Giant Wave-Drag Enhancement of Friction in Sliding Carbon Nanotubes

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Molecular dynamics simulations of coaxial carbon nanotubes in relative sliding motion reveal a striking enhancement of friction when phonons whose group velocity is close to the sliding velocity of the nanotubes are resonantly excited. The effect is analogous to the dramatic increase in air drag experienced by aircraft flying close to the speed of sound, but differs in that it can occur in multiple velocity ranges with varying magnitude, depending on the atomic level structures of the nanotubes. The phenomenon is a general one that may occur in other nanoscale mechanical systems.

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The miniaturization of mechanical devices has evolved to the point that microelectromechanical systems (MEMS) are now in widespread use in a growing number of commercial products. Miniaturization has many advantages including increased speed and sensitivity, reduced power consumption, and improvements in the positional and spatial precision with which complex functions can be performed. At all length scales the scale-relative properties of mechanical systems vary with their size due to increasing edge to surface and surface to volume ratios. However, as the nanometer length scale is approached, an entirely new set of phenomena are becoming relevant that provide great challenges to the further miniaturization of devices. The precise atomic and electronic structure of devices, the chemical reactivity of device components, the quantum mechanical behaviour of atoms, and the presence of large thermal fluctuations are all issues of increasing importance at this scale. Engineering useful nanoscale mechanical devices will require an understanding of these effects and their impacts on device efficiency and reliability.

Multiwalled carbon nanotubes (MWCNTs) can be fabricated with near structural perfection and their resistance to deformation and the smoothness with which coaxial tubes can move relative to one another suggest that they could be ideal materials from which to build nanoscale machinery. While useful nano-mechanical systems have not yet been constructed, some prototype mechanical elements with sub-micron dimensions have been made using MWCNTs\textsuperscript{1} and it seems unlikely that the ultimate lower size limit of functional mechanical devices has, on the micron scale, already been reached. Theory and simulation can help to investigate the limitations of nanoscale machinery and to improve their design by providing a more detailed understanding of energy loss mechanisms than can be inferred from experiment. This Letter is concerned with one of the most important gross features of nanoscale phononic friction: Its dependence on velocity.

Small volume (mass) to surface (interaction area) ratios\textsuperscript{2,3} mean that components of nanoscale devices can move with enormous accelerations and velocities. Simulations of an oscillator composed of double-walled carbon nanotubes(CNT)\textsuperscript{4} showed that even the weak van der Waals interactions between layers of a MWCNT are sufficient to accelerate nanotubes to velocities of thousands of meters per second\textsuperscript{2,3}.

In this work, some results of molecular dynamics (MD) simulations of CNT “shuttles”, of varying length, sliding within a larger radius CNT under periodic boundary conditions\textsuperscript{3} are reported. The focus is on a striking phenomenon, involving an enhancement of friction of several orders of magnitude, that occurs in certain velocity

![Graph showing sliding velocity vs. time and friction force vs. sliding velocity](image-url)
FIG. 2: The difference in \((\omega, k)\) space between the power spectra of the \((10,10)\) CNT calculated at sliding velocities of \(1100\,\text{ms}^{-1}\) and \(1000\,\text{ms}^{-1}\). Inset: Frequency-dependent power spectrum at different sliding velocities.

FIG. 3: The lines are the phonon bands of an isolated \((10,10)\) nanotubes at 0 K. The RB and LA modes are indicated by dashed lines. The underlying color plot is the \(k\)-resolved phonon power spectrum of the \((10,10)\) CNT when the \((5,5)\) shuttle is moving inside it at \(1000\,\text{ms}^{-1}\). Circles indicate the positions of the peaks seen in Fig. 2 while the boxes are points that satisfy the proposed conditions for enhanced friction.

FIG. 4: Average over time and azimuthal angle of the local radius, \(r_{ave}\) of the \((10,10)\) CNT as a function of displacement from one end of the \((5,5)\) CNT at three different sliding velocities. The value of \(v_s\) that is used here is the average velocity in each simulation, and not \(v_0\). As expected from Fig. 1(a), there is a massive increase in \(F_f\) near \(v_s = 1100\,\text{ms}^{-1}\) for the \((10,10)-(5,5)\) system. Figure 1(b) also shows \(F_f\) vs \(v_s\) (scaled by a factor of two) for a \((5,5)\) shuttle sliding within a (chiral) \((16,2)\) CNT. In this case the shuttle and the outer tube are incommensurate and there are peaks in \(F_f\) at many different velocities. Although the peaks in the \(F_f\) are suggestive of resonances, and the possibility of resonant friction between CNTs has previously been proposed, it will be shown that this effect cannot be explained by a resonant mechanism alone. Due to its relative simplicity, the analysis in the remainder of this Letter is focused on the \((10,10) - (5,5)\) system.
tion of the corrugated surface potentials directly excites particular phonons. A phonon excited in this way can be resonantly excited if its frequency coincides with the “washboard frequency,” \( \omega_{wb} = 2\pi v_s/a \), where \( v_s \) is the sliding velocity, and \( a \) is a length scale that is common to both surfaces and that plays a role in the excitation of that particular phonon. \( \omega_{wb} \) can take any value depending on the value of the \( v_s \).

To identify which phonons are involved in the observed enhancement of friction the displacement-dependent velocity-velocity correlation function (i.e. \( A(z_i - z_j, t_1 - t_2) = \langle v_i(t_1) v_j(t_2) \rangle \), where \( z_i, z_j \) are the displacements along the axis of the outer nanotube of the primitive unit cells to which atoms \( i \) and \( j \) belong, and \( v_i(t_1), v_j(t_2) \) are their velocities at times \( t_1 \) and \( t_2 \), of the (10,10) tube was Fourier transformed in time and space. This gives the frequency \( \omega \) and wavevector \( k = 2\pi/\lambda \) dependence of its phonon power spectrum. Figure 2 shows the difference in \( (\omega, k) \) space between the power spectra calculated at 1100 ms\(^{-1} \) and at 1000 ms\(^{-1} \). One of the two peaks in this difference spectrum is centered at \( (\omega, \lambda) = (146 \text{ cm}^{-1}, \infty) \) and the other, less symmetric peak, has maximum intensity at \( (\omega, \lambda) = (140 \text{ cm}^{-1}, 3.68 \text{ nm}) \) and a substantial shoulder at \( (137 \text{ cm}^{-1}, 3.16 \text{ nm}) \). The inset to Fig. 2 demonstrates that, apart from these modes at \( \omega \approx 140 \text{ cm}^{-1} \) that are strongly excited at \( v_s = 1100 \text{ ms}^{-1} \), the phonon power spectrum is rather insensitive to the sliding velocity.

The phonon band structure of the (10,10) CNT at 0 K was calculated by diagonalizing its dynamical matrix. These bands are superimposed in Fig. 3 on an image depicting the \( k \)-resolved power spectrum calculated from the MD simulations at 1000 ms\(^{-1} \). The phonon energetics are obviously different in the 300 K system with the outer nanotube that moves at close to the value of \( v_s \) and \( v_{wb} \), of the phonon that is excited by the sliding motion is close to the value of \( v_s \) required for a match between the phonon frequency, \( \omega(k) \), and \( \omega_{wb} \), i.e. the following two criteria are simultaneously met: (i) \( v_s(k) = d\omega(k)/dk \approx v_s \) and, (ii) \( \omega_{wb}(v) \approx \omega(k) \Rightarrow v_{wb} = \omega_{wb}a/2\pi \approx v_s \). Evidence to support this hypothesis will now be provided. This evidence consists of a demonstration that these criteria can only be met at velocities close to 1100 ms\(^{-1} \) by phonons that have frequencies and wavevectors consistent with the anomalous phonon excitation demonstrated in Fig. 2 and furthermore, that these phonon modes are those that would, most obviously, be excited by the washboard mechanism - the axially symmetric radial breathing (RB) and longitudinal acoustic (LA) modes. The LA mode and the RB mode both have perfect axial symmetry and they mix strongly with each other and with other phonon modes near \( (\omega, k) = (140, 0.1) \) where a band anticrossing occurs.

In Fig. 3 circles mark the positions in \( (\omega, k) \) space of the peaks seen in Fig. 2. Four other points are marked with boxes. At these points criteria (i) and (ii) are both met. Criteria (i) and (ii) are also met at a number of other points, however, none of these other phonons either have axial symmetry or group velocities in the range \( 950 \text{ ms}^{-1} < v_g < 1250 \text{ ms}^{-1} \). Furthermore, of these phonons, all but one (for which \( \omega \approx 128 \text{ cm}^{-1} \)) have frequencies outside the range \( 120 \text{ cm}^{-1} < \omega < 160 \text{ cm}^{-1} \). The four marked points in Fig. 3 mostly have the character of RB modes with some LA character in the points closest to the band anti-crossing.

The agreement between two of these four phonons and the simulation results presented in figures 1 and 2 is remarkably good given that they have been calculated from the 0 K phonons of an isolated (10,10) tube. The values of \( v_s \) and \( v_{wb} \) are close to 1100 ms\(^{-1} \) and they are close in \( (\omega, k) \) space to the positions at which anomalous phonon excitation occurs in the MD simulations. Two other phonons do not, at first, appear consistent with figures 1 and 2. The first of these has a group velocity of \( \sim 500 \text{ ms}^{-1} \) and no peak is observed at this velocity in Fig. 1. However, the wavelength of this phonon is \( \lambda \approx 24.5 \text{ nm} \) which is larger than the length of the simulation cell, and therefore it cannot be excited in the MD simulations. The second point, at \( \lambda \approx 1.5 \text{ nm} \), has a group velocity close to 1100 ms\(^{-1} \) but no anomalous phonon excitation at \( \lambda \approx 1.5 \text{ nm} \) is observed in the MD simulations. A possible reason for this is provided below.

The MD simulations have been repeated with shuttles of lengths (in nm, and excluding the lengths of the caps) 10, 8, 5.3, 2.6 and 0 (i.e. a C\(_{60}\) molecule). No enhancement of \( F_f \) was observed near 1100 ms\(^{-1} \) or at any other velocity for the C\(_{60}\) molecule. There were peaks at \( v_s = 1100 \text{ ms}^{-1} \) for all other shuttle lengths and the heights of the peaks increased almost linearly with the shuttle length. For the 2.6 nm shuttle, there was anomalous phonon excitation at the smaller wavelength \( \lambda \approx 3.2 - 3.7 \text{ nm} \) but no anomalous phonon excitation at \( \lambda \to \infty \). As the length of the shuttle was increased, the \( \lambda \to \infty \) peak began to grow at the expense of the smaller wavelength peak until, for the longest shuttle, this smaller wavelength peak had almost disappeared. It is reasonable that short shuttles cannot excite very long wavelength phonons and that, as the shuttle length increases, phonons with a larger wavelength are excited. Furthermore, when the washboard excitation is in-phase with a phonon at a given point along the outer tube
it must be in anti-phase with a point a distance $\lambda/2$ away. Therefore, a shuttle cannot resonantly excite a phonon whose wavelength is substantially smaller than twice its length. This explains the shift in intensity of the anomalous phonon excitation from shorter to longer wavelengths as the shuttle length increases, and the absence of anomalous excitation of phonons at a wavelength of $\sim 1.5$ nm.

A physical mechanism is now proposed for the results of the MD simulations: When $v_s$ is sufficiently close to the group velocity of a RB mode of the outer nanotube that is being resonantly excited by the relative motion of the corrugated surface potentials of the nanotubes, a wave packet remains in the vicinity of the shuttle for long enough that it is reinforced and amplified. The build up of a radial distortion in the outer nanotube that moves along with the shuttle results. This distortion, in turn, changes the interaction between the two nanotubes - a feedback that makes the response of the outer nanotube to the presence of the shuttle strongly non-linear. The details of this non-linearity are likely to be complicated and sensitively velocity-dependent, but the general effect is analogous to the sharp increase in drag experienced by airplanes travelling at close to the speed of sound. At low speeds, an airplane creates a disturbance in the air and sends out pressure pulses that travel ahead of it to separate the air flow, thereby allowing air to move smoothly around it. As Mach 1 is approached, however, pressure waves build up to form a shock front just ahead of the airplane. Air in the airplane’s path has little warning of its arrival and so the air’s compressibility plays a crucial role in the flow dynamics. The dynamics become strongly nonlinear, instabilities appear in the flow, and lots of energy is dissipated by the shock front. The drag that results is commonly known as “wave drag” and its maximum as a function of velocity is the “sound barrier”. At supersonic speeds the flow stabilizes and the drag is reduced.

Further evidence that a similar mechanism is responsible for the enhancement of friction between CNTs is provided in Fig. 4 which shows the average radius, $r_{ave}$, of the outer CNT as a function of displacement, $Z$, from one end of the shuttle. The average is performed over the azimuthal angle and over time. At velocities of 500 ms$^{-1}$ and 1000 ms$^{-1}$, $r_{ave}$ is very similar, however, at 1100 ms$^{-1}$ there is a large change, suggesting that a wave builds up around the shuttle at this velocity. The fact that the difference between $r_{ave}$ at 1100 ms$^{-1}$ and at 1000 ms$^{-1}$ is of the same order of magnitude as the variations in $r_{ave}$ at either 500 ms$^{-1}$ or 1000 ms$^{-1}$ implies that the inter-tube interaction is strongly affected by the distortion. This, in turn, implies a non-linear response of the outer CNT to the shuttle that could explain the sharp increase in the rate of energy dissipation.

A vast literature exists on wave-drag and related phenomena in the context of aeronautics and much of the theory may be adaptable to the nanoscale. One important difference at the nanoscale, however, is that, depending on the atomic level structure, this effect can occur in the same device at multiple velocities. Particularly for incommensurate systems, there may be more than one length scale that is common to the contacting surfaces and that can cause washboard resonances. Furthermore, multiple phonons with different group velocities may satisfy the criteria necessary for this effect to occur. The (16, 2) – (5, 5) system shown in figure 4(b) is an example in which there are many peaks in $F_j$ vs $v_s$.

In conclusion, a mechanism by which huge increases in mechanical energy dissipation at well defined velocities can occur at the nanoscale has been demonstrated using atomistic simulations of coaxial carbon nanotubes sliding relative to one another. As demonstrated in Fig. 4(a), this effect can have a dramatic impact on the dynamics of a nano-mechanical system.

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