Supplementary information for "Canalized polaritons as virtual waveguides for nanoscale emitters"

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CONTENTS

S1. QUASI-ELECTROSTATIC APPROXIMATION

The quasi-electrostatic (or high-k) approximation applies when the electromagnetic field varies on a length scale much smaller than the free-space wavelength. This condition can be written as

$$\nabla \sim \frac{1}{l} \gg k_0 = \frac{2\pi}{\lambda_0}, \tag{S1}$$

where l is the characteristic length scale of spatial variation, defined by $|\mathbf{E}(\mathbf{r}+\mathbf{l}) - \mathbf{E}(\mathbf{r})| \sim |\mathbf{E}(\mathbf{r})|$, and k_0 and λ_0 are the free-space wavevector and wavelength, respectively.

To analyze the consequences of this condition, we introduce the scalar potential φ and vector potential $\mathbf{A}$,

$$\mathbf{E} = -\nabla\varphi - \frac{1}{c}\frac{\partial \mathbf{A}}{\partial t}, \tag{S2}$$

$$\mathbf{B} = \nabla \times \mathbf{A}. \tag{S3}$$

Substituting Eqs. (S2) and (S3) into the Maxwell equation

$$\nabla \times \mathbf{B} = \frac{4\pi}{c}\frac{\partial \mathbf{D}}{\partial t}, \tag{S4}$$

yields

$$\nabla \times \nabla \times \mathbf{A} = -\frac{4\pi}{c}\frac{\partial}{\partial t}\overleftrightarrow{\varepsilon}\left(\nabla\varphi + \frac{1}{c}\frac{\partial \mathbf{A}}{\partial t}\right). \tag{S5}$$

Using the condition in Eq. (S1), we estimate the relative magnitude of the terms:

$$|\nabla \times \nabla \times \mathbf{A}| \sim \frac{|\mathbf{A}|}{l^2} \gg k_0^2|\overleftrightarrow{\varepsilon}\mathbf{A}| \sim \left|\frac{1}{c}\frac{\partial}{\partial t}\overleftrightarrow{\varepsilon}\frac{1}{c}\frac{\partial \mathbf{A}}{\partial t}\right|. \tag{S6}$$

Thus, the term containing $\partial^2\mathbf{A}/\partial t^2$ can be neglected in Eq. (S5), leading to the estimate

$$|\mathbf{A}| \sim k_0 l|\varphi|. \tag{S7}$$

Consequently,

$$|\nabla\varphi| \sim \frac{|\varphi|}{l} \gg k_0^2 l|\varphi| \sim k_0|\mathbf{A}|, \tag{S8}$$

which justifies neglecting the time-derivative term in Eq. (S2). In addition, from Eq. (S4) one finds $\mathbf{B} \sim k_0 l\mathbf{D}$, implying $\mathbf{B} \ll \mathbf{E}$. Therefore, the electromagnetic field is fully determined by the scalar potential,

$$\mathbf{E} = -\nabla\varphi, \tag{S9}$$

and the problem reduces to an electrostatic one. The field distribution is then governed by

$$\nabla\overleftrightarrow{\varepsilon}\nabla\varphi = -4\pi\rho, \tag{S10}$$

where ρ is the external charge density.

We now consider plane-wave solutions in this approximation. For a potential of the form $\varphi = \varphi_0 e^{i\mathbf{k}\cdot\mathbf{r}}$, the electric field reads

$$\mathbf{E} = -\nabla\varphi = -i\varphi_0 \mathbf{k} e^{i\mathbf{k}\cdot\mathbf{r}}. \tag{S11}$$

Outside the layer, the field is evanescent and takes the form

$$\mathbf{E} \propto \begin{pmatrix} k_x \\ k_y \\ i\sqrt{k_x^2 + k_y^2} \end{pmatrix} e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel - kz}, \tag{S12}$$

while inside the anisotropic layer

$$\mathbf{E} \propto \begin{pmatrix} k_x \\ k_y \\ k_z \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{S13}$$

with the dispersion condition

$$\mathbf{k} \cdot \overleftrightarrow{\varepsilon} \mathbf{k} = 0. \tag{S14}$$

S2. CLASSICAL SOLUTION

Here we consider electromagnetic modes in a biaxial layer within the quasi-electrostatic approximation, i.e., assuming $k = |\mathbf{k}| \gg k_0 = \omega/c$, where $\mathbf{k}$ is the in-plane wavevector, ω is the angular frequency, and c is the speed of light. In this regime, only the electric field, $\mathbf{E} = -\nabla\varphi$, needs to be retained, while the magnetic field can be neglected. We describe the material using the Lorentz oscillator model, which accurately captures the optical response of polar dielectrics, including α-MoO_3 [1]. For simplicity, we retain a single phonon resonance along each crystallographic direction. Owing to the narrow spectral width of optical phonons, this approximation is valid in the frequency range of interest and ensures that the dispersion can be treated as a single-valued function $\omega = \omega(\mathbf{k})$.

We consider an anisotropic layer of thickness d, oriented perpendicular to the z-axis and placed between two isotropic, non-dispersive semi-infinite dielectrics with permittivities ε_1 and ε_2, i.e., $\varepsilon(z > d/2) = \varepsilon_1$ and $\varepsilon(z < -d/2) = \varepsilon_2$. The optical response of the anisotropic layer is modeled as a set of anisotropic harmonic oscillators with mass tensor $\overleftrightarrow{\mathbf{m}}$, resonance-frequency tensor $\overleftrightarrow{\omega}_0$, charge e, and concentration n. These oscillators are embedded in a background medium with non-dispersive anisotropic permittivity $\overleftrightarrow{\varepsilon}_\infty$. The corresponding parameters can be extracted from the measured dielectric function of α-MoO_3 [1], $\overleftrightarrow{\varepsilon}$:

$$\varepsilon_i(\omega) = \varepsilon_{\infty_i} + \frac{ne^2}{\varepsilon_0 m_i \left(\omega_{0i}^2 - \omega^2\right)}. \tag{S15}$$

where $i = x, y, z$. Since the permittivity tensor of α-MoO_3 is diagonal, we assume that all tensors, including $\overleftrightarrow{\mathbf{m}}$, $\overleftrightarrow{\omega}_0$, and $\overleftrightarrow{\varepsilon}_\infty$, are diagonal in the same basis.

Within the continuous-medium description, we introduce the oscillator displacement field $\mathbf{X}(\mathbf{r}, t)$. The Lorentz oscillator equation of motion reads

$$\overleftrightarrow{\mathbf{m}}\ddot{\mathbf{X}}(\mathbf{r}, t) + \overleftrightarrow{\mathbf{m}}\overleftrightarrow{\omega}_0^2\mathbf{X}(\mathbf{r}, t) = e\mathbf{E}(\mathbf{r}, t) \tag{S16}$$

where $\mathbf{X}(\mathbf{r}, t)$ is the displacement from equilibrium. The polarization density is then $\mathbf{P}(\mathbf{r}, t) = en\mathbf{X}(\mathbf{r}, t)$, and the corresponding charge density is $\rho(\mathbf{r}, t) = -\nabla\mathbf{P}(\mathbf{r}, t)$. In the quasi-electrostatic limit, Maxwell's equations reduce to

$$\nabla\mathbf{D}(\mathbf{r}, t) = \rho(\mathbf{r}, t), \tag{S17}$$

where the displacement field contains only the background permittivity, i.e., $\mathbf{D} = \varepsilon_0\overleftrightarrow{\varepsilon}_\infty\mathbf{E}$ inside the anisotropic layer and $\mathbf{D} = \varepsilon_0\varepsilon_{1,2}\mathbf{E}$ outside. The classical Hamiltonian of the coupled oscillator–field system is therefore

$$H = \int d^3\mathbf{r} \left(\frac{n}{2}\dot{\mathbf{X}}\overleftrightarrow{\mathbf{m}}\dot{\mathbf{X}} + \frac{n}{2}\mathbf{X}\overleftrightarrow{\mathbf{m}}\overleftrightarrow{\omega}_0^2\mathbf{X} - en\varphi\nabla\mathbf{X} - \frac{\varepsilon_0}{2}\nabla\varphi\overleftrightarrow{\varepsilon}_\infty\nabla\varphi\right). \tag{S18}$$

We seek solutions of Eqs. (S16) and (S17) in the form

$$\varphi_{\mathbf{k}}(\mathbf{r}, t) = e^{i\mathbf{kr} - i\omega t} \begin{cases} \left(\varphi_1 \cos k_z \frac{d}{2} + \varphi_2 \sin k_z \frac{d}{2}\right) e^{-k\left(z - \frac{d}{2}\right)}, & z > \frac{d}{2} \\ \varphi_1 \cos k_z z + \varphi_2 \sin k_z z, & -\frac{d}{2} < z \le \frac{d}{2} \\ \left(\varphi_1 \cos k_z \frac{d}{2} - \varphi_2 \sin k_z \frac{d}{2}\right) e^{k\left(z + \frac{d}{2}\right)}, & z \le -\frac{d}{2} \end{cases}. \tag{S19}$$

The corresponding electric field, $\mathbf{E_k}(\mathbf{r},z,t) = -\nabla\varphi_\mathbf{k}(\mathbf{r},z,t)$, is

$$\mathbf{E_k}(\mathbf{r},z,t) = \left(\varphi_1 \cos k_z \frac{d}{2} + \varphi_2 \sin k_z \frac{d}{2}\right) \begin{pmatrix} -ik_x \\ -ik_y \\ k \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r} - k\left(z-\frac{d}{2}\right) - i\omega t}, \qquad z > \frac{d}{2} \tag{S20}$$

$$\mathbf{E_k}(\mathbf{r},z,t) = \begin{pmatrix} -ik_x \left(\varphi_1 \cos k_z z + \varphi_2 \sin k_z z\right) \\ -ik_y \left(\varphi_1 \cos k_z z + \varphi_2 \sin k_z z\right) \\ k_z \left(\varphi_1 \sin k_z z - \varphi_2 \cos k_z z\right) \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r} - i\omega t}, \qquad -\frac{d}{2} < z \leq \frac{d}{2} \tag{S21}$$

$$\mathbf{E_k}(\mathbf{r},z,t) = \left(\varphi_1 \cos k_z \frac{d}{2} - \varphi_2 \sin k_z \frac{d}{2}\right) \begin{pmatrix} -ik_x \\ -ik_y \\ -k \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r} + k\left(z+\frac{d}{2}\right) - i\omega t}, \qquad z \leq -\frac{d}{2} \tag{S22}$$

The charge density distribution, $\rho(\mathbf{r},t) = \nabla\mathbf{D}(\mathbf{r},t)$, is

$$\begin{aligned} \rho(\mathbf{r},t) = \varepsilon_0 \left(\varepsilon_{\infty_x} k_x^2 + \varepsilon_{\infty_y} k_y^2 + \varepsilon_{\infty_z} k_z^2\right) \left(\varphi_1 \cos k_z z + \varphi_2 \sin k_z z\right) \theta\left(\frac{d^2}{4} - z^2\right) e^{i\mathbf{k}\cdot\mathbf{r} - i\omega t} + \\ + \varepsilon_0 \left[\varepsilon_1 k \left(\varphi_1 \cos \frac{k_z d}{2} + \varphi_2 \sin \frac{k_z d}{2}\right) - \varepsilon_{\infty_z} k_z \left(\varphi_1 \sin \frac{k_z d}{2} - \varphi_2 \cos \frac{k_z d}{2}\right)\right] \delta\left(z - \frac{d}{2}\right) e^{i\mathbf{k}\cdot\mathbf{r} - i\omega t} - \\ - \varepsilon_0 \left[\varepsilon_{\infty_z} k_z \left(\varphi_1 \sin \frac{k_z d}{2} + \varphi_2 \cos \frac{k_z d}{2}\right) - \varepsilon_2 k \left(\varphi_1 \cos \frac{k_z d}{2} - \varphi_2 \sin \frac{k_z d}{2}\right)\right] \delta\left(z + \frac{d}{2}\right) e^{i\mathbf{k}\cdot\mathbf{r} - i\omega t}, \end{aligned} \tag{S23}$$

where $\theta(x)$ is the Heaviside step function and $\delta(x)$ is the Dirac delta function. The oscillator displacement field is

$$\mathbf{X_k}(\mathbf{r},z,t) = \begin{pmatrix} \frac{-iek_x}{m_x\left(\omega_{0x}^2 - \omega^2\right)} \left(\varphi_1 \cos k_z z + \varphi_2 \sin k_z z\right) \\ \frac{-iek_y}{m_y\left(\omega_{0y}^2 - \omega^2\right)} \left(\varphi_1 \cos k_z z + \varphi_2 \sin k_z z\right) \\ \frac{ek_z}{m_z\left(\omega_{0z}^2 - \omega^2\right)} \left(\varphi_1 \sin k_z z - \varphi_2 \cos k_z z\right) \end{pmatrix} \theta\left(\frac{d^2}{4} - z^2\right) e^{i\mathbf{k}\cdot\mathbf{r} - i\omega t}. \tag{S24}$$

Applying the condition $\rho(\mathbf{r},t) = -\nabla\mathbf{P}(\mathbf{r},t)$ inside the layer yields

$$\varepsilon_x k_x^2 + \varepsilon_y k_y^2 + \varepsilon_z k_z^2 = 0 \tag{S25}$$

where ε_i is given by Eq. (S15). At the interfaces, the boundary conditions become:

$$\begin{cases} \varepsilon_1 k \left(\varphi_1 \cos k_z \frac{d}{2} + \varphi_2 \sin k_z \frac{d}{2}\right) = \varepsilon_z k_z \left(\varphi_1 \sin k_z \frac{d}{2} - \varphi_2 \cos k_z \frac{d}{2}\right) \\ \varepsilon_2 k \left(\varphi_1 \cos k_z \frac{d}{2} - \varphi_2 \sin k_z \frac{d}{2}\right) = \varepsilon_z k_z \left(\varphi_1 \sin k_z \frac{d}{2} + \varphi_2 \cos k_z \frac{d}{2}\right) \end{cases}. \tag{S26}$$

Solving system (S26) gives the relation between φ_1 and φ_2,

$$\varphi_2 = \varphi_1 \frac{(\varepsilon_2 - \varepsilon_1)k(1 + \cos k_z d)}{2\varepsilon_z k_z(1 + \cos k_z d) + (\varepsilon_1 + \varepsilon_2)k \sin k_z d} \tag{S27}$$

and the dispersion relation

$$\tan k_z d = \frac{(\varepsilon_1 + \varepsilon_2)\varepsilon_z k_z k}{\varepsilon_z^2 k_z^2 - \varepsilon_1 \varepsilon_2 k^2}. \tag{S28}$$

Equation (S28) has the explicit solution [2]

$$k = \frac{\xi}{d}\left(\arctan\frac{\xi\varepsilon_1}{\varepsilon_z} + \arctan\frac{\xi\varepsilon_2}{\varepsilon_z} + \pi l\right), \quad l \in \mathcal{N}, \tag{S29}$$

where $\xi = \sqrt{\frac{-\varepsilon_z}{\varepsilon_x \cos^2\phi + \varepsilon_y \sin^2\phi}}$, and ϕ is the in-plane angle defining the direction of $\mathbf{k}$. In what follows, we consider the solution with the lowest positive k, corresponding to $l = 0$ or $l = 1$.

A. Dispersion relation of canalized polaritons

Canalization occurs when the IFC becomes nearly flat, so that the group velocity, which is normal to the IFC, is aligned along the canalization direction for all relevant modes. In a single anisotropic layer, this direction must coincide with a principal crystal axis by symmetry. Without loss of generality, we consider canalization along the x-axis. Ideally, the IFC then consists of two straight lines perpendicular to the x-axis; in practice, it is sufficient that $k_x \approx \mathrm{const}(\phi)$, as in Fig. 2 of the main text. Multiplying Eq. (S29) by $\cos\phi$, we obtain

$$k_x = \frac{1}{d}\sqrt{\frac{-\varepsilon_z \cos^2\phi}{\varepsilon_x \cos^2\phi + \varepsilon_y \sin^2\phi}}\left(\arctan\frac{\xi\varepsilon_1}{\varepsilon_z} + \arctan\frac{\xi\varepsilon_2}{\varepsilon_z} + \pi l\right), \quad l \in \mathcal{N}, \tag{S30}$$

If $\varepsilon_y = 0$, the prefactor becomes independent of ϕ. In the idealized case $\varepsilon_1 = -\varepsilon_2$, canalization follows directly, and

$$k_x = \frac{\pi l}{d}\sqrt{\frac{-\varepsilon_z}{\varepsilon_x}}, \quad l = 1, 2, \ldots \tag{S31}$$

In practice, however, the condition $\varepsilon_1 = -\varepsilon_2$ cannot be satisfied exactly: a negative permittivity always has a positive imaginary part, and even a nanometer-scale air gap between the anisotropic layer and the substrate modifies the effective substrate response. Nevertheless, approximate canalization remains possible. If $\varepsilon_1 + \varepsilon_2 < 0$, the fundamental mode corresponds to $l = 1$, and the canalization condition is approximately satisfied when both arctangent terms are small enough to be neglected, recovering Eq. (S31). Since the tensor components of anisotropic materials are typically large, $\varepsilon_{x,y,z} \gg 1$, this requirement can be fulfilled even for a substrate with $|\varepsilon_2| \sim 1$.

A different and experimentally relevant regime occurs for a substrate with large negative permittivity, $\varepsilon_2 \to -\infty$, as for a metallic substrate in the infrared and lower-frequency ranges, which can be approximated as a perfect electric conductor. In this limit, the second arctangent term approaches $-\pi/2$ and becomes independent of ϕ. If, in addition, $\sqrt{-\varepsilon_x \varepsilon_z} \gg \varepsilon_1$, the first arctangent term is negligible and canalization is achieved. The resulting dispersion relation is

$$k_x = \frac{\pi}{d}\left(l - \frac{1}{2}\right)\sqrt{\frac{-\varepsilon_z}{\varepsilon_x}}, \quad l = 1, 2, ... \tag{S32}$$

S3. PHONON POLARITON HAMILTONIAN

Note that $\mathbf{X}$ and $n\overleftrightarrow{\mathbf{m}}\dot{\mathbf{X}}$ are canonical variables of the system described by the Hamiltonian (S18). We introduce bosonic annihilation and creation operators, $a_{\mathbf{k}}$ and $a_{\mathbf{k}}^{+}$, satisfying

$$\left[a_{\mathbf{k}}, a_{\mathbf{k}'}^{+}\right] = \delta(\mathbf{k} - \mathbf{k}'), \tag{S33}$$

and construct the displacement operator as

$$\hat{\mathbf{X}}(\mathbf{r}, z, t) = \int \frac{d^2\mathbf{k}}{2\pi} \begin{pmatrix} \frac{-iek_x}{m_x\left(\omega_{0x}^2 - \omega^2\right)} \left(\varphi_1(\mathbf{k}) \cos k_z z + \varphi_2(\mathbf{k}) \sin k_z z\right) \\ \frac{-iek_y}{m_y\left(\omega_{0y}^2 - \omega^2\right)} \left(\varphi_1(\mathbf{k}) \cos k_z z + \varphi_2(\mathbf{k}) \sin k_z z\right) \\ \frac{ek_z}{m_z\left(\omega_{0z}^2 - \omega^2\right)} \left(\varphi_1(\mathbf{k}) \sin k_z z - \varphi_2(\mathbf{k}) \cos k_z z\right) \end{pmatrix} \theta\left(\frac{d^2}{4} - z^2\right) a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r} - i\omega t} + h.c. \tag{S34}$$

where $\varphi_2(\mathbf{k})$ is determined by Eq. (S27), while $\varphi_1(\mathbf{k})$ is fixed by the canonical commutation relations. We assume $\varphi_{1,2}(\mathbf{k})$ to be real and even functions of momentum, i.e., $\varphi_{1,2}(\mathbf{k}) = \varphi_{1,2}(-\mathbf{k})$.

Because the layer is finite along the z-axis, the boundary conditions uniquely determine the out-of-plane field profile for a given $\mathbf{k}$. The system is therefore effectively two-dimensional: the in-plane wavevector $\mathbf{k}$ remains in momentum space, while the in-plane position $\mathbf{r}$ is treated in real space. We integrate the commutation relation over the z-axis:

$$\int_{-\frac{d}{2}}^{\frac{d}{2}} dz \left[\hat{\mathbf{X}}(\mathbf{r}, z), n\overleftrightarrow{\mathbf{m}}\hat{\dot{\mathbf{X}}}(\mathbf{r}', z)\right] = i\hbar\delta\left(\mathbf{r} - \mathbf{r}'\right). \tag{S35}$$

Substituting Eq. (S34) into Eq. (S35) yields

$$d\left(\varphi_1^2+\varphi_2^2\right)\left(\frac{ne^2k_x^2\omega}{m_x\left(\omega_{0x}^2-\omega^2\right)^2}+\frac{ne^2k_y^2\omega}{m_y\left(\omega_{0y}^2-\omega^2\right)^2}+\frac{ne^2k_z^2\omega}{m_z\left(\omega_{0z}^2-\omega^2\right)^2}\right)+$$
$$+\frac{\varphi_1^2-\varphi_2^2}{k_z}\sin k_zd\left(\frac{ne^2k_x^2\omega}{m_x\left(\omega_{0x}^2-\omega^2\right)^2}+\frac{ne^2k_y^2\omega}{m_y\left(\omega_{0y}^2-\omega^2\right)^2}-\frac{ne^2k_z^2\omega}{m_z\left(\omega_{0z}^2-\omega^2\right)^2}\right)=\hbar \quad \text{(S36)}$$

In terms of the dielectric permittivity, this condition can be rewritten as

$$\left(\varphi_1^2+\varphi_2^2\right)\left(\frac{d\varepsilon_x}{d\omega}k_x^2+\frac{d\varepsilon_y}{d\omega}k_y^2+\frac{d\varepsilon_z}{d\omega}k_z^2\right)+\left(\varphi_1^2-\varphi_2^2\right)\frac{\sin k_zd}{k_zd}\left(\frac{d\varepsilon_x}{d\omega}k_x^2+\frac{d\varepsilon_y}{d\omega}k_y^2-\frac{d\varepsilon_z}{d\omega}k_z^2\right)=\frac{2\hbar}{\varepsilon_0d} \quad \text{(S37)}$$

Equations (S37) and (S27) together determine φ_1 and φ_2.

The operators $\hat{\varphi}(\mathbf{r},z,t)$, $\hat{\rho}(\mathbf{r},z,t)$, and $\hat{\mathbf{E}}(\mathbf{r},z,t)$ are constructed in the same way. In particular,

$$\hat{\mathbf{E}}(\mathbf{r},z,t)=\int\frac{d^2\mathbf{k}}{2\pi}\left\{\theta\left(z-\frac{d}{2}\right)\left(\varphi_1\cos k_z\frac{d}{2}+\varphi_2\sin k_z\frac{d}{2}\right)\begin{pmatrix}-ik_x\\-ik_y\\k\end{pmatrix}e^{-k\left(z-\frac{d}{2}\right)}+\right.$$
$$+\theta\left(\frac{d^2}{4}-z^2\right)\begin{pmatrix}-ik_x\left(\varphi_1\cos k_zz+\varphi_2\sin k_zz\right)\\-ik_y\left(\varphi_1\cos k_zz+\varphi_2\sin k_zz\right)\\k_z\left(\varphi_1\sin k_zz-\varphi_2\cos k_zz\right)\end{pmatrix}+$$
$$\left.+\theta\left(-z-\frac{d}{2}\right)\left(\varphi_1\cos k_z\frac{d}{2}-\varphi_2\sin k_z\frac{d}{2}\right)\begin{pmatrix}-ik_x\\-ik_y\\-k\end{pmatrix}e^{k\left(z+\frac{d}{2}\right)}\right\}a_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}-i\omega t}+h.c. \quad \text{(S38)}$$

The PhP Hamiltonian is then

$$\hat{H}_{PhP}=\int d^2\mathbf{r}\int dz\left(\frac{n}{2}\hat{\dot{\mathbf{X}}}\overleftrightarrow{\mathbf{m}}\hat{\dot{\mathbf{X}}}+\frac{n}{2}\hat{\mathbf{X}}\overleftrightarrow{\mathbf{m}}\overleftrightarrow{\omega}_0^2\hat{\mathbf{X}}+\hat{\varphi}\hat{\rho}-\frac{\varepsilon_0}{2}\hat{\mathbf{E}}\overleftrightarrow{\varepsilon}_\infty\hat{\mathbf{E}}\right). \quad \text{(S39)}$$

To carry out the integration over $d^2\mathbf{r}$, we use

$$\int d^2\mathbf{r}e^{i(\mathbf{k}\pm\mathbf{k}')\cdot\mathbf{r}}=4\pi^2\delta\left(\mathbf{k}\pm\mathbf{k}'\right) \quad \text{(S40)}$$

Performing the integration over $d^2\mathbf{k}'$, we obtain

$$\hat{H}_{PhP} = \int d^2\mathbf{k} \left\{ \frac{1}{2} \int\limits_{-\frac{d}{2}}^{\frac{d}{2}} dz \left[\varepsilon_0 \left(\varepsilon_x k_x^2 + \varepsilon_y k_y^2 + 2\varepsilon_{\infty_z} k_z^2 \right) \left(\frac{\varphi_1^2 + \varphi_2^2}{2} + \frac{\varphi_1^2 - \varphi_2^2}{2} \cos 2k_z z \right) + \right.\right.$$
$$\left. + \varepsilon_0 \left(\varepsilon_z - 2\varepsilon_{\infty_z} \right) k_z^2 \left(\frac{\varphi_1^2 + \varphi_2^2}{2} - \frac{\varphi_1^2 - \varphi_2^2}{2} \cos 2k_z z \right) \right] + \frac{ne^2 k_z \left(\varphi_1^2 - \varphi_2^2 \right)}{m_z \left(\omega_{0z}^2 - \omega^2 \right)} \sin k_z d -$$
$$-\varepsilon_0 \varepsilon_2 k^2 \left(\varphi_1 \cos k_z \frac{d}{2} - \varphi_2 \sin k_z \frac{d}{2} \right)^2 \int\limits_{-\infty}^{-\frac{d}{2}} dz \left[e^{2k\left(z + \frac{d}{2}\right)} \right] - \varepsilon_0 \varepsilon_1 k^2 \left(\varphi_1 \cos k_z \frac{d}{2} + \varphi_2 \sin k_z \frac{d}{2} \right)^2 \times$$
$$\left. \times \int\limits_{\frac{d}{2}}^{\infty} dz \left[e^{-2k\left(z - \frac{d}{2}\right)} \right] \right\} a_{\mathbf{k}} a_{\mathbf{k}} e^{-2i\omega t} + \int d^2\mathbf{k} \left\{ \frac{1}{2} \int\limits_{-\frac{d}{2}}^{\frac{d}{2}} dz \left[\varepsilon_0 \left(\varepsilon_x k_x^2 + \frac{2ne^2 k_x^2 \omega^2}{\varepsilon_0 m_x \left(\omega_{0x}^2 - \omega^2 \right)^2} + \varepsilon_y k_y^2 + \right.\right.\right.$$
$$\left. + \frac{2ne^2 k_y^2 \omega^2}{\varepsilon_0 m_y \left(\omega_{0y}^2 - \omega^2 \right)^2} + 2\varepsilon_{\infty_z} k_z^2 \right) \left(\frac{\varphi_1^2 + \varphi_2^2}{2} + \frac{\varphi_1^2 - \varphi_2^2}{2} \cos 2k_z z \right) + \varepsilon_0 \left(\frac{2ne^2 \omega^2}{\varepsilon_0 m_z \left(\omega_{0z}^2 - \omega^2 \right)^2} + \varepsilon_z - \right.$$
$$\left.\left. - 2\varepsilon_{\infty_z} \right) k_z^2 \left(\frac{\varphi_1^2 + \varphi_2^2}{2} - \frac{\varphi_1^2 - \varphi_2^2}{2} \cos 2k_z z \right) \right] + \frac{ne^2 k_z \left(\varphi_1^2 - \varphi_2^2 \right)}{m_z \left(\omega_{0z}^2 - \omega^2 \right)} \sin k_z d - \varepsilon_0 \varepsilon_2 k^2 \left(\varphi_1 \cos k_z \frac{d}{2} - \right.$$
$$\left. \left. - \varphi_2 \sin k_z \frac{d}{2} \right)^2 \int\limits_{-\infty}^{-\frac{d}{2}} dz \left[e^{2k\left(z + \frac{d}{2}\right)} \right] - \varepsilon_0 \varepsilon_1 k^2 \left(\varphi_1 \cos k_z \frac{d}{2} + \varphi_2 \sin k_z \frac{d}{2} \right)^2 \int\limits_{\frac{d}{2}}^{\infty} dz \left[e^{-2k\left(z - \frac{d}{2}\right)} \right] \right\} \times$$
$$\times a_{\mathbf{k}} a_{\mathbf{k}}^{+} + h.c. \quad \text{(S41)}$$

where we have already used Eq. (S15) and the identity $\int\limits_{-\frac{d}{2}}^{\frac{d}{2}} \sin\left(2k_z z\right) dz = 0$. Performing the integration over dz and using Eq. (S25), we arrive at

$$\hat{H}_{PhP} = \int d^2\mathbf{k} \frac{\varepsilon_0}{2} \left[\varepsilon_z k_z \left(\varphi_1^2 - \varphi_2^2 \right) \sin k_z d - (\varepsilon_1 + \varepsilon_2) k \left(\frac{\varphi_1^2 + \varphi_2^2}{2} + \frac{\varphi_1^2 - \varphi_2^2}{2} \cos k_z d \right) - \right.$$
$$\left. - (\varepsilon_1 - \varepsilon_2) k \varphi_1 \varphi_2 \sin k_z d \right] \left[a_{\mathbf{k}} a_{\mathbf{k}} e^{-2i\omega t} + a_{\mathbf{k}} a_{\mathbf{k}}^{+} + h.c. \right] + \int d^2\mathbf{k} \left[\left(\frac{ne^2 k_x^2 \omega^2}{m_x \left(\omega_{0x}^2 - \omega^2 \right)^2} + \right.\right.$$
$$\left. + \frac{ne^2 k_y^2 \omega^2}{m_y \left(\omega_{0y}^2 - \omega^2 \right)^2} \right) \left(d \frac{\varphi_1^2 + \varphi_2^2}{2} + \frac{\varphi_1^2 - \varphi_2^2}{2k_z} \sin k_z d \right) + \frac{ne^2 k_z^2 \omega^2}{m_z \left(\omega_{0z}^2 - \omega^2 \right)^2} \left(d \frac{\varphi_1^2 + \varphi_2^2}{2} - \right.$$
$$\left.\left. - \frac{\varphi_1^2 - \varphi_2^2}{2k_z} \sin k_z d \right) \right] \left[a_{\mathbf{k}} a_{\mathbf{k}}^{+} + a_{\mathbf{k}}^{+} a_{\mathbf{k}} \right]. \quad \text{(S42)}$$

Substituting φ_2 from Eq. (S27) and $\tan k_z d$ from Eq. (S28), one finds that the first term in Eq. (S42) vanishes, as required for a time-independent Hamiltonian. Using the commutation relation (S33) and shifting the zero of energy to the ground state, we obtain

$$\hat{H}_{PhP} = \int d^2\mathbf{k} \left[d\left(\varphi_1^2 + \varphi_2^2\right) \left(\frac{ne^2k_x^2\omega^2}{m_x\left(\omega_{0x}^2 - \omega^2\right)^2} + \frac{ne^2k_y^2\omega^2}{m_y\left(\omega_{0y}^2 - \omega^2\right)^2} + \frac{ne^2k_z^2\omega^2}{m_z\left(\omega_{0z}^2 - \omega^2\right)^2} \right) + \right.$$
$$\left. \frac{\varphi_1^2 - \varphi_2^2}{k_z} \sin k_z d \left(\frac{ne^2k_x^2\omega^2}{m_x\left(\omega_{0x}^2 - \omega^2\right)^2} + \frac{ne^2k_y^2\omega^2}{m_y\left(\omega_{0y}^2 - \omega^2\right)^2} - \frac{ne^2k_z^2\omega^2}{m_z\left(\omega_{0z}^2 - \omega^2\right)^2} \right) \right] a_{\mathbf{k}}^+ a_{\mathbf{k}} \quad \text{(S43)}$$

Comparing Eq. (S43) with Eq. (S36) finally gives the phonon-polariton Hamiltonian

$$\hat{H}_{PhP} = \int d^2\mathbf{k}\hbar\omega(\mathbf{k}) a_{\mathbf{k}}^+ a_{\mathbf{k}}. \quad \text{(S44)}$$

S4. LOSSES OF POLARITONIC MODES

In a real system, material losses lead to attenuation of the modes, which makes the dielectric permittivity complex:

$$\varepsilon_i(\omega) = \varepsilon_{\infty_i} + \frac{ne^2}{\varepsilon_0 m_i\left(\omega_{0i}^2 - \omega^2 - i\gamma_i\omega\right)}. \quad \text{(S45)}$$

This is equivalent to modifying the classical oscillator equation of motion as

$$\overleftrightarrow{\mathbf{m}}\ddot{\mathbf{X}}(t) + \overleftrightarrow{\gamma}\overleftrightarrow{\mathbf{m}}\dot{\mathbf{X}}(t) + \overleftrightarrow{\mathbf{m}}\overleftrightarrow{\omega}_0^2\mathbf{X}(t) = e\mathbf{E}(t). \quad \text{(S46)}$$

In the quantum description, such damping can be modeled by coupling the system to a bath consisting of a continuum of harmonic oscillators.

A. One-oscillator Hamiltonian

We begin with a single one-dimensional oscillator, whose homogeneous classical equation of motion is

$$\ddot{x}(t) + \gamma\dot{x}(t) + \omega_0^2 x(t) = 0. \quad \text{(S47)}$$

For weak damping, $\omega_0 \gg \gamma$, the solution can be written as

$$x(t) = x(0)e^{-\frac{\gamma}{2}t + i\omega_0 t}, \quad \text{(S48)}$$

so that the oscillator energy decays as $e^{-\gamma t}$. In the quantum picture, this corresponds to $\langle\psi(t)|\hat{H}_0|\psi(t)\rangle = \hbar\omega_0 e^{-\gamma t}$, where $\hat{H}_0 = \hbar\omega_0 a^+ a$ and $|\psi(0)\rangle = |1\rangle = a^+|0\rangle$.

A Hamiltonian producing this behavior can be written as

$$\hat{H} = \hbar\omega_0 a^+ a + \hbar \int\limits_0^\infty d\omega \left(\omega a_\omega^+ a_\omega + g(\omega) a a_\omega^+ + g^*(\omega) a^+ a_\omega\right). \tag{S49}$$

We seek the solution of the Schrödinger equation in the form

$$|\psi(t)\rangle = c(t)|1,0\rangle + \int\limits_0^\infty d\omega b_\omega(t)|0,1_\omega\rangle. \tag{S50}$$

with $c(0) = 1$ and $b_\omega(0) = 0$. Introducing $\tilde{c}(t) = c(t)e^{i\omega_0 t}$ and $\tilde{b}_\omega(t) = b_\omega(t)e^{i\omega t}$, we obtain

$$\dot{\tilde{c}}(t) = -i \int\limits_0^\infty d\omega g^*(\omega)\tilde{b}_\omega(t) e^{-i(\omega-\omega_0)t}, \tag{S51}$$

$$\dot{\tilde{b}}_\omega(t) = -ig(\omega)\tilde{c}(t)e^{-i(\omega_0-\omega)t}. \tag{S52}$$

Integrating Eq. (S52) and substituting the result into Eq. (S51) gives

$$\dot{\tilde{c}}(t) = -\int\limits_0^\infty d\omega |g(\omega)|^2 \int\limits_0^t dt' \tilde{c}(t') e^{-i(\omega-\omega_0)(t-t')}. \tag{S53}$$

If $g(\omega) = g = \text{const}$, Eq. (S53) yields

$$\dot{\tilde{c}}(t) = -|g|^2 \int\limits_0^\infty d\omega \int\limits_0^t dt' \tilde{c}(t') e^{-i(\omega-\omega_0)(t-t')} \approx -\pi g^2 \tilde{c}(t). \tag{S54}$$

Therefore, $|c(t)|^2 = e^{-2\pi|g|^2 t}$, which reproduces the classical decay if we choose $g = i\sqrt{\frac{\gamma}{2\pi}}$. Using the standard expressions

$$a = \frac{m\omega_0\hat{x} + i\hat{p}}{\sqrt{2\hbar m\omega_0}}, \qquad a^+ = \frac{m\omega_0\hat{x} - i\hat{p}}{\sqrt{2\hbar m\omega_0}}. \tag{S55}$$

and analogous expressions for a_ω and a_ω^+, the Hamiltonian can be rewritten as

$$\hat{H} = \frac{\hat{p}_0^2}{2m} + \frac{m\omega_0^2\hat{x}_0^2}{2} + \int\limits_0^\infty d\omega \left(\frac{\hat{p}_\omega^2}{2m} + \frac{m\omega^2\hat{x}_\omega^2}{2} + \sqrt{\frac{\gamma}{2\pi}} \frac{\omega_0\hat{x}_0\hat{p}_\omega - \omega\hat{x}_\omega\hat{p}_0}{\sqrt{\omega\omega_0}}\right). \tag{S56}$$

B. Phonon polariton non-Hermitian Hamiltonian

The simplest way to incorporate losses into the Hamiltonian (S44) is to assume that phonon-polariton modes couple directly to a continuum of bath modes. The full Hamiltonian then reads

$$\hat{H} = \int d\omega d\phi \left[\hbar\omega a^+_{\omega,\phi} a_{\omega,\phi} + \hbar \int\limits_0^\infty d\omega_b \left(\omega_b a^+_{\omega_b} a_{\omega_b} + i\sqrt{\frac{\omega''(\omega,\phi)}{\pi}} a_\omega a^+_{\omega_b} - \right.\right.$$
$$\left.\left. -i\sqrt{\frac{\omega''(\omega,\phi)}{\pi}} a^+_\omega a_{\omega_b}\right)\right], \quad \text{(S57)}$$

where $\omega''(\omega, \phi)$ is obtained from the classical dispersion relation.

The complex dielectric permittivity reads [1]

$$\varepsilon_i(\omega) = \varepsilon_{\infty i} + \frac{ne^2}{\varepsilon_0 m_i \left(\omega_{0i}^2 - \omega^2 - i\gamma_i\omega\right)} = \varepsilon_i'(\omega) + i\varepsilon_i''(\omega), \quad \text{(S58)}$$

where $\varepsilon_i'(\omega)$ and $\varepsilon_i''(\omega)$ are the real and imaginary parts of the dielectric permittivity, respectively, and $\omega = \omega' - i\omega''$ is the complex frequency. For weak losses, i.e., $\gamma_i \ll \omega_{0i}$ and $\omega'' \ll \omega'$, one may write

$$\varepsilon_i(\omega) \approx \varepsilon_i'(\omega') + \frac{d\varepsilon_i'(\omega')}{d\omega'}\left(\frac{\gamma_i}{2} - \omega''\right). \quad \text{(S59)}$$

The dispersion relation

$$\varepsilon_x k_x^2 + \varepsilon_y k_y^2 + \varepsilon_z \frac{\pi^2}{d^2} = 0, \quad \text{(S60)}$$

can then be expanded to first order in the losses:

$$\varepsilon_x' k_x^2 + \varepsilon_y' k_y^2 + \varepsilon_z' \frac{\pi^2}{d^2} = 0, \quad \text{(S61)}$$

$$\frac{d\varepsilon_x'(\omega')}{d\omega'}\left(\frac{\gamma_x}{2} - \omega''\right) k_x^2 + \frac{d\varepsilon_y'(\omega')}{d\omega'}\left(\frac{\gamma_y}{2} - \omega''\right) k_y^2 + \frac{d\varepsilon_z'(\omega')}{d\omega'}\left(\frac{\gamma_z}{2} - \omega''\right) \frac{\pi^2}{d^2} = 0. \quad \text{(S62)}$$

From this, the imaginary part of the frequency is

$$\omega'' = \frac{1}{2}\frac{\frac{d\varepsilon_x'(\omega')}{d\omega'} k_x^2\gamma_x + \frac{d\varepsilon_y'(\omega')}{d\omega'} k_y^2\gamma_y + \frac{d\varepsilon_z'(\omega')}{d\omega'}\frac{\pi^2}{d^2}\gamma_z}{\frac{d\varepsilon_x'(\omega')}{d\omega'} k_x^2 + \frac{d\varepsilon_y'(\omega')}{d\omega'} k_y^2 + \frac{d\varepsilon_z'(\omega')}{d\omega'}\frac{\pi^2}{d^2}}. \quad \text{(S63)}$$

S5. INTERACTION HAMILTONIAN

We consider a set of two-level systems (TLSs) with transition frequencies Ω_i and transition dipole moments $\mathbf{d}_i$, located at $(\mathbf{r}_i, z_i)$, where $\mathbf{r}_i = (x_i, y_i)$ or, equivalently, in polar coordinates, $\mathbf{r}_i = (r_i, \theta_i)$. The full Hamiltonian of the system is

$$\hat{H} = \int d^2\mathbf{k}\hbar\omega(\mathbf{k}) a^+_{\mathbf{k}} a_{\mathbf{k}} + \sum_{i=1}^{2} \hbar\Omega_i \sigma_i^+ \sigma_i^- + \hat{H}_{int}. \quad \text{(S64)}$$

where the interaction Hamiltonian is

$$\hat{H}_{int} = -\sum_{i=1}^{2} \hat{\mathbf{d}}_i \hat{\mathbf{E}}(\mathbf{r}_i, z_i, t), \tag{S65}$$

and $\hat{\mathbf{d}}_i = \mathbf{d}_i \left(\hat{\sigma}_i^+ + \hat{\sigma}_i^-\right)$ is the dipole moment operator of the ith emitter. From this point on, we consider emitters placed in the upper half-space, $z_i = h_i + d/2 > d/2$, with vertically oriented dipole moments, $\mathbf{d}_i = (0, 0, d_i)^T$. In the calculations, we choose d_i such that the free-space spontaneous-emission rate, $\Gamma = \frac{\omega^3 d_i^2}{3\pi\hbar\varepsilon_0 c^3}$, is of the order of 1 ns^{-1} [3]; specifically, we use $d_i = 10^{-26}$ Cm, which gives $\Gamma = 1.74$ ns^{-1}. Within the rotating-wave approximation (RWA), the interaction Hamiltonian takes the form

$$\hat{H}_{int} = -\int \frac{d^2\mathbf{k}}{2\pi} k \left(\varphi_1 \cos\frac{k_z d}{2} + \varphi_2 \sin\frac{k_z d}{2}\right) \sum_{i=1}^{2} d_i e^{-kh_i} \left[\hat{\sigma}_i^+ a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_i} + \hat{\sigma}_i^- a_{\mathbf{k}}^+ e^{-i\mathbf{k}\cdot\mathbf{r}_i}\right]. \tag{S66}$$

A. Simplification for the thin layer limit

In this limit, we consider the $l = 0$ mode and assume that $\varepsilon_{x,y,z} \gg \varepsilon_{1,2}$ and $d \ll 1/k$. Then, according to Eq. (S27), $\varphi_2 \ll \varphi_1$, so we set $\varphi_2 = 0$. Equation (S37) then gives

$$\varphi_1 = \sqrt{\frac{\hbar}{\varepsilon_0 d \left(\frac{d\varepsilon_x}{d\omega} k_x^2 + \frac{d\varepsilon_y}{d\omega} k_y^2\right)}}. \tag{S67}$$

The interaction Hamiltonian, therefore, reduces to

$$\hat{H}_{int} = -\int \frac{d^2\mathbf{k}}{2\pi} \sqrt{\frac{\hbar k^2}{\varepsilon_0 d \left(\frac{d\varepsilon_x}{d\omega} k_x^2 + \frac{d\varepsilon_y}{d\omega} k_y^2\right)}} \sum_{i=1}^{2} d_i e^{-kh_i} \left[\hat{\sigma}_i^+ a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_i} + \hat{\sigma}_i^- a_{\mathbf{k}}^+ e^{-i\mathbf{k}\cdot\mathbf{r}_i}\right]. \tag{S68}$$

B. Simplification for the canalization condition

Canalization in a single anisotropic layer arises when $\varepsilon_2 \to -\infty$. In this case, the fundamental mode corresponds to $l = 1$, with $k_z d = \pi/2$ and $\varphi_1 = \varphi_2$. Equation (S37) then yields

$$\varphi_1 = \varphi_2 = \sqrt{\frac{\hbar}{\varepsilon_0 d \left(\frac{d\varepsilon_x}{d\omega} k_x^2 + \frac{d\varepsilon_y}{d\omega} k_y^2 + \frac{d\varepsilon_z}{d\omega} \frac{\pi^2}{4d^2}\right)}}. \tag{S69}$$

Accordingly, the interaction Hamiltonian becomes

$$\hat{H}_{int} = -\int \frac{d^2\mathbf{k}}{2\pi} \sqrt{\frac{2\hbar k^2}{\varepsilon_0 d \left(\frac{d\varepsilon_x}{d\omega} k_x^2 + \frac{d\varepsilon_y}{d\omega} k_y^2 + \frac{d\varepsilon_z}{d\omega} \frac{\pi^2}{4d^2}\right)}} \sum_{i=1}^{2} d_i e^{-kh_i} \left[\hat{\sigma}_i^- a_{\mathbf{k}}^+ e^{-i\mathbf{k}\cdot\mathbf{r}_i} + \hat{\sigma}_i^+ a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_i}\right] \tag{S70}$$

C. Frequency-angle space

In some cases, it is convenient to parametrize the polaritonic modes by frequency ω and in-plane propagation angle ϕ, rather than by the wavevector $\mathbf{k}$. This is useful because the dispersion relation, Eq. (S28), provides an explicit expression for $\mathbf{k}(\omega,\phi)$, Eq. (S29), whereas the inverse relation $\omega(\mathbf{k})$ is not generally available in closed form. The change of variables under the integral is performed according to

$$\int f(\mathbf{k}) d^2\mathbf{k} = \int \tilde{f}(\omega,\phi) \det(\mathbf{J}_{\mathbf{k}}(\omega,\phi)) d\omega d\phi. \tag{S71}$$

where $\mathbf{J}_{\mathbf{k}}(\omega,\phi)$ is the Jacobian of the transformation:

$$\det(\mathbf{J}_{\mathbf{k}}(\omega,\phi)) = \det\begin{pmatrix} \frac{\partial k_x}{\partial \omega} & \frac{\partial k_x}{\partial \phi} \\ \frac{\partial k_y}{\partial \omega} & \frac{\partial k_y}{\partial \phi} \end{pmatrix} = \det\begin{pmatrix} \frac{\partial k}{\partial \omega}\cos\phi & \frac{\partial k}{\partial \phi}\cos\phi - k\sin\phi \\ \frac{\partial k}{\partial \omega}\sin\phi & \frac{\partial k}{\partial \phi}\sin\phi + k\cos\phi \end{pmatrix} = \frac{1}{2}\frac{\partial k^2}{\partial \omega}. \tag{S72}$$

We introduce the creation and annihilation operators in the new variables, $a_{\omega,\phi}$ and $a^+_{\omega,\phi}$, while preserving the commutation relation in the form

$$\left[a_{\omega,\phi}, a^+_{\omega',\phi'}\right] = \delta(\omega-\omega')\delta(\phi-\phi'). \tag{S73}$$

The new operators are therefore related to $a_{\mathbf{k}}$ by

$$a_{\omega,\phi} = a_{\mathbf{k}}\sqrt{\det(\mathbf{J}_{\mathbf{k}}(\omega,\phi))}. \tag{S74}$$

The Hamiltonian becomes

$$\hat{H} = \int \hbar\omega a^+_{\omega,\phi} a_{\omega,\phi} d\omega d\phi + \sum_{i=1}^{2} \hbar\Omega_i \sigma_i^+ \sigma_i^- + \hat{H}_{int}. \tag{S75}$$

with

$$\hat{H}_{int} = -\int \frac{d\omega d\phi}{2\pi} k \left(\varphi_1 \cos\frac{k_z d}{2} + \varphi_2 \sin\frac{k_z d}{2}\right) \sqrt{\det(\mathbf{J}_{\mathbf{k}}(\omega,\phi))} \sum_{i=1}^{2} d_i e^{-kh_i} \times$$
$$\times \left[\hat{\sigma}_i^- a^+_{\omega,\phi} e^{-ikr_i\cos(\phi-\theta_i)} + \hat{\sigma}_i^+ a_{\omega,\phi} e^{ikr_i\cos(\phi-\theta_i)}\right], \tag{S76}$$

where $r_i = |\mathbf{r}_i|$ and θ_i is the in-plane angle of $\mathbf{r}_i$.

Under canalization conditions, the Jacobian determinant becomes

$$\det(\mathbf{J}_{\mathbf{k}}(\omega,\phi)) = \frac{1}{2}\frac{\partial k^2}{\partial \omega} = \frac{\partial}{\partial \omega}\frac{-\pi^2}{8d^2\left(\frac{\varepsilon_x}{\varepsilon_z}\cos^2\phi + \frac{\varepsilon_y}{\varepsilon_z}\sin^2\phi\right)} = \frac{k^4 d^2}{8\pi^2\varepsilon_z}\left[\left(\frac{d\varepsilon_x}{d\omega} - \frac{\varepsilon_x}{\varepsilon_z}\frac{d\varepsilon_z}{d\omega}\right)\cos^2\phi + \right.$$
$$\left. + \left(\frac{d\varepsilon_y}{d\omega} - \frac{\varepsilon_y}{\varepsilon_z}\frac{d\varepsilon_z}{d\omega}\right)\sin^2\phi\right]. \tag{S77}$$

Accordingly, the interaction Hamiltonian can be written as

$$\hat{H}_{int} = -\int \frac{d\omega d\phi}{4\pi^2} k^2 \sqrt{\frac{\hbar d}{\varepsilon_0 \varepsilon_z}} \sum_{i=1}^{2} d_i e^{-kh_i} \left[\hat{\sigma}_i^- a^+_{\omega,\phi} e^{-ikr_i\cos(\phi-\theta_i)} + \hat{\sigma}_i^+ a_{\omega,\phi} e^{ikr_i\cos(\phi-\theta_i)}\right]. \tag{S78}$$

S6. RADIATION SPECTRUM

To further characterize the behavior of emitters coupled to phonon polaritons, we calculate the radiation spectra for the three cases considered in the main text: a single emitter, two emitters placed along the canalization direction away from the BIC condition, and two emitters under the BIC condition. In the lossless case, the spectrum can be obtained by summing the occupations of all modes at a given frequency after the emitters have fully decayed. For a state of the form

$$|\psi(t)\rangle = c_1(t)|1,0,0\rangle + c_2(t)|0,1,0\rangle + \int\int d\omega d\phi a_{\omega,\phi}(t)|0,0,1_{\omega,\phi}\rangle. \tag{S79}$$

The spectrum is

$$S(\omega) = \int d\phi |a_{\omega,\phi}(\infty)|^2. \tag{S80}$$

In the presence of losses, however, Eq. (S80) is no longer applicable because the polaritonic modes themselves decay into the bath. Assuming that each polaritonic mode couples to its own continuum of bath modes, the emitted spectrum can instead be extracted from the bath population. We then consider the state vector

$$\begin{aligned}|\psi(t)\rangle = c_1(t)|1,0,0,0\rangle + c_2(t)|0,1,0,0\rangle + \int\int d\omega d\phi \left[a_{\omega,\phi}(t)|0,0,1_{\omega,\phi},0\rangle + \right. \\ \left. + \int d\omega_b b_{\omega,\phi,\omega_b}(t)|0,0,0,1_{\omega,\phi,\omega_b}\rangle\right].\end{aligned} \tag{S81}$$

The spectrum is then given by

$$S(\omega) = \int d\phi \int_0^\infty d\omega_b |b_{\omega,\phi,\omega_b}(\infty)|^2, \tag{S82}$$

where, according to Eq. (S52),

$$b_{\omega,\phi,\omega_b}(\infty) = e^{-i\omega_b t}\sqrt{\frac{\omega''(\omega,\phi)}{\pi}} \int_0^\infty dt' \tilde{a}_{\omega,\phi}(t') e^{-i(\omega-\omega_b)t'}, \tag{S83}$$

with $\tilde{a}_{\omega,\phi}(t') = a_{\omega,\phi}(t')e^{i\omega t'}$. Substituting these expressions into Eq. (S82) gives

$$\begin{aligned}S(\omega) = \int d\phi \frac{\omega''(\omega,\phi)}{\pi} \int_0^\infty \int_0^\infty dt' dt'' \tilde{a}_{\omega,\phi}(t') \tilde{a}^*_{\omega,\phi}(t'') \int_0^\infty d\omega_b e^{-i(\omega-\omega_b)(t'-t'')} \approx \\ \approx 2\int d\phi \omega''(\omega,\phi) \int_0^\infty \int_0^\infty dt' dt'' \tilde{a}_{\omega,\phi}(t') \tilde{a}^*_{\omega,\phi}(t'') \delta(t'-t'').\end{aligned} \tag{S84}$$

Finally, we obtain

$$S(\omega) = 2\int d\phi \omega''(\omega,\phi) \int_0^\infty dt' |a_{\omega,\phi}(t')|^2. \tag{S85}$$

S7. BIC CONDITIONS DERIVATION

The time evolution of the system can be expressed in terms of the resolvent operator $\hat{G}(E)$:

$$\hat{U}(t-t') = \int_{-\infty}^{\infty} \frac{dE}{2\pi i} \left(\hat{G}(E-i0) - \hat{G}(E+i0)\right) e^{-i\frac{E(t-t')}{\hbar}}, \tag{S86}$$

where

$$\hat{G}(E) = \frac{1}{E-\hat{H}}. \tag{S87}$$

We consider an initial state corresponding to an excitation of the emitters, $|\psi(0)\rangle = |\alpha\rangle = (c_1\sigma_1^+ + c_2\sigma_2^+)|0\rangle$; $|c_1|^2+|c_2|^2=1$. The state at time $t>0$ is then

$$|\psi(t)\rangle = -\int_{-\infty}^{\infty} \frac{dE}{2\pi i} e^{-i\frac{Et}{\hbar}} \hat{G}(E+i0)|\alpha\rangle. \tag{S88}$$

The relevant matrix element of the resolvent is

$$G_{\alpha\alpha}(E+i0) = \langle\alpha|\hat{G}(E+i0)|\alpha\rangle = \frac{1}{E-\langle\alpha|\hat{H}_0|\alpha\rangle - \Sigma_{\alpha\alpha}(E+i0)+i0}, \tag{S89}$$

where

$$\hat{H}_0 = \int d^2\mathbf{k}\hbar\omega(\mathbf{k})a_\mathbf{k}^+a_\mathbf{k} + \sum_{i=1}^{2}\hbar\Omega_i\sigma_i^+\sigma_i^-, \tag{S90}$$

and the self-energy correction is

$$\Sigma_{\alpha\alpha}(E+i0) \approx \sum_\beta \frac{\left|\langle\alpha|\hat{H}_{int}|\beta\rangle\right|^2}{E-\langle\beta|\hat{H}_0|\beta\rangle+i0}. \tag{S91}$$

Bound states are determined by the pole condition

$$E_B = \langle\alpha|\hat{H}_0|\alpha\rangle + \Sigma_{\alpha\alpha}(E_B+i0). \tag{S92}$$

For symmetric and antisymmetric emitter states,

$$|\pm\rangle = \frac{1}{\sqrt{2}}(\sigma_1^+ \pm \sigma_2^+)|0\rangle,$$

we obtain

$$E_B^\pm = \hbar\frac{\Omega_1+\Omega_2}{2} + \int d^2\mathbf{k}\frac{\left|\langle\pm|\hat{H}_{int}a_\mathbf{k}^+|0\rangle\right|^2}{E_B^\pm - \hbar\omega(\mathbf{k})+i0}. \tag{S93}$$

Substituting Eq. (S70) yields

$$E_B^\pm = \hbar\frac{\Omega_1+\Omega_2}{2} + \int\frac{d^2\mathbf{k}}{E_B^\pm-\hbar\omega(\mathbf{k})+i0}\frac{\hbar k^2\left|d_1e^{-kh_1}e^{i\mathbf{k}\cdot\mathbf{r}_1} \pm d_2e^{-kh_2}e^{i\mathbf{k}\cdot\mathbf{r}_2}\right|^2}{4\pi^2\varepsilon_0 d\left(\frac{d\varepsilon_x}{d\omega}k_x^2 + \frac{d\varepsilon_y}{d\omega}k_y^2 + \frac{d\varepsilon_z}{d\omega}\frac{\pi^2}{d^2}\right)}. \tag{S94}$$

Without loss of generality, we set $\mathbf{r}_1 = (l/2, 0)^T$ and $\mathbf{r}_2 = (-l/2, 0)^T$, which gives

$$E_B^{\pm} = \hbar\frac{\Omega_1+\Omega_2}{2} + \mathcal{P}\int \frac{d^2\mathbf{k}}{E_B^{\pm} - \hbar\omega(\mathbf{k})} \frac{\hbar k^2 \left| d_1 e^{-kh_1} e^{i\frac{k_x l}{2}} \pm d_2 e^{-kh_2} e^{-i\frac{k_x l}{2}} \right|^2}{4\pi^2\varepsilon_0 d\left(\frac{d\varepsilon_x}{d\omega}k_x^2 + \frac{d\varepsilon_y}{d\omega}k_y^2 + \frac{d\varepsilon_z}{d\omega}\frac{\pi^2}{d^2}\right)} -$$
$$- i\int d^2\mathbf{k} \frac{\hbar k^2 \left| d_1 e^{-kh_1} e^{i\frac{k_x l}{2}} \pm d_2 e^{-kh_2} e^{-i\frac{k_x l}{2}} \right|^2}{4\pi\varepsilon_0 d\left(\frac{d\varepsilon_x}{d\omega}k_x^2 + \frac{d\varepsilon_y}{d\omega}k_y^2 + \frac{d\varepsilon_z}{d\omega}\frac{\pi^2}{d^2}\right)} \delta(E_B^{\pm} - \hbar\omega(\mathbf{k})). \quad \text{(S95)}$$

For a bound state in the continuum (BIC), the imaginary part must vanish. Since the integrand is non-negative, this requires that the numerator vanishes for all $\mathbf{k}$ satisfying the on-shell condition $E_B^{\pm} = \hbar\omega(\mathbf{k})$, yielding

$$d_1 e^{-kz_1} e^{i\frac{k_x l}{2}} \pm d_2 e^{-kz_2} e^{-i\frac{k_x l}{2}} = 0 \quad \text{(S96)}$$

This condition implies identical emitters, $d_1 = d_2$, and equal distances from the layer, $h_1 = h_2 = h$, also, $1 \pm \cos(k_x l) = 0$. Thus,

$$k_x = k_c = \text{const}, \qquad l_+ = \frac{\pi + 2\pi n}{k_c}, \quad l_- = \frac{2\pi n}{k_c}, \quad n \in \mathbb{N}.$$

Since this condition must hold for all modes contributing to the integral, it requires canalization, i.e., a fixed longitudinal wavevector $k_x = k_c$. Consequently, the bound-state energy is fixed by the canalized mode, $E_B^{\pm} = \hbar\omega_c$. Equation (S95) then reduces to

$$\hbar\omega_c = \hbar\frac{\Omega_1+\Omega_2}{2} + \mathcal{P}\int \frac{d^2\mathbf{k}}{\omega_c - \omega(\mathbf{k})} \frac{k^2\left(1 \pm \cos k_x l\right) d_1^2 e^{-2kh}}{2\pi^2\varepsilon_0 d\left(\frac{d\varepsilon_x}{d\omega}k_x^2 + \frac{d\varepsilon_y}{d\omega}k_y^2 + \frac{d\varepsilon_z}{d\omega}\frac{\pi^2}{d^2}\right)}. \quad \text{(S97)}$$

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