Review

Renlong Zhou*, Kaleem Ullah*, Sa Yang*, Qiawu Lin, Liangpo Tang, Dan Liu, Shuang Li, Yongming Zhao and Fengqiu Wang

Recent advances in graphene and black phosphorus nonlinear plasmonics

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Abstract: Over the past decade, the plasmonics of graphene and black phosphorus (BP) were widely recognized as promising media for establishing linear and nonlinear light-matter interactions. Compared to the conventional metals, they support significant light-matter interaction of high efficiency and show undispersed optical properties. Furthermore, in contrast to the conventional metals, the plasmonic properties of graphene and BP structure can be tuned by electrical and chemical doping. In this review, a deep attention was paid toward the second- and third-order nonlinear plasmonic modes of graphene and BP. We present a theoretical framework for calculating the lifetime for surface plasmons modes of graphene and BP assisted by the coupled mode theory. The effect of the Fermi energy on the second-order and third-order nonlinear response is studied in detail. We survey the recent advances in nonlinear optics and the applications of graphene and BP-based tunable plasmonic devices such as light modulation devices, switches, biosensors, and other nonlinear photonic devices. Finally, we highlight a few representative current applications of graphene and BP to photonic and optoelectronic devices.

Keywords: graphene; black phosphorus; tunable nonlinear plasmonics; lifetime.

***Corresponding authors: Renlong Zhou and Sa Yang**, School of Physics and Information Engineering, Guangdong University of Education, Guangzhou 510303, China, e-mail: rlzhoupc@sina.com (R. Zhou); yangs7014@sina.com (S. Yang).

<https://orcid.org/0000-0002-1251-8910> (R. Zhou); and

Kaleem Ullah, School of Electronic Science and Engineering, Nanjing University, Nanjing 210093, China, e-mail: kaleem758@gmail.com

Qiawu Lin, Liangpo Tang, Dan Liu, Shuang Li and Yongming Zhao: School of Physics and Information Engineering, Guangdong University of Education, Guangzhou 510303, China

Fengqiu Wang: School of Electronic Science and Engineering, Nanjing University, Nanjing 210093, China

1 Introduction

The surface plasmons (SPs) can control and confine electromagnetic (EM) field in subwavelength scales. To control SPs, several structures such as topological defect, prism, cavity, and metamaterials can be employed [1]. Coupled with electrons, phonons, or photons, surface plasmons in metal and 2D material were investigated theoretically as well as experimentally for the development of linear and nonlinear photonics nanoplatform [2–7]. Previously, the novel plasmonic materials such as Ag, Al, and Au were used in broad applications for nonlinear plasmonic device [8, 9], temperature and gas sensor [10–13], flexible electrode [14], graphite-based batteries [15], terahertz metamaterial controlled with thermal [16], symmetry-breaking-induced nonlinearity in a microcavity [17], and nonlinearity in the dark [18]. However, these noble metals have large energy dissipation due to radiative and Ohmic losses. Moreover, these conventional metals exhibit a poor tunability. To solve this problem, the plasmonic of 2D materials, especially graphene and BP, attracted a deep interest owing to their high electrical tunability in contrast to noble metals.

The atomic thin 2D nanomaterial sheet presents significant light-matter interaction phenomena owing to quantum confinement effect. Their electronic structures and optical properties are distinctive from their bulk morphology. The carrier (both electrons and holes) density in graphene can be engineered by changing their surrounding dielectric environment or grating structure [19, 20]. The important role of BP plasmonics is an easy control on nanoscale electromagnetic field compared to the conventional materials [21, 22]. The frequency region for efficient wave localization can be up to mid-infrared and terahertz frequencies, and thus, it verifies that graphene and BP can be utilized in several promising applications such as the surface plasmons [23], photodetectors [24–28], enhanced Raman scattering [29], gate-tuning nano-imaging [30], optical modulator [31–36], ultrafast nonlinear ultrafast laser [37, 38], sensors [39], optical switches [40, 41], energy

conversion [42], optical force [43], plasmonic logic gate [44], and broadband light absorption in multiple graphene nanoresonator [45]. Several investigations on the BP plasmonic were carried out in literature such as the polarization-dependent symmetric and anti-symmetric plasmonic modes of BP pairs, which were found due to the anisotropic dispersion [46] and defect engineering in few-layer BP [47]. The graphene and BP plasmonic with a large nonlinear optical constant can be used as a photonic platform to study the nonlinear optical interaction within 2D material [48–53]. The graphene and BP plasmonic have the large third harmonic generation (THG) and strong Kerr nonlinearity [3, 54–56], and induced second harmonic generation (SHG) [57–59]. A detailed description of the graphene and BP plasmonic concepts, computational methods, and Z-scan measurements in nonlinear plasmonics can be found in a few excellent reviews [60–64]. The coexistence of local field enhancement and strongly induced nonlinearity in properly designed 2D materials open up an alternative route for high-efficiency tunable nonlinear devices. It is demonstrated that a broadband THz emission of 2D materials has a giant efficiency, which is about two orders stronger compared to that of a conventional metasurfaces [65, 66]. The 2D material plasmonics opens up opportunities for not only THz technology but also on-chip nonlinear nano-optical devices.

There are mainly two types of the methods for preparing graphene, BP, and 2D materials, which include dissection methods (top-down routes) and the growth methods (bottom-up routes). In layered materials such as graphene and BP, the atoms with strong in-plane chemical bonds are vertically stacked through a weak physical electrostatic force (van der Waals interaction) to form bulk crystals. This weak interaction between the layers provides us a possibility of top-down routes including micromechanical exfoliation method and liquid phase exfoliation [67, 68]. The bottom up routes for preparing the 2D materials are called the thin film deposition methods such as chemical vapor deposition (CVD), magnetron sputtering, atomic layer deposition (ALD), molecular beam epitaxy (MBE), pulsed-laser deposition (PLD), and so on [69–72]. Among these thin film deposition methods, the most famous method is CVD. In the CVD method, the synthesized 2D materials through the CVD method were grown on a substrate, where the source materials may be either carried by inert gas (e.g. argon) and then deposited on the target substrates or pre-deposited on the target substrates and then converted to 2D sheets [73].

Compared to the traditional mode-locked fiber applications [37, 74–92], the ultrafast saturable absorber (SA) of 2D material for wavelength insensitive plays an important

role in many fields [93–98]. A few-layer BP-based saturable absorber was experimentally demonstrated, which could be acted as an optical modulator used to generate short laser pulses in solid-state lasers [99]. MoS₂/graphene nanocomposites can act as ultrafast photonics and lasers [100, 101]. Tunable, electrically controlled graphene was investigated as a planar plasmonic filter [102, 103]. With the improvement in the oxygen evolution reaction activity, BP grown on a carbon nanotube network can show better activity in the electrocatalyst [104]. Ultrathin BP sheets exhibit efficient photothermal therapy due to strong near-infrared (NIR) light absorption and excellent photothermal performance [105]. BP has biomedicine application due to its unique properties of bio-compatibility [106]. Few-layer BP can also be applied as electrocatalysts due to highly efficient oxygen evolution reaction [107]. Graphene oxide and BP flake aerogels have an enhanced photothermal character for its robust thermo stability [108]. Light activation of BP applied for cancer therapy in hydrogel nanostructures can be used for the smart NIR light-controlled drug release [109]. At room temperature, the 2D material such as graphene and BP can be used for a THz detector with high performance [110], plasmonic fiberoptic biosensor [111], Faraday rotator [112], and chirped nonlinear photonic crystal cavity [113]. The composite structure of isotropic graphene and anisotropic BP has strong coherent coupling property, and their strong coupling between graphene and BP is described with an oscillator model [114].

In this review, we provide graphene and BP tunable nonlinear plasmonics information and its applications in the physical system. The weak light absorption of ultrathin graphene and BP sheets hinders their application, but the light absorption can be enhanced in graphene and BP using grating structure or by making a strong light-matter interaction system. Furthermore, the graphene and BP plasmonic properties can be modified by electrical and chemical doping. In section 2, we will present the theoretical basis including the optical conductivity and optical nonlinear properties, which are required to understand the graphene and BP tunable nonlinear plasmonics. In section 3, a detailed summary of the research results will be provided, which described the linear and nonlinear properties of the graphene and BP. We present a theoretical framework for calculating the lifetime for surface plasmon modes of graphene and BP assisted by coupled mode theory (CMT). The linear and nonlinear plasmonic modes of graphene and BP can be modified by changing Fermi energy and refractive index of medium. The polarization-resolved second and third harmonic generation emission were also analyzed. Furthermore, we study the

relation progress with topics: graphene plasmonic logic gate, graphene devices with bright visible light emission, photothermal therapy, and plasmonic nonamer antenna-graphene phototransistor, etc. Finally, we design the 2D material array chips, which illustrate the graphene and BP plasmonic nonlinear photonics nanoplatform with its applications and perspectives.

2 Plasmonic nonlinearity and theoretical background

In this section, we introduce the concepts of conductivity for graphene and BP plasmonic. We start with a brief description of the surface conductivity and nonlinear optical processes of graphene and BP tunable plasmonic. Finally, we continue with a more discussion of a lifetime using CMT.

2.1 Optical conductivity of graphene and BP

The optical feature of graphene can be expressed by the surface conductivity σ_{gra} composed of the intraband part and the interband part, $\sigma_{\text{gra}} = \sigma_{\text{intra}} + \sigma_{\text{inter}}$ [115]. The intraband electron-photon scattering contribution can be described by:

$$\sigma_{\text{intra}}(\omega) = -\frac{ie^2 k_B T}{\pi \hbar^2 (\omega - i\tau^{-1})} \left[\frac{E_f}{k_B T} + 2 \ln \left(e^{\frac{E_f}{k_B T}} + 1 \right) \right] \quad (1)$$

The interband transition contribution is described by:

$$\sigma_{\text{inter}}(\omega) = i \frac{e^2}{4\pi \hbar} \ln \left[\frac{2|E_f| - \hbar(\omega + i\tau^{-1})}{2|E_f| + \hbar(\omega + i\tau^{-1})} \right] \quad (2)$$

In addition, for the case including $\hbar\omega \gg k_B T$ and $|E_f| \gg k_B T$, in the mid-infrared and THz frequency region, the optical feature of graphene can be expressed by the surface conductivity σ_{gra} [116]:

$$\sigma_{\text{gra}}(\omega) = -ie^2 E_f / [\pi \hbar^2 (\omega - i\tau^{-1})] \quad (3)$$

Here, the carrier relaxation time is $\tau = (\mu E_f) / (e v_f^2)$. The Boltzmann's constant, reduced Planck's constant, temperature, electric charge, Fermi velocity, Fermi energy or chemical potential, and carrier mobility are k_B , $\hbar$, T , e , v_f , E_f , and μ , respectively. The transient optical conductivity and dielectric function of 2D material samples can be

experimentally measured [116–118]. The linear electric current response of the graphene in Maxwell equations is given by $\mathbf{J} = \sigma_{\text{gra}}(\omega) \mathbf{E} / \Delta$. The thickness of 2D material is set as Δ .

For another BP material, the electronic density n_s of BP is given by a diagonal tensor:

$$n_s = \begin{pmatrix} m_1 & 0 \\ 0 & m_2 \end{pmatrix} \frac{k_B T}{d\pi \hbar^2} \ln \left[1 + \exp \left(\frac{E_f - E_c}{k_B T} \right) \right] \quad (4)$$

where m_1 and m_2 denote the in-plane electron effective masses near the Γ point within the Hamiltonian model. m_1 and m_2 can be written with $m_1 = \hbar^2 / (2\gamma^2 / \Delta + \eta_c)$ and $m_2 = \hbar^2 / v_c$. Conduction band edge energy is noted with E_c . The conductivity of few-layer BP is given as [119, 120]:

$$\sigma_j = iD_j / (\pi(\omega + i\eta_e / \hbar)), \quad D_j = \pi e^2 n_{sj} / m_j (j=1, 2) \quad (5)$$

where, $D_j = \pi e^2 n_{sj} / m_j$ is the Drude weight along the j -axis, and j denotes the x or y direction. η_e describes the scattering rate. The BP flake is treated as a thin layer with the anisotropic Drude model. The linear electric current of the BP in Maxwell equations is given by $\mathbf{J}_j = \sigma_j \mathbf{E} / \Delta$.

2.2 Nonlinear optical processes of 2D materials

The electromagnetic response of the 2D materials is determined by the polarization $\mathbf{P}(\mathbf{r}, t)$. In order to study the second-order and third-order nonlinearity of an optical medium, the polarization $\mathbf{P}(\mathbf{r}, t)$ of the 2D materials is expanded in the power series using the electric dipole approximation [121, 122]: $\mathbf{P}(\mathbf{r}, t) = \epsilon \chi^{(1)}(\mathbf{r}, t) \mathbf{E}(\mathbf{r}, t) + \chi^{(2)}(\mathbf{r}, t) : \mathbf{E}(\mathbf{r}, t) \mathbf{E}(\mathbf{r}, t) + \chi^{(3)}(\mathbf{r}, t) : \mathbf{E}(\mathbf{r}, t) \mathbf{E}(\mathbf{r}, t) \mathbf{E}(\mathbf{r}, t)$, where, $\chi^{(i)}$ is the i -th-order susceptibility.

2.2.1 Second-order nonlinearity of graphene and BP

For the second-order nonlinearity of 2D materials such as graphene and BP, the incident electromagnetic field is composed of two monochromatic plane waves with frequencies ω_1 and ω_2 :

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_1 e^{i(\mathbf{k}_1 \cdot \mathbf{r} - \omega_1 t)} \mathbf{E}_2 e^{i(\mathbf{k}_2 \cdot \mathbf{r} - \omega_2 t)} \quad (6)$$

where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the corresponding wave vectors. For the second-order nonlinear polarization $\mathbf{P}^{(2)}(\mathbf{r}, t)$, we can obtain:

$$\begin{aligned} \mathbf{P}^{(2)}(\mathbf{r}, t) = & \varepsilon_0 \chi^{(2)}(\mathbf{r}, t) : [\mathbf{E}_1 \mathbf{E}_2 e^{2i(\mathbf{k}_1 \mathbf{r} - \omega_1 t)} \\ & + \mathbf{E}_2 \mathbf{E}_2 e^{2i(\mathbf{k}_2 \mathbf{r} - \omega_2 t)} 2\mathbf{E}_1 \mathbf{E}_2 e^{i[(\mathbf{k}_1 + \mathbf{k}_2) \mathbf{r} - (\omega_1 + \omega_2)t]} \\ & + 2\mathbf{E}_1 \mathbf{E}_2^* e^{i[(\mathbf{k}_1 - \mathbf{k}_2) \mathbf{r} - (\omega_1 - \omega_2)t]} + c.c.] \\ & + 2\varepsilon_0 \chi^{(2)}(\mathbf{r}, t) : (\mathbf{E}_1 \mathbf{E}_1^* + \mathbf{E}_2 \mathbf{E}_2^*) \end{aligned} \quad (7)$$

The terms in the Eq. (7) have the second-order nonlinearity including SHG at $2\omega_1$ and $2\omega_2$, difference frequency generation (DFG) at $(\omega_2 - \omega_1)$, sum frequency generation (SFG) at $(\omega_1 + \omega_2)$ and optical rectification, respectively. The last term of the right side in Eq. (7) is a static polarization. The polarization $\mathbf{P}^{(2)}(\mathbf{r}, t)$ in Eq. (7) describes the nonlinear optical processes in 2D materials, including SHG, SFG, and DFG.

The local response approximation model, such as the Drude model, is used to model metal plasmonic nanostructures. Nowadays, nanoscale fabrication techniques for 2D material plasmonic nanodevices allow us to investigate the new phenomena occurring in physical regimes of 2D materials. When an optical field strongly couples to a 2D material plasmonic structure, it interacts with free electrons resulting in intrinsic nonlinear optical response including SHG, SFG, DFG effects, and other higher harmonic generation.

The linear and second-order nonlinear optical responses between light and BP are described by Maxwell equations. The second-order nonlinear theory of the 2D material plasmonic nanostructures is based on the nonlinear Lorentz force like that in metal. The electrons are described by the velocity field $\mathbf{u}_e(\mathbf{r}, t)$ and electronic number density $n_e(\mathbf{r}, t)$. The total electronic charge density is defined as $en_e(\mathbf{r}, t) = [en_0(\mathbf{r}) - \rho(\mathbf{r}, t)]$. The electronic current density was defined with the following equation: $\mathbf{j}(\mathbf{r}, t) = -en_e \mathbf{u}_e = (\rho - en_0) \mathbf{u}_e$. The force density $\mathbf{F}$ consists of the electric and magnetic part of the Lorentz force, and force density $\mathbf{F}$ has the term: $\mathbf{F} = -en_e(\mathbf{E} + \mathbf{u}_e \mathbf{B}) - m_e \gamma n_e \mathbf{u}_e$. The evolution for electronic current density is expressed with: $\partial \mathbf{j} / \partial t = -e/m_e \mathbf{F} - \sum_k \partial(\mathbf{j}_k / (en_0 - \rho)) / \partial r_k$.

Instead of the force density $\mathbf{F}$ in the expression, $\partial \mathbf{j} / \partial t$ yields the time derivative for nonlinear current density:

$$\frac{\partial \mathbf{j}}{\partial t} = -\gamma \mathbf{j} + \beta \mathbf{E} + \sum_k \frac{\partial}{\partial r_k} \left(\frac{\mathbf{j}_k}{en_0 - \rho} \right) - \frac{e}{m_e} [\rho \mathbf{E} + \mathbf{j} \times \mathbf{B}] \quad (8)$$

The second-order nonlinear source $\mathbf{S}^{(2)}$ for second-order nonlinearity can be described with:

$$\mathbf{S}^{(2)} = \sum_k \frac{\partial}{\partial r_k} \left(\frac{\mathbf{j}^{(1)} \mathbf{j}_k^{(1)}}{en_0} \right) - \frac{e}{m_e} [\varepsilon_0 (\nabla \cdot \mathbf{E}^{(1)}) \mathbf{E}^{(1)} + \mathbf{j}^{(1)} \times \mathbf{B}^{(1)}] \quad (9)$$

Here, k is noted as the coordinates x , y , and z . The variables $\mathbf{j}^{(1)}$, $\mathbf{B}^{(1)}$, and $\mathbf{E}^{(1)}$ is the current density, magnetic

flux intensity, and electric field vector of fundamental frequency wave (FFW), respectively, which are noted with superscript (1). m_e and n_0 stand for effective electron mass and ion density, respectively. The $\mathbf{S}^{(2)}$ is the nonlinear source of second harmonic wave (SHW) noted with superscript (2). The electric part and magnetic part of the Lorentz force contribute to the nonlinear optics. Using the electric dipole approximation, the electric field of SHW can be related to the fundamental frequency wave:

$$\mathbf{E}^{(2)}(2\omega) \propto \chi^{(2)} \cdot \mathbf{E}^{(1)}(\omega) \mathbf{E}^{(1)}(\omega) \quad (10)$$

where $\chi^{(2)}$ is the susceptibility of the second-order nonlinear for graphene and BP.

2.2.2 Third-order nonlinearity of graphene

The third-order nonlinear optical property in ultra-thin graphene plasmonics is investigated here. The Kerr effect of the third-order nonlinear polarization of graphene is expressed as following:

$$\mathbf{P}^{(3)}(\omega) = 3\varepsilon_0 \chi^{(3)} |\mathbf{E}(\omega)|^2 \mathbf{E}(\omega) \quad (11)$$

In Eq. (11), the third-order susceptibility $\chi^{(3)}$ can be obtained in nonlinear graphene or BP materials.

The graphene lattice with D_{6h} space group is centrosymmetric. A direct implication of this property is that second-order nonlinearity is forbidden. However, nonlinearity for THG is allowed and particularly strong in graphene. The quadratic optical nonlinearity of graphene can be described using the nonlinear optical conductivity tensor σ_3 . The current density of the third-order nonlinearity is $\mathbf{j}^{3nl}(\mathbf{r}, t) = \sigma_3 E(\mathbf{r}, t) |E(\mathbf{r}, t)|^2$. The third-order optical conductivity has a dependence on the frequency and Fermi energy. The nonlinear conductivity σ_3 of graphene has the following form [123–125]:

$$\sigma_3(\omega) = -i3e^2(\nu_f e)^2 / (32\pi E_f \omega^3) \quad (12)$$

Here, the imaginary part of the nonlinear conductivity σ_3 is negative, which describes the self-focusing type nonlinear response in graphene [126]. Both linear conductivity σ_{gra} and σ_3 nonlinear conductivity are highly dependent on Fermi energy, which could provide a way to get an electrically controlled optical bistability [127]. After considering the nonlinear effect of graphene, the conductivity σ of graphene can be written as $\sigma = \sigma_{\text{gra}} + \sigma_3 |\mathbf{E}(\mathbf{r}, t)|^2$, and the dielectric constant of graphene can be set as $\varepsilon = \varepsilon_{\text{gra}} + \chi^{(3)} |\mathbf{E}(\mathbf{r}, t)|^2$.

2.2.3 Third-order nonlinearity of BP

In the space group, BP has an orthorhombic crystal structure. For an orthorhombic crystal exhibiting THG, the contracted third-order nonlinear susceptibility tensor of BP can be written as follows [128]:

$$\chi_{BP} = \begin{bmatrix} \chi_{11} & 0 & 0 & 0 & 0 & \chi_{16} & 0 & \chi_{18} & 0 & 0 \\ 0 & \chi_{22} & 0 & \chi_{24} & 0 & 0 & 0 & 0 & \chi_{29} & 0 \\ 0 & 0 & \chi_{33} & 0 & \chi_{35} & 0 & \chi_{37} & 0 & 0 & 0 \end{bmatrix} \quad (13)$$

where “1, 2, 3” is the first subscript, which refers to “x, y, z,” respectively. The second subscript signifies the following meaning in Table 1. In our paper, the BP sheet is set along the x-y plane. If we only excite the x-y plane, all components containing a “z” term can be set as zero. Only non-zero elements ($\chi_{11}, \chi_{22}, \chi_{18}, \chi_{29}$) can determine the third-order nonlinearity of BP. So, the output electric field intensity can be expressed as follows:

$$E_x^2 \propto [\chi_{11} \cos^3(\theta) + \chi_{18} \cos(\theta) \sin^2(\theta)]^2 \quad (14)$$

$$E_y^2 \propto [\chi_{22} \sin^3(\theta) + \chi_{29} \sin(\theta) \cos^2(\theta)]^2 \quad (15)$$

Here, θ is the polarization angle.

2.3 The linear and nonlinear coupled mode theory

The coupled mode theory has a long history of application in optics, where it offers a good description on the propagation of light in a set of coupled linear and nonlinear optical waveguides. The characteristics of the SP mode can be analyzed with the CMT method. The resonance modes have the energy amplitude A_m ($m=1, 2, 3$), where $dA_m/dt = -i\omega A_m$. The incoming and outgoing waves are depicted by $S_{\pm, \text{in(out)}}$. The amplitude A_m has the expression [129, 130]:

$$\begin{aligned} \frac{dA_m}{dt} = & \left(-i\omega_m - \frac{1}{\tau_{im}} - \frac{1}{\tau_{wm}} \right) A_m + S_{+, \text{in}} \sqrt{\frac{1}{\tau_{wm}}} \\ & + S_{-, \text{in}} \sqrt{\frac{1}{\tau_{wm}}} - \sum_{p \neq m} i\mu_{mp} A_p \end{aligned} \quad (16)$$

Table 1: List of the second subscript “m” number meaning in third-order susceptibility uses in Eq. (13) such as χ_{29} (second subscript is 9).

jkl	xxx	yyy	zzz	yzz	yyz	xzz	xxz	xyy	xyx	xyz
m	1	2	3	4	5	6	7	8	9	0

where, τ_{im} is the lifetime of decay process due to the intrinsic loss for the m-th mode. τ_{wm} is the lifetime of energy coupling process for the m-th mode. μ_{mp} ($m, p=1, 2, 3$ and $m \neq p$) is the coupling coefficient between the three resonant modes. The absorption of the 2D materials system can be expressed as:

$$A(\omega, E_f) = 1 - |t|^2 - |r|^2 \quad (17)$$

where the transmission function of 2D material plasmonic system is: $t(\omega, E_f) = 1 - [(\tau_{w1})^{-1/2} + (\tau_{i1})^{-1/2}] P_1/P_0 - [(\tau_{w2})^{-1/2} + (\tau_{i2})^{-1/2}] P_2/P_0 - [(\tau_{w3})^{-1/2} + (\tau_{i3})^{-1/2}] P_3/P_0$. In addition, the reflection function is $r(\omega, E_f) = -(\tau_{w1})^{-1/2} P_1/P_0 - (\tau_{w2})^{-1/2} P_2/P_0 - (\tau_{w3})^{-1/2} P_3/P_0$. P_0, P_1, P_2 , and P_3 are the functions of $\omega, \omega_m, \tau_{im}, \tau_{wm}$, and μ_{mp} . After adjusting the parameters of τ_{wm}, τ_{im} , and μ_{mp} , we can precisely compare the intensity, resonant wavelength, and spectral width of absorption peaks $A(\omega)$ in Eq. (17) with that of the absorption spectrum simulated by the FDTD method. Then, we can choose the appropriate value of the parameters ω_m, τ_{im} , and τ_{wm} . The theoretical descriptions and data fitting will make it useful in applying the methods for future 2D material plasmonic applications.

In particular, for a second-order nonlinearity with mode $\omega_{s,1}$ and its second harmonic $\omega_{s,2}$, the amplitudes $a_{s,1}$ and $a_{s,2}$ of the nonlinear coupled mode equations take the following form:

$$\begin{aligned} \frac{da_{s,1}}{dt} = & \left(-i\omega_{s,1} - \frac{1}{\tau_{s,i1}} - \frac{1}{\tau_{s,w1}} \right) a_{s,1} + S_{+, \text{in}} \sqrt{\frac{1}{\tau_{s,w1}}} \\ & + S_{-, \text{in}} \sqrt{\frac{1}{\tau_{s,w1}}} - i\omega_{s,1} \beta_1 a_{s,1}^* a_{s,2} \end{aligned} \quad (18)$$

$$\begin{aligned} \frac{da_{s,2}}{dt} = & \left(-i\omega_{s,2} - \frac{1}{\tau_{s,i2}} - \frac{1}{\tau_{s,w2}} \right) a_{s,2} + S_{+, \text{in}} \sqrt{\frac{1}{\tau_{s,w2}}} \\ & + S_{-, \text{in}} \sqrt{\frac{1}{\tau_{s,w2}}} - i\omega_{s,2} \beta_2 a_{s,1}^2 \end{aligned} \quad (19)$$

Similarly, for a third-order nonlinearity with mode $\omega_{t,1}$ and its third harmonic mode $\omega_{t,3}$, the amplitudes $a_{t,1}$ and $a_{t,3}$ of the coupled mode equations take the form:

$$\begin{aligned} \frac{da_{t,1}}{dt} = & \left[-i\omega_{t,1} (1 + \eta_{11} |a_{t,1}|^2 + \eta_{13} |a_{t,3}|^2) - \frac{1}{\tau_{t,i1}} - \frac{1}{\tau_{t,w1}} \right] a_{t,1} \\ & + S_{+, \text{in}} \sqrt{\frac{1}{\tau_{t,w1}}} + S_{-, \text{in}} \sqrt{\frac{1}{\tau_{t,w1}}} - i\omega_{t,1} \beta_1 (a_{t,1}^*)^2 a_{t,3} \end{aligned} \quad (20)$$

$$\frac{da_{t,3}}{dt} = \left[-i\omega_{t,3}(1 + \eta_{33}|a_{t,3}|^2 + \eta_{31}|a_{t,1}|^2) - \frac{1}{\tau_{t,3}} - \frac{1}{\tau_{t,w3}} \right] a_{t,3} + S_{+,in} \sqrt{\frac{1}{\tau_{t,w3}}} + S_{-,in} \sqrt{\frac{1}{\tau_{t,w3}}} - i\omega_{t,3}\beta_3(a_{t,1})^3 \quad (21)$$

In derivative equations of $a_{t,1}$ and $a_{t,3}$, one kind of nonlinearity depends on one of the field amplitudes. For the $\chi^{(3)}$ nonlinearity, this effect is known as self-phase and cross-phase modulation. Another kind of nonlinearity originates from transferring energy between the modes [131].

3 Surface plasmons and their nonlinearity in graphene and BP

A single layer of graphene sample was grown on a copper foil [132]. The doping of the graphene layer was controlled by changing the duration of nitric acid vapor treatment. The reflectance R of the samples was measured with a

Fourier transform infrared spectrometer. The absorptivity can be obtained with $A=1-R$. The angle-selective perfect light absorption was experimentally achieved in Figure 1A.

Construction of such an angle-selective absorption structure is important for the applications such as energy harvesting, photo-detection, and sensing. The weak light absorption of the graphene layer hinders their application for the incident angle around $\theta=0^\circ$. The light absorption in graphene or BP can also be tuned with the grating structure below. The two-dimensional material-based tunable plasmonic nonlinear photonics nanoplatform can be established based on the grating structure or other strong light-matter interaction systems.

3.1 Linear and nonlinearity of graphene plasmonic

Graphene has a gate-voltage-dependent feature that the Fermi energy of graphene can be tuned dynamically using the bias voltage. We consider a periodic grating, formed

Figure 1: Angle-selective, grating-selective absorption and their lifetime in graphene structure.

(A) The angle-selective absorption and the planar graphene structure [132]. (B) The nanostructured DGN surrounded with substrate and superstrate such as aqueous solution. (C) The simulated linear-optical absorption with FDTD simulation (black circles) and the theoretical CMT analysis results (red line). τ_{w1} , τ_{w2} , τ_{l1} , and τ_{l2} various with (D) Fermi energy E_f and (E) refractive index n_1 .

by the double graphene nanoribbon (DGN) arrays, which has a lattice constant $L=200$ nm in the xy -plane. The graphene grating is placed on the dielectric substrate as shown in Figure 1B. The graphene nanoribbons have the cuboid shape (length $\times$ width $\times$ thickness) with $L_1 \times L_2 \times \Delta$. The graphene nanoribbons are set as length $L_1=160$ nm, width $L_2=40$ nm, and thickness $\Delta=2$ nm. Other parameters of the graphene are chosen as $v_f=10^6$ m/s, $\mu=10,000$ cm²/(V $\cdot$ s), and $E_f=0.64$ eV. Using the FDTD method and CMT theory, the absorption (marked with red dotted) of the DGN grating is investigated in Figure 1C. The theoretical CMT results (black line) are in accordance with the FDTD simulation (red dotted).

The lifetime τ_{wm} and τ_{im} ($m=1, 2$) can be modulated with the Fermi energy E_f and refractive index n_1 in Figure 1D and E, respectively. The lifetimes τ_{wm} and τ_{im} ($m=1, 2$) strongly depend on the parameter of Fermi energy E_f , which can be through doping or an applied gate voltage, thus, resulting in the sensing application with applied voltage bias. The lifetimes τ_{w1} , τ_{w2} , τ_{i1} , and τ_{i2} can be modulated with the refractive index n_1 of the superstrate. The theoretical descriptions of the tunable lifetime will make it useful in applying the theory for future 2D material-sensing applications by changing the refractive index n_1 of the superstrate such as aqueous solution.

For example, the incident electromagnetic field is composed of two monochrome plane waves. The incident electric field with two frequencies ω_1 and ω_2 can be written as:

$$\mathbf{E}^{(1)}(\mathbf{r}, t) = \mathbf{E}_0^{(1)}(\mathbf{r}) (e^{-i\omega_1 t} + e^{-i\omega_2 t}) \quad (22)$$

In the case of the incident wavelengths $\lambda_1=4.4$ μm and $\lambda_2=4.9$ μm in Figure 2A, the second-order nonlinearity including SHG as well as SFG and DFG can be obtained in Figure 2B. We can obtain the SHW including SHG ($\lambda_3=2.2$ μm , $\lambda_4=2.45$ μm) as well as the SFG ($\lambda_5=2.32$ μm) and DFG ($\lambda_6=38.6$ μm) in Figure 2B. The second-order nonlinear conversion efficiency is large compared to that in single graphene nanoribbon grating.

Let us now assume that the incident field is composed of two short pluses. The short pluses with two central frequencies ω_1 and ω_2 can be written as $\mathbf{E}^{(1)}(t) = \exp(-(t-t_0)/(t_d)) (e^{-i\omega_1 t} + e^{-i\omega_2 t})$, where t_0 is the delay of time, and $t_d=200$ fs. For Fermi energy $E_f=0.64$ eV, the time evolution of lifetimes for FFW at wavelengths λ_1 and λ_2 are shown in Figure 2C. In addition, time evolution of lifetimes for SHW at wavelengths λ_3 , λ_4 , and λ_5 are shown in Figure 2D [133]. The carrier dynamic of a saturable absorber plays an important role to determine how a short pulse can be produced in passively mode-locked laser.

Illuminated with an x -polarized plane wave at the fundamental frequencies λ_1 and λ_2 , the polarization state of the second harmonic emission for total electric field energy ($|E|^2$) at (e) λ_3 , (f) λ_4 , (g) λ_5 , and (h) λ_6 from an array of double graphene nanoribbons are shown in Figure 2E–H. The second harmonic signal is a function of the measuring angle (not the polarization of the incident field) $\theta=0$ corresponds to the x direction.

Here, we consider the third-order nonlinear optical property in nonlinear 2D material graphene plasmonics. In the case of the incident wavelengths $\lambda_1=4.4$ μm , the THG signal of the total electric field energy ($|E|^2$) with three different Fermi energies $E_f=0.4$ eV, 0.6 eV, and 0.8 eV are shown in Figure 3A. The polar diagram of the polarization state of the second harmonic emission at $\lambda_1/3$ and $E_f=0.6$ eV is shown in Figure 3B. In addition, the polarization state of the second harmonic emission for the total electric field energy ($|E|^2$) at $\lambda_1/3$ is a function of the measuring angle (not the polarization of the incident field $\theta=0$) corresponding to the x direction.

The sandwiched graphene structure is composed of two dielectric layers and a single graphene layer in Figure 3C. There was a weak light absorption of the 2D material layer for the incident angle around $\theta=0^\circ$ as shown in Figure 1A. Here, we consider the incident angle $\theta=75^\circ$. The refraction index of dielectric 2 and dielectric 3 is n_2 and n_3 , respectively. Dielectrics 1 and 4 are set as air. The thicknesses of dielectric slabs 2 and 3 are set as d_2 and d_3 , respectively. The graphene layer is sandwiched between the superstrate and substrate dielectric slab 2 and slab 3. Without considering the external field and under the random-phase approximation, the surface conductivity of the graphene is $\sigma_{\text{gra}} = \sigma_0$. The nonlinear surface conductivity of graphene is σ_3 , E_i , E_r , and E_t are the amplitude of incident, transmitted and reflected magnetic fields, respectively. An amplitude E_i of a plane wave is incident on the graphene structure at the incident angle θ .

The x axis is parallel along with the graphene layer, and the z axis is norm to the graphene layer. The electric fields and magnetic field in dielectric layers 1, 2, 3, and 4 are noted as (E_{1x}, E_{1z}, H_{1y}) , (E_{2x}, E_{2z}, H_{2y}) , (E_{3x}, E_{3z}, H_{3y}) , and (E_{4x}, E_{4z}, H_{4y}) , respectively. In addition, $k_x = k_0 n_1 \sin\theta$, and $k_{iz} = \text{sqrt}(k_0^2 n_i^2 - k_x^2)$, $j=1, 2, 3, 4$. θ is the incident angle. By considering the boundary condition of electromagnetic field at position $z=-d_2, 0, d_3$, there is the relation of transmitted electric field and incident electric field E_i and E_t : $|E_i|^2 = |E_t|^2 |U(E_i)|^2$. Clearly, the incident electric field is the function of the transmitted electric field, and hence, optical bistability can be obtained with appropriate conditions [134, 135]. The dependencies of the transmitted electric field on the input electric field at wavelength 60 μm

Figure 2: Wavelength and polarization dependent SHG of nanostructured DGN.

(A) The amplitude of Fourier spectrum for FFW $E_x^{(1)}$ with two wavelengths λ_1 and λ_2 for the nanostructured DGN. (B) The amplitude of Fourier spectrum for SHW $E_x^{(2)}$ with the four wavelengths λ_3 , λ_4 , λ_5 , and λ_6 . (C) Lifetime of FFW at λ_1 and λ_2 changing with time. (D) Lifetime of SHW at λ_3 , λ_4 , and λ_5 changing with time. The polarization state of total electric field energy ($|E|^2$) for second harmonic emission at (E) λ_3 , (F) λ_4 , (G) λ_5 , and (H) λ_6 from an array of DGN illuminated with an x-polarized plane wave at the fundamental frequency λ_1 and λ_2 .

for the different Fermi energies and index of superstrate dielectric as shown in Figure 3D and E, respectively.

The plasmonic devices were fabricated with chemical vapor deposition-grown monolayer graphene that was transferred onto a silicon oxide on lightly doped silicon in Figure 4A. Figure 4B illustrates the experimental setup that was used to investigate the nonlinear terahertz response. Measured relative change in various transmissions with pump-probe time delay Δt is depicted for several pump fluences in Figure 4C. In all cases, the pump causes an increase in transmission, which is accompanied with a decrease in absorption [133]. The observed nonlinear response decays is about 10 ps.

The strong enhancement of light absorption in graphene can be realized using periodically patterned structure and optical graphene-cavity system [136]. The polarization directions of input and output light are shown in Figure 5A. The monolayer graphene reduces the cavity reflection and reduces the cavity quality factor. The cavity-enhanced Raman spectroscopy is obtained in the graphene sample. The attenuation at the peak of reflection increases with 20 dB by inserting a graphene layer in Figure 5B. The graphene's complex refractive index can also be measured in the coupled graphene-cavity system.

The second-order susceptibility $\chi^{(2)}$ of graphene/gold nanohole array/glass structure [137] was studied with

Figure 3: Polarization and Fermi energy dependent THG in sandwiched graphene sheet. (A) THG signal of total electric field energy ($|E|^2$) for three different Fermi energies $E_f = 0.4$ eV, 0.6 eV, and 0.8 eV of FW mode at λ_1 in the nanostructured DGN. (B) Polar diagram of polarization state of the second harmonic emission at $\lambda_1/3$. (C) The sandwiched graphene sheet structure surrounded with substrate and superstrate dielectric. The dependencies of the transmitted electric field on the input electric field at a wavelength of 60 μm for different (D) Fermi energies and (E) index of superstrate dielectric.

different polarizations in Figure 5C. The SHG image was collected with the graphene-coated sieve by a CCD camera in Figure 5D. The second-order susceptibility of the experimental results depends on the Fermi level and can be retrieved with a simple classical model $\chi_{\text{eff}}^{(2)} = e/(2m_e\omega^2)(e^w - 1)$. The second-order susceptibility $\chi^{(2)}$ can be controlled with Fermi level, which is the advantage of graphene plasmonic compared with that of conventional metal.

3.2 Linearity and nonlinearity of BP plasmonic

We considered a nanostructured BP nanoflake with period $P = 200$ nm in the xy -plane. The BP square array is placed on the SiO_2 substrate in Figure 6A. The BP nanoflake is shown in Figure 6B. The two BP nanoflakes have a cuboid shape (length $\times$ width $\times$ thickness) with $L \times L \times \Delta$. The parameters about the BP flake are set as side length $L = 70$ nm and thickness $\Delta = 2$ nm. The superstrate is the dielectric with refractive index n_1 . For the nanostructured BP in the mid-IR and far-IR wavelength regimes, the parameters can be set as follows: the band gap Δ is about 0.8 eV for thickness $d = 2$ nm, and the band gap is thickness dependent. $\epsilon_f = 5.76$, $\gamma = 4a/\pi\text{eV}\cdot\text{m}$ ($a = 0.223$ nm is scale length), $\eta_c = \hbar^2/(0.4 m_0)$, $v_c = \hbar^2/(0.7 m_0)$, $\eta_e = 10^{-3}$ eV, and $E_f - E_c = 0.6$ eV. The electron mass $m_0 = 9.10938 \times 10^{-31}$ kg.

The normalized linear-optical absorption [138] (black circles) of FFW through the nanostructured BP flake is investigated with the FDTD simulation as shown in Figure 6C. The theoretical CMT result is noted with a red line in Figure 6C. For the incident fundamental wave at two different wavelengths $\lambda_1 = 4.35$ μm and $\lambda_2 = 5.1$ μm in Figure 6D, we can obtain the SHW including SHG ($\lambda_3 = 2.18$ μm , $\lambda_4 = 2.55$ μm) as well as SFG ($\lambda_5 = 2.36$ μm) in Figure 6E.

Figure 4: Pump probe measurements of graphene gratings on SiO_2 Substrate [133]. (A) Cross-sectional diagram of device with graphene grating on SiO_2 . (B) Experimental setup for the pump-probe measurements. (C) Measured relative change in transmission along the time axis Δt under different pump fluencies [133].

Figure 5: SHG measurement results of graphene-cavity structure [136].

(A) Cross-polarization confocal microscope. HWP, half-wave plate; PBS, polarized beam splitter. (B) Reflection before and after the deposition of graphene. Inset: attenuation of the cavity with graphene [136]. (C) Schematic of SHG measurements in transmission mode in graphene/gold nanohole array/glass substrate structure. (D) The SHG image is collected for the graphene-coated sieve by a CCD camera [137].

Figure 6: Absorption, FFW, SHW and lifetime of FFW in nanostructured BP.

(A) The nanostructured BP nanoflake on SiO_2 substrate. (B) The atomic arrangement of the BP structure. (C) The linear-optical absorption with FDTD simulation (black circles) and the theoretical CMT (red line). (D) The amplitude of Fourier spectrum for FFW $E_x^{(1)}$ with two wavelengths peaks λ_1 and λ_2 . (E) Amplitude of Fourier spectrum for SHW $E_x^{(2)}$ with the three wavelengths peaks λ_3 , λ_4 , and λ_5 . (F) Lifetime τ_{w1} , τ_{w2} , τ_{i1} , and τ_{i2} as a function of various relative Fermi energies ($E_f - E_c$).

The conversion efficiency η of the second-order nonlinearity is about 10^{-10} . The second-order nonlinear conversion efficiency is larger than that in the single BP nanoflake grating [111]. The strongly enhanced electric field and noncentrosymmetry increase the signals of second-order nonlinearity. In the process of energy coupling between each mode and light field, the lifetime τ_{w1} and τ_{w2} of the plasmon modes with various relative Fermi energies ($E_f - E_c$) are shown in Figure 6F. In the process of intrinsic loss for the m -th mode, the lifetime for τ_{i1} and τ_{i2} with various ($E_f - E_c$) is also shown in Figure 6F. The intensity, wavelengths, and spectral width of the resonance peaks can be precisely modulated with the Fermi energy here. Enhanced absorption can be obtained using the periodically patterned structures and graphene cavity [139, 140]. We present a theoretical framework for calculating the lifetime of BP surface plasmon modes here. The carrier dynamics with two different decay times, 873 fs and 96.9 ps, and nonlinear optical properties were experimentally investigated in the quasi-BP system via transient transport and the Z-scan method [141]. The investigation of wavelength conversion and all-optical switching in the BP analog 2D material could be helpful for all optical signal processes [142].

According to the angle-resolved pump-probe reflection spectroscopy, the dynamical response for BP under ultrafast photo excitation is studied in Figure 7A. Under

Figure 7: Polarization dependent linear reflection contrast and THG spectra of BP flake [143, 144].

(A) Polarization-resolved reflection experiment. (B) Measured relative change in reflection varying with pump-probe time delay Δt (arrow: pump polarization along 0° and 90°) [143]. The data is fitted with biexponential decay function (decay time constants: $\tau_1 = 5.96$ ps, $\tau_2 = 87.6$ ps). (C) The THG of BP flake. (D) The experiment measurement equipment for THG. (E) Polar plot of the measured angular dependence of the THG from a 26-nm BP flake on a glass substrate. The red and purple arrows indicate the AC and ZZ crystal directions obtained from polarized Raman measurements [144].

pump polarization along 0° and probe polarization at 90° , the relative change in transient reflection as a function of pump-probe time delay Δt is measured as shown in Figure 7B. The data is fitted using the biexponential decay function with two decay time constants: $\tau_1 = 5.96$ ps, $\tau_2 = 87.6$ ps [143]. This experimental work provides a way for understanding angle-sensitive properties of BP-based devices. The anisotropic third harmonic generation with third-order susceptibility $\chi^{(3)}$ about $\chi^{(3)} = 1.64 \times 10^{-19}$ m² V⁻² in exfoliated BP was studied using a fast scanning multiphoton characterization method in Figure 7C. The exfoliated samples of BP were prepared with mechanical exfoliation in bulk BP with scotch magic tape onto glass or SiO₂/Si substrates. The thickness of a BP flake is 26 nm. Then, the samples were encapsulated using an AlO_x coating grown. The THG was measured with a multiphoton microscope. A description of the equipment for experimental measurement is shown in Figure 7D. The polar plot of the measured angular dependence of the THG from a BP flake on a substrate is shown in Figure 7E. After the polarized Raman measurements, the red and

purple arrows indicate the AC and ZZ crystal directions [144].

The nonlinear optical response of oxidized black phosphorus (OBP) was experimentally investigated using femtosecond Z-scan technology. The ultrafast nonlinear optics is obtained in the few-layer OBP. The saturable absorption and self-defocusing are observed here. These optical properties are useful in many optoelectronic devices, such as mode locker, wavelength converter, optical switch, and laser beam shaper.

If we only consider the exciting third-order nonlinearity of BP in the x - y plane here, we can set all components that contain a z term to zero in the third-order nonlinear susceptibility tensor in Eq. (13) [128]. Only four non-zero matrix elements ($\chi_{11}, \chi_{22}, \chi_{18}, \chi_{29}$) are left, which determine the THG in BP. Because the third-harmonic electric field is proportional to the nonlinear susceptibility, we can write the THG output intensity as follows: $E_x^2 \propto [\chi_{11} \cos^3(\theta) + \chi_{18} \cos(\theta) \sin^2(\theta)]^2$ and $E_y^2 \propto [\chi_{22} \sin^3(\theta) + \chi_{29} \sin(\theta) \cos^2(\theta)]^2$, where θ is the polarization angle for the x -axis of the BP crystal. The solid lines in Figure 8B are fitted to the measured intensity

Figure 8: Anisotropic THG in multilayer BP [128].

(A) Dependence of THG on the incident polarization. Zero degrees corresponds to the x -axis (armchair direction) of the BP crystal. (B) The same data as in (A) with theoretical fits to the intensity polarized in the x (black squares) and y (red dots) directions. Blue triangles correspond to the total intensity. (C) Calculated THG current induced in BP. (D) Atomic force microscopy image of a BP flake containing many layers with various thicknesses. (E) Position-dependent THG signal measured by scanning the position of the beam relative to the sample. Different thicknesses can be resolved via contrast in the THG signal.

in Figure 8A in the x and y directions using the relationships. One can see good agreement with the overall fit, excluding some asymmetry in the $|E_y|^2$ data. This asymmetry is likely caused by some residual polarization dependence in the transmission of the dichroic mirror that we were not able to account for during the measurement. The calculated THG current induced in BP is also shown in Figure 8C.

The third harmonic generation in BP is observed using an ultrafast NIR laser. The THG experiment results are observed using atomic force microscopy in Figure 8D. The third harmonic emission is dependent on the incident polarization. In addition, the third harmonic emission varies with the number of layers due to phase-matching condition and signal depletion. To prepare the samples, the author used Scotch tape to exfoliate single-crystal BP onto glass slides covered with polydimethylsiloxane. A femtosecond, near-IR fiber laser was used as the pump to probe the THG response of the samples. A short-pass

dichroic filter was used to reflect the fundamental frequency toward the sample while passing the third harmonic. Position-dependent THG signal is measured by scanning the position of the beam relative to the sample in Figure 8E. Different thicknesses can be resolved via contrast in the THG signal. An integrated optical device composed of an erbium-doped fiber, coupler, isolator, SA, and wavelength division multiplexer is studied [145]. A mechanically exfoliated BP was fabricated to realize the optical SA, which can be applied in the field such as ultra fast laser and ultra fast photonics [89, 146–148].

4 Application and perspectives for graphene and BP plasmonics

The graphene and BP plasmonic have become a fertile ground for the connection of the nonlinear photonics

community and their application. The graphene plasmonic has tunability because the carriers can be controlled with electrical doping and bias voltage. The graphene plasmonic logic gate and dual-gate BP FET are based on the tunability of surface plasmons with bias voltage. Graphene-based tunable plasmonic linear and nonlinear photonics also have the bright visible light emission. Moreover, plasmonic-induced hot electronics and quantum dot p-doping can be used to create a diode and an n-p-n transistor. Graphene and BP plasmonic-based tunable linear and nonlinear photonics play an important role in the nanoplatform with the various linear or nonlinear optical phenomena and their applications.

Graphene plasmonic logic gates were studied at the mid-infrared wavelengths in Figure 9A [149]. The electro-optical graphene plasmonic modes are controlled by an external gate voltage. Graphene plasmonic logic gates are superior to the conventional optical logic gates. The six basic logic gates (i.e. NOR/AND, XNOR/XOR, NAND/OR) are designed. The state of “ON/OFF” of each input channel can be controlled by tuning the optical conductivity of graphene. The six basic logic gates can also be controlled with the external gate voltage. This structure of graphene logic gate is compact, and the geometrical size is small.

The graphene plasmonic logic gates have an important application in photonic integrated circuits [152].

A dual-gate FET with a 12-nm-thin BP channel was fabricated on a glass substrate in Figure 9B. The Al_2O_3 dielectrics, which were 30-nm thick, were deposited on the top and bottom of the BP channel. In addition, BP FETs have a patterned Au-gate architecture out of the Al_2O_3 dielectric layers. Top Al_2O_3 dielectric was also used as an encapsulation layer for the FET device. The measured maximum ON currents were about $5\text{ }\mu\text{A}$ and $50\text{ }\mu\text{A}$ at drain voltages of -0.1 V and -1 V , respectively. The dual-gate BP FETs can nicely demonstrate the OLED switching for green OLED and particularly for blue OLED. The dual-gate BP FETs also can demonstrate NOR logic functions separately using top-input and bottom-input. The patterned dual gate BP FETs display promising perspectives for light-emitting pixel and logics [150]. The photographic images of blue OLED were displayed under turn-off state and turn-on state as shown in Figure 9C.

Graphene layer is deposited on SiO_2 substrates. In the mid infrared frequency range, graphene can act as a low-efficiency emitter with the electrical bias voltage. For electrically biased suspended graphene devices, the bright visible light emission was observed using false-color

Figure 9: Graphene plasmonic logic gate, blue OLED in BP FET and visible light emission in graphene [149–151].

(A) Basic switching unit of the graphene plasmonic MZI logic gate [149]. (B) Schematic 3D view of the dual-gate BP FET. (C) Photographic images display blue OLED under turn-off and -on states [150]. (D) The suspended monolayer graphene devices with bright visible light emission under electrically bias voltage. (E) ID-VSD relation in graphene. Solid curves are the calculated results with the transport model [151].

scanning electron microscopy image in Figure 9D. In these devices, heat transport in graphene is largely reduced because the hot electrons are mainly localized inside the graphene layer. A thermal radiation efficiency has reached 1000-fold enhancement in this structure [151]. The ID-VSD relation for suspended mechanically exfoliated monolayer graphene obtained after the measurement of visible light emission spectra is shown in Figure 9E. Solid curves are the calculated results using the transport. The spontaneous emission (SE) rate of a monolayer 2D material also can be modulated in a planar photonic crystal nanocavity [153].

A monolayer graphene sheet was synthesized by the CVD method on a copper foil. A native oxide layer was thermally grown on a silicon wafer. The graphene was transferred onto the native oxide layer. The drain electrode, source electrode, and plasmonic nonamer antenna was patterned onto the monolayer graphene sheet with EBL and Au evaporation in Figure 10A. The plasmonic nonamer antenna consists of eight gold disks surrounding a larger center disk [154]. A nonamer antenna is located on the graphene sheet with back-gated voltage. Under light irradiation, the hot electrons were generated in the gold nonamer antennas due to plasmon excitation. Then, the hot electrons were injected into the monolayer graphene sheets, which can induce n-type doping in graphene sheets. The scattering and absorption of the plasmonic nonamer antenna are shown in Figure 10B. An interesting

concept is the optically induced hot electronics. The distinct regions of an undoped graphene structure can be patterned using plasmonic antennas or quantum dots. Then, illuminated by light with a different (or the same) wavelength, it is possible to create a p-n junction and a simple electronic circuitry. The photons in visible and NIR wavelength region can be converted into electrons in graphene-antenna sandwich photodetector with 800% enhancement of the relative photocurrent [155]. The near field of the antenna enhanced the excitation of the intrinsic graphene electrons due to plasmon resonance. The photocurrent induced by the hot electrons was measured in the graphene-based vertical photodetector [156].

After the schematic illustration of optically induced hot electronics, a diode and an n-p-n transistor are possible using quantum dot p-doping and nanoantenna n-doping in Figure 10C and D, respectively. Some regions of the undoped graphene structures are covered by plasmonic antennas (tuned to red), while other regions are covered with quantum dots (tuned to blue). When a specific pattern of light with requisite wavelengths illuminates the structure, hot electrons from the plasmonic antennas induce n-doping. In addition, the holes from the quantum dots injected the graphene sheet, which induced p-doping in the graphene layer. In addition, the microsecond doped carrier relaxation time scale could enable the development of a wide variety of optically induced electronics, graphene photodetectors, and graphene switches.

The ultra thin BP nanosheets were prepared by liquid exfoliation of the bulk sample. Thereafter, the stacked flakes were dispersed in IPA liquid, as shown in Figure 11A. An annular pump beam was focused into the Er:YAG. The spiral-propagation symmetry and degeneracy of LG0, +1 and LG0, -1 modes were expected to be broken through introducing different Fresnel reflection losses. The

Figure 10: Phototransistor based plasmonic nonamer antenna and graphene hybrid structure [154]. (A) Nonamer antenna-graphene phototransistor. (B) FDTD calculated scattering (blue) and absorption (red) of a plasmonic nonamer. Schematic illustration of doped graphene sheet by n-doping and quantum dot p-doping, inducing (C) a diode or (D) an n-p-n transistor, respectively [154].

Figure 11: Vortex laser based on a black phosphorus plate [157]. (A) BP liquid sample. (B) Experimental setup used to excite the pulsed vortex laser and the intensity profile of the output. (C) The interference patterns with different tilt angles of BP [157].

uncoated polished side of BP is used with the discriminating Poynting vectors [157]. Using a Mach-Zehnder interferometer, the interference patterns are recorded in Figure 11B. Through optimizing the position and fine tuning the angle of the BP, the interference fringes could be stable at one handedness when the BP was tilted to 4° (or -3.5°), while this helicity selection mechanism was invalid, and the inference fringes turned into a disordered structure when the tilted angle was close to 0° (as shown in Figure 11C). Thus, we are able to confirm that BP can efficiently function as a helicity controller in this vortex laser [38].

A liquid exfoliation method was designed to produce phosphorene with a controllable size and layer number in large quantities [67]. The layer number of phosphorene can be determined with the experimental Raman scattering. The linear and nonlinear optical properties of the chemically exfoliated phosphorene were investigated by the Z-scan technique and UV-vis-NIR absorption spectrophotometry. The experimental synthesized ultrasmall BP quantum dots exhibited excellent nonlinear optical response with a modulation depth of 36% and a saturable intensity of 3.3 GW/cm^2 with the Z-scan measurement technique [158, 159]. BP quantum dots can act as an ultrafast nonlinear optical saturable absorber, which can be applied in the mode-locked fiber laser [160]. The saturable absorber BP films, fabricated by evaporating the solutions on glass wafers, were attached onto the end facet of a waveguide. A stable Q-switched ultrafast waveguide laser can operate under a pump laser at 810 nm [161]. Compared with traditional therapies, it is noted that BP analog tin sulfide nanosheets can serve as NIR photothermal agents and drug delivery platforms for cancer therapy [162]. Photothermal therapy has great application in cancer immunotherapy. In addition, the formulation of a biomimetic BP quantum dot is prepared to induce cancer cells by laser irradiation to eliminate the metastatic and residual cancer cells [163–165].

Graphene and BP have an important role in the field of tunable plasmonic nonlinear photonics [166, 167]. The advantage of graphene and BP plasmonic is the tunability of surface plasmons compared to the conventional metal. The linear and nonlinear susceptibility can also be controlled with Fermi level. The graphene nanostructure chips with electrically bias voltages G_1 and G_2 are designed in Figure 12A. In the graphene or BP chip, bright visible or THz light emission, THz detector, light modulation, switches, superstrate sensing, etc., can be realized in the 2D material-based tunable plasmonic nonlinear photonics nanoplatfroms. Figure 12B illustrates the various linear or nonlinear optical phenomenon and their application in graphene and BP-based nanoplatfrom.

Figure 12: Tunable graphene and BP nonlinear plasmonics photonics nanoplatfrom.

(A) The 2D material array chip applications including the bright visible or THz light emission, THz detector, light modulation, switches, photothermal therapy, biosensors, etc. (B) The illustration of tunable graphene and BP nonlinear plasmonics photonics nanoplatfrom with the various linear or nonlinear optical phenomenon and their applications.

The graphene and BP plasmonic can be achieved, which may launch a revolution in nonlinear photonics nanoplatfrom with the prospects for future related research such as linear and nonlinear photonics [47, 168–171], ultrafast photonics [172–174], metamaterials [175–177], photo-detector [178–189], saturable absorber [190], cancer therapy [191–197], photonic diodes [166], supercapacitor [198, 199], and transistor [200]. By designing the graphene and BP-based plasmonic tunable devices, they can be fabricated for novel THz active devices such as light modulation devices [201–204], switches [179, 205–208], and biosensors [209–211]. These impending future developments will be spurred not only by the possibility to discover new 2D materials but also by our increased ability to integrate these material-based tunable plasmonic in novel nonlinear photonics nanodevices [212].

5 Outlook

In this review, we presented the linear and nonlinear optical properties of graphene and BP-tunable plasmonic modes. We present the theoretical framework for calculating the lifetime of tunable plasmonic modes assisted by the coupled mode theory. The plasmonic conditions and lifetime of graphene and BP plasmonic modes can be modified by electrical and chemical doping, which has an advantage over that of conventional metals. The effect of the Fermi energy on the linear, second-order and third-order nonlinear response is presented in detail. In particular, we presented the linearity and nonlinearity of such 2D material plasmonic using theoretical

modeling, simulations, experimental methods, and nonlinear photonic applications. This is a rapidly growing field of science as it has been exciting linear and nonlinear optical phenomena and novel photonic nanodevice applications due to their large surface area-to-volume ratio and fluorescence characteristics. Especially, it also provides an exciting possibility to integrate these 2D materials with medical mark, medical diagnosis, and treatment through their excellent optical response and photothermal properties. There are still some problems that need to be solved. First of all, it is very important to find novel experimental methods to fabricate graphene and BP with uniform large sized, periodic nano-array patterns, great controllability, and high production. Second, the induced mechanism, physical model, and the test approach in graphene and BP nonlinear plasmonic should be established and investigated to achieve smaller side effects and quantum effects. The fabrication and synthesis techniques should enable the manipulation of graphene and BP nonlinear plasmonic down to the molecular and even atomic level. Finally, the plasmonic nanodevices and drug delivery platform should be combined with graphene and BP to achieve better optoelectronic characteristic and therapeutic effects.

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