

Structural superlubricity and ultralow friction across the length scales

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Structural superlubricity, a state of ultralow friction and wear between crystalline surfaces, is a fundamental phenomenon in modern tribology that defines a new approach to lubrication. Early measurements involved nanometre-scale contacts between layered materials, but recent experimental advances have extended its applicability to the micrometre scale. This is an important step towards practical utilization of structural superlubricity in future technological applications, such as durable nano- and micro-electromechanical devices, hard drives, mobile frictionless connectors, and mechanical bearings operating under extreme conditions. Here we provide an overview of the field, including its birth and main achievements, the current state of the art and the challenges to fulfilling its potential.

Friction is one of the oldest phenomena examined and used by humankind¹. It has diverse implications in many scientific and technological fields, ranging from physics and chemistry to biology and engineering^{2–5}. In the macroscopic world, friction is an inherent phenomenon in the operation of any mechanical system. Whereas in some cases it is essential for the proper function of the device, friction is often responsible for considerable energy loss and wear⁶. In fact, it has been estimated that about one-third of the energy supplied by fossil fuel in automotive vehicles is consumed in overcoming various forms of frictional dissipation⁷. Friction-induced wear becomes a severe problem when reducing the system size to the nanoscale. Here, owing to the intrinsically high surface-to-volume ratio, even the slightest surface wear may hinder device operation and reduce its durability. One may naively suggest traditional lubrication approaches as a remedy for this problem. However, standard liquid-phase lubricants—for example, organic oils—have been shown to either become highly viscous when confined to nanoscale constrictions⁸ or completely evacuate the junction under external pressure leaving behind a bare frictional interface⁹. As the world strives to miniaturize electronic and mechanical technologies towards ever smaller length scales, new approaches are therefore required to decrease or even eliminate friction and wear at reduced dimensions.

The natural world suggests many alternative strategies for friction reduction. The most common example is lubricated friction, where surface fluid molecules adsorbed at the sliding interface serve as a tribological buffer layer. For instance, polymer brushes and water solvation shells have been suggested to provide the remarkable durability of skeletal joints operating under extreme loads^{4,10–12}. In this respect, the superlubric properties of such brush architectures have been investigated down to the single molecule level¹³. Recently, ionic liquids have emerged as alternative fluid lubricants allowing electro-tunable ultralow friction to be achieved in non-aqueous environments^{15,16}. Solid lubrication constitutes a different approach to the reduction of friction, and relies on the introduction of micrometre-scale or nanoscale particles into the contact region¹⁷. These particles can serve as miniature bearings as well as a source of lubricating flakes via successive layer exfoliation or complete collapse of onion-type structures¹⁷.

The schemes described above follow the standard paradigm that friction reduction requires the introduction of external lubricants into the sliding junction. Notably, ultralow friction can be achieved in the

complete absence of such lubricants. One such scheme, first studied in the early 1970s, involved the use of layered materials, such as graphite, for ultralow friction substrates^{18,19}. This approach enabled the reduction of kinetic friction coefficients down to 5×10^{-3} at nano- and micro-scale contacts under relatively low loads. More recently, amorphous diamond-like carbon appeared as a promising coating for achieving super-low friction and wear at even larger-scale junctions²⁰. Such films are already used in many industrial applications, including razor blades, magnetic hard drives, engine parts, mechanical face seals, scratch-resistant glasses, invasive and implantable medical devices, as well as micro-electromechanical systems²¹. An alternative approach to the reduction of dry friction and wear involves mechanical modulation of the normal and lateral forces applied to the interface, resulting in the elimination of stick-slip motion and hence decrease in energy dissipation^{22–24}.

In this Perspective, we focus on a different, inherent, type of lubricant-free friction reduction scheme that appears in incommensurate crystalline contacts. This mechanism, often termed structural superlubricity²⁵, is one of the most interesting concepts in modern tribology, and holds promise for the achievement of even lower friction coefficients²⁶. It relies on the lattice misfit between two flat and rigid crystalline surfaces leading to effective cancellation of the lateral forces during sliding motion²⁷. The major advantage of structural superlubricity is the ability to circumvent the need for external additives or mechanical manipulation by using chemically clean and stiff surfaces that preserve their crystal lattice structure under shear stress, thus maintaining their incommensurate configuration. This provides intrinsic lubrication that can be adjusted, by the nature of the contacting materials and the junction geometry, to be robust even under extreme conditions, such as high pressures, high and low temperatures, as well as vacuum.

The realization and characterization of structural superlubricity at microscale contacts recently became feasible with advances in the fabrication of large-scale pristine single-crystal layered materials and the development of supersensitive manipulation devices. This constituted an increase of scale of three orders of magnitudes with respect to nanoscale contact experiments performed only a decade ago. In combination with recent advances in the computational modelling of such junctions, this marks out the field of structural superlubricity research as timely and exciting with great promise for realistic technological applications. Here we provide a general overview of the field, starting from its inception,

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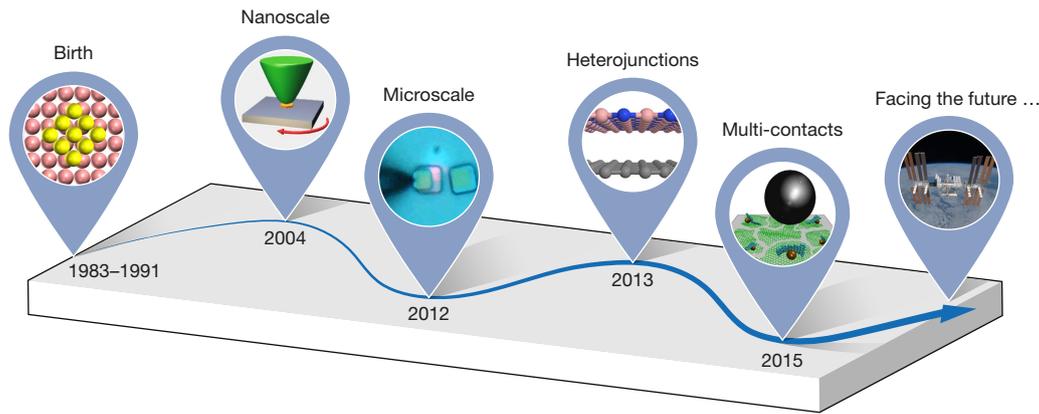


Fig. 1 | Timeline of major milestones in structural superlubricity research. The timeline starts with the first theoretical prediction of vanishing static friction, made in 1983²⁸, and the computational study of ultralow kinetic friction states in 1991²⁹ ('Birth'). This is followed by the pioneering experimental demonstration of nanoscale superlubricity in graphitic contacts in 2004⁴¹, which led to the first observations of microscale superlubricity in 2012⁴² and to the suggestion of heterojunctions⁸⁶ in 2013 and of multi-contact configurations⁷ in 2015 (multi-contacts schematic image adapted with permission from ref. 7,

American Association for the Advancement of Science) as possible routes to achieve robust superlubricity at large length scales. This path taken by the scientific community in recent years opens the door to the scaling-up of structural superlubricity towards the macroscale, with substantial technological implications and applications, such as solid lubricants for satellite solar panel motors operating under the extreme conditions encountered in space ('Facing the future ...'; satellite image adapted from https://www.nasa.gov/multimedia/imagegallery/image_feature_1314.html, NASA).

discuss the main achievements and the state of the art, and foresee the challenges that are yet to be overcome towards achieving this goal. We aim to turn the attention of the scientific community to this phenomenon and to trigger new fundamental and applied research for its scaling-up to the macroscale.

The birth of structural superlubricity

The first theoretical prediction of such a state of vanishing static friction in crystalline interfaces was given by Peyrard and Aubry²⁸ for infinite incommensurate contacts in 1983 (see Fig. 1). The term superlubricity was coined by Hirano and co-workers²⁹ almost a decade later (see Fig. 1), referring to the suppression of stick-slip motion via the elimination of a particular energy dissipation channel related to elastic instabilities. Such stick-slip dynamics, commonly associated with the squeaky sound of opening unoled doors (widely used in horror movies), is a major source of energy dissipation, hence its suppression results in considerable reduction of dynamic friction. Nevertheless, in practice, there always exist alternative energy dissipation routes (for instance, the excitation of lattice vibrations induced by variations of long-range interactions) and wear mechanisms resulting in residual friction even in the absence of stick-slip motion. Therefore, unlike other critical phenomena, such as superconductivity and superfluidity, frictional energy dissipation never truly vanishes. In light of this, the criterion for the onset of superlubricity is commonly chosen as the reduction of the friction coefficient (the derivative of the friction force with respect to the normal load) to below 10^{-3} – 10^{-4} .

Insight into the phenomenon of structural superlubricity can be gained by considering the interactions between two surfaces made from plastic foam, each bearing an 'egg-box' pattern of peaks and troughs (see Fig. 2a, b). When the corrugated surfaces of the two foams are put in registry, one can hardly induce lateral sliding because many high barriers have to be crossed simultaneously over the entire interface. Nevertheless, when one foam is slightly laterally rotated with respect to the other, the lattices are taken out of registry. In this case, when one surface slides upon the other some of its peaks are forced to climb uphill while others go downhill. For sufficiently large interfaces these local opposite motions result in effective cancellation of the global friction force. A similar mechanism holds true for micro- and nanoscale interfaces, with the corrugated foam surfaces being replaced by the potential energy landscape of the inter-surface interactions (see Fig. 2c).

Naturally, realistic material interfaces are more complicated than implied by the rigid egg-box foam model. Specifically, the elasticity

of contacting materials may affect superlubric behaviour. Such effects are already appearing in single-particle phenomenological treatments such as the Prandtl–Tomlinson model, where a point mass, dragged by an external support via an elastic spring of stiffness k , slides atop a periodic sinusoidal potential of periodicity a_0 and amplitude V_0 , representing the underlying surface^{30,31}. Here, a transition from stick-slip motion to smooth sliding occurs when the dimensionless parameter $\eta = 4\pi V_0/(ka_0^2)$ exceeds the critical value $\eta = 1$. At this point the mechanical instability resulting from the competition between the driving spring force and the opposing frictional force, exerted by the potential energy landscape, is eliminated. A more realistic description of contacting surfaces requires extension to a multi-particle treatment, such as the Frenkel–Kontorova model³². This introduces intra-surface elasticity that allows the slider atoms to accommodate to the underlying potential, as directly demonstrated by a recent experiment using cold atom chains residing on an optical lattice³³. As a result, above a critical contact size that depends on the ratio between the intra-surface elasticity and interfacial stiffness, locally commensurate regions may form, leading to pinning effects and enhancement of friction^{25,34–36}. Notably, this theoretical prediction was recently verified experimentally³⁷ for antimony particles sliding atop MoS₂. We note that such pinning effects are not limited to the context of tribology but are of rather general nature and well known to the superconductivity community^{25,38}. An important advantage of layered materials is their extremely stiff intra-layer structure and low inter-layer potential energy surface corrugation that may shift the critical length to macroscopic scales.

Experimental evidence of structural superlubricity was reported as early as in 1993, for homogeneous MoS₂ interfaces³⁹. This was further supported by experiments on nanoscale heterogeneous MoS₂/MoO₃ junctions, which exhibited the anisotropic friction characteristic of these systems⁴⁰. A decade later (see Fig. 1), the first detailed experimental exploration of the mechanisms of structural superlubricity in nanoscale graphitic contacts was undertaken, demonstrating controllable and reproducible superlubric motion⁴¹. This triggered extensive experimental investigations that resulted in promising realizations of superlubricity in microscopic graphitic contacts as well as in centimetre-long carbon nanotubes, as detailed below^{42–45}. These impressive recent advances constitute important milestones towards the achievement of macroscale superlubricity, which holds great technological promise for the reduction of friction and wear in actual mechanical devices. Nevertheless, with increasing contact size, factors such as in-plane elasticity and out-of-plane corrugation as well as surface defects and

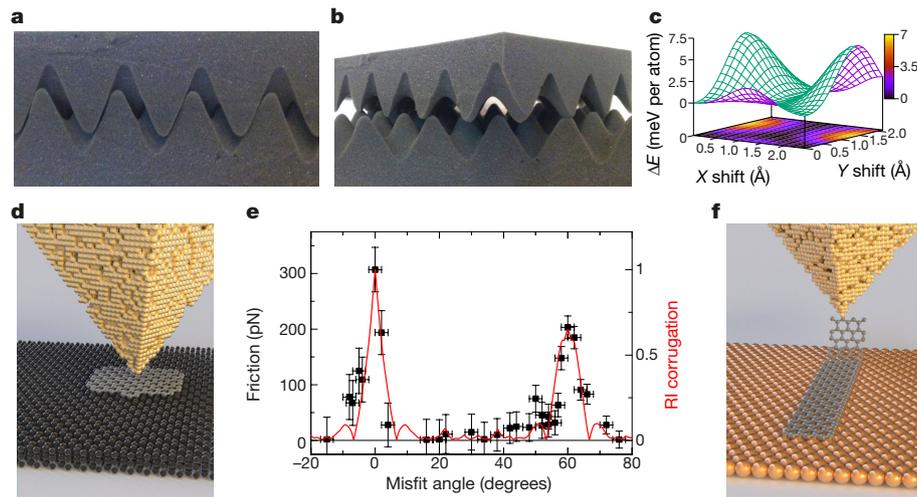


Fig. 2 | Nanoscale structural superlubricity. **a, b**, Egg-box foam models of commensurate **(a)** and incommensurate **(b)** lattices illustrating the origin of finite and vanishing interfacial friction states, respectively (both images reproduced with permission from ref. ⁴⁷, American Physical Society). Each individual foam surface represents the potential energy landscape experienced by a single atom sliding atop the atomic lattice of a rigid two-dimensional layered surface. **c**, The resulting inter-layer sliding energy landscape of a graphene bilayer, obtained by a dedicated inter-layer potential calibrated against advanced density functional theory calculations (image adapted with permission from ref. ⁹⁹, American

Chemical Society). **d, e**, The first experimental demonstration of such a transition from finite to vanishingly small friction was obtained for graphitic flakes sliding atop graphite **(d)**, where friction was shown to reduce by orders of magnitude upon relative rotation of the originally aligned contacting surfaces⁴¹ (**e**; image adapted with permission from ref. ⁴⁷, American Physical Society). Here, the symbols display the experimental results and the full red line represents results of a purely geometric model. **f**, Recently, unidirectional sliding under superlubric conditions was observed for graphene nanoribbons dragged on gold surfaces¹⁴.

chemical contamination may hinder superlubricity by introducing new energy dissipation channels and mechanical wear. Therefore, further scaling-up efforts involve several challenges that are yet to be overcome before the practical application of superlubricity can be realized. To this end, one first has to gain comprehensive understanding of the tribological processes occurring at nanoscale contacts.

Nanoscale superlubricity

As mentioned above, the theoretical predictions of Peyrard and Aubry²⁸ and Shinjo and Hirano²⁷ (later revisited by Consoli et al.⁴⁶), and the pioneering experiments of Dienwiebel et al.⁴¹, set the foundations for our present perception of nanoscale structural superlubricity. Specifically, in ref. ⁴¹, the sliding friction of nanoscale graphite flakes dragged across a pristine graphitic surface was measured as a function of the flake/substrate relative angular orientation (see Fig. 2d, e). The measured friction was found to be vanishingly small (below experimental error) throughout the range of misfit angles studied except for narrow regions, of 60° periodicity, that exhibited stick–slip motion with substantially increased friction. This friction enhancement was associated with an aligned contact, in which the lattice vectors of the contacting hexagonal graphene layers forming the interface become parallel. Notably, the width of these friction peaks was found to reduce with increasing contact size, mainly due to better cancellation of the lateral forces that act on the flake atoms resulting in averaging-out of the net friction force^{47–49}.

The directional character of superlubricity in graphitic contacts was further demonstrated in free sliding experiments, in which nanoscale graphene flakes were pushed out of their optimal commensurate stacking configuration and allowed to slide above a graphene surface⁵⁰. In their superlubric state, the flakes were found to slide freely up to 100 nm at a temperature of 5 K, and were stopped only by their realignment with the underlying substrate. Owing to the enhancement of the thermally induced reorientation processes, the free-sliding length decreased with increasing temperature.

Extending the scope of super-low friction studies beyond rigid layered material interfaces, the motion of graphene nanoribbons across Au(111) surfaces has been recently studied (see Fig. 2f)¹⁴. In this case, extremely small frictional forces, of the order of piconewtons, were

measured for graphene nanoribbons pulled along the $[-1, 0, 1]$ direction of the underlying gold surface. The forces were found to be nearly independent of the length of the nanoribbons, indicating superlubric motion of the incommensurate contact with residual friction forces originating from edge effects.

It was further demonstrated that superlubricity is not limited to fully crystalline contacts, but can also be realized in incommensurate junctions consisting of crystalline and disordered materials. In particular, the friction between a graphite surface and amorphous antimony or crystalline gold clusters was measured⁵¹. Within the context of the scale-up of superlubricity, the main focus of these studies was devoted to investigating the contact size dependence of the kinetic friction force. The traditional tribological view suggests a classical linear dependence, as often observed in the macroscopic world. This, indeed, is the case for commensurate nanoscale friction contacts²⁵. Remarkably, for clean contacts between the amorphous clusters and graphite the friction force was found to scale with the square root of the contact area. This scaling was rationalized by the fact that the lateral atomic forces in disordered surfaces average-out, following the central limit theorem⁵². In misaligned crystalline contacts between gold and graphite, where cancellation of the lateral forces results from the lattice incommensurability, even lower scaling exponents of the friction force with flake size were measured. It was further argued that in this case the exponent value is not unique but rather depends on the contact shape and scan-line direction^{51,53,54}. Interestingly, recent computational studies predicted that under certain conditions the frictional behaviour of incommensurate layered material contacts can become completely independent of the contact size^{53,55,56}. This finding suggests the intriguing possibility of scaling-up structural superlubricity towards the micro- and even macroscales.

Notably, similar effects of friction reduction due to inter-lattice commensuration effects have also been observed for interfaces of soft materials such as colloidal suspensions⁵⁷. The microscale dimensions of the individual colloids used in such set-ups allows for direct observation and investigation of the mechanisms underlying the transition from stick–slip to superlubric sliding. Furthermore, they facilitate explicit control over inter-particle and particle/substrate interaction parameters, thus enabling various frictional regimes to be explored. Apart

from their fundamental importance, such studies, combined with large-scale realistic simulations of appropriate model systems⁵⁸, shed light on important factors that govern friction and wear in crystalline contacts.

These striking examples of the strong interplay between experiment and theory are characteristic of the field of nanoscience in general and of nanotribology in particular. At such reduced length scales, theory and computation can provide highly reliable description of the physical processes underlying the measured phenomena. Hence, they can help in both the analysis and the rationalization of experimental observations and in the prediction of novel material behaviours. To this end, theory offers a spectrum of approaches, ranging from coarse-grained descriptions relying on geometric considerations to fully atomistic elaborate simulations. These allow deep understanding of nanoscale tribological effects to be gained and rational deductions about micro- and macroscale contacts to be made.

As discussed above, when considering wearless friction in rigid crystalline interfaces, the friction is found to be strongly related to the inter-lattice commensurability^{39–41,59}. In such cases, substantial insights can be gained from simple geometric descriptions of the contact that quantify the degree of lattice registry^{60,61}. As an example, in Fig. 2e we show that accounting for geometric considerations by using the registry index approach (red line) can capture the variation of the measured sliding friction with misfit angle (black points) in graphitic contacts⁶⁰. However, such descriptions neglect important effects such as in- and out-of-plane elasticity of the substrate and slider, dynamic reorientations, thermal fluctuations, energy dissipation processes, effects of chemical contact contamination, and possible wear. In principle, all these effects can be captured by ab initio molecular dynamics simulations, but these are prohibitively computationally expensive even for nanoscale contact models. Therefore, one often resorts to fully atomistic classical molecular mechanics simulations that rely on dedicated force fields, specifically parameterized to capture the corresponding intra- and inter-layer interactions^{62–68}. Even this approach is restricted by the simulated timescales limiting the calculation to interfacial shear velocities that are orders of magnitude higher than in typical friction force experiments. Here, semi-phenomenological approaches that focus on reduced dynamics of few important degrees of freedom may help bridge the gap between the timescales of experiments and those of simulations⁶⁹. In particular, such approaches have shown that friction often exhibits a logarithmic dependence on velocity, thus justifying the validity of fully atomistic simulations in the study of tribological processes in nanoscale junctions.

One of the most important contributions of computational simulations to the field of nanotribology is the ability to identify mechanisms that eliminate superlubricity and suggest ways to overcome them. In this respect, an important extrinsic factor that may suppress superlubricity was found to be the incorporation of contaminants, such as chemical adsorbates and various nanoparticles, within the frictional junction. These often lead to pinning of incommensurate surfaces, resulting in the appearance of static friction and enhancement of kinetic friction^{51,70–72}. Surface heating may lead to contaminant desorption, thus recovering the bare surfaces and reducing the adsorbate-related friction^{51,73–75}. Such heating would be most effective as a pre-treatment applied to the exposed surfaces before the formation of the junction. An alternative in situ approach was further suggested, in which mechanical oscillations of the frictional contact lead to substantial decontamination of the interface, thus restoring its super-low friction characteristics^{76,77}.

The normal load experienced by the frictional junction constitutes another extrinsic factor limiting superlubricity. Obviously, above a certain (system-dependent) normal load, any contact should exhibit increased friction leading to enhanced wear. It is therefore desirable to identify conditions under which superlubricity can be sustained with practically applied loads. Computational studies have revealed that for incommensurate contacts the edge atoms of a finite sliding flake are most prone to the effects of an increased normal load. This, in turn, may lead to enhanced friction via their pinning to the underlying surface⁷⁸. The importance of such effects, however, was shown to reduce with increasing contact size^{56,75}.

Furthermore, most experiments demonstrating superlubricity to date have been performed at relatively low driving velocities, of the order of micrometres per second, whereas practical applications, such as mechanical hard-disk drive read/write components (see below), often operate at considerably higher velocities of tens of metres per second. This, in turn, may enhance energy dissipation and wear effects, resulting in the elimination of superlubricity. Microscale understanding of these processes may be gained by molecular dynamics simulations that are most suitable for describing frictional behaviour at such high velocities.

Frictional junctions also possess intrinsic properties that may eliminate superlubricity. For example, considering the results of Dienwiebel et al.⁴¹, one may conclude that in order to achieve superlubricity in practical applications it is sufficient to bring two rigid crystalline surfaces into incommensurate contact. Nevertheless, both experiments and simulations have shown that dynamic reorientations of the sliding surfaces tend to lock the system into its commensurate high-friction configuration, thus limiting the realization of superlubricity to short timescales⁴⁹. Furthermore, as discussed above, intrinsic layer elasticity has been theoretically predicted to increase friction in incommensurate contacts. Here the typical out-of-plane stiffness of the individual layers is comparable to the effective stiffness of the inter-layer shear force hence providing accessible energy dissipation channels that may enhance friction already in nanoscale contacts^{71,72,79–82}. Furthermore, while the corresponding in-plane stiffness is considerably higher, its effects are expected to already be manifested in microscale contacts, where elastic deformations may become sufficiently large⁸³ that commensurate regions can develop, leading to an increase in friction^{35,36,84}.

To address these issues, nanoscale heterogeneous junctions formed between graphite and hexagonal boron nitride⁸⁵ have been suggested theoretically to provide an accessible route towards robust superlubricity (see Fig. 1)^{56,86,87}. It was shown that, above a certain contact size, the corrugation of their sliding potential energy surface is considerably reduced even for aligned contacts. This effect, associated with the appearance of moiré patterns resulting from the intrinsic inter-layer lattice constant mismatch in heterojunctions, suggests that superlubricity should be preserved even under interfacial reorientations^{86,87}. It was further shown that superlubricity in heterogeneous interfaces of graphene and hexagonal boron nitride can sustain considerably higher loads than in their homogeneous graphitic counterparts⁵⁶. This was attributed to the intrinsic incommensurability that reduces the effects of edge atom pinning under high normal loads. Recent experimental evidence showing that heterogeneous microscale graphite and hexagonal boron nitride contacts exhibit superlubricity that is nearly orientationally independent supports that prediction⁸⁸. Interestingly, a related experiment⁴⁵ demonstrated similar behaviour for both heterogeneous graphene/hexagonal boron nitride and homogeneous graphitic contacts. Here, the polycrystalline nature of one of the contacting surfaces further prohibited the formation of fully commensurate contacts⁸⁹. Additional support was also provided by experimental observations of self-orientation of microscale hexagonal boron nitride flakes on graphitic surfaces⁹⁰.

Micrometre- and centimetre-scale superlubricity

At the forefront of the field of superlubricity stands the effort to demonstrate the robustness of the effect against intrinsic elimination mechanisms and external perturbations at ever-growing contact dimensions. Recently, two decades after the first experimental demonstration of nanoscale structural superlubricity, evidence of frictionless sliding in micrometre- and centimetre-sized crystalline contacts has been reported^{142–45,88}.

The first experimental observation of microscale structural superlubricity was reported in 2012⁴² (see Fig. 1). Clean single-crystalline interfaces between two graphitic stacks were formed via shear-force-induced mechanical exfoliation of a multilayer graphitic mesa (see Fig. 3a)⁴². The shear force was applied by an external tip to the mesa top causing a stacking fault at the weakest interface that divides it into

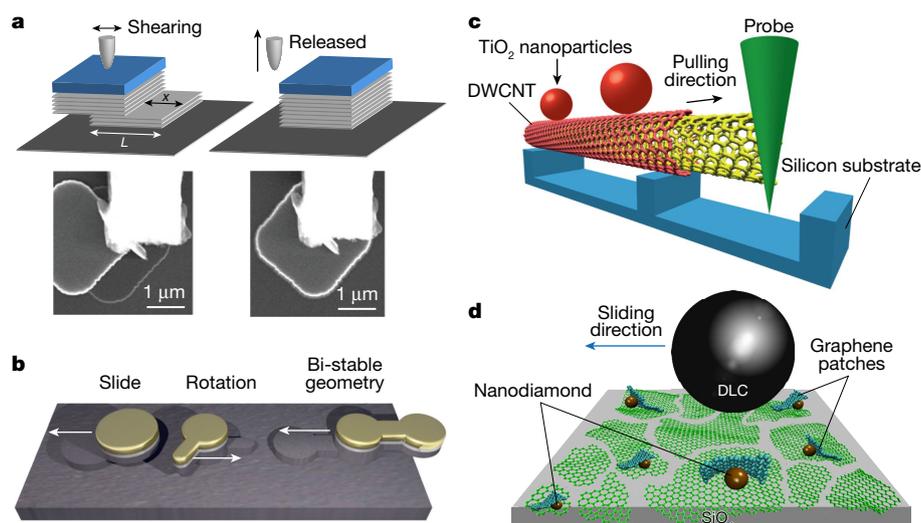


Fig. 3 | Microscale superlubricity. **a**, Self-retraction motion of a microscale square graphitic mesa. An initially sheared mesa (illustrated and imaged in the upper and lower left subpanels, respectively) self-retracts to its original position (illustrated and imaged in the upper and lower right subpanels, respectively) (image reprinted with permission from ref. ⁴², American Physical Society). **b**, Shear force measurement in mesoscopic graphite contacts (image reprinted with permission from

ref. ⁴⁴, American Association for the Advancement of Science). **c**, Inter-wall telescopic superlubric motion in centimetre-long double-walled carbon nanotubes (DWCNT; image reprinted with permission from ref. ¹⁰⁰, Springer Nature). **d**, Multi-contact superlubricity in microscopic interfaces between diamond-like carbon (DLC) and graphene scrolls wrapped around diamond nanoparticles (image adapted with permission from ref. ⁷, American Association for the Advancement of Science).

two weakly interacting graphite stacks. The upper stack could then be repeatedly sheared against the lower one in different directions and relative angular orientations. Notably, upon release, most sheared stacks returned to their original position with no external aid, exhibiting self-retraction due to an adhering restoring force that drives the system towards minimum interfacial energy⁹¹. For specific orientations, of six-fold rotational symmetry, a lock-in effect was demonstrated with no evident self-retraction. This clearly indicates that the microscale interface is constructed from single-crystalline graphene layers that exhibit superlubric self-retraction motion when placed at incommensurate configurations. Further support for this conclusion was recently provided⁷⁵, when quantitative measurements of the tribological properties of this system demonstrated dynamic friction coefficients well within the superlubric regime for the misaligned contact up to external normal loads of 1.67 MPa. Importantly, the superlubric behaviour was found to sustain not only vacuum conditions but also ambient conditions at various humidities⁴² and high sliding velocities up to⁷³ 25 m s⁻¹. Additional experimental evidence of superlubricity in microscale graphitic contacts was recently provided when measurements of the dynamic friction force as a function of contact size yielded power-law scaling with a typical exponent of 0.35, lower than the value of 0.5 characteristic of amorphous contact, thus indicating the formation of an incommensurate crystalline contact (see Fig. 3b)⁴⁴. However, a major drawback of such microscale homogeneous graphitic junctions is that friction increases dramatically when the contacting surfaces are aligned. As mentioned above, recent theoretical predictions⁸⁶ and experiments⁸⁸ on graphene/hexagonal boron nitride contacts demonstrated that heterojunctions in layered materials may offer a remedy for this problem.

One of the most recent advances in the field of superlubricity involved the extension of the scope of superlubricity to the centimetre regime⁴³. This became possible by taking advantage of the intrinsic inter-wall incommensurability in pristine bichiral double-walled carbon nanotubes. An inner-shell pull-out experiment was used to measure the inter-wall friction in coaxial centimetre-long double-walled carbon nanotubes, yielding friction forces as low as 1 nN independent of the axial shift extension (see Fig. 3c). Such systems hold the technological potential to serve as low-energy dissipative gigahertz oscillators^{92,93}. We note that the actual contact area in this experiment was comparable to that of the self-retracting graphitic mesas discussed

above. Nevertheless, centimetre-scale single-crystalline graphene surfaces are already achievable today and hold great potential for large-scale tribological applications (see Fig. 4)⁹⁴. Furthermore, the fact that one of the system dimensions extends to the centimetre scale provides evidence that using judicious geometrical designs can help suppress intrinsic elimination mechanisms to allow superlubricity at large length scales.

Another promising route to obtain large-scale superlubricity involves multi-contact interfacial geometries (see Fig. 1). In this respect, it was recently demonstrated that ultralow friction coefficients can be achieved in mesoscale contacts formed between a diamond-like carbon sphere and a silicon dioxide surface covered by graphene patches and diamond nanoparticles (see Fig. 3d)⁷. Under shear, the graphene patches tend to scroll around the diamond nanoparticles forming an incommensurate contact with the surface of the diamond-like carbon sphere. As mentioned above, another realization of the multi-contact approach to microscale superlubricity was recently provided, in which the friction between a graphene coated microsphere and a silicon dioxide surface covered by graphene or hexagonal boron nitride was shown to be considerably lower than between the bare surfaces⁴⁵. The underlying mechanism relies on the roughness of the sphere surface that incorporates elevated asperities covered by randomly oriented graphene patches. These form multiple nanoscale contacts of different registry with the substrate, prohibiting the formation of a fully commensurate contact. Importantly, these examples demonstrated that the multi-contact approach, involving the cumulative effect of many nanoscale contacts that are randomly distributed on the surface, is less susceptible to effects of external loads and humidity.

These recent experimental demonstrations of superlubricity in microscale contacts constitute an important milestone on the way to achieving superlubricity in macroscale contacts, which is essential for practical applications (see Fig. 4).

Challenges and future directions

Superlubricity holds great technological promise for reducing energy loss and friction-induced wear in actual mechanical devices. However, fulfilling this potential requires the scaling-up of superlubricity to macroscopic contacts. To meet this challenge several issues have to be addressed, as follows.

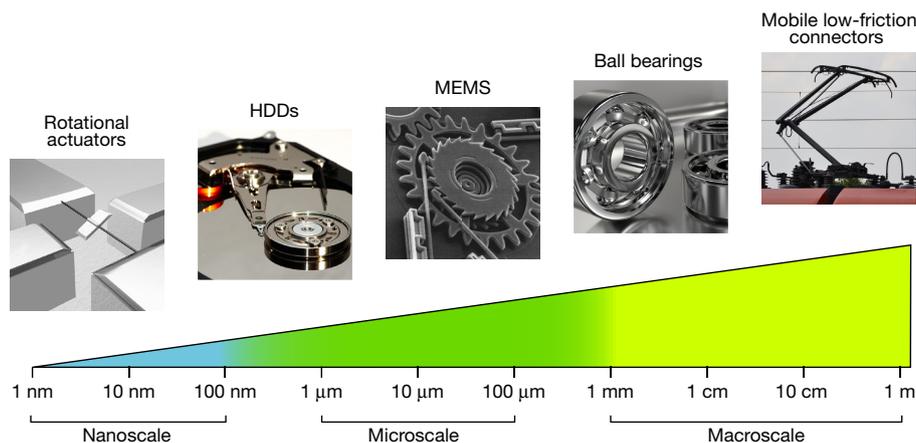


Fig. 4 | Demonstrative applications of structural superlubricity at different length scales. The bar is labelled underneath with the approximate positions of the length scales. Above the bar are images representing the applications, as follows (left to right): nanotube-based frictionless rotational actuators (image reproduced with permission from ref. ¹⁰¹, Springer Nature); wear-free nanoscale read/write contacts in hard-disk drives (HDDs; image reproduced with permission from Magnus

Hagdorn); durable micro-electro-mechanical systems (MEMS; image reproduced with permission from ref. ¹⁰², Annual Reviews); low-friction ball bearings; and efficient mobile low-friction connectors (image adapted from https://commons.wikimedia.org/wiki/File:Pantograph_of_a_DBAG_Class_423.JPG, published under a CC BY 3.0 DE licence (<https://creativecommons.org/licenses/by/3.0/de/deed.en>)).

Edge effects

Because the circumference-to-surface ratio of planar contacts decreases as the inverse contact dimension one could naively expect that edge effects would reduce with the interface size. However, considering the fact that the central contact area exhibits superlubric behaviour, edge pinning effects are often responsible for the residual friction and hence may become dominant with increasing contact size.

Surface roughness

In any practical application, layered materials will be deposited on substrates that will exhibit roughness at various length scales. When the surface corrugation becomes substantially larger than the typical length scales of the two-dimensional layered material crystal coat, friction may be dictated by the overall surface roughness rendering commensurability effects unimportant.

Surface defects and contaminants

Although pristine interfaces can be readily fabricated at the nanoscale, surface defects and contaminants are expected to appear with increasing contact size. Specifically, inter-layer covalent bonding and bulky molecular adsorbates that are present under ambient conditions may cause interfacial chemical pinning, which can considerably increase friction. Since the corresponding unbinding energy scales are typically very large even at low densities, such imperfections are expected to induce a considerable effect on the measured friction.

Elasticity

When incommensurate surfaces are put into contact, elasticity effects can support inter-lattice readjustment that leads to the formation of locally commensurate regions⁹⁵ characterized by strong lock-in forces. This effect grows with the contact size and hence is expected to enhance friction at macroscale junctions²⁵.

External conditions

Some of the less-explored factors in the field of superlubricity are the effects of external normal load, sliding velocity and distance, as well as temperature. Most experiments to date have considered normal loads and velocities that are substantially lower than those typically experienced in macroscale systems. It is expected that high loads and high sliding velocities will lead to enhanced wear of the two-dimensional material surface coating, which will reduce the interface durability⁹⁶. Furthermore, most measurements aim at understanding basic tribological phenomena and are thus performed at ambient temperature

and with very short sliding distances. In this respect, it should be noted that while under superlubric conditions the residual wear is expected to be low, it will not completely vanish as the friction coefficients remain finite. Hence, for practical applications experiments on superlubricity should consider large sliding distances as well as extreme temperatures and extreme normal loads.

On the basis of the substantial experimental and theoretical advances achieved over the past decade in the field of superlubricity, three promising routes can be suggested to overcome these challenges. First, the idea of obtaining robust structural superlubricity via the use of heterogeneous contacts of rigid layered materials can be extended to the macroscale. Here the intrinsic lattice misfit between the contacting surfaces leads to incommensurate positioning of the edge atoms that inhibits the formation of coherent edge pinning effects, thus considerably reducing the residual edge friction⁵⁶. In fact, structural superlubricity is not limited to the realm of layered materials and can be achieved under more general conditions. What is essential for the realization of the discussed mechanism is the formation of a contaminant-free interface between atomically smooth stiff surfaces that are out of registry. Moreover, the notion of disorder-induced incommensurability in heterojunctions formed between a crystal surface and an amorphous counterpart can also be extended to the macroscale. When the contacting surfaces are rigid—for example, diamond-like carbon—ultralow friction and wear can be achieved^{21,97}.

Second, multi-contact configurations can serve as a venue for resolving some of the above-mentioned problems, taking advantage of surface roughness and/or polycrystallinity to effectively transform macroscale junctions into a large collection of nanoscale contacts^{45,89}. Under appropriate conditions, where the various contacts are coated by randomly oriented patches of two-dimensional material, robust superlubricity is expected to prevail even under high external loads and high sliding velocities. Furthermore, multi-asperity configurations effectively decouple the individual nanoscale contacts, thus diminishing undesirable elasticity effects. Nevertheless, within this approach the effective contact edge-to-surface ratio considerably increases and that, in turn, may result in undesirable edge pinning effects and additional friction.

Third, a way to reduce in-plane elasticity effects would involve the deposition of two-dimensional material coatings on rigid surfaces and/or the usage of multi-layer stacks. The supporting bulk reduces the tendency of the contacting layers at the frictional interface to adjust their lattices, therefore diminishing the formation of locally commensurate regions and supporting superlubricity. Further reduction of inter-lattice adjustment effects may be achieved via extension or compression

stresses applied to the interfacing layers. Notably, in contrast to such in-plane elasticity effects, interfacial out-of-plane deformations may promote the occurrence of superlubricity via soliton-like smooth sliding of elevated moiré ridges that can extend up to the macroscale⁵⁶. Multi-layer systems may also increase interface rigidity and reduce roughness effects to some extent by eliminating surface rippling and decoupling the frictional interface from the underlying corrugated surface⁷¹. We note that the external normal loads that are often considered to enhance friction due to increased steric repulsions may also suppress surface rippling if applied uniformly to extended contacts. Therefore, one may expect to find regimes of negative friction coefficients, where the friction forces reduce with the external load due to flattening effects⁹⁸. Furthermore, similarly to the microscopic contact case^{76,77}, the use of external loads to induce lateral mechanical oscillations can serve as an interface pre-treatment procedure to dynamically eliminate surface contaminants.

In view of all of the above, we believe that to meet the challenge of achieving superlubricity in macroscopic contacts under realistic operation conditions one should adopt a synergistic strategy combining the advantages of the various approaches discussed herein. Of particular potential would be the use of multi-contact polycrystalline heterojunctions of clean multi-layer stacks. The combination of polycrystallinity and heterogeneous coatings is required to prohibit the formation of commensurate regions and reduce edge pinning effects over the entire interface. Multi-asperity configurations can harness the advantages of nanoscale contacts even at the macroscale and when underlying multi-layer stacks they can further eliminate undesirable elasticity effects. Finally, keeping such interfaces clean to avoid pinning effects should lead to robust macroscale superlubricity. Identifying material junctions that can satisfy these conditions will be a technological breakthrough that will revolutionize many engineering and industrial concepts and shape new paradigms in friction-induced energy loss and wear. The space, automotive and electronics industries, as well as medical manufacturers and information storage centres, among many others, may all benefit greatly from this forthcoming technology (see Fig. 4). In an age when natural resources are becoming limited and the environmental impact of their usage is affecting Earth's atmosphere and ecosystem, macroscale superlubricity may contribute to the reduction of both global energy consumption and pollutant emission.

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Additional information

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