Collective fluorescence and decoherence of a few nearly identical quantum dots

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We study the collective interaction of excitons in closely spaced artificial molecules and arrays of nearly identical quantum dots with the electromagnetic modes. We discuss how collective fluorescence builds up in the presence of a small mismatch of the transition energy. We show that a superradiant state of a single exciton in a molecule of two dots with realistic energy mismatch undergoes a two-rate decay. We analyze also the stability of subdecoherent states for non-identical systems.

I. INTRODUCTION

Confinement of carriers in semiconductor quantum dots (QDs) leads to spectrally isolated states which may be optically controlled at a high level of coherence. A single QD offers at most two degrees of freedom (a biexciton) which may be coherently manipulated by optical fields in various ways, allowing one to demonstrate the simplest non-trivial quantum logical operations. In order to overcome this two-qubit limitation one needs to develop manufacturing methods and control schemes for arrays of two and more QDs. Conditional control of such systems, indispensable in both classical and quantum computing schemes, requires interaction between the QDs in the array, which may be provided, e.g., by dipole interaction between confined excitons. Therefore, much experimental effort has been devoted to the investigation of the coupling between QDs and its signatures in the optical response and correlation statistics of quantum dot molecules (QDMs) built of two coupled QDs. It turns out that dephasing of excitons in QDMs differs considerably from that of individual QDs. Even without interaction, optical properties of QDMs and QD arrays may be strongly modified due to collective coupling of sufficiently closely spaced QDs to the electromagnetic (EM) field. These collective effects have been extensively studied for atomic systems, where they manifest themselves by superradiant emission, i.e., an outburst of radiation from the excited sample, markedly different from any exponential decay. On the other hand, the collective interaction leads to the appearance of subradiant states for which the probability amplitudes for photon emission interfere destructively, leading to decoupling from the EM reservoir and to infinite lifetime. It has been proposed to use these states for noiseless encoding of quantum information. For the analogous problem of coupling to phonon modes of a semiconductor, “subdecoherent states” of QD arrays have been suggested as a possible noiseless implementation. Compared to atomic samples, QDs may be easier to arrange in a regular array but the perfectly identical transition energy characteristic of natural atoms is extremely hard to reach for these artificial systems.

The purpose of this paper is twofold. First, we deal with the general, theoretical problem of the stability of collective interaction, including noiseless encoding, against variations of the transition energies. Second, we look for clear manifestations of interaction between the QDs in the experimentally observable optical properties of the QD arrays, depending on the difference (mismatch) of the transition energies of individual QDs. Thus, we study the interaction between small, slightly inhomogeneous arrays of QDs and their EM environment. The system evolution is described within the Weisskopf–Wigner approach. We show how the coherent interaction is destroyed by growing inhomogeneity of the transition energies in the regime where the latter is comparable to decay rate. As important examples, we discuss the buildup of superradiant emission from a few QDs and the decay of “subdecoherent” states built on non-identical QDs. We show that interaction between dots in a regular array may, to some extent, stabilize the coherence, in contrast to randomly distributed atomic systems, where it leads to dephasing. We discuss also how the interplay of the transition energy mismatch and interaction strength determines the time evolution of the luminescence of a more realistic QDM. We show that the decay of luminescence varies from exponential with a single QD rate (for weakly interacting dots), through nonexponential (for interaction compared to energy difference), again to exponential with doubled rate (when interaction energy prevails).

The paper is organized as follows. In Section II we define the model used to describe the system. Section III describes the collective fluorescence and stability of quantum states in a molecule built of two dots. Next, in Section IV we extend the discussion to arrays of four dots. The final discussion and conclusions are contained in Section V.

II. THE SYSTEM

We consider an array of QDs located at points \( r_i \). We assume that each dot may either be empty or contain one ground state exciton of fixed polarization with an interband transition energy \( E_i \), hence can be described as a two-level system. The dots interact with transverse EM modes with frequencies \( \omega_k = \omega \), where \( \omega \) is the wavenumber and \( c \) is the speed of light. We will describe the system in the interaction picture with respect to the
Hamiltonian $H_0 = E \sum_j \hat{n}_j + \sum_{k,\lambda} \omega_k \hat{b}^\dagger_{k,\lambda} \hat{b}_{k,\lambda} (\hbar = 1)$, where $\hat{b}_{k,\lambda}, \hat{b}^\dagger_{k,\lambda}$ are photon creation and annihilation operators ($\lambda$ labels polarizations), $E$ is the average of the energies $E_j$, and $\hat{n}_j$ is the occupation operator for the $j$th dot, $\hat{n}_j = \sigma^{(j)}_++\sigma^{(j)}_-$, where $\sigma^{(j)}_+ = \sigma^{(j)}_x + i\sigma^{(j)}_y$ and $\sigma^{(j)}_y$ are Pauli matrices acting on the $j$th two-level system. The Hamiltonian of the system is then $H = H_X + H_1$. The first component describes the excitons,

$$H_X = \sum_j \Delta_j \hat{n}_j + \sum_{i \neq j} V_{ij} \sigma^{(i)}_+ \sigma^{(j)}_-,$$  

(1)

where $\Delta_j = E_j - E$ are the energy deviations from the average and $V_{ij}$ are Förster couplings between the QDs,

$$V_{ij} = \frac{1}{4\varepsilon_0 \varepsilon_r r_{ij}^3} \left( d^2 - \frac{3|\textbf{d} \cdot \textbf{r}_{ij}|^2}{r_{ij}^3} \right), \quad \textbf{r}_{ij} = \textbf{r}_j - \textbf{r}_i,$$

where $\textbf{d}$ is the interband dipole moment (for simplicity equal for all dots), $\varepsilon_0$ is the vacuum dielectric constant, and $\varepsilon_r$ is the relative dielectric constant of the semiconductor. For self-assembled dots, typical values for this coupling range from $\mu$eV for the distance $r_{ij}$ of order of 100 nm to meV for closely stacked dots separated by $\sim 10$ nm. Another contribution to the coupling may come from the polariton effect (coupling to transverse field).

The second term in the Hamiltonian accounts for the interaction with the EM modes in the dipole approximation and rotating wave approximation (RWA)

$$H_1 = \sum_l \sigma^{(l)}_+ \sum_{k,\lambda} g_{k\lambda} e^{i(\omega_k - E)\tau} \hat{b}^\dagger_{k,\lambda} + \text{H.c.},$$

(2)

with $g_{k\lambda} = id \cdot \hat{\epsilon}_\lambda(k) \sqrt{\omega_k/(2\varepsilon_0 \varepsilon_r v)}$, where $\hat{\epsilon}_\lambda(k)$ are unit polarization vectors and $v$ is the normalization volume for EM modes. The QDs are placed at distances much smaller than the relevant photon wavelength so that the spatial dependence of the EM field may be neglected (the Dicke limit). For wide-gap semiconductors with $E \sim 1$ eV, zero-temperature approximation may be used for any reasonable temperature.

In the present discussion, we disregard the coupling of the carriers with phonons. Let us note that the quantum confinement of excitons leads to a separation of at least a few meV between the ground exciton state involved in our analysis and the lowest excited state in a single dot. Therefore, no real phonon-induced transitions may take place in a single dot as long as the temperature is low enough. It has been shown that the combination of dipole interaction and phonon coupling may lead to phonon-assisted Coulomb transfer between the dots, which might be responsible for the unidirectional transfer observed in the experiment. However, the estimated rate reaches its maximum of $\sim 2$ ns for the energy separation of a few meV and decreases considerably away from this point. Therefore, we neglect this effect in the present considerations. For extremely closely spaced dots, with strongly overlapping carrier wave functions, phonon-assisted tunneling processes might also take place on time scales comparable to those characteristic of the radiative decay. Similar to the phonon-assisted Coulomb transfer, such processes would lead to thermalization of the state of a QDM or QD array which, in general, might suppress the dynamics described in the following sections.

Another phonon effect on the exciton state is pure dephasing. In QDMs, like in individual QDs, such processes affect only the first few picoseconds of the optical response of a QDM, while our present discussion is focused on the radiative decay that develops at much longer times. Due to this separation of time scales, the evolution related to the radiative processes may be discussed separately from this pure dephasing effect. If the system state is prepared by an ultrafast pulse, the initial phonon dynamics may result in a certain reduction of the coherence of the initial state, as we qualitatively discuss in the following sections.

III. QUANTUM DOT MOLECULES (2 QDS)

We will start our discussion with quantum dot molecules composed of two QDs. In the present section, we will first discuss the decay of sub- and super-radiant single exciton states in terms of the formal quantum fidelity with respect to the unperturbed state and in terms of the experimentally measurable exciton occupation. Then, we proceed to the decay of the biexciton state which will be studied again in terms of fidelity and in terms of the measurable photon emission rate.

A. Single-exciton states

The RWA Hamiltonian conserves the number of excitations (excitons plus photons). Let us first consider the initial subradiant state $|\psi(0)\rangle = (|01\rangle - |10\rangle)/\sqrt{2}$, where the two-digit kets denote the occupations of the respective dots. Since there is only one excitation in this state it may, in general, evolve into

$$|\psi(t)\rangle = c_{01}(t)|01\rangle + c_{10}(t)|10\rangle + \sum_{k,\lambda} c_{00k\lambda}(t)|00, k\lambda\rangle,$$

where the last ket denotes the state with no excitons and with one photon in the mode $(k, \lambda)$. The Schrödinger equation leads to the system of equations for the coefficients

$$i\dot{c}_{01} = \Delta c_{01} + V c_{10} + \sum_{k,\lambda} g_{k\lambda} c_{00k\lambda} e^{i(E - \omega_k)t},$$

(3a)

$$i\dot{c}_{10} = -\Delta c_{10} + V c_{01} + \sum_{k,\lambda} g^*_{k\lambda} c_{00k\lambda} e^{i(E - \omega_k)t}$$

(3b)

$$i\dot{c}_{00k\lambda} = g_{k\lambda}(c_{01} + c_{10}) e^{-i(E - \omega_k)t},$$

(3c)
In order to test the stability of the ideally subradiant state of the art of QD manufacturing, the differences between the transition energies of the two dots are rather small. If the QDs are close enough, the Förster interaction becomes effective. Since the sub- and superradiant states are eigenstates of the Förster Hamiltonian separated by an energy \( 2V \), the transition from the initially subradiant state to the superradiant state is suppressed if the magnitude of the Förster coupling exceeds the energy difference \( \Delta \). This is shown in Fig. 1(b). It is clear that the decay rate is reduced when \( V \sim \Delta \) and the subradiance is recovered for \( V \gg \Delta \). Note that, apart from the trivial limiting cases, the decay is markedly non-exponential and its modulation yields information on the origin of the energy level splitting in the system. Indeed, the decay of the superradiant state \( |\psi(0)\rangle = (|10\rangle + |10\rangle)/\sqrt{2} \) shown in the inset to Fig. 2(a) is clearly different for two systems with the same energy splitting, depending on whether the splitting originates from the difference between the dots or from the interaction.

The signatures of collective interaction with the electromagnetic field may also be found in the evolution of measurable quantities. As an example, let us consider the average number of excitons in the QDM. In the present state of the art of QD manufacturing, the differences between the transition energies of the two dots are rather small in the meV than in the \( \mu eV \) range discussed in the previous case. Therefore, let us consider the evolution of exciton occupations for the initial states \( |\psi(0)\rangle = (|10\rangle \pm |10\rangle)/\sqrt{2} \) for a QDM with \( \Delta = 1 \text{ meV} \). The results are shown in Fig. 2. As can be seen, in this case the decay of the occupation shows no oscillations. For \( V \ll \Delta \), both states show simple exponential decay with the rate \( \Gamma \). In the opposite limit, \( V \gg \Delta \), the subradiant state becomes the state of the art of QD manufacturing, the differences between the transition energies of the two dots are rather small in the meV than in the \( \mu eV \) range discussed in the previous case. Therefore, let us consider the evolution of exciton occupations for the initial states \( |\psi(0)\rangle = (|10\rangle \pm |10\rangle)/\sqrt{2} \) for a QDM with \( \Delta = 1 \text{ meV} \). The results are shown in Fig. 2. As can be seen, in this case the decay of the occupation shows no oscillations. For \( V \ll \Delta \), both states show simple exponential decay with the rate \( \Gamma \).
The number of excitons therefore evolves as $n(t) = |c(t)|^2 = \sin^2(\varphi + \pi/4)e^{2\text{Re} \lambda_- t} + \cos^2(\varphi + \pi/4)e^{2\text{Re} \lambda_+ t}$.

Due to the almost perfect orthogonality of the eigenvectors $u_\pm$, the interference term vanishes and the occupation decay is a combination of two exponentials with different rates, as shown in Fig. 2(b). In the inset to this figure we show the values of the two decay constants as a function of $V$ for $\Delta = 1$ meV.

If the initial sub- or superradiant state of a QDM is prepared by an ultrafast optical pulse it will partly lose its coherence within a few picoseconds of the system evolution due to phonon-induced pure dephasing. The details of this dephasing process differs from that of a single QD and depends on the system geometry. Nonetheless, its essential effect is to perturb the superposition state towards a mixture of two states, each of which undergoes the usual exponential decay. Therefore, one may expect a decrease of the amplitude of the oscillations in Figs. 1 and 3 and a shift of the decay curves in Fig. 2 towards the monoexponential decay with the usual decay rate. For special values of the energy mismatch, the results may also be modified by the phonon-assisted Coulomb transfer.
where we denoted $f_{ij} = \rho_{j,i,j}, l, j = 0, 1,$ and $p = \rho_{01,10}$.

The photon emission rate $\gamma = -(2f_{11} + f_{01} + f_{10})$ for the initial state $|11\rangle$ is plotted in Fig. 3. In the case of $V = 0$ [Fig. 3(a)] we see that the photon emission loses its superradiant behavior for growing energy mismatch between the dots, tending to the usual exponential decay for large $\Delta$. Like in the previous case, removing the degeneracy between the sub- and superradiant single-exciton states by including the Förster coupling stabilizes the collective fluorescence [the dotted line in Fig. 3(b) coincides with the $\Delta = 0$ line in Fig. 3(a)].

IV. QUANTUM DOT ARRAYS (4 QDS)

In this section, we study arrays of four QDs in a very special, regular arrangement. The resulting symmetry of the Förster term leads to symmetric eigenstates and, as we show below, to the stabilization of collective effects.

In general, the Weisskopf–Wigner equations lead to the Lindblad equation for the evolution of the reduced density matrix of the charge subsystem

$$\dot{\rho} = -i[H_X, \rho] + \mathcal{L}[\rho], \quad (7)$$

with

$$\mathcal{L}[\rho] = \Gamma \left[ \Sigma_{-}\rho \Sigma_{+} - \frac{1}{2} \{ \Sigma_{+}, \Sigma_{-}, \rho \} + \right],$$

where $\Sigma_{\pm} = \sum_j \sigma_{\pm}^{(j)}$. We now use Eq. (7) to study the evolution of four QDs forming a square array in the $xy$ plane. The energy deviations of individual dots are now $\Delta_i = \alpha_i \Delta$, where $\sum_i \alpha_i = 0$ and $\sum_i \alpha_i^2 = 1$, so that $\Delta$ is the mean square variation of the transition energies. The details of the system evolution depend on the particular choice of $\alpha_i$, but the general behavior is only governed by the interplay of $\Delta$ and $V$ (unless some particularly symmetric choice is made). We arbitrarily fix $\alpha_1 = 0$, $\alpha_2 = -0.8$, $\alpha_3 = 0.27$, $\alpha_4 = 0.54$ and use the mean square variation $\Delta$ as a parameter. The Förster interaction is parameterized by its magnitude $V$, with $V_{12} = V_{23} = V_{34} = V_{41} = V$ and $V_{13} = V_{24} = 2^{-3/2}V$ (the dots are numbered clock-wise).

First, let us choose the subradiant initial states $|\psi_{(0)}\rangle = (|1010\rangle - |0101\rangle + |0110\rangle - |1010\rangle)/2$ and $|\psi_{(1)}\rangle = (|1001\rangle - |0101\rangle + |0110\rangle - |1100\rangle)/2$, which span the subspace of logical qubit states that may be used for noiseless encoding of quantum information on four physical qubits. Obviously, for non-identical dots the phases in these superpositions will rotate and the state will be driven out of the initial noiseless subspace which leads to a decrease of fidelity, as shown in Fig. 4. Out of the two states, only $|\psi_{(1)}\rangle$ is a non-degenerate eigenstate of the Fröhlich interaction for the square array. As a result, as can be seen in Fig. 4 only this state is fully stabilized by the Förster interaction for $V \gg \Delta$ (the lines for $V = 100 \mu eV$ in Fig. 4 are very close to the asymptotic case of $V \to \infty$). Since the other state $|\psi_{(0)}\rangle$ is never completely stable the entire “noiseless subspace” of logical states remains stable only for an extremely homogeneous array of QDs.

Finally, let us study the photon emission rate from a superradiant state of four excited QDs, $|\psi(0)\rangle = |1111\rangle$ (Fig. 5). Now, a clear superradiant peak of photon emission develops for identical dots but vanishes as the dots become different. Again, interaction between the dots in a regular array stabilizes the collective emission. It is interesting to note that the superradiant emission is close to ideal already for $V \sim \Delta$, while the subradiant states are stabilized only when the interaction exceeds the energy difference by an order of magnitude.

It should be stressed that the stabilization effect results from the special, highly symmetric arrangement of the QDs. It should be contrasted with the dephasing induced by analogous interactions in the randomly distributed atomic samples. Likewise, in an irregular...
ensemble of QDs obtained by spontaneous self-assembly no stabilization effect should be expected. However, recent progress in the pre-patterned and strain-engineered growth of QDs\textsuperscript{22,23} shows great promise for the manufacturing of QD arrays with a desired geometry.

V. CONCLUSIONS

We have shown that collective interaction of carries in QDs with their EM environment is extremely sensitive to the homogeneity of the QD array. Already for the ensemble of non-interacting QDs seems highly unlikely. Likewise, implementing collective fluorescence effects in an ensemble of non-interacting QDs appears impractical due to the homogeneity of the QD array. This can stabilize the subradiance of a state of 2 QDs and the superradiant emission from 4 QDs but for a square alignment it cannot assure stability of the entire noiseless subspace implemented on 4 QDs.

When the energy mismatch between the dots is of order of meV, like in the currently fabricated artificial molecules of two QDs, the oscillations disappear and one observes a decay of the excitation (exciton occupation number) composed of two exponentials. For such a realistic energy mismatch, the two decay rates for non-interacting dots are practically equal to the free decay rate $\Gamma$. However, with growing interaction strength they approach $2\Gamma$ (superradiant component) and 0 (subradiant). Thus, their values carry information on the origin of the energy splitting (interaction vs. energy mismatch).

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The destructive effect of inhomogeneity can be, to some extent, overcome by excitation-transfer coupling ( Förster or tunneling) between the dots placed in a regular array. This can stabilize the subradiance of a state of 2 QDs and the superradiant emission from 4 QDs but for a square alignment it still cannot assure stability of the entire noiseless subspace implemented on 4 QDs.

The full minimal-coupling Hamiltonian would yield a cutoff of $g_{\kappa \lambda}$ at $\omega_{\kappa} \sim c/l$, where $l$ is the QD size. For our discussion it is only important that this frequency is extremely high, which justifies the Markov approximation. In this approximation, only the coupling at frequencies close to
Δ is relevant for the radiative damping. Strictly speaking, the absence of the biexciton component requires either a large enough biexcitonic shift or an excitation weak enough to neglect the higher-order biexcitonic occupation.