The Smallest Archimedean Screw: Facet Dynamics and Friction in Multi-Walled Nanotubes

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Abstract

We identify a new material phenomenon, where minute mechanical manipulations induce pronounced global structural reconfigurations in faceted multi-walled nanotubes. This behavior has strong implications on the tribological properties of these systems and may be the key to understand the enhanced inter-wall friction recently measured for boron-nitride nanotubes with respect to their carbon counterparts. Notably, the fast rotation of helical facets in these systems upon coaxial sliding may serve as a nanoscale Archimedean screw for directional transport of physisorbed molecules.

Keywords

nanotube, friction, faceting

Introduction

Nanotubes 1–5 form a paradigmatic family of quasi-one-dimensional materials playing a central role in the design of many nano-electromechanical systems. 6–19 Traditionally, they are perceived as miniature cylinders of nanoscale circular cross-section. Nevertheless, if the chirality of neighboring shells within a concentric multi-walled nanotube is correlated, extended circumferential facets may form. 20–26 The resulting polygonal cross section induces geometric inter-wall locking that can considerably enhance their mechanical rigidity. 15

Despite their remarkable structural similarity, faceting is more commonly observed in multi-walled boron-nitride nanotubes (MWBNNTs) 15,17,25,26 than in their carbon counterparts (MWCNTs). 21–24 This can be attributed to three important factors: (i) Stronger long-range dispersive attractive interactions exhibited by the former 27–29 that provide higher inter-wall adhesion thus favoring facet formation; (ii) Softer ZA modes of h-BN 30 that al-
low for sharper vertices thus promoting the formation of wider planar facet regions; and (iii) Higher inter-wall chiral angle correlation exhibited by MWBNNTs over MWCNTs that induces extended lattice registry patterns between adjacent tube shells and dictates the nature of the facets. The latter is mainly due to the additional inter-wall electrostatic interactions between the partially charged ionic centers in the hetero-nuclear BNNT network. While being relatively weak locally, when summed over extended commensurate facet regions these Coulomb interactions can foster energetic stabilization.

Similar to macroscopic objects, the strain developing within the hexagonal lattice of nanotubes under small external mechanical manipulations is proportional to the applied stress. Due to their exceptionally high rigidity this usually leads to minor structural deformations. In the present study, however, we discover a new material phenomenon, occurring in faceted double-walled nanotubes (DWNTs), where minute mechanical manipulations induce pronounced global superstructure reconfiguration. For monochiral DWNTs that exhibit axially aligned facets even the slightest inter-wall rotation induces significant circumferential facet revolution, and minor inter-wall telescoping can lead to complete unfaceting. Similar manipulations applied to bichiral DWNTs result in global screw-like motion of their elongated helical facets reminiscent of an Archimedean screw. Importantly, these superstructure evolutions under coaxial sliding open new collective energy dissipation channels that enhance inter-wall dynamic friction. This, in turn, suggests that the relative abundance of faceting in MWBNNTs plays a central role not only in their enhanced torsional stiffness but also in the significantly higher inter-wall friction that they exhibit with respect to MWCNTs.

Facet Superstructure Reconfiguration

To demonstrate the phenomenon of superstructure reconfiguration we consider a set of four representative double-walled BNNTs (DWBNNTs) including the achiral armchair (104,104)@109,109) and zigzag (180,0)@188,0) DWBNNTs that present axial facets; the bichiral (70,70)@77,74) system, whose small inter-wall chiral angle difference of $\Delta \Theta = 0.65^\circ$ induces helical facets; and the achiral mixed (179,0)@108,108) DWBNNT ($\Delta \Theta = 30^\circ$) that does not form circumferential facets. Here, the notation $(n_1, m_1)@(n_2, m_2)$ represents a $(n_1, m_1)$ inner tube concentrically aligned within an outer $(n_2, m_2)$ tube, where $n_i$ and $m_i$ are the corresponding tube indices.

Focusing first on inter-wall rotations of the achiral systems, we perform a set of constrained energy minimizations starting from a circular DWBNNT configuration and relaxing the geometry at several fixed inter-wall angular orientations ranging from 0$^\circ$ to 2$^\circ$. Fig. 1 presents the corresponding relaxed structures of the armchair (104,104)@109,109) (first row) and the zigzag (180,0)@188,0) (third row) DWBNNTs. As is evident from the figure, the angular orientation of the facets shows strong dependence on the inter-wall rotation angle. In the armchair case, which presents an optimal structure of pentagonal cross section, a dramatic 41.8$^\circ$ revolution of the facet superstructure is obtained for every nominal inter-wall rotation of 2$^\circ$. Similarly, the octagonal circumferential superstructure of the zigzag case revolves by as much as 45$^\circ$ at a similar inter-wall rotation of 2$^\circ$.

All the more pronounced structural variations arise in response to inter-wall telescoping. For the armchair DWBNNTs considered (second row of Fig. 1) the number of facets doubles from 5 to 10 and their angular orientation rotates by 18$^\circ$ upon inter-wall telescoping of 1.25 Å. Notably, for the zigzag DWBNNT (lowest row of Fig. 1) almost complete unfaceting is observed upon an axial shift of merely $\sim 1.7$ Å. The entire structural variation progression obtained during an adiabatic pull-out of 4.2 Å is reported in Supporting Information Movie 1.

The most remarkable structural response is exhibited by the bi-chiral (70,70)@77,74) DWBNNT. The chiral facets appearing in this system couple the translational and rotational
degrees of freedom. Hence, inter-wall telescoping induces global rotation of the entire helical superstructure reminiscent of an Archimedean screw (see Fig. 2 and Supporting Information Movie 3). This represents the smallest device exhibiting unidirectional helical motion that may be utilized as a nanoscale arterial thoroughfare for molecular transport.

On the contrary, the achiral mixed zigzag@armchair (179,0)@(108,108) DWBNNT that possesses the maximal inter-wall chiral angle difference of $\Delta \Theta = 30^\circ$ presents a featureless circular cross section (not shown) regardless of the inter-wall position.

Potential Energy Landscapes

The significant superstructure variations described above are expected to have distinct manifestation in the mechanical and tribological characteristics of faceted nanotube structures. To evaluate these, we compare, in Fig. 3, the potential energy surface (PES) for inter-wall rotation and telescoping of the faceted DWBNNTs (for the carbon counterpart see Supporting Information Fig. S1) considered with those of their circular cross-section counterparts (see Structures and Methods section for technical details regarding the calculation).

Focusing first on the armchair (104,104)@(109,109) DWBNNT (left column in Fig. 3) we find, as expected, that the potential energy corrugation for inter-wall rotations of the circular con-
Figure 2: **Bi-chiral DWNT facet rotation** – Perspective view of the bi-chiral (70,70)@(77,74) DWBNNT at two inter-wall configurations $\theta/z$ of 0.2°/2.4 Å (left) and 0.2°/3.2 Å (right). These correspond to configurations close to maximum and minimum potential energy, respectively (see top-right panel of Fig. 3). Blue and red atom false coloring represents high and low interlayer energy, respectively. Facet dynamics during a pull-out simulation at an inter-wall velocity of 0.01 Å/ps is reported in Supporting Information Movie 3.

The zigzag (180,0)@(188,0) DWBNNT exhibits similar behavior with smooth inter-wall rotation energy landscapes, indicating a smooth process. On the contrary, for the circular configuration, inter-wall telescoping is associated with potential energy variations that follow the mutual hexagonal lattice periodicity, $p_z$, along the zigzag axial direction of the two nanotube walls ($p_z = l\sqrt{3} \approx 2.498\,\text{Å}$, where $l = 1.442\,\text{Å}$ is the equilibrium BN bond length). The latter now follows the periodicity, $p_a$, of the armchair axial direction of the two nanotube walls ($p_a = 3l = 4.326\,\text{Å}$). Unlike the armchair DWBNNT case discussed above, here the circular zigzag DWBNNT configuration presents considerably higher corrugation (8.4 meV/atom) than the faceted one (1.4 meV/atom). This results from the fact that the inter-wall distance in the frustrated circular system, 3.18 Å, is smaller than the optimal value. Upon facet formation the inter-facet distance now increases to a nearly optimal value of 3.25 Å. This, in turn, results in lower barriers along the unfaceting and refaceting sequence obtained throughout the pull-out process.

An overall lower PES corrugation is presented by the circular bichiral (70,70)@(77,74) DWBNNT (right columns of Fig. 3) with relatively smooth telescoping and inter-wall rotation energy profiles (2.3·10^{-8} and 1.3·10^{-4} meV/atom, respectively). This mainly results from the fact that the inter-wall distance at this configuration, 3.78 Å, is considerably larger than the equilibrium value. Similar to the case of the armchair system described above, the appearance of facets effectively re-
duces the inter-facet distance toward the equilibrium value resulting in an increase of the PES corrugation. Nevertheless, while the translational and rotational degrees of freedom remain decoupled in the achiral systems that present axial facets, here they are strongly coupled by the helical facets as demonstrated by the tilted (rather than vertical or horizontal) PES ridges.

Interestingly, the achiral mixed (179,0)@(108,108) DWBNNT has an inter-wall distance of 3.21 Å, comparable to that of the zigzag (180,0)@(188,0) DWBNNT and smaller than the equilibrium value. One might therefore conclude that the two systems should present similar PES corrugation. Nevertheless, the former presents a completely flat translational-rotational PES for both the constrained circular and the fully relaxed configurations (not shown). This may be attributed to the incomensurability of the two hexagonal lattices in both the axial and circumferential directions obtained at the maximal inter-wall chiral angle misfit of 30°.

### Inter-wall Static Friction

A twofold effect of circumferential faceting on the inter-wall PES of DWBNNTs is thus found: (i) Facet restructuring during inter-wall displacements results in inter-wall distance variations that may increase or decrease PES corrugation depending on the corresponding distance within the unfaceted system; (ii) Helical facets, appearing in bichiral DWBNNTs, couple the translational and rotational degrees of freedom. The immediate physical manifestation of these effects is expected to appear in the static inter-wall friction exhibited by the DWNT.

The static friction force is defined as the minimal force required to initiate relative motion between the two nanotube walls that are initially interlocked in a (local) free-energy minimum. Despite the general non-uniformity of real telescopic sliding, also depending on the pulling mode, static friction may, in the low temperature limit \( T \to 0 \) K, be estimated from the inter-wall telescoping-rotation PES by evaluating the energy barrier required to lift the interface out of the equilibrium state. To this end, we plot the energy variations during adiabatic axial inter-wall pull-out and rotation and fit them to a sinusoidal curve of the form \( E(z) = (E_c/2) \sin(2\pi z/\Delta z) \) (See Supporting Information Fig. S2). The static friction is then extracted from the maximal derivative of the fitted curve given by \( F_s = \pi E_c/\Delta z \). A summary of the obtained PES corrugation and the corresponding static friction values appears in Supporting Information Tab. S1.

As may be expected, for the achiral armchair and zigzag systems the static friction force required to initiate inter-wall rotational motion is negligible compared to that necessary to trigger telescopic sliding for both the circular and the faceted configurations. The pull-out static friction force of the armchair DWBNNT at the circular geometry is 0.17 meV/Å per atom, much lower than the corresponding value of the zigzag system (11.8 meV/Å). As discussed above, in the relaxed configuration the inter-facet distance approaches the equilibrium value in both systems resulting in similar friction forces of 1.91 and 2.04 meV/Å per atom for the armchair and zigzag DWBNNTs, respectively.

The bichiral system in its circular geometry presents a negligible static friction force for axial shifts \((3.8 \cdot 10^{-8} \text{ meV/Å per atom})\), while a larger value, yet considerably smaller than the characteristic forces exhibited by the achiral systems, is obtained for inter-wall rotations \((2.2 \cdot 10^{-4} \text{ meV/Å per atom})\). At the faceted configuration the static friction forces for both telescoping and rotation increase yielding values of about \(3.5 \cdot 10^{-3}\) and \(2.5 \cdot 10^{-3}\) meV/Å per atom, respectively. It is clear from the upper right panel of Fig. 3 that a combined rotation and telescoping displacement path which follows the facet helicity will result in a considerably lower static friction force. For the mixed achiral DWBNNT considered that, as mentioned above, exhibits completely flat rotation-telescoping PES maps for both the circular and relaxed (unfaceted) geometries, we could not extract any meaningful static friction force values.
Dynamic Friction

Not only do the superstructure reconfigurations described above impact the static nanotube inter-wall friction but they provide a key to understanding the surprisingly high inter-wall dynamic friction recently measured for MWBNNTs with respect to their carbon counterparts.\(^{47}\) The underlying mechanism relates to the fact that the facet superstructural collective degrees of freedom introduce auxiliary energy dissipation routes that enhance dynamic friction. This is true for both translational and rotational inter-wall motion even when the latter presents negligible PES corrugation and static friction forces.

To quantify these effects we performed fully atomistic molecular dynamics inter-wall sliding simulations (see Structures and Methods section for details) of the DWBNNTs considered. When following the structural variations occurring during the telescopic pull-out of the armchair and zigzag DWBNNTs’ inner shells at a relative velocity of 0.01 Å/ps we observe a full unfaceting and refaceting superstructure cycle, superposed on asymmetric deformations induced by inertial effects (see Supporting Information Movie 2). For the bi-chiral DWBNNT we find that telescopic motion, at the same relative axial velocity, induces circumferential rotation of the helical facets with an angular velocity of about 0.24°/ps (evaluated from the simulated time evolution in Supporting Information Movie 3). This corresponds to a linear superstructure surface velocity of ≈0.2 Å/ps (assuming an average tube diameter of ≈10 nm), which is more than 20 times faster than the applied axial velocity.

Following Newton’s first law, we define the instantaneous dynamic friction force as the force required to maintain a constant velocity relative inter-wall sliding motion. To allow for comparison between DWNTs of different diameters we extract the shear stress by normalizing the calculated forces to the nominal surface contact area. In Fig. 4 the temporal shear stress traces obtained during constant velocity inner shell pull-out (see Structures and Methods section) are reported. We start by considering the achiral armchair (104,104)@ (109,109) and zigzag (180,0)@ (188,0) DWBNNTs (blue lines in the upper left and middle panels, respectively). To evaluate the effect of facet reconfiguration on the dynamic friction force we perform reference calculations on narrow armchair (31,31)@ (36,36) and zigzag (55,0)@ (63,0) DWBNNTs green lines in the upper left and middle panels, respectively that are below the critical diameter for facet formation.\(^ {15,20}\)

Due to their axial inter-wall translational
symmetry, the achiral DWNTs present periodic dynamic friction force variations with large peak values reflecting increased interfacial commensurability. Interestingly, the overall amplitude variations of the shear stress traces of the faceted DWBNNTs are comparable to those of the narrower circular systems. Nevertheless, while the circular systems present a nearly-sinusoidal smooth behavior, the faceted DWBNNTs show a complex pattern of rapid force fluctuations with clear asymmetry between the positive and negative shear stress regions. This is a clear manifestation of the effects of superstructure reconfigurations occurring during the pull-out dynamics in the presence of facets. As a consequence, the dynamic friction force, evaluated as the time averaged shear stress over an integer number of periods, is found to be 5-17 times larger in the faceted achiral DWBNNTs than in the circular systems studied.

We may therefore conclude that faceting which, as discussed above, is considerably more prevalent in MWBNNTs than in MWCNTs, may be responsible for the enhanced friction measured for the former. To understand how the inter-wall friction of the less abundant faceted MWCNTs compares to that of their BNNT counterparts, we have repeated our calculations for the corresponding achiral DWCNTs (see lower panels of Fig. 4). Similar to the case of DWBNNTs, the circular achiral DWCNTs show a much smoother and more symmetric shear stress trace (see light-grey lines in the lower left and lower middle panels of Fig. 4) resulting in considerably smaller dynamic friction forces than the faceted achiral systems (dark-grey lines). Interestingly, even for the latter, the kinetic friction force extracted is smaller by a factor of 3.4-3.8 than that of the corresponding faceted DWBNNTs with the force-field parameters used herein (see Structures and Methods section). Importantly, this is true also for the zigzag (180,0)@(188,0) DWNTs considered, where the PES corrugation of the BN based system was found to be comparable to that of its carbon counterpart (Figs. 3 and S1).

For the bi-chiral (70,70)@(77,74) DWBNNT considered no periodic kinetic friction force variations are observed (see upper right panel of Fig. 4). Furthermore, following some initial transient dynamics, smooth steady-state sliding...
motion with nearly-constant drag is obtained. This can be attributed to the reduced interwall commensurability and PES corrugation in this system (see right panels of Fig. 3). Consequently, the average dynamic friction force recorded in this case (~3.2 pN/nm²) is 20-fold times smaller than that of the faceted achiral systems. Nevertheless, it remains nearly an order of magnitude larger than the value measured for the corresponding bi-chiral DWCNT (0.4 pN/nm², see lower right panel of Fig. 4) and a factor of 80-320 larger than the kinetic friction measured for the achiral circular DWCNTs considered.

Finally, we study the velocity dependence of the interlayer sliding friction of DWBNNTs and DWCNTs in the range of 0.2-1.0 m/s (see Supporting Information Fig. S5). Our results show nearly linear increase of the friction force with the sliding velocity at the velocity range considered. For the axially commensurate armchair DWNTs the friction extrapolates to a finite value at zero velocity. This can be attributed to the finite static friction exhibited by these systems. For the incommensurate bi-chiral DWNTs the friction extrapolates to zero at vanishing interwall sliding velocity. This is in line with the experimental observation of viscous inter-wall telescopic motion in multi-walled NTs, where sliding is expected to occur at the weakest incommensurate interface. The calculated inter-wall friction forces in both DWCNTs considered are found to be weakly dependent on the sliding velocity and are consistently lower than those obtained for the corresponding DWBNNTs. This further supports the experimental observations of increased inter-wall friction in MWBNNTs over MWCNTs.

Conclusions

The resulting screw-like motion of the faceted helical pattern establishes the smallest realization of an Archimedean screw with the potential to achieve directional transport of weakly adsorbed molecules along the surface of the tube. We note that the super-structure variations discussed above may be viewed as the nanotube analogues of the soliton-like motion of moiré patterns occurring in sliding incommensurate planar interfaces. Nevertheless, due to geometric frustration in the tubular configuration, the extended circumferential registry patterns result in considerably larger structural deformations. The latter exhibit much richer dynamic behavior with marked influence on the mechanical, tribological, and electronic properties of the system.

The motion of such collective degrees of freedom opens new dissipative channels that enhance dynamic friction beyond the excitation of localized phonon modes. Since faceting is more commonly observed in MWBNNTs than in their carbon counterparts this rationalizes recent experimental findings showing that the former exhibit an order of magnitude larger dynamic friction. Furthermore, even when compared to the less abundant case of faceted DWCNTs, the BN systems exhibit 3-8 times larger dynamic friction forces. Hence, when designing smooth nanoscale bearings one should resort to unfaceted MWCNTs whereas if torsional and axial rigidities are desired facetted MWBNNTs should be the material of choice.

Finally, several other, more speculative but highly intriguing, consequences of the striking facet evolutions discussed herein can be envisioned. First, we have shown that facet dynamics strongly depends on the relative chirality of adjacent nanotube walls. Therefore, the inter-wall pulling force trace should encode information about the identity of the various tube shells. This, in turn, opens new opportunities for novel material characterization techniques that may provide access to the specific sequence of chiralities of successive nanotube walls. Furthermore, electronic effects, not discussed herein, may also exhibit unexpected behavior. Specifically, surface states that typically localize at sharp edges, such as the circumferential vertices of the polygonal cross-section, may also be pumped along the surface of nanotubes in an Archimedean manner.
Structures and Methods

DWNTs can have inner and outer walls that are either zigzag (ZZ), armchair (AC), or Chiral (Ch). In the present study four types of carbon and boron nitride (BN) DWNTs have been considered including the achiral AC@AC (104,104)@(109,109) and ZZ@ZZ (180,0)@(188,0) systems; the mixed achiral ZZ@AC (179,0)@(108,108); and the bi-chiral AC@Ch (70,70)@(77,74). Here, the notation $(n_1,m_1)@(n_2,m_2)$ represents a $(n_1,m_1)$ inner tube concentrically aligned within an outer $(n_2,m_2)$ tube, where $n_i$ and $m_i$ are the corresponding tube indices. Monochiral DWNTs that have chiral walls with matching chiral angles present axial facets like the achiral systems and are therefore not considered herein.

A summary of the relevant geometric parameters of the unrelaxed DWNTs appears in Table S2.

The structural and frictional properties of all DWNTs considered have been described using dedicated intra- and inter-layer classical force-fields as detailed below. For DWCNTs the intra-layer interactions have been described using the Tersoff potential adopting the parameterization of Lindsay and Broido. The inter-layer interactions of these systems have been described by the registry-dependent Kolmogorov-Crespi potential in its RDP1 form.

For the intra-layer interactions of DWBNNTs we have used the Tersoff force-field as parameterized by Sevik et al. for BN based systems, along with our recently developed $h$-BN interlayer potential with fixed partial charges. We note that suppressing the coulombic interactions between the partially charged atomic centers in the DWBNNTs studied ($q_B = +0.47\, e$, $q_N = -0.47\, e$) results in a reduction of merely $\sim 3.5\%$ in their calculated PES corrugation (see Supporting Information Fig. S3). Corrugation and adhesion energy profiles for rigid planar bi-layer of $h$-BN and graphene, as obtained by the above set of interlayer potentials, are reported in Supporting Information Fig. S4.

Periodic boundary conditions (PBC) along the tube axis have been applied to all DWNTs considered, resulting in a very small ($<0.1\%$) stress in the case of the bi-chiral and mixed systems due to the different lattice constants of the inner and outer tubes. In all the cases, an initial step of relaxation of the cell vectors has been performed in order to minimize any PBC-related stress effects.

In the pull-out/rotation potential energy surface calculations (Fig. 3) each point has been obtained by placing the two unrelaxed cylindrical nanotube walls at the corresponding relative axial and angular position followed by geometry optimization using FIRE quenched dynamics, while nullifying the center of mass (c.o.m.) axial and angular velocity of each nanotube wall. All the constrained relaxations were stopped after 5000 FIRE iterations, providing energy evaluations that are converged to within 0.1% of the highest energy obtained across the PES maps. The reported energy per-atom has been obtained by dividing the converged energy by the total number of atoms in the DWNT. We note that this procedure corresponds to an adiabatic relative motion of the tubes that can, in principle, be realized in experiment by adhering the outer tube wall(s) to a fixed stage and applying a slowly varying external force on the inner shells via the manipulation of an external tip. Although in typical experimental setups the external force is applied at one edge of the inner shells, their extreme stiffness permits the instantaneous propagation of the stress along the entire tube length. Hence, the calculated PESs should reliably describe the corresponding inter-wall energy variations measured in the experiment.

Dynamic friction calculations have been performed by numerically propagating the Langevin equation of motion using the standard molecular dynamics velocity-Verlet algorithm. The simulations have been performed in the underdamped regime by applying viscous damping to all degrees of freedom apart from the c.o.m. motion of both tubes. The dynamic friction force is evaluated from the inter-wall shear force required to keep the two nanotube walls at constant relative velocity motion $v_{\text{ext}}$. To this end, we have fixed the c.o.m. of the internal tube and applied a uniform force $F_{\text{ext}}$ to each of the $N$ atoms of the external tube so...
that

\[ v^1_i = v^0_i + \left( \frac{F_i + F_{ext}}{m_i} - \gamma (v^0_i - v^0_{cm}) \right) \Delta t \quad (1) \]

\[ \sum_{i=1}^{N} v^1_i = N v_{ext} \quad (2) \]

where \( v^0_i \) and \( v^1_i \) are the \( i \)-th atom velocities at times \( t_0 \) and \( t_1 \), respectively, \( \Delta t = t_1 - t_0 \) is the numerical propagation timestep, \( v^0_{cm} \) is the c.o.m. velocity of the external tube at \( t = t_0 \), \( m_i \) the atomic mass, \( F_i \) is the total force on atom \( i \) due to the chosen set of interatomic potentials, and \( \gamma = 0.1 \text{ ps}^{-1} \) is the viscous damping coefficient used in the simulation to avoid system overheating. Since the viscous damping is not applied to the c.o.m. motion of the tubes, the computed friction results weakly dependent on the adopted \( \gamma \) value, the latter mainly determining the steady-state temperature of the sliding system. In our typical simulations, which were run in the underdamped regime, we measured steady-state temperatures below 1 K, suggesting a negligible role of \( \gamma \) on the measured friction.

From Eqs. 1-2 we obtain

\[ F_{ext} = \frac{\bar{m} \bar{v}_{ext}}{\Delta t} + \bar{m} (\gamma - \frac{1}{\Delta t}) \bar{v} - \bar{m} \gamma v^0_{cm} - \bar{m} \bar{a} \quad (3) \]

where

\[ \bar{m} = N \left( \sum_{i=1}^{N} \frac{1}{m_i} \right)^{-1} \quad (4) \]

\[ \bar{a} = \frac{1}{N} \sum_{i=1}^{N} \frac{F_i}{m_i} \quad (5) \]

\[ \bar{v} = \frac{1}{N} \sum_{i=1}^{N} v^0_i \quad (6) \]

Since \( F_{ext} \) is applied to all the atoms of the external tube, the instantaneous friction force of the entire surface, \( F_{fric} \), is simply expressed by

\[ F_{fric} = N F_{ext} \quad (7) \]

Finally, the obtained dynamic friction force \( F_{fric} \) is normalized to the inter-wall contact area evaluated from the average diameter of the unrelaxed-cylindrical configuration (see Table S2), leading to the system-specific shear stress value. This allows for a direct comparison among forces calculated for DWNTs of different type and dimensions. The resulting shear stress has been averaged over a time window of at least 1 ns during the steady-state motion, after the initial transient dynamics has decayed, covering an integer number of oscillations in the case of periodic force traces.

We note that using this procedure a direct quantitative comparison with experimental data is hard to achieve, due to the large sliding velocities, to which MD simulations are limited, compared to those accessible in realistic experimental conditions. Despite this, our dynamic simulations allow for a comparative study of the tribological properties of faceted and unfaceted DWNTs of different chemical composition.

### Associated Content

**Supporting Information**

- PES of armchair, zigzag, and bi-chiral DWCNTs (relaxed and cylindric); example of sinusoidal fitting of a PES to estimate static friction; comparison of the PES of armchair DWBNNT with and without partial charges contribution; corrugation and adhesion energy profiles for a bilayer of graphene and of h-BN; effect of sliding velocity on the average friction of armchair and bi-chiral DWCNTs and DWBNNTs; table reporting evaluated static friction and maximum corrugation energy for the reference DWNTs set; table reporting chiral angle difference, average diameter, and average interlayer spacing for the reference DWNTs set; movie of the cross sectional view of armchair and zigzag DWNTs relaxed at different relative axial positions \( z \); movie showing a comparison of the cross sectional view of the armchair DWBNNT in telescopic motion at zero
(adiabatic motion) and at large pulling velocity; movie of the bi-chiral DWBNNT at large relative pulling velocity; movie of the bi-chiral DWCNT at large relative pulling velocity.

Acknowledgments

Work in Trieste was carried out under ERC Grant 320796 MOPHYSLANPRICT. EU COST Action MP1303 is also gratefully acknowledged. O.H. acknowledges the Lise-Meitner Minerva Center for Computational Quantum Chemistry and the Center for Nanoscience and Nanotechnology at Tel-Aviv University for their generous financial support.

Author contributions

R.G. and I.L. developed the simulation code and performed the calculations. R.G. conceived the method to evaluate dynamic friction, and produced the figures and the movies. All the authors actively participated in the data analysis and in the writing of the manuscript.

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