Graphene for Tunable Nanophotonic Resonators

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(Invited Paper)

Abstract—Local electro-optic modulation of nanophotonic structures is important for many applications. Here, we show that one can achieve efficient tunability of the nanostructures by coupling them to graphene. The easy electrical control of graphene allows a large modulation of these graphene-clad nanophotonic devices. We describe electrostatic tuning of a metallic as well as a dielectric resonator coupled to graphene. A large change, both in resonator linewidth and resonance frequency is observed. The experimental data match the theory very well indicating that the changes in the resonator are solely coming due to graphene.

Index Terms—Plasmons, cavity resonators.

I. INTRODUCTION

THE ability to manipulate light at the nanoscale enables many different applications, including but not limited to, building efficient optical interconnect system [1] and sensors [2]. To this end, significant progress in the field of nanophotonics has been achieved in the past two decades. With sophisticated nanofabrication techniques, feature size approaching tens of a nanometer is being designed reliably, and light can be confined to an unprecedentedly small volume, enabling very strong light-matter interaction [3]. However, one major challenge in these nanostructures is the tunability: how much the amplitude or the phase of the light can be changed with low energy and fast speed? Such tunability can be realized by incorporating the field-effect transistor structure in a photonic device, where semiconductors inthe vicinity of a device work as an active absorption medium with different electrostatic gating voltages [4]. However, modulation at the optical frequency has been hampered by the weak optical response of the gate-induced free carriers in a semiconductor.

Graphene, a 2-D material of carbon atoms arranged in a honeycomb geometry, has generated considerable interest in recent years for its unusual electronic and optical property [5]. Due
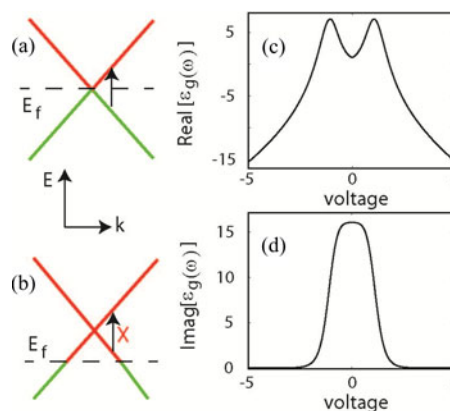
Fig. 1. (a) Band diagram of graphene showing the linear dispersion of the carriers near the Dirac point, where the conduction and the valence band meet. The Fermi energy is at the Dirac point, and graphene can absorb a light with the frequency corresponding to the arrow in the figure. (b) If the Fermi energy is shifted under the application of an electric field, then the light cannot get absorbed in graphene anymore. (c), (d): Real and imaginary part of the refractive index of graphene as a function of the applied gate voltage using ion gel as the gating media.

to its unique atomic arrangement, the conduction and valance bands meet at a single point, known as the Dirac point, in the dispersion diagram [see Fig. 1(a)]. Close to this Dirac point the band dispersion of the electrons and holes is linear, in contrast to a parabolic dispersion in other materials. Such a band structure leads to an extremely high carrier mobility, enabling high-speed operation. Due to the linear dispersion, graphene can absorb light over a broad frequency range. Moreover, the density of states of carriers near the Dirac point is low, and hence the Fermi energy of graphene can be tuned significantly with relatively low electrical energy [6], which in turn changes the refractive index of graphene [see Fig. 1(a) and (b)]. Thus, combining graphene with nanophotonic structures has tremendous potential for achieving electro-optical tunability of light transmission. In this paper, we present experimental result on two different tunable nanophotonic resonators, a single gold nanorod, and a silicon photonic crystal cavity, where the tunability is achieved by electrical gating of graphene placed on top of the nanorod and the cavity. Although, in this paper we mainly focus on nanophotonic resonators, such an electro-optic tunability can be used in conjunction with other nanophotonic structures for applications like phased-array optical antenna [7] or an optical display [8].

II. ELECTRICAL CONTROL OF GRAPHENE

In this section, we analyze the effect of an electric field on graphene. We will consider only the contribution of π-electrons, because only they can be modified significantly through

electrical gating. The complex dielectric function $\varepsilon_g(\omega)$ of graphene π-electrons can be obtained from the corresponding optical conductivity $\sigma(\omega) = \sigma_1(\omega) + i\,\sigma_2(\omega)$ using the relation $\varepsilon_g(\omega) = 1 + i\,\sigma(\omega)/(\omega\varepsilon_o d_g)$, where d_g is the thickness of the graphene layer. As the graphene layers are separated by ~ 0.34 nm in graphite, we use $d_g \sim 0.34$ nm for the calculation. Under random phase approximation and by using Kramer–Kronig relation, we can write the real and imaginary part of the optical conductivity σ as [9]

$$\sigma_1(\omega) = f(\omega) + \frac{q^2\Gamma}{2\pi\hbar^2}\frac{1/\tau}{\omega^2 + (1/\tau)^2}\log\left(2\cosh\frac{2E_F}{\Gamma}\right)$$

$$\sigma_2(\omega) = -\frac{2\omega}{\pi}\int_0^\infty \frac{f(\omega') - f(\omega)}{\omega'^2 - \omega^2}d\omega$$

$$+ \frac{q^2\Gamma}{2\pi\hbar^2}\frac{\omega}{\omega^2 + (1/\tau)^2}\log\left(2\cosh\frac{2E_F}{\Gamma}\right)$$

with

$$f(\omega) = \frac{q^2}{8\hbar}\left[\tanh\left(\frac{\hbar\omega + 2|E_F|}{\Gamma}\right) + \tanh\left(\frac{\hbar\omega - 2|E_F|}{\Gamma}\right)\right].$$

Here q is the electronic charge and Γ is the interband transition broadening. The free carrier scattering rate $1/\tau$ is in terahertz range, and can be neglected for electromagnetic waves in the near infrared and visible frequencies. We plot the real and imaginary part of the dielectric function $\varepsilon_g(\omega)$ as a function of the applied voltage assuming the wavelength of interest; in this case the wavelength is 1.5 μm [see Fig. 1(c) and (d)]. We find that that the imaginary part of $\varepsilon_g(\omega_r)$ (which is responsible for the light absorption) decreases monotonically with increasing voltage, especially when the $2|E_F|$ is larger than the resonance energy $E_r = \hbar\omega_r$ and blocks the relevant interband transitions. On the other hand, the real part of $\varepsilon_g(\omega_r)$ is a nonmonotonic function of the applied voltage. This is because $\sigma_2(\omega)$ has significant contribution both from intraband and interband transition. Contribution from the intraband transition $\left(\frac{q^2\Gamma}{2\pi\hbar^2}\frac{\omega}{\omega^2 + \left(\frac{1}{\tau}\right)^2}\log\left(2\cosh\frac{2E_F}{\Gamma}\right)\right)$ increases monotonically with increasing $|E_F|$. However, interband transition contribution has a minimum at $2|E_F| = E_r$ [see Fig. 1(c)]. We note that in our experiment, we are applying a voltage V to the graphene layer, and the Fermi level of graphene and applied voltage can be related by the equation

$$E_F = \hbar v_f \sqrt{\pi\left(n_o + \frac{C|V|}{q}\right)}$$

where C is the effective capacitance per unit area, $v_f \sim 10^6$ m/s is the Fermi velocity for graphene, and n_o is the intrinsic carrier concentration. These changes in the refractive index of graphene can modify the nanophotonic resonances, when the resonators are coupled with graphene. More specifically, the resonance linewidth depends on the imaginary part of the refractive index, and the resonance frequency depends on the real part of the refractive index. In the experiment, we find that the resonance wavelength (λ_R) and the cavity linewidth (Γ_R) have a linear relation with the complex dielectric constant of graphene, i.e., $\Gamma_R = \Gamma_R^0 + \alpha \text{Im}\left[\varepsilon_g(\omega)\right]$ and $\lambda_R = \lambda_R^0 + \beta\text{Re}\left[\varepsilon_g(\omega)\right]$.
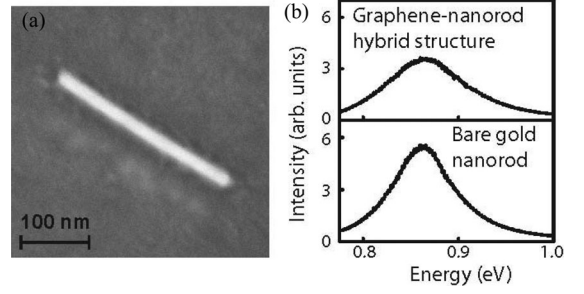

Fig. 2. (a) A representative high-resolution SEM of a single gold nanorod covered by graphene. (b) Rayleigh scattering spectra of a graphene-nanorod hybrid structure (top) and a bare gold nanorod (bottom) [Adapted with permission from [12]. Copyright (2012) American Chemical Society.]

III. METALLIC CAVITY: GOLD NANOROD

First, we analyze the effect of graphene on a metallic resonator. When surface plasmon, collective oscillation of free carriers, is confined in a nanostructure, it forms electromagnetic resonance. Surface plasmon resonance, with its unique capability to concentrate light into subwavelength volume, has enabled great advances in photon science. Despite fascinating potential for applications ranging from nanoantenna to metamaterials, most active device demonstrations have been shown at terahertz frequency due to weak free-carrier response in a semiconductor at optical frequencies [10].

On the other hand, graphene has a unique and tunable optical property over a broad frequency range as described in the previous section. This enables graphene to tune plasmon resonance of metallic nanostructures at optical frequency. In our experiment, we demonstrate a model system, where plasmon resonance in a gold nanorod at optical frequency is controlled efficiently by gating graphene. We note that, the change in the plasmonic resonance via gating graphene is also demonstrated in midinfrared frequencies elsewhere [9], [11]. Fig. 2(a) shows a scanning electron micrograph (SEM) of a hybrid structure, where graphene drapes nicely over a gold nanorod on a substrate. A gold nanorod with a resonance wavelength at 1.5 μm is chosen for demonstration at optical frequency. The resonance feature is found to be damped in a hybrid structure with graphene [see Fig. 2(b)]. The figure shows that the hybrid structure has a significantly broader full width half maximum than a bare gold nanorod (93 meV in the hybrid structure and 70 meV in a bare gold nanorod).

Such damping originates from energy loss due to interband absorption in graphene. Considering the thickness of graphene, ~ 0.34 nm, it is quite surprising that plasmonic resonance can be changed by such a large amount. This underlines the remarkably strong interband optical absorption of graphene at optical frequencies. As pointed out in the introduction, such strong optical absorption of graphene can be conveniently modulated by electrical gating. For experimental demonstration, electrical gating is enabled by ionic liquid as top electrolyte. Fig. 3(a) shows that damped plasmon resonance becomes sharp resonance with increasing gate voltage. The modulation of plasmon resonance with gate-induced change in Fermi level is summarized in Fig. 3(b) [bottom]. It shows nice switching behavior with a threshold at a gate voltage, where $2|E_F|$ becomes a plasmon
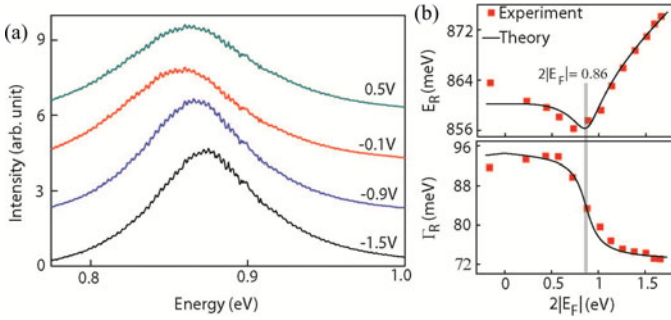
Fig. 3. (a) Rayleigh scattering spectra of the hybrid structure at different gate voltage. (b) Gate-dependent resonance frequency shift (top) and linewidth change (bottom). Experimental data (red dots) agrees well with theory (black solid line). [Adapted with permission from [12]. Copyright (2012) American Chemical Society.]
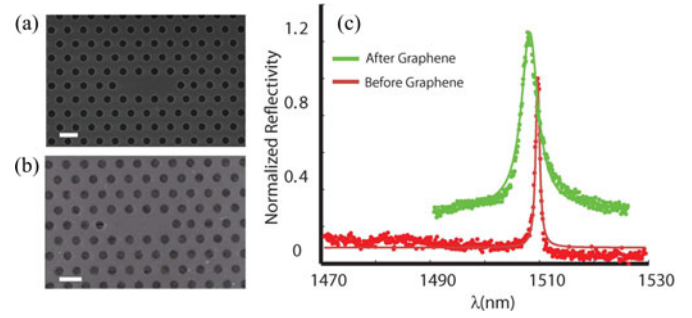

Fig. 4. Effect of graphene on a silicon photonic crystal cavity: (a) and (b) the SEM images of the cavity fabricated in SOI platform. (c) The reflectivity spectra of the cavity with and without graphene. With graphene the spectrum is significantly broadened. [Adapted with permission from [19]. Copyright (2013) American Chemical Society.]

resonance energy. The theoretical model explains linewidth change quantitatively with an excellent agreement (black solid curve). On the other hand, resonance energy has changed by 20 meV with gate voltages. This resonance energy shift can be explained by change in gate-induced local dielectric screening effect near a nanorod. In theory, such absorption and dielectric screening can be quantified by a complex dielectric constant. Our theoretical model, where resonance feature changes in a linear fashion with change with complex dielectric constant, explains experimental data with excellent agreement.

Besides an efficient modulation of plasmon resonance, numerical simulation (finite-element method) gives interesting prediction [12]. In fact, a significant amount of resonance feature changes results from the two ends of the nanorod, where electric field is enhanced, so-called hot spots. Our simulation result (not shown here) shows that only contribution from the hot spots (defined by two 20 nm × 20 nm area) can lead to a 4% increase in plasmon scattering intensity, an easily observable effect.

IV. DIELECTRIC CAVITY: PHOTONIC CRYSTAL CAVITY

Although a metallic resonator can be modulated significantly using graphene, as shown in the last section, the large loss in metal limits its utility for many applications, for example, electro-optic modulation. For such modulation dielectric nanostructures are better suited. Hence, in this section, we focus on dielectric cavities, where one can achieve a much larger quality factor compared to a metallic cavity. More specifically, we tune a graphene-clad photonic crystal cavity electrostatically. We note that graphene-waveguide system for electro-optic modulation is recently demonstrated [13], but with a cavity we can largely reduce the physical footprint of the device.

The experiments are performed with linear three hole defect silicon photonic crystal cavities fabricated in silicon-on-insulator (SOI) platform. The device thickness d is 250 nm, with photonic crystal lattice periodicity $a = 450$ nm, and radius $r = 90$ nm. The two holes at the end of the cavities are shifted by $0.15a$ to increase the cavity quality factor. The photonic crystals are fabricated by electron-beam lithography, followed by plasma etching and finally by removing the silicon oxide underneath, via buffered oxide etching to make a free standing silicon-photonic crystal membrane. On the top of the cavities, we transferred a large-area graphene grown by chemical vapor deposition using

the standard growth and transfer processes [14], [15]. For electrostatic gating of grapheme, we used a top electrolyte gating with ions [16]. Fig. 4(a) and (b) shows the SEM images of a fabricated photonic crystal cavity before and after the graphene transfer, respectively.

We characterize the photonic crystal cavities in cross-polarized reflectivity measurement setup with a supercontinuum laser, where the cavity is kept at a 45° angle, the incident probe laser is vertically polarized, and we collect horizontally polarized light [17]. We note that our cavity is linearly polarized, and thus by using a cross-polarized reflectivity, we ensure that the collected light consists of only the light that circulated inside the cavity. The collected light is analyzed by a spectrometer equipped with an InGaAs array detector. A quality (Q) factor of ∼1000–1500 is observed for the fabricated cavities without graphene. We note that although a much higher quality factor can be obtained in a silicon photonic crystal cavity (∼20 000), we want to keep the Q-factor moderate to achieve a relatively large spectral bandwidth as well as a higher speed of modulation. With graphene on top, the Q-factor drastically reduces to ∼300–500, as shown in Fig. 4(c). The significant broadening of the cavity linewidth arises mainly from the graphene absorption as noted previously. This is also consistent with another recent experimental observations on graphene-photonic crystal cavity system [18].

Then, we study the effect of the electric field on the graphene-cavity device. We gate the graphene layer by means of the ion-gel (refractive index ∼1.43) [20], that we spin-coat on the device. Fig. 5 shows the effect of the electric field on the graphene-cavity device. We simultaneously measure the cavity reflection spectrum and graphene resistance, while varying the gate voltage at a step of 10 mV/s. Fig. 5(a) shows the cavity reflectivity spectra for different voltages. A narrowing of the cavity linewidth as well as an increase in the cavity reflectivity is clearly observed with an increased gating of the graphene. We fit all the spectra with a Lorentzian line-shape to extract the cavity-resonance frequencies and cavity linewidths. Fig. 5(b) and (c) shows the cavity linewidths and the resonance frequencies as a function of the applied voltage. From the transport data (not shown here), we find that the charge neutral point is at around 0.5 V. This small deviation from the 0 V (as expected from an undoped graphene) can be ascribed to the slight p-doping of graphene during the graphene transfer process. We observe a
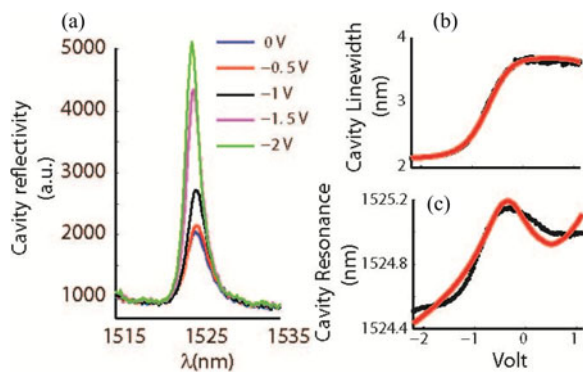
Fig. 5.   (a) Effect of the electric field on the cavity resonance: with an increasing electric field we observe a narrowing of the cavity linewidth as well as increase in the reflectivity. With a Lorentzian fitting to the cavity spectra we can estimate. (b) the cavity linewidth and (c) the cavity resonance. They follow very closely the imaginary and the real part of the refractive index of graphene showing that the change is solely coming due to graphene. [Adapted with permission from [19]. Copyright (2013) American Chemical Society.]

narrowing of the cavity linewidth with increasing voltage, consistent with the fact that increased gating of the graphene reduces its absorption. Both the linewidth and the resonance wavelength match the theory well [shown by the red line in Fig. 5(b) and (c)], as explained in Section II. We also confirmed that in this voltage range, the ion-gel does not affect the cavity resonance (data not shown here).

However, ion-gel gating as used in the experiment cannot be employed for fast dynamic electro-optic modulation due to the slow response of ions under the electric field. Instead, one should employ semiconductor field-effect transistor structure to gate graphene and achieve ultrafast electro-optic modulation. Our simulation suggests that a graphene-cavity device modulated at a speed of 100 GHz with 10 s of femto joule energy should be achievable with current state-of-the-art nanofabrication.

V. Conclusion

We experimentally demonstrate electrostatic tuning of graphene-clad nanophotonic resonators. With an increased light-matter interaction in cavity, even a single-atom-thick graphene can significantly modify the nanophotonic resonators. We believe, such tunability can be exploited in conjunction with other nanophotonic structures for various applications like optical interconnect, phased array optical antenna, or 3-D display.

References

[1] D. A. B. Miller, "Device requirements for optical interconnects to silicon chips," *Proc. IEEE*, vol. 97, no. 7, pp. 1166–1185, Jul. 2009.
[2] J. N. Anker, W. P. Hall, O. Lyandres, N. C. Shah, J. Zhao, and R. P. Van Duyne, "Biosensing with plasmonic nanosensors," *Nat. Mater.*, vol. 7, pp. 442–453, 2008.
[3] K. J. Vahala, "Optical microcavities," *Nature*, vol. 424, pp. 839–846, 2003.
[4] Q. Xu, B. Schmidt, S. Pradhan, and M. Lipson, "Micrometre-scale silicon electro-optic modulator," *Nature*, vol. 435, pp. 325–327, 2005.
[5] A. K. Geim and K. S. Novoselov, "The rise of graphene," *Nat. Mater.*, vol. 6, pp. 183–191, 2007.
[6] F. Wang, Y. Zhang, C. Tian, C. Girit, A. Zettl, M. Crommie, and Y. R. Shen, "Gate-variable optical transitions in graphene," *Science*, vol. 320, pp. 206–209, Apr. 11, 2008.
[7] J. Sun, E. Timurdogan, A. Yaacobi, E. S. Hosseini, and M. R. Watts, "Large-scale nanophotonic phased array," *Nature*, vol. 493, pp. 195–199, 2013.
[8] D. Fattal, Z. Peng, T. Tran, S. Vo, M. Fiorentino, J. Brug, and R. G. Beausoleil, "A multi-directional backlight for a wide-angle, glasses-free three-dimensional display," *Nature*, vol. 495, pp. 348–351, 2013.
[9] N. K. Emani, T.-F. Chung, X. Ni, A. V. Kildishev, Y. P. Chen, and A. Boltasseva, "Electrically tunable damping of plasmonic resonances with graphene," *Nano Lett.*, vol. 12, pp. 5202–5206, 2012.
[10] L. Ju, B. Geng, J. Horng, C. Girit, M. Martin, Z. Hao, H. A. Bechtel, X. Liang, A. Zettl, Y. R. Shen, and F. Wang, "Graphene plasmonics for tunable terahertz metamaterials," *Nat. Nanotechnol.*, vol. 6, pp. 630–634, 2011.
[11] Y. Yao, M. A. Kats, P. Genevet, N. Yu, Y. Song, J. Kong, and F. Capasso, "Broad electrical tuning of graphene-loaded plasmonic antennas," *Nano Lett.*, vol. 13, pp. 1257–1264, 2013.
[12] J. Kim, H. Son, D. J. Cho, B. Geng, W. Regan, S. Shi, K. Kim, A. Zettl, Y.-R. Shen, and F. Wang, "Electrical control of optical plasmon resonance with graphene," *Nano Lett.*, vol. 12, pp. 5598–5602, 2012.
[13] M. Liu, X. Yin, E. Ulin-Avila, B. Geng, T. Zentgraf, L. Ju, F. Wang, and X. Zhang, "A graphene-based broadband optical modulator," *Nature*, vol. 474, pp. 64–67, 2011.
[14] X. Li, W. Cai, J. An, S. Kim, J. Nah, D. Yang, R. Piner, A. Velamakanni, I. Jung, E. Tutuc, S. K. Banerjee, L. Colombo, and R. S. Ruoff, "Large-area synthesis of high-quality and uniform graphene films on copper foils," *Science*, vol. 324, pp. 1312–1314, Jun. 5 2009.
[15] K. S. Kim, Y. Zhao, H. Jang, S. Y. Lee, J. M. Kim, K. S. Kim, J.-H. Ahn, P. Kim, J.-Y. Choi, and B. H. Hong, "Large-scale pattern growth of graphene films for stretchable transparent electrodes," *Nature*, vol. 457, pp. 706–710, 2009.
[16] B. J. Kim, H. Jang, S.-K. Lee, B. H. Hong, J.-H. Ahn, and J. H. Cho, "High-Performance flexible graphene field effect transistors with ion gel gate dielectrics," *Nano Lett.*, vol. 10, pp. 3464–3466, 2010.
[17] D. Englund, A. Majumdar, A. Faraon, M. Toishi, N. Stoltz, P. Petroff, and J. Vučković, "Resonant excitation of a quantum dot strongly coupled to a photonic crystal nanocavity," *Phys. Rev. Lett.*, vol. 104, no. 7, pp. 073904–073913, 2010.
[18] X. Gan, K. F. Mak, Y. Gao, Y. You, F. Hatami, J. Hone, T. F. Heinz, and D. Englund, "Strong enhancement of light—Matter interaction in graphene coupled to a photonic crystal nanocavity," *Nano Lett.*, vol. 12,, no. 11, pp. 5626–5631, 2012.
[19] A. Majumdar, J. Kim, J. Vuckovic, and F. Wang, "Electrical control of silicon photonic crystal cavity by graphene," *Nano Lett.*, vol. 13, pp. 515–518, 2013.
[20] T. Buffeteau, J. Grondin, Y. Danten, and J.-C. Lassègues, "Imidazolium-Based ionic liquids: Quantitative aspects in the far-infrared region," *J. Phys. Chem. B.*, vol. 114, pp. 7587–7592, 2010.

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