Thermal-Induced Edge Barriers and Forces in Interlayer Interaction of Concentric Carbon Nanotubes

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Thermal-induced edge barriers and forces in interlayer interaction of concentric carbon nanotubes

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Molecular dynamics simulations reveal that thermal-induced edge barriers and forces can govern interlayer interaction of double walled carbon nanotubes. As a result, friction in such systems depends on both the area of contact and the length of contact edges. The latter effect is negligible in macroscopic friction and provides a feasible explanation for the seemingly contradictory laws of interlayer friction reported in the literature. The temperature-dependent edge forces can be utilized as a driving force in carbon nanotube thermal actuators, and has general implications on nanoscale friction and contact.

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Friction between two surfaces in contact is one of the most common processes in nature. From the atomic point of view, the origin of friction is related to irreversible energy transfers from translational kinetic energy into internal energy, irrespective of the length scale of investigation. However, frictional laws at the nanoscale are often found to be distinct from those at the macroscale, which has resulted in a new research field referred to as nanotribology.

At the macroscale, the friction force is generally regarded as proportional to the normal force or the real contact area [1, 2]. In contrast, the nanoscale friction force can be linearly or sub-linearly dependent on the normal load [3-8]. Understanding friction at the nanoscale remains a key challenge in tribology [9-11]. Some researchers concluded that continuum mechanics theories of friction break down at the nanoscale [3-6, 12], while others showed that these theories can still be applicable with appropriate care [8]. Whether and how continuum mechanics breaks down at nanoscale and what should be modified are still interesting questions open to debate. In particular, it is of importance to know whether, what, and why new effects arise at such a small scale.

Owing to their coaxial structure and nearly perfect lattice, multiwalled carbon nanotubes (MWCNTs) are ideal materials available for investigating underlying physics involved in nanoscale friction [13-22]. However, contradicting friction laws have been observed. While some studies [14] showed that dynamic interlayer friction is related to the contact area between tubes, others [17] found that friction is independent of the contact area and determined only by the dimensions of tube ends. Clearly, a better understanding of nanoscale friction laws is needed to explain such apparent contradictions.

In this letter, we focus on the edge effect on the interlayer interactions of a commensurate armchair double walled CNT (DWCNT). We show by molecular dynamics simulations that, when one tube end contacts with the wall surface of another tube, an edge barrier can be induced in the van der Waals (vdW) potential field along the surface. The existence of such edge barrier will lead to energy dissipation when the two tubes slide with respect to each other, and induce a resistance to the relative motion. This finding indicates a significant cause for the breakdown of continuum theories of friction at very small scales, and suggests a novel nanoscale friction law. Moreover, we will demonstrate that the edge barrier results in an edge force which can be utilized as a driving force for CNT thermal actuators. Our findings are therefore helpful to both the understanding of nanoscale friction and the design of thermal actuators.

All simulations [23] are carried out using classical molecular dynamics based on Brenner potential [24], as in our previous studies [25, 26]. Unless otherwise specified, a typical system of (15, 15)/(10, 10) DWCNT with a long inner tube (length L = 20 nm) and a short outer tube (length l = 7.3 nm) is studied.

To provide an atomic level insight into the interactions between the inner and outer tubes, we show in Fig. 1a the longitudinal distribution of the vdW potential field of the inner tube [23]. It is seen clearly that the presence of a short outer tube modifies the vdW potential field of the inner tube: a potential well is formed in the region where the outer tube is present, and considerable potential gradients exist in the sections near both ends of the outer tube. This is somewhat like a heavy object sitting on a soft substrate (see the inset of Fig. 1a): the substrate surface deforms and a gravitational potential well is generated. The well depth (edge barrier) should be closely related to energy dissipation

when the object is sliding on the substrate. Analogously, when the outer tube is sliding along the inner one, the vdW potential barrier could lead to extra energy dissipation and thus induces friction. That is to say, the dynamic friction between two tubes depends not only on the contact area but also on the edge barrier.

To give a quantitative assessment of the edge effect on the interlayer friction, we perform calculations [23] of the above system in which the outer tube slides along the inner tube at a center of mass velocity of 400 m/s. Our calculations show that the friction on the edge atoms (of the last two rings of both ends) at 100, 200 and 300 K contributes respectively 23%, 50% and 74% to the total friction. The average friction on edge atoms increases with temperature (Fig. 1b). This can be explained by the fact that the edge barrier is an increasing function of temperature (Fig. 1c). However, the friction on inner atoms shows a decreasing temperature dependence. At room temperature, the friction on an edge atom (0.023 pN/atom) may be 20 times of that on an inner atom (0.0012 pN/atom). It is clear that the shorter the tube, the less the contact area, and the stronger the edge effect. Although it is difficult to make a quantitatively accurate prediction of the interlayer friction in CNTs as the choices of both thermostat [27] and interlayer potential [18, 22] may influence the calculations, the interlayer friction we calculated at room temperature is in good agreement with the experimentally measured value (below 0.0014 pN/atom) by Kis et al. [19].

The existence of a temperature-dependent edge barrier could provide a fundamental understanding for seemingly contradictory laws of interlayer friction in MWCNTs reported in the literature. At low temperatures, the edge barrier is shallow, and thus friction is mainly caused by the interlayer shear strength and hence related to the contact

area, as shown by Guo et al. [14]. However, at high temperatures, the edge barrier becomes considerably deep and thus plays a dominant role in energy dissipation, which consequently leads to a tube end dependent friction, as reported by Tangney et al. [17]. Neither shear strength nor end effect alone can reflect the true mechanism of nanoscale friction.

There is no obvious reason to refute that similar edge barrier should exist whenever two nanoscale surfaces contact with each other. In this sense, our finding suggests a novel nanoscale friction law: nanoscale friction is related to both contact area and contact edges. The edge effect is negligible in macroscopic friction because of the small edge-to-area ratio, while at the nanoscale, it can play a dominant role due to large edge-to-area ratios, which may cause a breakdown of continuum mechanics friction theories in which only the contribution of contact area is considered.

What is the origin of the edge barrier? To answer this question, consider a system made of a long inner tube and a short outer tube. Suppose the system temperature is zero, we know that all atoms of the inner tube are in their equilibrium positions, and the vdW potential field of the inner tube is flat (with appropriate corrugations) along the axial direction. However, when the inner tube temperature is nonzero, all atoms vibrate around their equilibrium positions, leading to a change in the vdW potential field of the inner tube. The thermal vibration of inner tube atoms confined inside the outer tube should be different from that of inner tube atoms outside the confinement, since the atoms inside the outer tube are more constrained. Meanwhile, the intensity of the vdW potential field of the inner tube at a point is mainly determined by the inner tube atoms nearest to the point. For a point on the inner surface of the outer tube, all of its nearest neighbor atoms

on the inner tube are inside the outer tube. However, for a point at the end of the outer tube, one part of its nearest neighbor atoms on the inner tube lies inside the outer tube, while the other part does not. As a result, the vdW potential between the end atoms of the outer tube and the inner tube is different from that between the inner atoms of the outer tube and the inner tube. Potential gradients (edge barriers) therefore exist near the outer tube ends. This means that the edge barrier is indeed induced by the thermal vibrations. The results shown in Fig. 1c essentially substantiate this speculation.

According to the principle of virtual work, the edge barrier will result in an edge force on the outer tube end. This is confirmed by our simulations [23]. The longitudinal distribution of the interlayer vdW force exerted on the outer tube by the inner tube is shown in Fig. 2a. The forces exerted on the edge atoms are significantly larger than those on the inner atoms, indicating the existence of an edge force, F_e. We can see also that the edge force is toward to the center of the short tube, indicating there is a pinch force (produced by the two edge forces) exerted the short tube by the long tube.

The longitudinal force exerted on an extended layer by a truncated layer has been extensively studied in the context of MWCNTs, and it has been shown that the truncated layer may provide an attractive force, F_a, on the extended layer [13, 28-30]. This indicates that a longitudinal compressive force is exerted on the contact part of the long tube by the short tube in a DWCNT. Therefore, we can see that, when a short tube is present on a long tube, both the short tube and the contact part of the long tube are compressed. We note that this does not conflict with Newton's third law, because F_e and F_a are not action and reaction forces (Fig. 2b). The origins of F_e and F_a are quite different. Unlike F_a, which is mainly induced by the direct interlayer vdW interactions, F_e comes

from a coupling effect of the vdW interactions and thermal fluctuations. When the system temperature is zero, F_a still exists but F_e vanishes. At room temperature, our simulations show that F_e (about $0.015d$ nN/nm, where d is the outer tube diameter) is approximately 50 times lower than F_a (about $0.8d$ nN/nm [29, 30]). Like the edge barrier, F_e too exhibits a linear dependence on the inner tube temperature, but independent of the outer tube temperature, as shown in Fig. 2c.

Recently, thermal gradient on a CNT was experimentally [31] and numerically [32-35] found to be capable of driving nanoscale objects to move along the tube. Since there is a thermal gradient along the CNT, a fundamental question is whether the edge force contributes significantly to the thermophoretic force. We consider a DWCNT (with L = 24 nm and l = 7.3 nm) in which a thermal gradient is imposed on the inner tube. Our simulations [23] indicate that the unbalanced pinch force (acting only on atoms of last two rings of the outer tube ends) contributes approximately 40 percent of the total longitudinal thermophoretic force, while the usual gradient force produced by the thermal gradient along the inner tube (acting on all the other atoms of the outer tube) contributes 60 percent. This means that, although the edge barrier leads to energy dissipation when the two layers slide with respect to each other, the edge force provided by the edge barrier can act as a driving force in thermal gradient driven nanodevices.

In summary, edge effect in the interfacial interactions of nanoscale contact is investigated using double walled carbon nanotubes. We demonstrate that there exist an edge barrier and an edge force when the edge of one layer contacts another layer. Both the edge barrier and the edge force are due to a coupling effect between van der Waals interactions and atomic thermal vibrations, and are linearly dependent on the temperature.

The existence of the edge barrier indicates that nanoscale friction is related to both contact area and contact edges; the latter effect is negligible in macroscopic friction and unique to nanoscale contact. The edge force may be used as a driving force for nanoscale thermal actuators.

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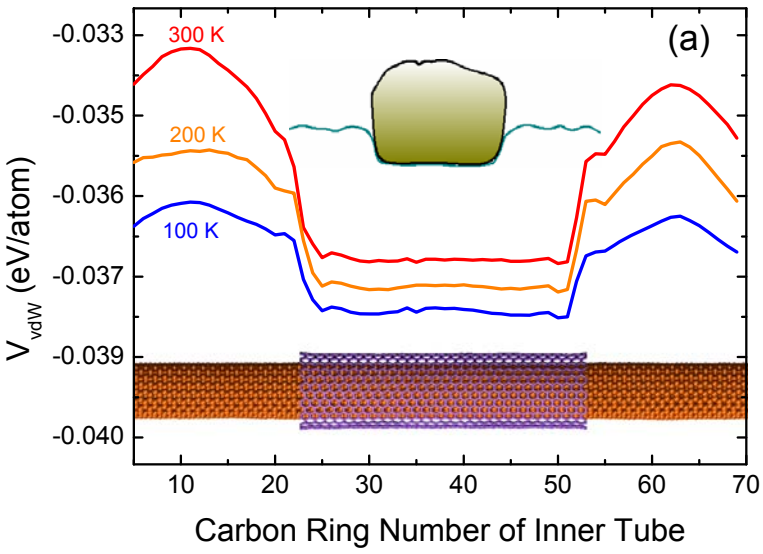
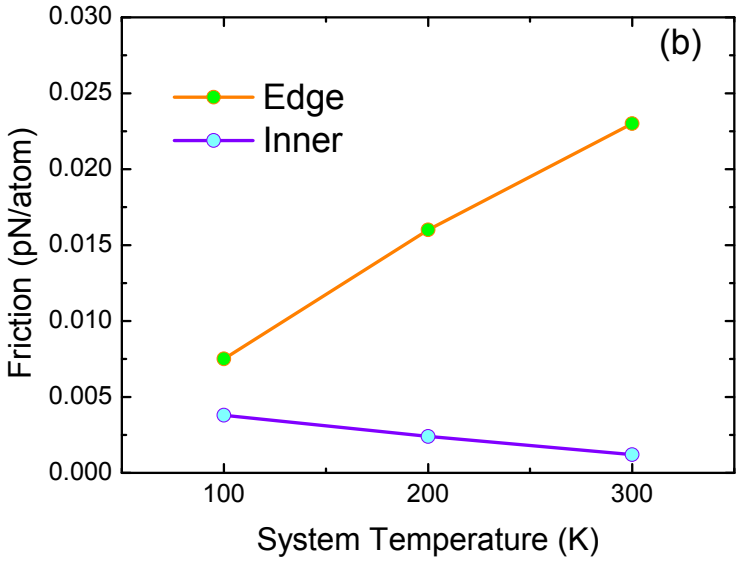
Figure Captions

Figure 1 (a) van der Waals potential between an outer atom and the inner tube. A potential well is induced by the outer tube. (b) Friction force versus system temperature when the outer tube slides at a velocity of 400 m/s along the inner tube. (c) Linear dependence of the potential well depth on the system temperature.

Figure 2 (a) Typical longitudinal distribution of the van der Waals force exerted on the outer tube by the inner tube. Significantly large edge forces can be observed. (b) Schematic illustration of interlayer interactions between two solid surfaces in contact. Both the truncated layer and the contact region of the extended layer are compressed. Arrows indicate forces between atoms. (c) Dependence of the edge force on the inner and outer tube temperature.

Fig. 1


(a)
300 K
200 K
100 K
-0.033
-0.035
-0.036
-0.037
-0.039
-0.040
V_{vdW} (eV/atom)
10
20
30
40
50
60
70
Carbon Ring Number of Inner Tube


(b)
Edge
Inner
0.030
0.025
0.020
0.015
0.010
0.005
0.000
Friction (pN/atom)
100
200
300
System Temperature (K)

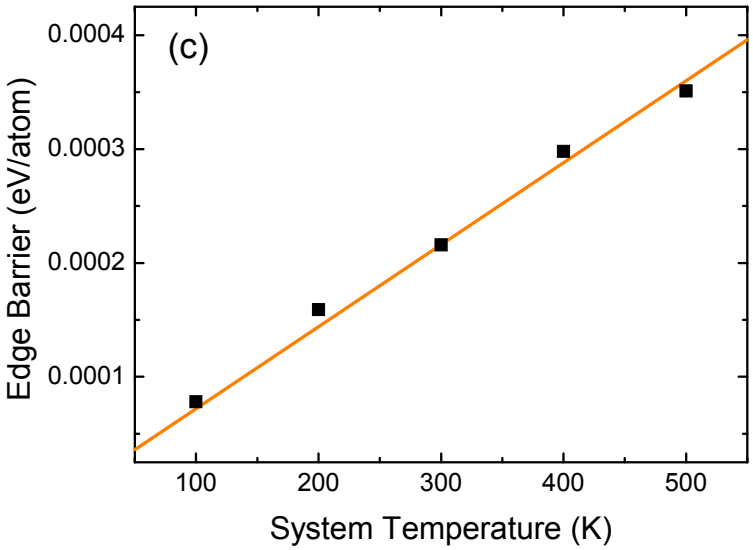
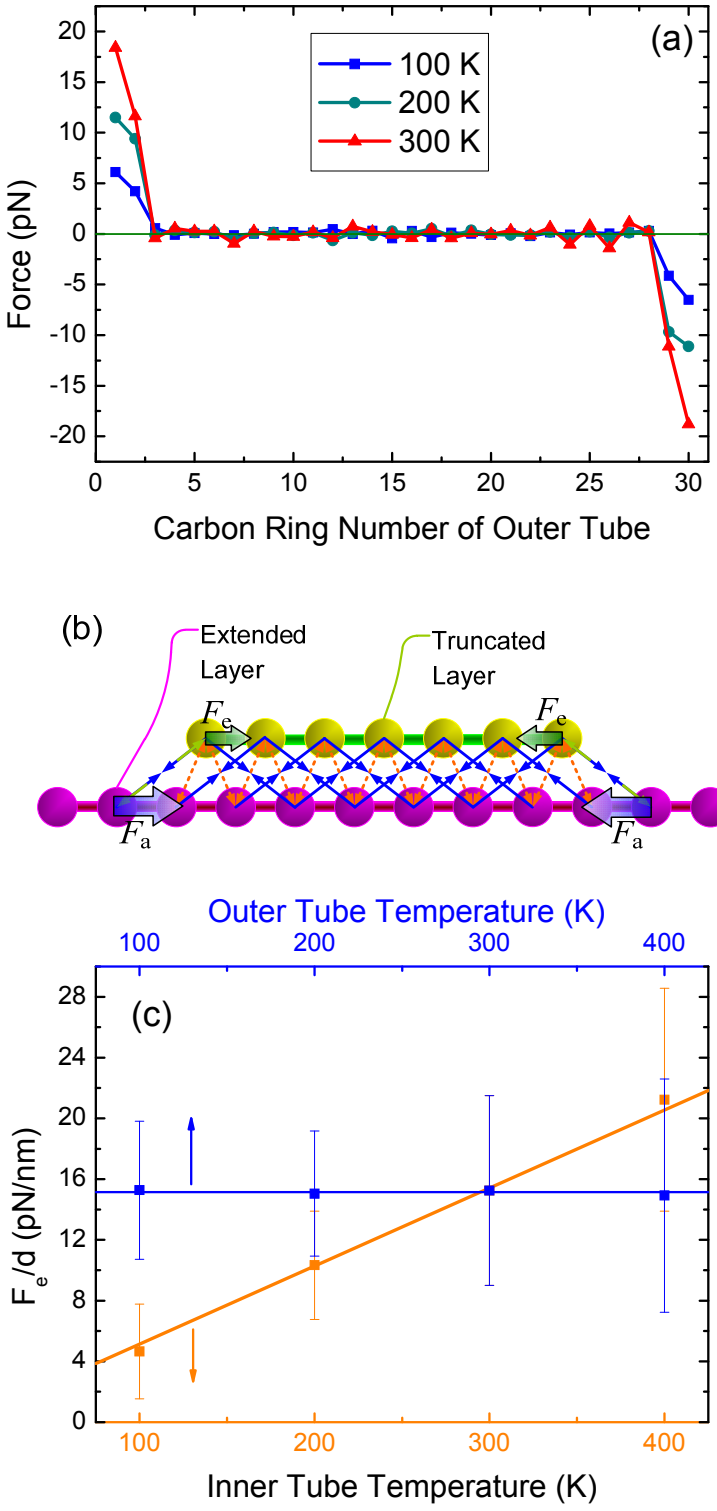

(c)
0.0004
0.0003
0.0002
0.0001
Edge Barrier (eV/atom)
100
200
300
400
500
System Temperature (K)

Fig.2


(a)
100 K
200 K
300 K
Force (pN)
Carbon Ring Number of Outer Tube
(b)
Extended Layer
Truncated Layer
F_e
F_a
(c)
Outer Tube Temperature (K)
F_e/d (pN/nm)
Inner Tube Temperature (K)