Supplementary Material for "Dimensionality and Anisotropicity Dependence of Radiative Recombination in Nanostructured Phosphorene"

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I. COMPUTATIONAL METHODS

The ground state properties are obtained from density functional theory (DFT) calculations using open source plane-wave code Quantum-Espresso[1, 2] with Perdue-Burke-Ernzehorf (PBE) exchange-correlation functional [3], ONCV norm-conserving pseudopotentials [4, 5] and planewave basis set up to a wavefunction cutoff of 70 Ry. The atom positions are fully relaxed in all systems. The periodic direction of MBP nanoribbons is along either zigzag or armchair directions. Both MBP nanoribbons and quantum dots are hydrogen terminated due to the large edge effect from the dangling bonds which could be easily passivated in an experimental condition[6]. The k point mesh is kept as 10 along the armchair direction and 14 along the zigzag direction in both 2D and 1D cases. The SCF total energy convergence threshold is 10^{-8} Ry and the force convergence threshold is 10^{-3} Ry/Bohr.

The band gaps are computed through GW approximation with the WEST-code, which avoids explicit empty states and inversion of dielectric matrices[7, 8]. We implemented 2D, 1D and 0D Coulomb truncations[9, 10] for the dielectric matrix and correlation part of GW self-energies in the WEST-code, in order to truncate the long-range Coulomb interaction and speed up the vacuum convergence. We adopted 20 Å vacuum size in most of our calculations. The Gygi-Baldereschi trick plus the extrapolation at $G = 0$ term has been applied to the exchange part of the self energy[11, 12] in GW calculations for all dimensions.

The absorption spectra and exciton properties are computed by solving the Bethe-Salpeter equation (BSE) in the Yambo-code[13] for MBP nanoribbons and monolayer systems. For MBP quantum dots, a BSE implementation without explicit empty states and inversion of dielectric matrix is applied instead[14–16], because a much larger number of empty bands is required to converge the BSE calculations in the 0D case compared with the 1D and 2D cases. A scissor operator based on the GW calculation is applied to the BSE calculations. Both *GW* and BSE calculations used a smaller planewave basis set cutoff of 20 Ry. The number of k-points along periodic directions in *GW* are doubled (20 or 28) compared with ground state calculations and are four times (40 or 56) in BSE calculations in order to achieve the convergence within 0.1 eV. More k point convergence tests can be found in section VII.

II. EXCITON RADIATIVE LIFETIME IN 0D, 1D AND 2D ANISOTROPIC SYSTEMS

Summary of the notations used in this work:

- $\boldsymbol{Q}_{\mathrm{ex}}$, $\boldsymbol{Q}$: exciton wavevector (for simplicity the subscript "ex" is neglected in $\boldsymbol{Q}$)
- $\boldsymbol{q}_{\mathrm{L}}$, $\boldsymbol{q}$: photon wavevector (for simplicity subscript "L" is neglected in $\boldsymbol{q}$)
- q, q_{L} : the norm of $\boldsymbol{q}$ ($|\boldsymbol{q}|$)
- Q, Q_{ex} : the norm of $\boldsymbol{Q}$ ($|\boldsymbol{Q}|$)
- θ, θ_{L} : polar angle of photon wavevector. Without subscript it is always refer to the photon.
- θ_{ex} : polar angle of exciton wavevector
- φ, φ_{L} : azimuth angle of photon wavevector. Without subscript it is always refer to the photon.
- φ_{ex} : azimuth angle of exciton wavevector.

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- q', θ', φ' : An exciton with wavevector $\boldsymbol{Q}$ can only emit photons of specific wavevectors due to energy conservation and momentum conservation. Those constrained components are denoted as q', θ', φ'. This can be computed from $\boldsymbol{Q}$ and $\Omega(\boldsymbol{Q})$ (see following discussions)
  - 0D : No constraint for $\boldsymbol{Q}$
  - 1D : $\cos\theta' = cQ_z/\Omega(\boldsymbol{Q})$
  - 2D : $\sin\theta' = cQ_{xy}/\Omega(\boldsymbol{Q})$, $\varphi' = \varphi_{\text{ex}}$
  - 3D : $q' = Q$, $\theta' = \theta_{\text{ex}}$, $\varphi' = \varphi_{\text{ex}}$ ($\boldsymbol{q} = \boldsymbol{Q}$)
- Ω: $\hbar\Omega$ is exciton energy
- Ω_0 : Ω at $\boldsymbol{Q} = 0$
- μ_i : $i = x, y, z$, exciton dipole matrix element, can be obtained by solving BSE.
- μ_A : the normal of the exciton dipole moment ($\mu_A = \sqrt{|\mu_x|^2 + |\mu_y|^2 + |\mu_z|^2}$)
- M_i : $M_i = \mu_i/\mu_A$ is the ratio between the matrix element with a given polarization direction μ_i and its norm μ_A.
- $\boldsymbol{M}$: $\boldsymbol{M} = (M_x, M_y, M_z)$

Without loss of generality, we choose the coordinate system of low-dimensional systems as follows:

- 2D : 2D material plane as the xy-plane
- 1D : 1D periodic direction as z-axis

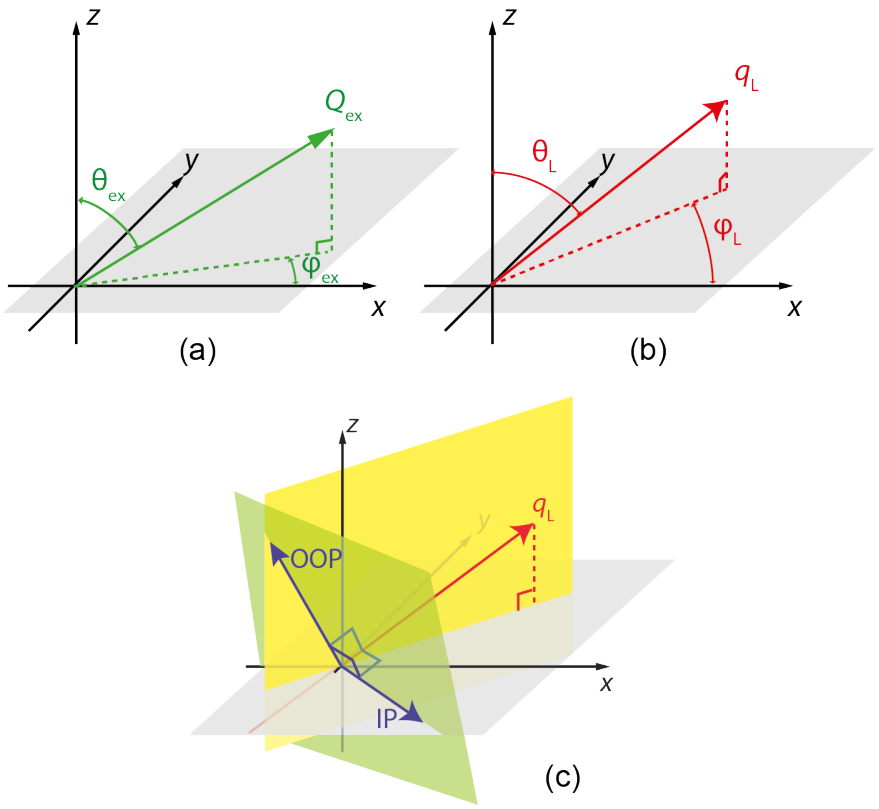

FIG. S1: The definition of exciton wavevector $\boldsymbol{Q}$, photon wavevector $\boldsymbol{q}$, two polarization directions in-plane (IP) and out-of-plane (OOP) as basis vectors. $\boldsymbol{q}$ and $\boldsymbol{Q}$ can be represented by polar angle (θ_{ex} and θ_{L}) and azimuth angle (φ_{L} and φ_{ex}). $\boldsymbol{Q}$ may be constrained along z-axis (1D) or in the xy-plane (2D). The OOP direction is in the plane of z-axis and $\boldsymbol{q}$, while the IP direction is in the xy-plane. Both are perpendicular to $\boldsymbol{q}$.

The Hamiltonian describing the electron-photon interaction can be written as

$$H^{\text{int}} = -\frac{e}{mc}\int \mathrm{d}\boldsymbol{r}\Psi(\boldsymbol{r})^{\dagger}\boldsymbol{A}(\boldsymbol{r})\cdot\boldsymbol{p}\Psi(\boldsymbol{r}) \tag{1}$$

where $\boldsymbol{p}$ is the momentum operator and $\Psi(\boldsymbol{r})^{\dagger}$ and $\Psi(\boldsymbol{r})$ are field operators creating or annihilating an electron in the position $\boldsymbol{r}$. In Eq. 1

$$\boldsymbol{A}(\boldsymbol{r}) = \sum_{\boldsymbol{q}\lambda}\sqrt{\frac{2\pi\hbar c}{Vq_L}}\boldsymbol{\epsilon}_{\boldsymbol{q}\lambda}[a^{\dagger}_{\boldsymbol{q}\lambda}e^{-i\boldsymbol{q}\cdot\boldsymbol{r}} + \text{H.c.}] \tag{2}$$

is the vector potential with $a^{\dagger}_{\boldsymbol{q}\lambda}$ denoting the creation operator of a photon with polarization vector $\boldsymbol{\epsilon}_{\boldsymbol{q}\lambda}$.

To discuss the light polarization, we can choose two polarization directions: in-plane (IP) and out-of-plane (OOP), with IP in the xy plane and OOP in the plane formed by z and $\boldsymbol{q}$ (see Fig. S1). The photon wavevectors and polarization are defined as follows:

$$\cos\theta = q_z/q \tag{3}$$
$$\tan\varphi = q_y/q_x \tag{4}$$
$$\boldsymbol{\epsilon}_{\boldsymbol{q}1} = (-\sin\varphi, \cos\varphi, 0) \tag{5}$$
$$\boldsymbol{\epsilon}_{\boldsymbol{q}2} = (-\cos\theta\cos\varphi, -\cos\theta\sin\varphi, \sin\theta) \tag{6}$$
$$\boldsymbol{\epsilon}_{\boldsymbol{q}1}\cdot\langle G|\boldsymbol{r}|S\rangle = -\mu_x\sin\varphi + \mu_y\cos\varphi \tag{7}$$
$$\boldsymbol{\epsilon}_{\boldsymbol{q}2}\cdot\langle G|\boldsymbol{r}|S\rangle = -\mu_x\cos\theta\cos\varphi - \mu_y\cos\theta\sin\varphi + \mu_z\sin\theta. \tag{8}$$

In previous work the exciton radiative lifetime of 1D systems was derived by considering only the z-component of $\langle G|\boldsymbol{r}|S\rangle$ [17]. Here we extend this formalism to include all components of $\langle G|\boldsymbol{r}|S\rangle$, as required to correctly describe the lifetimes of phosphorene nanoribbons (see main text):

$$\begin{aligned}
\gamma(\boldsymbol{Q}) =& \sum_{q,\lambda=1,2}\gamma(\boldsymbol{Q},\boldsymbol{q},\lambda)\\
=&\frac{4\pi^2e^2}{\hbar c^2V}\Omega(\boldsymbol{Q})^2\sum_{\boldsymbol{q}\lambda}\frac{1}{q}\left|\boldsymbol{\epsilon}_{\boldsymbol{q}\lambda}\cdot\langle G|\boldsymbol{r}|S\rangle\right|^2\delta(\frac{\Omega(\boldsymbol{Q})}{c}-q)\\
=&\frac{4\pi^2e^2}{\hbar c^2V}\Omega(\boldsymbol{Q})^2\frac{L_xL_y}{(2\pi)^2}\int\mathrm{d}q_x\int\mathrm{d}q_y\sum_{q_z}\delta_{q_z,Q_z}\\
&\frac{1}{q}\sum_{\lambda}\left|\boldsymbol{\epsilon}_{\boldsymbol{q}\lambda}\cdot\langle G|\boldsymbol{r}|S\rangle\right|^2\delta\left(\frac{\Omega(\boldsymbol{Q})}{c}-\sqrt{q_x^2+q_y^2+q_z^2}\right)\\
=&\frac{e^2\Omega(\boldsymbol{Q})^2}{\hbar c^2L_z}\int_0^{\infty}\mathrm{d}q_{xy}\int_0^{2\pi}\mathrm{d}\varphi q_{xy}\\
&\frac{1}{q}\sum_{\lambda}\left|\boldsymbol{\epsilon}_{\boldsymbol{q}\lambda}\cdot\langle G|\boldsymbol{r}|S\rangle\right|^2\delta\left(\frac{\Omega(\boldsymbol{Q})}{c}-\sqrt{q_{xy}^2+Q_z^2}\right)
\end{aligned}$$

From the momentum conservation in 1D systems we have

$$q_z = Q_z \tag{9}$$

and from the energy conservation

$$q^2 = \Omega(\boldsymbol{Q})^2/c^2. \tag{10}$$

The specific values of q, q_z, q_{xy}, θ determined by the above conditions are denoted as $q', q'_z, q'_{xy}, \theta'$.

By simplifying the delta function and approximating $\Omega(\boldsymbol{Q}) \approx \Omega_0$, we obtain:

$$\begin{aligned}\gamma(\boldsymbol{Q}) \approx & \frac{e^2\Omega_0^2}{\hbar c^2 L_z}\int_0^{\infty} \mathrm{d}q_{xy}\int_0^{2\pi}\mathrm{d}\varphi\frac{q_{xy}}{q} \\ & \sum_{\lambda}|\boldsymbol{\epsilon}_{\boldsymbol{q}\lambda}\cdot\langle G|\boldsymbol{r}|S\rangle|^2\,\frac{\delta(q_{xy}-\sqrt{\Omega(\boldsymbol{Q})^2/c^2-Q_z^2})}{(1-\frac{Q_z^2c^2}{\Omega(\boldsymbol{Q})^2})^{1/2}} \\ = & \frac{e^2\Omega_0^2}{\hbar c^2 L_z}\int_0^{2\pi}\mathrm{d}\varphi\sum_{\lambda}|\boldsymbol{\epsilon}_{\boldsymbol{q}\lambda}\cdot\langle G|\boldsymbol{r}|S\rangle|^2 \\ = & \frac{e^2\Omega_0^2}{\hbar c^2 L_z}\pi\left((1+\cos^2\theta')(\mu_x^2+\mu_y^2)+2\sin^2\theta'\mu_z^2\right) \end{aligned} \tag{11}$$

With respect to Ref. 17, additional μ_x and μ_y present in the equations, which are essential for anisotropic systems and have been neglected in the past work [17].

In summary, the radiative decay rate of excitons $\gamma(\boldsymbol{Q})$ (the inverse of exciton lifetime) for different dimensionalities are:

- 0D:

$$\gamma_0 = \frac{4e^2}{3\hbar c^3}\Omega_0^3\mu_A^2 \tag{12}$$

$$\gamma(\boldsymbol{Q}) = \gamma_0 \tag{13}$$

- 1D with xyz components:

$$\gamma_0 = \frac{2\pi e^2}{\hbar c^2 L_z}\Omega_0^2\mu_A^2 \tag{14}$$

$$\gamma(\boldsymbol{Q}) = \gamma_0\frac{M_z^2 2\sin^2\theta' + (M_x^2+M_y^2)(1+\cos^2\theta')}{2} \tag{15}$$

$$\cos\theta' = cQ_z/\Omega_0 \tag{16}$$

- 2D with the xy component and M_z is approximately zero:

$$\gamma_0 = \frac{4\pi e^2}{\hbar c L_x L_y}\Omega_0\mu_A^2 \tag{17}$$

$$\begin{aligned}\gamma(\boldsymbol{Q}) = & \gamma_0\cos^{-1}\theta' \\ & \Big((-M_x\sin\varphi'+M_y\cos\varphi')^2 + \\ & \cos^2\theta_{\mathrm{L}}\left(M_x\cos\varphi'+M_y\sin\varphi'\right)^2\Big)\end{aligned} \tag{18}$$

$$\begin{aligned}\sin\theta' &= cQ_{xy}/\Omega_0 \\ \varphi' &= \varphi_{\mathrm{ex}}\end{aligned} \tag{19}$$

In the equations above, M_i denotes the normalized dipole moments along the directions $i = x, y, z$ defined as $M_i = \mu_i/\mu_A$, where μ_i is the i-th component of the dipole moment (e.g. $\mu_x = \langle G|x|S\rangle$) and $\mu_A = \sqrt{|\mu_x|^2+|\mu_y|^2+|\mu_z|^2}$; Ω_0 represent the exciton energy at $\boldsymbol{Q} = 0$ ($\Omega_0 = E(\boldsymbol{Q}=0)/\hbar$). $L_i = N_i l_i$ where l_i is the unit cell parameter and N_i is the number of k-point in the direction $i = x, y, z$; the angles θ_{L} and φ_{L} determining the directions of photon wavevectors $\boldsymbol{q}$ are defined in Fig. S1.

We note that the expressions in Eqs. 14-15 have never been discussed before in literature but are necessary to describe the peculiar behavior of the 1D nanostructures considered in this work. Indeed, in the systems studied in previous work [17] only light polarized along the periodic direction was relevant for the exciton lifetime (namely only M_z was providing a sizeable contribution). As discussed below, because of the interplay between anisotropicity and the truncated periodicity in certain directions, for some phosphorene nanoribbons all the three light polarization directions provide a comparable contribution to the radiative lifetime.

III. EXCITON LIGHT EMISSION INTENSITY IN 0D, 1D AND 2D ANISOTROPIC SYSTEMS

The light emission intensity with specific photon wavevector $\boldsymbol{q}$ and polarization λ emitted from all possible occupied excitonic states is

$$I(\boldsymbol{q},\lambda)=\sum_{\boldsymbol{Q}} n(\boldsymbol{Q})\gamma(\boldsymbol{Q},\boldsymbol{q},\lambda) \tag{20}$$

where $n(\boldsymbol{Q})$ is the occupation number of the excitonic state with momentum $\boldsymbol{Q}$.

Since we focus our discussion on the first exciton, the upper bound of the exciton wavevector $Q=\Omega_0/c$ is small and at thermal equilibrium we have

$$\frac{n(\boldsymbol{Q})}{n(\boldsymbol{Q}=0)}=\exp(-(E(\boldsymbol{Q})-E_0)/k_BT)\approx 1, \tag{21}$$

which means $n(\boldsymbol{Q})\approx n(\boldsymbol{Q}=0)$. For simplicity we define $n_0=n(\boldsymbol{Q}=0)$.

In this work we mainly focus on the angular dependence of light emission and, accordingly, below we will consider only the following expression

$$I(\theta,\varphi,\lambda)=\int dqI(\boldsymbol{q},\lambda), \tag{22}$$

obtained by integrating Eq. 20 over the norm of $\boldsymbol{q}$. In order to obtain explicit expressions for different dimensionalities, it is necessary to consider that along a periodic dimension i, $\gamma(\boldsymbol{Q},\boldsymbol{q},\lambda)\neq 0$ only when $q_i=Q_i$ due to the momentum conservation condition; along a non-periodic dimension j, we have $Q_j=0$. Additionally, because of the energy conservation condition, the norm of $\boldsymbol{q}$ is determined by $\hbar cq=\hbar\Omega(\boldsymbol{Q})\approx\hbar\Omega_0$.

According to these considerations, Eq. 22 can be written as

$$I(\theta,\varphi,\lambda)=\int \mathrm{d}qI(\boldsymbol{q},\lambda)=n_0\Gamma_0\left|\boldsymbol{\epsilon}_{\boldsymbol{q}\lambda}\cdot\boldsymbol{M}\right|^2, \tag{23}$$

where the difference among different dimensionalities is contained only in the angle-independent term Γ_0:

- 0D:

$$\Gamma_0=\frac{e^2\Omega_0\mu_A^2}{2\pi\hbar c}, \tag{24}$$

- 1D:

$$\Gamma_0=\frac{e^2\Omega_0\mu_A^2}{\hbar cL_z}, \tag{25}$$

- 2D:

$$\Gamma_0=\frac{2\pi e^2\Omega_0\mu_A^2}{\hbar cL_xL_y}. \tag{26}$$

Finally the angle-resolved light emission intensity with IP and OOP polarizations can be written in a general form applicable to systems of any dimensions:

$$I(\theta,\varphi,\mathrm{IP})=n_0\Gamma_0\left(-M_x\sin\varphi_\mathrm{L}+M_y\cos\varphi\right)^2, \tag{27}$$

$$I(\theta,\varphi,\mathrm{OOP})=n_0\Gamma_0(-M_x\cos\theta\cos\varphi-M_y\cos\theta_\mathrm{L}\sin\varphi+M_z\sin\theta)^2. \tag{28}$$

We note that the results obtained from Eq. 27, 28, and 26 for 2D systems is 1/2 of the ones from Eq. 4 and Eq. 5 in Ref. 18. This comes from a factor of 2 considering one $\boldsymbol{Q}$ corresponds to two $\boldsymbol{q}$ (q_x,q_y,q_z) and $(q_x,q_y,-q_z)$ instead of one $\boldsymbol{q}$ used in Ref. 18.

IV. POLARIZATION OF LIGHT DETERMINED FROM STOKES PARAMETERS

The polarization of light can be determined by Stokes parameters. The Stokes parameters are a set of values that can be defined with two perpendicular directions: horizontal (0°) and vertical (90°)[19]:

$$\boldsymbol{S} = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix}, \tag{29}$$

- S_0 : the total intensity of the light,
- S_1 : the intensity of the light passing through a horizontal linear polarizer*2 - total intensity,
- S_2 : the intensity of the light passing through a 45° linear polarizer*2 - total intensity,
- S_3 : the intensity of the light passing through a left circular polarizer*2 - total intensity.

In our work, we derived the Stokes parameters as a function of θ and φ. We define the coordinate system as follows: IP as the horizontal axis and OOP as the vertical axis. The related polarization vectors can be computed from the geometric relationship:

- Horizontal (IP): $\boldsymbol{\epsilon_{q\lambda}}[H] = (-\sin\varphi, \cos\varphi, 0)$
- Vertical (OOP) : $\boldsymbol{\epsilon_{q\lambda}}[V] = (-\cos\theta\cos\varphi, -\cos\theta\sin\varphi, \sin\theta)$
- 45° : $\boldsymbol{\epsilon_{q\lambda}}[45] = \frac{1}{\sqrt{2}}(\boldsymbol{\epsilon_{q\lambda}}[H] + \boldsymbol{\epsilon_{q\lambda}}[V])$
- Left circular: $\boldsymbol{\epsilon_{q\lambda}}[L] = \frac{1}{\sqrt{2}}(\boldsymbol{\epsilon_{q\lambda}}[H] + i\boldsymbol{\epsilon_{q\lambda}}[V])$

By substituting the above polarization vectors into Eq. 23, we can get a series of equations.

$$S_0 = n_0\Gamma_0(|\boldsymbol{\epsilon_{q\lambda}}[H]\cdot\boldsymbol{M}|^2 + |\boldsymbol{\epsilon_{q\lambda}}[V]\cdot\boldsymbol{M}|^2) \tag{30}$$
$$S_1 = n_0\Gamma_0(|\boldsymbol{\epsilon_{q\lambda}}[H]\cdot\boldsymbol{M}|^2 - |\boldsymbol{\epsilon_{q\lambda}}[V]\cdot\boldsymbol{M}|^2) \tag{31}$$
$$S_2 = n_0\Gamma_0(2\,|\boldsymbol{\epsilon_{q\lambda}}[45]\cdot\boldsymbol{M}|^2 - |\boldsymbol{\epsilon_{q\lambda}}[H]\cdot\boldsymbol{M}|^2 - |\boldsymbol{\epsilon_{q\lambda}}[V]\cdot\boldsymbol{M}|^2) \tag{32}$$
$$S_3 = n_0\Gamma_0(2\,|\boldsymbol{\epsilon_{q\lambda}}[L]\cdot\boldsymbol{M}|^2 - |\boldsymbol{\epsilon_{q\lambda}}[H]\cdot\boldsymbol{M}|^2 - |\boldsymbol{\epsilon_{q\lambda}}[v]\cdot\boldsymbol{M}|^2) \tag{33}$$

The Stokes parameters can be normalized without altering the polarization. Expressing Eqs. 30-33 as a function of angles θ and φ for photon wavevectors and normalizing Stokes parameters by dividing by $n_0\Gamma_0$, we get:

$$\begin{aligned} S_0' = {} & |-M_x\sin\varphi + M_y\cos\varphi|^2 \\ & + |-M_x\cos\theta\cos\varphi - M_y\cos\theta\sin\varphi + M_z\sin\theta|^2 \end{aligned} \tag{34}$$
$$\begin{aligned} S_1' = {} & |-M_x\sin\varphi + M_y\cos\varphi|^2 \\ & - |-M_x\cos\theta\cos\varphi - M_y\cos\theta\sin\varphi + M_z\sin\theta|^2 \end{aligned} \tag{35}$$
$$\begin{aligned} S_2' = {} & |-M_x\sin\varphi + M_y\cos\varphi \\ & - M_x\cos\theta\cos\varphi - M_y\cos\theta\sin\varphi + M_z\sin\theta|^2 - S_0 \end{aligned} \tag{36}$$
$$\begin{aligned} S_3' = {} & |-M_x\sin\varphi + M_y\cos\varphi \\ & - iM_x\cos\theta\cos\varphi - iM_y\cos\theta\sin\varphi + iM_z\sin\theta|^2 - S_0 \end{aligned} \tag{37}$$

Because the component of $\langle G|\boldsymbol{r}|S\rangle$ perpendicular to the 2D material plane is always zero, at least one of M_x, M_y and M_z can be chosen to be zero. Here we set $M_z = 0$, and keep M_x and M_y as variables. Then we can obtain the normalized Stokes parameters of light emission of excitons in low dimensional materials:

$$
\begin{aligned}
S_0' =&(M_x \sin\varphi - M_y \cos\varphi)(M_x^* \sin\varphi - M_y^* \cos\varphi) \\
&+ \cos^2\theta (M_x \cos\varphi + M_y \sin\varphi)(M_x^* \cos\varphi + M_y^* \sin\varphi) && (38)\\
S_1' =&(M_x \sin\varphi - M_y \cos\varphi)(M_x^* \sin\varphi - M_y^* \cos\varphi) \\
&- \cos^2\theta (M_x \cos\varphi + M_y \sin\varphi)(M_x^* \cos\varphi + M_y^* \sin\varphi) && (39)\\
S_2' =&\cos\theta (M_x M_x^* \sin(2\varphi) - M_x M_y^* \cos(2\varphi) \\
&- M_y M_y^* \sin(2\varphi) - M_x^* M_y \cos(2\varphi)) && (40)\\
S_3' =&i \cos\theta (-M_x M_y^* + M_x^* M_y) && (41)
\end{aligned}
$$

V. ANGLE DEPENDENCE OF LIGHT EMISSION INTENSITY

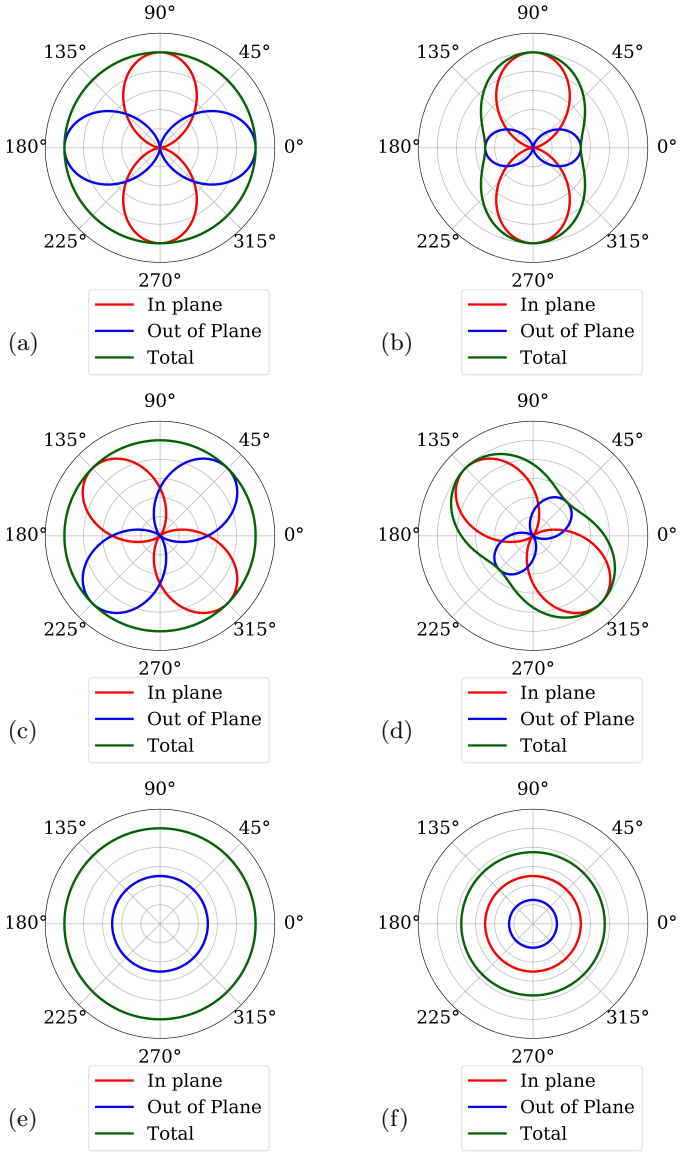

FIG. S2: The angle dependence of light emission intensity. The intensity along IP and OOP polarization directions at different φ_{L} (azimuthal angle that defines photon wavevector) with given θ_{L} (polar angle) of different M_x, M_y, M_z combination computed from Eq. 27 and Eq. 28.

- (a)(b): $\boldsymbol{M} = (1, 0, 0)$ (Arbitrary M_x)
- (c)(d): $\boldsymbol{M} = \frac{1}{\sqrt{2}}(1, 1, 0)$
- (e)(f): $\boldsymbol{M} = \frac{1}{\sqrt{2}}(1, i, 0)$
- (a)(c)(e) $\theta_{\mathrm{L}} = 0°$
- (b)(d)(f) $\theta_{\mathrm{L}} = 45°$

From Eqs. 23 to 28, we can see that the angle dependence of light emission intensity ($I/(n(\boldsymbol{Q})\Gamma_0)$) on the photon wavevector direction ($\theta_{\mathrm{L}}, \varphi_{\mathrm{L}}$) is not directly related to the systems' dimensionality, but rather depends on the dot product between the polarization vector $\boldsymbol{\epsilon_{q\lambda}}$ and the normalized dipole moment $\boldsymbol{M}$. For the case of MBP nanostructures, regardless of dimensionalities, we can take the x-axis along the armchair direction, y-axis along the zigzag direction and z-axis perpendicular to the material plane. Because the first exciton is only bright when light polarized along the armchair direction as discussed earlier, the first exciton dipole moments of 0D, 1D and 2D MBP nanostructures can all be described by the same condition: $M_x = 1, M_y = M_z = 0$. Therefore, the light emission intensities of MBP nanostructures follow the same angle dependence of photon wavevector for the first exciton, independent of their dimensionality.

Because $\boldsymbol{M} = (1, 0, 0)$ in MBP nanostructures discussed above, a $\cos^2 \varphi_{\mathrm{L}}$ relationship between the light emission intensity and the photon wavevector azimuth angle φ_{L} can be always obtained from Eq. 27 and Eq. 28, as shown in Fig. S2(a) and (b). From Eq. 23, the intensity reaches maximum when the polarization vector $\boldsymbol{\epsilon_{q\lambda}}$(IP) or $\boldsymbol{\epsilon_{q\lambda}}$(OOP) is along the armchair direction of MBP. When $\theta = 45°$ the OOP polarization direction can never be along the armchair direction, so the corresponding maximum intensity is smaller than IP at any θ and OOP at $\theta = 0°$.

In comparison, the angle dependence of light emission intensity is different in in-plane isotropic low-dimensional materials (such as monolayer MoS_2 and h-BN) from an in-plane anisotropic system like MBP. Both MoS_2 and h-BN belong to the hexagonal group symmetry and the lowest direct transition is at K to K'. The valley-degeneracy of K and K' gives extra degree of freedom: the two excitons on K, K' valleys can mix depending on the incident light[18], which is able to give arbitrary exciton dipoles. Two possible values of $\boldsymbol{M}$ in MoS_2 and h-BN different from MBP are shown in Fig. S2: $\boldsymbol{M} = 1/\sqrt{2}(1, 1, 0)$, $\boldsymbol{M} = 1/\sqrt{2}(1, i, 0)$. Specifically, the angle dependence of light emission intensity with $\boldsymbol{M} = 1/\sqrt{2}(1, 1, 0)$ is plotted in Fig. S2 with $\theta_{\mathrm{L}} = 0°$(c) and $\theta_{\mathrm{L}} = 45°$(d). This case is exactly the same as rotating the x and y axis by $\pi/4$ in Fig. S2 (a) and (b) with $\boldsymbol{M} = (1, 0, 0)$. However, with $\boldsymbol{M} = 1/\sqrt{2}(1, i, 0)$, the light emission intensity is spherically symmetric in the xy-plane in Fig. S2 (e) and (f), which is distinct from MBP and can not be obtained from the case of MBP in (a) and (b) through rotation of axis.

In summary, the emitted photon wavevector angle dependence for in-plane anistropic systems like MBP nanostructures is always $\cos^2(\varphi_L)$ as Fig. S2 (a) and (b), and can not be spherical symmetric as Fig. S2 (e) and (f) regardless of the excitation laser polarization in angle-resolved PL measurements. This is determined by the anisotropic nature of MBP, and is applicable to all 0D, 1D and 2D MBP nanostructures. Instead, in-plane isotropic systems can have various angle dependence including spherical symmetric light emission intensity due to the mixed exciton states between K and K' valleys.

VI. IMAGINARY PART OF DIELECTRIC FUNCTION OF NANOSTRUCTURED PHORSPHORENE FROM SOLVING THE BETHE-SALPETER EQUATION

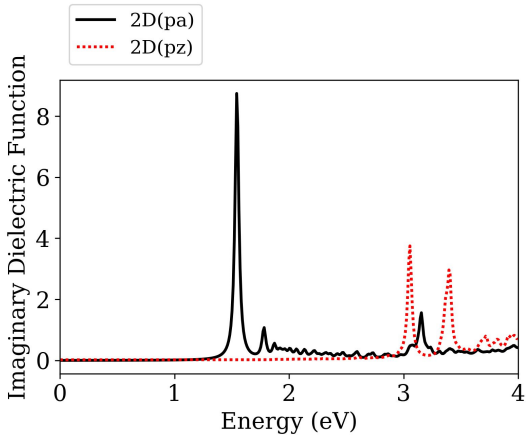

FIG. S3: The imaginary part of the dielectric function of 2D MBP ("pa" denotes polarization along the armchair direction and "pz" denotes the zigzag direction) computed by solving the Bethe-Salpeter equation.

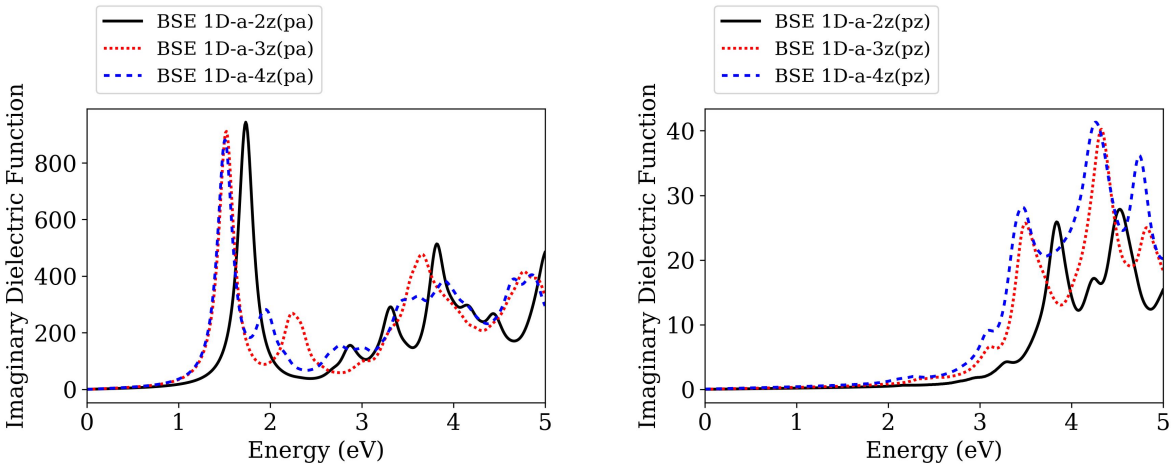

FIG. S4: The imaginary part of dielectric function of 1D-a-Nz MBP nanoribbons computed by solving the Bethe-Salpeter equation (The armchair direction is periodic; "pa" denotes the light polarization along the armchair direction and "pz" denotes along zigzag direction). For comparison the dielectric function is normalized by multiplying the volume / the number of electrons. (V/N_{e}).

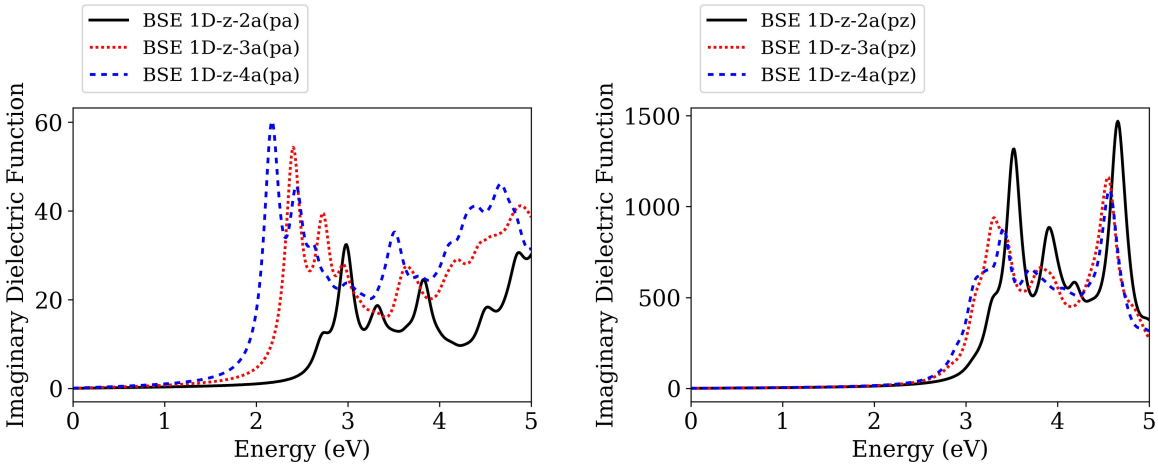

FIG. S5: The imaginary part of dielectric function of 1D-z-Na MBP nanoribbons by solving the Bethe-Salpeter equation (the zigzag direction is periodic; "pa" means polarization along the armchair direction and "pz" means zigzag direction). For comparison the dielectric function is normalized by multiplying the volume / the number of electrons. (V/N_{e}).

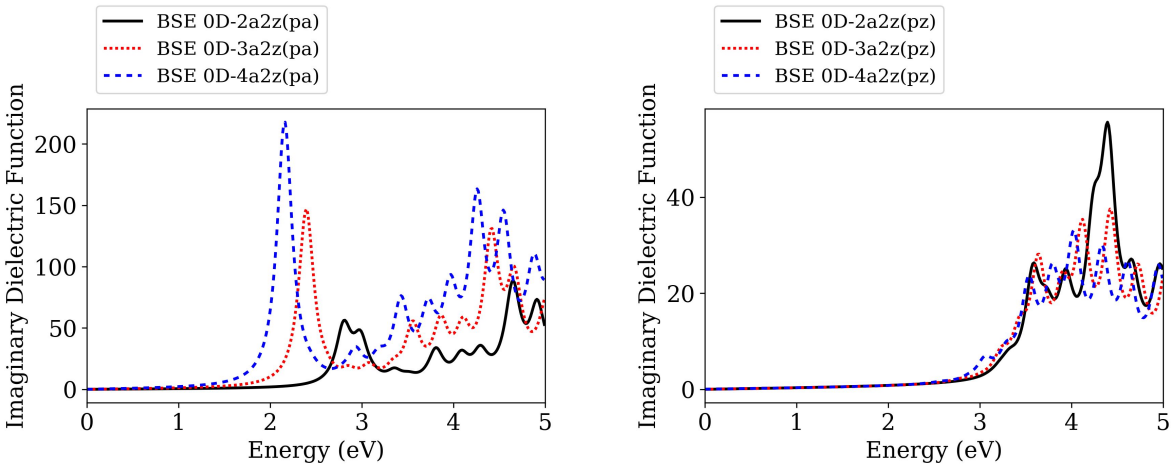

FIG. S6: The imaginary dielectric function of 0D-Na-2z MBP quantum dots by solving the Bethe-Salpeter equation (extended along the armchair direction; "pa" denotes light polarization along the armchair direction and "pz" denotes light polarization along the zigzag direction). For comparison the dielectric function is normalized by multiplying the volume / the number of electrons. (V/N_{e}.)

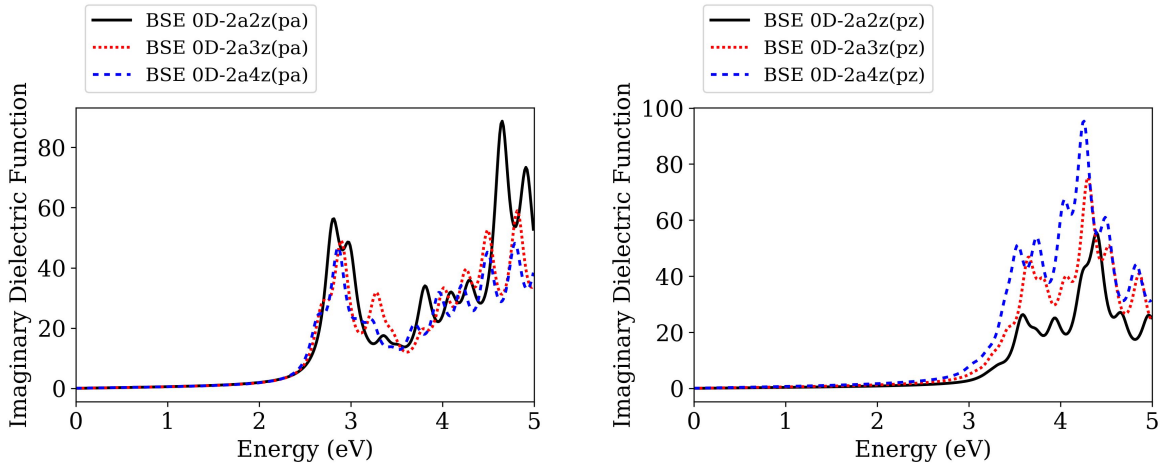

FIG. S7: The imaginary part of dielectric function of 0D-2a-Nz MBP quantum dots by solving the Bethe-Salpeter equation (extended along the zigzag direction; "pa" denotes polarization along the armchair direction and "pz" denotes along the zigzag direction). For comparison the dielectric function is normalized by multiplying the volume / the number of electrons. (V/N_{e}).

VII. FUNDAMENTAL BAND GAPS AND OPTICAL PROPERTIES OF NANOSTRUCTURED PHOSPHORENE

TABLE S1: Band gaps and first exciton properties of 2D, 1D and 0D nanostuctured phosphorene with light polarized along the armchair direction. This exciton is only bright with light polarized along the armchair direction.

System	Width Armchair (Å)	Width Zigzag (Å)	GW Gap (eV)	Exciton Peak (eV)	Exciton Binding (eV)	Lifetime at 0K
0D-2a2z	9	9	5.62	2.80	2.81	57 ns
0D-2a3z	9	12	5.28	2.68	2.60	125 ns
0D-2a4z	9	15	5.05	2.64	2.41	113 ns
0D-3a2z	13	9	4.77	2.39	2.38	20 ns
0D-4a2z	18	9	4.29	2.16	2.13	14 ns
1D-a-2z	∞	9	3.37	1.73	1.64	21 ps
1D-a-3z	∞	12	2.93	1.52	1.41	21 ps
1D-a-4z	∞	15	2.75	1.51	1.24	17 ps
1D-z-2a	9	∞	4.59	2.72	1.87	1.0 ns
1D-z-3a	13	∞	3.81	2.40	1.41	130 ps
1D-z-4a	18	∞	3.37	2.17	1.20	110 ps
2D	∞	∞	2.22	1.54	0.68	99 fs

"1D-a" and "1D-z" denote that the periodic direction is along the armchair (a) or zigzag (z) direction. The number M and N in "1D-a-Nz" , "1D-z-Na" and "0D-Na-Mz" gives the number of unit cells in given directions.

TABLE S2: Band gaps and first exciton properties of 2D and 1D nanostuctured phosphorene compared with previously reported values.

System	Width Armchair (Å)	Width Zigzag (Å)	DFT Gap (eV)	GW Gap (eV)	Exciton Peak (eV)	Exciton Binding (eV)
1D-a-2z [This work]	∞	9	1.35	3.37	1.73	1.64
1D-a-2z [20]	∞	9	1.3	3.4	1.9	1.5
2D [This work]	∞	∞	0.91	2.22	1.54	0.68
2D [20]	∞	∞	0.95	2.3	1.5	0.8
2D [21]	∞	∞	0.85	2.0	1.2	0.8

"1D-a" and "1D-z" denote that the periodic direction is along the armchair (a) or zigzag (z) direction. The number M and N in "1D-a-Nz" , "1D-z-Na" and "0D-Na-Mz" gives the number of unit cells in given directions.

TABLE S3: Convergence of k-points in *GW* calculations of nanostructured phosphorene.

System	Property	K-points	Value (eV)
2D	*GW* gap	7x5	3.05
		11x7	2.57
		14x10	2.41
		Extrapolated ∞	2.22
1D-a-1z	*GW* gap	10	4.85
		20	4.64
		40	4.60

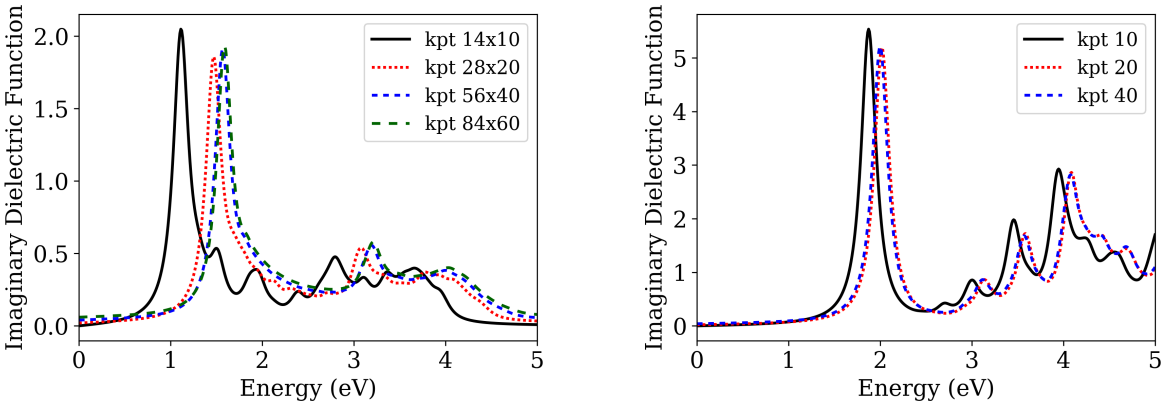

FIG. S8: The imaginary part of dielectric function of 2D MBP (left) and 1D-z-Na MBP (right) nanoribbons as a function of k points by solving the Bethe-Salpeter equation with light polarized along the armchair direction. BSE calculations are done with a fixed scissor operator across all k-points.

From Table S3 and Fig. S8, we can see that BSE with 56x40 k-points converges to 0.1 eV and *GW* with 20 k-points converges to 0.1 eV. For the 2D MBP *GW* calculation, due to the computational cost, the 2D GW band gap is extrapolated from 11x7 and 14x10 to infinity number of k-points based on the $1/N_k$ asymptotic behavior[22].

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