Anisotropic Plasmonic Response of Black Phosphorus Nanostrips in Terahertz Metamaterials

Volume 10, Number 3, June 2018

Qingqing Fo
Ling Pan
Xieyu Chen
Quan Xu
Chunmei Ouyang
Xueqian Zhang
Zhen Tian
Jianqiang Gu
Liyuan Liu
Jiaguang Han
Weili Zhang

DOI: 10.1109/JPHOT.2018.2842059
1943-0655 © 2018 IEEE
Anisotropic Plasmonic Response of Black Phosphorus Nanostrips in Terahertz Metamaterials

Qingqing Fo, Ling Pan, Xiyue Chen, Quan Xu, Chunmei Ouyang, Xueqian Zhang, Zhen Tian, Jianqiang Gu, Liyuan Liu, Jiaguang Han, and Weili Zhang

1Center for Terahertz Waves and College of Precision Instrument and Optoelectronics Engineering, and the Key Laboratory of Optoelectronics Information and Technology, Ministry of Education, Tianjin University, Tianjin 300072, China
2Automation Department, Chongqing Industry Polytechnic College, Chongqing 401120, China
3School of Electrical and Computer Engineering, Oklahoma State University, Stillwater, OK 74078 USA

DOI:10.1109/JPHOT.2018.2842059
1943-0655 © 2018 IEEE. Translations and content mining are permitted for academic research only. Personal use is also permitted, but republication/redistribution requires IEEE permission. See http://www.ieee.org/publications_standards/publications/rights/index.html for more information.

Abstract: Two-dimensional black phosphorus (BP) recently emerged as an outstanding material for optoelectronics and nanophotonics applications. In contrast to graphene, BP has a sufficiently large electronic bandgap and its high carrier mobility allows for efficient free-carrier absorption in the infrared and terahertz regimes. Here, we present a reflective structure to enhance the response of nanostructured monolayer BP at terahertz frequencies and investigate localized surface plasmon resonances in BP nanostrip arrays. Anisotropic absorption is observed in the proposed BP metamaterials due to the puckered crystal structure of the monolayer BP, and further investigations show that the plasmonic resonances are strongly depending on the geometric parameters of the nanostrips and the coupling between the adjacent nanostrips. We expect that the monolayer BP is an outstanding candidate of highly anisotropic plasmonic material for ultrascaled optoelectronic integration.

Index Terms: Black phosphorus, surface plasmon, terahertz.

1. Introduction

Strong light-matter interaction makes atomically thin materials attractive in the areas of sensitive-sensing [1], [2], super-resolution [3], [4], optoelectronics [5], [6] and nanophotonics [7], [8]. In recent years, thin two-dimensional (2D) materials, such as graphene, transition metal dichalcogenides (TMDs) and black phosphorus (BP), with atomic-scale thicknesses, have been attracting extensive attention [9]–[11]. Extensive works were carried out on interactions of graphene with terahertz radiation at subwavelength scales [5], [12]–[14]. Graphene has a high carrier mobility and strong interaction with light when it is structure patterned. Although there exists immense potential for novel applications, challenges such as serious scattering loss, absence of band gap and low absorption efficiency in graphene remain unsolved [15], which limit its further applications under conditions...
where strong light-matter interactions are involved. In contrast, transition-metal dichalcogenides (like MoS$_2$, WS$_2$ and MoSe$_2$) have the band gap ranging from 1.0 to 2.0 eV and their electromagnetic properties depend on various factors such as thickness [16], temperature [17] and illumination intensity [18]. However, TMDs have low electronic mobility, which limit their applications in field-effect transistors and infrared or far infrared optoelectronics [19].

Currently, another emerging 2D material: black phosphorus, the most thermodynamically stable allotrope of phosphorus, has been noticed in the area of nanophotonics [11], [20]–[22]. In contrast to graphene, the monolayer BP has a direct band gap reaching 2.0 eV [23]–[26], thus showing many exotic properties featuring high-contrast inter-band excitation [27], linear dichroism [28] and tunable direct band gap [29]. The possibility of collective electronic motion with light to form surface polaritons has made BP a versatile platform for extreme light confinement [30], tailored nanophotonics [31], [32] and ultrafast switching [33]. Therefore, monolayer BP is considered as a natural candidate for broadband optical applications, including the infrared and terahertz branches of the spectrum. In particular, as a plasmonic platform, the atomically thin BP enables to confine plasmons in an extremely small domain, enhancing the light-mater interactions.

In this article, we propose the first detailed demonstration of localized surface plasmons in a 2D BP based metamaterial in the terahertz regime, showing that localized surface plasmons can be excited and efficiently manipulated in the nanostructured monolayer BP. To observe obvious surface plasmon resonances, in the simulation, we employed a reflective structure that consists of periodic monolayer BP strips, a transparent insulator and a full reflective metallic mirror. The metallic mirror is used to reflect the terahertz wave and suppress the transmission. Compared with free-standing BP nanostrips, the dielectric layer between the BP and the metallic mirror forms a Fabry-Perot cavity and therefore increases the interaction of the terahertz wave with monolayer BP strips. Dielectric thickness can be chosen to maximize the absorption using Fabry-Perot interference. Different electromagnetic responses were observed in the proposed BP metamaterial when patterning BP nanostrips in the Armchair and Zigzag directions, due to its puckered crystal structure. Anisotropic behaviours of the nanostructured BP will open a new functionality and perspective towards developing novel plasmonic devices.

2. Atomic Arrangement and Electrical Model of Monolayer BP

Similar to graphite, BP consists of layered sheets of atoms held together by the van der Waals force. The difference is that, the anisotropy of the electric and optical properties is a unique feature distinguishing monolayer BP from graphene and many other 2D materials. The fundamental cause of this feature is that the atoms in 2D BP are arranged to form a puckered hexagonal honeycomb structure with ridges due to sp$^3$ hybridization, which leads to strong in-plane anisotropic electrical and optical properties [34]–[36]. As shown in Fig. 1, the atomic structure arrangement is different between the named Armchair and Zigzag directions. Thus, we anticipate that different plasmonic responses will be observed when the nanostrps are along the Armchair and Zigzag direction.

Here, we employ a semiclassical Drude model to describe the photonic properties of the 2D BP layer. Permittivities $\varepsilon_1$ and $\varepsilon_2$ are denoted to be along the $x$ and $y$ directions, respectively. They can be expressed by the following formula:

$$\varepsilon_j = \varepsilon_r + \frac{i \sigma_j}{\omega \varepsilon_0} (j = 1, 2), \quad (1)$$

Where $\varepsilon_r$ is relative permittivity of the monolayer BP, $t$ is thickness of the monolayer BP (chosen to be 1 nm in the simulation), $\sigma_j$ is BP surface conductivity, $\varepsilon_0$ is free space permittivity and $\omega$ is frequency of the incident light. The relative permittivity $\varepsilon_r$ is taken as 5.76 for the monolayer BP [37]. From Eq. (1), we can see that the value of $\varepsilon_j$ is determined by the surface conductivity $\sigma_j$. In the Drude model, the conductivity $\sigma_j$ can be given as [23], [35]:

$$\sigma_j = \frac{i D_j}{\pi \left( \omega + \frac{\omega^2}{\rho_j} \right)} (j = 1, 2), \quad (2)$$
The parameter $\hbar$ is the reduced Planck constant. $\eta$ describes the relaxation rate and is chosen to be 10 meV [35]. $D_j$ is the Drude weight and can be described by:

$$D_j = \frac{\pi e^2 n}{m_j} (j = 1, 2)$$  \hspace{1cm} (3)

Where $e$ is the electron charge, the electron doping $n = 10^{13} \text{cm}^{-2}$ is taken in the simulation [36].

The parameters $m_1$ and $m_2$ denote the electron mass of 2D BP along the Armchair and Zigzag directions, respectively, which can be given as:

$$m_1 = \frac{\hbar^2}{2\gamma + \eta_c}, \quad m_2 = \frac{\hbar}{2v_c}. \hspace{1cm} (4)$$

For the monolayer BP [34], we have:

$$\gamma = \frac{4t}{\pi} \text{eVm}, \quad \eta_c = \frac{\hbar^2}{0.4m_0}, \quad v_c = \frac{\hbar}{1.4m_0} \text{ and a band gap } \Delta = 2 \text{ eV}.$$

3. Simulated Results and Analysis

In order to observe electric-field localization and light confinement resulted from the BP nanostrips easily, we employed a reflective structure to enhance the light-matter interaction. A schematic diagram of the proposed BP metamaterial is shown in Fig. 2(a). The structure consists of periodic monolayer BP nanostrips, a transparent insulator and a reflective metallic mirror. The reflective mirror suppresses the transmission of the incident light, forming a Fabry-Perot cavity together with the insulator and the BP layer, and therefore increases the interaction of light with the monolayer BP.

The schematic diagram of the BP nanostrip arrays and the unit cell and its geometrical parameters are shown in Fig. 2(a). In the simulations, we chose 13.5-μm-thick quartz as the transparent insulator and 3-μm-thick aluminum as the metallic mirror.

In the CST Microwave Studio simulations, the BP film thickness $t$ is chosen to be 1 nm with a mesh of 0.25 nm. This assumption helps reducing the simulation time, meanwhile, ensuring the precision. We performed full-field electromagnetic simulations with frequencies between 0–18 THz. The plane terahertz wave with polarization along the BP nanostrip is incident from the top surface of the structure (see Fig. 2(a)). Absorptivity ($A$) can be calculated with a formula $A = 1-R-T$. Since the metallic mirror prevents the transmission of the terahertz wave, thus transmission ($T$) can be regarded as zero. Then we can calculate the absorptivity using $A = 1-R$. 
Firstly, we performed electromagnetic simulations for the full-plane monolayer BP and BP nanostrips (along Armchair direction), respectively. As shown in Fig. 2(a), the patterned BP nanostrip is along the x-direction. The absorption spectra are plotted in Fig. 2(b). When a continuous monolayer BP film (with Armchair direction along the x-axis) is applied on the dielectric layer, due to the Fabry-Perot effect, multiple resonance absorption peaks at 2.89, 8.62 and 14.42 THz are observed in the concerned frequency range. These frequencies are well consistent with theoretical calculations based on the Fabry-Perot effect with a formula $\lambda = 4 nh/(2m+1)$, where $\lambda$ is the resonance wavelength, $m$ is a positive integer, $n = 1.94$ is the refractive index of quartz, and $h = 13.5 \, \mu m$ is the thickness of quartz. Then, we plot the electromagnetic response curve of the BP nanostrips (along the Armchair direction) in Fig. 2(b). Differing from the response of the full-plane BP, due to the localized plasmon resonances of the BP nanostrips, the strongest absorption peak is located at 8.78 THz and the absorption intensity is about 0.46, stemming from the combination effect of the Fabry-Perot effect and the resonances of the BP nanostrips. For the first absorption peak, the frequency position is nearly not changed while the intensity is much smaller compared with the full-plane BP case because of the domination by the plasmon resonances of the BP nanostrips. Meanwhile, the third absorption peak is caused mainly by the Fabry-Perot effect and therefore, there is no significant change in the frequency position and absorption intensity.

Furthermore, the side view electric field profile of the vertical component $E_z$ with the case of the BP nanostrip arrays along the Armchair direction at resonance frequency of 8.78 THz is displayed in Fig. 2(c). It is seen that a typical localized plasmon resonance has been excited by the incident terahertz wave, indicated by the positive and negative dipoles, in which the electric field is mostly concentrated around the ends of the BP nanostrip.

Due to different permittivities along the Armchair and Zigzag directions of 2D BP originating from its instinct anisotropic structure, the optical responses of the BP nanostrips in the Armchair direction...
and Zigzag directions are different. To verify this, we performed electromagnetic simulations for the monolayer nanostrips in the Armchair and Zigzag directions, respectively. The structural parameters are the same for the two cases. The absorption spectra are shown in Fig. 3. The result shows that the major absorption peak position is located at 3.35 THz in the Zigzag direction (blue line), while that is 8.78 THz in the Armchair direction (red line). It suggests that, compared with the Zigzag direction, the major absorption peak frequency in the Armchair direction is higher and its absorption rate reaches 0.46, while it is only 0.10 in the Zigzag direction. This is because the smaller mass along the Armchair direction indicates higher resonance frequency. Compared with the Zigzag direction, the absorption peak in Armchair direction is higher and the resonance line width is slightly narrower. This result suggests that BP has more optical loss in the Zigzag direction at corresponding resonance frequency. Thus, the localized electric field and plasmonic resonance of the BP nanostrips are much stronger in the Armchair direction.

In order to investigate the influence of the structural parameters of the proposed BP metamaterial unit cell on its plasmon behaviour, we further performed simulations for different lengths of the BP nanostrip as well as different periodicities. While ensuring that the other parameters remain unchanged, the length of the BP nanostrip (l) is changed from 120 to 160 nm. The absorption spectra with the BP nanostrip in the Armchair and Zigzag directions are plotted in Fig. 4(a) and (b), respectively. It is seen that the major absorption peak position exhibits a red shift when increasing
Fig. 5. (a), (b) Absorption spectra for various transverse periods of the two directions. The insets are the major absorption peaks on an expanded scale over 7–11 THz and 1–5 THz, respectively. (c), (d) Absorption spectra for various lengths of the BP nanostrip of the two directions, for a fixed ratio $l/P_x = 2/5$. The dotted lines indicate the Fabry-Perot resonance frequency positions.

$l$ in the Armchair direction, which follows the regular rules of resonance of the strip structures. However, the change of the corresponding absorption intensity is not monotonic, which increases firstly and then decreases as resonance frequency moves towards to lower frequencies. As the BP nanostrip is lengthened to 140 nm, its resonance frequency moves to 8.62 THz, which approaches the Fabry-Perot resonance frequency given by $\lambda = 4nh/(2m+1)$ for $m = 2$. Therefore, the absorption rate reaches maximum. To further increase $l$, the mismatch leads to attenuation of the resonance. In contrast, for the Zigzag direction, since the major absorption peak position is far from the Fabry-Perot resonance frequency for $m = 1$, the absorption peak intensity increases consistently as the resonance frequency slightly moves towards to lower frequencies (see Fig. 4(b)). Additionally, there is no significant change observed in the other two absorption peaks for the two directions with increasing $l$. This is because these two absorption peaks originate from the Fabry-Perot effect, not influenced by the structural parameters of the BP nanostrips.

Another anisotropic behaviour has been observed when varying the transverse period $P_x$, as shown in Fig. 5(a) and (b). The other parameters remain the same with that in Fig. 2, we only changed the transverse period $P_x$ ranging from 160 to 300 nm. For the Armchair and Zigzag-direction BP nanostrips, their absorption spectra are shown in Fig. 5(a) and (b), respectively. If the transverse period is small enough (the ratio $l/P_x > 1/2$), there is a longitudinal capacitive coupling between the neighboring BP nanostrips along x-axis, which cannot be ignored, especially for the Armchair direction. When we gradually increase the transverse period $P_x$ to 260 nm, it is seen that differing from changing $l$, the major absorption peak position shows a slight blue shift attributing to the weakened coupling. However, when we continue to increase $P_x$ to the ratio $l/P_x < 1/2$, the coupling between the neighboring nanostrips is negligible and therefore, the absorption peak
Fig. 6. (a), (b) Absorption spectra for various longitudinal periods of the two directions. Absorption spectra for various numbers of nanostrips per unit area along (c) the Armchair direction and (d) Zigzag direction.

position holds steady, which follows the expected results of the strip structure resonance. In addition, the absorption intensity decreases due to the smaller duty cycle of BP with increasing $P_x$. The similar phenomenon was observed in graphene ribbons [34]. Furthermore, as shown in Fig. 5(a) and (b), the change of $P_x$ did not influence the other two absorption peaks for the two directions, due to unchanged thickness of the dielectric layer (unchanged Fabry-Perot resonance). Since the above results are attributed to the combined effects of the Fabry-Perot effect, the resonances of BP nanostrips and BP duty cycle, here we consider the transverse period ($P_x$) and the length ($l$) change at a fixed ratio $l/P_x = 2/5$ (the influence of capacitive coupling and duty cycle could be negligible), the major absorption spectra are shown in Fig. 5(c) and (d). It is seen from the absorption spectra that the major absorption peak position shows a red shift with the increase of $P_x$ and $l$ (with $l$ ranging
from 140 to 240 nm). The absorption peak intensity reaches maximum when $l = 180$ nm, matching the Fabry-Perot resonance with the localized surface plasmon resonance of the BP nanostrips in the Armchair direction. Increasing $P_x$ and $l$ makes the plasmon resonance of the BP nanostrips in the Zigzag direction close to the Fabry-Perot resonance and therefore, the major absorption peak intensity increases gradually with increasing $P_x$ and $l$.

Furthermore, we investigated the transverse capacitive coupling between the neighboring BP nanostrips along the y-axis in two ways. Firstly, we fixed the other values and only changed the longitudinal period $P_y$ ranging from 40 to 80 nm. The simulated absorption spectra along the Armchair and Zigzag directions are plotted in Fig. 6(a) and (b), respectively. We can see that the major absorption peak intensity reduces significantly with increasing $P_y$ for the two cases, stemming from the decrease of the duty cycle of BP in the unit cell. In addition, the absorption peak frequency exhibits a red shift in the two directions. This is because the transverse coupling is weakened with the increase of $P_y$. A similar phenomenon was observed when we fixed the longitudinal period $P_y$ to 180 nm and increased the number of the nanostrips from 1 to 4, as shown in Fig. 6(c) and (d). The distance $d$ between the two adjacent nanostrips remains 20 nm. For both the Armchair and Zigzag directions, the major absorption peak values are enhanced as the number of the BP nanostrips increases, companied by a blue shift of the resonance frequency which is influenced by the transverse capacitive coupling between the BP nanostrip dipole resonances.

For the reflective setup designed in our simulations, the thickness $h$ of the quartz dielectric layer will affect the results inevitably. Therefore, we analyzed the variation of the absorption response with various quartz thicknesses as well. Fig. 7 shows the absorption maps of the two directions with the thickness $h$ changing from 1 to 40 μm. The transverse and longitudinal periods remain 190 and 40 nm, respectively. The major resonance peak positions are about 9.1 and 4.0 THz in the two directions, respectively, and the resonance positions are unchanged with varying thickness $h$. In addition, we observed that the absorption reached maximum at certain thickness. This phenomenon can be explained with the Fabry-Perot resonance formula $\lambda = 4nh/(2m + 1)$. When $m$ is an integer, the corresponding $h$ is the thickness that enables the absorption rate to the maximum. Since the refractive index of the quartz layer is 1.94, so we can calculate the thicknesses for the first maximum absorption ($m = 1$) in the Zigzag and Armchair directions are 9.66 and 4.24 μm, respectively, agreeing well with the simulation results.

4. Conclusions

We investigated anisotropic plasmonic responses of 2D BP by patterning nanostrips along the Armchair and Zigzag directions, respectively. In the proposed BP terahertz metamaterial, to enhance the light-matter interaction, a sandwiched structure consisting of periodic BP nanostrips, a transparent insulator and a metallic mirror was employed. The results suggest that by combing the Fabry-Perot effect with the dipole resonance of the BP nanostrips, the absorption intensity has been enhanced and approached 0.51 in Armchair direction. Stemming from the intrinsic anisotropic structure of BP, different plasmonic responses were observed when the BP nanostrips were along the Zigzag direction. Furthermore, we analysed their anisotropic behaviours when changing the structural parameters in the unit cell. In addition, we investigated two types of coupling between the dipole resonances of the BP nanostrips. Although challenges still exist, such as weak-absorption and difficulty to obtain large monolayer BP film at present, the proposed BP metamaterial has proved that the monolayer BP could be used to build 2D anisotropic plasmonic devices, which may be verified experimentally in the future.

References


