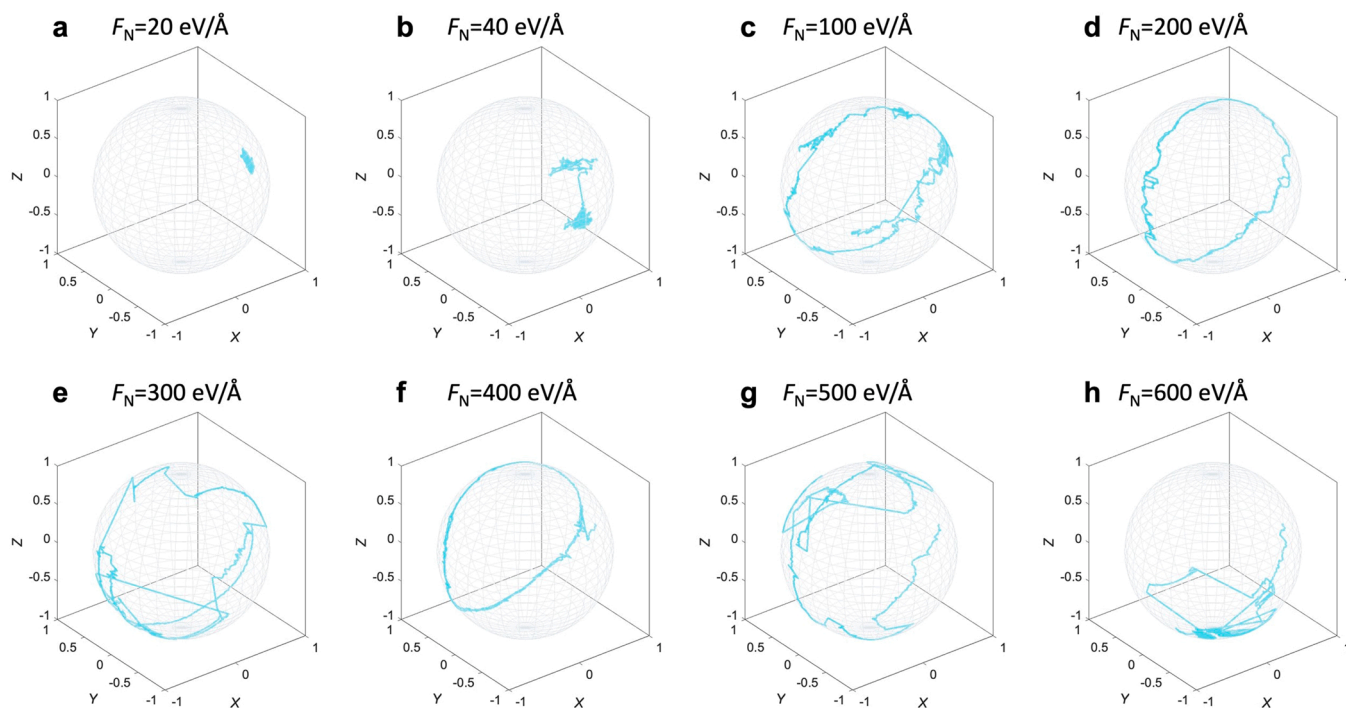


Fig. 3. Representative rolling trajectories on the unit sphere surface. (a-h) Trajectory of a point on a single fullerene sphere in the ensemble for various normal forces, see panel titles. The trajectory is shown for a shear duration of 10 ns in the sandwich model. $T = 5$ K. $N = 40$ mol, $V = 0.01$ ps/Å.

nanoparticles during shear, with the scalar variable $C(t) \in [-1, 1]$. When the particles roll in sync, their orientation vectors rotate together, and $C(t)$ shows a sinusoidal shape, as shown in Fig. 4a at intermediate applied load. In contrast, when sliding prevails and rotation is suppressed, C approaches a constant as is indeed observed for low and very high applied load in Fig. 4a. Hence, the transition from constant C at low load, to sinusoidal behavior at intermediate applied load indicates the transition from wiggling to rolling of the particles, in agreement with the individual particle trajectory in Fig. 3; the increasing amplitude of the sinusoidal function then indicates the increasing correlation in the particle's rolling angles under increasing applied load. At even higher normal force, however, the amplitude of the sinusoidal function decreases again until it vanishes almost completely, indicating that the motion becomes dominated by sliding or strong decorrelation in the rotation angles. This is indeed in agreement with Fig. 3g and h showing less rotation at high load. At room temperature, the trend remains; yet, the particles decorrelate significantly more at low loads due to their

pronounced rotational diffusion, as shown in Fig. S4 of the SI. The situation changes drastically at high particle density (Fig. 4b at $T = 5$ K), where sinusoidal behavior is no longer observed. Here, $C(t)$ merely shows a small decay, indicating that rolling is suppressed. The decay is somewhat more pronounced at room temperature (inset in Fig. 4b), but still limited to $C(t) \sim 0.9$. We hypothesize that the steric interactions of the neighboring particles in the dense ensemble restrict the rotational motion and the particles slide along the slab. This is confirmed when we look at the average rotation angle of the particles as a function of time, shown in SI Fig. S6. At high coverage, the rotation angle evolves much more slowly than at lower coverages, indicating suppression of rolling. Interestingly, this does not lead to a higher friction force; on the contrary, the highest particle coverage is associated with the lowest friction coefficient (Fig. 1c), indicating that sliding along the MoS₂ slab leads to the most superlubric behavior.

The contact-related sliding of the nanoparticles is further evident from the height and velocity distribution of the fullerene atoms, as

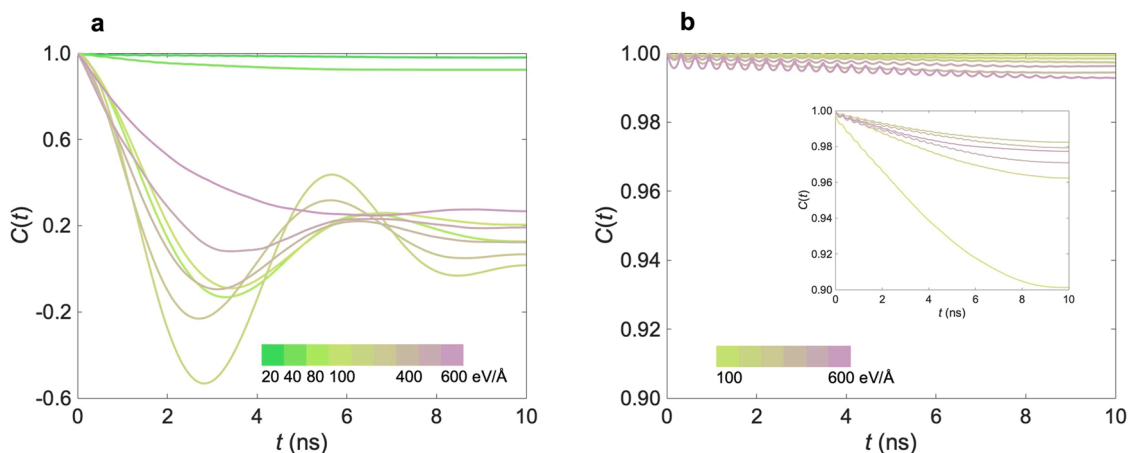


Fig. 4. Rotational autocorrelation function versus delay time. Time evolution of the rotational autocorrelation function at $T = 5$ K and $V = 0.01$ ps/Å, for different normal forces, increasing from the green to purple, see color bar. (a) $N = 40$ mol and (b) $N = 80$ mol, inset in (b) shows the same autocorrelation function at $T = 300$ K.

shown in Fig. 5. At low coverage, sharp peaks in the height distributions evolving with increasing normal force (Fig. 5a) indicate the formation of an adhesive binding area between the MoS₂ and the fullerenes [56]; at the same time, the gradient in the height-dependent velocity (Fig. 5b) indicates the rolling of the fullerenes. Both the structural and velocity profiles together thus indicate adhesion-induced rolling. The signatures diminish at higher coverage (Fig. 5c and d), indicating that the loss of adhesion contact leads to a transition from rolling to sliding.

To obtain full atomistic insight into the fullerene-MoS₂ adhesion interaction, we investigate the atomic stacking at the contact interface. We first investigate the zenith angle ψ that changes continuously when the particle is rolling. We define ψ as the angle between the normal vector of the nearest pentagonal or hexagonal motif of the fullerene facing the MoS₂ and the normal vector of the MoS₂ plane, as defined in the inset of Fig. 6a. This angle is widely distributed at low normal force as shown in the main panel Fig. 6a, as expected for the rolling of the fullerenes. Yet, when the normal force is increased, the distribution

narrows, indicating the increasing alignment of the fullerenes' polygon motif with the atomic motif of the MoS₂ slab as the fullerene flattens under the applied load. This alignment further substantiates the formation of a contact area with increasing load in Fig. 5a. As the fullerene coverage increases, the peak broadens again (see Fig. 6b and c), in line with the loss of a clearly defined contact area in Fig. 5c. Furthermore, the distributions for low and high normal load become more similar, reflecting the increasing load-bearing ability of the fullerenes, and resulting lack of flattening and contact formation at higher fullerene density. We also investigate the in-plane twist angle ϕ between the carbonate ring orientation and the zigzag direction (armchair direction along the y-axis) of the MoS₂ slab, see inset of Fig. 6d. This twist angle shows a similar trend as the zenith angle ψ : the flat distributions sharpen as the normal force is increased, reflecting the increasing contact interactions with the MoS₂ as the fullerenes flatten under the increasing load. This is especially observed for low fullerene coverage (Fig. 6d), for which the largest normal forces cause a nearly aligned state with $\phi \sim 0^\circ$.

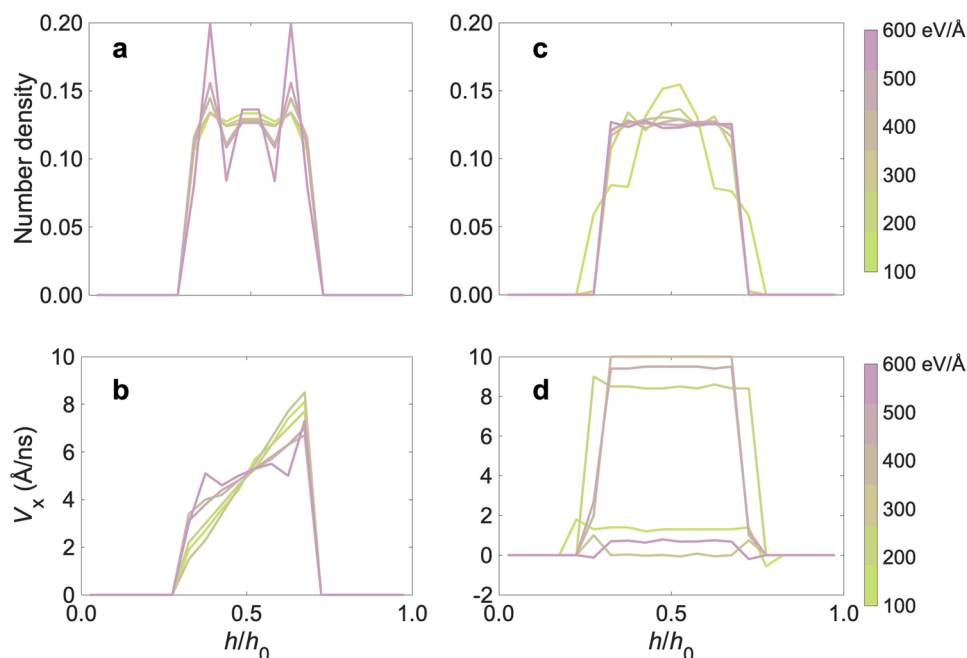


Fig. 5. Structural profile and velocity distribution. (a, b) Time-averaged height profile (a) and velocity distribution in the shearing direction (b) of carbon atoms under low fullerene coverage, $N = 40$. (c, d) Same quantities for high fullerene coverage, $N = 80$.

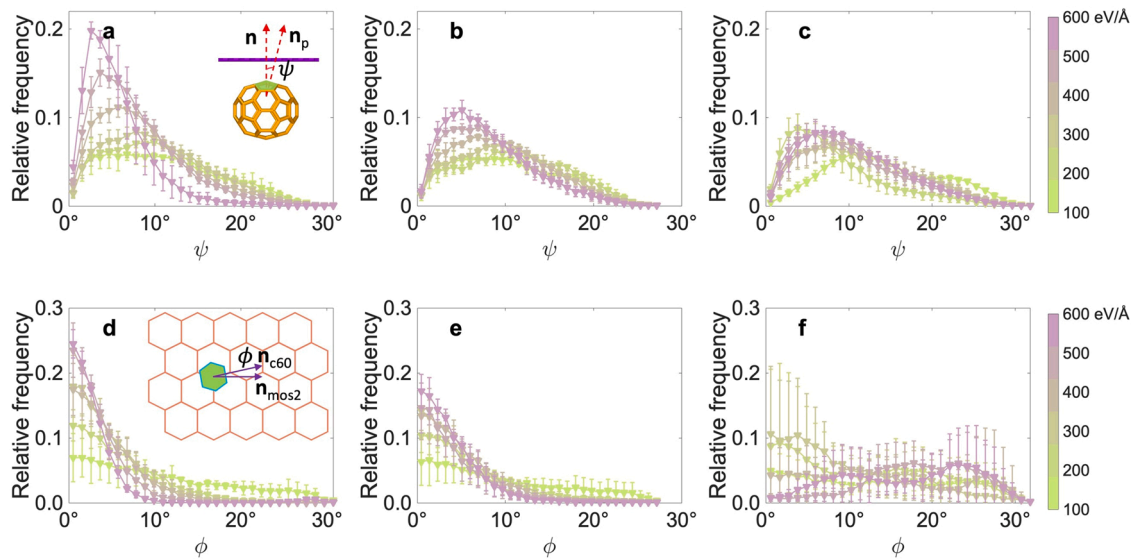


Fig. 6. Atomic alignment at the fullerene-MoS₂ contact. (a-c) Probability distributions of the zenith angle (ψ) under different normal forces (see color bar), for fullerene coverage of (a) $N = 40$ mol, (b) $N = 60$ mol and (c) $N = 80$ mol. The out-of-plane zenith contact angle ψ between the C₆₀ and MoS₂ is defined in the inset of (a), where $\mathbf{n}_p$ is the normal vector of the fullerene motif nearest to the MoS₂, and $\mathbf{n} = [0\ 0\ 1]$ is the normal vector of the MoS₂ plane. (d-f) Corresponding probability distribution of the twist angle (ϕ) at the same fullerene coverages as in (a-c). The twist angle ϕ is defined between the edge normal vector ($\mathbf{n}_{C60}$) of the nearest hexagons of C₆₀ and the zigzag direction ($\mathbf{n}_{MoS2}$) of MoS₂ plane, see inset in (d). Only the contacts of unbroken fullerenes with the nearest six-membered ring are taken into consideration. In all distributions, error bars show the standard deviation in the relative frequency of the angles.

This alignment is in line with the simulation results obtained by Silva et al. [50], who studied the rotational energy landscape of a layered heterostructure of MoS₂/graphene which showed a stable alignment at 0° or 60°. With increasing coverage (Fig. 6e, f), the distinction between high and low normal force diminishes, as fullerene-fullerene interactions arise and compete with the fullerene-substrate interactions. For the highest coverage $N = 80$, the trend even reverts: the misalignment angle jumps closer to 30°, as shown in Fig. 6f. This twisted alignment of C₆₀/MoS₂ is similar to the experimental observations of frequently aligned orientation in the MoS₂ growth on epitaxial graphene by Liu et. al [57], who also observed a 30° rotated growth between the graphene and MoS₂ lattice. In our case of fullerenes, the transition from aligned to misaligned stacking is caused by the fullerene-fullerene interactions: an isolated C₆₀ molecule can rotate freely and primarily interacts with the MoS₂ substrate, favoring a stable ($\phi \sim 0^\circ$) alignment with the substrate. This aligned stacking promotes rolling, which, however, needs to overcome a higher energy barrier causing significant friction. With increasing fullerene coverage, lateral interactions between neighboring fullerenes arise, restricting rolling and resulting in a misaligned angle with the substrate. Thus, sliding becomes a more favorable pathway. The frictional property is therefore not only a result of atomic alignment, but is also strongly influenced by fullerene-fullerene interactions that can suppress rolling by collective jamming effects. Counterintuitively, the resulting sliding dissipates less energy than the rolling motion as shown by the ultra-low friction behavior under high coverage in Fig. 1. These results are in agreement with simulations of larger carbon nanoparticles, which similarly observed a friction reduction when transitioning from rolling to slip [43,44]. Furthermore, the higher rolling friction of monolayer C₆₀ was experimentally observed by Coffey and Krim [42] using a quartz crystal microbalance. Our results provide insight into the ultralow friction coefficient observed experimentally for a simple model of a C₆₀/MoS₂ heterojunction [46]. Experiments on C₆₀ nanoparticle mobility on the surface of MoS₂ films could provide further experimental insight into the C₆₀-MoS₂ interaction and establish a more direct connection with our computational model.

4. Conclusions

The molecular-dynamics simulations of monolayer C₆₀ nanoparticles in between MoS₂ slabs unravel the lubrication mechanism of confined carbon nanoparticles: contact formation between the fullerenes and the MoS₂ layers lead to alignment-induced adhesion interactions and to rolling of the particles, while strong applied load leads to transition from rolling to sliding as the fullerenes are strongly deformed. This transition is clearly reflected in the rotational trajectories of the nanoparticles that evidence the onset of rolling at low applied load, and its partial loss at high loads. We ascribe this transition to the normal-force induced deformation of the particles, leading to formation of multi-contact interfaces between the fullerenes and MoS₂ due to spreading from point to area contact.

With increasing coverage of the fullerenes, their deformation is reduced, lowering the amount of rolling, until fullerene-fullerene interactions set in, which suppress rolling. The resulting sliding of the MoS₂ slab along the fullerenes reduces the friction and leads to nearly superlubric behavior. Hence, counterintuitively, a larger coverage of nanoparticles causes friction reduction despite their emerging interactions and blocking of their rotation, due to the transition from rolling to sliding. At the atomic contact level, this is related to a shift of the alignment angle from aligned to misaligned stacking. Hence, the ball bearing effect of C₆₀ particles is not beneficial for friction reduction in the heterojunction system, even though the nanoparticles rotate directionally. In contrast, in the jamming state of the ball-plate heterojunction, an adaptable misalignment of the large-area contact, controlled by the applied load, leads to nearly frictionless sliding. Our results on multi-contact interactions between fullerenes and MoS₂ slabs provide physical insight into the frictional behavior, and offer guidelines for carrying the nanoscale superlubric property over to the macroscopic scale for applications.

Statement of originality

Hereby, we state that the submitted paper is based on original work by the authors, and is not submitted or under consideration at another journal.

CRediT authorship contribution statement

Dongdong Zhou: Writing – original draft, Investigation, Funding acquisition, Conceptualization. **Kedong Bi:** Writing – review & editing, Conceptualization. **Abhishek Gupta:** Writing – review & editing, Validation. **Peter Schall:** Writing – review & editing, Supervision, Investigation, Funding acquisition, Conceptualization. **Astrid de Wijn:** Writing – review & editing, Supervision, Investigation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors would like to thank the China Scholarship Council (CSC) research program for providing funding for D. Zhou. Furthermore, A.K. G, A.S. de W. and P. S. acknowledge funding from the HORIZON-EIC-2021-PATHFINDEROPEN-01 N. 101046693, SSLIP project funded by the European Union. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or European Commission. Neither the European Union nor the granting authority can be held responsible for them.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.triboint.2025.111317](https://doi.org/10.1016/j.triboint.2025.111317).

Data availability

Data will be made available on request.

References

- [1] Novoselov KS, Mishchenko A, Carvalho A, Castro Neto AH. 2D materials and van der Waals heterostructures. *Science* 2016;353(6298):aac9439.
- [2] Geim AK, Grigorieva IV. Van der Waals heterostructures. *Nature* 2013;499(7459):419–25.
- [3] Ding Z, Pei Q-X, Jiang J-W, Huang W, Zhang Y-W. Interfacial thermal conductance in graphene/MoS₂ heterostructures. *Carbon* 2016;96:888–96.
- [4] Lee C, Wei X, Kysar JW, Hone J. Measurement of the elastic properties and intrinsic strength of monolayer graphene. *Science* 2008;321(5887):385–8.
- [5] Koren E, Leven I, Lortscher E, Knoll A, Hod O, Duerig U. Coherent commensurate electronic states at the interface between misoriented graphene layers. *Nat Nanotechnol* 2016;11(9):752–7.
- [6] Novoselov KS, Geim AK, Morozov SV, Jiang D, Zhang Y, Dubonos SV, et al. Electric field effect in atomically thin carbon films. *Science* 2004;306(5696):666–9.
- [7] Britnell L, Ribeiro RM, Eckmann A, Jalil R, Belle BD, Mishchenko A, et al. K S. Strong light-matter interactions in heterostructures of atomically thin films. *Science* 2013;340(6138):1311–4.
- [8] Hirano M, Shinjo K, Kaneko R, Murata Y. Observation of superlubricity by scanning tunneling microscopy. *Phys Rev Lett* 1997;78(8):1448–51.
- [9] Hirano M, Shinjo K. Atomistic locking and friction. *Phys Rev B* 1990;41(17):11837–51.
- [10] Tang H, Wang Y, Ni X, Watanabe K, Taniguchi T, Jarillo-Herrero P, et al. On-chip multi-degree-of-freedom control of two-dimensional materials. *Nature* 2024;632(8027):1038–44.
- [11] Wang W, Shen J, He QC. Microscale superlubricity of graphite under various twist angles. *Phys Rev B* 2019;99(5):054103.
- [12] Song Y, Mandelli D, Hod O, Urbakh M, Ma M, Zheng Q. Robust microscale superlubricity in graphite/hexagonal boron nitride layered heterojunctions. *Nat Mater* 2018;17(10):894–9.
- [13] Dienwiebel M, Verhoeven GS, Pradeep N, Frenken JW, Heimberg JA, Zandbergen HW. Superlubricity of graphite. *Phys Rev Lett* 2004;92(12):126101.
- [14] Liao M, Nicolini P, Du L, Yuan J, Wang S, Yu H, et al. Ultra-low friction and edge-pinning effect in large-lattice-mismatch van der Waals heterostructures. *Nat Mater* 2022;21(1):47–53.
- [15] Pierucci D, Henck H, Avila J, Balan A, Naylor CH, Patriarche G, et al. Band alignment and minigaps in monolayer MoS₂-graphene van der Waals heterostructures. *Nano Lett* 2016;16(7):4054–61.
- [16] Ji Z, Zhao Y, Chen Y, Zhu Z, Wang Y, Liu W, et al. Opto-twistronic Hall effect in a three-dimensional spiral lattice. *Nature* 2024;634(8032):69–73.
- [17] Liu Z, Yang JR, Grey F, Liu JZ, Liu YL, Wang YB, et al. Observation of microscale superlubricity in graphite. *Phys Rev Lett* 2012;108(20):205503.
- [18] Vu CC, Zhang S, Urbakh M, Li Q, He QC, Zheng Q. Observation of normal-force-independent superlubricity in mesoscopic graphite contacts. *Phys Rev B* 2016;94(8):081405.
- [19] Li J, Li J, Chen X, Liu Y, Luo J. Microscale superlubricity at multiple gold-graphite heterointerfaces under ambient conditions. *Carbon* 2020;161:827–33.
- [20] Zhang R, Ning Z, Zhang Y, Zheng Q, Chen Q, Xie H, et al. Superlubricity in centimetres-long double-walled carbon nanotubes under ambient conditions. *Nat Nanotechnol* 2013;8(12):912–6.
- [21] Lee C, Li Q, Kalb W, Liu XZ, Berger H, Carpick RW, et al. Frictional characteristics of atomically thin sheets. *Science* 2010;328(5974):76–80.
- [22] Spear JC, Custer JP, Batteas JD. The influence of nanoscale roughness and substrate chemistry on the frictional properties of single and few layer graphene. *Nanoscale* 2015;7(22):10021–9.
- [23] Fang L, Liu DM, Guo Y, Liao ZM, Luo JB, Wen SZ. Thickness dependent friction on few-layer MoS₂, WS₂, and WSe₂. *Nanotechnology* 2017;28(24):245703.
- [24] Urbakh M, Meyer E. Nanotribology: the renaissance of friction. *Nat Mater* 2010;9(1):8–10.
- [25] Schweizer P, Dolle C, Dasler D, Abellan G, Hauke F, Hirsch A, et al. Mechanical cleaning of graphene using in situ electron microscopy. *Nat Commun* 2020;11(1):1743.
- [26] Li Z, Kozbial A, Nioradze N, Parobek D, Shenoy GJ, Salim M, et al. Water protects graphitic surface from airborne hydrocarbon contamination. *ACS Nano* 2016;10(1):349–59.
- [27] Wang C, Han R, Wang Y, Chen S, Xu H, Huang S, et al. Controlling friction energy dissipation by ultrafast interlayer electron-phonon coupling in WS₂/graphene heterostructures. *Nano Energy* 2024;132:110371.
- [28] Minkin AS, Lebedeva IV, Popov AM, Knizhnik AA. Atomic-scale defects restricting structural superlubricity: Ab initio study on the example of the twisted graphene bilayer. *Phys Rev B* 2021;104(7):075444.
- [29] Chiu HC, Ritz B, Kim S, Tosatti E, Klinker C, Riedo E. Sliding on a nanotube: interplay of friction, deformations and structure. *Adv Mater* 2012;24(21):2879–84.
- [30] Berman D, Deshmukh SA, Sankaranarayanan SK, Erdemir A, Sumant AV. Macroscale superlubricity enabled by graphene nanoscroll formation. *Science* 2015;348(6239):1118–22.
- [31] de Wijn AS, Fasolino A, Filippov AE, Urbakh M. Low friction and rotational dynamics of crystalline flakes in solid lubrication. *Europhys Lett* 2011;95(6):66002.
- [32] van Wijk MM, de Wijn AS, Fasolino A. Collective superlubricity of graphene flakes. *J Phys Condens Matter* 2016;28(13):134007.
- [33] Berman D, Narayanan B, Cherukara MJ, Sankaranarayanan S, Erdemir A, Zinovev A, et al. Operando tribochemical formation of onion-like-carbon leads to macroscale superlubricity. *Nat Commun* 2018;9(1):1164.
- [34] Meirzadeh E, Evans AM, Rezaee M, Milich M, Dionne CJ, Darlington TP, et al. A few-layer covalent network of fullerenes. *Nature* 2023;613(7942):71–6.
- [35] Hou L, Cui X, Guan B, Wang S, Li R, Liu Y, et al. Synthesis of a monolayer fullerene network. *Nature* 2022;606(7914):507–10.
- [36] Ying P, Hod O, Urbakh M. Superlubric graphitefullerene. *Nano Lett* 2024;24(34):10599–604.
- [37] Peng B. Stability and strength of monolayer polymeric C₆₀. *Nano Lett* 2023;23(2):652–8.
- [38] Ribeiro LA, Pereira ML, Giazza WF, Tromer RM, Galvão DS. Thermal stability and fracture patterns of a recently synthesized monolayer fullerene network: A reactive molecular dynamics study. *Chem Phys Lett* 2022;807:140075.
- [39] Jafary-Zadeh M, Reddy CD, Zhang Y-W. Effect of rotational degrees of freedom on molecular mobility. *J Phys Chem C* 2013;117(13):6800–6.
- [40] Savin AV. Friction, mobility, and thermophoresis of carbon nanoparticles on a graphene sheet. *Phys Rev B* 2022;106(20):205410.
- [41] Liang Q, Tsui OK, Xu Y, Li H, Xiao X. Effect of C₆₀ molecular rotation on nanotribology. *Phys Rev Lett* 2003;90(14):146102.
- [42] Coffey T, Krim J. C₆₀ molecular bearings and the phenomenon of nanomapping. *Phys Rev Lett* 2006;96(18):186104.
- [43] Zhao W, Duan F. Friction properties of carbon nanoparticles (nanodiamond and nanoscroll) confined between DLC and a-SiO₂ surfaces. *Tribol Int* 2020;145:106153.
- [44] Ewen JP, Gattinoni C, Thakkar FM, Morgan N, Spikes HA, Dini D. Nonequilibrium molecular dynamics investigation of the reduction in friction and wear by carbon nanoparticles between iron surfaces. *Tribol Lett* 2016;63(3):38.
- [45] Braun OM, Tosatti E. Molecular rolling friction: The cogwheel model. *J Phys Condens Matter* 2008;20(35):354007.
- [46] Nakagawa H, Kibi S, Tagawa M, Umeno M, Ohmae N. Microtribological properties of ultrathin C₆₀ films grown by molecular beam epitaxy. *Wear* 2000;238(1):45–7.
- [47] Stuart SJ, Tutein AB, Harrison JA. A reactive potential for hydrocarbons with intermolecular interactions. *J Chem Phys* 2000;112(14):6472–86.
- [48] Jiang JW, Park HS, Rabczuk T. Molecular dynamics simulations of single-layer molybdenum disulphide (MoS₂): Stillinger-Weber parametrization, mechanical properties, and thermal conductivity. *J Appl Phys* 2013;114(6):064307.
- [49] Le M-Q. Cohesive energy in graphene/MoS₂ heterostructures. *Meccanica* 2016;52(1-2):307–15.
- [50] Silva A, Claerhout VEP, Polcar T, Kramer D, Nicolini P. Exploring the stability of twisted van der Waals heterostructures. *ACS Appl Mater Interfaces* 2020;12(40):45214–21.
- [51] Plimpton S. Fast parallel algorithms for short-range molecular-dynamics. *J Comput Phys* 1995;117(1):1–19.

- [52] Miura K, Kamiya S, Sasaki N. C₆₀ molecular bearings. *Phys Rev Lett* 2003;90(5): 055509.
- [53] Okita S, Matsumuro A, Miura K. Tribological properties of a C₆₀ monolayer film. *Thin Solid Films* 2003;443(1-2):66–70.
- [54] Braun OM. Simple model of microscopic rolling friction. *Phys Rev Lett* 2005;95 (12):126104.
- [55] Seo KJ, Kim DE. Molecular dynamics investigation on the nano-mechanical behaviour of C₆₀ fullerene and its crystallized structure. *Nanoscale* 2020;12(17): 9849–58.
- [56] Xia D, Xue Q, Xie J, Chen H, Lv C, Besenbacher F, et al. Fabrication of carbon nanoscrolls from monolayer graphene. *Small* 2010;6(18):2010–9.
- [57] Liu X, Balla I, Bergeron H, Campbell GP, Bedzyk MJ, Hersam MC. Rotationally commensurate growth of MoS₂ on epitaxial graphene. *ACS Nano* 2016;10(1): 1067–75.