Supplementary Material for "Anisotropic Dependence of Radiation from Exciton in Ga_2O_3/MoS_2 Heterostructure"

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S1 Computational Details

In this paper, our calculations are based on density functional theory (DFT) and many-body perturbation theory, including the GW and Bethe-Salpeter equation (BSE) method, implemented in BerkeleyGW package.[1, 2, 3] We first calculated the mean-field wavefunctions and eigenvavlues based on the density functional theory (DFT), implemented in the Quantum Espresso package, within the local density approximation (LDA) functional with the plane wave basis.[4, 5] As the Ga_2O_3 monolayer in this paper is ferroelectrics , to better understand the interactions of the adjacent layers in the heterostructure, the van der Waals correction is included with the type of DFT-D2[6] in the functionals. The pseudopotential is used in a norm-conserving type with a energy cutoff 60 Ry. The force and electronic convergence tolerance is set to 0.001 Ry/Bohr and 10^{-8} Ry. A Monkhorst-Pack-Γ-centered k grid is set to $11\times11\times1$ in the structure optimization, while it is set to $15\times15\times1$ in the self-consistent calculation. As the ferroelectric properties of Ga_2O_3 monolayer, the vacuum slab is set to 30 Å. The exciton wave function is visualized with a supercell of 10×10.

To verify the thermal stability of the heterostructure, by setting the threshold of 10^{-14}, the force sets can be calculated by Quantum Espresso, with the displacement method in a supercell with the k grid of $15\times15\times1$. In the GW and BSE calculations, the mean-field calculations are performed by using Quantum Espresso package. Then the self-energy in GW calculation is calculated with k grid of $21\times21\times1$. In the BSE calculations, the k-point is doubled by interpolating with a finer k-mesh of $41\times41\times1$. To make sure the convergence, the number of empty band is set to 500, which is 10 times larger than the band number of occupied states and makes sure the

convergences of band gap below 0.05eV. The convergence of k-points and numbers of bands are shown in table 1 and figure 1. From table 1 and figure 1, we notice that the GW calculations with $21x21$ converges to 0.05eV, and the BSE calculations with 21×21 (interpolated with a finer k-mesh of 41×41) converges to 0.05 eV.

Table 1: Convergence of k-points and band numbers in GW calculations of FE-ZB/MoS_2 heterostructures. In the calculation of the convergence of band numbers, the k-point is set to $21x21$.

k-points	E_{gw}(eV)
11x11	2.68
15x15	2.33
21x21	2.18
35x35	2.15
number of bands	**E_{gw}(eV)**
200	2.49
500	2.18
2000	2.16

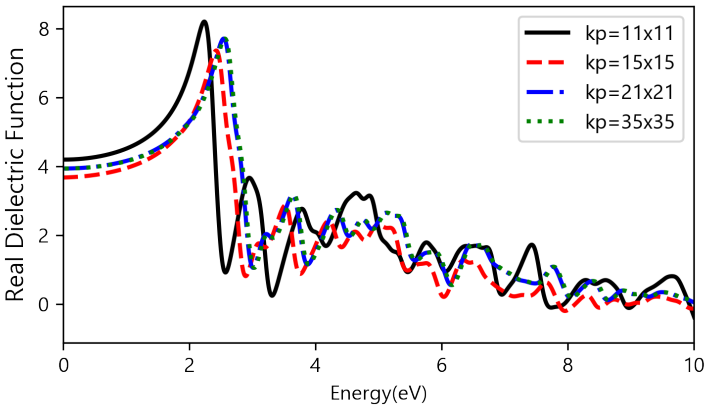

Figure 1: The real dielectric function of FE-ZB/MoS_2 heterostructure in the BSE calculations.

S2 Intensity of Radiation

Our approach to computing the lifetimes of radiation is based on the Fermi's golden rule formula,

$$\gamma = \frac{2\pi}{\hbar}\left|\langle G|H_{\text{int}}|S\rangle\right|^2 \delta(E_S - \hbar\omega_k) \tag{1}$$

where γ represents the rate of emission of a photon with angular frequency ω_k, and E_s is the exciton energy. The Hamiltonian describing the electron-photon interaction can be expressed as,[7]

$$H_{\text{int}} = -\frac{e}{m}\int \mathrm{d}\mathbf{r}\psi^\dagger(\mathbf{r})\mathbf{A}(\mathbf{r})\cdot\mathbf{p}\psi(\mathbf{r}) \tag{2}$$

where $\mathbf{p}$ is the electron momentum operator and the vector-potential operator $\mathbf{A}(\mathbf{r})$ is,[8]

$$\mathbf{A}(\mathbf{r}) = \sum_{\mathbf{k}}\sum_{\lambda=1,2}\sqrt{\frac{\hbar}{2\varepsilon_0 V\omega_k}}\boldsymbol{e}_{\mathbf{k}\lambda}[a^\dagger_{\mathbf{k}\lambda}\mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} + h.c.]. \tag{3}$$

The difficulty is to evaluate $|\langle G|H_{\text{int}}|S\rangle|^2$. Inserting equation (3) into (1), with the relation between operators $\mathbf{r}$ and $\mathbf{p}$ ($\mathbf{p} = -im/\hbar[\mathbf{r}, H]$), γ can be written as,

$$\begin{aligned}\gamma &= \frac{e^2\pi}{m^2\varepsilon_0 V}\sum_{\mathbf{k}}\sum_{\lambda=1,2}\frac{1}{\omega_k}|\langle G|\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\mathbf{p}|S\rangle|^2\delta(E_s-\hbar\omega_k)\\ &= \frac{e^2\pi}{m^2\varepsilon_0 Vc}\sum_{\mathbf{k}}\sum_{\lambda=1,2}\frac{1}{k}|\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\langle G|\mathbf{p}|S\rangle|^2\delta(E_s-\hbar ck)\\ &= \frac{E_s^2e^2\pi}{\hbar^2\varepsilon_0 Vc}\sum_{\mathbf{k}}\sum_{\lambda=1,2}\frac{1}{k}|\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\boldsymbol{\mu}|^2\delta(E_s-\hbar ck)\end{aligned} \tag{4}$$

As

$$\sum_{\mathbf{k}}\longrightarrow\int \mathrm{d}^3k,$$

$$\begin{aligned}\gamma &= \frac{E_s^2e^2}{8\pi^2\hbar^2\varepsilon_0 c}\sum_{\lambda=1,2}\int \mathrm{d}^3k\frac{1}{k}|\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\boldsymbol{\mu}|^2\delta(E_s-\hbar ck)\\ &= \frac{E_s^2e^2}{8\pi^2\hbar^3\varepsilon_0 c^2}\sum_{\lambda=1,2}|\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\boldsymbol{\mu}|^2\int \mathrm{d}^3k\frac{1}{k}\delta(\frac{E_s}{\hbar c}-k)\\ &= \frac{E_s^2e^2}{8\pi^2\hbar^3\varepsilon_0 c^2}\sum_{\lambda=1,2}\int \mathrm{d}\Omega|\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\boldsymbol{\mu}|^2\int \mathrm{d}k\,k\delta(\frac{E_s}{\hbar c}-k)\\ &= \frac{E_s^3e^2}{8\pi^2\hbar^4\varepsilon_0 c^3}\sum_{\lambda=1,2}\int \mathrm{d}\Omega|\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\boldsymbol{\mu}|^2\\ &= \frac{E_s^3e^2\mu^2}{8\pi^2\hbar^4\varepsilon_0 c^3}\sum_{\lambda=1,2}\int \mathrm{d}\Omega|\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\boldsymbol{e}_{\boldsymbol{\mu}}|^2\\ &= \gamma_0\int \mathrm{d}\Omega\ \mathrm{Y}(\theta,\phi)\end{aligned} \tag{5}$$

$$\gamma_0 = \frac{E_s^3e^2\mu^2}{8\pi^2\hbar^4\varepsilon_0 c^3}$$

where $\int \mathrm{d}\Omega = \int \sin\theta \mathrm{d}\theta\mathrm{d}\phi$ is the integral over solid angle. $\gamma(\theta,\phi) = \sum_\lambda |\boldsymbol{e}_{\mathbf{k}\lambda}\cdot\boldsymbol{e}_{\boldsymbol{\mu}}|^2 = \gamma_{\text{IP}} + \gamma_{\text{OP}}$, $\boldsymbol{\mu} = |\langle G|\mathbf{r}|S\rangle| = (\mu_x, \mu_y, \mu_z)$.

The unit vector of light propagating direction:

$$\boldsymbol{e}_{\boldsymbol{k}} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$$

The unit vector of in-plane polarization:

$$\boldsymbol{e}_{\text{IP}} = (\sin\phi, -\cos\phi, 0)$$

The unit vector of out-of-plane polarization:

$$\boldsymbol{e}_{\text{OP}} = (\cos\theta\cos\phi, \cos\theta\sin\phi, -\sin\theta)$$

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