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# Effect of uni-axial strain on THz/far-infrared response of graphene

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We present polarized optical transmission study of uniaxially strained large scale graphene in THz/far-infrared (IR) frequency region. Graphene was supported on stretchable polyethylene substrate and they were elongated up to 20% ( $\Delta L/L_o = 0.2$ ) by applying tensile force. For the IR light polarized along the strain direction ( $E_{IR} \parallel \text{strain}$ ), the optical conductivity  $\sigma_1(\omega)$  of graphene changes from Drude response into strongly non-Drude-like behavior with a peak formed at finite energy  $\sim 10$  meV. In contrast, the coherent Drude conductivity is preserved along the direction perpendicular to the strain ( $E_{IR} \perp \text{strain}$ ). Possible origin of the strain-induced non-Drude  $\sigma_1(\omega)$ -behavior is discussed. © 2012 American Institute of Physics. [doi:10.1063/1.3680095]

The remarkable mechanical flexibility is an important advantage of graphene in addition the other well known property such as the high carrier mobility and the near-perfect optical transparency. Large scale CVD-grown graphene can be stretched as large as 25% by applying uni-axial tensile strain without interrupt by structural failure.<sup>1</sup> The high dc-conductivity and the carrier mobility of un-strained pristine graphene are preserved over many cycles of repeated deformation without being degraded. The excellent mechanical flexibility under enforced stretching/bending when combined with the good electrical-optical property enables large scale application of graphene as flexible, foldable transparent conductor/display, and electronic paper.<sup>2,3</sup>

When graphene is uniaxially stretched Kim *et al.* showed that dc-resistance increases along the strain direction whereas it remains unchanged for the perpendicular direction. The anisotropic dc-transport suggests that the uniaxial strain induces certain effects on the Dirac band and the massless Dirac particle. Theory predicts that the stretching deforms the linear Dirac band leading to anisotropic Fermi surface. Also the Dirac point shifts in the k-space. Further, at high-strain, opening of band gap is expected.<sup>4,5</sup> The lattice elongation also changes the Fermi surface and the Fermi velocity of the massless Dirac carrier,<sup>6</sup> which will contribute to some extent to the observed anisotropic dc-transport. In contrast to the theoretical studies that have accumulated significant results and predictions, experimental investigation of the uni-axial strain effect, in particular on the large scale graphene (LSG), is yet very rare. From the dc-transport result, one can infer that the strain-induced effect may also appear in ac-response of graphene such as in the transmission spectrum at infrared (IR) frequency. To probe the strain effect in the finite frequency domain is important not only for the fundamental understanding but also, ultimately, for the flexible electrical/optical device application of graphene.

In this work, we performed far-infrared transmission measurement on large scale CVD graphene under uniaxial strain. In un-strained pristine graphene, the free carrier shows Drude response in this frequency range.<sup>7</sup> Previously, we have studied the Drude peak of CVD graphene on various substrates such as SiO<sub>2</sub>, metal-oxide compounds, and organic polymer thin films which revealed that the Drude response is observed consistently for all types of the substrate materials.<sup>8</sup> Here, we perform polarized-transmission measurement to study how the Drude response changes as the uniaxial strain is applied.

LSG was grown by CVD method and subsequently transferred on IR-transparent and stretchable low density polyethylene (LDPE) substrate.<sup>9</sup> We mounted LSG/LDPE on home-made mechanical device and applied tensile force to stretch graphene as  $\Delta L/L_o = 0\% \rightarrow 5\% \rightarrow 10\% \rightarrow 15\% \rightarrow 20\%$  ( $L_o =$  initial length 1 cm). Adhesion of graphene to LDPE was strong enough and no slippage of graphene took place during the stretching. Fig. 1 shows schematic view of IR transmission measurement. IR was selectively polarized along parallel or perpendicular direction to the uniaxial strain,  $E_{IR} \parallel \text{strain}$ , and  $E_{IR} \perp \text{strain}$ , respectively. The transmission spectrum through LSG/LDPE sample ( $= T_s$ ) and LDPE reference ( $= T_r$ ) were measured at each  $\Delta L/L_o$  using Fourier transform interferometer (FTIR, Bomem DA8). We used Hg-lamp as far-IR source and liquid He-cooled bolometer as detector. All spectra were taken at room-temperature. From  $T_s$  and  $T_r$ , we obtain the relative transmission  $T_R(\omega) = T_s/T_r$ . The black curve in Fig. 2(a) shows  $T_R(\omega)$  of un-strained graphene  $\Delta L/L_o = 0$ . The transmission level decreases as frequency decreases which represents the Drude absorption by the free carrier of graphene. The periodic wiggle of  $T_R(\omega)$  is due to interference of IR in the substrate. LDPE is an insulator with constant transmission level (87%, not shown) in the far-IR region. To compare with the data, we calculate  $T_R(\omega)$  of LSG/LDPE theoretically. Here, we use Drude conductivity for the graphene layer as

$$\sigma(\omega) = \frac{\omega_p^2}{4\pi} \cdot \frac{i}{\omega + i\Gamma}, \quad (1)$$

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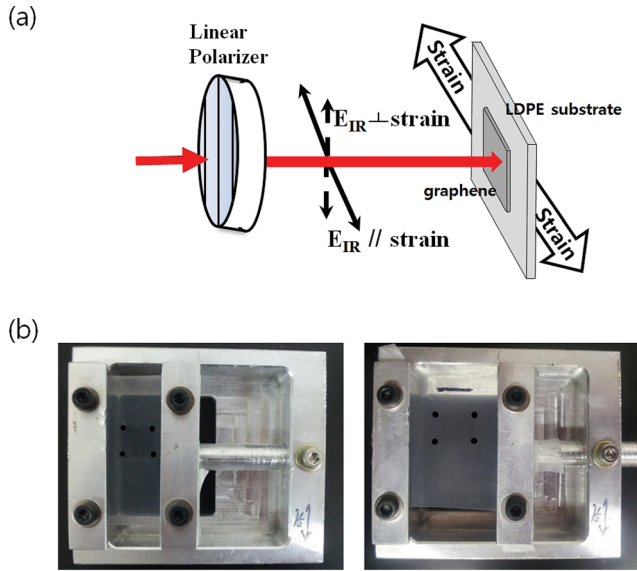


FIG. 1. (Color online) (a) Schematic view of IR transmission measurement on strained graphene. Large scale graphene is supported on IR transparent and stretchable LDPE substrates which are uniaxially stretched by tensile strain. IR is selectively polarized either in parallel ( $E_{IR} // \text{strain}$ ) or perpendicular ( $E_{IR} \perp \text{strain}$ ) direction to the applied strain. (b) Graphene/substrate mounted on mechanical vice before (left) and after (right) the strain is applied. The four small dots in the pictures indicate the (barely visible) monolayer graphene sheet.

where  $\omega_p^2$  and  $\Gamma$  are plasma frequency and carrier scattering rate of free carrier, respectively.<sup>7,8,10</sup> For LDPE layer (0.1 mm thick), dielectric constant  $\epsilon_1 = 2.9$  ( $n = 1.7$ ) is used and the interference effect is included. We used the REFIT simulation program for the two-layer transmission calculation.

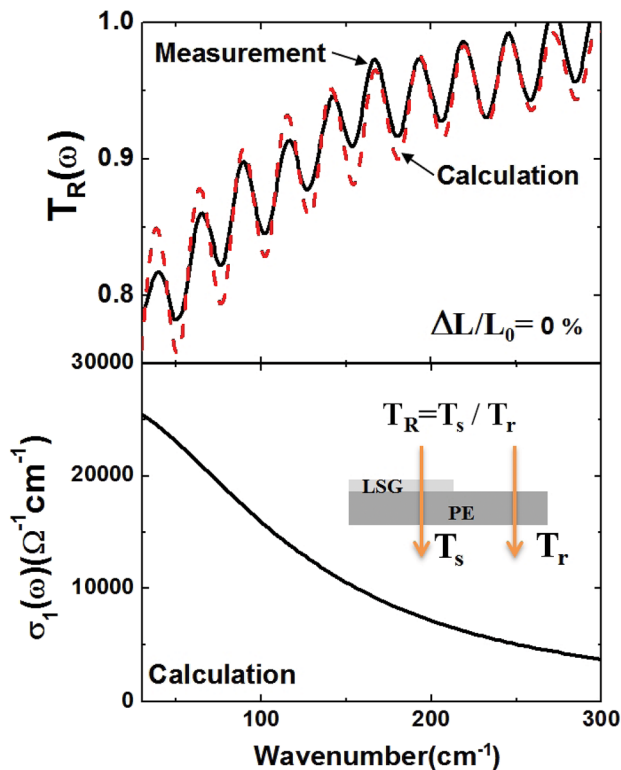


FIG. 2. (Color online) (a) Normalized transmission data  $T_R(\omega)$  of graphene before the strain is applied,  $\Delta L = 0$  (black solid curve) and theoretical simulation result of  $T_R(\omega)$  (red curve). (b) Drude conductivity  $\sigma_1(\omega)$  used in the  $T_R(\omega)$  calculation.

tion.<sup>11</sup> When  $\omega_p$  and  $\Gamma$  are adjusted to  $\omega_p = 11\,000\text{ cm}^{-1}$  and  $\Gamma = 97\text{ cm}^{-1}$ , the calculated  $T_R(\omega)$  reproduces the data closely. These  $\omega_p$  and  $\Gamma$  correspond to the carrier density  $N = 0.7 \times 10^{12}\text{ cm}^{-2}$  and mobility  $\mu = 5900\text{ cm}^2/\text{V} \cdot \text{s}$ .<sup>8</sup> The curve in Fig. 2(b) shows the real part of the Drude conductivity  $\sigma_1(\omega)$  used in the  $T_R(\omega)$  simulation.

Fig. 3 shows  $T_R(\omega)$  of the graphene stretched by  $\Delta L/L_0 = 0.15$ . For IR polarized parallel to the strain direction ( $E_{IR} // \text{strain}$ ),  $T_R(\omega)$  falls faster than the unstrained curve and rises up at low frequency  $\omega < 150\text{ cm}^{-1}$ . To test whether this change is caused by graphene or by the substrate, we repeated the stretching experiment on the bare LDPE without graphene mounted on it. The inset shows the stretched transmission normalized by the un-stretched one. The graphene/LDPE data exhibits the low- $\omega$  upturn while it is absent in LDPE data, which concludes that the non-Drude behavior is coming from the graphene under strain. The periodic wiggles in the transmission ratio are due to that LDPE becomes thinner by the stretching and subsequently the interference pattern is changed. For IR polarized perpendicular to the strain ( $E_{IR} \perp \text{strain}$ ), the Drude response is preserved.

In Fig. 4, the left column displays the detailed change of  $T_R(\omega)$  along with the step-by-step increase of strain. As the strain is applied, the Drude-fall becomes flat at  $\omega < 100\text{ cm}^{-1}$  and then evolves into the upturn behavior. We fit  $T_R(\omega)$  using the ReFit analysis program to obtain optical conductivity  $\sigma_1(\omega)$  of graphene, making use of the Kramers-Kronig constrained two-layer variational dielectric function method.<sup>11</sup>  $\sigma_1(\omega)$  displayed in the right column shows that the Drude conductivity is suppressed at low  $\omega$ -region at low

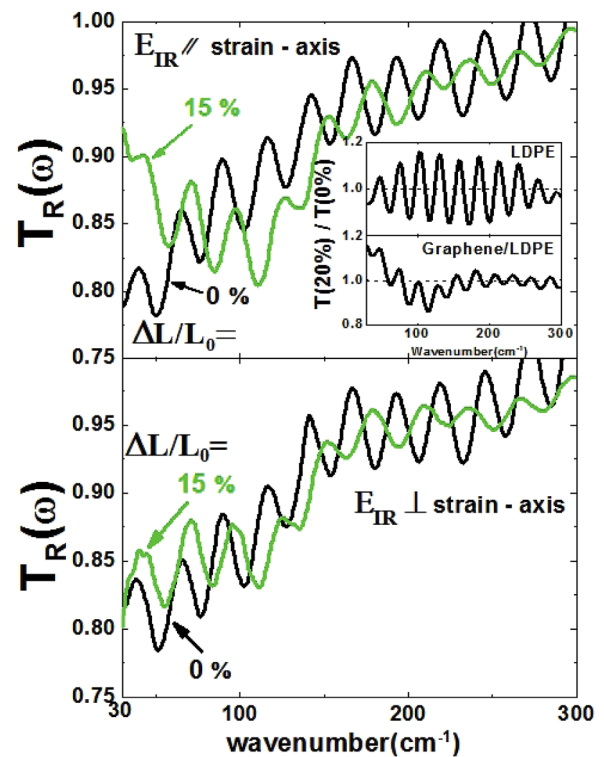


FIG. 3. (Color online)  $T_R(\omega)$  measured with the uniaxial strain applied by  $\Delta L/L_0 = 0.15$ . IR is polarized in parallel ( $E_{IR} // \text{strain}$ ) and perpendicular ( $E_{IR} \perp \text{strain}$ ) to the strain direction, respectively. Inset shows the ratio of transmission after and before the stretching for the graphene/substrate and for the bare LDPE substrate.



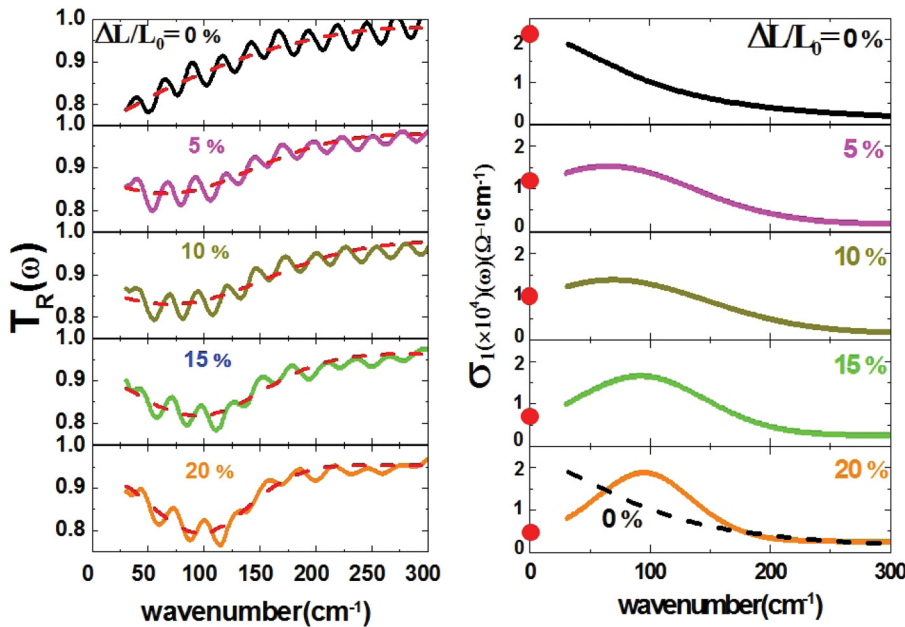


FIG. 4. (Color online) (left):  $T_R(\omega)$  for  $E_{IR}$ // strain polarization with increasing uniaxial strain  $\Delta L/L_0 = 0\%$ , 5%, 10%, 15%, and 20%. (right): Optical conductivity  $\sigma_1(\omega)$  of graphene extracted by fitting  $T_R(\omega)$ . The dot symbols at  $\omega = 0$  represent dc-conductivity  $\sigma(0)$  estimated by extrapolating  $\sigma_1(\omega)$  to  $\omega \rightarrow 0$ .

strain. The suppression continues and eventually a peak appears at finite energy  $\hbar\omega = 100 \text{ cm}^{-1}$  (12 meV). Note that the peak is formed by gaining its spectral weight from that lost in the low- $\omega$  region as can be seen by comparing the two  $\Delta L/L_0 = 0\%$  and 20% data. The low- $\omega$  suppression also leads to the decrease of the dc-conductivity  $\sigma(0)$ . We estimate  $\sigma(0)$  by extrapolating  $\sigma_1(\omega)$  to  $\omega = 0$  as indicated by the red dots. It changes from 2 to  $0.5 \Omega^{-1} \cdot \text{cm}^{-1}$  for  $0 \rightarrow 20\%$ , resulting in the conductivity ratio  $\sigma(20\%)/\sigma(0\%) = 0.25$ . This ratio is consistent with the measured dc-resistance result  $\sigma(20\%)/\sigma(0\%) = 0.3$  in Ref. 1.

When graphene is stretched, the C-C bond of the hexagonal lattice is elongated. As result, the carrier hopping and, accordingly, the dc-conductivity  $\sigma(0)$  will decrease along the strain direction. Noting that Drude conductivity in Eq. (1) is related with  $\sigma(0)$  as  $\sigma(\omega) = \frac{\sigma(0)}{1+i(\frac{\omega}{\Gamma})}$ , one can expect that even

under strain  $\sigma_1(\omega)$  should maintain the Drude form (with possible change of  $\Gamma$ ) although the Drude strength can be reduced. However, this simple picture is denied by the non-Drude  $\sigma_1(\omega)$  observed, which points to the need of advanced theory. First, principle calculation showed that the uniaxial strain brings about several effects on the band structure of graphene, namely, shift of the Dirac point in k-space, anisotropic group velocity, change in the density of state and the work function. These multiple effects can be, fortunately, integrated into the single equation—Weyl’s equation—which however goes beyond the simple tight-binding approximation.<sup>4</sup> Therefore, one should compute optical conductivity based on it and investigate whether the non-Drude  $\sigma_1(\omega)$  is produced within the band theory. In particular, band calculation predicts an energy gap of  $\sim 45 \text{ meV}$  opened at  $\sim 26\%$  of uniaxial strain.<sup>4</sup> The 10 meV peak in our  $\sigma_1(\omega)$  may be signalling the gap opening. In the mean time, in low-dimensional solid with strong correlation, non-Drude  $\sigma_1(\omega)$  and the gap-like low- $\omega$  suppression is often observed.<sup>12,13</sup> Such effect is determined generally by  $U/W$  where  $U$  and  $W$  represent the correlation strength and the carrier’s kinetic energy, respectively. If we conjecture that the on-site  $U$  is playing some role, although

not significant, in graphene, then the increase of  $\Delta L/L_0$  will reduce  $W$  and consequently enhance  $U/W$  leading to non-Drude behavior such as seen in  $\sigma_1(\omega)$ .

In conclusion, we studied effect of uniaxial strain on far-IR response of large scale graphene from polarized transmission measurement. As graphene is stretched out, Drude optical conductivity  $\sigma_1(\omega)$  exhibits the anisotropic change: For IR polarized along the strain direction ( $E_{IR}$ //strain),  $\sigma_1(\omega)$  evolves into non-Drude profile resulting from the low- $\omega$  suppression and the prominent peak forming at  $\hbar\omega = 10 \text{ meV}$ . The decrease of dc-conductivity observed previously in dc-transport measurement results from the non-Drude suppression of  $\sigma_1(\omega)$ . For IR polarized perpendicular to the strain ( $E_{IR} \perp$  strain), the Drude behavior of the un-strained graphene is preserved up to  $\Delta L/L_0 = 0.2$ . Further study is needed to understand the origin of the non-Drude incoherent  $\sigma_1(\omega)$  for  $E_{IR}$ //strain. As first step toward it, we are pursuing calculation of  $\sigma_1(\omega)$  within the band theory for elongated graphene. Finally, the response of  $\sigma_1(\omega)$  under strain provides useful information for large scale and flexible THz/far-IR device application of graphene.

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