Electronic manipulation of near-field nanofocusing in few-layer graphene-based hybrid nanotips

Yanhong Dong (董彦宏)¹, Qing Yang (杨 青)², Guangqing Du (杜广庆)^{1,*}, Feng Chen (陈 烽)^{1,**}, Noor Uddin¹, Dayantha Lankanath¹, and Xun Hou (侯 洵)¹

¹State Key Laboratory for Manufacturing System Engineering and Key Laboratory of Photonics Technology for Information of Shaanxi Province, School of Electronics & Information Engineering, Xi'an Jiaotong University, Xi'an 710049, China

²School of Mechanical Engineering, Xi'an Jiaotong University, Xi'an 710049, China *Corresponding author: guangqingdu@mail.xjtu.edu.cn; **corresponding author: chenfeng@mail.xjtu.edu.cn Received February 12, 2019; accepted April 18, 2019; posted online July 4, 2019

In this Letter, we propose the electronic manipulation of localized surface plasmon resonance for active tuning in near-field nanofocusing. We theoretically studied the excited graphene tuning of the nanofocusing field in fewlayer graphene (FLG)-based hybrid nanotips. It is revealed that the normalized enhanced electric field can be significantly promoted to more than 300 times. It is also observed that resonant peaks can be unprecedently modified by the electron state of excited graphene that is embedded in the substrate. It shows the possibility of flexible tuning of plasmon resonances via controlling the electron excitation state of graphene for specific advanced near-field nanofocusing applications.

OCIS codes: 250.5403, 000.6800, 260.2110. doi: 10.3788/COL201917.072501.

Near-field nanofocusing has drawn much attention in the past few years because of its unique properties of spatially localized near-field enhancement on the nanoscale for potential applications in the fields of nanolithography, photothermal nanosurgery, and nanoimaging $\lfloor \underline{1} - \underline{3} \rfloor$. Various structures for near-field nanofocusing have been studied and reported so far, such as nanowires, nanorings, nanoshells, nanorices, and some other complex structures of bowtie, like the nanotrench [4-7]. There have been tremendous efforts to design and optimize nanostructures in order to obtain more excellent nanofocusing properties to meet specific applications. Among them, the tip-based structures exhibit great importance for the concentration of light into nanoscale spots, and the noble-metaltip-based nanostructures have been widely studied for near-field nanofocusing applications^[8].

Compared to the noble-metal-based nanotip structures, the non-metal-based nanotips, such as semiconductors and dielectrics, are mostly selected as the candidate for plasmonic nanofocusing. The intrinsic optical losses can be significantly reduced in non-metal plasmonic materials⁹. Nevertheless, the lacking of active tuning of plasmonic near-field nanofocusing is still a major challenge due to the fact that the nanofocusing properties of plasmonic nanostructures closely depend on their shape, size, and material properties [10]. On the other hand, it is usually in the cost of the fabrication engineering's complexity for obtaining the ideal geometry, which presents a challenge in the nanofabrication process. In recent years, graphene, a monolayer of hexagonally arranged carbon atoms^[11], has been widely concerned and vigorously pursued for the plasmonic applications because of its unique mechanical, electric, magnetic, and

thermal properties. The electron state of excited graphene can be flexibly tuned by external excitations, such as electrical sources and optical sources, without consideration of the shape or size. The active tuning of near-field nanofocusing in graphene-based hybrid nanotips can be explained as graphene plasmonics hybridization in nanostructures, leading to the possibility of electronic manipulation of localized surface plasmon resonance (LSPR). However, the intrinsic graphene-based plasmon resonances typically exhibit limited tunability due to the fact that the graphene layer is atomically thin and thus only interacts with a very small portion of the plasmon $\mathrm{modes}^{[\underline{12}]}$. It is believed that the few-layer graphene (FLG)-based hybrid nanotips could be more beneficial for electronic manipulation of LSPR compared to atom-layered intrinsic graphene plasmonics^[13]. FLG can be treated as thin graphite, as the atom layer number is less than five^[14]. Experimentally, the FLG can be fabricated by mechanical or ion beam milling of graphite to a few atom layers¹⁵. Especially, the typical metallic nanotips with respect to the excited graphene hybrid nanostructures can be interesting for manipulatable near-field nanofocusing applications. It is currently challenging to obtain more flexible and active tuning of nanofocusing based on the modified plasmon resonance process for the graphene-noble nanotip structure. To the best of our knowledge, the active tunability mechanism in excited graphene-based metallic plasmonic hybrid structures is still an open topic, especially in the FLG-based hybrid system. It is urgent to understand the LSPR tunability mechanism, which is greatly beneficial for promoting near-field nanofocusing applications.

In this Letter, by combining graphene with gold nanotips, we theoretically investigated the electronic manipulation of the LSPR for active tuning near-field nanofocusing in excited graphene-noble nanotips based on the finite element method (FEM). We started with electronic tuning of plasmons via modifying the Fermi energy of excited graphene, which may be modified by an external laser or electrostatic excitation. The potential physical mechanism of the electronic manipulation of the LSPR in the excited graphene-noble hybrid system is examined in details. The significant results of tunable near-field nanofocusing in aspects of the enhancement and resonant peak are explored. The results provide a large possibility for promoting high-performance nearfield nanofocusing applications.

Figure 1 illustrates the cross-section view of the graphenebased nanotip structure. A graphene-coated gold conical tip is suspended above a substrate, in which the FLG is embedded in the SiO_2 substrate. The graphene coated on the gold tip is contributive to near-field nanofocusing, and it can serve as a protective layer thanks to the high mechanical strength of graphene. The scattered light from the nanotip-based structure is totally absorbed through the perfectly matched layer (PML) in the far-field, and the boundary condition at the interface between graphene layers is treated as a continuous one. The field is defined throughout the calculated region by using the Helmholtz equation, except for the PML. Then, the fields are calculated after inserting the gold conical tip and the FLG-based substrate. We used the RF module of a commercial package, COMSOL, to solve for the fields. When the graphene-based hybrid nanostructure is excited by the incident wave, the electron system of the nanotip and the graphene is excited; synchronously the properties on the thermal excitation zone could be actively tuned via modulating the electron state of the FLG. Numerically, the Helmholtz electromagnetic



Fig. 1. Schematic of the simulated FLG-based nanotip hybrid system. The graphene-coated Au tip is modeled as a conical taper terminated by a hemisphere of radius R as its point and elevated a distance d above a SiO₂–graphene–SiO₂ substrate. An electromagnetic plane wave is incident at an angle θ with respect to the surface normal. A 300-nm-thick perfectly matched layer (PML) encloses the simulation domain.

equation is solved by an iterative algorithm based on FEM. In addition, the parameters of excited graphene with respect to the Fermi energy modifications are modeled with the surface optical conductivity that is obtained via the Kubo–Greenwood formula^[16,17]:

$$\tilde{\sigma}_{\text{intra}} = \frac{2ie^2k_BT}{\hbar^2\pi(\omega + i\tau_g^{-1})} \ln\left[2\cosh\left(\frac{E_F}{2k_BT}\right)\right], \quad (1)$$

$$\tilde{\sigma}_{\text{inter}} = \frac{e^2}{4\hbar} \left[\frac{1}{2} + \frac{1}{\pi} \arctan\left(\frac{\hbar\omega - 2E_F}{2k_B T}\right) \right] \\ - \frac{e^2}{4\hbar} \left[\frac{i}{2\pi} \ln \frac{(\hbar\omega + 2E_F)^2}{(\hbar\omega - 2E_F)^2 + (2k_B T)^2} \right].$$
(2)

In the equation set, T is the temperature (300 K), k_B is the Boltzmann constant, and E_F is the Fermi energy of excited graphene. The scattering lifetime of electrons in excited graphene is given by

$$\tau_g = \frac{\mu E_F}{e v_F^2},\tag{3}$$

where the impurity-limited dc conductivity $\mu \approx 10,000 \text{ cm}^2/(\text{V} \cdot \text{s})$, and the graphene Fermi velocity $v_F = 10^6 \text{ m/s}^{(\underline{18},\underline{19})}$.

As for the tunability of this excited graphene-based nanotip hybrid structure, changes of the Fermi energy would lead to a simultaneous change in the interband threshold and the interband transition owing to the unique linear dispersion relation of graphene^[20]</sup>. In other words, the interband transitions are allowed at all frequencies at a Dirac point; however, as the Fermi energy is increased, some of the interband transitions are blocked, hence leading to a narrower resonance. As a result, tunable Fermi energy leads to tunable surface optical conductivity, which includes the contributions from intraband transitions and interband transitions of excited graphene. As reported, most works consider FLG to be graphene sheets with no more than five layers $\frac{21-23}{2}$, and the optical conductivity of FLG is $N\sigma(\omega)^{[21]}$, where $\sigma(\omega)$ is the surface optical conductivity of monolayer graphene (MLG) calculated by the Kubo formula, and N is the number of layers. In this model, we treat the FLG as few-atom-layer graphite, and the force of van der Waals of different atom layers of intrinsic graphite is only considered. However, the interaction of different graphene layers is ignored. As a result, the dielectric function of FLG can be straightforwardly obtained from its optical conductivity $\sigma(\omega)$ and the thickness of graphene $t_a^{[24]}$ as

$$\varepsilon_r(\omega) = i\sigma(\omega)/(\omega\varepsilon_0 t_g).$$
 (4)

Both the real and imaginary parts of permittivity of excited graphene can be modified via tuning the Fermi energy and the thickness of the substrate graphene. The relation does not hold for more than five layers, as interactions between adjacent layers cannot be ignored anymore^[17]. It is reasonable that the introduction of a tunable number of graphene layers would greatly enrich the diversity of both real and imaginary parts of the FLG permittivity, as well as the tuning modes of the FLG-based electronic manipulation of near-field nanofocusing.

The localized electric fields (e-fields) for near-field nanofocusing in the excited mono-graphene-based nanotip structure with respect to different excitation wavelengths are shown in Fig. 2. The permittivity of Au is cited in Ref. [25]. The graphene-based hybrid nanotip is biased to 0.8 eV. As shown, the incident wavelengths range from 15.0 to $15.75 \ \mu m$, leading to different normalized e-field enhancement (from 1.05 to 70.93). It can be seen that the normalized e-fields for near-field nanofocusing vary rapidly in amplitude with the change of incident wavelength. As observed in Fig. 2(f), a more enhanced e-field of 70.93 can be obtained when the incident light travels in the Xdirection at a wavelength of $15.75 \,\mu\text{m}$. The localized e-fields of near-field nanofocusing in a mono-graphene-based tip structure show sensitive tunability of both field distribution and field enhancement. In addition, the nanotip is almost completely dark in the calculated images, which means that there is little electromagnetic field inside. The resonant wavelength of this mono-graphene-based hybrid nanotip structure is in the mid-infrared part of the spectrum, which is one of the typical characteristics of graphene plasmonics ^[20,24,26]. The results will be instructional for sensitively tuning a highly localized strong e-field for promoting the applications of near-field nanofocusing.

Figure <u>3</u> shows the simulation results of the electronic manipulation of near-field nanofocusing in the FLG-based tip structure, as well as the permittivity of graphene with respect to different layers of graphene and different Fermi energies. As shown in Fig. <u>3(a)</u>, the complex permittivity (ε_r) of MLG depends on the incident wavelength and the Fermi energy in the mid-infrared region. The real part of



Fig. 2. Cross-section of mono-graphene-based nanotip structure. Calculated images of the e-field distributions in the case of mono-graphene in the substrate (The curvature radius of the nanotip is R = 30 nm. The vertical spacing between the nanotip and substrate is d = 20 nm, and incident light travels along the X direction).



Fig. 3. Permittivity of MLG with respect to different Fermi energies. (a) The red, blue, black, green, and orange lines are corresponding to 0.1, 0.2, 0.3, 0.4, and 0.5 eV, respectively. (b) The normalized e-field enhancement and resonant frequency of the nanotip hybrid system depending on the Fermi energy of excited graphene. (c) Real and (d) imaginary parts of graphene permittivity with respect to the layers of FLG changing from 1 to 5 (E_F is biased to 0.5 eV).

 ε_r (solid line) is negative, and the imaginary part of ε_r (dotted line) is positive. The inset in Fig. 3(a) shows the details of the imaginary part of ε_r . The real part of ε_r decreases as either the Fermi energy or wavelength increases, while the imaginary part of ε_r presents opposite dependence on the Fermi energies and incident wavelengths. It can be understood from Fig. 3(a) that this MLG with tunable Fermi energy shows relatively low loss due to the calculated value of $|Im(\varepsilon)/Re(\varepsilon)|$ being less than 0.1. Figure $\underline{3}(\underline{b})$ presents the maximal values of the normalized resonant e-field enhancement and the resonant frequency dependence on the Fermi energy of excited graphene. It can be seen that the electronic manipulation of near-field nanofocusing in the FLG-based tip structure possesses strong tuning ability in the normalized resonant field enhancement (from 25.7 to 391.7) and the resonant frequency (from 4.7 to 18.7 THz). It is evident from Fig. 3(b) that the resonant peaks can be modified by the Fermi energy of FLG that is embedded in substrate as well. Figures 3(c) and 3(d) show the variation of real and imaginary parts of graphene permittivity versus incident wavelength. The black arrow indicates the curves group as the number of graphene layers increases from N = 1 to 5. It is seen that $\operatorname{Re}(\varepsilon)$ shifts to higher values when the number of graphene layers increases, while $\text{Im}(\varepsilon)$ shifts to lower values simultaneously. The insets in Figs. 3(c) and 3(d) show the details of $\operatorname{Re}(\varepsilon)$ and $\text{Im}(\varepsilon)$ of permittivity of the FLG, respectively, versus the incident wavelength (from 5 to 20 μ m). It can be observed that the absolute values of $\operatorname{Re}(\varepsilon)$ and $\operatorname{Im}(\varepsilon)$ of the permittivity of MLG are about five times larger than those of five-layer FLG. As the Fermi energy of graphene increases, the carrier concentration of graphene increases consequently and finally causes the blue shifts of the wavelength. It can be explained as a significant role in active tuning of the graphene-noble structure due to plasmon hybriding, resulting in the abundant tunable electron plasmons being compared to that of intrinsic graphene. It can be observed that graphene's conductivity is strongly dispersive and extremely sensitive to the bias Fermi level, which suggests the potential for graphene's usage in tunable terahertz (THz) metamaterials and metadevices.

Figure 4 shows the simulation results of the electronic manipulation of the LSPR for active tuning of near-field nanofocusing in a five-layer FLG-based structure with respect to different Fermi energies. When the Fermi energy varies from 0.7 to 1.4 eV, as shown in Fig. 4(a), a peak-shifted enhancement profile is exhibited, corresponding to different Fermi energies. The resonant peak, namely the normalized resonant e-field enhancement, becomes stronger with the increase of Fermi energy. A blue shift of the resonant wavelength from 24.876 to $17.671 \ \mu m$ is observed with increasing Fermi energy, and the wavelength range is red-shifted compared to that of the mono-graphene-based tip structure in Fig. 2. Figure 4(b) presents the resonant wavelength and Fermi energy dependence on the carrier concentration of the excited FLG. The resonant wavelength decreases rapidly with the increasing carrier concentration, as the carrier concentration is less than 2×10^{17} m⁻². The carrier concentration $n_{a,2D}$ in the graphene sheet is given by^[10]

$$n_{g,2D} = k_F^2 / \pi = \left(\frac{E_F}{\hbar v_F}\right)^2 / \pi.$$
 (5)

An increase in the number of carriers inside the FLG leads to the graphene being more metal-like due to the promoted Fermi energy. As a result, the increase of the carrier concentration of FLG leads to obvious blue shifts in the resonant wavelengths, which is in agreement with the charging effect in previous theoretical studies and experimental measurements. As reported, the carrier



Fig. 4. Simulation results of the electronic manipulation of nearfield nanofocusing in a five-layer FLG-based tip structure. (a) The normalized e-field enhancement spectra with respect to different Fermi energies (The Fermi energy decreases with 0.1 eV step size from left to right). (b) The Fermi energy of FLG (black line) and the resonant wavelength (red line) are plotted as a function of the carrier concentration of FLG. The inset illustrates the calculated image of the localized e-field.

concentration can be tuned by means such as doping and femtosecond laser ionization, and those fundamental researches provide an important foundation for the electronic manipulation of LSPR for actively tuning near-field nanofocusing. The inset in Fig. <u>4(b)</u> shows the calculated image of the localized e-field at $E_F = 1.4$ eV when the incident light travels in the X direction, and the enhanced e-field in the nanofocusing region can be promoted to 304.3 times. The FLG-based hybrid nanotip structure will hold significant potential for promoting a wide range of the near-field nanofocusing applications, as the active tunability is fully taken into account for electronic manipulation of the field distribution.

In conclusion, we have theoretically investigated the electronic manipulation of LSPR for actively tuning the spatially highly concentrated near-field nanofocusing in FLG-based hybrid nanotips. It is revealed that the introduction of multilayered graphene leads to not only the increase of tuning ranges in resonant peaks, but also the promoted e-field enhancement for nanofocusing. It shows that the maximal normalized e-field enhancement in the nanofocusing region of the five-layer FLG-based tip structure reaches up to 304.3 times in the THz band. The resonant wavelengths of the nanofocusing hotspot can be actively modified from 17.671 to 24.876 µm. The results are important for understanding graphene-based plasmonics hybridization and promoting a wide range applications of specific advanced near-field nanofocusing, such as near-field imaging, nanolithography, and photothermal nanosurgery.

This work was supported by the National Key Research and Development Program of China (No. 2017YFB1104700), the National Natural Science Foundation of China (Nos. 51335008, 61775177, and 61475124), the NSAF (No. U1630111), the Collaborative Innovation Center of Suzhou Nano Science and Technology, and the China Postdoctoral Science Foundation (No. 2014M560778).

References

- 1. S. Kawata, Y. Inouye, and P. Verma, Nat. Photon. 3, 388 (2009).
- 2. X. Huang and M. A. J. El-Sayed, Alexandria J. Med. 47, 1 (2011).
- Q. Xu, X. Q. Zhang, Y. H. Xu, C. M. Ouyang, Y. F. Li, J. G. Han, and W. L. Zhang, Chin. Opt. Lett. 16, 050002 (2018).
- G. Du, Q. Yang, C. Feng, L. Yu, B. Hao, J. Yong, and H. J. Xun, Appl. Phys. B **120**, 47 (2015).
- G. Du, Q. Yang, C. Feng, L. Yu, O. Yan, J. Yong, and H. J. Xun, Appl. Phys. A **122**, 185 (2016).
- S. X. Xia, X. Zhai, L. L. Wang, Q. Lin, and S. C. J. Wen, Opt. Express 24, 16336 (2016).
- S. Y. Cao, W. X. Yu, C. Wang, and Y. Q. Fu, Chin. Opt. Lett. 12, 012401 (2014).
- A. Wiener, A. I. Fernández-Domínguez, A. P. Horsfield, J. B. Pendry, and S. A. J. Maier, Nano Lett. 12, 3308 (2012).
- P. West, S. Ishii, G. Naik, N. Emani, V. M. Shalaev, and A. J. L. Boltasseva, Laser Photon. Rev. 4, 795 (2010).
- N. K. Emani, A. V. Kildishev, V. M. Shalaev, and A. Boltasseva, Nanophotonics 4, 214 (2015).

- K. S. Novoselov, V. I. Fal'Ko, L. Colombo, P. R. Gellert, M. G. Schwab, and K. J. Kim, Nature 490, 192 (2012).
- Z. Wu, L. Jaekwang, N. Jagjit, S. T. Pantelides, S. J. Pennycook, and I. J. Juan-Carlos, Nat. Nanotechnol. 7, 161 (2012).
- J. Niu, Y. J. Shin, Y. Lee, J. H. Ahn, and H. J. Yang, Appl. Phys. Lett. 100, 183 (2012).
- H. Seyed, N. Mohammad, F.-D. Reza, L. Max, H. B. Peter, C.-A. Albert, A. Eduard, and S. Abadal, Nanomaterials 8, 577 (2018).
- H. Yenny, N. Valeria, L. Mustafa, F. M. Blighe, S. Zhenyu, D. Sukanta, I. T. Mcgovern, H. Brendan, B. Michele, and Y. K. Gun'Ko, Nat. Nanotechnol. 3, 563 (2008).
- 16. L. A. Falkovsky and A. A. Varlamov, Eur. Phys. J. B 56, 281 (2007).
- 17. L. A. Falkovsky, J. Phys.: Conf. Series 115, 012004 (2008).
- K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, J. Sci. **306**, 666 (2004).

- C. Johan, M. Alejandro, T. Sukosin, F. H. L. Koppens, and F. Javier Garcia de Abajo, ACS Nano 6, 431 (2012).
- 20. L. Tony and A. Phaedon, ACS Nano
 ${\bf 8},$ 1086 (2014).
- C. Lin, Z. Tian, H. Wei, X. Zhou, and L. Xun, J. Lightwave Technol. 32, 4199 (2014).
- C. Casiraghi, A. Hartschuh, E. Lidorikis, H. Qian, H. Harutyunyan, T. Gokus, K. S. Novoselov, and A. C. Ferrari, Nano Lett. 7, 2711 (2007).
- 23. C. H. Gan, Appl. Phys. Lett. 101, 071109 (2012).
- N. K. Emani, T. F. Chung, X. Ni, A. V. Kildishev, Y. P. Chen, and A. Boltasseva, Nano Lett. **12**, 5202 (2012).
- P. B. Johnson and R.-W. Christy, Phys. Rev. B 6, 4370 (1972).
- 26. D. Sun, M. Wang, Y. Huang, Y. Zhou, M. Qi, M. Jiang, and Z. Ren, Chin. Opt. Lett. 15, 051603 (2017).