Supporting information for: Quantum Plexcitonics: A New Approach to Strongly Interacting Plasmons and Excitons

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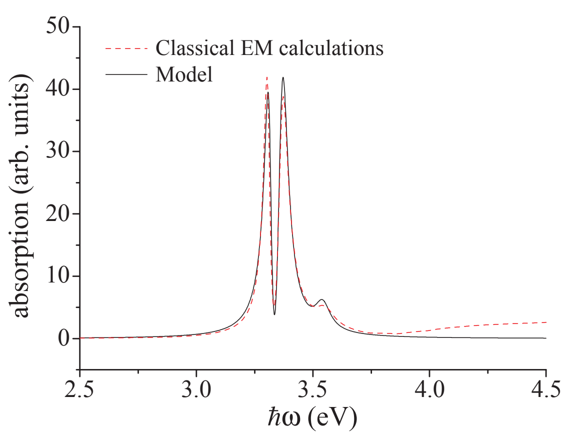

Figure 1: Comparison of the optical absorption spectrum of the dimer-quantum-emitter system obtained with classical EM theory (dashed line), and the model developed using the Zubarev's Green function method [see Eq. (19) of the main paper] (solid line). We take the emitter excitation energy $\varepsilon_c = 3.35\,\mathrm{eV}$, while the rest of the model parameters are given in Figure 1(b) of the main paper.

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Derivation of the optical absorption spectrum in terms of Zubarev's Green functions

Within the approximation of linear response, the optical absorption cross section is given by Fermi's Golden rule according to

$$\sigma(\omega) \propto \sum_f \left| \langle f; n-1 | \mathscr{H}' | i; n \rangle \right|^2 \delta(\varepsilon_f - \hbar\omega), \tag{1}$$

where $|i\rangle$ represents the initial ground state of the system under consideration; $|f\rangle$ is the corresponding final state, separated from the former by an energy difference ε_f; n is the number of external photons with frequency ω; and $\mathscr{H}'$ is the hamiltonian that couples the system to the external photon field, which is given by the expression

$$\mathscr{H}' \propto Aa^+ + A^+ a. \tag{2}$$

Here, a and A (a^+ and A^+) are annihilation (creation) operators for the external photons and the excitations of the system, respectively, and therefore, A connects the initial and final states, governing the optical absorption properties.

Using the hamiltonian of Eq. (2) and the identity

$$\delta(x) = \frac{1}{\pi} \mathrm{Im} \left\{ \frac{1}{x - i0^+} \right\}, \tag{3}$$

we can recast Eq. (1) into

$$\sigma(\omega) \propto \mathrm{Im} \left\{ \sum_f \frac{\langle i; n | \mathscr{H}' | f; n-1 \rangle \langle f; n-1 | \mathscr{H}' | i; n \rangle}{\varepsilon_f - \hbar\omega - i0^+} \right\}. \tag{4}$$

Then, taking into account the action of the photon annihilation operator ($a|n\rangle = \sqrt{n}|n-1\rangle$) and the orthogonality of the photonic states ($\langle n| |n'\rangle = \delta_{n,n'}$), we obtain

$$\sigma(\omega) \propto \mathrm{Im}\left\{\sum_f \frac{\langle i|A|f\rangle \langle f|A^+|i\rangle}{\varepsilon_f - \hbar\omega - i0^+}\right\}. \tag{5}$$

At this point, we assume that A^+ connects the initial ground state with a set of final states that present a common energy ε_f. This is a good approximation for the systems considered in this paper, in which the external photons couple through excitation of either a single particle plasmon or two degenerate plasmons in a dimer. Therefore, we can extract the denominator of Eq. (5) from the sum over final states, and using the closure relation for final states $\sum_f |f\rangle \langle f| = \mathbb{I}$ (within the ε_f reachable-energy shell), the optical absorption spectrum reduces to

$$\sigma(\omega) \propto \mathrm{Im}\left\{\frac{\langle i|AA^+|i\rangle}{\varepsilon_f - \hbar\omega - i0^+}\right\}, \tag{6}$$

where $\langle i|AA^+|i\rangle$ is the expectation value of AA^+ in the initial ground state.

We now consider the definition of the retarded Zubarev Green function for the operators that annihilate and create a system excitation [see Eq. (1) of the main paper]:

$$\langle\langle A;A^+\rangle\rangle_{\omega+i0^+} = -\frac{i}{\hbar}\int_0^\infty dt e^{i(\omega+i0^+)t}\left[\left\langle A(t)A^+(0)\right\rangle - \eta\left\langle A^+(0)A(t)\right\rangle\right], \tag{7}$$

where we have switched to the Heisemberg picture, and therefore, the free evolution of the operator A is given by the Heisemberg equation of motion,[1] which for fixed excitation energy ε_f leads to

$$A(t) = A(0)e^{-i\varepsilon_f t/\hbar}. \tag{8}$$

This is consistent with the noted assumption of a single final-state energy ε_f. Eq. (8) allows us to solve the integral of Eq. (7). We find

$$\langle\langle A;A^+\rangle\rangle_{\omega+i0^+} = -\frac{\langle A(0)A^+(0)\rangle}{\varepsilon_f - \hbar\omega - i0^+}, \tag{9}$$

where we have taken into account that the second term of Eq. (7) vanishes when the system is in the ground state. Finally, noticing that $\langle i|AA^{+}|i\rangle$ and $\langle A(0)A^{+}(0)\rangle$ are both the same magnitude in the Schrödinger and the Heinsemberg picture, respectively, we can substitute Eq. (9) into Eq. (6) to obtain the desired expression [see Eq. (2) of the main paper].

Derivation of the equation of motion for the retarded Zubarev's Green functions

The retarded Zubarev's Green function in the time domain can be obtained from the Fourier transform of Eq. (1) of the main paper, as

$$\langle\langle A(t);B(0)\rangle\rangle = -\frac{i}{\hbar}\theta(t)\left\langle [A(t),B(0)]_{\eta}\right\rangle. \tag{10}$$

Differentiating with respect to the time, we find

$$i\frac{d}{dt}\langle\langle A(t);B(0)\rangle\rangle = \frac{1}{\hbar}\frac{d\theta(t)}{dt}\left\langle [A(t),B(0)]_{\eta}\right\rangle + \langle\langle i\frac{dA(t)}{dt};B(0)\rangle\rangle, \tag{11}$$

which can be simplified using the Heisenberg equation of motion[1] to yield

$$i\hbar\frac{d}{dt}\langle\langle A(t);B(0)\rangle\rangle = \delta(t)\left\langle [A(t),B(0)]_{\eta}\right\rangle + \langle\langle [A(t),\mathscr{H}];B(0)\rangle\rangle. \tag{12}$$

Here, $\mathscr{H}$ is the hamiltonian of the system. Finally, taking the Fourier transform of Eq. (12), we readily obtain the desired equation of motion for for the retarded Zubarev's Green functions [see Eq. (3) of the main paper].

References

(1) A. Messiah, *Quantum Mechanics* (Dover, New York, 1999)