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Beat vibration of hybrid boron nitride-carbon nanotubes

– A new avenue to atomic-scale mass sensing

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Abstract: In this letter a beat phenomenon is reported in molecular dynamics simulations for vibrating boron nitride-carbon nanotubes (BN-CNTs) and then analysed based on a continuum mechanics theory. It was shown that the distinctive dynamic behaviour is a result of the superposition of two orthogonal transverse vibrations whose frequencies are slightly different due to the oval cross-section of the hybrid nanotubes. In particular, the interaction between the two vibrations in BN-CNTs will facilitate to resolve the fundamental issue in developing mass nanosensors for atomic-scale mass measuring. To reach this goal, efforts should be made to maintain high quality factor of the BN-CNT oscillating system by minimising the damping effect of its surrounding environment. This issue turns out to be essential for the beat mode-based nanosensors as large damping will reduce the hybrid nanotubes to conventional resonators with only one transverse vibration same as that reported for homogeneous nanotubes.

In the past decades, superior mechanical and other physical properties of quasi-one-dimensional nanomaterials such as carbon nanotubes (CNTs) and boron nitride nanotubes

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(BNNTs) have triggered a great deal of interest due to a broad range of their potential engineering applications [1, 2]. Among the most promising applications are the nanoresonators with ultra-high frequency and the nanosensors for measuring atomic-scale mass [3-8], which exploit the extreme stiffness, super high strength and relatively low mass density of CNTs and BNNTs, and the sensitivity of their nature frequency to the mass absorbed on the nanotubes. Nevertheless, a long-standing issue remains in measuring the mass of a single atom as the frequency shift varies with not only the mass of the atom but also its position on the nanotube [9, 10]. To locate the position of the attached atom, one has to resort to additional experimental equipment, such as a scanning electron microscope [11, 12] and optical microscope [13]. This will make the measurement much more complicated and time-consuming [12]. Under this circumstance, efforts are required to resolve the issue on the nanotube-based mass sensor by achieving a novel design that enables one to correlate the mass of adsorbate directly to the shift of the frequency. Recently, Gil-Santos [9] confirmed that such a mass sensor can be achieved via a resonator exhibiting two orthogonal vibrations with different frequencies. This indeed provides a new avenue towards the design of a nanotube-based mass sensor.

Obviously, homogenous nanotubes, e.g., CNTs or BNNTs, only vibrate in one transverse direction due to their axisymmetric cross-sections [14-16]. Thus, newly fabricated hybrid nanotubes are expected to provide a solution to the long-standing issue in achieving the nanosensors for measuring atomic-scale mass. Inspired by this idea, the present work is focused on the vibration of boron nitride-carbon nanotubes (BN-CNTs), which are fabricated by doping a BN segment into the CNTs. These novel nanotubes have attracted increasing attention due to their unique physical properties and thus, great potential for the next-generation nanodevices [17-19]. The first-principles calculations and molecular dynamics (MD) simulations were employed to

measure the geometric and elastic properties for BN-CNTs [20-22]. The results show that the structural stability of BN-CNTs is similar to their CNT and BNNT counterparts [20] and their Young's modulus is comparable to CNTs and BNNTs [21, 22]. Such similarities between BN-CNTs and CNTs/BNNTs suggest that BN-CNTs are also promising for the design of nanoresonators. The vibrations of pristine CNTs and BNNTs have been studied extensively in the past two decades [14-16, 23-25]. Different vibration frequencies were obtained for CNTs and BNNTs even with similar geometric size [16]. This observation infers that the frequency of hybrid BN-CNTs should change with the vibration (or bending) direction due to the fact that the bending stiffness of the hybrid nanotubes varies with the bending axis. These may lead to two orthogonal vibrations of the hybrid nanotubes with different frequencies as described in Ref. [9]. It is thus of great interest to explore the unique vibrational responses of BN-CNTs.

In this letter, the vibrational behaviours of BN-CNTs were investigated based on MD simulations. A beat phenomenon was observed, which is unique for the vibration of BN-CNTs but is absent from vibrating CNTs and BNNTs. Effort was then invested to reveal the physical mechanisms of the beat phenomena by using the continuum mechanics theory. It is found that such two-dimensional vibration characteristics make BN-CNTs an ideal candidate for the nanosensors for atomic-scale mass sensing. In addition, case studies were carried out to exam the influence of key parameters on the performance of BN-CNTs-based nanoresonators.

In the present study, we considered (10, 0) zigzag BN-CNTs with two linear C-B (N) junctions along the tube axis as shown in Fig. 1a. The composition of BN-CNTs is represented by the molar fraction of BN atoms c , which was taken as the ratio of the number of BN atoms to the total number of atoms in the system. Here c was taken as 0.55 and the length L of the nanotubes was assumed to be 100 Å. In this study, the classical MD simulations were employed to investigate

the transverse vibration characteristics of BN-CNTs. The Tersoff-like potential was adopted to model the interactions between atoms, which is in the form of an interactive empirical bond order potential [26]. The values of the parameters in Tersoff potentials were taken from Refs. [27, 28]. Such interatomic interactions in the C-B-N system have been successfully employed to evaluate the mechanical and thermal transport properties of hybrid BN-C nanosheets [29, 30] and study the mechanical behaviours of BN-CNTs under static loading [21, 22]. These studies showed clear evidence that Tersoff potentials are reliable in measuring the mechanical properties of BN-CNT structures. In each simulation, a BN-CNT was initially created using the lattice constant of CNTs, and then relaxed to a minimum energy state using the conjugate gradient algorithm. Subsequently, MD simulations were performed to study the vibration behaviours of the BN-CNT on the energy-minimized configuration, which was detailed in the supplementary material [31] of this letter.

In Figs. 1a and 1b we showed the oscillation mode of the vibrating BN-CNTs, where a non-planar whirling motion is observed for the nanotubes (Fig. 1a), characterized by an elliptical trajectory of their cross-sections (Fig. 1b). It is noted that such a non-planar whirling motion of hybrid nanotubes is in contrast to the conventional transverse vibration reported for CNTs and BNNTs [14-16], where the oscillation trajectory of the cross-section is a straight line. To further study the non-planar whirling motion observed for BN-CNTs we plotted the kinetic energy time history in Fig. 1c for the vibrating nanotubes. It is seen from the figure that the amplitude of the kinetic energy exhibits a periodic pulsation pattern that, in the classical vibration theory, is recognized as a distinctive feature of beat vibration [32], which theoretically is induced by a combination of two simple harmonic vibrations with almost the same frequency. After applying the fast Fourier transform (FFT) to the kinetic energy time history, we obtained the corresponding frequency spectrum as shown in Fig. 1d, where two peaks of the vibration amplitude are identified

associated with the frequency of 390 GHz and 415 GHz, respectively. These results indicate that the BN-CNT is exposed to a combined vibration that is comprised of two orthogonal vibrational components whose frequencies f are, respectively, 195 GHz and 207.5 GHz (Note that the kinetic energy fluctuates at a frequency two times of the vibration frequency of the nanotubes). Thus, the beat mode achieved in MD simulations for the nanotubes (Fig. 1) is possibly a result of the interaction between the two orthogonal transverse vibrations. To identify the origin of the two orthogonal vibrations, in Fig. 2a we illustrated the cross-section of the BN-CNT after the relaxation. It is observed that boron, nitrogen and carbon atoms move away from their original positions on the circular perimeter leading to an oval cross-section with its short and long axis respectively in x and y directions. This is a central symmetric structure differing from the circular symmetric cross-section found in pristine CNTs and BNNTs. Such a non-circular symmetric cross-section was also observed in recent density functional calculations for BN-CNTs [33]. Thus, if approximating the hybrid nanotube as an elastic beam with an elliptic cross-section, from the classical beam theory [32] its frequency can be estimated by $f = 3.56\sqrt{EI/(\rho AL^4)}$ for the doubly-clamped nanotubes. Here, E is Young's modulus, ρ is the mass density, A is the cross-sectional area. Specifically, for the elliptic cross-section the moment of inertia is [34] $I = I_x \approx \pi(3ba^2 + a^3)h/32$ and $I = I_y \approx \pi(3ab^2 + b^3)h/32$ for the transverse vibration along x and y directions, respectively. Here, a and b ($a < b$) are the length of the short axis and long axis of the elliptic cross-section, and h is the effective thickness of the nanotubes (assumed to be a constant). Thus, I_y is greater than I_x and accordingly, the frequency of BN-CNTs vibrating along the y direction (noted as f_y) is greater than that along the x direction (noted as f_x). The combination of the two orthogonal vibrations (in x and y directions) with different frequencies

finally leads to the non-planar whirling motion of the hybrid nanotubes as observed in MD simulations. Thus, it is concluded that the two-dimensional oscillation of the BN-CNT is primarily a result of the non-circular symmetric cross-section of nanotubes leading to different I for vibrations along x and y directions. Moreover, besides the present BN-CNT whose c is 0.55, oval cross-sections are also observed in Fig. 2b for other BN-CNTs with c increasing from 0.15 to 0.85. Obviously, we shall see the changes in the moment of inertia and thus the frequency of the vibration in x and y directions in this process. These suggest that the frequency and the vibration amplitude of BN-CNTs should be tunable via the variation of the molar fraction of BN atoms.

Next, we shall establish a mechanics model to account for the proposed physical mechanisms underlying the formation of the beat phenomenon. It is well known that the first transverse resonant mode of a nanotube along the x or y direction can be described by a sinusoidal function. For simplicity, the oscillating system of the hybrid nanotube can be represented by the spring-mass system illustrated in Fig. 2c. In particular, two elastic constants, k_x and k_y , are respectively assigned to the springs in x and y directions giving the structural stiffness in two elementary orientations of BN-CNTs. Thus, the harmonic vibration in the x or y direction can be described by $U_i = A_i \sin(\omega_i t)$, where $i = x$ and y referring to the x and y directions, ω_i is the angular frequency, A_i is the amplitude with no phase lag and t is time. The velocity of the vibrating nanotube is $V_i = dU_i / dt$ and its total kinetic energy (E_{ke}) reads:

$$E_{ke} = \frac{1}{2} m_e \sum_{i=x,y} V_i^2 = \frac{1}{2} m_e \sum_{i=x,y} A_i^2 \omega_i^2 \cos^2(\omega_i t). \quad (1)$$

Here, m_e is the mass of BN-CNTs. Under the condition $\omega_x \approx \omega_y$ (Fig. 1d) or $\sin[(\omega_x - \omega_y)t] \approx 0$

Eq. 1 reduces to:

$$E_{ke} = A_{ke} - A_{ke} \cos[(\omega_x - \omega_y)t] \cos[(\omega_x + \omega_y)t], \quad (2)$$

where $A_{ke} = m_e (A_x^2 \omega_x^2 + A_y^2 \omega_y^2) / 4$ is the amplitude of the total external energy. Eq. 2 gives a nearly harmonic vibration with a relatively low frequency $f_b = |\omega_x - \omega_y| / 2\pi = |f_x - f_y|$ [32, 35], i.e., the beat vibration achieved in MD simulations under the condition $\omega_x \approx \omega_y$. This theoretical analysis confirmed that the two orthogonal vibrations of BN-CNTs (Fig. 1d) satisfy the existence condition of the beat mode ($f_b = 12.5$ GHz), i.e., the frequencies are different but very close to each other. Thus, the beat mode achieved in MD simulations (Fig. 1c) is indeed the combination of the two orthogonal vibrations.

In what follows, the established mechanics model and the vibration theory will be further employed to characterise the evolution of the cross-sectional trajectory in the beat vibration. As shown in Fig. 3a the BN-CNT is first excited to vibrate transiently along a straight line in the first and third quadrants of the coordinate system (Fig. 2a). Then, a clockwise whirling motion is observed due to the increasing phase difference in the two non-synchronous vibrations of the BN-CNT along x and y directions. As a result, in Fig. 3b an approximately elliptic trajectory is shown with the major axis being in the first and third quadrants. After this, in Fig. 3c the elliptic trajectory gradually expands and finally evolves into a circular one, which means that the phase difference of the two vibrations of the BN-CNT reaches 90° . With greater phase difference, Fig. 3d shows an elliptic trajectory again. However, this time its major axis is found in the second and fourth quadrants. Finally, in Fig. 3e the length of the primary axis increases with time leading to a straight line trajectory in the second and fourth quadrants. This indicates that the vibrations along the x and y directions now are nearly 180° out of phase. Figs. 3f-3h show the evolution of the vibration patterns in the second half period, which is found to be similar to the one in the first half period except that the cross-section rotates in an anticlockwise direction.

As confirmed by Gil-Santos et al. [9], the beat vibration of BN-CNTs with the two-dimensional whirling motion of the cross-section offers a new approach for mass sensing at the nanoscale. The novelty here is that the mass of the adsorbate can be directly determined by the frequency shifts of the two component vibrations [9]. The basic idea of this method is that in addition to relative frequency shifts which are determined by the mass and position of the adsorbate, this novel method provides a new sensing parameter, the difference between the relative frequency shifts of two component vibrations, which only depends on the position of the adsorbate [9]. In practical nanoresonator application, a BN-CNT is usually excited by an external force generated by the electrostatic electric field [3-5], and vibrates with damping resulting from various extrinsic and intrinsic sources [36]. To model this situation, we consider the BN-CNT to act as a damped mass-spring system along each elementary direction shown in the inset of Fig. 4, where c_x and c_y are coefficients of the damping, respectively, in x and y directions, F the actuation force, and θ the actuation angle. Suppose F is a time harmonic force, i.e., $F = F_0 \sin(\omega t)$ with F_0 being the force amplitude and ω the force frequency, the dimensionless vibration amplitude (α) of the BN-CNT induced by F is [31]

$$\alpha = \frac{\cos^2 \theta}{\sqrt{(r - \lambda^2)^2 + 4s^2 \lambda^2 \xi^2}} + \frac{\sin^2 \theta}{\sqrt{(1 - \lambda^2)^2 + 4\lambda^2 \xi^2}}. \quad (3)$$

Here $r = k_x / k_y$ is the ratio of the elastic constants of the springs. Since $\omega_i = \sqrt{k_i / m_e}$ ($i = x$ and y) we have $r = \omega_x^2 / \omega_y^2 = f_x^2 / f_y^2$. In addition, $s = c_x / c_y$ is the ratio of damping coefficients, $\xi = c_y / (2m_e \omega_y)$ is the dimensionless damping and $\lambda = \omega / \omega_y$ is the dimensionless force frequency. In Fig. 4 the dimensionless frequency-response curve is plotted for the BN-CNT under different actuation angles. The results associated with dimensionless damping $\xi = 0.006$ and 0.06

are shown in Figs. 4a and 4b, respectively. The corresponding quality factor $Q = 1/(2\xi)$ [32] is 83 and 8.3, which covers the Q value ($3 \leq Q \leq 80$) measured experimentally for most nanotube-based nanoresonators [3, 36, 37]. In the present study, we consider $r = 0.88$ as calculated in our MD simulations and $s = 1$, i.e., the environmental condition is identical in x and y directions. We can see from Fig. 4a that when the damping effect is relatively small (or relatively high Q) two peak amplitudes are identified for all vibrations with $0^\circ < \theta < 90^\circ$, corresponding to the two resonant frequencies of the vibrations along the two elementary orthogonal directions (i.e., $\theta = 0^\circ$ and $\theta = 90^\circ$). In addition, in Fig. 4a the difference between the two peak amplitudes is found to change with θ , i.e., it reaches its minimum value at $\theta = 45^\circ$ and then increases when θ rises (or decreases) from 45° to 90° (or 0°). In contrast, at a much lower Q we can see from Fig. 4b that only one peak amplitude is detected for the resonator irrespective of the actuation direction. In other words, when Q decreases from 83 to 8.3 by an order of one magnitude the beat mode with a two dimensional vibration of the cross-section will reduce into the conventional transverse vibration with the cross-section oscillating along a straight line. Such a mode change due to the increasing damping effect was observed experimentally for a nanowire-based nanoresonator [9]. This observation suggests that minimising the damping effect of the surrounding environment is essential for the appropriate performance of this novel BN-CNT-based nanoresonator.

In summary, the vibrations of BN-CNTs were studied based on MD simulations. A beat phenomenon is achieved, which is comprised of two orthogonal vibrations of the hybrid nanotubes. Such a beat vibration of BN-CNTs is found to originate from the symmetry breaking of their cross-sections due to the different bond characters of carbon-carbon, boron-nitrogen, carbon-boron and carbon-nitrogen bonds. This results in two slightly different principal moments of inertia and accordingly, two orthogonal vibrations with close but different frequencies, the superposition of

which finally leads to the beat mode. The beat vibration of BN-CNTs is of engineering interest as its two component vibrations provide a solution to a long-standing issue on the development of mass sensors and thus a new pathway to atomic-scale mass sensing. Specifically, in practical applications, the quality factor Q turns out to be of particular importance for BN-CNT-based nanosensors as decreasing Q will not only lead to low sensitivity of mass sensors but also transform the BN-CNTs vibrating in beat mode to conventional resonators showing one-dimensional vibration of their cross-sections.

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Figures

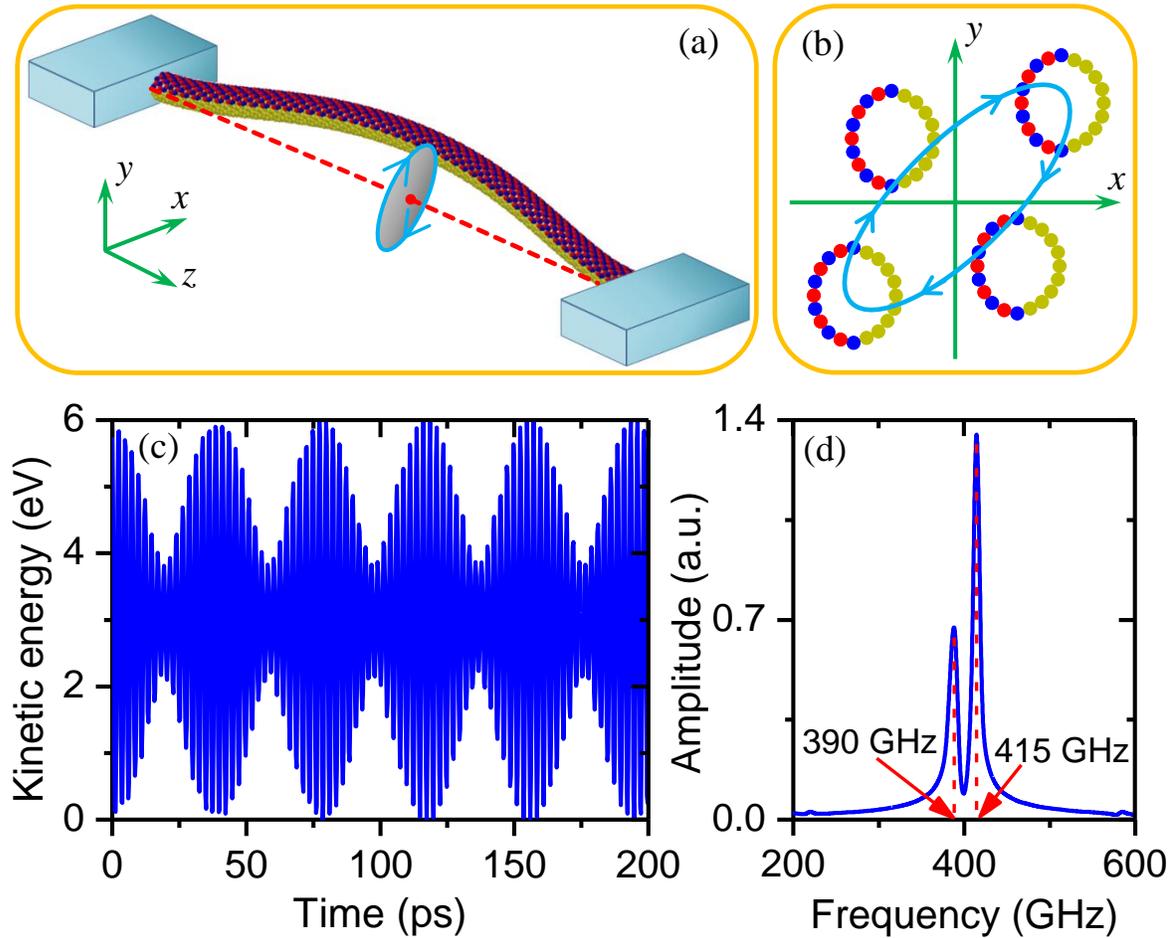


Fig. 1. (a) A schematic showing a vibrating BN-CNT modelling in the present study (see the video). Here the carbon atoms are denoted as yellow balls, while the boron and nitrogen atoms are represented by red and blue balls. (b) A typical displacement pattern of the cross-section during the vibration. (c) Kinetic energy time history of the vibrating BN-CNT. (d) Fast Fourier transform of the kinetic energy time history.

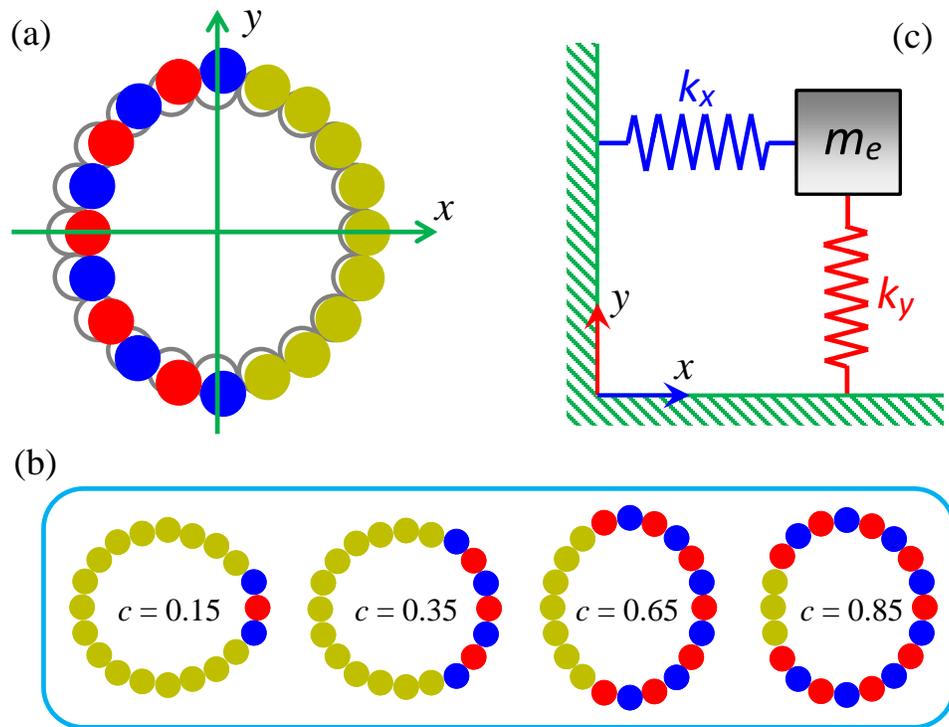


Fig. 2. (a) Cross-sectional geometry of the BN-CNT with a molar fraction of BN atoms c of 0.55 after the initial relaxation. Here the hollow and solid circles respectively represent the atomic position before and after relaxation. (b) Cross-sectional geometry of BN-CNTs with different molar fractions of BN atoms. (c) Simplified spring-mass model of BN-CNTs.

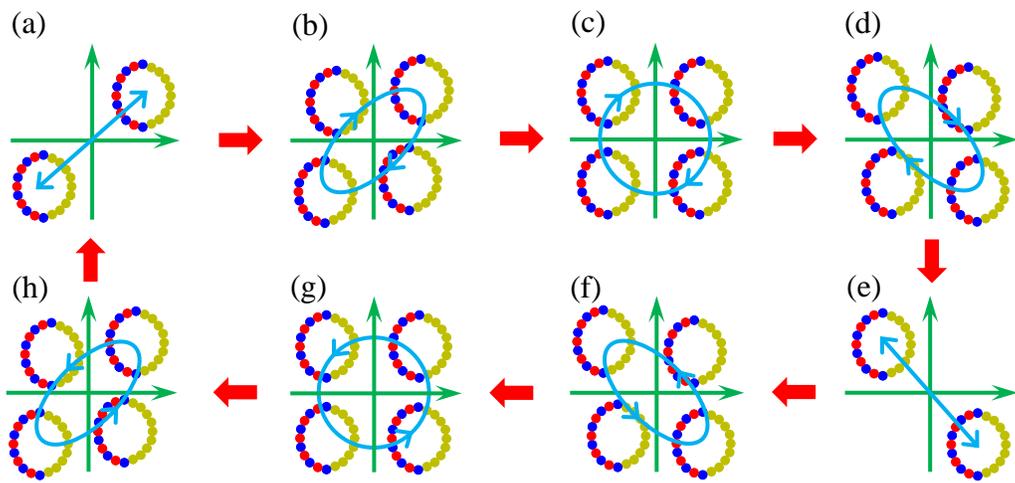


Fig. 3. Evolution of the displacement patterns of the cross-section during a beat vibration period of the vibrating BN-CNT.

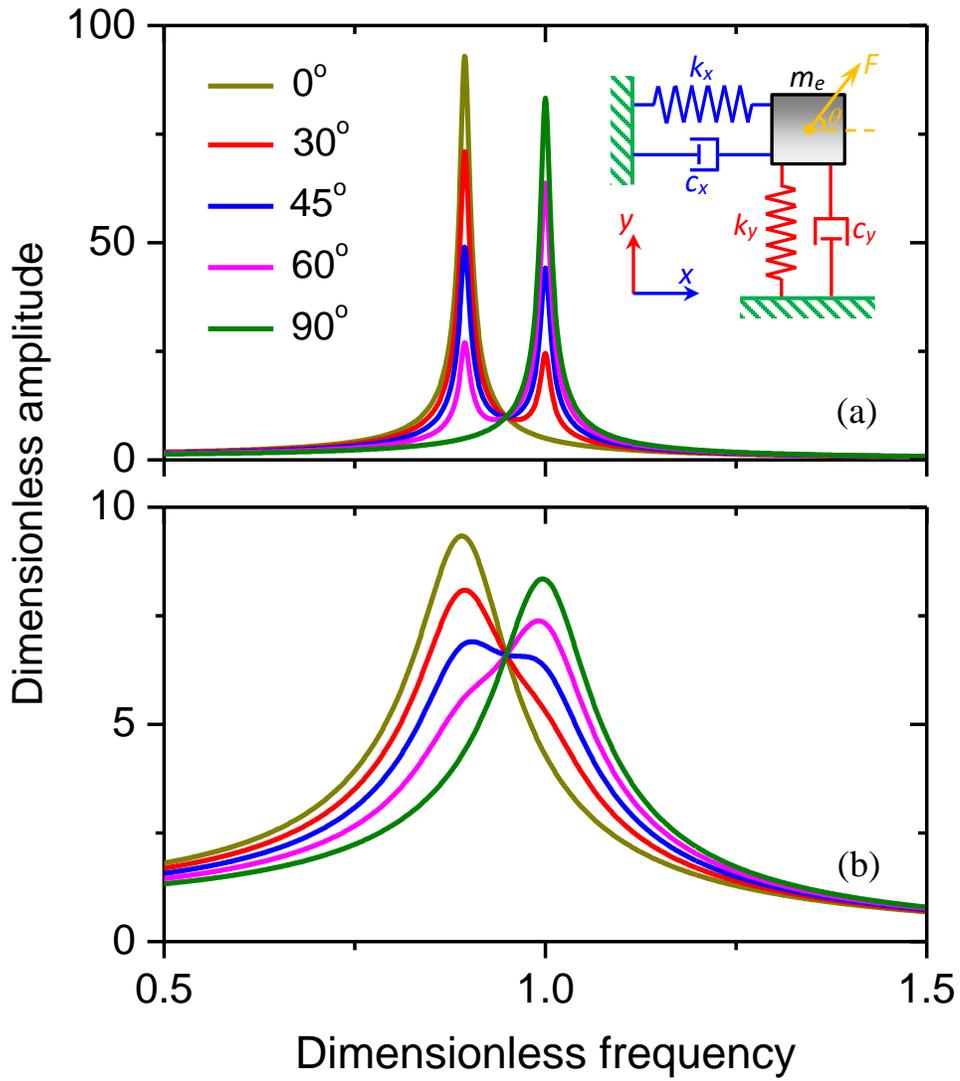


Fig. 4. The dimensionless vibration amplitude as a function of the dimensionless force frequency for the BN-CNT-based resonator with the quality factor respectively being (a) 83 and (b) 8.3. The inset shows the damped spring-mass model of the BN-CNT-based resonator.