Applied Physics Letters

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Cite as: Appl. Phys. Lett. **98**, 063101 (2011); https://doi.org/10.1063/1.3551574
Submitted: 24 September 2010 . Accepted: 10 December 2010 . Published Online: 07 February 2011

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Dynamic ripples in single layer graphene

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(Received 24 September 2010; accepted 10 December 2010; published online 7 February 2011)

Similar to the water wave on a pond caused by dropping a pebble, the formation of ripples is shown in a single layer graphene (SLG) when it is stroked by a C60 molecule, no matter whether the graphene is plane or corrugated. The controllable ripple in SLG is investigated by molecular dynamics simulations. Noticeable diffraction and interference of the ripples are observed. This study indicates that the ripple propagation in graphene can be used to detect defects. © *2011 American Institute of Physics*. [doi:10.1063/1.3551574]

Since Novoselov *et al.* fabricated the isolated graphene via mechanical exfoliation of graphite,[1] graphene has arose intense research interest due to its distinctive and outstanding properties such as quantum electronic transport,[2,3] superior electronic structure,[4–6] extremely high elasticity,[7,8] and large thermal conductivity.[9–11] Those properties, which depend on the unique structures of two dimensional (2D) carbon atoms sheets, are crucial for graphene application.[12,13] Recently, it has been found that a freestanding isolated graphene layer can be intrinsically corrugated to produce ripples, which are expected to affect its conductivity.[14] Another approach to create periodic ripples into graphene has been reported by Bao and colleagues by utilizing the negative expansion coefficient of graphene sheet.[15] The formation of ripple, which is related to the periodic variation of electronic properties of graphene, is expected to be used for synthesizing graphene based electronic devices.[16]

At present, there are two different understandings on the formation of ripples in graphene. Meyer and co-workers revealed that random ripples could intrinsically across the freely suspended graphene sheets.[14] On the other hand, Kim and Castro Neto provided another theoretical explanation that graphene behaves as an electronic membrane, in which the ripples can also be induced by changing the electrochemical potentials.[17]

Here, we report a molecular dynamics (MD) simulation of ripple propagation in graphene. In the simulation, a C60 is used to strike a single layer graphene (SLG) to initiate ripple formation. To mimic the classical single or double slit experiments, we also demonstrate the diffraction and interference of ripples in graphene.

In the MD simulation, the second-generation reactive empirical bond order (REBO) potential is used to describe the C–C interaction,[18,19] and the Lennard-Jones 12-6 potential is used to calculate the long range van der Waals interaction. It is important to note that the ultrathin 2D graphene sheet [its effective thickness is only ~0.088 nm (Ref. 10)] is the medium of ripple propagation. Graphenes of two different sizes are studied. One graphene sheet has a size of 197 Å (zigzag direction) × 169 Å (armchair direction) (containing 12 880 atoms). The size of another is 254 Å (armchair direction) × 153 Å (zigzag direction) (containing 14 040 atoms). In order to observe the wave diffraction and interference, two symmetrical slits with a width of 3 Å are created along a centerline of the latter graphene (all C atoms on the centerline, except those that belong to the slits, are fixed during the simulation). Initially, a C60 molecule is placed 6.5 Å above the graphene. At time zero, the C60 starts to impact the SLG with an initial vertical velocity of 15 Å/ps. The time step of the MD simulation is 1 fs and each trajectory runs for 10 ps.

A snapshot of circular ripples propagating in the SLG during the simulation is shown in Fig. 1(b). It is interesting that these circular ripples are very similar to the water waves stirred by dropping a pebble into a pond [Fig. 1(a)]. Different from the water wave in a pond, the ripple in SLG is arisen by distortion of C–C bonds. As vibration energy is passing through the graphene from the impacting center to another place, the ripples are formed. The ripple propagation in the SLG actually means that the "strong" impact energy is transformed into the "soft" wave energy, illustrating that the graphene can be used as an energy buffer to withstand shocks.

The onset of the graphene ripple and its propagation in the SLG are demonstrated in Fig. 2(a). The z-direction displacements (Δz) of atoms on the centerline versus time curves [Fig. 2(b)] are used to illustrate the transverse waves. At 800 fs, the displacement of the atoms at the impacting

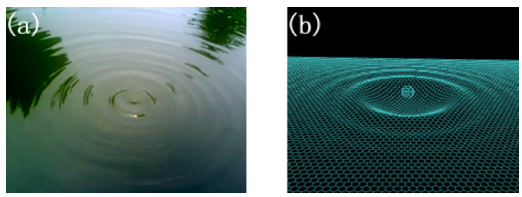

FIG. 1. (Color online) Snapshots of water wave and graphene ripple. (a) Water waves are generated by means of throwing a pebble to the static water pond (Nikon coolpix L22); (b) graphene ripples are generated on the graphene which is stroked by an energic C60 molecule (enhanced online). [URL: http://dx.doi.org/10.1063/1.3551574.1]

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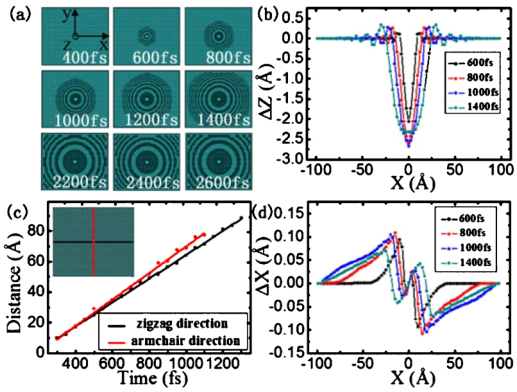

FIG. 2. (Color online) (a) The onset of the graphene ripple generated by striking with C60 molecule and its propagation on the surface of the graphene. The original ripples and the reflective ripples arouse interference pattern in the last three snapshots. (b) and (d) show the z-direction displacements (Δz) and x-direction displacements (Δx) of atoms along the centerline [colored black in (c)], respectively. (c) The traveling distance of the ripples along different directions (enhanced online). [URL: http://dx.doi.org/10.1063/1.3551574.2]

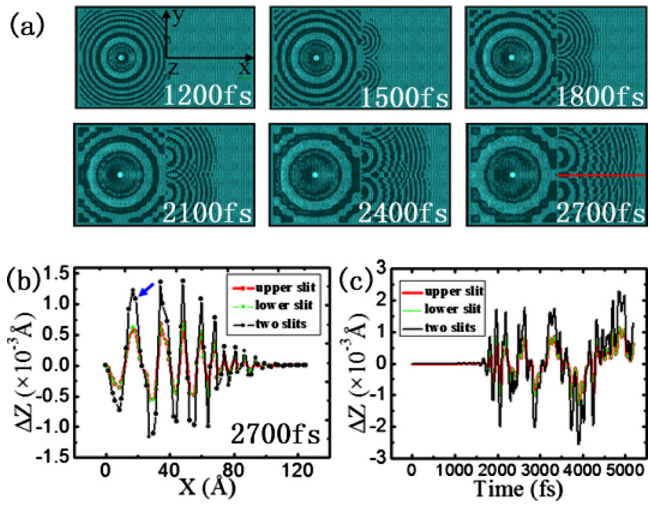

FIG. 3. (Color online) (a) Snapshots of interference of ripples coming from two slits. (b) The z-direction displacements (Δz) of atoms on the centerline [indicated in (a)]; (c) the z-direction displacements of the atom pointed by an arrow in (b) at different time (enhanced online). The red and green lines indicate the displacements caused by the diffraction wave when the two slits are closed, respectively, while the black line indicates the displacement caused by the diffraction wave when the two slits are both open (enhanced online). [URL: http://dx.doi.org/10.1063/1.3551574.3]

point is about 2.6 Å. The amplitude of the nearest wave crest of the impacting point is 0.26 Å. The amplitude of the wave decays as the following exponential function:

$$A(x) = 2.599\,43 \exp(-|x|/7.339\,93) - 0.013\,51,$$

where x (in angstrom) is the horizontal position and $A(x)$ (in angstrom) is the corresponding amplitude. The distances of ripple propagation at different time are shown in Fig. 2(c). The average propagating speed of ripples along the zigzag direction is 78.32 Å/ps, while the propagating speed of ripples along the armchair direction is 87.02 Å/ps. The small difference between the two displacement lines results in a hexagonal symmetry of the ripples rather than complete circles [as shown in Fig. 2(a)]. Meyer *et al.* mentioned that, under the condition of thermal equilibrium, ripples in freestanding graphene are intrinsic, random, and uncontrollable.[14] However, this study shows that, by C60 striking, the ripple forming on the surface of SLG is reproducible and controllable. For example, the amplitude of ripple can be easily tuned by varying the impacting speed of the C60.

The x-direction displacements (Δx) of atoms along the centerline are shown in Fig. 2(d). The atoms near the zero point do not move along the x-direction. With the increase of distance from the impacting point, Δx fluctuates between positive and negative. By comparing Δx with Δz, it is found that the z-direction displacement of atoms is much larger than the x-direction displacement. Graphene shrinks as any other 2D-membranes due to transverse bending-vibrations. Muñoz *et al.* discussed the ballistic heat conductivity of graphene and showed that the soft bending mode dominated the graphene thermal conductivity at low temperature.[10] This is in agreement with our simulation that propagation of the transverse wave is free in the graphene, which means that the free path of bending mode phonons is very large.

When the ripples reach a narrow slit, the atoms in the slit vibrate like a point source. Noticeable diffraction is shown in Fig. 3(a). The diffraction wave resembles a circular ripple with the slit as its center. Our results indicate that this mechanical ripple can spread out along the narrow slit (one atom distance), which would provide a possible technique to detect defects in graphene. For example, if there is a vacancy defect in the graphene, the shape of ripple would be different due to the wave diffraction at the vacancy defect. When the ripple arrives at the vacancy point, the smooth circle ripple at the defect point would form a small crater or even be separated into two parts, which is dependent on the size and shape of the vacancy. If lots of vacancy defects exist, the circle ripple would become quite unsmooth with some concaves. Therefore, we can determine the existence of vacancy defect by observing the shape of ripple.

When the two diffraction waves encounter in the double slit interference experiment, interference is generated [shown in Fig. 3(a)]. Figure 3(b) shows the displacements of a string of atoms along the centerline at 2700 fs. The two point sources of the diffraction waves are also bilaterally symmetrical due to two localized bilaterally symmetrical slits. When the two slits are closed, respectively, it is found that the two displacement curves would almost fully overlap. However, once the two slits are both open, the resultant displacement is equal to the sum of the above two displacements, which accords with the principle of superposition of waves. In this way, the vibrations of all the atoms along the centerline are strengthened. Then, a single atom is picked from the central row of atoms in order to study the change of displacement as a function of time [shown in Fig. 3(c)]. It is found that the displacement-time curves of the single atom are also corresponded to the above superposition principle. The above phenomena indicate that ripples propagating in a 2D single atom thick media can produce interference. We can use this concept of "superposition of waves" to design a "lens" that focuses wave energy on the SLG sheet, which would open an exciting possibility of the fabrication of nanodevices (e.g., signal transducer).

The above mentioned graphenes are perfect 2D crystals which are not stable according to both theory and experiment.[14,20,21] There should be intrinsic ripples at finite temperatures due to its thermodynamic stability. When a

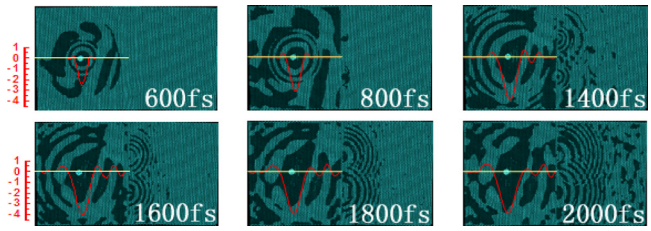

FIG. 4. (Color online) The propagation, diffraction, and interference of impacting ripples in the corrugated graphene. The inset curve of each graph is the z-direction displacements of atoms on the centerline. Tick labels on the left are in units of angstrom.

graphene with intrinsic ripples is stroked by a C60 molecule, ripples also exist at the impacting point and propagate outside as shown in Fig. 4. It is found that impacting ripples on the plane graphene are different from those on the corrugated graphene. On one hand, the intrinsic ripples would inevitably affect the impacting ripples. The bending and stretching of the graphene layer result in the intrinsic ripples which are distributed randomly and uncontrollably. Therefore, the graphene surface is no longer plane and symmetric. During the propagating process, the impacting ripples vary with the different directions. On the other hand, the impacting ripples would affect the intrinsic ripples in turn. The propagation of impacting ripples breaks the original stability of graphene and thus changes the intrinsic ripples. As shown in Fig. 4, there are many cloudlike shades outside the impacting ripples, which means that the amplitudes of intrinsic ripples in these areas have decreased. When the impacting ripples arrive at a slit, diffraction still occurs and brings cloudlike shades. However, interference of diffraction ripples coming from the two slits is not as clear as that of plane graphene. Although the intrinsic ripples disturb the impacting ripples, the signal carried by the impacting ripples does not fade away but still propagates forward. Intrinsic ripples in graphene are expected to strongly influence its electronic properties by introducing spatially varying potentials or effective magnetic fields.[22,23] Different from intrinsic ripples, the impacting ripples made from C60 will disappear after the system is fully relaxed. However, the impacting ripple has its own excellent feature such as reducing local excessive deformation, transferring signals caused by striking, and detecting cracks or defects.

In this study, nanoscale dynamic ripples have been observed on the graphene no matter whether the graphene is plane or corrugated, although there are lots of differences between them. A graphene sheet may be a well promising material to be served as an energy buffer to withstand shocks. The propagation orientation and amplitude of ripples can be controlled via changing the locally insert slits and the speed of C60. The graphene ripple is a kind of surface wave which carries a lot of surface information, and the wave energy could also be focused due to the interference of ripples. The controlled ripples and their diffraction and interference in graphene are of great significance to detect the cracks and defects in the graphene sheet by receiving the wave signals. Our findings provide deep insight into the vibrating properties of graphene and lead to an improved understanding of suspended graphene devices.

We would like to acknowledge the support from the National Natural Science Foundation of China (Grant Nos. 50971081 and 50831003). We also thank the support from the Natural Science Fund for Distinguished Young Scholars of Shandong (Grant No. JQ200817). This work was also supported by the Natural Science Fund of Shandong Province (Grant No. ZR2009FM043), by the Ph.D. Dot Programs Foundation of the Ministry of Education of China (Grant No. 20090131110025) and by the National Science Fund for Distinguished Young Scholars (Grant No. 2009JQ014) from Shandong University.

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