Supporting Information for: TMDC-based topological nanospaser: single and double threshold behavior

Rupesh Ghimire,[*,†] Fatemeh Nematollahi,[*,†] Jhih-Sheng Wu,[*,‡] Vadym Apalkov,[*,†] and Mark .I. Stockman[*,†]

†*Center for Nano-Optics (CeNO) and Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA*

‡*Department of Photonics, College of Electrical and Computer Engineering, National Chiao Tung University, Hsinchu 30010, Taiwan*

E-mail: aaryun11@gmail.com; fnematallohi1@gsu.edu; b91202047@gmail.com; vapalkov@gsu.edu; mstockman@gsu.edu

Metallic Oblate Spheroid: Geometry and Modes

We consider an oblate spheroid, which in the Cartesian coordinate system is described by the following equation

$$\frac{x^2+y^2}{a^2}+\frac{z^2}{c^2}=1\ , \tag{S.1}$$

where a and c are semi-axes, $\varepsilon=\sqrt{1-\frac{c^2}{a^2}}$ is the eccentricity of the spheroid.

It is convenient to introduce the spheroidal coordinates, ξ, η and φ, which are related to the Cartesian coordinates, x, y and z through the following expressio:[1]

$$x = f\sqrt{\xi^2+1}\sqrt{1-\eta^2}\ \cos(\varphi), \tag{S.2}$$

$$y = f\sqrt{\xi^2+1}\sqrt{1-\eta^2}\ \sin(\varphi), \tag{S.3}$$

$$z = f\xi\eta, \tag{S.4}$$

where $0 \le \xi < \infty, \quad -1 \le \eta \le 1, \quad 0 \le \varphi < 2\pi$ and $f = \varepsilon a$.

Then the surface plasmon eigenmodes of the metal spheroid are described by the quasistatic equation[2]

$$\nabla\left[\theta(\mathbf{r})\nabla\phi_m\right] = s_{\mathrm{sp}}\nabla^2\phi_m, \tag{S.5}$$

where s_{sp} is the eigenvalue of the corresponding mode ϕ_m. Here $\theta(\mathbf{r})$ is the characteristic function that is 1 inside the metal and 0 elsewhere. For oblate spheroid, the eigenmodes are characterized by multipole quantum number l and magnetic quantum number m. For the relevant modes of topological nanospaser, the multipole quantum number is 1, $l = 1$. Then the corresponding eigenmodes are described by the following expressions

$$\phi_m = C_{\mathrm{N}} P_1^m(\eta)e^{im\phi}\begin{cases}\frac{P_1^m(i\xi)}{P_1^m(i\xi_0)}, & 0 < \xi < \xi_0,\\ \frac{Q_1^m(i\xi)}{Q_1^m(i\xi_0)}, & \xi_0 < \xi,\end{cases} \tag{S.6}$$

where $P_l^m(x)$ and $Q_l^m(x)$ are the Legendre functions of the first and second kind, respectively, and $\xi_0 = \frac{\sqrt{1-\varepsilon^2}}{\varepsilon}$. The constant C_{N} is determined by normalization condition,

$$\int_{\text{All Space}} |\nabla\phi(\mathbf{r})_m|^2 d^3\mathbf{r} = 1. \tag{S.7}$$

Due to axial symmetry of the nanospheroid, the corresponding eigenvalues, s_{sp}, do not depend on m. They can be also found from the following expression[3,4]

$$s_{\text{sp}} = \frac{\displaystyle\int_{\text{All Space}} \theta(\mathbf{r})|\nabla\phi_m(\mathbf{r})|^2 d^3\mathbf{r}}{\displaystyle\int_{\text{All Space}} |\nabla\phi_m(\mathbf{r})|^2 d^3\mathbf{r}}. \tag{S.8}$$

Using explicit expression (S.6) for ϕ_m, we derive the final equation for the eigenvalue

$$s_{\text{sp}} = \left. \frac{\frac{dP_1^m(x)}{dx}}{\frac{dP_1^m(x)}{dx} - \frac{P_1^m(x)}{Q_1^m(x)}\frac{dQ_1^m(x)}{dx}} \right|_{x=i\xi_0} . \tag{S.9}$$

To find the plasmon frequency, ω_{sp}, and the plasmon relaxation rate, γ_{sp}, we use the following relations:[3,4]

$$s_{\text{sp}} = \text{Re}[s(\omega_{\text{sp}})], \tag{S.10}$$

$$r_{\text{sp}} = \frac{\text{Im}[s(\omega_{\text{sp}})]}{s'_{\text{sp}}}, \qquad s'_{\text{sp}} \equiv \left. \frac{d\text{Re}[s(\omega)]}{d\omega} \right|_{\omega=\omega_{\text{sp}}}, \tag{S.11}$$

where the Bergman spectral parameter is defined as

$$s(\omega) = \frac{\epsilon_d}{\epsilon_d - \epsilon_m(\omega)} . \tag{S.12}$$

Here ϵ_d is the dielectric constant of surrounding medium, and $\epsilon_m(\omega)$ is the dielectric function of the metal (silver). In our computations, for silver, we use the dielectric function from Ref.[5]

TMDC Parameters

In our calculations the TMDC (MoS_2) monolayer is characterized by its bandgap and the dipole matrix elements between the conduction and valence bands at the K and K' points. To find these parameters we have used a three-band tight binding model.[6] The calculated values of the transition dipole matrix elements and the bandgap are given in Table S1. The

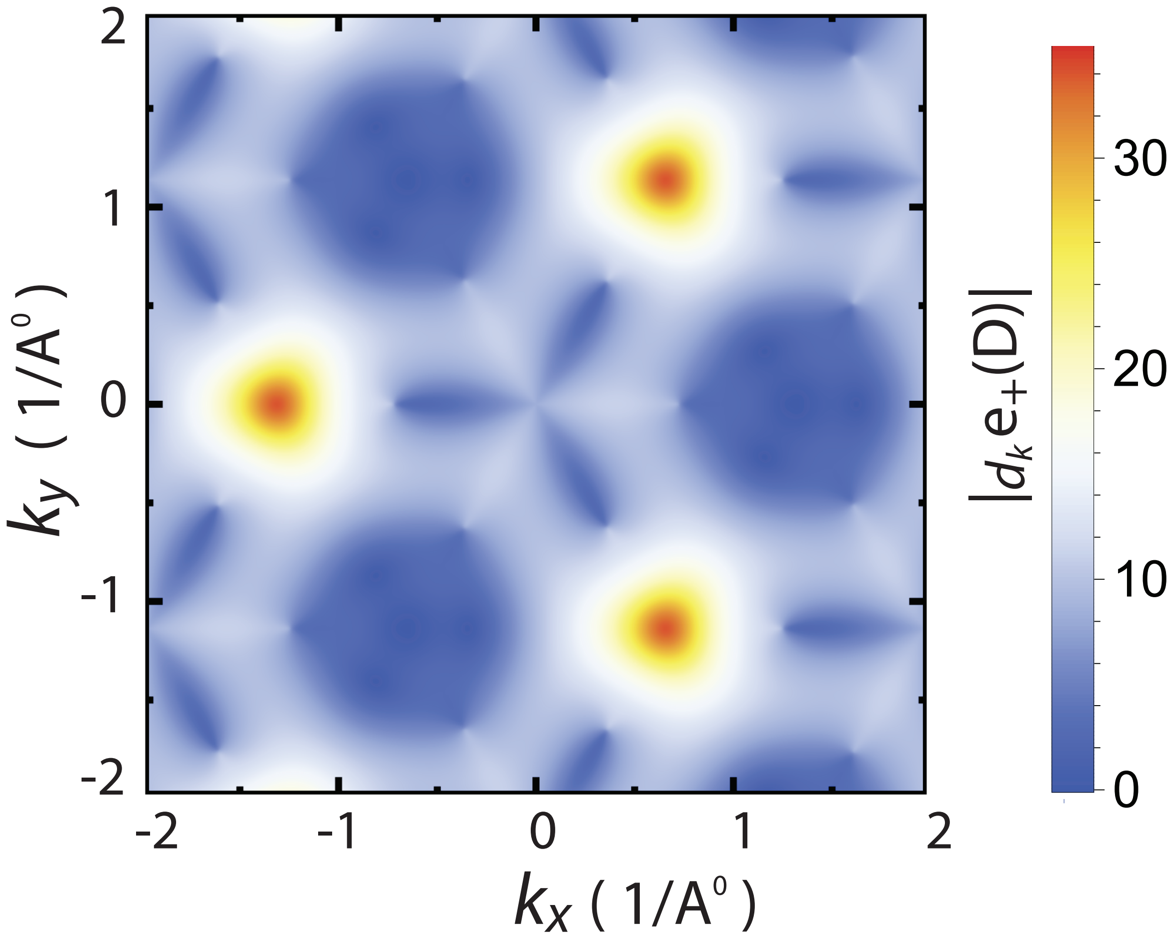

Fig. S1: Absolute value of the left-rotating chiral dipole component, $\mathbf{d}_- = \mathbf{e}_+\mathbf{d}$, in MoS_2

dipole matrix elements at the K and K' points are purely chiral. They are proportional to $\mathbf{e}_{\pm} = 2^{-1/2}\left(\mathbf{e}_x \pm i\mathbf{e}_y\right)$, where $\mathbf{e}_x$ and $\mathbf{e}_y$ are the Cartesian unit vectors. The plot of the absolute value of the chiral dipole, $|\mathbf{d}_{\pm}|$, where $\mathbf{d}_{\pm} = \mathbf{e}_{\pm}^{*}\mathbf{d}$, is shown in Fig. S1.

Table S1: Parameters employed in the calculations: Semi-principal axes of the spheroids, and the dipole matrix elements and band gaps of the TMDCs.

TMDC	Semi-principal axis c (nm)	Dipole elements (D) $\mathbf{d_K}$	Dipole elements (D) $\mathbf{d_{K'}}$	Band gap (eV)
MoS_2	1.20	$17.68\mathbf{e}_+$	$17.68\mathbf{e}_-$	1.66
$MoSe_2$	1.45	$19.23\mathbf{e}_+$	$19.23\mathbf{e}_-$	1.79
WSe_2	0.85	$18.38\mathbf{e}_+$	$18.38\mathbf{e}_-$	1.43
$MoTe_2$	1	$20.08\mathbf{e}_+$	$20.08\mathbf{e}_-$	1.53

Stationary solution

For the large radius of TMDC nanopatch and for the gain rate larger than the critical value, in the stationary regime, two types of plasmons, co-rotating ($m = -1$) and counter-rotating ($m = 1$), are generated. Although the co-rotating mode is more strongly coupled to the K valley of TMDC then the counter-rotating one, the number of generated counter-rotating plasmons is larger then the number of co-rotating ones, $N_{-1} > N_1$. To understand this relation we consider the following approximation for the Rabi frequency dependence on the position within the TMDC nanopatch: (i) the $m = 1$ plasmon mode is coupled to the K valley of TMDC at $r < r_0 \approx 12$ nm and to the K' valley of TMDC at $r_1 > r > r_0$; (ii) the $m = -1$ plasmon mode is coupled to the K' valley of TMDC at $r < r_0$ and to the K valley of TMDC at $r_1 > r > r_0$. Here r_1 is the radius of TMDC nanopatch. Under this approximation, there two uncoupled systems: system "1": $m = 1$ plasmons, K valley of TMDC at $r < r_0$, and K' valley of TMDC at $r_1 > r > r_0$; system "2": $m = -1$ plasmons, K' valley of TMDC at $r < r_0$, and K valley of TMDC at $r_1 > r > r_0$.

Then the stationary equations for system "1" become (see Eqs. (10)-(12) of the main text)

$$
\begin{aligned}
&\gamma_{\rm sp} a_1 = i\nu \int_{S_0} d^2\mathbf{r} \rho^*_K(\mathbf{r}) \tilde{\Omega}^*_{1,K}(\mathbf{r}) + i\nu \int_{S_1} d^2\mathbf{r} \rho^*_{K'}(\mathbf{r}) \tilde{\Omega}^*_{1,K'}(\mathbf{r}) \ , \\
&4{\rm Im}\left[\rho_K(\mathbf{r}) \tilde{\Omega}_{1,K}(\mathbf{r}) a_1\right] = g_K \left[1 - n_K(\mathbf{r})\right] - \gamma_{2K} \left[1 + n_K(\mathbf{r})\right], \\
&\Gamma_{12} \rho_K(\mathbf{r}) = i n_K(\mathbf{r}) \tilde{\Omega}^*_{1,K} a^*_1 \ , \\
&4{\rm Im}\left[\rho_{K'}(\mathbf{r}) \tilde{\Omega}_{1,K'}(\mathbf{r}) a_1\right] = -\gamma_{2K} \left[1 + n_{K'}(\mathbf{r})\right], \\
&\Gamma_{12} \rho_{K'}(\mathbf{r}) = i n_{K'}(\mathbf{r}) \tilde{\Omega}^*_{1,K'} a^*_1 \ .
\end{aligned}
$$

Here S_0 and S_1 are defined by the conditions $r < r_0$ and $r_0 < r < r_1$, respectively. We also take into account that only the K valley is pumped by a circularly polarized light. Similar system of equations can be written for the system "2"

$$
\begin{aligned}
&\gamma_{\rm sp} a_{-1} = i\nu \int_{S_1} d^2\mathbf{r} \rho^*_K(\mathbf{r}) \tilde{\Omega}^*_{-1,K}(\mathbf{r}) + i\nu \int_{S_0} d^2\mathbf{r} \rho^*_{K'}(\mathbf{r}) \tilde{\Omega}^*_{-1,K'}(\mathbf{r}) \ , \\
&4{\rm Im}\left[\rho_K(\mathbf{r}) \tilde{\Omega}_{-1,K}(\mathbf{r}) a_{-1}\right] = g_K \left[1 - n_K(\mathbf{r})\right] - \gamma_{2K} \left[1 + n_K(\mathbf{r})\right], \\
&\Gamma_{12} \rho_K(\mathbf{r}) = i n_K(\mathbf{r}) \tilde{\Omega}^*_{-1,K} a^*_{-1} \ , \\
&4{\rm Im}\left[\rho_{K'}(\mathbf{r}) \tilde{\Omega}_{-1,K'}(\mathbf{r}) a_{-1}\right] = -\gamma_{2K} \left[1 + n_{K'}(\mathbf{r})\right], \\
&\Gamma_{12} \rho_{K'}(\mathbf{r}) = i n_{K'}(\mathbf{r}) \tilde{\Omega}^*_{1,K'} a^*_{-1} \ .
\end{aligned}
$$

From the above systems of equation, assuming that $|\Omega_{m,\boldsymbol{\kappa}}|^2 a_m^2 \gg g_K \Gamma_1 2$, we obtain

$$
N_1 = |a_1|^2 = \frac{\nu}{4\gamma_{\rm sp}} \left[g_K S_0 - \gamma_{2K}(S_0 + S_1)\right] \tag{S.13}
$$

and

$$
N_{-1} = |a_{-1}|^2 = \frac{\nu}{4\gamma_{\rm sp}} \left[g_K S_1 - \gamma_{2K}(S_0 + S_1)\right], \tag{S.14}
$$

where S_0 and S_1 are the areas of the corresponding regions. Thus, above the threshold, the number of generated plasmons is proportional to $S_0 = \pi r_0^2$ for co-rotating plasmons and to $S_1 = \pi(r_1^2 - r_0^2)$ for counter-rotating plasmons. If $r_1 > \sqrt{2} r_0 \approx 16$ nm, i.e., $S_1 > S_0$, then the number of counter-rotating plasmons is larger than the number of co-rotating ones, $N_{-1} > N_1$.

Far-field radiation

The total dipole moment of the spaser can be expressed in the following form

$$\mathbf{d_{total}} = \mathbf{d_{metal}} + \mathbf{d_{tmdc}}, \tag{S.15}$$

where $\mathbf{d_{metal}}$ is the dipole moment of the metal nanospheroid and $\mathbf{d_{tmdc}}$ is the dipole moment of the TMDC nanoflake.

Dipole moment of the metal nanospheroid

The electric field inside the metal, which is produced by generated plasmon modes, both $m = 1$ and $m = -1$, is uniform and is given by the following expression

$$\mathbf{F}_m(\mathbf{r}, t) = -\sum_{m=1,-1} A_{\rm sp}(\nabla\phi_m \hat{a}_m e^{-i\omega t} + \nabla\phi_m^* \hat{a}_m^* e^{i\omega t}), \tag{S.16}$$

where

$$A_{\rm sp} = \sqrt{\frac{4\pi\hbar s(\omega)}{\epsilon_d s'(\omega)}} \tag{S.17}$$

and

$$\mathrm{s}(\omega) = \frac{\epsilon_d}{\epsilon_d - \epsilon_{m(\omega)}}, \tag{S.18}$$

Then the dipole moment of the metal nanospheroid can be found from the following expression

$$\mathbf{d_{metal}} = \int_V \mu \mathbf{F_m}(\mathbf{r}, \mathbf{t})\, dv \tag{S.19}$$

where

$$\mu = \frac{\mathrm{Re}[\epsilon_{metal} - \epsilon_d]}{4\pi}. \tag{S.20}$$

Taking into account that the electric field inside the metal is a constant, $E_0 = |\nabla \phi_m|$, we derive the following expressions for the dipole moment of the metal nanospheroid

$$\mathbf{d_{metal,x}} = -\mu \mathrm{A_{sp}} \mathrm{E_0} \mathrm{V} \Big((\hat{a}_1 e^{-i\omega t} + \hat{a}_1^* e^{i\omega t}) + (\hat{a}_{-1} e^{-i\omega t} + \hat{a}_{-1}^* e^{i\omega t}) \Big) \tag{S.21}$$

$$\mathbf{d_{metal,y}} = -\mu \mathrm{A_{sp}} \mathrm{E_0} \mathrm{V} \Big(i(\hat{a}_1 e^{-i\omega t} - \hat{a}_1^* e^{i\omega t}) - i(\hat{a}_{-1} e^{-i\omega t} - \hat{a}_{-1}^* e^{i\omega t}) \Big) \tag{S.22}$$

Dipole moment of TMDC monolayer

The density matrix of TMDC nanoflake has the following structure

$$\hat{\rho}_{\boldsymbol{\mathcal{K}}}(\mathbf{r}, t) = \begin{pmatrix} \rho^{(\mathrm{c})}_{\boldsymbol{\mathcal{K}}}(\mathbf{r}, t) & \rho_{\boldsymbol{\mathcal{K}}}(\mathbf{r}, t) e^{i\omega t} \\ \rho^*_{\boldsymbol{\mathcal{K}}}(\mathbf{r}, t) e^{-i\omega t} & \rho^{(\mathrm{v})}_{\boldsymbol{\mathcal{K}}}(\mathbf{r}, t) \end{pmatrix}. \tag{S.23}$$

where $\boldsymbol{\mathcal{K}}$ is the valley index, K or K'. The off-diagonal elements, i.e., coherences, determine the dipole moment of TMDC system

$$\mathbf{d_{tmdc}} = \sum_{\mathrm{S}} \sum_{\boldsymbol{\mathcal{K}}=\mathbf{K},\mathbf{K'}} (\rho_{\boldsymbol{\mathcal{K}}}(\mathbf{r}) \mathbf{d}_{\boldsymbol{\mathcal{K}}} e^{i\omega t} + \rho^*_{\boldsymbol{\mathcal{K}}}(\mathbf{r}) \mathbf{d}^*_{\boldsymbol{\mathcal{K}}} e^{-i\omega t}) + h.c., \tag{S.24}$$

where $\sum_{\mathrm{S}}$ is the sum (integral) over all points $\mathbf{r}$ of TMDC nanoflake.

The coherences satisfy the following stationary equation (see Eq. (12) of the main text)

$$[-i(\omega-\omega_{21})-\Gamma_{12}]\rho_{\mathcal{K}}(\mathbf{r})+in_{\mathcal{K}}(\mathbf{r})\sum_{m=1,-1}\tilde{\Omega}^{*}_{m,\mathcal{K}}(\mathbf{r})a^{*}_{m}=0, \tag{S.25}$$

where Γ_{12} is the polarization relaxation rate, $n_{\mathcal{K}}$ is the population inversion defined as

$$n_{\mathcal{K}}\equiv\rho^{(\mathrm{c})}_{\mathcal{K}}-\rho^{(\mathrm{v})}_{\mathcal{K}}\ , \tag{S.26}$$

and

$$\tilde{\Omega}_{m,\mathcal{K}}(\mathbf{r})=-\frac{1}{\hbar}A_{\mathrm{sp}}\nabla\phi_{m}(\mathbf{r})\mathbf{d}_{\mathcal{K}}\ . \tag{S.27}$$

From Eq. (S.25) we can find the stationary coherences of TMDC monolayer

$$\rho_{\mathcal{K}}(\mathbf{r})=-\frac{in_{\mathcal{K}}(\mathbf{r})\sum_{m=1,-1}\tilde{\Omega}^{*}_{m,\mathcal{K}}(\mathbf{r})a^{*}_{m}}{-(\omega-\omega_{21})+i\Gamma_{12}}. \tag{S.28}$$

We substitute Eq. (S.28) into Eq. (S.24) and obtain the following expression for the dipole moment of TMDC

$$\mathbf{d}_{\mathbf{tmdc}}=\mathrm{f}_{\mathbf{K}}\mathbf{d}_{\mathbf{K}}e^{i\omega t}+\mathrm{f}_{\mathbf{K}'}\mathbf{d}_{\mathbf{K}'}e^{i\omega t}+h.c., \tag{S.29}$$

where the following notations were introduced

$$\mathrm{f}_{\mathbf{K}}=-\nu\sum_{\mathrm{S}}\frac{in_{\mathbf{K}}(\mathbf{r})\sum_{m=1,-1}\tilde{\Omega}^{*}_{m,\mathbf{K}}a^{*}_{m}}{-(\omega-\Delta_{\mathrm{g}})+i\Gamma_{12}} \tag{S.30}$$

$$\mathrm{f}_{\mathbf{K}'}=-\nu\sum_{\mathrm{S}}\frac{in_{\mathbf{K}'}(\mathbf{r})\sum_{m=1,-1}\tilde{\Omega}^{*}_{m,\mathbf{K}'}a^{*}_{m}}{-(\omega-\Delta_{\mathrm{g}})+i\Gamma_{12}}. \tag{S.31}$$

Taking into account that $\mathbf{d}_{\mathbf{K}}=\mathrm{d}_0(1,i)$ and $\mathbf{d}_{\mathbf{K}'}=\mathrm{d}_0(1,-i)$ we obtain the x and y components of the dipole moment

$$\mathbf{d_{tmdc,x}} = \mathrm{f}_{\mathbf{K}}\mathrm{d}_0 e^{i\omega t} + \mathrm{f}_{\mathbf{K}'}\mathrm{d}_0 e^{i\omega t} + h.c. \tag{S.32}$$

$$\mathbf{d_{tmdc,y}} = i\mathrm{f}_{\mathbf{K}}\mathrm{d}_0 e^{i\omega t} - i\mathrm{f}_{\mathbf{K}'}\mathrm{d}_0 e^{i\omega t} + h.c. \tag{S.33}$$

Far field dipole radiation

The total dipole moment of the spaser system is the sum of the dipole moment of the metal nanospheroid and TMDC nanoflake. Its x and y components can be expressed as

$$\begin{aligned}\mathbf{d_{total,x}} = & -\mu \mathrm{A_{sp}} \mathrm{E}_0 \mathrm{V}\left(\hat{a}_1 e^{-i\omega t} + \hat{a}_{-1} e^{-i\omega t} + h.c.\right) \\ & + \left(\mathrm{f}_{\mathbf{K}}\mathrm{d}_0 e^{i\omega t} + \mathrm{f}_{\mathbf{K}'}\mathrm{d}_0 e^{i\omega t} + h.c.\right)\end{aligned} \tag{S.34}$$

$$\begin{aligned}\mathbf{d_{total,y}} = & -\mu \mathrm{A_{sp}} \mathrm{E}_0 \mathrm{V}\left((i\hat{a}_1 e^{-i\omega t} - i\hat{a}_{-1} e^{-i\omega t} + h.c.\right) \\ & + \left(i\mathrm{f}_{\mathbf{K}}\mathrm{d}_0 e^{i\omega t} - i\mathrm{f}_{\mathbf{K}'}\mathrm{d}_0 e^{i\omega t} + h.c.\right)\end{aligned} \tag{S.35}$$

These expressions have the following structure

$$\mathbf{d_{total,x}} = 2\mathrm{Re}\left[B_x e^{i\omega t}\right], \tag{S.36}$$

$$\mathbf{d_{total,y}} = 2\mathrm{Re}\left[B_y e^{i\omega t}\right], \tag{S.37}$$

where,

$$B_x = -\mu \mathrm{A_{sp}} \mathrm{E}_0 \mathrm{V}(\hat{a}_1^* + \hat{a}_{-1}^*) + \mathrm{f}_{\mathbf{K}}\mathrm{d}_0 + \mathrm{f}_{\mathbf{K}'}\mathrm{d}_0, \tag{S.38}$$

$$B_y = i\mu \mathrm{A_{sp}} \mathrm{E}_0 \mathrm{V}(\hat{a}_1^* - \hat{a}_{-1}^*) + i\mathrm{f}_{\mathbf{K}}\mathrm{d}_0 - i\mathrm{f}_{\mathbf{K}'}\mathrm{d}_0. \tag{S.39}$$

The total dipole moment of the system determines the far-field radiation of the spaser. The polarization of radiation is characterized by the x and y components of the far electric field, which are proportional to the corresponding components of the dipole moment, i.e., $\mathbf{d}_{\mathbf{total,x}}$ and $\mathbf{d}_{\mathbf{total,y}}$, while the total radiation power is given by the following expression

$$\begin{aligned} I &= \frac{4}{3}\left(\frac{\omega}{c_0}\right)^3 \frac{(\epsilon_{\mathrm{d}})^{1/2}}{\hbar}\langle|\mathbf{d}_{\mathbf{total}}|^2\rangle \\ &= \frac{8}{3}\left(\frac{\omega}{c_0}\right)^3 \frac{(\epsilon_{\mathrm{d}})^{1/2}}{\hbar}(|B_x|^2+|B_y|^2) \end{aligned} \tag{S.40}$$

where $\langle \ldots \rangle$ means the time average.

References

(1) M. Willatzen and L. C. L. Y. Voon. Oblate spheroidal coordinates. In *Separable Boundary-Value Problems in Physics*, pages 155–164. Wiley-VCH, Weinheim, Germany, 2011.

(2) M. I. Stockman, S. V. Faleev, and D. J. Bergman. Localization versus delocalization of surface plasmons in nanosystems: Can one state have both characteristics? *Phys. Rev. Lett.*, 87:167401–1–4, 2001.

(3) D. J. Bergman and M. I. Stockman. Surface plasmon amplification by stimulated emission of radiation: Quantum generation of coherent surface plasmons in nanosystems. *Phys. Rev. Lett.*, 90:027402–1–4, 2003.

(4) M. I. Stockman. The spaser as a nanoscale quantum generator and ultrafast amplifier. *Journal of Optics*, 12:024004–1–13, 2010.

(5) P. B. Johnson and R. W. Christy. Optical constants of noble metals. *Phys. Rev. B*, 6:4370–4379, 1972.

(6) G. B. Liu, W. Y. Shan, Y. G. Yao, W. Yao, and D. Xiao. Three-band tight-binding

model for monolayers of group-VIB transition metal dichalcogenides. *Phys. Rev. B*, 88:085433–1–10, 2013.