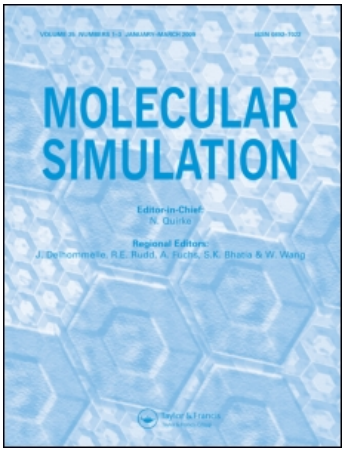

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MOLECULAR DYNAMICS SIMULATIONS OF CARBON NANOTUBE ROLLING AND SLIDING ON GRAPHITE

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Molecular dynamics simulations were carried out to investigate the origin of friction for carbon nanotubes on graphite substrates. In an initial simulation, a (10,10) nanotube was placed in an 'in-registry' starting position where the hexagonal lattice of the substrate matched that of the nanotube. In a second simulation, the substrate was oriented 90 degrees to the nanotube. A uniform force was applied to the nanotubes for 500 fs to set them into motion. The simulation was then run until the nanotubes stopped moving relative to the substrate. Only sliding was observed in the out-of-registry simulation, while periodic sliding and rolling was observed in the in-registry simulation. The latter is a result of the relatively larger surface corrugation for the in-registry case and occurs to avoid direct atomic collisions between nanotube and substrate atoms as the nanotube is moved along the substrate. Analysis of the kinetic energy suggests that the transition between sliding and rolling contributes to enhanced energy dissipation and higher net friction. These results are consistent with preliminary experimental observations by Superfine and coworkers.

Keywords: Nanotube; nanotribology; nanomanipulation

INTRODUCTION

Friction between carbon nanotubes and different materials plays an important roll in utilizing nanotubes in various applications. For example, one application is the use of nanotubes as fiber reinforcement in composites, where knowledge of frictional behavior would assist in the selection of matrix materials with high resistance to fiber pull-out. Another example is

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the use of nanotubes as solid lubricants for next generation nanoscale analogs of microelectrical-mechanical devices.

Superfine and coworkers have recently characterized the frictional properties of multiwalled nanotubes on graphite using an atomic-force microscope with a unique tactile interface [1]. These experiments show that nanotubes have preferred orientations on graphite substrates in which the structure of the nanotube is in registry with that of the substrate [2]. These experiments have also suggested that nanotubules can either slide or roll when pushed depending on this registry. Preliminary data, for example, suggests that nanotubes tend to slide when out of registry and roll when in registry, with the former having an apparent lower friction. Signatures of complicated motion with periodicities matching that of the substrate have also been observed. Recent calculations by Lu and Buldum, which are to be discussed in this meeting, have confirmed that nanotubes energetically prefer in-registry orientations, and that slide *versus* rolling behavior is dependent on this orientation [3].

This paper is a preliminary report of molecular dynamics simulations of the motion of a single-walled (10,10) nanotube on a graphitic substrate. These calculations predict differences in motion for nanotubes in different orientations, with the out-of-registry orientation showing sliding, and the in-registry orientation showing relatively complicated slide-roll behavior. The frictional force, as determined by slowing of the nanotube after being initially accelerated, is also found to depend on the mechanism of motion. The results are consistent with experiment, although the difference in friction coefficient is much smaller between and in and out of registry nanotubes is smaller than inferred from experiment [2].

COMPUTATIONAL METHODS

The nanotube-graphite system was modeled as follows. The nonbonded interaction between the nanotube and substrate was modeled using a Lennard-Jones 6–12 potential with parameters derived by Girifalco [4, 5]. Two-dimensional periodic boundaries were enforced in the plane of the graphite substrate. This resulted in a nanotube of infinite length, and kept the nanotube in the desired orientation. Two layers of graphite were used for the substrate. The top layer was flexible, with a temperature maintained at 300 K using a generalized Langevin thermostat. This allowed energy to be dissipated from the nanotube, resulting in a net frictional force. The bottom layer was held rigid as an approximation to a many-layer graphite substrate.

The intratomic interactions within the nanotube and the substrate were modeled with a many-body bond order potential [6]. This allows deformation of the nanotube in response to the nanotube-substrate interactions. Intratube van der Waals interactions were neglected in this simulation.

Two simulations were carried out. In the first simulation, a (10,10) nanotube was placed in an in-registry orientation where the hexagonal lattice of the substrate matched that of the nanotube. In the second simulation, the substrate was oriented at 90° to the tubule. To match periodic boundaries, a ratio of 3 graphite unit cells to 5 nanotube unit cells was used. For an exact match, a 4% compressive strain was applied to the substrate in the direction of the tube axis.

After relaxation, the nanotube flattened slightly as a result of adhesive forces between tube and substrate. The resulting structures were very similar to those predicted by Hertel *et al.* [7]. A uniform force was then applied to the nanotube to initiate translation. The force was released after 500 fs and the tube was allowed to travel along the substrate. The resulting coordinate output was captured every 100 fs.

RESULTS

Analysis of animated sequences from the simulations reveals that the in-registry nanotube slides as it travels along the substrate for the first 15 ps of the simulation, and then begins an alternating slide-roll behavior. At approximately 100 ps the tube comes to a complete stop. The out-of-registry nanotube slides along the substrate rather than rolling. The deceleration of the nanotubes was estimated by taking the second derivative of a quadratic equation fit to the position of the nanotubes *versus* time. The friction of the out-of-registry case is approximately 80% that of the in-registry case.

Plotted in Figure 1a are the various contributions to the kinetic energy for the in-registry simulation; plotted in Figure 1b is the corresponding data for the out-of-registry case. The out-of-registry simulation shows a relatively smooth decrease in the total and translational energy and very little rotational kinetic energy. The in-registry case shows a relatively complex periodic behavior as it decreases. To analyze this periodic behavior, the translational kinetic energy was normalized using the equation

$$y_i' = y_i - (mx_i + b) \tag{1}$$

where y_i' is the normalized value of the kinetic translational energy, y_i is the initial translational kinetic energy, m and b are the slope and intercept

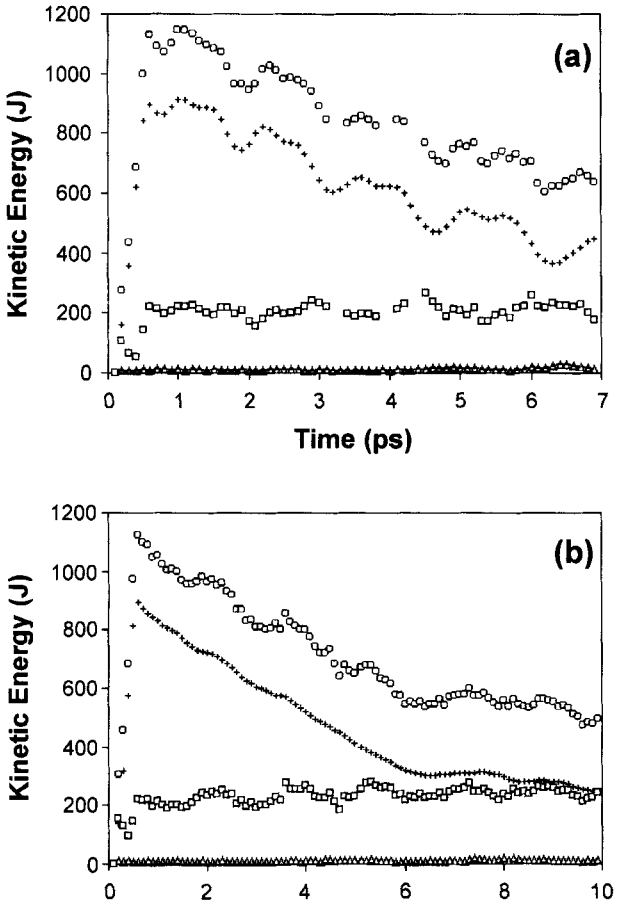

FIGURE 1 Total, translational, vibrational, and rotational kinetic energy (shown as circles, crosses, squares and triangles, respectively) plotted as a function of time. (a) For an in-registry nanotube (b) For an out-of-registry nanotube.

from a linear fit to the translational data from Figure 1a, and x_i is the center of mass position of the tubule along the substrate. The result is plotted in Figure 2a (indicated by crosses). Superimposed on Figure 2a is the rotational kinetic energy (indicated by open triangles). The average period in the data from Eq. (1) is 4.58 angstroms, with a sub-period of 1.54 angstroms for each of the double maxima. These distances approximately equal to the graphite lattice repeat distance (4.2612 angstroms) and carbon–carbon bond length (1.4204 angstroms). The rotational kinetic energy begins to peak as the translational kinetic energy decreases. This

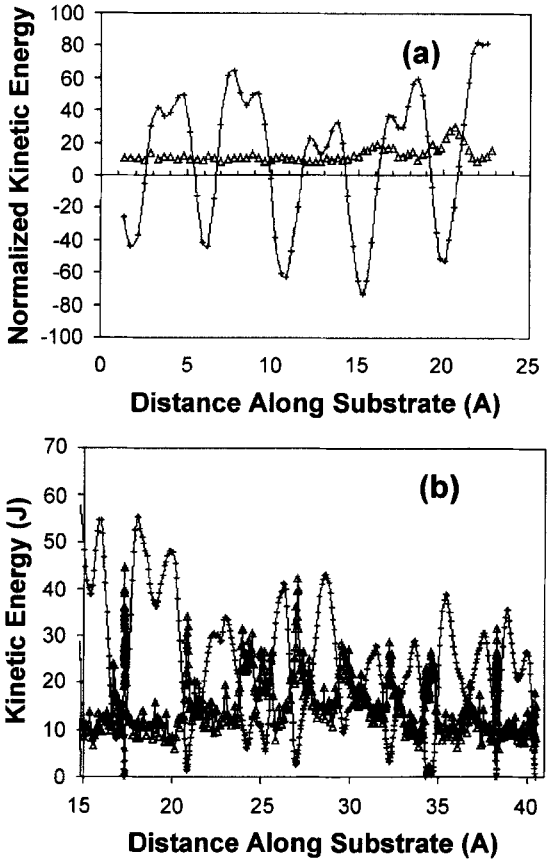

FIGURE 2 Normalized translational kinetic energy and rotational energy of the nanotube as a function of distance traveled.

indicates the start of a transition to rolling. Similar analysis for the out-of-registry case shows no significant periodic behavior.

Plotted in Figure 2b is the translational and kinetic energy *versus* substrate position for the in-registry nanotube from approximately 45 to 100 ps. Spikes occur in the rotational kinetic energy where transitions between rotation and translation occur. These spikes can be correlated with the structure of the substrate. During the sliding portion of the trajectory, the nanotube atoms are staggered with those in the substrate. This produces a relatively low-energy configuration. As the nanotube continues to move, atoms within the nanotube begin to collide with a row of atoms in the substrate. Rather than the nanotube and substrate atoms passing over one another, which is a relatively high-energy configuration, the nanotube rolls

until the atoms are again staggered, after which sliding is reinitiated. Energy loss, and hence friction, is greatest as the rotation is initiated. This is apparent by the dips in the translational energy in Figure 2b that correspond to the spikes in rotational energy. In contrast, the out-of-registry nanotube is not able to reach the low energy configuration in which substrate and nanotube atoms are completely staggered. Therefore the potential surface appears much smoother, there are no transitions to rotation, and the overall friction is lower.

CONCLUSIONS

Relative friction coefficients and mechanisms of energy loss for a (10,10) carbon nanotube with in-registry and out-of-registry orientations on a graphite substrate have been determined using molecular dynamics simulations. The simulations show a relative friction coefficient for out-of-registry orientations is approximately 80% of that for the in-registry orientations. Although with larger value, this is qualitatively consistent with recent experimentally observed phenomena, where values are observed ranging between 15% and 30% [2]. The greater friction coefficient for the in-registry case is due to a transition between sliding and rotation as atoms in the nanotube and substrate begin to pass over one another. Rotation for the in-registry case is consistent with experimental observations on multiwalled nanotubes [2], and with other theoretical analysis by Lu and Buldum [3]. Finally, we note that the general mechanism leading to alternating sliding and rolling is independent of tubule radius, and likely remains valid for multiwalled nanotubes. However, larger nanotubes will have larger contact areas and more degrees of freedom for energy dissipation. Both could conceivably lead to greater relative energy dissipation for in-registry *versus* out-of-registry orientations, and could account for the larger difference in frictional force between these two cases inferred from experiment.

Acknowledgements

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