

## Supplementary Materials for

### **Cavity quantum-electrodynamical polaritonically enhanced electron-phonon coupling and its influence on superconductivity**

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## Section S1. Relevant photon modes in cavity

In this work we consider a 2D material on a dielectric substrate in a nanocavity. We impose reflecting mirror boundary conditions with  $\vec{n} \cdot \vec{B} = 0$  and  $\vec{n} \times \vec{E} = 0$  for the magnetic  $\vec{B}$  and electric  $\vec{E}$  components of the photonic field, and  $\vec{n} = \hat{z}$  the surface normal. The size of the cavity in  $z$  direction is  $L_z$ . If the dielectric substrate has a very high dielectric constant, such as for SrTiO<sub>3</sub> at low temperature, it can be considered almost metallic and  $L_z$  is reduced accordingly in our effective description.

Assuming periodic boundary conditions in the  $x - y$  plane, we obtain for example for the vacuum electric field, obeying the wave equation  $\nabla^2 E - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = 0$  with  $c$  the speed of light

$$E_x(x, y, z, t) = E_1 \exp(ik_x x) \exp(ik_y y) \sin(k_z z) \exp(-i\omega_{\text{phot}}(\vec{k})t) \quad (\text{S1})$$

$$E_y(x, y, z, t) = E_2 \exp(ik_x x) \exp(ik_y y) \sin(k_z z) \exp(-i\omega_{\text{phot}}(\vec{k})t) \quad (\text{S2})$$

$$E_z(x, y, z, t) = E_3 \exp(ik_x x) \exp(ik_y y) \cos(k_z z) \exp(-i\omega_{\text{phot}}(\vec{k})t) \quad (\text{S3})$$

with  $\omega_{\text{phot}}(\vec{k}) = c|\vec{k}|$ , and

$$k_x = \frac{2\pi l}{L_x}, \quad l \in \mathbb{N}_0 \quad (\text{S4})$$

$$k_y = \frac{2\pi m}{L_y}, \quad m \in \mathbb{N}_0 \quad (\text{S5})$$

$$k_z = \frac{\pi n}{L_z}, \quad n \in \mathbb{N}_0 \quad (\text{S6})$$

We assume  $L_x$  and  $L_y$  to be large to obtain a fine momentum grid in the  $x - y$  plane. By contrast  $L_z$  is assumed to be small ( $L_z \ll L_x, L_y$ ), implying that for  $n = 1$  the photon energy is at least  $c\frac{\pi}{L_z}$  well above typical phonon energy scales and thus irrelevant to the problem of our interest. We retain only the  $n = 0$ ,  $k_z = 0$  component that has constant mode amplitude along the  $z$  direction. Thus we will use only one mode for each in-plane momentum  $\vec{q} = (q_x, q_y)$  with

$$E_x(x, y, z, t) = 0 \quad (\text{S7})$$

$$E_y(x, y, z, t) = 0 \quad (\text{S8})$$

$$E_z(x, y, z, t) = E_3 \exp(iq_x x) \exp(iq_y y) \exp(-i\omega_{\text{phot}}(\vec{k})t) \quad (\text{S9})$$

## Section S2. Phonon-photon Hamiltonian

We consider the generic Hamiltonian for phonon-photon coupling (41)

$$H_{\text{phon-phot}} = H_0 + H' \quad (\text{S10})$$

$$H_0 = \Omega \sum_{\vec{q}} b_{\vec{q}}^\dagger b_{\vec{q}} + \sum_{\vec{q}} \omega_{\text{phot}}(\vec{q}) a_{\vec{q}}^\dagger a_{\vec{q}} \quad (\text{S11})$$

$$H' = -\frac{e}{Mc} \sum_j \vec{P}_j \cdot \vec{A}(\vec{R}_j) + \frac{e^2}{2Mc^2} \sum_j \vec{A}(\vec{R}_j) \cdot \vec{A}(\vec{R}_j) \quad (\text{S12})$$

Throughout we approximate the phonon dispersion relevant for FeSe/SrTiO<sub>3</sub> with a dispersionless  $\Omega = 92$  meV (29). Here  $\vec{q}$  summations are over the first Brillouin zone  $[-\pi, \pi)^2$  in the 2D square lattice with lattice constant  $a = 1$ , implying a high-frequency cutoff to the photons, which is irrelevant to the electron-boson physics happening at much lower energy. For the photon, we take only the mode polarized along the  $\hat{z}$  direction parallel to the phonon dipoles, and restrict it to the lowest branch  $q_z = 0$  due to cavity confinement as discussed above, implying  $\omega_{\text{phot}}(\vec{q}) = c|\vec{q}| = c\sqrt{q_x^2 + q_y^2}$ .

We write the phononic dipole current operator via bosonic operators

$$\vec{J}_j \equiv \frac{e}{M} \vec{P}_j = ie \sum_{\vec{q}} \left( \frac{\Omega}{2NM} \right)^{1/2} \hat{\xi}_{\vec{q}} \left( b_{\vec{q}}^\dagger - b_{-\vec{q}} \right) e^{-i\vec{q}\vec{R}_j} \equiv \sum_{\vec{q}} \frac{1}{\sqrt{N}} \vec{J}(\vec{q}) e^{-i\vec{q}\vec{R}_j} \quad (\text{S13})$$

with polarization vector  $\hat{\xi}_{\vec{q}} = \hat{z}$ , and similarly for the relevant  $z$  component of the photonic vector potential

$$A_z(\vec{R}_j) \equiv \sum_{\vec{q}} \left( \frac{2\pi c^2}{\omega_{\text{phot}}(\vec{q}) \nu_0} \right)^{1/2} \left( a_{\vec{q}}^\dagger + a_{-\vec{q}} \right) e^{-i\vec{q}\vec{R}_j} \equiv \sum_{\vec{q}} \frac{c}{\sqrt{\nu_0}} A_\mu(\vec{q}) e^{-i\vec{q}\vec{R}_j} \quad (\text{S14})$$

assuming periodic boundary conditions inside the 2D plane. Here  $b_{\vec{q}}^\dagger$  ( $b_{\vec{q}}$ ) creates (annihilates) a phonon with wavevector  $\vec{q}$ ;  $a_{\vec{q}}^\dagger$  ( $a_{\vec{q}}$ ) creates (annihilates) a cavity photon with wavevector  $\vec{q}$ .  $N$  is the number of unit cells,  $V$  the system volume,  $\nu_0 \equiv V/N$  the unit cell volume, and  $e$  and  $M$  the ionic charge and reduced mass, respectively, related to the relative motion of positively and negatively charged ions in the optical phonon mode. In momentum space we have

$$J_z(\vec{q}) \equiv ie \left( \frac{\Omega}{2M} \right)^{1/2} \left( b_{\vec{q}}^\dagger - b_{-\vec{q}} \right) \quad (\text{S15})$$

$$A_z(\vec{q}) \equiv \left( \frac{2\pi}{\omega_{\text{phot}}(\vec{q})} \right)^{1/2} \left( a_{\vec{q}}^\dagger + a_{-\vec{q}} \right) \quad (\text{S16})$$

Now we first diagonalize the bare photon plus  $A^2$  terms of the Hamiltonian

$$H_{0,\text{phot}} = \sum_{\vec{q}} \omega_{\text{phot}}(\vec{q}) a_{\vec{q}}^\dagger a_{\vec{q}} \quad (\text{S17})$$

$$= \frac{1}{2} \sum_{\vec{q}} (P_{A,\vec{q}} P_{A,-\vec{q}} + \omega_{\text{phot}}(\vec{q})^2 X_{A,\vec{q}} X_{A,-\vec{q}}) \quad (\text{S18})$$

$$H_{A^2} = \frac{1}{2} \sum_{\vec{q}} \omega_{\text{P}}^2 X_{A,\vec{q}} X_{A,-\vec{q}} \quad (\text{S19})$$

Here we introduced canonical position and momentum operators for photon degrees of freedom

$$X_{A,\vec{q}} \equiv \sqrt{\frac{1}{2\omega_{\text{phot}}(\vec{q})}} (a_{\vec{q}} + a_{-\vec{q}}^\dagger) \quad (\text{S20})$$

$$P_{A,\vec{q}} \equiv -i\sqrt{\frac{\omega_{\text{phot}}(\vec{q})}{2}} (a_{-\vec{q}} - a_{\vec{q}}^\dagger) \quad (\text{S21})$$

We also defined the phononic plasma frequency

$$\omega_{\text{P}} \equiv \sqrt{\frac{4\pi e^2}{M\nu_0}} = \sqrt{\frac{4\pi e^2}{M\nu_{0,2D}L_z}} \quad (\text{S22})$$

which for the 2D system in the cavity is governed by the length of the vacuum inside the cavity in  $z$  direction,  $L_z$ , and the 2D unit cell area  $\nu_{0,2D}$ . The expressions above are given in cgs units. In the SI system,  $\omega_{\text{P}}^{\text{SI}} = \sqrt{\frac{e^2}{M\epsilon_0\nu_{0,2D}L_z}}$  with the vacuum permittivity  $\epsilon_0$ .

The bilinear  $J \cdot A$  coupling term is written as

$$H_{J \cdot A} = -\frac{1}{\sqrt{\nu_0}} \sum_{\vec{q}} \vec{J}(\vec{q}) \cdot \vec{A}(-\vec{q}) \quad (\text{S23})$$

$$= -\sum_{\vec{q}} \omega_{\text{P}} X_{A,\vec{q}} P_{B,\vec{q}} \quad (\text{S24})$$

where it is convenient to introduce canonical position and momentum operators for the phonons

$$X_{B,\vec{q}} \equiv \sqrt{\frac{1}{2\Omega}} (b_{\vec{q}} + b_{-\vec{q}}^\dagger) \quad (\text{S25})$$

$$P_{B,\vec{q}} \equiv -i\sqrt{\frac{\Omega}{2}} (b_{-\vec{q}} - b_{\vec{q}}^\dagger) \quad (\text{S26})$$

Written in these operators, the bare phonon term  $H_{0,\text{phon}} \equiv \Omega \sum_{\vec{q}} b_{\vec{q}}^\dagger b_{\vec{q}}$  takes the form

$$H_{0,\text{phon}} = \frac{1}{2} \sum_{\vec{q}} (P_{B,\vec{q}} P_{B,-\vec{q}} + \Omega^2 X_{B,\vec{q}} X_{B,-\vec{q}}) \quad (\text{S27})$$

The total phonon-photon Hamiltonian is now written as pairs of coupled harmonic oscillators

$$H_{\text{phon-phot}} = H_{0,\text{phot}} + H_{0,\text{phon}} + H_{A^2} + H_{J,A} \quad (\text{S28})$$

$$\begin{aligned} &= \frac{1}{2} \sum_{\vec{q}} \left( P_{A,\vec{q}} P_{A,-\vec{q}} + P_{B,\vec{q}} P_{B,-\vec{q}} + (\omega_{\text{phot}}(\vec{q})^2 + \omega_{\text{P}}^2) X_{A,\vec{q}} X_{A,-\vec{q}} + \right. \\ &\quad \left. + \Omega^2 X_{B,\vec{q}} X_{B,-\vec{q}} - 2\omega_{\text{P}} X_{A,\vec{q}} P_{B,\vec{q}} \right) \end{aligned} \quad (\text{S29})$$

In order to diagonalize this Hamiltonian, we introduce a transformation

$$\tilde{P}_{B,\vec{q}} \equiv \Omega X_{B,\vec{q}}, \quad (\text{S30})$$

$$\tilde{X}_{B,\vec{q}} \equiv -\Omega^{-1} P_{B,\vec{q}} \quad (\text{S31})$$

which leaves the canonical commutator unchanged but interchanges position and momentum operators. The phonon-photon Hamiltonian is then compactly represented as

$$\begin{aligned} H_{\text{phon-phot}} &= \frac{1}{2} \sum_{\vec{q}} \begin{bmatrix} P_{A,\vec{q}} \\ \tilde{P}_{B,\vec{q}} \end{bmatrix}^T \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} P_{A,-\vec{q}} \\ \tilde{P}_{B,-\vec{q}} \end{bmatrix} + \\ &+ \frac{1}{2} \sum_{\vec{q}} \begin{bmatrix} X_{A,\vec{q}} \\ \tilde{X}_{B,\vec{q}} \end{bmatrix}^T \begin{bmatrix} \omega_{\text{phot}}(\vec{q})^2 + \omega_{\text{P}}^2 & \Omega\omega_{\text{P}} \\ \Omega\omega_{\text{P}} & \Omega^2 \end{bmatrix} \begin{bmatrix} X_{A,-\vec{q}} \\ \tilde{X}_{B,-\vec{q}} \end{bmatrix} \end{aligned} \quad (\text{S32})$$

Diagonalization is now achieved with the following unitary transformation to polariton canonical position and momentum operators

$$\begin{bmatrix} X_{+,\vec{q}} \\ X_{-,\vec{q}} \end{bmatrix} = \begin{bmatrix} \cos(\theta_{\vec{q}}) & \sin(\theta_{\vec{q}}) \\ -\sin(\theta_{\vec{q}}) & \cos(\theta_{\vec{q}}) \end{bmatrix} \begin{bmatrix} X_{A,\vec{q}} \\ \tilde{X}_{B,\vec{q}} \end{bmatrix} \quad (\text{S33})$$

$$\begin{bmatrix} P_{+,\vec{q}} \\ P_{-,\vec{q}} \end{bmatrix} = \begin{bmatrix} \cos(\theta_{\vec{q}}) & \sin(\theta_{\vec{q}}) \\ -\sin(\theta_{\vec{q}}) & \cos(\theta_{\vec{q}}) \end{bmatrix} \begin{bmatrix} P_{A,\vec{q}} \\ \tilde{P}_{B,\vec{q}} \end{bmatrix} \quad (\text{S34})$$

which leaves canonical commutation relations intact. The resulting phonon-photon Hamiltonian expressed in polaritonic operators is

$$\begin{aligned} H_{\text{phon-phot}} &= \frac{1}{2} \sum_{\vec{q}} \begin{bmatrix} P_{+,\vec{q}} \\ P_{-,\vec{q}} \end{bmatrix}^T \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} P_{+,-\vec{q}} \\ P_{-,-\vec{q}} \end{bmatrix} + \\ &+ \frac{1}{2} \sum_{\vec{q}} \begin{bmatrix} X_{+,\vec{q}} \\ X_{-,\vec{q}} \end{bmatrix}^T \begin{bmatrix} \omega_{+}(\vec{q})^2 & 0 \\ 0 & \omega_{-}(\vec{q})^2 \end{bmatrix} \begin{bmatrix} X_{+,-\vec{q}} \\ X_{-,-\vec{q}} \end{bmatrix} \end{aligned} \quad (\text{S35})$$

with polaritonic dispersions  $\omega_{\pm}(\vec{q})$  fulfilling

$$\omega_{\pm}(\vec{q})^2 = \frac{1}{2} \left( \omega_{\text{phot}}(\vec{q})^2 + \omega_{\text{P}}^2 + \Omega^2 \pm \sqrt{(\omega_{\text{phot}}(\vec{q})^2 + \omega_{\text{P}}^2 + \Omega^2)^2 - 4\omega_{\text{phot}}(\vec{q})^2\Omega^2} \right) \quad (\text{S36})$$

In particular, in the long-wavelength limit one obtains

$$\omega_{+}(\vec{q} \rightarrow 0) \rightarrow \sqrt{\Omega^2 + \omega_{\text{P}}^2} \quad (\text{S37})$$

$$\omega_{-}(\vec{q} \rightarrow 0) \rightarrow 0 \quad (\text{S38})$$

as shown for the semiclassical polariton dispersions in (41). The diagonalization condition is given by

$$\arctan(\theta_{\vec{q}}) = \frac{\omega_{\text{phot}}(\vec{q})^2 + \omega_{\text{P}}^2 - \Omega^2 + \sqrt{(\omega_{\text{phot}}(\vec{q})^2 + \omega_{\text{P}}^2 + \Omega^2)^2 - 4\omega_{\text{phot}}(\vec{q})^2\Omega^2}}{2\Omega\omega_{\text{P}}} \quad (\text{S39})$$

Defining bosonic operators for the upper ( $\lambda = +$ ) and lower ( $\lambda = -$ ) polariton branches

$$X_{\lambda, \vec{q}} \equiv \sqrt{\frac{1}{2\omega_{\lambda}(\vec{q})}} \left( \alpha_{\vec{q}, \lambda} + \alpha_{-\vec{q}, \lambda}^{\dagger} \right) \quad (\text{S40})$$

$$P_{\lambda, \vec{q}} \equiv -i\sqrt{\frac{\omega_{\lambda}(\vec{q})}{2}} \left( \alpha_{-\vec{q}, \lambda} - \alpha_{\vec{q}, \lambda}^{\dagger} \right) \quad (\text{S41})$$

we rewrite the phonon-photon Hamiltonian in a very compact polaritonic form

$$H_{\text{phon-photon}} = \sum_{\vec{q}, \lambda = \pm} \omega_{\lambda}(\vec{q}) \alpha_{\vec{q}, \lambda}^{\dagger} \alpha_{\vec{q}, \lambda} \quad (\text{S42})$$

The transformation from the initial phononic degrees of freedom to the final polaritonic ones is then given by

$$X_{B, \vec{q}} = \frac{1}{\Omega} (\sin(\theta_{\vec{q}}) P_{+, \vec{q}} + \cos(\theta_{\vec{q}}) P_{-, \vec{q}}) \quad (\text{S43})$$

For the bosonic operators, this implies

$$b_{\vec{q}} + b_{-\vec{q}}^{\dagger} = -i \sin(\theta_{\vec{q}}) \sqrt{\frac{\omega_{+}(\vec{q})}{\Omega}} (\alpha_{-\vec{q}, +} - \alpha_{\vec{q}, +}^{\dagger}) - i \cos(\theta_{\vec{q}}) \sqrt{\frac{\omega_{-}(\vec{q})}{\Omega}} (\alpha_{-\vec{q}, -} - \alpha_{\vec{q}, -}^{\dagger}) \quad (\text{S44})$$

which will give the transformation from electron-phonon to electron-polariton coupling in the following.

### Section S3. Electron-polariton Hamiltonian

The electron-polariton model Hamiltonian for FeSe/SrTiO<sub>3</sub> inside the cavity reads

$$H = H_{e\text{-phonon}} + H_{\text{phon-photon}} \quad (\text{S45})$$

$$H_{e\text{-phonon}} = \sum_{\vec{k},\sigma} \epsilon_{\vec{k}} c_{\vec{k},\sigma}^\dagger c_{\vec{k},\sigma} + \frac{1}{\sqrt{N}} \sum_{\vec{k},\vec{q},\sigma} g(\vec{k},\vec{q}) c_{\vec{k}+\vec{q},\sigma}^\dagger c_{\vec{k},\sigma} (b_{\vec{q}} + b_{-\vec{q}}^\dagger) \quad (\text{S46})$$

Here,  $c_{\vec{k},\sigma}^\dagger$  ( $c_{\vec{k},\sigma}$ ) creates (annihilates) an electron with wavevector  $\vec{k}$  and spin  $\sigma$ ;  $\epsilon_{\vec{k}}$  is the electronic band dispersion measured relative to the chemical potential  $\mu$ ;  $g(\vec{k},\vec{q})$  is the momentum dependent electron-phonon coupling. The direct electron-photon coupling of electrons in the FeSe plane to the photon branch of interest is neglected, which amounts to the assumption that the paramagnetic electronic current density  $\vec{j}$  inside the FeSe layer is perfectly two-dimensional, thus not coupling to the photonic vector potential  $\vec{A}$  which points perpendicular to the plane, implying  $\vec{j} \cdot \vec{A} \approx 0$ .

Adopting the FeSe/SrTiO<sub>3</sub> single-band model from Rademaker *et al.* (28), we take an electronic band dispersion  $\epsilon_{\vec{k}} = -2t[\cos(k_x a) + \cos(k_y a)] - \mu$ , where  $a$  is the in-plane lattice constant. We set  $t = 0.075$  eV and use as an initial guess  $\mu = -0.235$  eV, which is adjusted during the self-consistent calculations (see below) to a fixed band filling  $n_\uparrow = n_\downarrow = 0.07$  for each spin. We neglect the fermion momentum dependence in the electron-phonon coupling  $g(\vec{k},\vec{q}) = g(\vec{q})$ , where  $\vec{q}$  is the momentum transfer, and use  $g(\vec{q}) = g_0 \exp(-|\vec{q}|/q_0)$ . Here,  $g_0$  is adjusted to fix the total dimensionless coupling strength  $\lambda \approx 0.18$  of the electron-phonon interaction in absence of the cavity coupling, and  $q_0$  sets the range of the interaction in momentum space.

The electron-polariton expressed in polaritonic bosonic operators is obtained via Eq. (S44) as

$$H = \sum_{\vec{k},\sigma} \epsilon_{\vec{k}} c_{\vec{k},\sigma}^\dagger c_{\vec{k},\sigma} + \frac{1}{\sqrt{N}} \sum_{\vec{k},\vec{q},\sigma,\lambda=\pm} c_{\vec{k}+\vec{q},\sigma}^\dagger c_{\vec{k},\sigma} (g_\lambda^*(\vec{q}) \alpha_{-\vec{q},\lambda}^\dagger + g_\lambda(\vec{q}) \alpha_{\vec{q},\lambda}) + \sum_{\vec{q},\lambda=\pm} \omega_\lambda(\vec{q}) \alpha_{\vec{q},\lambda}^\dagger \alpha_{\vec{q},\lambda} \quad (\text{S47})$$

where

$$g_+(\vec{q}) = i \sin(\theta_{\vec{q}}) \sqrt{\frac{\omega_+(\vec{q})}{\Omega}} g_0 \exp(-|\vec{q}|/q_0) \quad (\text{S48})$$

$$g_-(\vec{q}) = i \cos(\theta_{\vec{q}}) \sqrt{\frac{\omega_-(\vec{q})}{\Omega}} g_0 \exp(-|\vec{q}|/q_0) \quad (\text{S49})$$

The couplings are thus fully determined through Eqs. (S48, S49) in connection with Eqs. (S36) and (S39). The polariton branches and couplings to the electrons are shown in Fig. 1 in the main text.

#### Section S4. Migdal-Eliashberg simulations

The electronic self-energy in Migdal-Eliashberg theory on the Matsubara frequency axis employing Nambu notation reads (28)

$$\hat{\Sigma}(\vec{k}, i\omega_n) = i\omega_n[1 - Z(\vec{k}, i\omega_n)]\hat{\tau}_0 + \chi(\vec{k}, i\omega_n)\hat{\tau}_3 + \phi(\vec{k}, i\omega_n)\hat{\tau}_1 \quad (\text{S50})$$

where  $\hat{\tau}_i$  are the Pauli matrices,  $Z(\vec{k}, i\omega_n)$  and  $\chi(\vec{k}, i\omega_n)$  renormalize the electronic single-particle mass and band dispersion, respectively, and  $\phi(\vec{k}, i\omega_n)$  is the anomalous self-energy, which vanishes in the normal state. In Migdal-Eliashberg theory, the self-energy corresponding to the Hamiltonian (S46) is computed by self-consistently evaluating

$$\hat{\Sigma}(\vec{k}, i\omega_n) = \frac{-1}{N\beta} \sum_{\vec{q}, m} |g(\vec{q})|^2 D^{(0)}(\vec{q}, i\omega_n - i\omega_m) \hat{\tau}_3 \hat{G}(\vec{k} + \vec{q}, i\omega_m) \hat{\tau}_3 \quad (\text{S51})$$

where  $D^{(0)}(\vec{q}, i\omega_\nu) = -\frac{2\Omega}{\Omega^2 + \omega_\nu^2}$  is the bare phonon propagator,  $\hat{G}^{-1}(\vec{k}, i\omega_n) = i\omega_n \hat{\tau}_0 - \epsilon_{\vec{k}} \hat{\tau}_3 - \hat{\Sigma}(\vec{k}, i\omega_n)$  is the dressed electron propagator,  $N$  is number of momentum grid points, and  $\beta = 1/(k_B T)$  is the inverse temperature.

Inside the cavity with  $\omega_P > 0$ , these well-known equations are modified to account for the Hamiltonian (S47) by using polariton branches  $\lambda = \pm$  instead of the phonon

$$\hat{\Sigma}(\vec{k}, i\omega_n) = \frac{-1}{N\beta} \sum_{\vec{q}, m, \lambda=\pm} |g_\lambda(\vec{q})|^2 D_\lambda^{(0)}(\vec{q}, i\omega_n - i\omega_m) \hat{\tau}_3 \hat{G}(\vec{k} + \vec{q}, i\omega_m) \hat{\tau}_3 \quad (\text{S52})$$

where  $D_\lambda^{(0)}(\vec{q}, i\omega_\nu) = -\frac{2\omega_\lambda(\vec{q})}{\omega_\lambda(\vec{q})^2 + \omega_\nu^2}$  is the bare polariton propagator.

In practice, we use an initial guess of 0.007 eV for the anomalous self-energy and run the self-consistency until a convergence to better than  $10^{-6}$  eV is achieved. The 2D momentum grid to sample the Brillouin zone is chosen as  $2000 \times 2000$  and convergence checked by comparing against  $4000 \times 4000$  grids in selected cases. For the patch around  $q = 0$  we avoid the point  $q = 0$  where the lower polariton branch becomes soft since the corresponding propagator diverges in the static  $\omega_\nu = 0$  case. Under the  $q$  integral this divergence is cured. We therefore apply a  $q$  coarse graining by averaging  $\frac{1}{N_{\text{small}}} \sum_q |g(\vec{q})|^2 D^{(0)}(\vec{q}, i\omega_\nu)$  over  $N_{\text{small}}$



small patches ( $\tilde{\sum}_q$  is the sum inside the momentum patch around  $q = 0$ ), and using this averaged function in lieu of  $|g(0)|^2 D^{(0)}(0, i\omega_\nu)$ , again checking convergence in the momentum grid. The momentum convolution in Equations (S51) and (S52) is performed by fast Fourier transforms to a real-space grid and products on the real-space grid. The Matsubara cutoff is 0.4 eV for the frequency summations, and convergence in this cutoff also checked.