Coulomb-assisted cavity feeding in the non-resonant optical emission from a quantum dot

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Recent experiments have demonstrated that for a quantum dot in an optical resonator off-resonant cavity mode emission can occur even for detunings of the order of 10 meV. We show that Coulomb mediated Auger processes based on additional carriers in delocalized states can facilitate this far off-resonant emission. Using a novel theoretical approach for a non-perturbative treatment of the Auger-assisted quantum-dot carrier recombination, we present numerical calculations of the far off-resonant cavity feeding rate and cavity mean photon number confirming efficient coupling at higher densities of carriers in the delocalized states. In comparison to fast Auger-like intraband scattering processes, we find a reduced overall efficiency of Coulomb-mediated interband transitions due the required electron-hole correlations for the recombination processes.

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Semiconductor quantum-dot (QD) microcavity devices offer many applications of strong current interest, such as lasers with improved emission properties and integrated sources of indistinguishable and entangled photons. In contrast to atomic-like isolated emitters, QDs exhibit an interesting peculiarity: even if the QD emission lines are significantly detuned from the cavity resonance, photons can be emitted into the cavity mode. As a result of intense experimental and theoretical investigations, different mechanisms are discussed in the literature. Firstly, the interaction of QD excitons with acoustic phonons has been shown to provide efficient off-resonant cavity feeding for an energetic mismatch of few meV. Secondly, when a QD can accommodate many single-particle bound states, the number of carrier configurations becomes quite large, and their Coulomb interaction results in a broad quasi-continuum of multie exciton transitions. Provided that these multi-exciton states are excited, their overlap with the cavity mode allows for a Purcell-enhanced photon production at larger detunings (~10 meV away from the single exciton line). Also the role of the interaction with the wetting layer (WL) in the formation of this multie exciton spectral background was recognized and Coulomb hybridization of QD bound states with the WL continuum was demonstrated.

In this letter, we quantify the role of Coulomb interaction with WL carriers for the off-resonant coupling of QD transitions to a cavity mode. Specifically, carriers in the WL can act as a thermal bath which compensate for the energy mismatch via Auger-like processes. We point out that this is an alternative mechanism to the Coulomb configuration interaction between carriers, as we consider the role of the Coulomb interaction not in the spectrum but in the dynamics of the exciton recombination. Its effect in opening a kinetic channel is present even for QDs hosting very few confined states, and as a proof of principle, we evaluate it for a QD with single electron and hole levels.

We find that optical interband transitions assisted by WL carriers via Coulomb interaction can provide off-resonant coupling, however, with smaller rates in comparison to ultrafast intraband Coulomb relaxation processes, which are known to allow carriers to efficiently bridge several tens of meV. We trace back the reason for the difference to electron-hole correlation effects.

To describe the system, we start from the Hamiltonian containing the Jaynes-Cummings (JC) part for the interaction of the QD exciton with photons of the cavity mode as well as the Coulomb part for the interaction between QD and WL states. Two different techniques are used to formally eliminate either (i) the exciton-photon or (ii) the Coulomb interaction part from the Hamiltonian. Specifically, the Schrieffer-Wolff transformation is used that amounts to a perturbative diagonalization of the JC interaction part. As an alternative second method, we exploit a novel approach, involving a unitary transform which is related to the one used in the independent boson model (IBM) for the treatment of the carrier-phonon interaction, but is now applied to the fermionic bath of WL carriers.

The cavity-QD system in the presence of the continuum of WL states is described by the Hamiltonian (see Appendix [A]):

\[ H = \varepsilon_X X^\dagger X + \omega b^\dagger b + g b^\dagger X + b X^\dagger + (1 - X^\dagger X) h_0 + X^\dagger X h_X, \]

\[ h_0 = \sum_{\lambda,k} \varepsilon_{k}^\lambda \lambda_k^\lambda, \quad h_X = h_0 + W, \]  

where \( X \) and \( X^\dagger \) are the exciton annihilation and creation operators corresponding to the QD-bound electron and hole s-states, \( b, b^\dagger \) are the photon operators, \( \varepsilon_X \) and \( \omega \) are the exciton and cavity mode energy, \( \lambda_k, \lambda_k^\dagger \) are the fermionic operators referring to the WL \( k \)-states (including the spin implicitly) for electrons and holes (\( \lambda = e, h \)) with the energies \( \varepsilon_k^\lambda \), and \( g \) is the JC coupling strength. In the presence of the exciton, the WL free Hamiltonian...
\( h_0 \) is modified by the interaction of the WL carriers with the electron-hole pair, described by the operator
\[
W = \sum_{\lambda,k,k'} W^\lambda_{kk'} \lambda_k \lambda_{k'},
\]
where
\[
W_{kk'} = W^{ce}_{kk'} - W^{he}_{kk'} - W^{ce}_{k,k'},
\]
\[
W_{kk'} = W^{eh}_{kk'} - W^{ce}_{k,k'} + W^{he}_{k,k'}.
\]
The interaction matrix elements \( W_{ij,kl}^{\lambda \lambda'} \) are given by
\[
\frac{1}{\pi} \sum_k w_\lambda \langle \phi_\lambda | e^{i\mathbf{q}\cdot\mathbf{r}} | \phi_{\lambda'} \rangle = \text{the static screening Coulomb potential} w_\lambda \text{contains the single particle states} |\phi_\lambda \rangle \text{of electrons and holes in the QD-WL system (see Ref.\textsuperscript{13}). The first two terms in the equations (3) and (4) represent the electrostatic (Hartree) interaction of the WL carriers with the exciton and cavity, which leads to an increasing feeding rate.}

The idea of the Schrieffer-Wolff approach is to use a unitary transformation: \( H' = e^{S} H e^{-S} = H + [S, H] + \frac{1}{2} [S, [S, H]] + \ldots \), with \( S \) selected such that the JC Hamiltonian is formally eliminated. This can be achieved by choosing \( S = -\frac{\kappa}{\Delta} (b\dagger X - b X\dagger) \), which shows that the generator \( S \) is of first order in a series expansion in the small parameter \( g/\Delta \), with \( \Delta = \varepsilon_X - \omega \) is the exciton-cavity detuning. Typically \( g \) is of the order of 0.1 meV and for large detunings \( \Delta \approx 1 \) to 10 meV a perturbative approach is justified. Neglecting terms of higher order than \( (g/\Delta)^2 \), and \( (g/\Delta)^4 \) one is lead to an effective JC interaction Hamiltonian
\[
H_{\text{int,SWA}} = -\frac{\kappa}{\Delta} W (b\dagger X + b X\dagger)
\]
that describes transitions between the QD exciton and the cavity photons, assisted by the Coulomb interaction with WL carriers.

When evaluating the QD exciton and photon dynamics under the influence of \( H_{\text{int,SWA}} \), the quasi-continuous nature of the delocalized WL-states allows for treating them as a bath. We consider the fermionic WL reservoir as being stationary and in thermal equilibrium. Following the standard Born-Markov approach\textsuperscript{22,23} for the system-bath interaction, we find a new Lindblad terms, \( L_{\xi,X} \) and \( L_{\zeta,X} \), with rates given by the Fourier transform of the reservoir correlator \( \langle W(t)W(0) \rangle - \langle W \rangle^2 \), taken at the energy \( \Delta \) lost in the transition corresponding to the exciton recombination. As a result we obtain
\[
\gamma_{\xi,X} = 2\pi \frac{\kappa^2}{\Delta} \sum_{\lambda,k,k'} |W^{\lambda}_{kk'}|^2 f_{k'} (1 - f_{k'}) \delta(\Delta + \varepsilon_{\lambda_{kk'}}),
\]
Here \( \varepsilon_{\lambda_{kk'}} = \varepsilon_{\lambda} - \varepsilon_k \), and the occupancies \( f_{k'} = \langle \lambda_{k'} \rangle \) are Fermi functions describing the WL carrier population. Similarly, \( \gamma_{\zeta,X} \) follows by changing \( \Delta \rightarrow -\Delta \). The first-order mean-field contribution of \( H'_{\text{int,SWA}} \) maintains the form of the JC Hamiltonian, but with a renormalized coupling strength \( \tilde{g} = \frac{g}{\Delta} \langle X \rangle \), where \( \langle X \rangle = -\sum_{\lambda,k} W^{\lambda}_{kk} f_{k} \). Consequently, the Schrieffer-Wolff procedure modifies the coherent energy exchange between the exciton and the cavity and at the same time gives rise to an incoherent exchange in the form of a new type of Lindblad terms.

In Fig. 1 we address the dependence of the Coulomb assisted cavity feeding rate \( \gamma_{\xi,X} \) on the detuning \( \Delta \) between exciton and cavity mode for different carrier densities \( n_{WL} \) in the delocalized WL states of at a temperature of 77K. In the calculation, we have used InGaAs parameters\textsuperscript{24} and assume a flat lens-shaped QD on a WL. For the QD wave functions of the energetically lowest confined states \( \phi_{\lambda}^a(r) \) Gaussian functions with standard deviation \( \alpha_{\lambda} \) are applied for the in-plane motion and in growth direction the solution of a finite height potential well is assumed. For the WL states \( \phi_{\lambda}^a(r) \) orthogonalized plane waves are used\textsuperscript{16}. With increasing WL-carrier density additional scattering channels can compensate for the energetic mismatch \( \Delta \) between exciton and cavity, which leads to an increasing feeding rate. For comparison, we show the spontaneous emission rate \( R = 4g^2(\kappa + \Gamma Ps)/[(\kappa + \Gamma Ps)^2 + (2\Delta)^2] \) caused by the JC coupling alone for typical QD-cavity parameter. At low carrier density Coulomb assisted cavity feeding is neg-
ligible in comparison to the spontaneous emission rate, which is in agreement with previous experiments performed under low excitation condition[24], in which only phonon signatures were found. However, for sufficiently high carrier densities (nWL > 10^{10}/cm^2) Coulomb assisted processes prevail even at large detuning and lead to a significant cavity feeding that is one order of magnitude stronger in comparison to the JC coupling alone. Secondly, there is a pronounced asymmetry between positive and negative detuning for low temperature. This is expected, since any thermal bath favors the process which lowers the system energy, all the more so at low temperatures.

Finally, we find a significant reduction of the off-resonant cavity feeding, if the wave functions for electrons and holes are similar (lines without dots). The reason is a large degree of compensation between the electrostatic (Hartree) Coulomb integrals contributing to Eq. (6). For identical electron and hole wave functions the exciton is bistable QD (lines without dots). A calculation beyond the SPA, as the perturbation acting on the WL is the Coulomb interaction part from the Hamiltonian with a unitary transform U = 1 − X^†X + X^†X S(0, −∞), which is in agreement with previous experiments performed under low excitation condition. The compensation effect between electron and hole positions: In the former case electrons and holes can scatter independently, while in the latter case the emission of a photon requires the presence of an exciton, i.e., a fully correlated electron-hole pair. As a consequence any formalism describing the off-resonant cavity feeding, which relies on an interaction of Coulombian origin, like QD-WL Auger interaction, or Fröhlich interaction of QD carriers with LO phonons, must obey the following local neutrality condition: for locally neutral excitons and discarding the exchange terms the off-resonant process should vanish exactly.

So far, we have presented results based on an approximate, perturbative diagonalization of the JC interaction part of the Hamiltonian. As an alternative approach, we introduce a non-perturbative treatment of the Coulomb interaction between QD excitons and WL carriers by using a suitable unitary transform. The transform is in many ways similar to the polaron transform of the IBM. The presence of the exciton generates an external field to which the lattice ions react by displacements of their oscillation centers. The polaronic transform connects the distorted lattice with the original one. Similarly, in the present case the exciton perturbs the WL-carrier system by an external field term W. The source of this field is localized around the QD, which thus acts as a scattering center for the continuum of extended WL states. The associated S-matrix is the unitary transform that connects h_X and h_0 (see e.g. 25). Even though the Coulomb interaction with the WL states cannot be treated exactly any more, it lends itself to a diagrammatic expansion. We term the method “scattering potential approach” (SPA). Specifically, one has

\[ \h_0 = \mathcal{S}(-\infty, 0) h_X \mathcal{S}(0, -\infty), \]  

with \( S \) generated by the scattering potential \( W \)

\[ S(t_1, t_2) = S(t_1 - t_2) = \mathcal{S}^\dagger(t_2, t_1) = T \exp \left[ -i \int_{t_2}^{t_1} W(t) \, dt \right], \quad t_1 > t_2, \]  

assuming that the requirements of the scattering theory are fulfilled 26. \( T \) is the time ordering operator and the interaction representation of the perturbation \( W(t) \) with respect to \( h_0 \) is used. We formally eliminate the QD-WL Coulomb interaction part from the Hamiltonian with a unitary transform \( U = 1 − X^†X + X^†X S(0, −\infty) \), which is in agreement with previous experiments performed under low excitation condition. The interaction with the WL appears now in the light-matter coupling term by the presence of the S-matrix

\[ H'_{\text{int,SPA}} = g \left[ b^\dagger X S(0, −\infty) + b X^\dagger S(−\infty, 0) \right] . \]  

The standard Born-Markov treatment of the system-reservoir interaction with respect to \( H'_{\text{int,SPA}} \) leads to Lindblad terms of the form \( \mathcal{L}_{b^\dagger X} \) and \( \mathcal{L}_{b X^\dagger} \). For the off-resonant cavity feeding rate \( \gamma_{b^\dagger X} \), we obtain

\[ \gamma_{b^\dagger X} = 2g^2 e^{\beta \Delta} \text{Re} \int_0^\infty e^{-i\Delta t} \langle \mathcal{S}(t, 0) \rangle \, dt, \]  

with the inverse temperature \( \beta = 1/(k_B T) \). The evaluation of \( \langle \mathcal{S}(t, 0) \rangle \) can be done using the linked cluster (cumulant) expansion 23-25. Details about the derivation are included in Appendix B.

In Fig. 2 we show the steady-state mean photon number for a typical QD-cavity system and compare the results obtained by the SWA (black line) and the SPA (red dash dotted line) for a high WL carrier density of \( n_{WL} = 10^{12}/cm^2 \), where we expect a pronounced off-resonant cavity feeding. To this end, we solve the time evolution of the system density operator by the von-Neumann Lindblad equation \( \frac{d}{dt} \rho = -i [H_{\text{sys}}, \rho] + \sum_{\mathcal{L}} \mathcal{L}(\rho) \) and include the developed cavity feeding Lindblad terms \( \mathcal{L}_{b^\dagger X} \) and \( \mathcal{L}_{b X^\dagger} \). We find a good qualitative agreement between both methods for large detunings. As an advantage, the non-perturbative SPA allows to extend the results to small detuning values and includes additional WL-induced renormalizations.

The compensation effect between electron and hole potentials that we discussed for the SWA also holds for the SPA, as the perturbation acting on the WL is the same one-particle operator \( W \). Thus, electrons and holes are not contributing independently, and this results in a reduced cavity mean photon number for a locally neutral QD (lines without dots). A calculation beyond the second-order Born-Markov approach for the system-bath treatment, e.g. by using quantum kinetic methods, would
A carrier density of $10^{12}$ cm$^{-2}$ and holes. Shown is the steady state mean photon number for the SPA result, as a consequence of uncorrelated electrons and holes. In Fig. 3: Influence of artificial renormalization/broadening of the SPA result, as a consequence of uncorrelated electrons and holes. Shown is the steady state mean photon number for a carrier density of $10^{12}$ cm$^{-2}$ by using the SWA (solid) and the SPA (dashed dotted) for a temperature of 77 K. Additionally, results for non-equal envelopes (lines with dots) and pure JC coupling (dotted) are shown. Typical parameters are used ($g = 0.1$ ps, $\kappa = 0.1$ ps, $\Gamma = 0.01$ ps), $P = 0.1$ ps.

Figure 2: Steady state cavity mean photon number generated by an off-resonant QD mediated by the continuum of WL states for a carrier density of $10^{12}$/cm$^2$ by using the SWA (solid) and the SPA (dashed dotted) for a temperature of 77 K. Additionally, results for non-equal envelopes (lines with dots) and pure JC coupling (dotted) are shown. Typical parameters are used ($g = 0.1$ ps, $\kappa = 0.1$ ps, $\Gamma = 0.01$ ps), $P = 0.1$ ps.

In conclusion, for the off-resonant coupling between QD emitters and optical cavity modes, the Auger-like recombination assisted by WL carriers has been identified as a possible cavity feeding channel. It coexists with other processes like phonon-assisted recombination, which is only efficient for small detuning, or multi-exciton effects, which require QDs with several shells. For large carrier densities in the delocalized states, we have demonstrated efficient coupling at large detunings up to 10 meV for a QD with only one confined shell. The off-resonant coupling of QD emitters has profound implications for the description and characterization of QD microcavity lasers, where a small number of close to resonant emitters are accompanied by many far-detuned emitters. Their background emission can enhance the photon production rate sufficiently to result in a reduced laser threshold and modified photon statistics. Finally, we point out that the described assisted interband scattering processes, while significant, are found to be much smaller in comparison to the intraband-relaxation processes. The reason lies in the correlation between electron and hole, that is required for the interband recombination, but not for the intraband scattering process.

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Appendix A: The exciton-WL interaction

The Coulomb interaction between the QD and the WL carriers is expressed as

$$ W = e^4 \epsilon \sum_{k,k'} \left[ W_{ek,k'e_k'k'} + W_{ek,k'e_k'k'}^h h_{k'k}^\dagger - W_{ek,k'e_k'k'}^e e_{k,k'}^\dagger - W_{ek,k'e_k'k'}^h h_{k'k}^\dagger \right] + \hbar^4 \sum_{k,k'} \left[ W_{ek,k'e_k'k'}^h h_{k'k}^\dagger h_{k'k}^\dagger - W_{ek,k'e_k'k'}^e e_{k,k'}^\dagger e_{k,k'}^\dagger - W_{ek,k'e_k'k'}^h h_{k'k}^\dagger h_{k'k}^\dagger \right]. $$

(A1)

The subscripts $k, k'$ describe WL continuum states, while electron $e, e^\dagger$ and hole $h, h^\dagger$ operators without subscripts correspond to the QD-localized $s$-states. The two lines involve quasi-particle properties with complex spectral structure instead of the free-particle energies. In the simplest approximation this leads to a Lorentzian broadening of the electron and hole energy spectrum. However, the renormalization of the exciton energy should also take into account the correlations between the two constituent particles and especially reflect the reduced QD-WL Coulomb interaction for a locally neutral exciton. In this case the exciton renormalization should be considerably smaller in comparison to the sum of independently broadened electron and hole energies. In Fig. 3 we show how artificially uncorrelated electrons and holes would influence the cavity feeding. To this end we introduce an additional renormalization to the QD exciton energy in Eq. (B5) by replacing $\epsilon_X \rightarrow \epsilon_X + i\gamma$, where $\gamma = \gamma_e + \gamma_h$ is the sum of the independently broadened electron and hole energies. It is seen that by increasing the total broadening $\gamma$ the off-resonant cavity feeding gains substantially. Being an artifact, this result underscores the need for the correct treatment of correlations between the electron and hole, which reduce the efficiency of interband transitions assisted by wetting layer carriers.
in Eq. (A1) define the contribution from the QD electron and hole respectively.

The Hilbert space of our problem is limited to neutral states. In other words the QD electron and hole are either both absent or both present, which prohibits an approach where Lindblad terms are derive from each of the two lines of Eq. (A1) separately, because the excitonic pair is highly correlated. This is taken into account by having $e^\dagger e = h^2 c = X^\dagger X$, where $X = \hbar c$, and leads to the exciton-WL Hamiltonian considered in the paper, which accumulates the terms of both lines. The ensuing compensation between the fields created by the exciton carriers, and its consequences, are discussed in the paper.

**Appendix B: Scattering potential approach**

We formally eliminate the QD-WL Coulomb interaction part from the Hamiltonian

$$H = \varepsilon X^\dagger X + \omega b^\dagger b + g(b^\dagger X + bX^\dagger) + (1 - X^\dagger X) h_0 + X^\dagger X h_X ,$$

(B1)

using the unitary transform

$$U = 1 - X^\dagger X + X^\dagger X S(0, -\infty) ,$$

(B2)

with the properties that $X' = U^\dagger X U = X S(0, -\infty)$, $(X^\dagger)' = S(-\infty, 0) X^\dagger$, while $X^\dagger X$ remains invariant. The important changes concern the WL part of the Hamiltonian of Eq. (B1)

$$U^\dagger [(1 - X^\dagger X) h_0 + X^\dagger X h_X] U = (1 - X^\dagger X) h_0 + X^\dagger X S(-\infty, 0) h_X S(0, -\infty) = h_0 ,$$

(B3)

from which the interaction is now removed. The same role is played by the polaron transform in the IBM.

For the transformed light-matter coupling Hamiltonian we obtain

$$H'_{\text{int,SPA}} = g b^\dagger X S(0, -\infty) + b X^\dagger S(-\infty, 0) .$$

(B4)

The standard Born-Markov treatment of the system-reservoir interaction with respect to $H'_{\text{int,SPA}}$ leads to Lindblad terms of the form $L_{b^\dagger X}$ and $L_{b X^\dagger}$, where the corresponding rates $\gamma_{b^\dagger X}$ and $\gamma_{b X^\dagger}$ are

$$\gamma_{b^\dagger X} = g^2 \int_{-\infty}^{\infty} e^{i\Delta t} \langle e^{ithat S(-\infty, 0)} e^{-ithat S(0, -\infty)} \rangle dt ,$$

$$\gamma_{b X^\dagger} = g^2 \int_{-\infty}^{\infty} e^{-i\Delta t} \langle e^{ithat S(0, -\infty)} e^{-ithat S(-\infty, 0)} \rangle dt ,$$

(B5)

with the averages taken over the thermal equilibrium of $h_0$. The rates obey the Kubo-Martin-Schwinger (KMS) relation

$$\gamma_{b^\dagger X} = e^{\beta \Delta} \gamma_{b X^\dagger}$$

(B6)

with the inverse temperature $\beta = 1/(k_B T)$. Therefore it is sufficient to compute only one of these rates. The simplest one is $\gamma_{b X^\dagger}$. The correlator $\langle e^{ithat S(0, -\infty)} e^{-ithat S(-\infty, 0)} \rangle$ can be rewritten as $\langle S(t, 0) \rangle$ by using successively $S(0, -\infty) e^{-ithat} = e^{-ithat} S(0, -\infty)$, in accordance with the unitary equivalence between $h_X$ and $h_0$

$$S(0, -\infty) h_0 = h_X S(0, -\infty) ,$$

(B7)

$e^{ithat} e^{-ithat t} = S(t, 0)$, and the semigroup property of the $S$-matrix. For the off-resonant cavity feeding rate $\gamma_{b^\dagger X}$, we obtain

$$\gamma_{b^\dagger X} = 2g^2 e^{\beta \Delta} \Re \int_{0}^{\infty} e^{-i\Delta t} \langle S(t, 0) \rangle dt ,$$

(B8)

where we have also used that $\langle S(-t, 0) \rangle = \langle S(0, t) \rangle = \langle S(t, 0) \rangle^\ast$.

The evaluation of $\langle S(t, 0) \rangle$ can be done using the linked cluster (cumulant) expansion. This is expressed as $\langle S(t, 0) \rangle = \exp[\Phi(t)]$, where $\Phi(t) = \sum_n L_n(t)$ is the sum over all connected diagrams $L_n$ with no external points, having $n$ internal ones and carrying a prefactor $1/n$. The internal points run in time from $0$ to $t$. Here, the interaction is an external potential, not a many-body one, and the elementary interaction vertex, $W_{\lambda k, k'}$, is represented in Fig. 1(a). The first terms in the linked cluster expansion are shown in Fig. 1(b). In what follows we restrict the calculation to the first two diagrams. Note that in the case of interaction with phonons the result is exact, because only the corresponding $L_2$ diagram is present in the IBM theory. One has

$$L_1(t) = -\sum_{\lambda, k} \int_{0}^{t} dt_1 W_{\lambda k, k}^\lambda G_{\lambda k}^0(t_1, t_1^+)$$

$$L_2(t) = -\frac{1}{2} \sum_{\lambda, k, k'} \left| W_{\lambda k, k'}^\lambda \right|^2$$

$$\times \int_{0}^{t} dt_1 \int_{0}^{t} dt_2 G_{\lambda k}^0(t_1, t_2) G_{\lambda k'}^0(t_2, t_1) ,$$

(B9)
where $G^0_{\lambda k}$ is the free Green’s function for the WL state $\lambda k$ and $t_1^+$ is infinitesimally later than $t_1$. Evaluating the time integration finally leads to

$$L_1(t) = -i \sum_{\lambda k} W^{\lambda}_{k,k} f^\lambda_k t$$

$$L_2(t) = \sum_{\lambda k,k'} \left| W^{\lambda}_{k,k'} \right|^2 (1 - f^\lambda_k) f^\lambda_{k'} \times \left( e^{-i\varepsilon_{kk'} t} - 1 + i \varepsilon_{kk'} t \right) \left( \varepsilon_{kk'} \right)^{-2}.$$ 

$L_1$ is purely imaginary and amounts to a shift of the excitonic energy, which we include in the detuning $\Delta$. Further renormalizations occur due to the imaginary part of the $L_2$ term. The real part of the latter shows a linear long-time asymptotics, according to $\langle \cos(\epsilon t) \rangle = -t \sin^2(\epsilon t/2)/\epsilon^2 t/2$ which for large times behaves like $-\pi \delta(\epsilon)$. This gives rise to an exponential decay of $\langle S(t,0) \rangle$ as $t \to \infty$, which implies a vanishing first-order mean-field contribution of the system-bath interaction $H_{\text{int},\text{SPA}}$. An exponential behavior of $\langle S(t,0) \rangle$ suggests a slow (Lorentzian) decay of its Fourier transform which would entail, according to Eq. (13), a divergent rate $\gamma_{b|X}$ in the limit of large $\Delta$. This is prevented by the quadratic behavior of $L_2$, and the corresponding Gaussian shape of $\langle S(t,0) \rangle$ in the low-time regime.

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