# Supporting Information for Engineering valley quantum interference in anisotropic van der Waals heterostructures

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## I. PURCELL EFFECT AND DECAY RATE CALCULATIONS

When a quantum emitter is placed in a in-homogeneous environment, the lifetimes are altered due to change in density of states which is commonly referred as the Purcell effect. In the weak light matter coupling regime, the Purcell factor is given by [1],

$$\frac{\gamma}{\gamma_0} = 1 + \frac{6\pi}{|\vec{\mu}_p|\, k_0} \vec{\mu^*}_p \cdot \mathrm{Im}\left[\overline{\overline{G}}_S\left(\vec{r}_0, \vec{r}_0, \omega\right)\right] \cdot \vec{\mu}_p \tag{S.1}$$

where $\frac{\gamma}{\gamma_0}$, $\mu_0$, $\mathbf{E}_\mathrm{s}$, are normalized decay rate with free space, transition matrix element of exciton and scattered component of the dyadic Green's function. The scattered Green's function is given by Eq.3 of Ref.[2]

The reflection tensor $\mathbf{\Gamma}$ can be obtained from the transfer matrix formalism defined in Ref.[3]. The transfer matrix $\mathbf{M_{4\times4}}^b$ connects left and right sides electric field for a given interface:

$$\begin{pmatrix} E_\mathrm{R}^{+,s} \\ E_\mathrm{R}^{+,p} \\ E_\mathrm{R}^{-,s} \\ E_\mathrm{R}^{-,p} \end{pmatrix} = \mathbf{M_{4\times4}}^b \begin{pmatrix} E_\mathrm{L}^{+,s} \\ E_\mathrm{L}^{+,p} \\ E_\mathrm{L}^{-,s} \\ E_\mathrm{L}^{-,p} \end{pmatrix} \tag{S.2}$$

where the transfer matrix for atomic layer is given by,

$$\mathbf{M}^b_{4\times4}\left(n_\mathrm{R}, \vartheta_\mathrm{R}, n_\mathrm{L}, \vartheta_\mathrm{L}, \boldsymbol{\sigma}\right) = \begin{pmatrix} \mathbf{M}^b_{11} & \mathbf{M}^b_{12} \\ \mathbf{M}^b_{21} & \mathbf{M}^b_{22} \end{pmatrix} \tag{S.3}$$

$$\mathbf{M}^b_{11} = \frac{1}{2}\begin{pmatrix} f_+ - \frac{2\alpha\sigma_{yy}}{n_R\cos\vartheta_R} & -\frac{2\alpha\sigma_{yx}\cos\vartheta_\mathrm{L}}{n_R\cos\vartheta_R} \\ -\frac{2\alpha\sigma_{xy}}{n_R} & g_+ - \frac{2\alpha\sigma_{xx}\cos\vartheta_L}{n_R} \end{pmatrix} \tag{S.4}$$

$$\mathbf{M}^b_{12} = \frac{1}{2}\begin{pmatrix} f_- - \frac{2\alpha\sigma_{yy}}{n_R\cos\vartheta_R} & \frac{2\alpha\sigma_{yx}\cos\vartheta_L}{n_R\cos\vartheta_R} \\ -\frac{2\alpha\sigma_{xy}}{n_R} & g_- + \frac{2\alpha\sigma_{xx}\cos\vartheta_L}{n_R} \end{pmatrix} \tag{S.5}$$

$$\mathbf{M}^b_{21} = \frac{1}{2}\begin{pmatrix} f_- + \frac{2\alpha\sigma_{yy}}{n_\mathrm{R}\cos\vartheta_\mathrm{R}} & \frac{2\alpha\sigma_{yx}\cos\vartheta_\mathrm{L}}{n_\mathrm{R}\cos\vartheta_\mathrm{R}} \\ -\frac{2\alpha\sigma_{xy}}{n_\mathrm{R}} & g_- - \frac{2\alpha\sigma_{xx}\cos\vartheta_\mathrm{L}}{n_\mathrm{R}} \end{pmatrix} \tag{S.6}$$

$$\mathbf{M}^b_{22} = \frac{1}{2}\begin{pmatrix} f_+ + \frac{2\alpha\sigma_{yy}}{n_R\cos\vartheta_R} & -\frac{2\alpha\sigma_{yx}\cos\vartheta_L}{n_R\cos\vartheta_R} \\ -\frac{2\alpha\sigma_{xy}}{n_R} & g_+ + \frac{2\alpha\sigma_{xx}\cos\vartheta_L}{n_R} \end{pmatrix} \tag{S.7}$$

$$f_\pm = 1 \pm \frac{n_\mathrm{L}\cos\vartheta_\mathrm{L}}{n_\mathrm{R}\cos\vartheta_\mathrm{R}} \text{ and } g_\pm = \frac{n_\mathrm{L}}{n_\mathrm{R}} \pm \frac{\cos\vartheta_\mathrm{L}}{\cos\vartheta_\mathrm{R}}$$

Here $\alpha = \frac{1}{2\varepsilon_0 c}$, $\vartheta_R(\vartheta_L)$, $\mathrm{n}_R(\mathrm{n}_L)$ are refracted(incident) angle, refractive index of right(left) side respectively, and $\boldsymbol{\sigma}$ is surface conductivity. We need to note that these transfer matrix calculations are done in the x-z plane. In general, the wavevector can also have a y-component. To the avoid the complexity of equations, we do a rotation in x-y plane such that only x component exists and replace the conductivity tensor($\boldsymbol{\sigma}$) with the transformed conductivity($\boldsymbol{\sigma'}$) tensor

$$\boldsymbol{\sigma'} = \boldsymbol{R}^T \boldsymbol{\sigma} \boldsymbol{R} \tag{S.8}$$

where

$$R = \begin{pmatrix} \cos(\varphi) & -\sin(\varphi) \\ \sin(\varphi) & \cos(\varphi) \end{pmatrix} = \frac{1}{k_\rho} \begin{pmatrix} k_x & -k_y \\ k_y & k_x \end{pmatrix} \tag{S.9}$$

The transfer matrix for a dielectric strip of thickness d with wavevector k and propagation angle $\vartheta$ is given by

$$\mathbf{M}^{\text{dieletric}}(d) = \begin{pmatrix} \mathrm{e}^{\mathrm{i}kd\cos\vartheta} & 0 & 0 & 0 \\ 0 & \mathrm{e}^{\mathrm{i}kd\cos\vartheta} & 0 & 0 \\ 0 & 0 & \mathrm{e}^{-\mathrm{i}kd\cos\vartheta} & 0 \\ 0 & 0 & 0 & \mathrm{e}^{-\mathrm{i}kd\cos\vartheta} \end{pmatrix} \tag{S.10}$$

The total transfer matrix of the heterostructure is obtained by multiplying transfer matrix at each interface.The total transfer matrix is represented as.

$$\mathbf{M_{4\times4}}^{\text{total}} = \begin{pmatrix} \mathbf{M}_{11} & \mathbf{M}_{12} \\ \mathbf{M}_{21} & \mathbf{M}_{22} \end{pmatrix} \tag{S.11}$$

From the total transfer matrix, the reflection coefficient is given by Ref.[3]

$$\mathbf{\Gamma} = -\left(\mathbf{M}_{22}\right)^{-1} \mathbf{M}_{21} \tag{S.12}$$

The poles of reflection coefficients give the dispersion relation for the surface plasmons. Therefore the dispersion can be written as

$$\det(\mathbf{M}_{22}) = 0 \tag{S.13}$$

Eq. S.13 is generalized and can be used for any multilayer system. We can use this equation to calculate isofrequency surfaces and the dispersion relation for any multilayer system.

## II. QUANTUM MASTER EQUATION

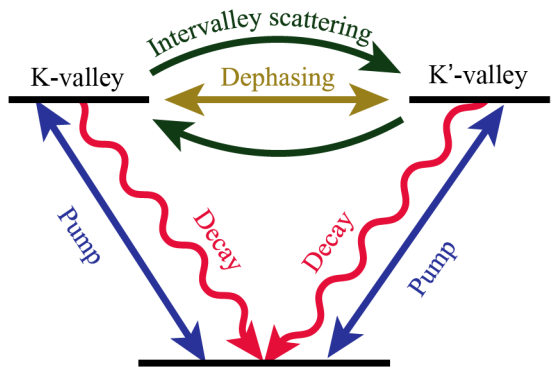


FIG. S1. V-level system

The excitons in K and K′ valleys can be modelled with three V-scheme system as shown in Fig.S1. The exciton in each valley can be modeled as a local in-plane circular dipole. From the quantum master equation given by Eq.1 of main text, the equations of motion of the excitonic population $\varrho_{\mathrm{KK,K'K'}}$ and the exciton intervalley coherence $\varrho_{\mathrm{KK'}}$ are given by

$$\frac{\partial}{\partial t}\varrho_{\mathrm{KK}} = -(\gamma_1 + R + \gamma_{\mathrm{s}})\varrho_{\mathrm{KK}} + \gamma_{\mathrm{s}}\varrho_{\mathrm{K'K'}} + R\varrho_{00} - (\alpha\varrho_{\mathrm{KK'}} + \mathrm{c.c.}) \tag{S.14}$$

$$\frac{\partial}{\partial t}\varrho_{\mathrm{K'K'}} = -(\gamma_2 + R + \gamma_{\mathrm{s}})\varrho_{\mathrm{K'K'}} + \gamma_{\mathrm{s}}\varrho_{\mathrm{KK}} + R\varrho_{00} - (\alpha\varrho_{\mathrm{K'K}} + \mathrm{c.c.}) \tag{S.15}$$

$$\frac{\partial}{\partial t}\varrho_{\mathrm{KK'}} = -(\gamma + R + \gamma_{\mathrm{s}} + \gamma_{\mathrm{dep}})\varrho_{\mathrm{KK'}} - \alpha^*\varrho_{\mathrm{K'K'}} - \alpha\varrho_{\mathrm{KK}} \tag{S.16}$$

$$\frac{\partial}{\partial t}\varrho_{00} = -\frac{\partial}{\partial t}\varrho_{\mathrm{KK}} - \frac{\partial}{\partial t}\varrho_{\mathrm{K'K'}} \tag{S.17}$$

where $\gamma_1 = \gamma_2 = \gamma = \frac{\gamma_{xx}+\gamma_{yy}}{2}$, $\alpha = \frac{\gamma_{xx}-\gamma_{yy}}{4} + i\delta_{KK'}$ and $\kappa = \frac{\gamma_{xx}-\gamma_{yy}}{2}$. Here $\gamma_{xx}$ ($\gamma_{yy}$) represents the decay rate of a dipole oriented along the x (y) axis, $\kappa$ represents the coupling constant and $\delta_{KK'}$ is the dipole-dipole frequency shift.

From the equations of motion of the excitonic population, the steady state DOLP is given by

$$DOLP(t \to \infty) = -\frac{Q}{\left(1 + \frac{R+\gamma_s+\gamma_{dep}}{\gamma}\right)} \tag{S.18}$$

where $Q = \frac{\kappa}{\gamma}$

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