Dicke states in multiple quantum dots

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We present a theoretical study of the collective optical effects which can occur in groups of three and four quantum dots. We define conditions for stable subradiant (dark) states, rapidly decaying super-radiant states, and spontaneous trapping of excitation. Each quantum dot is treated like a two-level system. The quantum dots are, however, realistic, meaning that they may have different transition energies and dipole moments. The dots interact via a short-range coupling which allows excitation transfer across the dots, but conserves the total population of the system. We calculate the time evolution of single-exciton and biexciton states using the Lindblad equation. In the steady state the individual populations of each dot may have permanent oscillations with frequencies given by the energy separation between the subradiant eigenstates.

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I. INTRODUCTION

Collective optical (super-radiant) effects appear in ensembles in which the distances between single emitters are much smaller than the radiation wavelength with which they interact (Dicke limit) [1]. The spatial dependence of the electromagnetic field within such ensembles is negligible and thus all the systems effectively interact with a common photon reservoir. This leads to formation of rapidly decaying (super-radiant) and optically inactive (subradiant) states [2] and consequently to the appearance of a vacuum-induced coherence effect which results in occupation trapping [3].

Although effects resulting from collective coupling of atoms to a radiative environment have been known for nearly 60 years, are very well described [1–5] and have been extensively investigated experimentally [6,7], they still attract much scientific attention. This is caused by the increasing variety of physical systems in which these phenomena may be observed, such as quantum dots (QDs) [8], Bose-Einstein condensates [9], superconducting qubits [10,11], ionic Coulomb crystals [12], or dipolaritons [13].

The investigation of super-radiant effects have been initiated and driven to a large extent by the promise which the short-living states show for optimization of lasers [14]. Recently the scientific interest has been focused on the concept of a "super-radiant laser" which allows the spectral purity of emitted light to be increased [15,16]. The technological realization of such a device presented in Ref. [17] allows the accuracy of atomic clocks [18,19] and thus measurements of gravity [20] and fundamental constants [21,22] to be increased. The experimental investigations of the collective effects have mostly been restricted to the analysis of the super-radiant states which appear spontaneously in cascade emission and manifest themselves as a maximum in the intensity or photon emission rate [2,6,23].

Although the observation of subradiance phenomena was also reported in atomic ensembles [7] as the opposite of the super-radiant states, the preparation of subradiant states is much more difficult, and therefore the possibilities they give have been much less investigated experimentally. Recently, the preparation of an optically inactive state was reported in a system of superconducting qubits [24] and in a diatomic molecule in an optical lattice [25]. The advantage of the subradiant states stems from their decoupling from the photon environment because of which they do not undergo radiative decoherence and thus may form decoherence-free subspaces [26,27]. This makes them useful for quantum information processing especially for noiseless encoding of quantum information [28,29]. The stable states also allow construction of a scalable quantum processor [30], quantum memories [31], and nonlinear sign-shift gates [32], and storage of time-bin qubits [33] for quantum cryptography. An interesting group of Dicke states is formed of single-excitation combinations belonging to the class of "W" states which have been widely considered for quantum information processing [34–37] or optimization of quantum clock synchronization [38].

Systems composed of two and more coupled QDs have attracted much scientific focus due to the richness of their properties, which pave the way to new technological applications. Already pairs of quantum dots allow for long-time storage of quantum information [39], conditional optical control of carrier states [40], implementation of a two-qubit quantum gate [41], optical writing of information on the spin state of the dopant Mn atom [42], and construction of quantum nanoantennas due to collective phenomena [43]. Systems of three QDs enable realization of two different kinds of entanglement [44], teleportation via super-radiance [45], controlled-NOT (CNOT) gates [46], and the control of spin blockades [47]. Moreover, in these systems the collective transport effects (electronic Dicke or Kondo-Dicke effects) may be realized [48,49] and lead to the enhancement of thermoelectric efficiency [50]. Arrays of QDs allow reduction of the effect of pure dephasing on quantum information encoded in excitonic states [51].

In this paper we analyze the collective optical effects in ensembles of three and four QDs. Compared to double QDs in which only one optically inactive state may be realized [2], ensembles of three and more two-level systems allow the realization of many stable states which occur at different exciton occupations of the single emitters. Although the

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super-radiant effects are well described in systems of identical atoms the description of such phenomena in QD ensembles requires taking into account properties characteristic for those systems which distinguish them from natural ones. Therefore we include in our model the fundamental energy mismatches, different dipole moments of single dots, and coupling which induces excitation transfer between single emitters, but conserves the total population of the ensemble. In our previous works concerning double QDs it has been shown that the collective optical effects are extremely sensitive to inhomogeneity of the fundamental transition energy which leads to the decay of the exciton occupation for energy splitting much below the present technological feasibility. This decoherence effect may be strongly reduced by sufficiently strong coupling between the dots [52–54] and fully overcome in double QDs with different decay rates [55]. In this paper we extend the results of two QDs and specify conditions which allow us to take advantage of the super-radiant phenomena in fully inhomogeneous QD systems. We analyze the dynamics of single electron-hole pairs and biexcitons. We show how to adjust the system parameters in such a way that an arbitrary dark single-exciton combination may be blocked in a multiple QD; we also specify the conditions which allow trapping of two excitons and preparation of a system in a biexcitonic state which allows recombination of only one electron-hole pair. Due to the existence of two or more subradiant single-exciton eigenstates and coupling between the dots the occupation of individual dots oscillates while the population of the whole ensemble remains stable; we show that the oscillation amplitudes may be strongly reduced if the system is initially prepared in a biexciton state.

The paper is organized as follows. In Sec. II we describe the system under study, define its model, and describe a method used to study the system evolution, in Sec. III we present and discuss our results, and we conclude the paper in Sec. IV.

II. THE SYSTEM

The investigated system consists of N ($N = 3,4$) quantum dots in which only the ground-level exciton states with fixed spin polarizations are taken into consideration. Due to the strong Coulomb coupling and absence of the external electric fields we may restrict the discussion to "spatially direct" excitonic states, i.e., states with electron-hole pairs residing in the same QD which in these conditions have much lower energy than the "dissociated" states referring to excitons formed of carriers residing in two different dots [56,57]. These assumptions allow us to treat every QD as a two-level system which may either be empty or contain an exciton and thus describe the set of N QDs as a 2^N-level system, with $|0\rangle$ denoting the ground (or "vacuum") state in which all N dots are empty, single-exciton states $|i\rangle$ corresponding to one exciton localized in the ith QD, biexcitonic states $|ij\rangle$ referring to electron-hole pairs residing in the ith and jth QDs, states $|ijk\rangle$ with three QDs, ith, jth, and kth ($1 \leqslant i,j,k \leqslant N$), occupied by an exciton etc.

Present manufacturing technology does not allow on demand production of QD systems with identical fundamental transition energies; therefore we assume different electron-hole binding energies of each dot defined as

$$E_i = E + \Delta_i,$$

where E is the average transition energy and $\Delta_i = \alpha_i \Delta$ is the energy mismatch of the ith QD. We impose $\sum_i \alpha_i = 0$ and $\sum_i \alpha_i^2 = 1$, such that Δ^2 is the mean square variation of the transition energies.

We analyze the system in a "rotating frame" defined by the evolution operator

$$U = \exp\left[-\frac{i}{\hbar}\left(E\sum_i^N \sigma_+^{(i)}\sigma_-^{(i)} + H_{\mathrm{rad}}\right)t\right],$$

where $\sigma_-^{(i)} = (\sigma_+^{(i)})^\dagger = |0\rangle\langle i| + \sum_j |j\rangle\langle ij| + \sum_{jk}|jk\rangle\langle ijk| + \cdots$ are the annihilation (creation) operators for the exciton in the ith QD, respectively, and $H_{\mathrm{rad}} = \sum_{\boldsymbol{k}\lambda}\hbar\omega_{\boldsymbol{k}} b_{\boldsymbol{k}\lambda}^\dagger b_{\boldsymbol{k}\lambda}$ is the standard free photon Hamiltonian with operators $b_{\boldsymbol{k}\lambda}^\dagger$ and $b_{\boldsymbol{k}\lambda}$ creating and annihilating radiation modes with wave vector $\boldsymbol{k}$ and polarization λ, while $\omega_{\boldsymbol{k}}$ is the corresponding frequency.

In this frame the Hamiltonian of the system is

$$H = H_{\mathrm{S}} + H_{\mathrm{S\text{-}rad}}.$$

The first term describes electron-hole pairs residing in the QD system. We assume that the ground state $|0\rangle$ corresponds to the zero energy level, so the excitonic Hamiltonian is

$$H_{\mathrm{S}} = \sum_{i=1} \Delta_i \sigma_+^{(i)}\sigma_-^{(i)} + \sum_{i,j=1} B_{ij}|ij\rangle\langle ij| + \sum_{i,j,k=1} B_{ijk}|ijk\rangle\langle ijk| + \cdots + \sum_{i,j=1} V_{ij}\sigma_+^{(i)}\sigma_-^{(j)}, \quad (1)$$

where B_{ij} are biexcitonic shifts due to the interaction of static dipole moments of the ith and jth QDs, B_{ijk} is the deviation of energy caused by interaction of the dipole moments of three dots, etc, and V_{ij} describes coupling between the dots.

In ensembles of QDs one may distinguish two types of interaction between the emitters: dipole (Förster) coupling which in the leading order decays as $1/r_{ij}^3$ with the QD separation (r_{ij}) [58–60] and short-range coupling resulting from a combination of tunneling (wave function overlap) and Coulomb correlations. Both types of interaction induce excitation transfer between the emitters but conserve the total exciton occupation of the ensemble. The short-range couplings allow enhanced emission in the energetically inhomogeneous ensembles to be rebuilt while in the case of dipole interaction a similar effect is achieved if the coupling is enhanced artificially by a factor of 400 [61]. To overcome the destructive effect of the transition energy mismatch the coupling between the dots must be of the order of the energy splitting, i.e., 1 meV for technologically feasible systems [52–54]. For a planar QD arrangement the distance between emitters is about 30 nm (the average value for the sample studied in Ref. [8]), and for such a distance the Förster coupling drops to about 1 μeV, which is not sufficient to stabilize collective effects.

The Förster coupling reaches a fraction of meV for QDs separated by only a few nanometers [62] which can be achieved in vertically stacked QD systems, and where indeed it can stabilize the collective effects [52,55]. In the present paper we describe a planar system and therefore we prefer to consider a short-range coupling, for which we choose an exponential model, $V_{ij} = V_0 \exp[-r_{ij}/r_0]$, where the subscripts i and j refer to the ith and jth QDs, respectively, V_0 is a constant amplitude, r_{ij} is the distance between the QDs, and r_0 is the spatial range of the interaction. Remarkably, this simple model reproduced well recent experimental results [61].

The eigenstates of the Hamiltonian (1) do not mix quantities associated with different exciton numbers, i.e. the eigenstates of the system are superpositions of the basis states restricted to a particular exciton number.

The second term of the Hamiltonian accounts for coupling between the QD system and quantum electromagnetic field

$$H_{\text{S-rad}} = \sum_{j=1}^{N} \sum_{\boldsymbol{k}\lambda} \sigma_{-}^{(j)} g_{\boldsymbol{k}\lambda}^{(j)} e^{-i(E/\hbar-\omega_k)t} b_{\boldsymbol{k}\lambda}^{\dagger} + \text{H.c.}, \tag{2}$$

where

$$g_{\boldsymbol{k}\lambda}^{(j)} = i\boldsymbol{d}_j \cdot \hat{e}_\lambda(\boldsymbol{k}) \sqrt{\frac{\hbar\omega_k}{2\epsilon_0\epsilon_r v}}$$

is a coupling constant for the jth QD. Here $\boldsymbol{d}^{(j)}$ is the interband dipole moment for the jth QD, $\hat{e}_\lambda(\boldsymbol{k})$ is the unit polarization vector of the photon mode with polarization λ, ϵ_0 is the vacuum dielectric constant, ϵ_r is the relative dielectric constant, and v is the normalization volume. We restrict our investigations to wide-gap semiconductors with electron-hole binding energies of the order of 1 eV which allows us to describe the photon modes within the zero-temperature approximation at any reasonable temperature.

To describe the evolution of the carrier subsystem we use an equation of motion for the reduced density operator in the Markov approximation. In the "rotating frame" it takes the form

$$\dot{\rho} = -\frac{i}{\hbar}[H_{\text{S}},\rho] + \mathcal{L}_{\text{rad}}[\rho],$$

where ρ is a reduced density matrix of the exciton subsystem and $\mathcal{L}_{\text{rad}}$ is a Lindblad dissipator,

$$\mathcal{L}_{\text{rad}}[\rho] = \sum_{i,j=1}^{N} \Gamma_{ij} \left[\sigma_{-}^{(i)} \rho \sigma_{+}^{(j)} - \frac{1}{2}\{\sigma_{+}^{(j)}\sigma_{-}^{(i)},\rho\} \right],$$

where

$$\Gamma_{ij} = \Gamma_{ij}^{*} = \frac{E^3}{3\pi\epsilon_0\epsilon_r\hbar^4c^3} \boldsymbol{d}_i \cdot \boldsymbol{d}_j^{*} \tag{3}$$

with c being the speed of light. Since for $i = j$ Eq. (3) describes the spontaneous decay rates of single QDs [63], the mixed (off-diagonal) decay rates (3) may be expressed in terms of single-QD quantities,

$$\Gamma_{ij} = \Gamma_{ji}^{*} = \sqrt{\Gamma_{ii}\Gamma_{jj}}\,\hat{\boldsymbol{d}}_i \cdot \hat{\boldsymbol{d}}_j^{*},$$

where $\hat{\boldsymbol{d}}_i = \boldsymbol{d}_i/d_i$ and $\hat{\boldsymbol{d}}_i \cdot \hat{\boldsymbol{d}}_j^{*} \approx e^{i\eta}(1-\theta_{ij}^2/2)$; here η is an irrelevant phase and θ_{ij} is a small angle between the dipole moments which depends on the light-hole admixture [55].

In the numerical simulations we assume constant energy mismatches with the parameters $\alpha_1 = 2/\sqrt{56}$, $\alpha_2 = 4/\sqrt{56}$, $\alpha_3 = -6/\sqrt{56}$, $\alpha_4 = 0$, and $\Delta = 1$ meV (except for Fig. 4). For the coupling amplitudes we take $V_0 = 5$ meV and $r_0 = 15$ nm. In Secs. III A, III B, and III C we assume parallel dipole moments, since the effect of the light-hole admixture in the absence of external electric fields is negligible [55].

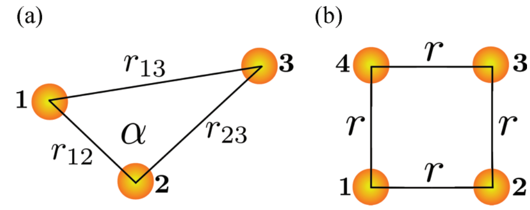

FIG. 1. (Color online) The planar arrangement of the three- (a) and four- (b) QD ensembles.

III. RESULTS

Below we present an analysis of the collective effects which occur in multiple QDs. We define the Dicke states for an arbitrary number of emitters and perform numerical simulations for ensembles of three and four QDs. In Fig. 1 we illustrate the numbering of the QDs and their spatial arrangement. In Secs. III A and III B we focus on single-exciton states. In Sec. III A we show the evolution of uncoupled systems with identical fundamental transitions and parallel dipole moments of different amplitudes. Then, in Sec. III B we analyze the same effects in a system composed of three coupled energetically inhomogeneous dots. The dynamics of biexciton states is presented in Sec. III C. In Sec. III D we show possibilities of controlling the exciton occupation given by a *p-i-n* junction.

A. Single-exciton states of ideal quantum dots

The collective effects were described first in ensembles of uncoupled identical atoms where all the emitters have the same transition energies and dipole moments [1]. Such optical effects, resulting from the interaction in the Dicke limit, are also present in uncoupled systems, even with different dipole moments, if all the emitters have identical transition energies. For the purpose of this paper we define such systems as ideal QDs.

The coupling of excitons to their radiative environment described in the Dicke limit by the Hamiltonian (2) and Fermi's golden rule, according to which the probability of releasing a photon through a transition from the initial to the final states is $P \sim |\langle\text{final}|H_{\text{S-rad}}|\text{initial}\rangle|^2$, allow definition of rapidly decaying (super-radiant) and optically inactive (subradiant) states, both also known as Dicke states. By definition the super-radiant states ($|\text{SUPER}\rangle$) correspond to the maximum transition probability, whereas the subradiant states ($|\text{SUB}\rangle$) refer to a vanishing probability. Due to the decoupling from the photon reservoir these are dark, optically inactive states. In the weak-excitation limit, i.e., for a single excitation in the system from which the sample may decay only to the ground state ($|0\rangle$), the proportionality $g_{\boldsymbol{k}\lambda}^{(j)} \sim \sqrt{\Gamma_{jj}}$ allows the short-living states to be written in the form

$$|\mathrm{SUPER}\rangle = \frac{\sum_{i=1}^{N} \sqrt{\Gamma_{ii}}|i\rangle}{\sqrt{\sum_{i=1}^{N} \Gamma_{ii}}}, \quad (4)$$

and the stable superpositions to be expressed as

$$|\mathrm{SUB}\rangle = \frac{\sum_{i=1}^{N} a_i \sqrt{\prod_{j=1}^{N} \frac{\Gamma_{jj}}{\Gamma_{ii}}}|i\rangle}{\sqrt{\sum_{i=1}^{N} |a_i|^2 \prod_{j=1}^{N} \frac{\Gamma_{jj}}{\Gamma_{ii}}}}, \quad (5)$$

where the coefficients a_i satisfy $\sum_i a_i = 0$. Irrespective of the number of emitters there is only one super-radiant state in a system of a particular number of QDs, whereas the only structure which realizes just one dark state is a double quantum dot. The systems of three and more QDs allow realization of an arbitrary number of dark states of the form (5) since there are many combinations of the parameters a_i for which the transition matrix element $\langle 0|H_{\text{S-rad}}|\mathrm{SUB}\rangle = 0$.

The consequence of the coexistence of rapidly decaying and stable states is the effect of spontaneous trapping of excitation [3,55]. An arbitrary single-exciton state $|s\rangle = \sum_i c_i|i\rangle$, where $|c_i|^2$ is the localization probability of the exciton on the ith QD and $\sum_i |c_i|^2 = 1$, may be expressed as a combination of the super-radiant state (4) and a dark state of type (5),

$$|s\rangle = \frac{\left(\sum_{i=1}^{N} c_i\sqrt{\Gamma_{ii}}\right)}{\sqrt{\sum_{i=1}^{N} \Gamma_{ii}}}|\mathrm{SUPER}\rangle + \sqrt{1 - \frac{\left(\sum_{i=1}^{N} c_i\sqrt{\Gamma_{ii}}\right)^2}{\sum_{i=1}^{N} \Gamma_{ii}}}|\mathrm{SUB}\rangle_s, \quad (6)$$

where the dark state is

$$|\mathrm{SUB}\rangle_s = \frac{\sum_{i=1}^{N} c_i\left[\left(\sum_{j\neq i}^{N} \Gamma_{jj}\right)|i\rangle - \sum_{j\neq i}^{N} \sqrt{\Gamma_{ii}\Gamma_{jj}}|j\rangle\right]}{\sqrt{\left(\sum_{i=1}^{N} \Gamma_{ii}\right)\left[\sum_{i=1}^{N} \Gamma_{ii} - \left(\sum_{i=1}^{N} c_i\sqrt{\Gamma_{ii}}\right)^2\right]}}. \quad (7)$$

The derivation of Eqs. (6) and (7) is done in the the Appendix. The collective coupling to the radiative surrounding induces emission only from the super-radiant state and thus the fraction of excitation initially spanned in the dark state,

$$1 - \left(\sum_{i=1}^{N} c_i\sqrt{\Gamma_{ii}} \Big/ \sqrt{\sum_{i=1}^{N} \Gamma_{ii}}\right)^2,$$

remains unaffected. Since the only single-exciton state which decays totally is the super-radiant state, we define a state $|s\rangle$ as being bright if it has a super-radiant component, i.e., if a system prepared in that state partially recombines and only a part of the initial exciton occupation remains trapped.

In Figs. 2(a) and 2(b) we show the dynamics of a single excitation induced by a common photon reservoir in an ideal system of three uncoupled ($V_{ij} = 0$) QDs with equal electron-hole binding energies ($\Delta = 0$) and parallel dipole moments ($\theta_{ij} = 0$) of different magnitudes. In Fig. 2(a) we show the evolution of the exciton occupations of a system prepared initially in a bright state $(5|1\rangle + |2\rangle)/\sqrt{26}$ and in Fig. 2(b) we show the corresponding coherences. As expected, the coupling to the photon reservoir spans the excitation into the sub- and super-radiant states according to Eq. (6) and induces emission only from the short-living state

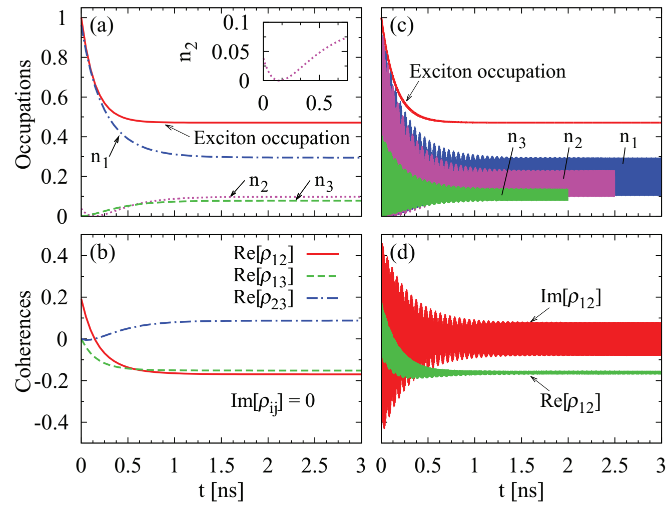

FIG. 2. (Color online) Exciton occupations [(a) and (c)] and coherences [(b) and (d)] of a system of three QDs prepared initially in a bright state $(5|1\rangle + |2\rangle)/\sqrt{26}$ for $\Gamma_{11} = 2.44\ \mathrm{ns}^{-1}$, $\Gamma_{22} = 3.31\ \mathrm{ns}^{-1}$, and $\Gamma_{33} = 1\ \mathrm{ns}^{-1}$. (a) and (b) show the results for uncoupled QDs ($V_{ij} = 0$) with identical transition energies ($\Delta = 0$), while (c) and (d) refer to energetically inhomogeneous ($\Delta \neq 0$) and coupled ($V_{ij} \neq 0$) systems arranged in a lateral array of an equidistant triangle shape with the side length $r = 30$ nm. The inset to (a) shows the initial evolution of the exciton occupation of the QD no. 2.

$$\frac{\sqrt{\Gamma_{11}}|1\rangle + \sqrt{\Gamma_{22}}|2\rangle + \sqrt{\Gamma_{33}}|3\rangle}{\sqrt{\Gamma_{11} + \Gamma_{22} + \Gamma_{33}}}$$

[Fig. 2(a)]. The excitation dynamics occurs until occupations of all dots stabilize at certain levels corresponding to the dark state defined in Eq. (7) which confirms that the state (7) is indeed unaffected by the photon reservoir and, after the decay of the super-radiant state, neither the total exciton occupation nor occupations of single dots ($n_{1,2,3}$) change due to radiative environment. The emission from the above state induces decay of the total exciton occupation and excitation transfer, which results in the redistribution of the occupations of single dots. Since all of the localized single-exciton states $|i\rangle$ contribute to the super-radiant state (4), the collective coupling spans the initial excitation in all of the dots even if some of them were initially empty. Therefore the population of initially unoccupied systems builds up spontaneously [n_3, green dashed line in Fig. 2(a)]. If the initial occupation of one of the dots is relatively small while the spontaneous decay rate from that system is sufficiently strong, then the exciton occupation of that dot may vanish at some point and then be restored due to the excitation transfer [magenta dotted line in Fig. 2(a) and the inset to Fig. 2(a)]. During the emission process also evolution of the off-diagonal density matrix elements is observed, the coherences related to the initially populated dots decay, while those corresponding to initially empty systems build up spontaneously due to the increasing occupations of those dots. When exciton dynamics in the system reaches population distribution corresponding to the optically inactive state also the off-diagonal density matrix elements stabilize at a certain nonzero level [Fig. 2(b)], defined by the dark contribution to the initial state.

B. Single-exciton states for inhomogeneous quantum dots

Technologically feasible QDs forming multiple structures differ in both fundamental transition energies ($\Delta \neq 0$) and dipole moments ($\boldsymbol{d}_i \neq \boldsymbol{d}_j$), and are coupled with each other ($V_{ij} \neq 0$). As shown in the previous section and, for a double QD in Ref. [55], the super-radiant character of the evolution of one exciton is present in ideal systems ($\Delta = 0$), but with parallel dipole moments ($\theta_{ij} = 0$). The collective evolution is very sensitive to the energy mismatches and is destroyed in ensembles with energy splittings of the order of the transition linewidth [52–55]. In such systems the localized eigenstates corresponding to different energies cannot form delocalized superpositions which would also be the system eigenstates. This destructive effect may be overcome by coupling between the dots (V_{ij}) which delocalizes the system eigenstates and different dipole moments allowing the super-radiant state to be a nonsymmetric superposition of the localized states $|i\rangle$ [Eq. (4)].

The single-exciton eigenstates of the system depend on the energy mismatches and coupling between the dots, while the Dicke states are defined by the interplay of decay rates [Eqs. (4) and (5)]. If the individual QD decay rates [Eq. (3) for $i = j$] are adjusted in such a way that the super-radiant state (4) corresponds to one of the system eigenstates, then the inhomogeneous ensemble of QDs interacts with its radiative environment in the "collective regime", i.e., allows many effects typically present only in systems with identical electron-hole binding energies. The amplitudes c_i of a single-exciton state orthogonal to the super-radiant state (4) must satisfy the equation $\sum_i c_i\sqrt{\Gamma_{ii}} = 0$ which implies the condition $\langle 0|H_{\text{S-rad}}|\text{SUB}\rangle = 0$ defining a subradiant state. Therefore, if one of the eigenstates has a super-radiant character then the other eigenstates of a system are optically inactive and thus defying the collective regime requires only specifying the super-radiant eigenstate.

In Figs. 2(c) and 2(d) we show the evolution of a realistic group of three QDs placed in the corners of an equilateral triangle, we assume nonequal fundamental transition energies ($\Delta = 1$ meV), nonvanishing coupling between the systems ($V_{ij} \neq 0$), and parallel dipole moments ($\theta_{ij} = 0$). We compare the results obtained for an inhomogeneous system coupled to the photon reservoir in the collective regime to the ideal case presented in Figs. 2(a) and 2(b), where the decay rates of individual dots take the same values as in Figs. 2(c) and 2(d). As can be seen in Fig. 2(c), coupled ensembles with energy mismatches of the order of meV allow trapping of the same fraction of excitation as ideal dots with the same decay rates [red solid lines in Figs. 2(a) and 2(c)]. Here, as in the ideal case, any single-exciton eigenstate may be decomposed into sub- and super-radiant components according to Eq. (6) and also in this case the super-radiant state is the only state which decays totally. For an arbitrary set of decay rates, which do not correspond to the super-radiant eigenstate, the exciton occupation of a system prepared initially in a state of the form (5) is quenched and the decay of the state (4) is slowed down compared to the collective regime.

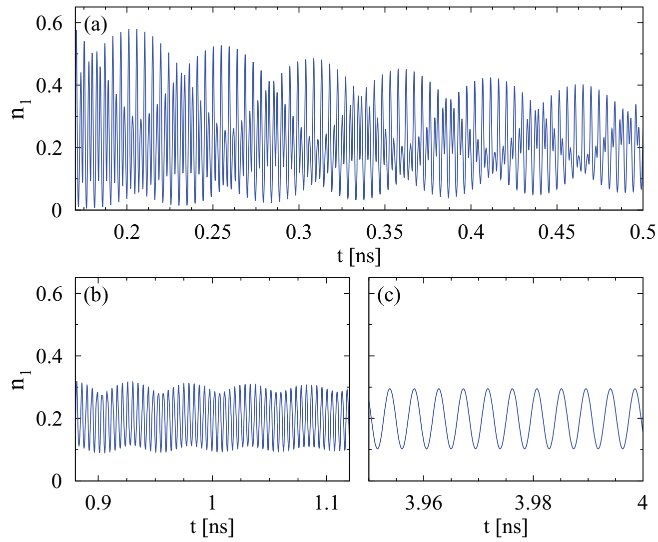

FIG. 3. (Color online) The exciton occupation of the QD no. 1 (n_1) shown by the blue line in Fig. 2(c). The time evolution is shown for three different time intervals at picosecond resolution scale.

Although sub- and super-radiant states may exist in appropriately designed realistic systems, the dynamics of individual QD occupations differs considerably from the ideal case discussed in the previous section, when the dots interact only through the common radiative reservoir. The situation changes when the dots communicate with each other via short-range coupling which induces excitation transfer between dots and thus oscillations in the evolution of single-QD populations. In a double-QD system the oscillation amplitudes decrease and the occupations stabilize at levels corresponding to the single dark state of the system [55]. In multiple QDs composed of three or more emitters oscillations of the individual dot populations never vanish [Figs. 2(c) and 3]. The excitation is trapped in the system because of the existence of dark states, which in such systems may be realized by many different amplitude combinations and thus also the final population number may be realized in various ways.

In Fig. 3 we enlarge the exciton occupation n_1 shown in Fig. 2(c) (blue line). As can be seen in Fig. 3(a) the initial oscillation pattern is relatively complicated; this is caused by the existence of three energy gaps defined by the differences between the super-radiant eigenstate and the subradiant ones, $|E_{\text{SUPER}} - E_{\text{SUB}_{1(2)}}|$, and by the energy splitting between the two subradiant eigenstates, $|E_{\text{SUB}_1} - E_{\text{SUB}_2}|$. The period of the envelope oscillations is a multiplication of the corresponding three periods, i.e., $T_{1(2)} = h/|E_{\text{SUPER}} - E_{\text{SUB}_{1(2)}}|$) and $T = h/|E_{\text{SUB}_1} - E_{\text{SUB}_1}|$. The period T itself defines the fine oscillations of the occupation. Due to the emission process, the super-radiant contribution attenuates and the interference pattern simplifies [Fig. 3(b)]. Finally, when the short-living state decays and the total exciton occupation becomes trapped, the evolution of the single-dot occupation shows a single-mode pattern which repeats with the time T and with amplitude depending on the initial occupation of individual dots [Fig. 3(c)].

A system of three and more QDs allows many different planar arrangements of the emitters. Since the coupling

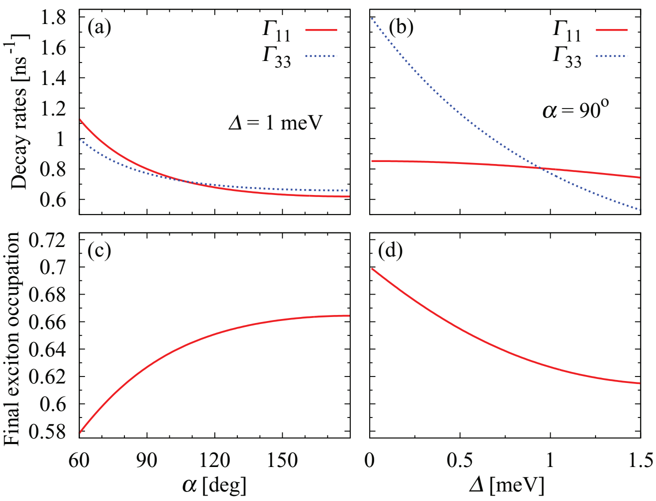

FIG. 4. (Color online) Decay rate (in the collective regime) dependence on the spatial arrangement of a triple QD, i.e., on the angle α defined in Fig. 1 (a) and on the energy mismatch (b), and the corresponding steady-state occupations for the initial state $(5|1\rangle + |2\rangle)/\sqrt{26}$ (c) and (d), respectively. For all panels we assume $r_{12} = 30$ nm and $r_{23} = 20$ nm and a constant decay rate of the QD No. 2 ($\Gamma_{22} = 2$ ns^{-1}).

amplitudes (V_{ij}) depend on the distances between emitters, the eigenstates of the system, and thus the decay rates for which the ensemble interacts collectively with its radiative environment, also depend on the arrangement of the dots. Using the geometry design defined in Fig. 1(a) we calculate the dependence of the decay rates Γ_{11} and Γ_{33} on the spatial arrangement of the system. We assume constant distances r_{12} and r_{23} and thus constant values of the coupling amplitudes V_{12} and V_{23} and change the angle α from 60° to a linear design, i.e., we increase the distance between dots 1 and 3. As seen in Fig. 4(a) the values of the decay rates necessary to form the collective regime slightly decrease with increasing angle α. The two similarly decreasing decay rates (while the third one is constant), according to Eq. (6) lead to increasing steady-state (final) exciton occupation which is shown in Fig. 4(c). If the coupling between two out of three QDs is much stronger than the coupling of those dots with the third one (e.g., $V_{13} \gg V_{12}, V_{23}, r_{13} \ll r_{12}, r_{23}$) then one of the system eigenstates has a large contribution from the localized state associated with the weakly coupled dot (no. 2) and to achieve collective regime the decay rate Γ_{22} must be much smaller than Γ_{11} and Γ_{33}. In the limiting case of vanishing couplings V_{12} and V_{23} the localized state $|2\rangle$ becomes the system eigenstate and the corresponding decay rate (Γ_{22}) vanishes. Consequently the pair of coupled dots acts as a double QD while the third dot does not contribute to the evolution.

In Fig. 4(b) we show the dependence of the decay rates forming the collective regime on the energy mismatch. Both calculated decay rates decrease with increasing energy separation, but one changes slowly while the second decays fast; this leads to decreasing [Eq. (6)] steady-state occupation shown in Fig. 4(d). For an appropriately arranged ensemble of dots the decay rates may decrease in such a way that for smaller energy mismatches the final exciton occupation decreases with increasing energy mismatch, but after exceeding a critical point it increases. Similar effects in the dynamics of single excitons are observed for electron-hole pairs confined in ensembles of four and more dots.

C. Biexciton states

In multiple QDs built out of three and four units more than one exciton may be delocalized and thus these systems allow for more complex collective effects. Below we focus on biexciton states which in general are described by a vector $|\text{biexciton}\rangle = \sum_{i,j,i\neq j}^{N} b_{ij}|ij\rangle$, where $\sum_{i,j,i\neq j}^{N} |b_{ij}|^2 = 1$. Realistic ensembles of QDs ($\Delta \neq 0$, $\boldsymbol{d}_i \neq \boldsymbol{d}_j$, $V_{ij} \neq 0$, and $B_{ij} \neq 0$) permit super-radiance phenomena in the two-exciton subspace if the biexcitonic, as well as single-exciton, eigenstates correspond to the Dicke states. As in the single-exciton case, the biexcitonic state is considered to be super-radiant if the exciton occupation of a system prepared in this state decays totally. Although eigenstates of the Hamiltonian (1) do not mix localized basis states associated with different exciton numbers, the biexcitonic super-radiant state may be formed provided one of the single-exciton eigenstates has a super-radiant character. Thus the biexcitonic super-radiant states occur only if collective effects are present in the single-exciton subspace. Due to the equal number of single-exciton eigenstates and QDs forming the ensemble, the collective regime in the single-exciton domain may be achieved by adjusting only the decay rates (dipole moments). As will be explained in detail below, the same rates Γ_{ij} (3) define the biexcitonic Dicke states. Thus, in order to achieve collective effects in the two-exciton subspace one has to also appropriately adjust the spatial arrangement of the dots or energies.

The super-radiant two-exciton superpositions may be spanned as well in ensembles of four emitters as in triple QDs and take the form

$$|\text{SUPER}\rangle_{\text{B}} = \frac{\sum_{i,j,i\neq j}^{N} \sqrt{\Gamma_{ii}\Gamma_{jj}}|ij\rangle}{\sqrt{\sum_{i,j,i\neq j}^{N} \Gamma_{ii}\Gamma_{jj}}}. \qquad (8)$$

Similarly to the single-exciton case, the biexcitonic super-radiant states are defined by the maximum value of the transition probability ($\sim |\langle\text{SUPER}|H_{\text{S-rad}}|\text{biexciton}\rangle|^2$), but this time from the initial biexciton state to the final single-exciton super-radiant one (4). The form of the condition is governed by the coupling to the radiative environment [Hamiltonian (2)] which induces decay of only one exciton at a time. Thus total quenching of two excitons must occur through formation of single-exciton super-radiant states. As can be seen in Fig. 5(a), due to the decay of the biexciton super-radiant state a part of the initial excitation is initially transferred to the single-exciton state, which reaches a maximum population and then is totally quenched, together with the biexciton state. The condition for the existence of the super-radiant state (8) implies that the transition to subradiant states (5) vanishes.

Two excitons can be blocked in a system if a transition from the biexciton state to any single-exciton state is forbidden, i.e., the transition matrix element $\langle\text{single}|H_{\text{S-rad}}|\text{biexciton}\rangle$ vanishes. Due to the infinite number of possible single-exciton states this condition reduces to the requirement $H_{\text{S-rad}}|\text{biexciton}\rangle = 0$. To simplify the

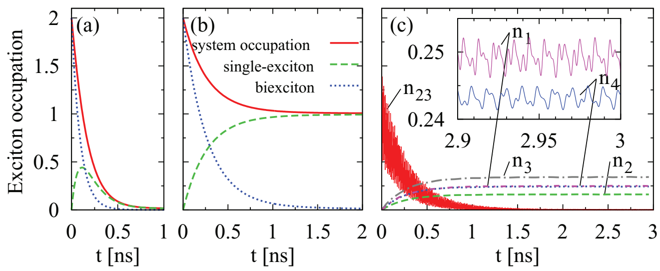

FIG. 5. (Color online) Exciton occupation for a biexciton initial state spanned in a system of four QDs placed at the corners of a square of side length $r = 30$ nm. (a) Super-radiant initial state. (b) and (c) Initial biexciton state which allows for a recombination of only one exciton. $d_{12} = \sqrt{\Gamma_{33}\Gamma_{44}}$, $d_{13} = -\sqrt{\Gamma_{22}\Gamma_{44}}$, $d_{14} = \Gamma_{44}(\Gamma_{33} - \Gamma_{22})\sqrt{\Gamma_{22}\Gamma_{33}}/(\Gamma_{22}\Gamma_{33} - \Gamma_{11}\Gamma_{44})$, $d_{23} = -\Gamma_{44}(\Gamma_{33} - \Gamma_{22})\sqrt{\Gamma_{11}\Gamma_{44}}/(\Gamma_{22}\Gamma_{33} - \Gamma_{11}\Gamma_{44})$, $d_{24} = \Gamma_{44}\sqrt{\Gamma_{11}\Gamma_{33}}/\Gamma_{11}$, and $d_{34} = -\Gamma_{44}\sqrt{\Gamma_{11}\Gamma_{22}}/\Gamma_{11}$. The values of the decay rates of individual dots are the same for all panels and take the values $\Gamma_{11} = 2.26$ ns^{-1}, $\Gamma_{22} = 2.75$ ns^{-1}, $\Gamma_{33} = 0.88$ ns^{-1}, and $\Gamma_{44} = 1.5$ ns^{-1}. The biexciton shifts are $B_{13} = B_{12} - 0.702$ meV, $B_{14} = B_{12} + 0.022$ meV, $B_{23} = B_{12} + 0.218$ meV, $B_{24} = B_{12} - 0.741$ meV, and $B_{34} = B_{12} + 0.042$ meV with B_{12} being an arbitrary parameter. The line types used in (b) are also valid for (a).

description we define non-normalized amplitudes d_{ij} in such a way that the amplitudes of the biexciton states $b_{ij} = d_{ij}/\sqrt{\sum_{i,j,i\neq j}|d_{ij}|^2}$. For a triple QD the condition leads to a system of three equations of the form

$$d_{ij}\sqrt{\Gamma_{jj}} + d_{ik}\sqrt{\Gamma_{kk}} = 0,$$

where every subscript i, j, and k takes the values 1, 2, and 3, respectively. The system is satisfied only in the case of vanishing amplitudes $d_{12} = d_{13} = d_{23} = 0$ which means that it is impossible to block two excitons in a triple QD. The coefficients of a stable biexcitonic state spanned in a system of four QDs must satisfy the system of four equations of the form

$$d_{ij}\sqrt{\Gamma_{jj}} + d_{ik}\sqrt{\Gamma_{kk}} + d_{il}\sqrt{\Gamma_{ll}} = 0,$$

where every subscript i, j, k, and l takes the values 1, 2, 3, and 4, respectively. The above equations lead to the condition for the d_{ij} numbers in the form

$$d_{12} = -\frac{\sqrt{\Gamma_{33}}d_{13} + \sqrt{\Gamma_{44}}d_{14}}{\sqrt{\Gamma_{22}}},$$
$$d_{23} = \sqrt{\frac{\Gamma_{11}\Gamma_{44}}{\Gamma_{22}\Gamma_{33}}}d_{14}, \quad d_{24} = \sqrt{\frac{\Gamma_{11}\Gamma_{33}}{\Gamma_{22}\Gamma_{44}}}d_{13}, \tag{9}$$
$$d_{34} = -\frac{\sqrt{\Gamma_{11}\Gamma_{33}}d_{13} + \sqrt{\Gamma_{11}\Gamma_{44}}d_{14}}{\sqrt{\Gamma_{33}\Gamma_{44}}}.$$

Although in a system of four QDs many dark states may be spanned, in realistic systems the parameters of biexciton states may be adjusted in such a way that only one particular (for specified values d_{13} and d_{14}) state is blocked. Thus, as in ideal systems and similarly to the double QDs [55], the contribution of any pair of dots to the total biexcitonic population is constant in time ($n_{ij} =$ const).

In realistic QDs one may realize either the super-radiant or subradiant biexciton eigenstate but never both simultaneously as in the ideal system. In both cases the basis is supplemented by a third kind of state which allows for a recombination of one electron-hole pair and trapping of the second exciton. The evolution of a four-QD system prepared in this state is shown in Figs. 5(b) and 5(c). While the population of the biexciton state decreases, the occupation of a single-exciton state increases until it stabilizes at the level corresponding to total trapping of one electron-hole pair [green dashed line in Fig. 5(b)]. The third basis state must be orthogonal to the super-radiant state (8), which equals the requirement of a vanishing transition matrix element between the biexciton state and the single-exciton super-radiant state ($\langle\text{SUPER}|H_{\text{S-rad}}|\text{biexciton}\rangle = 0$) and means that the state $H_{\text{S-rad}}|\text{biexciton}\rangle$ has a subradiant character. The orthogonality to the subradiant biexcitonic state (9) excludes contributions from the two-exciton dark states and thus population trapping occurs only due to formation of a single-exciton subradiant state (5). Since the transition to the state (4) and thus the decay of single-exciton states is forbidden, one exciton is blocked in the ensemble. Because the biexcitonic subradiant states cannot be formed in triple QDs, the requirement defining biexcitonic states which allow blocking of one exciton reduces to the orthogonality to the super-radiant state, and the states take the form

$$\frac{a_{12}\sqrt{\Gamma_{33}}|12\rangle + a_{13}\sqrt{\Gamma_{22}}|13\rangle + a_{23}\sqrt{\Gamma_{11}}|23\rangle}{\sqrt{|a_{12}|^2\Gamma_{33} + |a_{13}|^2\Gamma_{22} + |a_{23}|^2\Gamma_{11}}},$$

where $a_{12} + a_{13} + a_{23} = 0$. In contrast, due to the orthogonality to the super-radiant state the six amplitudes of the biexcitonic states spanned in ensembles of four QDs take the form

$$d_{ij} = a_{ij}\sqrt{\Gamma_{kk}\Gamma_{ll}},$$

where the coefficients (a_{ij}) must satisfy the equation $a_{12} + a_{13} + a_{14} + a_{23} + a_{24} + a_{34} = 0$ and, using further the orthogonality to the subradiant state relations,

$$a_{12}\Gamma_{33}\Gamma_{44} - a_{13}\Gamma_{22}\Gamma_{44} - a_{24}\Gamma_{11}\Gamma_{33} + a_{34}\Gamma_{11}\Gamma_{22} = 0,$$
$$a_{12}\Gamma_{33}\Gamma_{44} - a_{14}\Gamma_{22}\Gamma_{33} - a_{23}\Gamma_{11}\Gamma_{44} + a_{34}\Gamma_{11}\Gamma_{22} = 0.$$

Due to the existence of single-exciton and biexciton optically inactive states in ensembles of four QDs the initial biexcitonic states allow for many combinations of final occupation. An arbitrary fraction of exciton occupation ($\leqslant 2$) may be trapped by an appropriate combination of blocked single excitons and biexcitons due to the contribution to the initial state from dark states and basis states which allow only one electron-hole pair to recombine. Irrespective of the initial number of excitons, ensembles of three QDs allow spanning of only single-exciton subradiant states and thus block only single excitons.

It is important to emphasize that if the system were prepared in a bright biexcitonic state which leads to trapping of single-exciton occupation [Figs. 5(b) and 5(c)], then the pronounced oscillations due to coupling between the dots appear only in the occupation of localized biexciton states, while the amplitudes of oscillations in the populations of single dots are negligible [Fig. 5(c)] compared to the single-exciton initial state [Fig. 2(c)]. This means that the biexciton initial state allows well-defined stable single-exciton subradiant states to be achieved, which is important for the application of quantum computation.

D. *p-i-n* junction

The presence of optical collective effects in realistic QD systems requires high accuracy of the system parameters which may be controlled in the manufacturing stage or by external fields by, e.g., implementing the dots into the intrinsic region of a *p-i-n* junction. This structure provides the possibility of a separate injection of electrons and holes into QDs from both sides of a sample and control of the exciton dynamics and QD parameters through application of contacts on *n*- and *p*-type regions. It has been shown that control with a bias voltage carrier tunneling into a single QD in a *p-i-n* structure incorporated into a microcavity leads to regulated emission of single photons and pairs of photons [64]. The ideas were followed by technological realization of an electrically driven single-photon emitter with a layer of self-organized InAs QDs [65]. Gate voltages constructed over dots allow control of the energies of the dots and dipole moments, but the magnitudes of the decay rates of single QDs (3) depend on the average energy of the ensemble (E) and thus operations on one dot change the decay rates of all QDs in the system.

In order to simulate the possibility of controlling exciton occupation we have calculated the time evolution of the system of four QDs and we have changed the decay rates at some selected time points. The results are shown in Fig. 6 where the system is initially in the collective regime and has been prepared in a single-exciton subradiant state. We assume that the control electric fields are weak enough to exclude dissociated exciton states. As expected, initially the excitation is blocked in the system.

At time 2.5 ns a change of parameters occurs which destabilizes the system and induces quenching of the excitation. The effect may be produced by a variation of the internal electric field in the *p-i-n* junction, which is simulated here by a change of the decay rates of each single dot, and shown by the red solid line in Fig. 6. The decay rates have been changed as follows: $\Gamma_{11} \to 1.4\Gamma_{11}$, $\Gamma_{22} \to 1.3\Gamma_{22}$, $\Gamma_{33} \to 1.2\Gamma_{33}$, $\Gamma_{44} \to 1.1\Gamma_{11}$. In this case the quenching is relatively weak because of a small change in the ratio of the decay rates but the effect is visible. At time 7.5 ns the parameters of each dot are changed back and the system population is again stable.

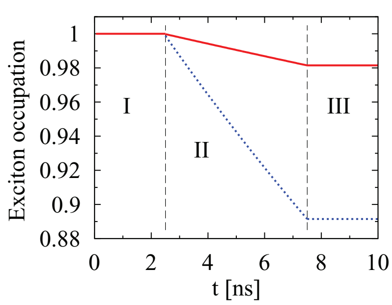

FIG. 6. (Color online) Exciton occupation for a system of four QDs placed at the vertices of a square of side length $r = 30$ nm and prepared initially in a single-exciton subradiant state $\sim(\sqrt{\Gamma_{22}\Gamma_{33}\Gamma_{44}}|1\rangle + \sqrt{\Gamma_{11}\Gamma_{33}\Gamma_{44}}|2\rangle + \sqrt{\Gamma_{11}\Gamma_{22}\Gamma_{44}}|3\rangle - 3\sqrt{\Gamma_{11}\Gamma_{22}\Gamma_{33}}|4\rangle)$. In regions I and III the decay rates are the same as in Fig. 5, i.e., selected such that the system interacts collectively with the radiative surrounding. The red solid line corresponds to changes of the decay rates of individual QDs, and the blue dotted line to the same changes plus changes of the relative angular orientation of the dipoles. In region II the decay rates and angles have been modified such that the ensemble does not interact collectively with its photon environment (see text).

The decay may be enhanced by changing the orientation of the dipoles as shown by the blue dotted line in Fig. 6. All dipoles are parallel in the regions I and III, i.e., all angles $\theta_{ij} = 0$. But now, in addition to the previous variations of Γ_{ij}, the angles are also modified in region II: $\theta_{12} = 0.1$, $\theta_{13} = 0.11$, $\theta_{14} = 0.2$, $\theta_{23} = 0.105$, $\theta_{24} = 0.2$, $\theta_{34} = 0.1$ rad. The misalignment of the dipole moments creates thus a stronger decay. The initial conditions may be restored at any time, which may result in the trapping of a desired fraction of the initial occupation.

IV. CONCLUSIONS

We have studied the optical collective effects due to interaction of multiple quantum dots built of three and four emitters with radiative surroundings. Ensembles of three and more emitters allow spanning of many subradiant states which facilitate preparation of the system in optically inactive single-exciton states and for ensembles of four emitters also in the biexciton subspace.

We specified the conditions which allow the super-radiance phenomena to occur in coupled inhomogeneous systems with different fundamental transition energies and dipole moments (and thus decay rates). We discussed the dynamics of single electron-hole pairs and biexcitons. Although many features typical for identical atoms, such as spontaneous trapping of excitation, may also occur in inhomogeneous QDs there are differences in the dynamics of these systems. In principle, coupling between the dots induces excitation transfer between the dots which together with the possibility to define many dark states in ensembles of three and more dots leads to oscillations in the occupation of single dots. The amplitudes of these oscillations may be considerably reduced if the system is prepared initially in an appropriate biexction state which allows for trapping of one electron-hole pair.

We envision that the presented collective effects may be controlled if the ensemble of dots is placed in the intrinsic region of a *p-i-n* junction with contacts constructed over the dots which due to sufficiently weak electric fields allows control of the dynamics of excitons and thus super-radiant effects.

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APPENDIX: DERIVATION OF EQ. (6)

To express an arbitrary single-exciton state in terms of the super-radiant state (4) and a subradiant state (5) we begin with a derivation of a formula for a localized state $|i\rangle$. The subradiant state which allows canceling out all localized contributions different from the state $|i\rangle$ to the super-radiant state has the form

$$|\mathrm{SUB}\rangle_i = \frac{\sum_{j\neq i}^{N}\Gamma_{jj}|i\rangle - \sqrt{\Gamma_{ii}}\sum_{j\neq i}^{N}\sqrt{\Gamma_{jj}}|j\rangle}{\sqrt{\sum_{j\neq i}^{N}\Gamma_{jj}\sum_{i=1}^{N}\Gamma_{ii}}} \tag{A1}$$

and is orthogonal to the super-radiant state for an arbitrarily chosen state $|i\rangle$. Therefore the localized single-exciton states may be decomposed into a superposition of the super-radiant (4) and subradiant state defined in the formula (A1) according to the equation

$$|i\rangle = \frac{\sqrt{\Gamma_{ii}}|\mathrm{SUPER}\rangle + \sqrt{\sum_{k\neq i}^{N}\Gamma_{kk}}|\mathrm{SUB}\rangle_i}{\sqrt{\sum_{i=1}^{N}\Gamma_{ii}}}. \tag{A2}$$

The above formula allows definition of a sub- and super-radiant component in every single-exciton state,

$$|\mathrm{s}\rangle = \sum_{i=1}^{N} c_i|i\rangle = |\mathrm{SUPER}\rangle' + |\mathrm{SUB}\rangle'.$$

Since there is only one super-radiant state in any system of N QDs, the short-living contribution

$$|\mathrm{SUPER}\rangle' = \frac{\sum_{i=1}^{N} c_i\sqrt{\Gamma_{ii}}}{\sqrt{\sum_{i=1}^{N}\Gamma_{ii}}}|\mathrm{SUPER}\rangle$$

is proportional to the state (4) with a weight factor

$$\frac{\sum_{i=1}^{N} c_i\sqrt{\Gamma_{ii}}}{\sqrt{\sum_{i=1}^{N}\Gamma_{ii}}},$$

while the stable part

$$|\mathrm{SUB}\rangle' = \frac{\sum_{i=1}^{N} c_i\sqrt{\sum_{k\neq i}^{N}\Gamma_{kk}}}{\sqrt{\sum_{i=1}^{N}\Gamma_{ii}}}|\mathrm{SUB}\rangle_i$$
$$= \frac{\sum_{i=1}^{N} c_i\left(\sum_{j\neq i}^{N}\Gamma_{jj}|i\rangle - \sum_{j\neq i}^{N}\sqrt{\Gamma_{ii}\Gamma_{jj}}|j\rangle\right)}{\sum_{i=1}^{N}\Gamma_{ii}}$$

is a combination of N subradiant states (A1), which as a sum of dark states remains optically inactive irrespective of the number of emitters. The component $|\mathrm{SUB}\rangle'$ is proportional to the subradiant state defined by the formula (7) with an amplitude

$$\sqrt{1-\left(\sum_{i=1}^{N} c_i\sqrt{\Gamma_{ii}} \bigg/ \sqrt{\sum_{i=1}^{N}\Gamma_{ii}}\right)^2}.$$

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