Collective optical (superradiant) 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 common photon reservoir. This leads to formation of rapidly decaying (superradiant) and optically inactive (subradiant) states\(^ {2}\) and consequently to the appearance of a vacuum-induced coherence effect which results in occupation trapping.\(^ {2}\)

Although effects resulting from collective coupling of atoms to radiative environment have been known for nearly sixty years, are very well described\(^ {1–4}\) and have been extensively investigated experimentally\(^ {5,6}\) in atomic systems, 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)\(^ {7}\), Bose–Einstein condensate\(^ {8}\), superconducting qubits\(^ {9,10}\), ion Coulomb crystals\(^ {11}\) or depolaritons.\(^ {12}\)

The investigation of superradiant effects have been initiated and driven to a large extend by the promise which the short-living states show for optimization of lasers.\(^ {13}\) Recently the scientific interest has been focused on the concept of “superradiant laser” which allows to increase spectral purity of emitted light.\(^ {14}\) The technological realization of such a device presented in Ref. \(^ {16}\) allows to improve accuracy of atomic clocks\(^ {17,18}\) and thus measurements of gravity\(^ {19}\) and fundamental constants.\(^ {20,21}\) The experimental investigations of the collective effects were mostly restricted to the analysis of the superradiant states which appear spontaneously in the cascade emission and manifest themselves as a maximum in the intensity and/or photon emission rate.\(^ {2,5,22}\)

Although the observation of the subradiance phenomena was also reported in the atomic ensembles\(^ {6}\) as the opposite of the superradiant states, the “on demand” preparation of subradiant states is much more difficult, and therefore the possibilities they give were much less investigated experimentally. Recently, the preparation of an optically inactive state was reported in a system of superconducting qubits\(^ {23}\) and in a diatomic molecule in an optical lattice.\(^ {24}\) 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.\(^ {25–26}\) This makes them useful for quantum information processing especially for noiseless encoding of quantum information.\(^ {27–28}\) The stable states also allow to construct a scalable quantum processor, quantum memories,\(^ {29,30}\) nonlinear sign-shift gate\(^ {31}\) and storing time-bin qubits\(^ {32}\) for quantum cryptography. Interesting group of Dicke states is formed of single-excitation blockade.\(^ {33–36}\) Systems composed of two and more coupled QDs attract much scientific focus due to the richness of their properties which pave the way to new technological application. Already pairs of quantum dots allow for long-time storage of quantum information,\(^ {37}\) conditional optical control of carrier states,\(^ {38}\) implementation of a two-qubit quantum gate,\(^ {39}\) optical writing of information on the spin state of the dopant Mn atom\(^ {40}\) and construct quantum nanoantennas due to the collective phenomena.\(^ {41}\) Systems of three QDs enable to realize two different kinds of entanglement\(^ {42}\) teleportation via superradiance\(^ {43}\), CNOT gates\(^ {44}\) and the control of spin blockade.\(^ {45}\) Moreover, in these systems the collective transport effects (electronic Dicke or Kondo-Dicke effect) may be realized\(^ {46,47}\) and lead to the enhancement of thermo-electric efficiency.\(^ {48}\) Arrays of QDs allow to reduce the effect of pure dephasing on quantum information encoded in excitonic states.\(^ {49}\)

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 to realize many stable states which occur at different exciton occupations of the single emitters. This allows to store quantum information in these systems and control light emission. Although the superradiant effects are well described in systems of identical atoms the description of such phenomena in QD ensembles requires taking into account characteristic for those system properties which distinguish them from natural atoms. Therefor we include in our model the fundamental energy mismatches, different dipole moments of single dots and coupling which induces excitation transfer between single dots, but conserves the total population of the ensemble. In our previous works concerning double QDs has been showed 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 the energy splitting much below the present technological feasibility. This decoherence effect may be strongly reduced by sufficiently strong coupling between the dots and fully overcome in double QDs with different decay rates. In this paper we extend the results of two QDs and specify conditions which allow to take advantage of the superradiant phenomena in fully inhomogeneous QDs systems/multiple QDs. We analyze the dynamics of one 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 to trap two excitons or prepare a system in a biexcitonic state which allows to recombine only one electron-hole pair. Due to existence of two or more subradiant single-exciton eigenstates and coupling between the dots the occupation of single 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, we conclude the paper in Sec. IV.

II. THE SYSTEM

The investigated system consists of $N$ ($N = 3, 4$) quantum dots (QDs) 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. These assumptions allow us to treat every QD as a two-level system which may be either empty or contain an exciton and thus describe the set of $N$ QDs as an $2^N$-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 $i$th QD, biexcitonic states $|ij\rangle$ referring to electron-hole pairs residing in the $i$th and $j$th QDs, states $|ijk\rangle$ with three QDs, $i$th, $j$th, and $k$th ($1 \leq i,j,k \leq N$), occupied with an exciton $\cdots$.

Present manufacturing technology does not allow to produce on demand systems of QDs 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 $i$th QD. We impose $\sum_i \alpha_i = 0$ and $\sum_i \alpha_i^2 = 1$, such that $\Delta^2$ is the mean square variation of the transition energies.

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

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

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

In this frame the Hamiltonian of the system is

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

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

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

where $B_{ij}$ are biexcitonic shifts due to the interaction of static dipole moments of $i$th and $j$th QD, $B_{i23}$ is a deviation of energy caused by interaction of the dipole moments of three dots, etc. $V_{ij} = V_0 \exp \left [ -r_{ij}/r_0 \right ]$ are the distant-dependent amplitudes of the coupling which is responsible for excitation transfer from the $i$th QD to the $j$th one and vice versa without a loss of the excitation, where $V_0$ is a constant amplitude, $r_{ij}$ is the distance between the $i$th and $j$th QD and $r_0$ the spatial range of the interaction. The eigenstate of the Hamiltonian $H$
do not mix quantities associated with different exciton numbers.

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

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

where

$$g_{k\lambda}^{(j)} = i d_j \hat{\varepsilon}_\lambda(k) \sqrt{\frac{\hbar \omega_k}{2 \varepsilon_0 \varepsilon_r v}}$$

is a coupling constant for the $j$th QD. Here $d_{(j)}$ is the inter-band dipole moment for the $j$th QD, $\hat{\varepsilon}_\lambda(k)$ is the unit polarization vector of the photon mode with polarization $\lambda$, $\varepsilon_0$ is the vacuum dielectric constant, $\varepsilon_r$ 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 a form

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

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

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

where

$$\Gamma_{ij} = \frac{E^3}{3\pi\varepsilon_0\varepsilon_r\hbar^4} d_i \cdot d_j^\ast.$$  

Since for $i = j$ the Eq. (3) describes the spontaneous decay rates of single QDs, the mixed (off-diagonal) decay rates may be expressed in terms of single QD quantities,

$$\Gamma_{ij} = \frac{1}{\sqrt{\Gamma_{ii} \Gamma_{jj}}} d_i \cdot d_j^\ast,$$

where $d_i = d_i / d_i$ and $d_i \cdot d_j = e^{i\eta} \cos \theta$, here $\eta$ is an irrelevant phase and $\theta$ is a small angle between the dipole moments which depends of light-hole admixture.

In the numerical simulations we assume constant energy mismatches with the parameters $\alpha_1 = 2\sqrt{3} \sqrt{6}$, $\alpha_2 = 4\sqrt{3} \sqrt{6}$, $\alpha_3 = -6\sqrt{3} \sqrt{6}$, $\alpha_4 = 0$ and $\Delta = 1$ meV. For the coupling amplitudes we take $V_0 = 5$ meV and $r_0 = 15$ nm.

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. II we illustrate the numbering of the QDs and their spatial arrangement. In Sec. 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.

A. Single-exciton states of ideal quantum dots

The collective effects were described for the first time in ensembles of uncoupled identical atoms where all the emitters have the same transition energies and dipole moments. 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. We consider different decay rates of single dots and hence different dipole moments.

The coupling of excitons to their radiative environment described in the Dicke limit by the Hamiltonian and the 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 to define rapidly decaying (superradiant) and optically inactive (subradiant) states. By definition the superradiant initial state ($\text{SUPER}$) corresponds to the maximum transition probability, whereas the subradiant states ($\text{SUB}$) refer to the 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 system may decay only to the ground state ($|0\rangle$), the proportionality $g_{k\lambda}^{(j)} \sim \sqrt{\Gamma_{jj}}$ allows to write the short-living.
states in the form
\[ |\text{SUPER}\rangle = \frac{\sum_{i=1}^{N} \sqrt{\Gamma_{ii}} |i\rangle}{\sqrt{\sum_{i=1}^{N} \Gamma_{ii}}}, \]
and express the stable superpositions as
\[ |\text{SUB}\rangle = \frac{\sqrt{\sum_{i=1}^{N} |a_i|^2 N \prod_{j=1}^{N} \Gamma_{jj}}}{\sum_{i=1}^{N} a_i}, \]
where the coefficients \(a_i\) satisfy \(\sum_{i=1}^{N} a_i = 0\). The above states are also known as the Dicke states. Irrespective of the number of emitters there is only one superradiant state in the system of a particular number of QDs, whereas the only structure which realizes only one dark state is a double quantum dot. The systems of three and more QDs allow to realize an arbitrary number of dark states of the form \(\text{SUB}\) since there are may combinations of the parameters \(a_i\), for which the transition matrix element \(\langle 0 | H_{\text{S-rad}} | \text{SUB}\rangle = 0\).

The consequence of the coexistence of rapidly decaying and stable states is the effect of spontaneous trapping of excitons. 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 QD \(i\), may be expressed as a combination of the superradiant state \(\text{SUPER}\) and a dark state of type \(\text{SUB}\),
\[
|s\rangle = \left(\frac{\sum_{i=1}^{N} c_i \sqrt{\Gamma_{ii}}}{\sqrt{\sum_{i=1}^{N} \Gamma_{ii}}}\right) |\text{SUPER}\rangle + \sqrt{1 - \left(\frac{\sum_{i=1}^{N} c_i \sqrt{\Gamma_{ii}}}{\sqrt{\sum_{i=1}^{N} \Gamma_{ii}}}\right)^2} |\text{SUB}\rangle_s, \tag{6}
\]
where the dark state is
\[
|\text{SUB}\rangle_s = \frac{\sum_{i=1}^{N} c_i \left[ \left(\sum_{j \neq i}^{N} \sqrt{\Gamma_{jj}}\right) |i\rangle - \sum_{j \neq i}^{N} \sqrt{\Gamma_{ii} \Gamma_{jj}} |j\rangle \right]}{\sqrt{\left(\sum_{i=1}^{N} \Gamma_{ii}\right) \left(\sum_{i=1}^{N} \Gamma_{ii} - \sum_{i=1}^{N} c_i \sqrt{\Gamma_{ii}}\right)^2}}. \tag{7}
\]

The derivation of the Eq. \(6\) and \(7\) is done in the the Appendix. The collective coupling to the radiative surrounding induces emission only from the superradiant state and thus the fraction of excitation initially spanned in the dark state, \(1 - \left(\frac{\sum_{i=1}^{N} c_i \sqrt{\Gamma_{ii}}}{\sqrt{\sum_{i=1}^{N} \Gamma_{ii}}}\right)^2\), remains unaffected. Since the only single-exciton state which decays totally is the superradiant state, we define a state \(|s\rangle\) as being bright if its superradiant contribution does not vanish, i.e., if a system prepared in that state partially recombines and only a part of the initial exciton occupation remains trapped.

In Fig. 2(a)-(c) we show the dynamics of a single-exciton state 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 \((\theta = 0)\) of different magnitudes. As can be seen...
in Fig. 2(a), the dynamics of a subradiant state
\[
2\sqrt{\Gamma_{22}\Gamma_{33}}|1\rangle + 3\sqrt{\Gamma_{11}\Gamma_{33}}|2\rangle - 5\sqrt{\Gamma_{11}\Gamma_{22}}|3\rangle
\]
\[
4\Gamma_{22}\Gamma_{33} + 9\Gamma_{11}\Gamma_{33} + 25\Gamma_{11}\Gamma_{22}
\]
is indeed unaffected by the photon reservoir and neither the total exciton occupation nor occupations of single dots \((n_{1,2,3})\) change due to radiative environment. In Figs. 2(b) we show the evolution of the exciton occupations of a system prepared initially in a bright state \((5|1\rangle + |2\rangle)/26\) and in Fig. 2(c) we show the corresponding coherences. As expected, the coupling to the photon reservoir spans the excitation into the sub- and superradiant states according to Eq. (9) and induces emission only from the short-living state
\[
\sqrt{\Gamma_{11}|1\rangle + \sqrt{\Gamma_{22}|2\rangle + \sqrt{\Gamma_{33}|3\rangle}}
\]
[Fig. 2(b)]. The emission from the above state induces decay of the total exciton occupation and excitation transfer which leads to the redistribution of the occupations of single dots. Since all of the localized single-exciton states \(|i\rangle\) contribute to the superradiant state \((1)\), 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 [blue and green lines in Fig. 2(b)]. 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 increase due to the excitation transfer [blue-dotted line in Fig. 2(b) and the inset to Fig. 2(b)]. The excitation dynamics takes place until occupations of all dots stabilizes at certain levels corresponding to the dark state defined in the Eq. 7. During the emission process also the 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 non-zero level [2(c)].

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 eigenstates of the system and different dipole moments allowing the superradiant state to be a non-equal superposition of the localized states \(|i\rangle\) [Eq. (3)].

The single-exciton eigenstates of the system depend on the energy mismatches and coupling between the dots, while the collective properties are defined by the inter-play of decay rates. If the single-exciton decay rates [Eq. (3)] for \(i = j\) are adjusted in such a way that the superradiant state \((1)\) corresponds to one of the eigenstates of the system, then the inhomogeneous system 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 (for more detail see the Appendix 13). The amplitudes \(c_i\) of a single-exciton state orthogonal to the superradiant state \((1)\) must satisfy the equation \(\sum_i c_i\sqrt{\Gamma_i} = 0\) which implies the condition \(\langle 0| H_{S-Rad}| SUB\rangle = 0\) defining a subradiant state. Therefore, if one of the eigenstates has a superradiant character then the other eigenstates of a system are optically inactive.

In Figs. 2(d) 2(f) we show the evolution of a realistic group of three QDs placed in the corners of an equilateral triangle, we assume non-equal fundamental transition energies \((\Delta = 1\text{ meV})\), non-vanishing coupling between the systems and parallel dipole moments \((\theta = 0)\). 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) 2(c), where the decay rates of single dots take the same values as in Figs. 2(d) 2(f). As can be seen in Fig. 2(d), ensembles with energy mismatches on the order of meV allow to realize perfectly stable subradiant states of the form \((3)\). Here, as in the ideal case, any single-exciton eigenstate may be decomposed into sub- and superradiant component according to Eq. (9) and also in this case the superradiant state is the only state which decays totally. For an arbitrary set of decay rates, which do not correspond to the superradiant eigenstate state, the exciton occupation of a system prepared initially in a state of the form \((3)\) is quenched and the decay of the state \((3)\) is slowed down compared to the “collective regime”.

Although sub- and superradiant states may exist in the appropriately designed realistic systems, the dynamics of single quantum dot occupations differs considerably from the discussed in the previous section ideal case, 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 the population of single QDs. In a double QD system the oscillation amplitudes decrease and the occupations stabilize at levels corresponding to the only one dark state of the system. In the multiple QDs
The system of three and more QDs allows many different planar arrangements of the dots. Since the coupling 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 geometry of the system. Using the geometry design defined in Fig. 4(a) we calculate the dependence of the decay rates \( \Gamma_{11} \) and \( \Gamma_{33} \) on the spatial arrangements 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 \( \alpha \) from 60 degrees to linear design, i.e., we increase the distance between dots 1 and 3. As seen in Figs. 4(a) and 4(b) the values of the decay rates necessary to form the “collective regime” slightly decrease with increasing angle \( \alpha \) and this effect is irrespective of the interplay of distances \( r_{12} \) and \( r_{23} \) [compare Figs. 4(a) and 4(b)]. Smaller decay rates result, according to Eq. 6 in increased number of trapped population [Figs. 4(c) and 4(d)]. From comparison of Figs. 4(c) and 4(d) it is seen that the differences in final occupation trapping resulting from spatial arrangement of the dots are much less pronounced in dense ensembles, where the coupling amplitude \( V_{13} \) change less with the angle \( \alpha \). If coupling between two out of three QDs is much stronger than coupling of that dots with the third one (e.g. \( V_{13} \gg V_{12}, V_{23}, r_{13} \gg r_{12}, r_{23} \)) then one of the system eigenstates has large contribution from the localized state \( |2⟩ \) and to achieve “collective regime” the decay rate \( \Gamma_{22} \) must be much smaller from \( \Gamma_{11} \) and \( \Gamma_{33} \). In the limiting case of

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**FIG. 3:** Oscillations in the exciton occupation of a single QD. All figures show picosecond scale intervals of the occupation of the QD number 1 \( (n_1) \) shown by the blue line in Fig. 2(e).

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**FIG. 4:** Decay rate dependence on the spatial arrangement of a system of three QDs (i.e., on the angle \( \alpha \) defined in Fig. 4(a) for \( r_{12} = 30 \text{ nm} \) (a) and \( r_{12} = 15 \text{ nm and } r_{23} = 20 \text{ nm} \) (b)). [(c) and (d)] Exciton occupations corresponding to arrangements presented in Figs. (a) and (b), respectively, for the initial state \( (5|1⟩ + |2⟩)/\sqrt{2} \) and for few values of the angle \( \alpha \). We assume constant decay rate of the QD number 2 \( \Gamma_{22} = 0.002 \text{ ns}^{-1} \).
vanishing couplings $V_{12}$ and $V_{23}$ the localized state $|2\rangle$ becomes the system eigenstate and the decay rate $\Gamma_{22}$ vanishes too. Consequently the pair of coupled dots acts as a double QD while the third dot does not contribute to the evolution. The similar effects in the dynamics of single excitation are observed for excitons confined in ensembles of four and more dots.

C. Biexciton states

Multiple QDs built out of three and four units allow to delocalize more than one exciton and thus produce more complex collective effects. Below we focus on biexciton states which in general are described by a vector $|\text{biexciton}\rangle = \sum_{i,j} N_{ij} b_{ij}|i\rangle|j\rangle$, where $\sum_{ij} |b_{ij}|^2 = 1$. The realistic ensembles of QDs ($\Delta \neq 0$, $V_{ij} \neq 0$ and $B_{ij} \neq 0$) permits superradiance 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 superradiant 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 superradiant state may be formed provided one of the single-exciton eigenstates has a superradant character. Thus the biexcitonic collective effects occur only if they are present in the single-exciton subspace. Due to the equal number of single-exciton eigenstates and single QD decay rates [4] the "collective regime" in the single exciton domain may be achieved by adjusting only the decay rates. As will be explained in detail below, the same rates $\Gamma_{ij}$ define the biexcitonic Dicke states. Thus, in order to achieve the "collective regime" in the two-exciton subspace one has to also appropriately adjust the spatial arrangement of the dots and/or energies.

The superradiant two exciton superpositions may be spanned as well in ensembles of four emitters as in triple QDs and take a form

$$|\text{SUPER}\rangle_B = \sum_{i,j,i\neq j}^{N} \sqrt{\Gamma_{ii} \Gamma_{jj}} |i\rangle |j\rangle. \quad (8)$$

Similarly to the single exciton case, the biexcitonic superradiant states are defined by the maximum value of the transition probability ($\sim |\langle \text{SUPER}|H_{S-rad}|\text{biexciton}\rangle|^2$), but this time from the initial biexciton state to the final single-exciton superradiant state [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 superradiant states. As can be seen in Fig. 5(a), due to the decay of the biexciton superradiant state the excitation is initially transferred to the single-exciton states which reaches a maximum population and then is totally quenched, together with the biexciton state. In this case the initial state has been selected such that the transition probability into subradiant states [5] vanishes.

Two excitons can be blocked in a system if the transition from the state $|\text{biexciton}\rangle$ to the single-exciton state is forbidden, i.e. the transition matrix element $\langle \text{single}|H_{S-rad}|\text{biexciton}\rangle$ vanishes. This situation is illustrated in in Fig. 5(b). Due to infinite number of possible single-exciton states this condition reduces to the requirement $H_{S-rad}|\text{biexciton}\rangle = 0$. To simplify the description we define non-normalized amplitudes $d_{ij}$ in such a way that the amplitudes of biexciton states $b_{ij} = d_{ij}/(\sum_{ij} d_{ij})$. For a triple QD the condition leads to a system of equations:

$$d_{12} \sqrt{\Gamma_{22}} + d_{13} \sqrt{\Gamma_{33}} = 0,$$

$$d_{12} \sqrt{\Gamma_{11}} + d_{23} \sqrt{\Gamma_{33}} = 0,$$

$$d_{13} \sqrt{\Gamma_{11}} + d_{23} \sqrt{\Gamma_{22}} = 0.$$
which is satisfied only for vanishing amplitudes $d_1 = d_2 = d_3 = 0$. This means that it is impossible to block two excitons in a system of three QDs. The coefficients of a stable biexcitonic state spanned in a system of four QDs must satisfy the system of equations

\[
\begin{align*}
    d_{12} \sqrt{\Gamma_{22}} + d_{13} \sqrt{\Gamma_{33}} + d_{14} \sqrt{\Gamma_{44}} &= 0, \\
    d_{12} \sqrt{\Gamma_{11}} + d_{23} \sqrt{\Gamma_{33}} + d_{24} \sqrt{\Gamma_{44}} &= 0, \\
    d_{13} \sqrt{\Gamma_{11}} + d_{23} \sqrt{\Gamma_{22}} + d_{34} \sqrt{\Gamma_{44}} &= 0, \\
    d_{14} \sqrt{\Gamma_{11}} + d_{24} \sqrt{\Gamma_{22}} + d_{34} \sqrt{\Gamma_{33}} &= 0,
\end{align*}
\]

which leads to the condition for the $d_{ij}$ numbers in a form

\[
\begin{align*}
    d_{12} &= -\frac{\sqrt{\Gamma_{33}} d_{13} + \sqrt{\Gamma_{44}} d_{14}}{\sqrt{\Gamma_{22}}}, \\
    d_{23} &= \frac{\sqrt{\Gamma_{11}} \sqrt{\Gamma_{44}} d_{14} - \sqrt{\Gamma_{22}} \sqrt{\Gamma_{33}} d_{13}}{\sqrt{\Gamma_{33}}}, \\
    d_{34} &= -\frac{\sqrt{\Gamma_{11}} \sqrt{\Gamma_{33}} d_{13} + \sqrt{\Gamma_{11}} \sqrt{\Gamma_{44}} d_{14}}{\sqrt{\Gamma_{33}} \sqrt{\Gamma_{44}}},
\end{align*}
\]

Although in a system of four QDs many dark states may be spanned, in realistic systems the energies of biexcitons 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 the ideal atomic systems and similarly to the double QDs, the contribution of any pair of QDs to the total biexcitonic population is constant in time.

In realistic QDs one may define either superradiant or subradiant eigenstate but never both simultaneously as in the ideal atomic system. In both cases the basis is supplemented by a third kind of states which allow for many combinations of final occupation. An arbitrary fraction of exciton occupation ($\leq 2$) may be trapped by an appropriate combination of blocked single-excitons and biexcitons due to contribution to the initial state of dark states and basis states which allow to recombine only one electron-hole pair. The occupation may be blocked in a single-exciton state if the subradiant biexciton state did not contribute to the initial state. Irrespective of the initial number of excitons, ensembles of three QDs allow to span only single-exciton subradiant states and thus block only single-excitations.

It is important to emphasis that if the system was prepared in a bright biexciton state which leads to trapping of single-exciton occupation [Figs. 3(b) and 3(d)], then the pronounced oscillations due to coupling between the dots appear only in occupation of localized biexciton states, while the amplitudes of oscillations in the populations of single dots are negligible [5(d)] compared to single-exciton initial state [Fig. 2(d) and 2(e)]. This means that the biexciton initial state allows to achieve nearly stable single-exciton subradiant states, 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 a system parameters which may be controlled on the manufacturing stage or by external fields by, e.g. implementing the dots into the intrinsic region of p-i-n junction. This structure provides a possibility of a separated injection of electrons and holes into QDs from both sides of a sample and control of exciton dynamics and QD parameters through application of contacts on n- and p- type regions. It has been shown that controlled 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\cite{1}. The ideas were followed by a technological realization of an electrically driven single-photon emitter with a layer of self-organized InAs QDs\cite{2}. The gate voltages constructed over dots allow to control energies of the dots and dipole moments, but the magnitudes of decay rates of single QDs \cite{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. This provides a possibility to control the final occupation of a system not only by appropriate preparation of the initial state but also during the system evolution.

In order to simulate this possibility 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 a single-exciton \((a)\) and biexciton \((b)\) subradiant state. We assume that the control electric fields are weak enough to exclude “dissociated” exciton states. As expected, initially the excitation(s) is (are) 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 lines in Fig. 6. The decay rates have been changed as follows: \(\Gamma_{11} \rightarrow 1.4\Gamma_{11}, \Gamma_{22} \rightarrow 1.3\Gamma_{22}, \Gamma_{33} \rightarrow 1.2\Gamma_{33}, \Gamma_{44} \rightarrow 1.1\Gamma_{44}\). In this case the quenching is relatively weak which shows that the collective effects in coupled ensembles are not very sensitive to the magnitudes of the dipole moments. At time 7.5 ns the parameters of each dot are changed back.

The decay may also be enhanced by changing the orientation of the dipoles as shown by the blue dotted lines 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 \(\Gamma_{ij}\) the angles are also modified in the regions II: \(\theta_{12} = 0.1, \theta_{13} = 0.11, \theta_{14} = 0.2, \theta_{23} = 0.105, \theta_{24} = 0.2, \theta_{34} = 0.1\) radians. The misalignment of the dipole moments creates thus a stronger decay. A contacted p-i-n structure allows the control of the orientation of the moments. The initial conditions may be restored at any time which may result in the trapping of a desired fraction of the initial occupation.

FIG. 6: Exciton occupation for a system of four QDs prepared initially in a single-exciton \((a)\) and biexciton \((b)\) subradiant state. In regions I and III of both panels the parameters are the same as in Fig. 5(b), i.e., selected such that the systems interact collectively with the radiative surrounding. The initial states in panel \((a)\) is \(\sim (\sqrt{11}\Gamma_{22}\Gamma_{33}\Gamma_{44}|1\rangle + \sqrt{11}\Gamma_{22}\Gamma_{33}\Gamma_{44}|2\rangle + \sqrt{11}\Gamma_{22}\Gamma_{33}\Gamma_{44}|3\rangle - 3\sqrt{11}\Gamma_{22}\Gamma_{33}\Gamma_{44}|4\rangle)\), and in panel \((b)\) like in Fig. 5(b). The red solid lines correspond to changes of the decay rates of individual QDs, whereas the blue dotted lines to the same changes plus changes of the relative angular orientation of the dipoles. In regions II the decay rates and angles have been modified such that neither of the system eigenstates corresponds to a subradiant state (see text).

IV. CONCLUSIONS

We have studied the optical collective effects due to interaction of multiple quantum dots built of three and four emitters with the radiative surrounding. Ensembles of three and more emitters allow to span many subradiant states which facilitate preparation of the system in an optically in active state and for ensemble of four emitters also in the biexciton subspace.

We specified the conditions which allow the superradiance 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 like spontaneous trapping of excitation, may also occur in inhomogeneous systems there are differences in the dynamics of these systems. In principle, coupling between the dots induces excitation transfer between the dots which together with a possibility to define many dark systems in ensembles of three and more dots lead to oscillations in the occupation of single dots.

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 allow to control energies of the dots and thus also decay rates and collective effects.

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Appendix A: Derivation of the equation \(\psi(t)\)

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

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

(A1)

and is orthogonal to the superradiant state for an arbitrarily chosen state $|i\rangle$. Therefore the localized single-exciton states may be decomposed into a superposition of a number of emitters. The component $\sum_{dark states}$ remains optically inactive irrespective of a number of emitters. The component $|\text{SUB}\rangle$ is proportional to the state $|\text{SUB}\rangle$ with a weight factor

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

while the stable part

$$|\text{SUB}\rangle = \frac{\sum_{i=1}^N c_i \Gamma_{ii}}{\sqrt{\sum_{i=1}^N \Gamma_{ii}}} |\text{SUB}\rangle_i$$

is a combination of $N$ subradiant states $A2$, which as a sum of dark states remains optically inactive irrespective of a number of emitters. The component $|\text{SUB}\rangle'$ is proportional to the subradiant state defined by the formula $A2$ with an amplitude $\left(1 - \left(\frac{\sum_{i=1}^N c_i \sqrt{\Gamma_{ii}}}{\sqrt{\sum_{i=1}^N \Gamma_{ii}}}\right)^2\right) \cdot \sqrt{\sum_{i=1}^N \Gamma_{ii}}$.

**Appendix B: Single-exciton eigenstates for a system of three QDs**

The eigenvalues of the Hamiltonian $A$ for $N = 3$ and $\Delta_3 = - \Delta_1 - \Delta_2$ are

$$E_1 = \sqrt{A - 3B^2} - B, \quad E_2 = -\sqrt{A - 3B^2} - B, \quad E_3 = 2B,$$

where

$$A = V_{23}^2 + V_{13}^2 + V_{12}^2 + \Delta_2^2 + \Delta_1 \Delta_2 + \Delta_1^2,$$

$$B = \frac{-\text{Im}(C + i) \sqrt{A}}{\sqrt{3(C^2 + 1)^2}},$$

$$C = \frac{\sqrt{4A^3/27 - D^2}}{D},$$

and

$$D = 2V_{12}V_{13}V_{23} - \Delta_1(V_{23}^2 - V_{12}^2) - \Delta_2(V_{13}^2 - V_{12}^2) - \Delta_1 \Delta_2 (\Delta_1 + \Delta_2).$$

The corresponding eigenstates are defined by the formula

$$|\Psi_i\rangle = \frac{1}{\sqrt{M_i}} \left( (V_{12}V_{23} + (E_i - \Delta_2)V_{13}) |1\rangle + \left( (V_{12}V_{13} + (E_i - \Delta_1)V_{23}) |2\rangle + \left( (E_i^2 - V_{12}^2 - E_i(\Delta_1 + \Delta_2) + \Delta_1 \Delta_2) |3\rangle \right) \right),$$

where

$$M_i = \left( (V_{12}V_{23} + (E_i - \Delta_2)V_{13})^2 + \left( (V_{12}V_{13} + (E_i - \Delta_1)V_{23})^2 + \left( (E_i^2 - V_{12}^2 - E_i(\Delta_1 + \Delta_2) + \Delta_1 \Delta_2)^2 \right) \right) \right).$$

The fully inhomogeneous (different transition energies and dipole moments) coupled system interacts collectively with its radiative environment if one of the eigenstates corresponds to the superradiant state $A2$. This means that amplitudes of one of the eigenstates $B3$ must satisfy a system of equations

$$\frac{V_{12}V_{23} + (E_i - \Delta_2)V_{13}}{\sqrt{M}} = \frac{\sqrt{11}}{\sqrt{11} + \Gamma_{22} + \Gamma_{33}},$$

$$\frac{V_{12}V_{13} + (E_i - \Delta_1)V_{23}}{\sqrt{M}} = \frac{\sqrt{22}}{\sqrt{11} + \Gamma_{22} + \Gamma_{33}},$$

$$\frac{E_i^2 - V_{12}^2 - E_i(\Delta_1 + \Delta_2) + \Delta_1 \Delta_2}{\sqrt{M}} = \frac{\sqrt{33}}{\sqrt{11} + \Gamma_{22} + \Gamma_{33}}.$$
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