Tuning the electrical and optical anisotropy of a monolayer black phosphorus magnetic superlattice

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Abstract
We investigate theoretically the effects of modulated periodic perpendicular magnetic fields on the electronic states and optical absorption spectrum in monolayer black phosphorus (phosphorene). We demonstrate that different phosphorene magnetic superlattice (PMS) orientations can give rise to distinct energy spectra, i.e. tuning the intrinsic electronic anisotropy. Rashba spin–orbit coupling (RSOC) develops a spin-splitting energy dispersion in this phosphorene magnetic superlattice. Anisotropic momentum-dependent carrier distributions along/perpendicular to the magnetic strips are demonstrated. The manipulations of these exotic electronic properties by tuning superlattice geometry, magnetic field and the RSOC term are addressed systematically. Accordingly, we find bright-to-dark transitions in the ground-state electron–hole pair transition rate spectrum and the PMS orientation-dependent anisotropic optical absorption spectrum. This feature offers us a practical way of modulating the electronic anisotropy in phosphorene by magnetic superlattice configurations and detecting this modulation capability by using an optical technique.

Keywords: phosphorene, magnetic superlattice, optical absorption spectrum, anisotropy modulation

1. Introduction
Two-dimensional (2D) semiconductor materials have a unique layer structure, in which each layer is vertically stacked by van der Waals forces. As a result, the crystal can be scaled down to atomic layer scale with significant changes in the physical properties. The changeable band gap and high mobility in some 2D materials offer exciting opportunities for development of high performance electronic and optical devices. To develop these fascinating applications based on their unique electronic and optical properties, a scheme of superlattices to manipulate the electronic states and charge flows in a 2D material based nanostructure has attracted increasing research interest. Since the work by Esaki and Tsu [1], a great deal of attention has been devoted to superlattice graphene, where external spatially periodic electric [2–8] and/or magnetic fields are applied to a graphene monolayer [9–13]. In these previous studies, people have demonstrated effective band engineering and optical modulation using a real superlattice structure as well as periodic external fields.

Recently, black phosphorus (BP), a rare allotrope of phosphorus, has become a new type of 2D material. The high electronic mobility has already been confirmed in
few-layer BP and applied to the applications of field-effect transistors [14]. The layer structure of BP held together by van der Waals forces can be exfoliated from a few layers to monolayer (phosphorene), with a layer-dependent direct band gap from 0.3 eV [15] (bulk) to 1.52 eV [16] (monolayer), leading to potential applications in optoelectronics, especially in the infrared regime. Interestingly, the concept of utilizing the strong anisotropic properties of 2D materials for novel optoelectronic and electronic device applications has been proposed. Strongly anisotropic conducting behavior [17–21], anisotropic exciton [22, 23] or optical response [24–26], anisotropic Landau levels [27], anisotropic Rashba spin–orbit coupling [28], as well as anisotropic structural flexibility [29], have been observed. Inspired by the superlattice structure where external spatially periodic electric and/or magnetic fields are applied to a graphene monolayer, people have paid attention to superlattice BP with varied external periodic potentials and the dependence of BP anisotropy on the periodic perturbation. The transmission probability of the wave packets with normal incidence can be tuned and controlled [30]. An analytical model is presented to relate the decrease in the direct band gap to the different orbital characters between the valence conduction band [31]. In addition to electric control, the effects of magnetic fields on BP’s electronic and optical properties have also been studied [32–36]. However, periodic magnetic modulations, especially for experimental measurable quantities such as conductance, optical absorption spectrum, have not been investigated thoroughly until now.

In this work, we propose a monolayer BP (or phosphorene) based phosphorene magnetic superlattice (PMS), which is an ideal system for archiving an anisotropic two-dimensional electron system with high flexibility and controllability. We show that the anisotropic energy dispersions of the phosphorene can be effectively tuned by different magnetic stripe configurations, the strength of the magnetic fields as well as Rashba spin–orbit coupling (RSOC) interactions. Accordingly we illustrate the impact of such a periodic magnetic field on the anisotropic transport properties, including the effective mass. We moreover demonstrate that the corresponding energy states of e–h pairs and optical transition rates can also be controlled by the periodic magnetic stripes. In addition to the crystalline-induced anisotropy along the two real-space orthogonal axes, the magnetic fields break the time-reversal symmetry and develop an in-line anisotropy along the reciprocal-space axes, e.g., the bright-to-dark transition in optical transition rate of e–h pairs, which also can be tuned by different magnetic stripe configurations and the RSOC interactions.

2. Theoretical model

We consider phosphorene (BP monolayer) coated by periodic magnetic stripes along the plane direction, as shown in figure 1. The magnetic field is applied perpendicular to the layer. The low-energy dispersion of bulk BP can be well described by a two-band \( \hbar \cdot \vec{p} \) effective mass Hamiltonian due to the \( D_{2h} \) point group invariance [37],

\[
H = H_0(p + eA) + H_R + H_Z,
\]

where \( H_0 \), \( H_R \) and \( H_Z \) are the two-band effective Hamiltonian, the RSOC term and the Zeeman term, respectively. The first term is given by [38–40],

\[
H_0 = \begin{bmatrix}
E_v + \alpha_v k_x^2 + \beta_v k_y^2 & \gamma k_x \\
\gamma k_x & E_v + \alpha_v k_x^2 + \beta_v k_y^2
\end{bmatrix}.
\]

The band parameters are \( \alpha_v = \hbar^2/2m_{xv}, \beta_v = \hbar^2/2m_{yv}, \alpha_e = \hbar^2/2m_{xv}, \beta_e = \hbar^2/2m_{yv}, m_{xv} = 0.793m_e, m_{yv} = 0.848m_e, m_{xv} = 1.363m_e, m_{yv} = 1.142m_e \). The x-direction periodic PMS induces periodic magnetic fields of pointing up and down sequentially. Accordingly the vector potential \( A \) in each cell is given by,

\[
B = \begin{cases}
(0, 0, B); & x \in \left[ -\frac{W}{2}, 0 \right] \\
(0, 0, -B); & x \in \left[ 0, \frac{W}{2} \right] \\
(0, 0, 0); & x \notin \left[ -\frac{W}{2}, \frac{W}{2} \right]
\end{cases}
\]

\[
A = \begin{cases}
(0, Bx + \frac{WB}{2}, 0); & x \in \left[ -\frac{W}{2}, 0 \right] \\
(0, -Bx + \frac{WB}{2}, 0); & x \in \left[ 0, \frac{W}{2} \right] \\
(0, 0, 0); & x \notin \left[ -\frac{W}{2}, \frac{W}{2} \right]
\end{cases}
\]

in which \( W \) is the width of each magnetic stripe and \( L \) is the periodic length of the PMS. The similar form for y-direction periodic PMS is given by,
The wave function can be calculated numerically in the basis set with the periodic boundary conditions in the crystal. The wave basis set. To make quantitative assessment of the anisotropic RSOC interaction, we discuss below in detail. First we plot the energy dispersions for electrons in phosphorene monolayer without any external magnetic field or with periodic magnetic field modulations along the x direction (armchair direction) and the y direction (zigzag direction), respectively. As sketched in figure 1, we find strongly PMS dependence on energy band gap enlargement, cone shift and dispersion modulation, which we will discuss below in detail. First we plot the energy dispersions of pristine phosphorene in figures 2(a) and (b). Note that the different slopes of the energy dispersions along the two orthogonal k_x and k_y directions indicate the anisotropic group velocities and effective masses in phosphorene (see figure 2(a)). This unique feature is also absent in graphene. When the RSOC interaction is incorporated into the calculations, we find the expected spin splitting in the conduction and valence bands, as shown by the dashed lines in figure 2(b). The spin-splitting energy is proportional to the wave vector. Its impact on the band structure of phosphorene is very small near the Γ point of the order of 0.0109 eVÅ, and the photon within the electrical dipole approximation is wE = |(E_f − E_i − iω)| |<f|H_int|i| > |^2. (9)
In which <f|H_int|i > =∑_{m,n}C^*_f,α,mC_i,α,nC_m,φ_{α,m}. Finally we can obtain the optical absorption rate by the integral of transition rates in k space.
Now the electron−light interaction induces transition from |i> to |f>, and |i> and |f> are written as Ψ_e(k_x, k_y). The resulting optical transition rate of e−h pairs between the valence and conduction bands is |<f|H_int|i| > |. The transition rate is given by.

\[ w_{if} = 2\pi\delta(E_f - E_i - i\omega)|<f|H_int|i| > |^2. \]

where \( R_{xx}, R_{yy}, R_{xy} \) are 0.0109 eVÅ and R_y = 0.0036 eVÅ [28]. The last term \( H_z = g_{\mu B}g_B \) represents the familiar Zeeman effect, and therefore our full Hamiltonian is \( H = H_0 + H_R + H_Z \).

The envelope functions may be combined into a four-component spinor \( \Psi = (\Psi_\uparrow, \Psi_\downarrow, \Psi_\uparrow, \Psi_\downarrow) \), which satisfies a Dirac equation \( H \Psi = E \Psi \). The electron wave functions are expanded in a plane wave basis confined by the large hard wall box. The wave function \( \Psi \) for electrons can be expanded as

\[
\Psi(k_x, k_y) = \sum_n C_n \phi_n(k_x, k_y) = \sum_n C_n \frac{1}{\sqrt{\Lambda}} e^{i(k_{x}x + k_{y}y)},
\]

where \( k_x (k_y) \) is the wave vector in the x (y) direction, and the expansion coefficient \( C_n \) is a four-component column vector. The wave function can be calculated numerically in the basis set with the periodic boundary conditions in the x (y) direction. Then the band structure can be obtained with this plane wave basis set. To make quantitative assessment of the modulated anisotropy in the PMS, we define an anisotropy index \( R_{xy} \equiv m_{\text{ax}}/m_{\text{ay}} \), in which the effective mass tensors are directly extracted from the energy dispersions.

The interaction Hamiltonian between the Dirac fermion and the photon within the electrical dipole approximation is \( H_{int} = H(p + e\bar{A}) - H(p) \), where the vector potential \( \bar{A} = (A_z + iA_y)e^{-i\omega t} \), corresponds to the \( \sigma^z \) circularly polarized light. |i> denotes the initial states in the lower cones that are hole or valence like, |f> denotes the final states in the upper cone states that are electron or conduction like. Now we include only the most important anisotropic RSOC term, which is linear in momentum, as given by [28].

In pristine phosphorene parabolic energy bands reflect the existence of a massive 2DEG with a direct band gap about 1.52 eV at the Γ point, being different from their counterparts in massless Dirac fermions in another well known as 2D material graphene of hexagonal symmetry. The electron−hole symmetry is broken owing to the interband coupling. The conduction band valence band coupling effects influence the band structure effectively and lead to distinct electronic and optical properties rather than being observed in graphene. We start by investigating the energy dispersion relationships of a pristine phosphorene monolayer without any external magnetic field or with periodic magnetic field modulations along the x direction (armchair direction) and the y direction (zigzag direction), respectively. As sketched in figure 1, we find strongly PMS direction dependent energy band gap enhancement, cone shift and dispersion modulation, which we will discuss below in detail. First we plot the energy dispersions of pristine phosphorene in figures 2(a) and (b). Note that the different slopes of the energy dispersions along the two orthogonal k_x and k_y directions indicate the anisotropic group velocities and effective masses in phosphorene (see figure 2(a)). This unique feature is also absent in graphene. When the RSOC interaction is incorporated into the calculations, we find the expected spin splitting in the conduction and valence bands, as shown by the dashed lines in figure 2(b). The spin-splitting energy is proportional to the wave vector. Its impact on the band structure of phosphorene is very small near the Γ point of the order of 0.0109 eVÅ, and the photon within the electrical dipole approximation is wE = |(E_f − E_i − iω)| |<f|H_int|i| > |^2. (9)
In which <f|H_int|i > =∑_{m,n}C^*_f,α,mC_i,α,nC_m,φ_{α,m}. Finally we can obtain the optical absorption rate by the integral of transition rates in k space,
magnetic phosphorene superlattices, however, are different in that the band gap is increased due to the external magnetic fields. The slopes of the conduction and valence energy bands decrease with the existence of a magnetic field. Along the periodic direction, a small gap is opened at the boundary of each superlattice Brillouin zone, e.g. $k_y (k_0) = \pm \pi / L + 2n\pi / L$, due to the perturbation potentials coming from the periodic magnetic fields. In the transverse direction, the energy spectrum $E[k_y(k_f)]$ becomes slightly asymmetric with respect to $k_x = 0$, which is absent in a phosphorene superlattice with electric potentials. It comes from the vector potentials accounting for time-reversal symmetry breaking by the magnetic fields, and therefore becomes more distinct as we increase the magnitude or the active area of the magnetic fields. For a preliminary summary, the impacts of the PMS with different configurations on the energy dispersion relationships are quite distinct. The energy spectrum in the $y$-direction PMS (corresponding to the transport direction along the zigzag direction) is much more sensitive to the external periodic magnetic fields than in its counterpart $x$-direction PMS.
Then we focus on the energy dispersion and anisotropy modulation in the y-direction PMS, since its interband coupling term gives rise to a more pronounced response to the periodic vector potential perturbations than that in the x-direction PMS. Heuristically, the magnetic fields tend to bend the carriers away from motion direction and thus suppress the group velocities of charge carriers as well as reduce energy dispersion. To get a quantitative assessment of the modulation effects on the electronic properties from the superlattice structure and magnetic field strength, we then plot the energy dispersions of the y-direction PMS along \( k_x \) and \( k_y \) with different periodic lengths and magnetic fields in figures 3 and 4. As the magnetic strip period increases, the aforementioned bi-directional anisotropy along the transversal direction (see figure 2(e)) becomes increasingly spectacular and develops two-band minima adjacent to the \( \Gamma \) point, as shown in figure 3(a). We note en passant that it is magnetic field induced in-line anisotropy [42], rather than the intrinsic in-plane anisotropy in pristine phosphorene. Along the periodic direction, more band gaps are opened arising from the periodic perturbation effect at a reduced Brillouin zone boundary. When the periodic length \( L \) is very large, the first subband is rather flat along the periodic direction, indicating the presence of heavy quasi particles in such a y-direction PMS, while the energy dispersion along the transversal direction almost remains. Therefore the effective mass and anisotropy can be effectively tuned by the proposed PMS, as shown in figures 3(c) and (d). Alternatively we can observe similar effects by increasing the strength of the magnetic fields. In addition band gap enlargement is more pronounced at the \( \Gamma \) point and at superlattice Brillouin zone boundaries. This trend is in contrast to the periodic electrical potential modulated phosphorene [30, 31].

So, the proposed PMS can effectively tune the anisotropy of energy dispersion and effective mass, since the energy dispersion along the transversal direction of the superlattice is almost immune to magnetic fields while the one along the longitudinal direction of the superlattice is effectively modulated. In addition to the unique anisotropy between the highly anisotropic band structures along the two crystalline directions of phosphorene, in-line bi-directional anisotropy is induced by the magnetic fields. Importantly, here we address that the modulation capabilities of different PMS periodic orientations are also highly anisotropic, as we have discussed for figure 2. For the configuration of the y-direction (zigzag...
direction) PMS, band structures more sensitively depend on the magnetic field strength $B$ and periodic cell size $L$.

Next, we turn our focus to the carrier probability distributions in a unit cell of the PMS with different configurations. The projected charge distribution of the electrons in the first conduction band with certain $k_x$ and $k_y$ are shown in figures 5 and 6. With the wave vector along the magnetic stripe (perpendicular to the periodic direction of the superlattice), the probability distributions exhibit anisotropic dependence on the wave vector, as shown by solid lines in figures 5(a) and (c) for the $x$-direction periodic and $y$-direction periodic superlattices, respectively. For a $x$-direction (or $y$-direction) PMS, the electrons from the first conduction band with positive $k_y$ (or $k_x$) tend to locate near the edges of a cell where no magnetic stripe is coated above, while for negative $k_y$ (or $k_x$), the electrons tend to locate in the center. This behavior directly arises from the Hall effect in the proposed PMS. Note that the spatial separation of electrons moving in the opposite direction is more pronounced with periodic magnetic modulations along the $y$ direction, as shown in figure 5(c). This feature is unique in phosphorene, since the effective vector potential $(\mathbf{A}_e)$ has distinct effects on the off-diagonal elements in Hamiltonian equation (2), while this impact is absent in the $x$-direction PMS. Along the superlattice direction, the probability distribution is isotropic for positive or negative wave vectors regardless of the $x$-direction or $y$-direction superlattice configuration (see figures 5(b) and (d)). The electrons tend to distribute in the central region of a cell underneath the magnetic stripe, since the perpendicular magnetic fields develop circular electron orbits and prevent them transmitting away. By incorporating the Zeeman term, we find that it has negligible impact on the energy spectrum but changes the probability distribution apparently, as shown by dashed lines in figure 5. In the plane wave basis, the diagonal matrix elements of the Zeeman term are canceled out and the off-diagonal elements are imaginary numbers with the given periodic magnetic field profile. Therefore the Zeeman term can hardly affect the eigenvalues but changes the eigenvector effectively. The RSOC term also hardly affects the density distributions with a wave vector along the direction of the magnetic stripes, as shown in figures 6(a) and (c). However, it tends to push the electrons with a wave vector along the superlattice direction out of the phosphorene plane, resulting in squeezed distributions, as shown in figures 6(b) and (d).

The anisotropy energy spectra and charge distributions are hard to measure directly. However, they apparently can affect the photoluminescence spectra. We therefore investigate the effect of the PMS on the energy to form electron–hole pairs by $\sigma+$ circularly polarized light and the transition rate. Figures 7 and 8 plot them as functions of the wave vector $k_x$ with $B = 1$ T and $R_0 = 0$ around the band bottom in an $x$-direction PMS. (b) The optical transition rate of $e$–$h$ pairs as a function of wave vector $k_x$. (c) and (d) are the same as (a) and (b) but including the RSOC term. (e)–(h) are the corresponding energy dispersions and the optical transition rates as a function of the wave vector $k_x$. The energy dispersions of $e$–$h$ pairs as a function of wave vector $k_x$ with $B = 1$ T and $R_0 = 0$ around the band bottom in an $x$-direction PMS. The optical transition rate of $e$–$h$ pairs as a function of wave vector $k_x$. (c) and (d) are the same as (a) and (b) but including the RSOC term. (e)–(h) are the corresponding energy dispersions and the optical transition rates as a function of the wave vector $k_x$.
to locate along the edge of a superlattice cell where no magnetic stripe is deposited above, regardless of positive or negative $k_y$. This results in a small overlap integral of the wave functions and a small transition rate in figure 7(b). When $k_y$ approaches zero, the charge carriers tend to distribute equally in a superlattice and thus increase the overlap integral of the wave functions. Then the optical transition rate is enhanced. Next we examine the effect of the RSOC term; one can see the expected spin splitting in the e–h pair energy spectrum (inset in figure 7(c)). Accordingly the RSOC term can affect the electron distribution at the superlattice Brillouin zone boundaries (figure 6(c)) and reduce the transition rate spectrum of the low-energy e–h pairs, so we can find a larger transition gap, as shown in figure 7(d). We also plot the e–h pair energy as a function of $k_y$ in figure 7(e), the asymmetrical behavior in the e–h pair energy spectrum is due to the anisotropic energy spectrum, as shown in figure 2. The optical transition rate spectrum along $k_x$ ($k_y$) exhibits in-line asymmetrical behavior with a dark-to-bright transition when $k_v$ varies from negative to positive. The electrons with positive $k_v$ tend to be located aside the superlattice cell while the ones with negative $k_v$ tend to be located in the center, as shown in figures 6(a) and (b). Different distributions result in different overlap integrals of wave functions and thus determine the optical transition rates as we observed. When the RSOC term is considered, we can find the spin splitting in the e–h pair energy and optical transition rate spectrum in figures 7(g) and (h). The optical transition rate is enhanced the same as that in figure 7(d) for the same reason.

Compared to the x-direction periodic PMS, the modulation effects by a y-direction PMS are much more pronounced. The ground-state e–h pair energy as a function of $k_y$ is asymmetrical due to time-reversal symmetry breaking in the presence of a magnetic field, as shown in figure 8(a) and spin splitting due to the RSOC interaction in figure 8(c). Because the electron with negative or positive $k_y$ shows separate distributions, as shown in figures 5(c) and 6(c), we can find the dark-to-bright transition of the ground-state e–h pair transition rate spectrum in figure 8(b). The RSOC term has little effect on the transition rate spectrum in $k_y$ accounting for its limited impact on the electron distribution (see figure 6(b)). The corresponding e–h pair energy spectrum in $k_y$ is symmetrical, as shown in figure 8(e). The e–h pair energy spectrum and optical transition rate spectrum of the y-direction periodic superlattice possesses larger gaps at the superlattice Brillouin zone boundaries, as shown figure 8, due to the enhanced interband coupling by the vector potential of the PMS. The optical transition rate spectrum of the ground e–h pairs is not bright (see figure 8(f)) due to the marginal distribution, as shown in figure 6(d), regardless of whether $k_y$ is negative or positive, although the spin splitting is shown in the energy spectrum but cannot be distinguished in the transition rate spectrum, as shown in figures 8(g) and (h). In brief summary, the optical transition rate of the y-direction PMS is different from that of the x-direction PMS, as we discussed for figure 7. Obviously the anisotropy can be adjusted by the strength of the external magnetic field, the periodic length and the RSOC term. So we can realize an external field controlled magneto-optical device base on different PMS configurations.

Finally the optical absorption spectrum of such a phosphorene magnetic superlattice is calculated, which can also be measured directly. In our calculation, we set the Fermi
level at zero energy, which is between the conduction and valence bands. This means the occupation of the valence band is full, while that for the conduction band is empty. We use a broadening factor of 0.15 meV to smooth the absorption spectrum. The optical absorption spectrum indicates useful broadening factor of 0.15 meV to smooth the absorption valence bands. This means the occupation of the valence band level at zero energy, which is between the conduction and valence bands. This means the occupation of the valence band is full, while that for the conduction band is empty. We use a broadening factor of 0.15 meV to smooth the absorption spectrum.

For a y-direction PMS, we can find a much higher absorption peak in the low-energy region and more oscillations when increasing the energy (frequency) of the incident light due to modulations from the y-direction PMS. Stronger coupling between the electrons and holes enhances the interband transition and optical absorption. Increasing the magnetic field can also increase the absorption rate. We note en passant that, for a linearly polarized light, the optical absorption is negligible when light is polarized along the y-direction [44], since the y-direction linearly polarized light (\( A_y = 0 \)) cannot affect the optic–electric interaction matrix in the off-diagonal element. We note that even in our PMS proposal, the y-direction linearly polarized light still has no effect on the optic–electric interaction matrix, i.e., no enhancement on the transition rate calculated in equation (9). Therefore, the y-direction PMS can hardly enhance the absorption spectrum of the y-direction linearly polarized light. The distinct optical absorption spectra of circular polarized light provide an effective way to detect the anisotropic energy properties of a PMS with different periodic orientations. The phosphorene magnetic superlattice is a promising platform for potential application in anisotropic magneto-optical devices.

### 4. Conclusions

In this work, we theoretically investigate the electronic and optical properties of a PMS utilizing the \( \vec{k} \cdot \vec{p} \) method. Our numerical results show that the anisotropic energy dispersions can be tuned by the PMS configurations, e.g., orientation, periodic length and strength. Accordingly the e–h pair energies also exhibit distinct differences between two configurations. We demonstrate that the y-direction (zigzag direction) superlattice gives rise to more pronounced modulation via the vector potential appearing in the electron–hole coupling term. As compared to the energy dispersion, charge distribution is much more sensitive to the external magnetic field or electric field (via RSOC). The magnetic fields and periodic orientations of the PMS proposed in this work play important roles in determining the e–h transition rates and the optical absorption spectrum. Our theoretical results shed new light on the potential applications of magneto-optical devices based on the anisotropic PMS.

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