# Supporting Information (SI)

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### (SI1) Single-mode approximation in the long-wavelength limit

We point out that in full minimal coupling each mode only couples to specific Fourier components of the matter subsystem [1, 2], which is just a reflection of the fact that the interaction conserves energy and momentum. This would mean that keeping only one mode would only couple to these specific states of the matter subsystem. In the long-wavelength approximation the photon modes couple to all states that have a Fourier component in the polarization direction of the mode. In our one-dimensional situation these are all electronic states, and hence the idea of just one effective mode is reasonable.

### (SI2) Charge transfer - quasi-classical driving by light-matter correlated dynamics

Not presented here is a slightly different setup where we select an initial state which is not an eigenstate but quench a system with electronic charge stronger localized on one side. Then, the cavity couples to this state and the evolving dipole leads to an electric displacement field which in turn acts back on the electronic structure. The following behavior leads to an evolution that strongly resembles electrons driven by a laser, that is, a mostly classically dominated Maxwell propagation. Depending on the coupling strength, the driving can be so strong that we ionize the electronic system by providing a sufficiently strong back-reaction from the initial dipole quench.

### (SI3) Hopfield coefficients

The Hopfield coefficients are a measure for how much a subsystem eigenstate contributes to a many-body eigenstate. This allows to highlight which character a specific many-body state will dominantly have and therefore illustrate its behavior in terms of its constituents. The coefficients are calculated here according to

$$h^n_{i,j} = tr_{\mathrm{ee}}\left[\Gamma^{(\mathrm{n})}_{\mathrm{e}}\Gamma^{(i,j)}_{\mathrm{e}}\right],$$

with $\Gamma^{(n)}_{\mathrm{e}}$ as in Eq. (4) for the $n$-th many-body eigenstate and

$$\Gamma^{(i,j)}_{\mathrm{e}} = \tfrac{1}{2}|(\phi^{\mathrm{D}}_i \otimes \phi^{\mathrm{A}}_j + \phi^{\mathrm{A}}_j \otimes \phi^{\mathrm{D}}_i)\rangle\langle(\phi^{\mathrm{D}}_i \otimes \phi^{\mathrm{A}}_j + \phi^{\mathrm{A}}_j \otimes \phi^{\mathrm{D}}_i)|.$$

Here $\phi^{\mathrm{A/D}}_i$ are the single-particle eigenstates $i, j$. The photonic contribution in, e.g., the middle polariton, is represented by the ground-state configuration $i = j = 0$, implying a photonic excitation.

1. Greiner W, Reinhardt J (1996) *Field quantization.* (Springer).
2. Ruggenthaler M et al. (2014) Quantum-electrodynamical density-functional theory: Bridging quantum optics and electronic-structure theory. *Phys. Rev. A* 90:012508.



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