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Radiative and phonon-induced dephasing in double quantum dots

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Abstract. A simple method for describing the evolution of a quantum state of a double quantum dot system interacting simultaneously with the electromagnetic environment and with the lattice modes is developed. It is shown that the joint action of the two reservoirs leads to nontrivial effects in the system dephasing. As an example, the impact of phonon-induced initial dephasing on the radiative decay of delocalized exciton states is discussed.

1. Introduction
Double quantum dots (DQDs) are systems composed of two quantum dots placed close enough to each other for new coherent and collective phenomena to occur which cannot be reduced to the known properties of individual dots. Such new phenomena related to the optical properties of the system include modification of the optical response due to inter-dot coupling [1; 2] or collective effects in the spontaneous emission [3; 4]. Another class of effects is related with carrier-phonon interaction. Examples include phonon-assisted exciton transfer between the dots [5–7], modification of optical spectra due to phonon packets traveling between the dots [8], or phonon-induced decay of entanglement [9]. The special properties of DQDs may open the way to new applications, like long-time storage of quantum information [10], conditional optical control [1] that may lead to an implementation of a two-qubit quantum gate [11], generation of entangled photons [12] or coherent optical spin control and entangling [13–15]. Although both spontaneous emission and phonon-related phenomena have been studied to some extent for DQD systems, the interplay between these two classes of effects has not been investigated.

In the present contribution, we formulate a theoretical description of the evolution of a double quantum dot coupled simultaneously to the two reservoirs: the quantum electromagnetic field (photon vacuum) and lattice vibrational modes (phonons). After defining the model (Sec. 2), the approach to the simulation of the quantum open system is presented (Sec. 3), followed by an example of an interplay between the two couplings (Sec. 4).

2. The system
The system under study is composed of two stacked self-assembled QDs with transition energies $\epsilon_1$ and $\epsilon_2$, interacting with their phonon and photon (radiative) environments. We restrict the discussion to the ground states of excitons in each dot and assume that the spin polarizations of the excitons are fixed. As the exciton dissociation energy in absence of external electric fields is rather large (several to a few tens of meV), we consider only spatially direct exciton states, i.e., such that the electron-hole pairs reside in one and the same dot.
Thus, the model includes four basis states: the state without excitons, \(|0\rangle\), the states \(|1\rangle\) and \(|2\rangle\) with an exciton in the first and second dot, respectively, and the ‘molecular biexciton’ state \(|3\rangle\) with excitons in both dots. The system evolution will be described in a ‘rotating basis’ defined by the unitary transformation

\[
U = e^{iEt(\langle 1|1|1\rangle + |2\rangle|2\rangle + |3\rangle|3\rangle)/\hbar},
\]

where \(E = (\epsilon_1 + \epsilon_2)/2\). Note that this is not equivalent to interaction picture with respect to the Hamiltonian of uncoupled dots. Upon the standard weak-coupling derivation of the Master equation, the latter would always lead to uncorrelated emission for non-identical dots. On the contrary, the present transformation allows us to explicitly keep the energy difference \(\Delta = (\epsilon_1 - \epsilon_2)/2\) which yields unitary evolution superposed on the dissipative one, consistent with the result of the Weisskopf–Wigner approach \[3\], and allows one to correctly account for the transition from “identical dots” to “different dots”. The Hamiltonian is then

\[
H = H_{\text{DQD}} + H_{\text{ph}} + H_{\text{rad}} + H_{\text{c-\text{ph}}} + H_{\text{c-\text{rad}}}.
\]

The first term describes exciton states in the DQD structure,

\[
H_{\text{DQD}} = \Delta (|1\rangle\langle 1| - |2\rangle\langle 2|) + V (|1\rangle\langle 2| + |2\rangle\langle 1|),
\]

where \(V\) is the coupling between the dots, which may originate either from the Coulomb (Förster) interaction or from tunnel coupling (it can be assumed real). It is convenient to introduce the parametrization \(\Delta = \mathcal{E}\cos 2\theta, \ V = \mathcal{E}\sin 2\theta\), where \(2\mathcal{E}\) is the energy splitting between the single-exciton eigenstates of \(H_{\text{DQD}}\) and \(\theta\) is the mixing angle of the single-exciton states. Electron and hole wave functions are modelled by identical anisotropic Gaussians with identical extensions \(l\) in the \(xy\) plane and \(l_z\) along \(z\) for both particles,

\[
\psi_{1,2}(r) \sim \exp \left[ -\frac{1}{2} \frac{x^2 + y^2}{l^2} - \frac{1}{2} \frac{(z \pm D/2)^2}{l_z^2} \right],
\]

where \(D\) is the distance between the dots.

The phonon modes are described by the free phonon Hamiltonian \(H_{\text{ph}} = \sum_k \hbar \omega_k b_k^\dagger b_k\), where \(b_k, b_k^\dagger\) are bosonic operators of the phonon modes and \(\omega_k\) are the corresponding frequencies. Interaction of carriers confined in the DQD with phonons is modelled by the independent boson Hamiltonian

\[
H_{\text{c-\text{ph}}} = (|1\rangle\langle 1| + |3\rangle\langle 3|) \sum_k f_k^{(1)} (b_k^\dagger + b_{-k}) + (|2\rangle\langle 2| + |3\rangle\langle 3|) \sum_k f_k^{(2)} (b_k^\dagger - b_{-k}),
\]

where \(f_k^{(1,2)}\) are system-reservoir coupling constants. For Gaussian wave functions, the coupling constants for the deformation potential coupling between confined charges and longitudinal phonon modes have the form \(f_k^{(1,2)} = f_k e^{\pm ik_z D/2}\), where

\[
f_k = (\sigma_e - \sigma_h) \sqrt{k} \sqrt{2\pi} \frac{l_z^2 k_z^2}{4} \exp \left[ -\frac{l_z^2 k_z^2}{4} \right].
\]

Here \(v\) is the normalization volume, \(k_{l/z}\) are momentum components in the \(xy\) plane and along the \(z\) axis, \(\sigma_e/h\) are deformation potential constants for electrons/holes, \(c_l\) is the speed of longitudinal sound, and \(\varrho\) is the crystal density. We assume that off-diagonal carrier–phonon couplings are negligible due to small overlap of the wave functions confined in different dots.
The third component in our modeling is the radiative reservoir (modes of the electromagnetic field), described by the Hamiltonian $H_{\text{rad}} = \sum_{\mathbf{k},\lambda} \hbar w_{\mathbf{k}\lambda} c_{\mathbf{k},\lambda}^\dagger c_{\mathbf{k},\lambda}$, where $c_{\mathbf{k},\lambda}, c_{\mathbf{k},\lambda}^\dagger$ are photon creation and annihilation operators and $w_{\mathbf{k}}$ are the corresponding frequencies ($\lambda$ denotes polarizations). The QDs are separated by a distance much smaller than the relevant photon wavelength $\lambda = 2\pi \hbar c / E$, so that the spatial dependence of the EM field may be neglected (theDicke limit). The Hamiltonian describing the interaction of carriers with the EM modes in the dipole and rotating wave approximations is

$$H_{\text{c-rad}} = \Sigma_- \sum_{\mathbf{k},\lambda} e^{-iEt/\hbar} g_{\mathbf{k}\lambda} c_{\mathbf{k},\lambda}^\dagger + \text{H.c.},$$

with $\Sigma_- = |0\rangle\langle 1| + |2\rangle\langle 3| + |0\rangle\langle 2| + |1\rangle\langle 3|$ and $g_{\mathbf{k}\lambda} = i \mathbf{d} \cdot \hat{\epsilon}_\lambda(\mathbf{k}) \sqrt{\frac{\hbar w_{\mathbf{k}}}{2\pi \epsilon_0 c^2}}$, where $\mathbf{d}$ is the interband dipole moment, $\epsilon_0$ is the vacuum permittivity, $\epsilon_\lambda$ is the dielectric constant of the semiconductor and $\hat{\epsilon}_\lambda(\mathbf{k})$ is the unit polarization vector of the photon mode with the wave vector $\mathbf{k}$ and polarization $\lambda$. For wide-gap semiconductors with $E \sim 1$ eV, zero-temperature approximation may be used for the radiation reservoir at any reasonable temperature.

3. The system evolution

In certain limiting cases, analytical formulas for the evolution of the DQD system may be found. For uncoupled dots ($V = 0$) interacting only with lattice modes (phonons), an exact solution is available [9]. If only the radiative decay is included, a solution in the Markov limit can be obtained [3]. Here, we propose a general description which allows one to deal with the simultaneous action of both these environments. We describe the evolution of the reduced density matrix of the DQD system in the interaction picture with respect to $H_{\text{DQD}}$ by the equation

$$\dot{\rho} = \mathcal{L}_{\text{rad}}[\rho] + \mathcal{L}_{\text{ph}}[\rho].$$

Here the first term describes the effect of the radiative decoherence in the Markovian limit in terms of the Lindblad dissipator

$$\mathcal{L}_{\text{rad}}[\rho] = \Gamma_{\text{rad}} \left[ \Sigma_-(t) \rho \Sigma_+(t) - \frac{1}{2} \{ \Sigma_+(t) \Sigma_-(t), \rho \} + \right],$$

where $\Sigma_-(t) = \Sigma_+^\dagger(t) = e^{iH_{\text{DQD}}t/\hbar} \sum_\mathbf{k} e^{-iE\mathbf{k}t/\hbar} c_{\mathbf{k}}^\dagger$ and $\Gamma_{\text{rad}} = E^3 |\mathbf{d}|^2 \sqrt{\epsilon_0 \epsilon_r} / (3\pi \epsilon_0 c^3 \hbar^4)$ is the spontaneous decay rate for a single dot. The second term accounts for the interaction with the non-Markovian phonon reservoir. We use the time-convolutionless equation

$$\mathcal{L}_{\text{ph}}[\rho] = - \int_0^t d\tau \text{tr}_{\text{ph}} \left[ H_{\text{c-ph}}(\tau), [H_{\text{c-ph}}(\tau), \rho(\tau) \otimes \rho_{\text{ph}}] \right],$$

where $H_{\text{c-ph}}(t) = e^{iH_{\text{DQD}}t/\hbar} H_{\text{c-ph}} e^{-i(H_{\text{DQD}}+H_{\text{ph}})t/\hbar}$ is the carrier-phonon interaction Hamiltonian in the interaction picture, $\rho_{\text{ph}}$ is the phonon density matrix at the thermal equilibrium, and $\text{tr}_{\text{ph}}$ denotes partial trace with respect to phonon degrees of freedom. While some phonon–assisted processes allow a Markovian limit, there are many effects that can only be reproduced if the reservoir memory is included (such as the initial phonon–assisted dephasing). Moreover, there seems to be no universal way to extract the Markov limit in various physical situations. By using the general Eq. (4), we obtain an all-purpose equation of motion at a modest computational cost.

In the evolution generated by the $H_{\text{DQD}}$, the states $|0\rangle$ and $|3\rangle$ are invariant and nontrivial evolution takes place only in the subspace spanned by $|1\rangle, |2\rangle$. The corresponding evolution
operator can therefore be found easily. Transforming Eq. (2) to the interaction picture and substituting to Eq. (4) we find the phonon-related contribution to the system evolution in the explicit form

$$\mathcal{L}_{\text{ph}}[\rho] = \sum_{i=1,2} [T_i(t)\rho(t)S_i(t) - S_i(t)T_i(t)\rho(t)] + \text{H.c.}$$

Here the nonzero elements of the matrices $S_i$ are

$$\langle 1|S_1|1\rangle = 1 - \langle 2|S_1|2\rangle = 1 - \langle 1|S_2|1\rangle = \langle 2|S_2|2\rangle = \frac{1}{2} \sin^2 2\theta \left( 1 - \cos \frac{2\xi t}{\hbar} \right),$$

$$\langle 1|S_1|2\rangle = \langle 2|S_1|1\rangle^* = -\langle 1|S_2|2\rangle = -\langle 2|S_2|1\rangle^* = \frac{i}{2} \sin 2\theta \sin \frac{2\xi t}{\hbar} - \frac{1}{4} \sin 4\theta \left( 1 - \cos \frac{2\xi t}{\hbar} \right),$$

$$\langle 3|S_1|3\rangle = \langle 3|S_2|3\rangle = 1,$$

and the matrices $T_i(t)$ are defined by

$$T_i(t) = \sum_{j=1,2} \int_0^t d\tau S_j(\tau) R_{ij}(t - \tau).$$

Here

$$R_{11}(t) = R_{22}(t) = \frac{1}{\hbar^2} \sum_k |f_k|^2 \left[ n_k e^{i\omega_k t} + (n_k + 1) e^{-i\omega_k t} \right],$$

$$R_{12}(t) = R_{21}(t) = \frac{1}{\hbar^2} \sum_k |f_k|^2 \cos k_z D \left[ n_k e^{i\omega_k t} + (n_k + 1) e^{-i\omega_k t} \right]$$

are memory functions with $n_k$ denoting the Bose distribution for phonon modes. The above equation of motion for the reduced density matrix strictly reproduces the results in the limiting cases mentioned above. Moreover, for the case of a DQD coupled to phonons with non-vanishing inter-dot coupling $V$, it yields results reasonably close to those obtained by a much more complex correlation expansion technique [5].

In numerical simulations, we take the parameters corresponding to a self-assembled InAs/GaAs system: $\sigma_e - \sigma_h = 9$ eV, $\rho = 5350$ kg/m$^3$, $c_1 = 5150$ m/s, the wave function parameters $l = 4.5$ nm, $l_z = 1$ nm, $D = 6$ nm, and the radiative recombination time (for a single dot) $1/\Gamma_{\text{rad}} = 400$ ps.

4. Example: Impact of phonon-induced pure dephasing on spontaneous emission

As an example of an application of the formalism developed above, let us study a simple example of interplay between phonon-related and radiative phenomena. First, we consider identical, uncoupled dots, that is, $V = 0$, $\Delta = 0$. In such case of two identical emitters, the spontaneous emission has a collective character which leads to a strong modification of the exciton recombination process [3]. In particular, out of the two delocalized single exciton states $|\psi_{\pm}\rangle = (|1\rangle \pm |2\rangle)/\sqrt{2}$, $|\psi_+\rangle$ has a “superradiant” character and decays twice faster than a single-dot excitation, while $|\psi_-\rangle$ is stable against recombination (“subradiant”). In Fig. 1(a), the decay of the state $|\psi_+\rangle$ in the absence of phonon perturbation is shown by the black dotted line.

For a single dot, phonon-induced dephasing (within an independent boson model with a harmonic reservoir) leads only to coherence decay on picosecond time scales and does not affect the radiative recombination on long time scales. This situation changes in quite an interesting manner in the two-dot case. As one can see in Fig. 1(a), in the presence of phonon-induced dephasing.
dephasing, the exciton recombination slows down and a long-lived occupation appears in the system. Moreover, the final exciton occupation grows as the temperature increases. Obviously, this effect must be due to some kind of coherence leading to occupation trapping in a subradiant state which is only possible in such a clear form because the model does not contain dephasing channels that would destroy this coherence (like anharmonicity-induced scattering [16]). Still, such a noise-induced coherence is remarkable.

The reason for this behavior is that during the first few picoseconds of the open system evolution the coherence between the single exciton states decays due to carrier-phonon dynamics according to \( \langle 1 | \rho | 2 \rangle = \frac{1}{2} e^{-\hbar(t)} \), where \( \rho \) is the density matrix of the exciton subsystem and

\[
\hbar(t) = 4 \sum_k \left| \frac{f_k}{\omega_k} \right|^2 \sin^2 \frac{k_z D}{2} (1 - \cos \omega_k t) e^{-\hbar t_{\text{ph}}} = 4 \sum_k \left| \frac{f_k}{\omega_k} \right|^2 \sin^2 \frac{k_z D}{2},
\]

where \( t_{\text{ph}} \) is the characteristic time of the phonon-induced initial dephasing which is of the order of a few picoseconds [9]. Actually, the above formulas are valid in the absence of spontaneous emission. However, on the time scales \( t \ll \Gamma \), when the dephasing takes place, radiative contribution can be approximately neglected. As a result, the initial density matrix \( \rho = |\psi_+ \rangle \langle \psi_+| \) is transformed into

\[
\rho' = \frac{1 + e^{-\hbar \infty}}{2} |\psi_+ \rangle \langle \psi_+| + \frac{1 - e^{-\hbar \infty}}{2} |\psi_- \rangle \langle \psi_-|.
\]

This dephased state is a mixture of a superradiant and a subradiant component. The latter does not decay radiatively and leads to the persistent occupation tail visible in Fig. 1(a).

In the more realistic case of dots that differ in their transition energies and are coupled [Fig. 1(b)], the radiative recombination process still depends on temperature but this dependence is reversed: Now, the emission speeds up as the temperature is increased. In any case, however, the decay in the presence of carrier-phonon coupling is slower than in the absence of phonons.

In order to explain this behavior, we note that although the eigenstates of \( H_{\text{DDQD}} \) in the presence of energy mismatch are not purely sub- and superradiant one of them still decays faster than the other [3]. For \( V > 0 \), the lower eigenstate has a partly subradiant character. Moreover, the coupling between the dots enables excitation transfer between these eigenstates [5]. Coupling to phonons provides a thermalization mechanism for the occupations of the two eigenstates. At \( T = 0 \) this means a transfer to the lower-energy, subradiant state, which strongly suppresses emission. At higher temperatures, however, the contribution from the higher-energy, superradiant state increases and the radiative decay speeds up.

**Figure 1.** Decay of the exciton occupation for the superradiant initial state \( |\psi_+ \rangle \) in the absence of phonon-induced dephasing (black dotted lines) and in the presence of phonon-induced dephasing at various temperatures as shown. (a) identical, uncoupled dots, \( \Delta = V = 0 \); (b) non-identical, coupled dots, \( \Delta = V = 1 \text{ meV} \).
5. Conclusion

Double quantum dots not only show a richer structure of exciton states than a single dot but also admit much more complicated dephasing channels. The approach to numerical simulation of the quantum open system dynamics of a single DQD structure presented in this contribution opens the way to a systematic study of these effects. Although much more accurate (sometimes even exact) methods can be used in the case of two dots interacting with a single reservoir [5; 7; 17] treating the evolution under joint action of both reservoirs at the same level is either impossible or very demanding numerically. The method presented here, based on the TNL equation for the non-Markovian phonon-related effects and the Lindblad equation for spontaneous emission, yields simple equations for the complete biexciton density matrix of the DQD system and allows one to calculate arbitrary exciton-related properties of an undriven system (or a system excited with a single ultrashort pulse).

Acknowledgments

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