Strain engineered linear dichroism and Faraday rotation in few-layer phosphorene

Cite as: Appl. Phys. Lett. **114**, 243102 (2019); https://doi.org/10.1063/1.5103172 Submitted: 24 April 2019 . Accepted: 02 June 2019 . Published Online: 17 June 2019

L. L. Li 匝, and F. M. Peeters 匝

ARTICLES YOU MAY BE INTERESTED IN

Topological valley currents in bilayer graphene/hexagonal boron nitride superlattices Applied Physics Letters **114**, 243105 (2019); https://doi.org/10.1063/1.5094456

Efficient broadband terahertz generation from organic crystal BNA using near infrared pump Applied Physics Letters **114**, 241101 (2019); https://doi.org/10.1063/1.5098855

Towards quantum phase slip based standard of electric current Applied Physics Letters **114**, 242601 (2019); https://doi.org/10.1063/1.5092271



Sensors, Controllers, Monitors from the world leader in cryogenic thermometry





Appl. Phys. Lett. **114**, 243102 (2019); https://doi.org/10.1063/1.5103172 © 2019 Author(s).

Strain engineered linear dichroism and Faraday rotation in few-layer phosphorene

Cite as: Appl. Phys. Lett. **114**, 243102 (2019); doi: 10.1063/1.5103172 Submitted: 24 April 2019 · Accepted: 2 June 2019 · Published Online: 17 June 2019



L. L. Li^{1,a)} (D) and F. M. Peeters^{1,2,b)} (D)

AFFILIATIONS

¹Department of Physics, University of Antwerp, Groenenborgerlaan 171, 2020 Antwerpen, Belgium ²School of Physics and Astronomy and Yunnan Key Laboratory for Quantum Information, Yunnan University, Kunming 650091, China

^{a)}Author to whom correspondence should be addressed: longlong.li@foxmail.com

^{b)}Electronic mail: francois.peeters@uantwerpen.be

ABSTRACT

We investigate theoretically the linear dichroism and the Faraday rotation of strained few-layer phosphorene, where strain is applied uniaxially along the armchair or zigzag direction of the phosphorene lattice. We calculate the optical conductivity tensor of uniaxially strained few-layer phosphorene by means of the Kubo formula within the tight-binding approach. We show that the linear dichroism and the Faraday rotation of few-layer phosphorene can be significantly modulated by the applied strain. The modulation depends strongly on both the magnitude and direction of strain and becomes more pronounced with increasing number of phosphorene layers. Our results are relevant for mechano-optoelectronic applications based on optical absorption and Hall effects in strained few-layer phosphorene.

Published under license by AIP Publishing. https://doi.org/10.1063/1.5103172

Phosphorene, a single layer of black phosphorus, has drawn a lot of attention from the research community since its first isolation in 2014.¹ This 2D material features the combined properties of direct bandgap, high carrier mobility, and strong in-plane anisotropy.^{2,3} Few-layer phosphorene is composed of two or more phosphorene monolayers that are vertically stacked and coupled via the van der Waals interaction. Compared to monolayer phosphorene, few-layer phosphorene exhibits a reduced bandgap due to interlayer electronic interaction. The bandgap decreases monotonically but remains direct with increasing number of phosphorene layers.^{4,5} These unique properties of monolayer and few-layer phosphorene make them promising for practical optoelectronic applications.

Recently, it was shown by using density-functional theory (DFT) calculations⁶ that phosphorene can sustain a tensile strain of up to 27% (30%) along its zigzag (armchair) direction without breaking the lattice stability, and that when compared to other 2D materials such as graphene, phosphorene exhibits superior mechanical flexibility with an order of magnitude smaller Young's modulus. These excellent mechanical properties of phosphorene make it promising for practical large-magnitude strain engineering. At present, various interesting properties induced by strain were theoretically and experimentally investigated for monolayer and few-layer phosphorene including, e.g., directional preference of charge transport,⁷ semiconductor-metal

phase transition,⁸ enhancement of room-temperature carrier mobility,⁹ and spatial modulation of quantized subbands.¹⁰

Although the optical properties (e.g., optical absorption) of fewlayer phosphorene were investigated previously,^{4,5} far less attention was paid to the effect of strain on linear dichroism and Faraday rotation. The linear dichroism refers to the property that light with two different polarizations are absorbed by different amounts, while Faraday rotation is the effect that the polarization of light can be rotated when transmitted through an optical medium in the presence of an external magnetic field. Faraday rotation is a direct consequence of the optical Hall effect. Previously, it was shown¹¹ that in few-layer phosphorene Faraday rotation is present even in the absence of an external magnetic field due to the reduced lattice symmetry, and that the linear dichroism and Faraday rotation of few-layer phosphorene can significantly be modulated by a perpendicular electric field. In this paper, we investigate theoretically the effect of strain on the linear dichroism and Faraday rotation of few-layer phosphorene, where strain is applied uniaxially along the armchair or zigzag direction of the phosphorene lattice. We show how these optical properties can be engineered by strain. Our investigation is important for mechanooptoelectronic devices based on strained few-layer phosphorene.

We consider *AB*-stacked few-layer phosphorene in the presence of an in-plane strain applied along the armchair or zigzag direction of the phosphorene lattice, as sketched in Fig. 1. It was shown experimentally¹² that such uniaxial strain can be produced uniformly. From DFT calculations,^{13,14} we know that AB stacking is energetically the most favorable for few-layer phosphorene. We employ the tight-binding approach to calculate the electronic band structure of uniaxially strained few-layer phosphorene. Once the electronic band structure is obtained, the optical conductivity tensor of few-layer phosphorene can be calculated by using the linear response Kubo formula, which is given by

$$\begin{aligned} \tau_{\alpha\beta}(\omega) &= \frac{e^2\hbar}{2\pi^2 i} \sum_{m,n} \int d\mathbf{k} \frac{f[E_m(\mathbf{k})] - f[E_n(\mathbf{k})]}{E_m(\mathbf{k}) - E_n(\mathbf{k})} \\ &\times \frac{\langle m \mathbf{k} | \mathbf{v}_{\alpha} | n \mathbf{k} \rangle \langle n \mathbf{k} | \mathbf{v}_{\beta} | m \mathbf{k} \rangle}{E_m(\mathbf{k}) - E_n(\mathbf{k}) + \hbar \omega + i\eta}, \end{aligned}$$
(1)

where $\alpha = x$, *y* is the tensor index, ω is the optical frequency, *m* is the band index, $\mathbf{k} = (k_x, k_y)$ is the 2D wave vector, f(E) is the Fermi-Dirac function, $E_m(\mathbf{k})$ is the eigenenergy, $|m\mathbf{k}\rangle$ is the eigenstate, \mathbf{v}_{α} is the α component of the velocity operator $\mathbf{v} = \hbar^{-1} \partial H / \partial \mathbf{k}$, and η is a finite broadening. The details of our theoretical approach are presented in the supplementary material. There are four in-plane components of the optical conductivity tensor: σ_{xx} and σ_{yy} for the longitudinal optical conductivities and σ_{xy} and σ_{yx} for the transverse optical conductivities (i.e., optical Hall conductivities). For the longitudinal components, we have $\sigma_{xx} = \sigma_{yy} (\sigma_{xx} \neq \sigma_{yy})$ for an isotropic (anisotropic) system. When calculating the optical conductivities of few-layer phosphorene, we took an energy broadening of 10 meV and the lattice temperature of 300 K (unless otherwise specified) for free-standing samples. The real parts of the longitudinal and transverse optical conductivities are related to the optical absorption effect and the optical Hall effect (e.g., Faraday rotation), respectively.

Because we consider undoped few-layer phosphorene in the absence and presence of strain, there are no free charge carriers



FIG. 1. (a) Sketch of strained few-layer phosphorene, excited by normal incident light with intensity (*I*) and polarization (λ), where strain (ϵ) is applied uniaxially along the in-plane direction (e.g., the armchair or zigzag direction of the phosphorene lattice). The two black rectangles show the top and side views of the phosphorene lattice. When light transmits through the sample, its intensity is reduced due to optical absorption and its polarization is rotated due to the optical Hall effect.

(electrons and holes) in the system, and so the Fermi energy of the system is located within the bandgap region of the system. In this situation, when the system is excited by normal incident light with certain polarization (as sketched in Fig. 1), only interband transitions from the occupied valence bands (VB) to the unoccupied conduction bands (CB) contribute to the optical conductivity of the system. In order to show the effect of strain more clearly, we consider a reasonably large strain (up to 10%) applied to few-layer phosphorene. The same order of magnitude of strain was demonstrated both theoretically⁶ and experimentally.¹²

In Figs. 2(a) and 2(b), we show the longitudinal optical conductivities (σ_{xx} and σ_{yy}) of strained bilayer phosphorene for different strain magnitudes and directions, with strain applied along the armchair (ϵ_{ac}) or zigzag (ϵ_{zz}) direction. Here, we define armchair (zigzag) directions along the *x* (*y*) direction and $\epsilon_{ac/zz} < 0$ ($\epsilon_{ac/zz} > 0$) as compressive (tensile) armchair/zigzag strain, and we use the band index pair (*m*, *n*) to denote the interband transition from the *m*th valence



FIG. 2. Effect of strain on the longitudinal optical conductivities (a) σ_{xx} and (b) σ_{yy} , on the band structure (c), on the optical Hall conductivity σ_{xy} (d), and on the Faraday rotation angles θ_F (e) and (f) of bilayer phosphorene. Here, $\sigma_0 = e^2/(4\hbar)$ is the universal optical conductivity, the strain is applied along the armchair (ϵ_{ac}) or zigzag (ϵ_{zz}) direction, the black curves show the unstrained results, and the band index pair (*m*, *n*) denotes the interband transition from the *m*th valence band (VB) to the *n*th conduction band (CB).

band (VB) to the *n*th conduction band (CB). Moreover, we took the magnitude of -10% (10%) for the compressive (tensile) strain.

As can be seen, in the absence of strain, there is a significant difference between σ_{xx} and σ_{yy} with the former being about 10 times larger than the latter, leading to the so-called "linear dichroism" that is typical for phosphorene systems. Applying a uniaxial strain (ϵ_{ac} or ϵ_{zz}) modulates significantly σ_{xx} and σ_{yy} . The energy cutoffs of these optical conductivities are red (blue)-shifted in the presence of compressive (tensile) strain along both the armchair and zigzag directions. The remarkable shift of the energy cutoff between the compressive and tensile strain was also confirmed by a recent experiment.¹⁰ Intuitively, we attribute this strain-induced shift in the energy cutoffs of σ_{xx} and σ_{yy} to the electronic bandgap change induced by the applied strain. In order to gain better insights, we have to look into the effect of strain on the band structure of bilayer phosphorene, as shown in Fig. 2(c). It is clear from the band structure that the bandgap increases (decreases) in the presence of tensile (compressive) strain for both the armchair and zigzag directions. More importantly, the intensities of σ_{xx} and σ_{yy} become enlarged (reduced) in the presence of compressive (tensile) strain. Furthermore, only optical transitions between the CBs and VBs with the same band indices are allowed, i.e., (m, n) = (1, 1) and (m, n)= (2, 2), irrespective of strain. In order to understand these features, we need to look into the interband momentum matrix elements for the x- and y-polarizations, i.e., $|M_{mn}^{xx}(\mathbf{k})|^2$ and $|M_{mn}^{yy}(\mathbf{k})|^2$, which are presented and discussed in supplementary material.

In Fig. 2(d), we show the optical Hall conductivity (σ_{xy}) of strained bilayer phosphorene for different strain magnitudes and directions. As can be seen, in the absence of strain, σ_{xy} is nonzero; however, it is about 100 (10) times smaller than σ_{xx} (σ_{yy}), which agrees with the results obtained from DFT calculations for other 2D materials such as GaS and GaSe multilayers.¹⁵ This small but nonzero optical Hall conductivity implies that Faraday rotation is present in bilayer phosphorene "even under zero magnetic field." This is due to the reduced symmetry of the phosphorene lattice (i.e., the puckered structure). According to the relation $\theta_F \simeq \sigma_{xy}/[(1 + n_{sub})c\epsilon_0]$, with θ_F being the Faraday rotation angle and n_{sub} the refractive index of the substrate,¹⁶ the frequency dependence of σ_{xy} indicates that the polarization of light can be rotated by simply varying the frequency of light transmitted through bilayer phosphorene. Now, we look at the effect of strain on the optical Hall conductivity. As can be seen, the intensity of σ_{xy} is almost unaffected by the compressive or tensile "zigzag" strain. However, it becomes enlarged by an order of magnitude in the presence of compressive or tensile "armchair" strain. Again, in order to understand these features, we need to look into the interband momentum matrix elements $|M_{mn}^{xy}(\mathbf{k})|^2$, which are presented and discussed in supplementary material. In Figs. 2(e) and 2(f), we show the Faraday rotation angles in units of degree/micrometer, corresponding to the optical Hall conductivities for the armchair and zigzag strain, as shown in Fig. 2(d). When calculating the Faraday rotation angle θ_F , we took $n_{sub} = 1$ throughout the paper, which models a free-standing sample. Recently, by using DFT calculations, magneto-optical effects (e.g., Faraday rotation) were predicted for the ferromagnetic monolayer $Cr_2Ge_2Te_6$.¹⁷ We notice that the rotation angle θ_F is smaller in both armchair- and zigzag-strained phosphorene ($\sim 10^{\circ}/\mu m$ and $\sim 1^{\circ}/\mu m$ μ m) than in the ferromagnetic monolayer of Cr₂Ge₂Te₆ (~120°/ μ m). The difference is mainly due to (i) different band properties of monolayer phosphorene and monolayer Cr2Ge2Te6 and (ii) different driving mechanisms: uniaxial strain (in phosphorene) vs local magnetic moment (in monolayer $Cr_2Ge_2Te_6$).

In addition, we see from Figs. 2(a)-2(f) that the band structure, the optical conductivity, and the Faraday rotation exhibit distinctively different responses to different strain directions (i.e., armchair and zigzag directions). This is a consequence of the structural anisotropy of the phosphorene lattice. Therefore, strain engineering in phosphorene might be even more interesting than in other 2D materials. Our numerical results indicate that the tuning of the Faraday rotation becomes less significant with decreasing magnitude of the applied strain, e.g., the Faraday rotation angle with 1% strain is smaller than that with 10% strain.

The mechano-optical modulation can be used to measure the change in the optical coefficients (such as optical absorption, reflection, and transmission) of a material due to the application of mechanical strain. For thin samples, a higher precision of such modulation can be obtained by performing optical transmission measurements. The reflection and transmission coefficients of polarized light through a thin sample can be obtained by solving the Maxwell equations with appropriate boundary conditions for the electromagnetic fields.¹⁸ For the case of normal incidence, the optical transmission coefficient is given by¹⁸

$$T(\omega) = \sqrt{\frac{\varepsilon_2}{\varepsilon_1}} \frac{4(\varepsilon_1 \varepsilon_0)^2}{\left|\left(\sqrt{\varepsilon_1 \varepsilon_2} + \epsilon_1\right)\varepsilon_0 + \sqrt{\varepsilon_1} Re\sigma(\omega)/c\right|^2},$$
 (2)

where ε_0 is the permittivity of vacuum, ϵ_1 and ϵ_2 are the relative permittivities of media on the top and bottom of the sample, and *c* is the speed of light in vacuum. In this paper, we took $\varepsilon_1 = \varepsilon_2 = 1$ unless otherwise specified, which models a free-standing sample. The strain modulation of the optical transmission is defined as $\Delta T/T = [T_{\epsilon}(\omega) - T_0(\omega)]/T_0(\omega)$, with the subscripts ϵ and 0 indicating the presence and absence of strain, respectively.

In Figs. 3(a) and 3(b), we show the strain modulation $\Delta T/T$ of the optical transmission of *N*-layer phosphorene (N = 1, 2, 3) for the *x* polarization, and in Figs. 3(c) and 3(d), we show the layer dependence of the strained optical Hall conductivity σ_{xy} . Here, the full/dashed curves in the top (bottom) panels represent the results of the compressive (tensile) strain applied along the armchair/zigzag directions, and the magnitude of the applied compressive (tensile) strain is fixed at -10% (10%). As can be seen, the modulation of the optical transmission and the optical Hall conductivity (or the Faraday rotation) depends strongly on both the magnitude and direction of strain and on the number of phosphorene layers. The modulation of the optical transmission, $\Delta T/T$, oscillates as a function of the photon energy. For the optical Hall conductivity (or the Faraday rotation), the modulation strength is always larger for the armchair strain than for the zigzag strain. Our numerical results also indicate that for the optical transmission, the modulation strength is much larger for the x-polarization than for the *y*-polarization.

In summary, we have investigated the linear dichroism and the Faraday rotation of strained few-layer phosphorene by using the Kubo formula within the tight-binding approach, where strain is applied uniaxially along the armchair or zigzag direction of the phosphorene lattice. We calculated the electronic band structure and the optical conductivity tensor of uniaxially strained few-layer phosphorene. We showed that linear dichroism and Faraday rotation of few-layer



FIG. 3. Strain modulation $\Delta T/T$ of the optical transmission of *N*-layer phosphorene (N = 1, 2, 3) for the *x*-polarization [(a) and (b)] and layer dependence of the strained optical Hall conductivity σ_{xy} [(c) and (d)]. Here, the full/dashed curves in the top (bottom) panels represent the results of the compressive (tensile) strain along the armchair/zigzag directions, and the magnitude of the compressive (tensile) strain magnitude is fixed at -10% (10%).

phosphorene can significantly be modulated by the applied strain. The modulation depends strongly on both the magnitude and direction of strain and on the number of phosphorene layers. Our results are related to strain tunable optical absorption and Hall effects in fewlayer phosphorene and can provide fundamental insights into potential applications of strained few-layer phosphorene in mechanooptoelectronic devices.

See supplementary material for (i) more details of theoretical approach and (ii) the numerical results of momentum matrix elements.

This work was financially supported by the Flemish Science Foundation (FWO-VI) and by the FLAG-ERA Project TRANS-2D-TMD.

REFERENCES

- ¹H. Liu, A. T. Neal, Z. Zhu, Z. Luo, X. Xu, D. Tománek, and P. D. Ye, ACS Nano 8, 4033 (2014).
- ²L. Kou, C. Chen, and S. C. Smith, J. Phys. Chem. Lett. **6**, 2794 (2015).
- ³A. Carvalho, M. Wang, X. Zhu, A. S. Rodin, H. Su, and A. H. C. Neto, Nat. Rev. Mater. 1, 16061 (2016).
- ⁴J. Qiao, X. Kong, Z.-X. Hu, F. Yang, and W. Ji, Nat. Commun. 5, 4475 (2014).
- ⁵V. Tran, R. Soklaski, Y. Liang, and L. Yang, Phys. Rev. B **89**, 235319 (2014).
- ⁶Q. Wei and X. Peng, Appl. Phys. Lett. **104**, 251915 (2014).
- ⁷R. Fei and L. Yang, Nano Lett. 14, 2884 (2014).
- ⁸A. Manjanath, A. Samanta, T. Pandey, and A. K. Singh, Nanotechnology 26, 075701 (2015).
- ⁹H. Morgan Stewart, S. A. Shevlin, C. R. A. Catlow, and Z. X. Guo, Nano Lett. 15, 2006 (2015).
- ¹⁰ J. Quereda, P. San-Jose, V. Parente, L. Vaquero-Garzon, A. J. Molina-Mendoza, N. Agraït, G. Rubio-Bollinger, F. Guinea, R. Roldán, and A. Castellanos-Gomez, Nano Lett. **16**, 2931 (2016).
- ¹¹L. L. Li, B. Partoens, W. Xu, and F. M. Peeters, 2D Mater. 6, 015032 (2019).
- ¹²R. Roldán, A. Castellanos-Gomez, E. Cappelluti, and F. Guinea, J. Phys.: Condens. Matter 27, 313201 (2015).
- ¹³D. Çakir, C. Sevik, and F. M. Peeters, Phys. Rev. B **92**, 165406 (2015).
- ¹⁴A. N. Rudenko and M. I. Katsnelson, Phys. Rev. B **89**, 201408 (2014).
- ¹⁵F. Li, X. Zhou, W. Feng, B. Fu, and Y. Yao, New J. Phys. **20**, 043048 (2018).
- ¹⁶Y. Ikebe, T. Morimoto, R. Masutomi, T. Okamoto, H. Aoki, and R. Shimano, Phys. Rev. Lett. **104**, 256802 (2010).
- ¹⁷Y. Fang, S. Wu, Z.-Z. Zhu, and G.-Y. Guo, Phys. Rev. B 98, 125416 (2018).
- ¹⁸T. Stauber, N. M. R. Peres, and A. K. Geim, Phys. Rev. B 78, 085432 (2008).