Probing built-in stress effect on the defect density of stretched monolayer graphene membranes

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Abstract

It is of great interest to link Raman scattering to the properties of disorders in graphene membranes, which provides an effective characterization method to probe atomic scale defects. The built-in stress effect on the defect densities of substrate-supported monolayer graphene membranes around wells is investigated. First, a modified phenomenological model is developed to depict the relationship between built-in stresses and defect-activated Raman intensities. To validate the rationality of the modified model, Raman spectroscopy is used to characterize stretched graphene membranes on different patterned substrates with micro-scale wells. The experimental data indicate that the intensity ratio of D mode to G mode $I_{D}/I_{G}$ increases with the Raman test point approaching the well edge. According to the modified model, the increase of $I_{D}/I_{G}$ means the rise of defect densities, which originates from the propagation of initial defects in graphene membranes under built-in tension. The underlying mechanism of defect density increasing phenomenon is that the built-in stresses provide the energy for defect propagations in stretched graphene membranes. Theoretical and experimental comparison well validates the rationality of the modified model. The work can provide a theoretical foundation for Raman characterization method of defect propagations in stretched graphene and applications of defective graphene-based nanodevices.

1. Introduction

Pristine graphene has attracted enormous interest due to its unique electronic structure [1] and remarkable mechanical, electronic, optical, and thermal properties [2–4]. All of these properties make it a potential material for both fundamental studies [5,6] and practical applications [7–10]. However, the structural defects such as atomic defects and grain boundaries are ubiquitous but seem noxious in realistic graphene samples [11,12], which can generate during high-temperature growth process [13,14], intentionally by electron or ion irradiation [15,16], or in the operational conditions of graphene-based devices [17,18].

The nature and amount of structural defects can remarkably degrade the physicochemical, electronic and mechanical properties of graphene, e.g. decreasing elastic modulus [2,19] and limiting electrical mobility [21]. On the other hand, well-controlled defects in graphene sheets can be employed to enhance the chemical reactivity [20], tailor a band gap of nanoelectronics [22] and selectively sieve molecules by introducing nanopore-like defects [23,24]. Besides, the surface functionality in graphene-based bio-devices is modified via the desired defects for DNA-sequencing [25]. Therefore, exploring the external conditions that can cause the defect propagations and the defect propagation mechanism of graphene under these conditions theoretically and experimentally is of vital importance for graphene-based devices.

To achieve this goal, a fast and concise method for characterizing defects in graphene membranes is required. Raman spectroscopy, as widely reported in the literature, has been proved to be an...
incredibly powerful tool for identifying the defects in graphene flakes [26]. Substantial efforts including theoretical modelling and experimental investigations have been focused on the Raman characterization of defects within the lattice of graphene. Due to the sensitivity to defects, the D band, appearing at ~1350 cm⁻¹, is typically utilized to characterize the lattice damage in graphene [15,16]. Actually, the D peak arises from the breathing modes of six-atom rings and requires a defect for its activation [27]. The intensity ratio of the defect-induced D band to the first-order G band, \( I_D/I_G \), provides a spectroscopy-based method to quantify the defect density of graphene. The well-known Tuinstra-Koenig relation, \( I_D/I_G = C_1 \lambda / L_0 \), is used to quantify the evolution of defects in graphene [28], where the coefficient \( C_1 \) depends on the excitation laser wavelength \( \lambda_L \), and \( L_0 \) is the crystal size. Furthermore, Luchese et al. [16] proposed a phenomenological model to investigate the density of point defects in ion-bombarded graphene. Tip-enhanced Raman spectroscopy is used to probe individual point defects for a graphene layer with an extremely low defect density [29]. Since \( I_D/I_G \) intensity ratio does not depend on the geometry of the defects [30], it can be used to investigate the nature of the defects [30]. Meanwhile, some external conditions during graphene transfer process, such as transfer methods and target substrate structures, may induce build-in stress to graphene, which will substantially affect the evolution of graphene defects. Consequently, the \( I_D/I_G \) intensity ratio and defect density of graphene membranes may be regulated by build-in stresses. However, the relationship between defect-activated Raman intensities and build-in stresses has not been explored systematically and a complete theory should be established to link defect-activated Raman intensities to build-in stresses.

In the paper, the effect of build-in stresses on the defect density of stretched graphene membranes is studied. First, a modified phenomenological model is proposed to reveal the relation between the build-in stress and the \( I_D/I_G \) intensity ratio. Second, the build-in stresses in substrate-supported graphene emerging during the transfer process are characterized via Raman spectroscopy. Third, Raman spectra of stretched monolayer graphene membranes on patterned substrates are measured near micro-wells to validate the rationality of the theoretical model. Finally, the theoretical and experimental comparison further reveals the underlying mechanism of build-in stress effect on the defect propagations of substrate-supported monolayer graphene membranes.

2. Modified phenomenological model

In order to link Raman scattering to the properties of structural defects in graphene membranes, a modified phenomenological model is presented to study the effect of tensile strains on the defect density in stretched graphene membranes. In this work, \( I_D/I_G \) is used to quantify the defect density of graphene [15,16], and the integrated intensity ratio between the D mode and the G mode and the intensity of the D mode are defined as

\[
\frac{I_D}{I_G} = C_2 \frac{r_A^2 - r_5^2}{2r_5^2} \left[ e^{-\pi r_5^2/L_0^2} - e^{-\pi (r_A^2 - r_5^2)/L_0^2} \right] + C_3 \left( 1 - e^{-\pi r_5^2/L_0^2} \right)
\]

where \( r_A \) and \( r_5 \) denote the radii of the activated and structurally defective areas, weighted by the parameters \( C_4 \) and \( C_5 \), respectively.

Because the built-in stresses are sufficiently small, the defects in our samples are at low defect concentration (\( L_D \gg r_A \)). For the large values of \( L_D \), Eq. (1) can be rewritten as

\[
\frac{I_D}{I_G} = C_4 \pi \left( r_A^2 - r_5^2 \right)/L_0^2 + C_5 \pi r_5^2/L_0^2
\]

As indicated by Eq. (2), \( I_D/I_G \) depends on not only the Raman-activated area, but also the structurally defective area, which can be weighted by parameters \( C_4 \) and \( C_5 \), respectively. However, in this work, the influence of the structurally defective area on the \( I_D/I_G \) can be not considered for the following reasons. For low defect density, namely large \( L_D \), the D band intensity is proportional to the defect density and dominated by the activated regions. Moreover, initial point-like defects in CVD-grown graphene before transfer such as Stone-Wales defects and vacancy-type defects are assumed to be homogeneously nucleated in this work. Since most of the defects are the breakdown of the C–C bonds, defect-structure determined \( C_5 \) should be null in the ideal case. Therefore, Eq. (2) can be approximated as

\[
I_D/I_G = C_4 \pi \left( r_A^2 - r_5^2 \right)/L_0^2
\]

As shown in Fig. S2, the defect density of two-dimensional graphene membranes can be estimated as

\[
n_D \left( m^{-2} \right) = 1/\pi (r_A + L_D)^2
\]

For defects with low concentration, \( L_D \gg r_A \), thus, Eq. (4) can be rewritten as

\[
n_D \left( m^{-2} \right) = 1/\pi L_0^2
\]

Combining Eq. (3) with Eq. (5), we can obtain the relation between the defect density and \( I_D/I_G \) as follows

\[
n_D \left( m^{-2} \right) = \frac{1}{C_4 \pi^2 (r_A^2 - r_5^2)} \left( \frac{I_D}{I_G} \right)
\]

where the parameter \( C_4 \) can be given as \( C_4 = (160 \pm 48)E_1^{-4} \) [15] and the laser excitation energies \( E_1 = 2.41 \) eV in this work.

In order to link defect propagations to tensile stresses by Raman spectra, the relation between the tensile strain and the distance of line defects \( L_D \) can be given by Ref. [12].

\[
\varepsilon = \sqrt{G\hbar/E_{2D} \left[ 2 \tanh(\lambda L_D/2) - \tanh(\lambda L_D) \right]}
\]

where \( G \) is the energy release rate, \( E_{2D} \) denotes the in-plane stiffness of graphene, 1/\( \lambda \) and \( h \) are the reference length and the thickness of graphene, respectively. Judge from Eq. (7), the defect distances of line defects decrease with the tensile strains, in other words, the defect density can increase due to the stretch of graphene membranes by the tensile stresses.

As shown in Fig. S2(b), due to the tensile stress, the point-like defects in graphene membranes can further propagate and some new defects maybe form. The initial radii of activated areas \( r_A \) can increase and the defect distances \( L_D \) can decrease. Consequently, the defect density \( n_D \) of graphene membranes can increase when the graphene is under tensile stresses. For the point-like defects, the alignments of Stone-Wales defects or vacancy defects in graphene membranes can form one-dimensional defects [31–33], especially for the defects in the graphene membranes under tensile stresses. Therefore, the multiple random point-like defects including Stone-Wales and vacancy-type defects can have similar
fracture strength characteristics with the line cracks [34], and Eq. (7) also is suitable for depicting defect density change behavior of point-like defects in the graphene membranes under tensile stresses.

By substituting Eq. (5) into Eq. (7), we can obtain the change of defect density in graphene membranes with the tensile strains $\varepsilon$ as follows

$$
\varepsilon = \frac{Gh}{E_{2D}} \frac{1}{2} \tanh \left( \frac{\lambda_c}{\sqrt{A_D}} \right) - \tanh \left( \frac{\lambda_c}{\sqrt{A_C}} \right) \tag{8}
$$

In order to facilitate the Raman characterization of the tensile stresses on the defect propagation in stretched graphene membranes, the relation between the $I_D/I_G$ and the tensile strain $\varepsilon$ can be obtained as

$$
\varepsilon = \frac{Gh}{E_{2D}} \frac{1}{2} \tanh \left( \frac{\lambda_c}{\sqrt{A_D}} \right) - \tanh \left( \frac{\lambda_c}{\sqrt{A_C}} \right) \tag{9}
$$

where $\lambda_c = \lambda / \sqrt{C_\varepsilon (r_A^3 - r_g^3)}$. Eq. (9) can be utilized to depict the defect propagation properties of graphene membranes under mechanical stresses though defect-activated Raman scattering parameters.

3. Experiment

In order to validate the rationality of the above phenomenological model, stretched graphene membranes near the wells are prepared and characterized by Raman spectroscopy.

3.1. Sample preparation

Before implementing graphene transfer process, patterned substrates with micro-wells are designed and fabricated. Standard micro fabrication processes are adopted to pattern different sizes and shapes of wells with depth of ~530 nm on the silicon dioxide (SiO$_2$) capped Silicon-On-Insulator (SOI) wafers [35].

Next, monolayer graphene membranes are transferred onto the patterned substrates with different micro-scale wells. Fig. 1 shows the transfer process of monolayer graphene membranes on target substrates by a combined dry and wet transfer method [35,36]. Firstly, graphene films are grown on germanium (Ge(110)) substrates by chemical vapor deposition (CVD) (see Fig. 1(a)). The corresponding Raman spectrum indicates that the CVD-grown graphene is monolayer (see Fig. 2(c)). Second, thin polymethyl methacrylate (PMMA) polymer is spin-coated onto a graphene/Ge composite, and a thick polydimethylsiloxane (PDMS) slice with a methacrylate (PMMA) polymer is spin-coated onto a graphene/Ge composite. The monolayer graphene membranes within the well area have severe plastic deformations due to the surface tension of liquid, which gives rise to the built-in tension, as shown in Fig. 1(f).

3.2. Built-in stress characterization and analysis

The built-in stresses emerging in substrate-supported graphene membranes around the wells during the transfer process are characterized via Raman spectroscopy. Fig. 2(c) shows the Raman spectrum of a graphene sheet transferred on a flat SiO$_2$/Si substrate. When the monolayer graphene is transferred onto a patterned substrate, the Raman spectra of graphene membranes near and far from the well area show a distinct difference. In order to define the graphene membranes near wells and far from wells, the built-in tensions $\tau_{\text{built-in}}$ of the stretched graphene membranes are introduced as: for graphene membranes near wells, $\tau_{\text{built-in}} > 0$ and for graphene membranes far from wells, $\tau_{\text{built-in}} = 0$ [35].

Compared to the Raman spectra tested at the flat substrate and far from well area of the patterned substrate, both the G and 2D peaks of Raman spectra at near well area shift along low frequency direction, which proves the existence of built-in stresses in graphene membranes around the wells caused by the transfer (see Fig. 2(d)). More experimental data on the G and 2D mode frequency shifts of substrate-supported graphene membranes around the wells with other sizes and shape, $\omega_G$ and $\omega_{2D}$, have been presented in Supplementary Fig. S1. As shown in Supplementary Fig. S1, both the $\omega_G$ and $\omega_{2D}$ downshift when the testing points move from far from the wells to near the wells. It is noted that we focus on the Raman spectra of substrate-supported graphene membranes, not the Raman spectra comparison between substrate-supported area and suspended area in Fig. 1(f), thus the doping effect from SiO$_2$ substrates on the G and 2D mode frequency shifts are the same when the testing point moves from far from wells to near the wells. According to the relation between tensile strains and the 2D band frequency shift ($\varepsilon = - \Delta \omega_{2D} / 27.1 \text{ cm}^{-1}$) given by the previous studies [35,37], the built-in tensile strains increase as the testing point approaches the well edge.

Bunch et al. [38–40] have also demonstrated that the graphene sheets across the trench or the well are under built-in strains resulting from the fabrication process. Depending on the thickness of spin-coated PMMA film, the graphene membranes of the well area will suspend over the well or stick onto the bottom of wells (partially free-standing) [36], as shown by the bottom diagram of Fig. 2(b). The dropped graphene membrane with partially free-standing area can provide higher deformation, and higher transverse deformations imply larger built-in strains of graphene membranes around the wells based on the theoretical analysis of our previous work [35]. Therefore, to obtain large built-in strains, in this work, we select thin PMMA films to obtain partially free-standing graphene for research. The SEM image of Fig. 2(a) shows the partially free-standing graphene membrane over the well. It can be found that the graphene suspends on the sidewall area and sticks onto the bottom of the well in the center area.

4. Results and discussions

4.1. Defect density increasing phenomenon

Raman spectra of substrate-supported graphene membranes around predefined wells under built-in stresses are measured by the Raman mapping, and the defect density increasing phenomenon is observed, experimentally. First, the nature of the initial defects in graphene membranes without built-in stresses is evaluated by defect-activated D and D’ Raman spectra. As shown in Fig. 2(c), the appearance of D peak indicates that the CVD-grown graphene on Ge(110) has initial defects. Moreover, the low D’ Raman peak in Fig. 2 (c) and (d) has also indicated that most of initial defects of
Fig. 1. Preparation of stretched monolayer graphene membranes on target substrates with different size wells by a combined dry and wet transfer method: (a) graphene sheets are grown by CVD on Ge(110) surface; (b) thin PMMA polymer is spin-coated onto graphene/Ge substrates and PDMS blocks are prepared to work as flexible frames to hold the PMMA/graphene film; (c) the Ge substrate is etched and the PDMS/PMMA/graphene composite is transferred onto the target substrate; (d) the heat treatment is used to make the PMMA/graphene composite sufficiently adhere to the target substrate; (e) the PMMA is removed by acetone solvent; (f) plastic deformations of suspended graphene membranes in the well, and built-in tensions of substrate-supported graphene membranes near the well can be introduced by the stretching of suspended graphene membranes due to the existence of wells. (A colour version of this figure can be viewed online.)

Fig. 2. Built-in stress characterization of CVD-grown graphene membranes before and after transfer: (a) the SEM image of a partially suspended graphene membrane onto a 2 μm-diameter square well, where the cyan dashed line shows the suspended area and the graphene of the other area in well is in dropped state; (b) schematic diagrams of graphene membranes in two states including free-standing graphene (top diagram) and dropped graphene (bottom diagram); (c) Raman spectra of graphene sheets on a flat SiO₂/Si surface, where the existence of D peak indicates that initial defects can emerge during the CVD process; (d) Raman spectra of graphene membranes far from and near the well, and the magnified views in (c) and (d) show the frequency downshift of the G mode at near well area and low D' peak near the G peak. (A colour version of this figure can be viewed online.)
graphene membranes after transfer are vacancy-like [30].

Fig. 3 shows the Raman mapping of the graphene membranes over two predefined wells and at the vicinity of the wells. An interesting phenomenon, the \( \text{ID}/\text{IG} \) intensity ratio increasing around the well, is observed. The \( \text{ID}/\text{IG} \) intensity ratio of the supported graphene membrane far from the 3 \( \mu \text{m} \)-diameter circular well (more than 4 \( \mu \text{m} \) distance to the well edge) is less than 0.2 (see Fig. 3(a)). When the testing point moves from \( y = -3.5 \mu \text{m} \) to \( y = -0.5 \mu \text{m} \), \( \text{ID}/\text{IG} \) intensity ratio changes from 0.349 to 0.817 (increases by 134.1%), as shown in Fig. 3(c); when the testing point moves from \( y = -5.5 \mu \text{m} \) to \( y = -2.5 \mu \text{m} \), \( \text{ID}/\text{IG} \) intensity ratio changes from 0.644 to 1.055 (increases by 63.8%), as shown in Fig. 3(f). Similar results can be observed from the monolayer graphene membrane over the 20 \( \mu \text{m} \times 2 \mu \text{m} \) well (see Fig. 3(d) and (f)). It is noted that the substrate effects on the \( \text{ID}/\text{IG} \) ratios of supported graphene near the wells and far from wells are statistically equivalent. The increase of \( \text{ID}/\text{IG} \) intensity ratios indicates that the defect densities of the substrate-supported and stretched graphene membranes significantly rise when the testing points approach the well edges.

Therefore, the explanation for the above defect density increasing phenomenon is that the initial defects in graphene membranes arising from CVD and transfer processes further propagate under the built-in tension, which can increase the defect density.

4.2. Theoretical and experimental comparison

To verify the validity of the modified model developed in Section 2, the theoretical and experimental comparison study of the built-in stress effect on the defect density of substrate-supported graphene membranes over the wells is presented in Fig. 4. As shown in the inset and lower-left corner of Fig. 4, when the testing point sweeps from \( x = -6 \mu \text{m} \) (far from the well) to \( x = 1 \mu \text{m} \) (near the well), the experimental \( \text{ID}/\text{IG} \) intensity ratio increases from 0.51 to 0.86. As demonstrated by our previous work [35], the built-in strain increases when the testing point moves from \( x = -6 \mu \text{m} \) to \( x = 1 \mu \text{m} \). Therefore, the \( \text{ID}/\text{IG} \) intensity ratio can increase with the built-in stresses, experimentally.
As shown in the upper-right corner of Fig. 4, the theoretical $I_D/I_G$ intensity ratio for $\lambda_c = \frac{\lambda E_r}{\sqrt{160\pi(r_A^2 - r_s^2)}}$ increases with the built-in strain. According to the relationship between the tensile strains and Raman frequency shift, $\epsilon = \frac{\Delta\nu_{2D}}{\nu_{2D}}/27.1$ %/cm$^{-1}$ [37], the built-in strains experimentally estimated by the 2D mode frequency shift are in the range of 0.2%–0.4% [35]. Moreover, the experimental built-in strains increases almost linearly when the testing point approaches the well edge [35]. Therefore, the theoretical built-in strains can be assumed to be linearly varied from 0.25% to 0.45% when the testing point moves from $x = -6$ μm to $x = 1$ μm in this work.

According to Eq. (9) and Fig. 4, the theoretical $I_D/I_G$ intensity ratio also increases with the built-in strain $\epsilon$, which has a similar tendency to the experimental data. Moreover, most of experimental data fall into the shadow area, in which the upper and lower limits are evaluated by Eq. (9) for $\lambda_c^- = \frac{\lambda E_r}{\sqrt{(160 - 48)\pi(r_A^2 - r_s^2)}}$ and $\lambda_c^+ = \frac{\lambda E_r}{\sqrt{(160 + 48)\pi(r_A^2 - r_s^2)}}$, respectively. Therefore, the experimental data agree with the theoretical results of the model, which can validate the rationality of the modified model. The reason is that the built-in strain increases when the testing point approaches the well edge. The increasing built-in strain can supply the adequate energy for the defect propagations, resulting in the increasing defect density.

Furthermore, the effect of built-in stresses on the intensities $I_D$ has also been discussed experimentally and theoretically. Fig. 5 has also indicated that the D mode intensities $I_D$ of supported-graphene membranes near the wells are also significantly affected by built-in tensions. Fig. S1(a)-(b) shows the line mapping results of the G-mode and 2D-mode frequencies of the supported graphene membranes around the 20 μm × 2 μm well with respect to $x$-axis. It is found from Fig. S1(b) that the frequencies of the G and 2D modes would downshift around the well, which proves the existence of tensile built-in stress of supported graphene membranes in the vicinity of the wells. As indicated from Fig. 5(b), $I_D$ of graphene membranes increases with the testing point approaching the well edge. As seen from Fig. 5(a)-(c), when the testing point moves from $y = 18$ μm (far from the well edge) to $y = 9$ μm (near the well edge), the intensities $I_D$ vary from 6.3 units to 111.1 units, increasing by 16.63 times.

![Fig. 4. Theoretical and experimental comparison of built-in stress effect on $I_D/I_G$ intensity ratios of substrate-supported and stretched graphene membranes around the well, where the dashed blue lines and markers in the lower-left corner show the experimental data, and the red lines in the upper-right corner denote the theoretical result of the $I_D/I_G$ intensity ratios for $\lambda_c = \frac{\lambda E_r}{\sqrt{160\pi(r_A^2 - r_s^2)}}$. The shadow area shows the upper and lower limits evaluated by Eq. (8), where $\lambda_c^- = \frac{\lambda E_r}{\sqrt{(160 + 48)\pi(r_A^2 - r_s^2)}}$ and $\lambda_c^+ = \frac{\lambda E_r}{\sqrt{(160 - 48)\pi(r_A^2 - r_s^2)}}$. The inset in the lower-left corner is the SEM image of the stretched graphene membranes over the 20 μm × 6 μm well and the line Raman mapping testing route along $x$ axis. (A colour version of this figure can be viewed online.)](image1)

![Fig. 5. The effect of built-in stresses on the $I_D$ intensities of the defective substrate-supported graphene membrane: (a) the SEM image of a graphene membrane, where the dashed line shows the 2D Raman mapping area and the $y$ axis shows the line mapping route; (b) the line mapping experimental results of the $I_D$ intensities and the inset shows the theoretical results of the defect distance $L_D$ with respect to the tensile strain calculated by Eq. (7); (c) 2D mapping results of the $I_D$ intensities; (d) the theoretical results of the defect density $n_0$ (m$^{-3}$) with respect to the tensile strain calculated by Eq. (8). (A colour version of this figure can be viewed online.)](image2)
As indicated by Eq. (7) and the inset of Fig. 5(b), the defect distance $L_0$ decreases with the tensile strain, theoretically. Due to $L_0 \propto n_D$, $n_D \propto 1/L_0^2$, the D mode intensities $I_D$ can increase with the tensile strain. In other word, as shown in Eq. (8) and Fig. 5(d), the defect densities of stretched graphene membranes can increase with the tensile strain, which agrees with the experimental results of Fig. 5(b). It should be attributed to the built-in stresses tearing the graphene membranes, which extend the existing defects emerging during the CVD and transfer processes and cause defect propagations in graphene membranes. Furthermore, a defect can introduce a local flexibility into the graphene membranes and the stress can sharply concentrate at the defect area [41]. The defects can further propagate to release the strain energy, and both the radii of structurally defective area and activated area $r_2$ and $r_4$ increase when the built-in stresses are applied on the graphene membranes. The propagations of point-like defects can decrease the defect distance $L_0$ and increase the defect density, as shown in Fig. 52(b). Therefore, both the experimental data and the theoretical results indicate that the increases of $I_D/L$ intensity ratios and $I_D$ intensities of substrate-supported graphene membranes around the wells are due to the increasing defect density under built-in stresses.

5. Conclusions

In this paper, the effect of built-in stresses on defect densities in substrate-supported monolayer graphene membranes has been investigated. A modified phenomenological model has been proposed to characterize the relation between the $I_D/L$ intensity ratios and built-in stresses. In order to verify the rationality of the modified model, Raman spectroscopy has been used to characterize substrate-supported and stretched graphene membranes on different patterned substrates with micro-scale wells. An expected defect density increasing phenomenon, both the $I_D/L$ intensity ratios and the $I_D$ intensities of the D mode with increase in the test points approaching the well, has been observed in substrate-supported graphene membranes near wells. Theoretical and experimental comparison well validates the rationality of the modified model. The discussions demonstrate that the initial defects in graphene membranes can further propagate under the built-in tension resulting in the increase of the defect density. The underlying mechanism is that the built-in stresses can increase the adequate energy for defect propagations, which causes the increase of the defect density. The work can provide a theoretical foundation for Raman characterization method of defect propagations in stretched graphene and applications of defective graphene-based nanodevices.

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Appendix A. Supplementary data

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References


