A translational nanoactuator based on carbon nanoscrolls on substrates

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Inspired by recent experimental studies on the fabrication of carbon nanoscrolls (CNSs) on solid substrates, we perform theoretical study and molecular dynamics simulations to investigate the translational rolling/unrolling of a CNS on rigid substrate. We show that a substrate-supported CNS can be controlled to roll forward and backward by tuning its interlayer interaction energy via an external field. The typical energy release rate per unit area of rolling for such controllable and reversible linear motion is estimated to be in the range of $(-0.06)-0.08$ nN/nm, indicating promising applications as actuators and motors in nanomechanical systems. © *2010 American Institute of Physics.* [doi:10.1063/1.3302284]

The development of nanotechnology has brought the concept of mechanical actuation to a new stage in which synthesized nanoactuators are broadly applied in nanoelectromechanical systems such as nanomotors, nanorobots, and nanosensors.[1–4] The materials considered for these applications are typically carbon nanotubes (CNTs) or nanobelts. Here we propose a class of controllable nanoactuators based on substrate-supported carbon nanoscrolls (CNSs) whose motion can be controlled over a broad size range by an applied dc/ac electric field. As a member of carbon family, CNS has attracted significant interests in recent years.[5–8] Unlike CNTs which have tubular structure, a CNS is made of a continuous basal graphene sheet rolled up in a spiral form. Due to their unique topology, CNSs have shown outstanding structural,[9–13] dynamical,[10] and electronic[8,9,14,15] properties.

It has been shown that the equilibrium core size of a CNS depends on the length and bending modulus of the basal graphene sheet, as well as the interlayer spacing and interaction energy in the CNS structure.[16] Upon externally applied stimuli which typically decrease the interaction energy,[17] the CNS would adjust to a new equilibrium state with an expanded inner core. In this way, the core size of a CNS can be controlled by tuning the effective surface energy via an applied dc/ac electric field.[18] The previous studies, however, have been focused on systems with suspended CNSs. Inspired by recent experiments on the fabrication of CNSs on solid substrates,[8] here we develop a theory of linear actuator based on substrate-supported CNSs and conduct molecular dynamics simulations to demonstrate the principle of such linear actuation through controlled forward and backward rolling of a CNS on a graphite substrate as its surface energy is tuned by an applied dc/ac electric field.

Figure 1 displays some snapshots from molecular dynamics simulations of a linear nanoactuator based on a CNS supported on a graphite substrate. A multiwalled CNT (MWCNT) has been added to the system to aid the initial formation of the CNS as well as to constrain its inner core

size during linear motion. With fixed CNS-substrate interaction, the CNS would roll rightward if the CNS–CNS interaction is strong enough to overcome the CNS–graphite interaction as well as the contribution from elastic bending of CNS [Fig. 1(a)]. On the other hand, it would roll leftward if the CNS–CNS interaction is reduced by an applied electric field [Fig. 1(b)]. In this manner, the CNS is controlled to move rightward or leftward by tuning the CNS–CNS interaction energy.

To develop a theory for the CNS linear actuator shown in Fig. 1, we consider a graphene sheet of length B and width L rolled up into a CNS on a rigid substrate. The nanoscroll has a fixed inner core radius r_0, outer radius R, and interlayer spacing h [Fig. 1(c)] and can be rolled out into a flat graphene sheet along the substrate. In this process, the nanoscroll part of the structure can be described by a radial function as $r = r_0 + h/2\pi\vartheta$, where B, h, R, and r_0 are related as $\pi(R^2 - r_0^2) = (B-x)h$, where x denotes the length of the rolled out part of the graphene sheet on substrate. The elastic energy per unit area of CNS is taken to be $dW(r)/dA = (D/2) \times (1/r^2)$, where D is the bending modulus. Noting that $dA \approx Lr\,d\vartheta$, the total elastic energy in the CNS can be obtained as $W = \pi DL/h\ln(R/r_0)$. Our simulation results, to be
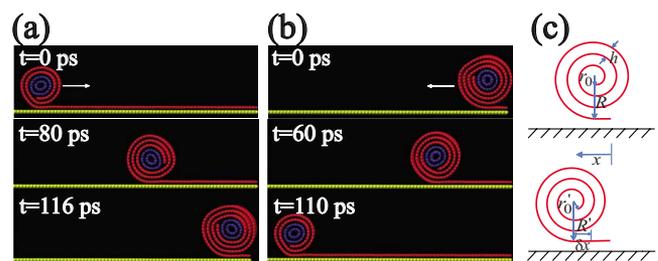
FIG. 1. (Color online) Snapshots of molecular dynamics simulation of CNS rolling along the graphite substrate. The tuning parameter for CNS–CNS interaction is (a) $\lambda_{CC} = 1.0$ and (b) $\lambda_{CC} = 0.6$. The tuning parameter for CNS–graphite interaction is $\lambda_{CS} = 0.8$ in both systems. To constrain the core size of CNS, multiwalled CNTs (MWCNTs) are inserted inside the core of CNS. (c) Schematic illustration of CNS on substrate. As the CNS rolls forward an infinitesimal displacement δx, the inner and outer radii of CNS change to r_0' and R' from r_0 and R, respectively. (enhanced online). [URL: http://dx.doi.org/10.1063/1.3302284.1]

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discussed in the next section, will show that a key to realizing the linear motion of CNS is to fix its inner core radius r_0 by inserting a CNT. Therefore, in the theory, we shall keep r_0 constant during the forward and backward rolling motion. As the CNS is unrolled by an infinitesimal distance δx, the outer radius R of the CNS would decrease by $2\pi R \delta R = -h \delta x$. The change in strain energy is then $\delta W = (\pi DL/h)(\delta R/R) = -DL\delta x/\{2[(B-x)h/\pi + r_0^2]\}$. At the same time, the total surface energy of the CNS is altered by $\delta \Gamma = L(\gamma_{CC} - \gamma_{CS})\delta x$, where γ_{CC} is the interlayer interaction energy of CNS and γ_{CS} is the interaction energy per unit area between the rolled out graphene sheet and substrate. Adding the contributions from elastic energy and surface energy, the change in total potential energy associated with an unrolling displacement δx is $\delta V/\delta x = \delta \Gamma/\delta x + \delta W/\delta x$. The energy release rate per unit area, corresponding to the energetic force associated with unrolling, is therefore

$$\frac{f}{L} = -\frac{1}{L}\frac{\delta V}{\delta x} = \gamma_{CS} - \gamma_{CC} + \frac{D}{2[(B-x)h/\pi + r_0^2]}. \quad (1)$$

The above equation shows that the net driving force can be either positive or negative, depending on the relative magnitudes of the self-affinity (γ_{CC}) of the graphene sheet and its affinity with the substrate (γ_{CS}). The equation for the translational motion can be derived from Lagrange's equation $[\partial(K-V)/\partial x] - d/dt\{[\partial(K-V)/\partial \dot{x}]\} = 0$, where K denotes the kinetic energy and V the potential energy of the system; t is the time and over-dot the time derivative. The potential energy can be derived as $V = \pi DL/h\ln(R/r_0) + L(\gamma_{CC} - \gamma_{CS})x + V_0$, where V_0 is a reference value independent of x and $\dot{x}$. To evaluate the kinetic energy in the system, we consider a rolling CNS as a rolling cylinder on a substrate with kinetic energy $K = 1/2M\dot{x}^2 + 1/2J_A\omega_A^2$, where $J_A = 1/2MR^2$ is the moment of inertia of the cylinder, $\omega_A = \dot{x}/R$ is the angular velocity and $M = \rho L(B-x) + M_0$ is the mass of the rolling CNS, ρ being the mass density of graphene and M_0 the mass of the constricting tube in the core. Combing all terms together, the Lagrange's equation of motion is derived as

$$6[\rho(B-x) + M_0/L]\ddot{x} - 3\rho\dot{x}^2 + 4(\gamma_{CC} - \gamma_{CS}) - 2D/[(B-x)h/\pi + r_0^2] = 0. \quad (2)$$

Given initial conditions and properties of the CNS and substrate, Eq. (2) can be integrated to predict the translational motion of the CNS.

To verify our theoretical model, we have conducted a series of MD simulations with bonded interaction among carbon atoms described by a Morse bond, a harmonic cosine of the bending angle and a twofold torsion potential.[19] The nonbonded van der Waals interactions are described by the Lennard–Jones (LJ) potential $U(r_{ij}, \lambda) = 4\lambda\varepsilon\{[(\sigma/r_{ij})]^{12} - [(\sigma/r_{ij})]^6\}$, where $\varepsilon = 0.3601$ kJ/mol, $\sigma = 0.34$ nm, and λ is a tuning parameter ($0 < \lambda \leq 1$) used to tune the strengths of CNS–CNS, CNS–CNT, and CNS–substrate interactions. For all simulations, the CNS–graphite and CNS–CNT interactions are fixed by setting the tuning parameter at 0.8 and 1.0, respectively. The CNS–CNS interactions, however, are tuned to different levels by setting the tuning parameter at $\lambda_{CC} = 0.5, 0.6, 0.7$ to unroll the CNS leftward, and $\lambda_{CC} = 0.9, 1.0$ to roll it rightward. (Experimentally, λ can be tuned by an applied electric field.[17]) Two layers of graphene with
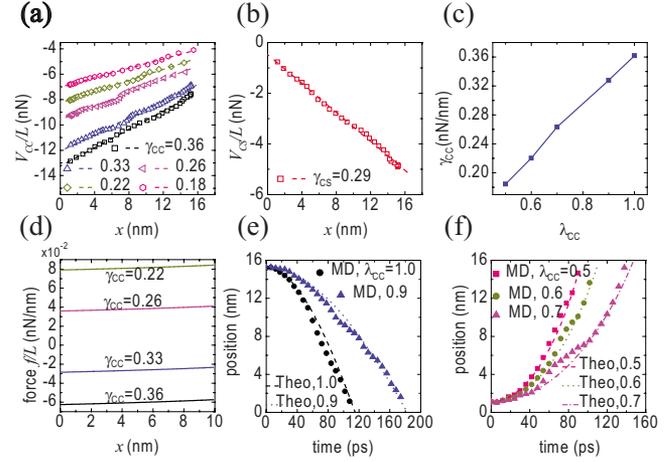
FIG. 2. (Color online) (a) The interaction energy of CNS–CNS as a function of the position of CNS. Five simulation results are shown with tuning parameters λ_{CC} at 1.0, 0.9, 0.7, 0.6, and 0.5, corresponding to different CNS surface energy 0.36 185, 0. 784, 0.2634, 0.22 024, and 0.18 452 nN/nm, respectively. (b) The interaction energy of CN32S–graphite as a function of the position of CNS with $\lambda_{CS} = 0.8$, corresponding to the CNS–graphite surface energy 0.28 557 nN/nm. (c) The surface energy of CNS γ_{CC} as a function of tuning parameter λ_{CC}. (d) The total force per unit length applied on CNS as a function of position of CNS for different levels of interaction. (e) Position of CNS as a function of time. Dashed lines show the theoretical results and scatters show the MD results. The CNS rolls rightward and (f) leftward with different CNS–CNS interactions.

dimension 31×3.8 nm^2 are used in the system. The bottom layer is fixed to model the graphite substrate and the top one is rolled up into a CNS with right end fixed. A MWCNT consisting of (5,5) and (10,10) SWCNTs is inserted into the core of CNS. Two initial configurations of partially rolled up CNSs are selected in the simulation: With $x = 15.2$ nm [Fig. 1(a)] and with $x = 1.1$ nm [Fig. 1(b)]. The initial velocities are set to be zero. The simulations are conducted using GROMACS4 (Ref. 20) with microcanonical (NVE) ensemble, with initial temperature 10 K and time step 2 fs.

To find the surface energy γ corresponding to different values of the tuning parameter λ, we plot the CNS–CNS interaction energies determined from MD simulations versus the rolling displacement of CNS. According to equation $\delta \Gamma = L(\gamma_{CC} - \gamma_{CS})\delta x$, the increment of CNS–CNS interaction energy $\delta \Gamma_{CC}$ is a linear function of the rolling displacement δx with a slope of $\gamma_{CC}L$. Therefore, the slope in Fig. 2(a) (dashed lines) corresponds to the surface energy. The CNS–substrate interaction energy is determined in a similar way, as shown in Fig. 2(b) for $\lambda_{CS} = 0.8$. It is shown in Fig. 2(c) that γ_{CC} and λ_{CC} obeys a linear relationship, which is consistent with the equation of LJ potential, where the interaction energy is linearly dependent on λ.

With the surface energy γ obtained, we calculate the energy release rate per unit area associated with the translational motion according to Eq. (1), with other parameters selected as $B = 31$ nm, $h = 0.34$ nm, $r_0 = 0.8$ nm, and $D = 0.11$ nN nm.[16,21] Figure 2(d) shows the variation in the force with CNS rolling along the substrate. Interestingly, in the present simulation system, the force only shows small variations as the CNS rolls forward or backward. According to Eq. (1), the variations are mainly due to the elastic bending part $D/2\{[(B-x)h/\pi] + r_0^2\}$, indicating that the elastic energy makes only minor contributions to the total force. We note that the energy release rate per unit area is on the order of -0.06 nN/nm for $\lambda_{CS} = 0.8$ and $\lambda_{CC} = 1.0$, and 0.08 nN/nm
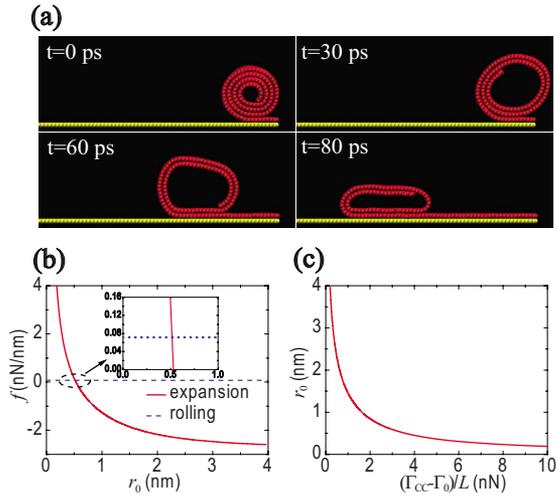
FIG. 3. (Color online) (a) Snapshots of molecular dynamics simulation of CNS expanding and rolling on the graphite substrate. The tuning parameter for CNS–CNS and CNS–graphite interactions are $\lambda_{CC}=0.6$ and $\lambda_{CS}=0.8$. (b) Competing core expansion and rolling motion of a CNS on substrate. The energy release rate associated with core expansion of a CNS is seen to easily dominate over that associated with the CNS rolling on a substrate as the system is perturbed away far from equilibrium by an external field. (c) The equilibrium core size as a function of the surface energy of CNS (after Ref. 16). The parameters are selected as $B=31$ nm, $h=0.34$ nm, $D=0.11$ nN nm, $\lambda_{CC}=0.6$, and $\lambda_{CS}=0.8$.

for $\lambda_{CS}=0.8$ and $\lambda_{CC}=0.6$. Therefore, the range of driving force is about $(-0.6)-0.8$ nN for a CNS-based nanoactuator with width 10 nm, and $(-6)-8$ nN with width 100 nm.

To further investigate the linear motion of CNS on substrate, we numerically solve Eq. (2), with additional parameters set as $\rho=12\times1.660\,54\times10^{-27}/(3\sqrt{3}/4\times0.142^2\times10^{-18})$ kg/m^2, $M_0=960\times12\times1.66054\times10^{-27}$ kg, and $L=3.8$ nm. The predicted motion of CNS for different tuning parameters, as shown in Figs. 2(e) and 2(f) (dashed lines), agrees well with the corresponding MD results (scatters), indicating that our theoretical model is capable of predicting the rolling motion of CNS on substrate.

We emphasize that constraining the core size of the CNS is essential in realizing the controlled translational motion of CNS on substrate. To demonstrate this, we repeat the simulation without any insertion into CNS. The results are shown in Fig. 3(a). It is seen that the CNS does not unroll leftward as the CNS–CNS interaction decreases. Instead it remains at the initial position while expanding its core [Fig. 3(a), $t=30$ ps]. To reveal the underlying mechanism, we compare the energy release rates associated with the rolling and expansion of CNS. The energy release rate for expansion of CNS has been obtained in Ref. 16 as $f_{expansion}=\pi D(R^2-r_0^2)/(hr_0R^2)-2\pi\gamma_{CC}(1+r_0/R)$, and that for rolling of CNS is given by Eq. (1). These energy release rates are plotted as a function of the core radius in Fig. 3(b) with typical parameters for the CNS on graphite. Figure 3(c) plots the core radius as a function of surface energy of CNS calculated in Ref. 16. As an external field is applied, the surface energy decreases while the equilibrium core radius increases. In this case, Fig. 3(b) shows that there is a tendency for the CNS core to expand toward the new equilibrium size, with energy release rate orders of magnitude higher than that associated with the rolling motion. Therefore, when an external field is

applied to perturb the equilibrium state of CNS, the dominant mode of response of CNS is core expansion. As the inner core r_0 increases, the energy release rate $f_{expansion}$ associated with core expansion drops rapidly, and eventually the rolling motion is initiated for further energy reduction. As shown in Fig. 3(a), the rolling motion starts only after substantial expansion of the inner core ($t=60$ ps), at which the shape of CNS has changed so much that the radial function can no longer be used to describe the elastic energy in the system. As the number of layers is reduced by core expansion, the CNS collapses on the substrate at $t=80$ ps. In this situation, the motion of CNS is rather complicated and unpredictable. We thus conclude that the core size of CNS must be fully constrained in order to design a controllable CNS-based nanoactuator.

In conclusion, a controllable translational nanoactuator based on the rolling and unrolling motion of CNSs on a substrate has been proposed and investigated by both theoretical modeling and molecular dynamics simulations. It is found that the theoretical model based on the classical Lagrangian dynamics is fully capable of predicting molecular dynamics simulation results. It is suggested that CNS based linear nanoactuator can be controlled by tuning the effective surface energy of the system via an applied dc/ac electric field as well as adjusting the width of CNS. These results suggest promising applications of CNSs as linear actuators and motors in nanomechanical systems.

[1] Y. N. Gartstein, A. A. Zakhidov, and R. H. Baughman, Phys. Rev. Lett. **89**, 045503 (2002).
[2] M. H. Zhao, Z. L. Wang, and S. X. Mao, Nano Lett. **4**, 587 (2004).
[3] R. Lefèvre, M. F. Goffman, V. Derycke, C. Miko, L. Forro, J. P. Bourgoin, and P. Hesto, Phys. Rev. Lett. **95**, 185504 (2005).
[4] W. H. Lin and Y. P. Zhao, Chaos Soliton. Fractals **23**, 1777 (2005).
[5] H. Shioyama and T. Akita, Carbon **41**, 179 (2003).
[6] L. M. Viculis, J. J. Mack, and R. B. Kaner, Science **299**, 1361 (2003).
[7] M. V. Savoskin, V. N. Mochalin, A. P. Yaroshenko, N. I. Lazareva, T. E. Konstantinova, I. V. Barsukov, and I. G. Prokofiev, Carbon **45**, 2797 (2007).
[8] X. Xie, L. Ju, X. F. Feng, Y. H. Sun, R. F. Zhou, K. Liu, S. S. Fan, Q. Q. Li, and K. L. Jiang, Nano Lett. **9**, 2565 (2009).
[9] Y. Chen, J. Lu, and Z. Gao, J. Phys. Chem. C **111**, 1625 (2007).
[10] S. F. Braga, V. R. Coluci, S. B. Legoas, R. Giro, D. S. Galvão, and R. H. Baughman, Nano Lett. **4**, 881 (2004).
[11] S. F. Braga, V. R. Coluci, R. H. Baughman, and D. S. Galvão, Chem. Phys. Lett. **441**, 78 (2007).
[12] V. R. Coluci, S. F. Braga, R. H. Baughman, and D. S. Galvão, Phys. Rev. B **75**, 125404 (2007).
[13] G. Mpourmpakis, E. Tylianakis, and G. E. Froudakis, Nano Lett. **7**, 1893 (2007).
[14] H. Pan, Y. Feng, and J. Lin, Phys. Rev. B **72**, 085415 (2005).
[15] R. Rurali, V. R. Coluci, and D. S. Galvão, Phys. Rev. B **74**, 085414 (2006).
[16] X. H. Shi, N. M. Pugno, and H. J. Gao, "Tunable core size of carbon nanoscrolls," J. Comp. Theo. Nanosci. (in press).
[17] X. H. Shi, Y. Cheng, N. M. Pugno, and H. J. Gao "Tunable water channels with carbon nanoscrolls" (unpublished).
[18] X. H. Shi, N. M. Pugno, Y. Cheng, and H. J. Gao, Appl. Phys. Lett. **95**, 163113 (2009).
[19] J. H. Walther, R. Jaffe, T. Halicioglu, and P. Koumoutsakos, J. Phys. Chem. B **105**, 9980 (2001).
[20] B. Hess, C. Kutzner, D. van der Spoel, and E. Lindahl, J. Chem. Theory Comput. **4**, 435 (2008).
[21] D. W. Brenner, O. A. Shenderova, J. A. Harrison, S. J. Stuart, B. Ni, and S. B. Sinnott, J. Phys.: Condens. Matter **14**, 783 (2002).