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Xia, Yuyu; Wang, Jie; Zhang, Yanbin; Shan, Yuwei; Dai, Yunyun; Chen, Ang; Shen, Tangyao; Wu, Shiwei; Liu, Xiaohan; Shi, Lei; Zi, Jian

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Published in:

ADVANCED OPTICAL MATERIALS

DOI:

[10.1002/adom.202000264](https://doi.org/10.1002/adom.202000264)

Published: 01/09/2020

Document Version

Peer reviewed version

Please cite the original version:

Xia, Y., Wang, J., Zhang, Y., Shan, Y., Dai, Y., Chen, A., Shen, T., Wu, S., Liu, X., Shi, L., & Zi, J. (2020). Transmission-Type Optical Modulator Based on Graphene Plasmonic Resonator Integrated with Off-Resonant Au Structure. *ADVANCED OPTICAL MATERIALS*, 8(18), [2000264]. <https://doi.org/10.1002/adom.202000264>

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Transmission-type optical modulator based on graphene plasmonic resonator integrated with off-resonant Au structure

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In this paper, a transmission-type graphene plasmonic modulator by introducing off-resonant Au structure is theoretically and experimentally investigated. It is found that the modulation efficiency and bandwidth could be dramatically enhanced compared with bare graphene plasmonic structure. The validity of this proposed method is verified both in the one- and two-dimensional graphene plasmonic structures: ribbons and holes. This work would open up a new path to design the high-efficiency modulators, switches and multispectral detectors.

I. INTRODUCTION

The compact, efficient, fast and broadband modulators for on-chip optical interconnects are becoming indispensable [1–5]. Graphene as a promising candidate has been intensively researched due to its unusual electro-optical properties [6–17]. To date, various methods have been adopted to design the modulator with high efficiency, including large chemical doping [18–20], coupling with an additional resonator [21–24] and integrating with Salisbury screens (consisting of a single reflective mirror) [25–30]. However, those methods also have some limitations. Firstly, using large chemical doping would degrade the tunable capability and durability of the graphene-based modulator [23]. Secondly, the method of coupling with an additional resonator would make the modulator working in a narrow bandwidth. If changing the operating frequency, the structure needs to be redesigned carefully. Lastly, the use of Salisbury screens would be only available in reflection-type modulators. The transmission-type modulators are rarely reported, which also have their valuable applications in optoelectronic devices such as spatial light modulator and signal processing [31–35].

In this paper, a transmission-type graphene plasmonic modulator integrated with off-resonant Au structure is theoretically and experimentally investigated. By integrating with Au structure, the modulation efficiency η could be dramatically enhanced and modulation bandwidth could also be broadened. Based on the coupled-modes theory (CMT) [36–38], η is determined by the ratio γ_r/γ_a between the radiation rate γ_r and the absorption rate γ_a . It is found that the larger γ_r/γ_a , the higher η of graphene modulator. For graphene plasmonic ribbons (GPRs) integrated with Au structure (GPRs-Au), γ_r/γ_a and η of this composite structure could be dramatically increased. The finite-difference time-domain (FDTD) simulation results show 45.4% maximum modulation

efficiency $\eta^{\max}$. And the modulation bandwidth is as wide as 1250 cm^{-1} , in which the modulation efficiency η is larger than that of the bare GPRs structure. Experimental results show 24% $\eta^{\max}$ and 800 cm^{-1} modulation bandwidth of GPRs-Au structure. In addition to one-dimensional graphene ribbons, two-dimensional graphene plasmonic holes (GPHs) integrated with off-resonant Au disks (GPHs-Au) are also designed and fabricated to realize polarization independent modulator. This work may pave the way for designing transmission-type graphene plasmonic modulator with high modulation efficiency and broad bandwidth.

II. DESIGN PRINCIPLE FOR HIGH-EFFICIENCY MODULATOR

Modulation efficiency η is one of important parameters of a modulator. For transmission-type electro-optical modulator based on graphene plasmons, η depends on the transmittance variation range tuned by electrostatic field. It is defined by $\eta = (T_{\max} - T)/T_{\max} = 1 - T/T_{\max}$, where T corresponds to the gate voltage-dependent transmittance and $T_{\max}$ is the maximum transmittance [23]. Due to the atomic thickness of graphene, $T_{\max}$ could be tuned to be about 100%. Thus, η can be simply expressed as $\eta = 1 - T$, which is equivalent to the extinction. Here T and η can be calculated by the CMT [36–41], which is phenomenological method to describe the interaction between a resonator and free-space light. In the CMT, the dynamic equation for a single resonator coupled with two ports (see Fig. 1(a)) can be written as

$$\frac{da}{dt} = (-i\omega_0 - \gamma_r - \gamma_a)a + kS_{1+}, \quad (1)$$

where ω_0 is the resonant frequency of the resonant mode, γ_r is the radiation coupling rate of the resonant mode and γ_a is the intrinsic absorption rate, respectively. S_{1+} denotes the incident light, k is the coupling coefficient. The CMT model is illustrated in Fig. 1(a). Due to energy

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FIG. 1. **Schematic and modulation efficiency η of an optical resonator.** (a) Schematic of an optical resonator coupled with free-space light. The arrows indicate the incoming and outgoing waves. Suppose that the light is incident from port 1, $S_{2+} = 0$. S_{1+} , S_{1-} and S_{2-} denote the incident, reflected and transmitted light, respectively. (b) η for an optical resonator as a function of $(\omega - \omega_0)/\gamma_a$ and γ_r/γ_a . The arrow shows that η increases most rapidly with γ_r/γ_a from zero at the resonant frequency ω_0 .

conservation and reciprocal theorem [40, 41], k and γ_r are related as $k = i\sqrt{\gamma_r}$. The transmittance is defined as

$$T = \left| \frac{S_{2-}}{S_{1+}} \right|^2. \quad (2)$$

where $S_{2-} = S_{1+} + ka$ is the transmitted light. Then the transmittance can be obtained as

$$\begin{aligned} T &= \frac{(\omega - \omega_0)^2 + \gamma_a^2}{(\omega - \omega_0)^2 + (\gamma_r + \gamma_a)^2} \\ &= \frac{\left(\frac{\omega - \omega_0}{\gamma_a}\right)^2 + 1}{\left(\frac{\omega - \omega_0}{\gamma_a}\right)^2 + (\gamma_r/\gamma_a + 1)^2}. \end{aligned} \quad (3)$$

Thus, η is

$$\eta = 1 - T = \frac{-1 + (\gamma_r/\gamma_a + 1)^2}{\left(\frac{\omega - \omega_0}{\gamma_a}\right)^2 + (\gamma_r/\gamma_a + 1)^2}. \quad (4)$$

From Eq. (4), it can be seen that η is determined by three parameters ω_0 , γ_r and γ_a . It is clear that η reaches its maximum at resonant frequency ω_0 ,

$$\eta_0 = 1 - \frac{1}{(\gamma_r/\gamma_a + 1)^2}. \quad (5)$$

When $\gamma_r \ll \gamma_a$, η_0 reaches its minimum, $\eta_0 = 0$. When $\gamma_r = \gamma_a$ namely critical coupling [40, 41], η_0 is equal to 75%. When $\gamma_r \gg \gamma_a$, η_0 is close to 100%.

As shown in Fig. 1(b), η as a function of γ_r/γ_a and $(\omega - \omega_0)/\gamma_a$ is shown on a contour. Obviously, η increases rapidly with γ_r/γ_a at the resonant frequency ω_0 [shown by the arrow in Fig. 1(b)]. This reveals that the design principle for high-efficiency modulator is to tune γ_r/γ_a of any optical resonator from zero to a value as large as possible. One way is to increase γ_r . Introducing off-resonant scatterer such as dielectric or metal structure could be applied to increase γ_r . The off-resonant scatterer assists graphene plasmons to be scattered to free

space. The stronger scattering ability the scatterer has, the easier the graphene plasmonic mode radiates to the free space and the larger γ_r will be. The scattering ability depends on scatterer material parameters, such as refractive index and geometry size. The details on how scatterer affects the modulation performance are shown in the supporting information. Considering the feasibility of the experiment, in this work the off-resonant Au structure are used as the scatterer to increase γ_r .

III. SIMULATION RESULTS

To investigate η_0 of bare GPRs structure, the transmittance spectra at normal incidence with different Fermi levels E_F [Fig. 2(a)] and ribbon widths [Fig. 2(b)] are obtained by FDTD simulations. In the simulations, the polarization of incident light is along the periodic direction of graphene ribbons. The graphene thickness and its optical constants are adopted from Ref. [42]. As shown in Fig. 2(a), with increasing graphene E_F from 0.2 to 0.8 eV for 90 nm ribbon width, the resonant frequency of graphene plasmonic mode displays a blue shift and the transmittance at resonant frequency T_0 decreases from 100% to 83%. Even under the high doping condition, $\eta_0 = 1 - T_0$ is only as less as 17%. As shown in Fig. 2(b), with increasing ribbon widths from 55 to 88 nm for 0.35 eV graphene, the transmittance still changes slightly. This means that through changing E_F and widths, it is difficult to tune η_0 of bare GPRs in a large range.

To explore the physical mechanism of low η_0 of bare GPRs structure with different E_F and widths, the corresponding decay rates γ_r and γ_a are obtained by fitting each transmittance spectrum in Fig. 2(a) and Fig. 2(b)

FIG. 2. **FDTD simulations of bare GPRs structure with different E_F and ribbon widths.** (a) Transmittance spectra of bare GPRs with different graphene E_F for 90 nm ribbon width. (b) Transmittance spectra of bare GPRs with different ribbon widths for 0.35 eV graphene E_F . The periodicity is 200 nm. (c) η_0 contour as a function of radiation rate γ_r and absorption rate γ_a . Circles and dots represent the decay rates fitted by the CMT for different widths and E_F , respectively. Inset exhibits η contour where black dots show the increase of γ_r/γ_a with E_F for 90 nm ribbon width.

FIG. 3. **FDTD simulations for bare GPRs and GPRs-Au structures.** (a) Transmittance spectra of bare Au, bare GPRs and GPRs-Au structures. Inset shows the schematic diagram of GPRs-Au structure. (b) Total electric field distributions at resonance without (upper) and with (lower) Au structure. (c) Transmittance spectra of bare GPRs (dotted curves) and GPRs-Au (solid curves) structures with different E_F . (d) η_0 contour as a function of γ_r and γ_a and the decay rates of both structures fitted from the spectra in (c) by the CMT. (e) Comparisons of γ_r/γ_a and η_0 of the two structures. Inset shows η contour and the change of γ_r/γ_a at resonant frequency as increasing E_F of both structures. (f) Comparison of the frequency-dependent η_f^{max} of both structures.

via the CMT. Both decay rates of different graphene widths and E_F are shown in Fig. 2(c). The purple circles show the decay rates with different ribbon widths for 0.35 eV graphene E_F . The red, black and blue dots show the decay rates of 70, 90 and 110 nm ribbon widths with different graphene E_F , respectively. η_0 as a function of γ_r and γ_a is also shown on a contour. Obviously, no matter what graphene widths and E_F are, γ_r is always one order less than γ_a leading to low η_0 of bare GPRs. The inset in Fig. 2(c) exhibits η contour where black dots show the change of γ_r/γ_a at resonant frequency with increasing E_F for 90 nm ribbon width. The small variation

range of γ_r/γ_a around 0 at resonant frequency results in small η_0 . Therefore, to design a high-efficiency modulator, it requires an effective way to increase γ_r/γ_a in a wide range.

Here, a simple and efficient method is proposed to increase γ_r/γ_a and design a high-efficiency graphene plasmonic modulator by integrating with off-resonant Au structure. The designed Au structure is almost transparent at the operating frequency, which will not interfere graphene plasmonic mode. This provides a possibility for realizing a broadband modulator.

To design a high η transmission-type modulator based

FIG. 4. **Experimental results for bare GPRs and GPRs-Au structures.** (a) Schematic diagram of GPRs-Au modulator. (b) Transmittance spectra of bare Au, bare GPRs and GPRs-Au structures with high doping level of graphene by Br_2 vapor. The widths of Au ribbon and graphene ribbon are 110 and 70 nm, respectively. The periodicity of the measured structures is 200 nm. Gate voltage-dependent transmittance spectra of (c) bare GPRs structure and (d) GPRs-Au structure. The corresponding minimum E_F is close to zero at 0.8 V voltage and the maximum E_F is about 0.5 eV at -2 V voltage. (c) The width of graphene ribbon and the periodicity of GPRs structure are 90 and 200 nm, respectively. (d) The width of graphene ribbon and the periodicity of GPRs-Au structure are 90 and 180 nm, respectively. (e) Comparison of η_0 as a function of gate voltage of both structures. (f) Comparison of the frequency-dependent $\eta_f^{\max}$ of both structures.

on one-dimensional GPRs, the off-resonant Au ribbons is introduced. The transmittance performance of this composite structure is studied by FDTD simulations. The schematic diagram is shown in the inset of Fig. 3(a). The width and periodicity of GPRs structure are set to be 90 and 200 nm, respectively. The parameters of Au ribbons are set as: the thickness $h = 30$ nm, the width $d = 100$ nm. Figure 3(a) shows the transmittance spectra of bare off-resonant Au ribbons, bare GPRs and GPRs-Au structures. The blue curve shows that the transmittance of bare Au structure is nearly 100% without introducing additional plasmonic modes.

The black curve shows that the transmittance at resonance frequency (T_0) of bare GPRs is about 92%. Thus, $\eta_0 = 1 - T_0$ of bare GPRs is only 8%. As a contrast, the red curve shows that, besides to a redshift of the resonant frequency (may caused by the changes of environmental refractive index, the details are shown in the supporting information), η_0 of the composite structure (GPRs-Au) dramatically increases to 24%, 3 times as large as that of bare GPRs structure. All these results imply that the off-resonant Au structure mainly enhances the coupling strength between graphene plasmonic mode and free-space light. The total electric field distributions at

resonance without (upper) and with (lower) Au structure are shown in Fig. 3(b). It shows that the near-field intensity around GPRs is extremely enhanced by Au structure which leads to larger η_0 of graphene plasmonic structure.

To further evaluate η_0 of this composite structure (GPRs-Au) with different E_F , the transmittance spectra are compared with that of bare GPRs structure as shown in Fig. 3(c). When changing E_F from 0.2 to 0.8 eV, the transmittance and η_0 of both structures could be tuned. The solid and dotted curves show the transmittance spectra of GPRs-Au structure and bare GPRs structure, respectively. T_0 decreases continuously as increasing E_F . Therefore, η_0 increases continuously. The minimum transmittance of GPRs-Au structure is 54.6% at 0.8 eV, thus $\eta^{\max}$ reaches 45.4%. In contrast, $\eta^{\max}$ of bare GPRs structure is only 17%. The enhancement effects of this off-resonant Au structure are analyzed through the CMT. Figure 3(d) shows the fitted γ_r and γ_a of GPRs-Au structure (red dots) and bare GPRs structure (black dots) at each E_F . It can be observed that by increasing E_F of graphene, although the decay rates of both structures have similar dynamic behavior, i.e. γ_a decreases and γ_r increases, both γ_r and its variation range of GPRs-Au structure are much larger than that of bare GPRs structure. The enhancement of γ_r directly enlarges γ_r/γ_a leading to the increase of η_0 . η_0 as a function of the two rates is also shown on a contour in Fig. 3(d). With increasing E_F , η_0 of GPRs-Au structure spans half contour map which is nearly 50% variation range and is much wider than that of bare GPRs structure. The comparisons of γ_r/γ_a and η_0 of the two structures are shown in Fig. 3(e). It is obvious that γ_r/γ_a and η_0 of GPRs-Au structure (red curves) are much larger than that of bare GPRs structure (black curves). It should be noted that η_0 of GPRs-Au structure can be further increased by increasing E_F . With increasing E_F , the rising trend of η_0 is similar to that of γ_r/γ_a which is nearly linear. The inset in Fig. 3(e) exhibits η contour where red and black dots represent the change of γ_r/γ_a at resonant frequency as increasing E_F of both structures. It indicates that the rising rate of γ_r/γ_a of GPRs-Au structure (red dots) is much faster than that of bare GPRs structure (black dots). The large γ_r/γ_a of GPRs-Au structure results in large η_0 .

Moreover, the proposed structure could also broaden the modulation bandwidth. As shown in Fig. 3(c), due to the blueshift of graphene plasmonic resonance with increasing E_F , for each frequency, there is a minimum transmittance and maximum modulation efficiency $\eta_f^{\max}$ of both structures. The frequency-dependent $\eta_f^{\max}$ of both structures are compared in Fig. 3(f). As shown in Fig. 3(f), $\eta_f^{\max}$ of GPRs-Au structure is larger than that of bare GPRs structure in a wide range from 500 to 1750 cm^{-1} . The corresponding modulation bandwidth is 1250 cm^{-1} . Therefore, the designed GPRs-Au structure would be valuable for applications of broadband modulators with high modulation efficiency.

IV. EXPERIMENTAL RESULTS

To show experimentally, the GPRs-Au structure is fabricated on the BaF_2 substrate. The experimental details are given in the method section. It should be noted that the fabricated sample is exposed to Br_2 vapor for 15 seconds to increase graphene E_F . The SEM image of GPRs-Au structure is shown in the inset of Fig. 4(b). Figure 4(b) shows the transmittance spectra of bare off-resonant Au ribbons, bare GPRs and GPRs-Au structures. In experiments, the transmittance spectrum of flat graphene T_b is used as a reference and the polarization direction of incident light is the same as that in simulations. The blue curve shows that the transmittance of bare Au ribbons is about 100%. The black curve shows that T_0 of bare GPRs is 96%. The corresponding η_0 is only 4%. And the red curve shows that η_0 of GPRs-Au structure reaches 30%, which is similar to that in simulation.

To further evaluate gate voltage-dependent η of this designed structure, ionic gel and electrodes are applied to realize the transmission-type modulator [43]. The schematic diagram of the device is shown in Fig. 4(a). The tuning range of graphene E_F is obtained from the threshold transmittance spectra in near infrared range caused by interband transitions [44]. The minimum E_F is close to zero at 0.8 V voltage and the maximum E_F is about 0.5 eV at -2 V voltage. The transmittance spectra as a function of gate voltage for bare GPRs and GPRs-Au structures are compared in Fig. 4(c) and Fig. 4(d). The spectra are measured in situ. And the background spectrum T_{Dirac} is measured at 0.8 V voltage as a reference, where graphene E_F is zero without plasmonic mode. Thus, T/T_{Dirac} in the whole measured frequency range should reach its maximum (100%) at 0.8 V voltage. By changing gate voltage from 0.8 to -2 V, the resonant frequency for both structures has blue shift and T_0 decreases continuously as shown in Fig. 4(c) and Fig. 4(d). For bare GPRs structure, the electrically controlled transmittance still maintains a high value ($> 93.5\%$) with small η ($< 6.5\%$). For GPRs-Au structure, T_0 decreases from 100% to 76% continuously. Thus η is tuned from 0 to 24%. η_0 of GPRs-Au structure (red curve) and bare GPRs structure (black curve) are compared in Fig. 4(e). Obviously, η_0 and its rising rate of GPRs-Au structure are much larger than that of bare GPRs structure. It verifies that the designed structure works well for high-efficiency modulation. To investigate the modulation bandwidth of the proposed structure, $\eta_f^{\max}$ of both structures are compared in Fig. 4(f). Here, frequency-dependent $\eta_f^{\max}$ for both structures can be simply expressed by η at -2 V voltage. $\eta_f^{\max}$ of GPRs-Au structure is larger than that of bare GPRs structure in a wide range from 650 to 1450 cm^{-1} . Thus, the modulation bandwidth in experiment reaches 800 cm^{-1} . Similarly, the modulation bandwidth could be broadened through further increasing graphene E_F . Besides, the several transmittance dips around 1200 cm^{-1} are due to the intrinsic absorp-

FIG. 5. **Experimental results for bare GPHs and GPHs-Au structures.** (a) Transmittance spectra of bare GPHs and GPHs-Au structures with high doping level of graphene by Br_2 vapor. Inset shows SEM image of GPHs-Au structure. (b) Gate voltage-dependent transmittance spectra of GPHs-Au structure. Inset shows η at 822 cm^{-1} as a function of gate voltage. The diameter of graphene hole is 120 nm. The periodicity of bare GPHs and GPHs-Au structures is 200 nm.

tion of ionic gel. Interestingly, the absorption of ionic gel is enhanced due to the coupling with graphene plasmon mode. The red curve shows that ionic gel can further broaden the bandwidth of modulator and increase the modulation efficiency.

Additionally, this design method could also be extended to the two-dimensional case. Here, the two-dimensional graphene plasmonic holes with Au disks (GPHs-Au) is investigated. The experimental steps are same as the one-dimensional device. The inset in Fig. 5(a) shows the SEM image of GPHs-Au structure. As shown in Fig. 5(a), the optimized structure also exhibits much lower T_0 than bare graphene plasmonic holes (GPHs) with high doping level by Br_2 vapor. Thus, η_0 of GPHs-Au structure is much larger than that of bare GPHs. η_0 of GPHs-Au structure is 14% while η_0 of bare GPHs is only 1.3%. η_0 of both structures are smaller than that of one-dimensional structures [see Fig. 4(b)] due to less effective graphene plasmons. Figure 5(b) shows electrical tuning performance of GPHs-Au structure. T_0 decreases continuously from 100% to 93%. The gate voltage-dependent η at 822 cm^{-1} is shown in the inset figure which increases from 0 to 7%. Compared with Fig. 5(a), it can be predicted that η of GPHs-Au structure could be further increased for higher graphene E_F . It should be noted that this composite two-dimensional structure could be applied to realize an isotropic optical modulator.

V. CONCLUSION

In summary, the transmission-type optical modulators with high η and broad bandwidth based on graphene plasmonic structures are designed under the guidance of the CMT. Both simulation and experimental results show that by integrating with off-resonant Au structure, η could be dramatically enhanced and the modulation bandwidth could be broadened as well. Specifically, FDTD simulation results show that GPRs-Au structure has 45.4% maximum modulation efficiency $\eta^{\max}$ when

maximum E_F of graphene reaches to 0.8 eV. And this proposed structure has nearly 1250 cm^{-1} modulation bandwidth to realize larger η than bare GPRs structure. Experimental results show 24% modulation efficiency of GPRs-Au structure which is about 4 times as large as that of bare GPRs structure. The modulation bandwidth is 800 cm^{-1} . Moreover, this design method could also be extended to the two-dimensional case such as graphene holes integrated with Au disks which could be applied to isotropic optical modulators. Therefore, this work may open up a new path to design the advanced electro-optical modulators, multispectral detectors and signal processing.

METHODS

A. Sample fabrications

A large-area CVD grown graphene sheet was first transferred onto a BaF_2 substrate. Graphene plasmonic structures were fabricated with $100 \times 100 \mu\text{m}^2$ area obtained by standard e-beam lithography using a PMMA resist. After development, the exposed graphene is etched by oxygen plasma etching. Compared with bare GPRs and bare GPHs structures, there is one more step for GPRs-Au and GPHs-Au structures before removing PMMA. The metallic layer was deposited by electron beam evaporation of Cr (5 nm) followed by Au (30 nm) with the patterned PMMA serving as a soft etch mask. Fabricated samples were then exposed to Br_2 vapor for 15 seconds to increase the doping level of graphene. The electrically controlled modulator was achieved by introducing ionic gel. The ionic gel solution was prepared according to the method in the Ref. [45]. And then it was diluted by 4 times with dichloromethane.

B. Spectral measurements

Experimental transmittance spectra of the structures were performed at room temperature by a Bruker Hyperion 2000 infrared microscope and a Fourier transform infrared (FTIR) spectrometer (Bruker Vertex 70) with liquid nitrogen cooled mercury cadmium telluride and mid-IR source. Transmitted light was collected using Hyperion 2000 IR microscope with a $15\times$ magnification objective and a numerical aperture of 0.4. The area covered by infrared light was about $100 \times 100 \mu\text{m}^2$. The spectral resolution was 16 cm^{-1} . In each sample measurement without gate voltage, transmitted light through the flat graphene and BaF_2 substrate was recorded as a background spectrum. For the sample with an applied gate voltage, the background spectrum T_{Dirac} is measured at 0.8 V voltage as a reference, where graphene E_F is zero without plasmonic mode.

ACKNOWLEDGEMENTS

The work was supported by China National Key Basic Research Program (Grants No. 2016YFA0301103,

No. 2016YFA0302000, and No. 2018YFA0306201) and National Science Foundation of China (Grants No. 11774063, No. 11727811, No. 91750102, No. 11604355, and No. 91963212). The research of L. S. was further supported by the Science and Technology Commission of Shanghai Municipality (Grants No. 17ZR1442300 No. 17142200100, and No. 19XD1434600)

Y.Y. Xia and J. W. contributed equally to this work.

KEYWORDS

graphene plasmons, coupled-modes theory, transmission-type modulator, graphene-metal composite structure

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