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Enhanced spontaneous optical emission from inhomogeneous ensembles of quantum dots is induced by short-range coupling

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We study theoretically the spontaneous emission from an inhomogeneous ensemble of quantum dots in the weak excitation limit. We show that collective, superradiancelike effects lead to an enhanced emission rate in the presence of sufficiently strong coupling between the dots, in agreement with experimental observations, which means that the quantum dot sample cannot be treated as an ensemble of individual emitters. We demonstrate also that the collective behavior of the quantum dot system relies on short-range interactions, while long-range dipole couplings are too weak to have any impact on the emission dynamics for a system with a realistic degree of inhomogeneity.

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The unique atomiclike optical properties of semiconductor quantum dots (QDs) allow one to transfer ideas and technologies from atomic systems to solid state structures and devices. Such quantum optics concepts successfully implemented in QD systems range from laboratory-scale coherent optical control experiments[1] to commercially available devices, such as QD lasers.[2] While similarities between atoms and QDs may be very useful, these two systems differ essentially in some respects. One of these differences is the inhomogeneity of the QD characteristics due to the inevitable randomness of the physical properties of these structures. This becomes important when collective interaction of QDs with their environment is essential, e.g., in lasers or some quantum information devices.[3] Understanding the collective evolution of nonidentical, coupled emitters can also be beneficial to the current study of other semiconductor,[4] plasmonic,[5] hybrid,[6] or atomic[7] systems where a similar interplay of collectivity, inhomogeneity, and interactions plays a crucial role.

In an experiment performed on a dense ensemble of CdSe QDs,[8] the decay of luminescence was shown to accelerate as the number of emitting dots increased. This means that, in their interaction with the radiation field, the QDs in the ensemble are not independent objects. The existence of such a cooperative effect for QDs is remarkable as the ensemble broadening of the transition energies is three to four orders of magnitude larger than the radiative broadening of the QD emission line. The experimentally observed effect was quite strong already for fewer than 100 QDs so that an accidental spectral overlap between two or more dots was clearly very unlikely. The cooperative effect was therefore attributed to the existence of long-range (LR) coupling between the dots.

Subsequent theoretical analysis[9–11] confirmed that coupling between nonidentical QD emitters stabilizes the collective effects in the spontaneous emission. However, in order to overcome the inhomogeneity the coupling must be at least comparable to the transition energy mismatch between the emitters,[9,10] while the LR dipole (Förster) couplings do not exceed a fraction of meV even for dots separated by a few nanometers distance[12] and drop down to about 1 μeV for interdot separations around 30 nm, which is the average value for the experimentally studied sample.[8] These values are at least two orders of magnitude lower than the transition energy inhomogeneity, which is on the order of a few tens of meV. Thus, the LR dipole couplings seem unlikely to underlie the enhanced emission. It is known, however, that also other couplings of a different nature exist in QD ensembles.[13,14]

In this Rapid Communication, we present the results of theoretical modeling of the spontaneous emission from an inhomogeneous ensemble of QDs and clarify the origin of the observed enhanced spontaneous emission. We extend the standard model of identical emitters[15,16] by including ensemble inhomogeneity and simulate the evolution of a planar ensemble of nonidentical, randomly distributed QDs in the weak excitation regime. Our analysis confirms that the cooperative interaction of the QDs with the surrounding electromagnetic (EM) vacuum can lead to an increased spontaneous emission rate. We show, however, that the dipole coupling between the QD emitters is insufficient for the appearance of cooperative behavior of these strongly inhomogeneous systems. Enhanced emission appears only if one includes short-range (SR) couplings between the dots, which may be due to a combination of tunneling (wave function overlap between the neighboring QDs) and Coulomb correlations.

We model a system of N QDs located at points $\boldsymbol{r}_\alpha$. The typical energy distance to p-shell exciton states in CdSe/ZnSe dots is at about 100 meV,[17] much larger than the inhomogeneous broadening of the s-shell states, which allows us to restrict the discussion to the fundamental transition. In addition, we fix the polarization. Hence each of the dots is represented as a two-level system with the basis states $|0\rangle_\alpha$ and $|1\rangle_\alpha$, where α labels the dots. The fundamental transition energy in the dot α is E_α. The dots are coupled by SR couplings described effectively by the coupling potentials $\Omega^{(\mathrm{T})}_{\alpha\beta}$. The Hamiltonian for the EM interactions is transformed to the dipole form using the Power-Zienau-Wooley transformation[18] and reads

$$H = \sum_{\alpha=1}^{N} E_\alpha \sigma_\alpha^\dagger \sigma_\alpha + \sum_{\alpha\neq\beta} \Omega^{(\mathrm{T})}_{\alpha\beta} \sigma_\alpha^\dagger \sigma_\beta + \sum_{\boldsymbol{k}\lambda} \hbar\omega_k b_{\boldsymbol{k}\lambda}^\dagger b_{\boldsymbol{k}\lambda} - \frac{1}{\varepsilon_0 \varepsilon_\mathrm{r}} \sum_{\alpha=1}^{N} (\boldsymbol{d}_0 \sigma_\alpha + \boldsymbol{d}_0^* \sigma_\alpha^\dagger) \cdot \boldsymbol{D}(\boldsymbol{r}_\alpha),$$

where the EM field is represented by the displacement operator

$$\boldsymbol{D}(\boldsymbol{r}) = i \sum_{\boldsymbol{k}\lambda} \sqrt{\frac{\hbar\varepsilon_0\varepsilon_\mathrm{r}\omega_k}{2V}}\, \hat{\boldsymbol{e}}_{\boldsymbol{k}\lambda} b_{\boldsymbol{k}\lambda} e^{i\boldsymbol{k}\cdot\boldsymbol{r}} + \mathrm{H.c.}$$

Here $b_{\boldsymbol{k}\lambda}$ is the annihilation operator for a photon with wave vector $\boldsymbol{k}$ and polarization λ, ω_k is the photon frequency, $\hat{e}_{\boldsymbol{k}\lambda}$ is the unit polarization vector, $\sigma_\alpha = (|0\rangle\langle 1|)_\alpha$ is the transition operator for the dot α, $\boldsymbol{d}_0$ is the interband matrix element of the dipole moment (we assume that all the interband dipoles are identical), c is the speed of light in vacuum, ε_0 is the vacuum permittivity, and ε_r is the dielectric constant of the semiconductor medium.

The evolution equation for any electronic operator Q can be obtained along the lines worked out in Ref. 19: One writes down the equations of motion for the electronic and photonic operators, eliminates the latter, neglects the off-resonant terms and radiation-induced energy shifts, and performs the Markov approximation. It is assumed that no external fields are present. The resulting evolution equation for the average value of the electronic operator Q has the form

$$\langle \dot{Q}\rangle = \frac{i}{\hbar}\sum_\alpha \epsilon_\alpha \langle[\sigma_\alpha^\dagger\sigma_\alpha, Q]\rangle + \sum_{\alpha\neq\beta} i\Omega_{\alpha\beta}\langle[\sigma_\alpha^\dagger\sigma_\beta, Q]\rangle + \sum_{\alpha,\beta}\Gamma_{\alpha\beta}\left\langle \sigma_\alpha^\dagger Q\sigma_\beta - \frac{1}{2}\{\sigma_\alpha^\dagger\sigma_\beta, Q\}\right\rangle, \tag{1}$$

where $\Omega_{\alpha\beta} = \Omega_{\alpha\beta}^{(\mathrm{rad})} + \Omega_{\alpha\beta}^{(\mathrm{T})}$ and

$$\Omega_{\alpha\beta}^{(\mathrm{rad})} = \Gamma G_{\alpha\beta}(k_0 r_{\alpha\beta}), \quad \Gamma_{\alpha\beta} = \Gamma F_{\alpha\beta}(k_0 r_{\alpha\beta}). \tag{2}$$

Here $\boldsymbol{r}_{\alpha\beta} = \boldsymbol{r}_\beta - \boldsymbol{r}_\alpha$, $\Gamma = |d_0|^2 k_0^3/(3\pi\varepsilon_0\varepsilon_\mathrm{r}\hbar)$ is the spontaneous emission (radiative recombination) rate for a single, isolated QD, $k_0 = nE/(\hbar c)$ is the average resonant wave vector in the dielectric medium with the refractive index n,

$$G_{\alpha\beta}(\xi) = \frac{3-9|\hat{d}_\parallel|^2}{4}\left(\frac{\sin\xi}{\xi^2}+\frac{\cos\xi}{\xi^3}\right) - \frac{3|\hat{d}_\perp|^2}{4}\frac{\cos\xi}{\xi},$$

$$F_{\alpha\beta}(\xi) = \frac{3-9|\hat{d}_\parallel|^2}{2}\left(\frac{\cos\xi}{\xi^2}-\frac{\sin\xi}{\xi^3}\right) + \frac{3|\hat{d}_\perp|^2}{2}\frac{\sin\xi}{\xi},$$

and $\hat{d}_\parallel$ and $\hat{d}_\perp$ are the components of $\hat{\boldsymbol{d}}_0 = \boldsymbol{d}_0/d_0$ parallel and perpendicular to $\hat{\boldsymbol{r}}_{\alpha\beta}$, respectively. The first expression in Eq. (2) describes the dipole couplings between the interband dipoles confined in the QDs (referred to as *Förster coupling* or *dispersion forces*). We assume that only heavy hole excitons are involved, so that $\boldsymbol{d}_0 = d_0(1, \pm i, 0)^\mathrm{T}/\sqrt{2}$, hence $|\hat{d}_\parallel|^2 = |\hat{d}_\perp|^2 = 1/2$ since the QDs are distributed in the xy plane. An admixture of light hole states would reduce the dipole coupling at short distances. However, a typical admixture of at most several percent[20] will only bring a small quantitative correction to our results and will not affect the qualitative conclusions of this Rapid Communication.

In order to find the evolution of the total number of excitons, $N_X = \sum_\alpha\langle\sigma_\alpha^\dagger\sigma_\alpha\rangle$, we use Eq. (1) to find the equations of motion for the quantities $x_{\alpha\beta} = \langle\sigma_\alpha^\dagger\sigma_\beta\rangle$. These quantities are dynamically coupled to higher order terms of the form $\langle\sigma_\alpha^\dagger\sigma_\beta^\dagger\sigma_\gamma\sigma_\delta\rangle$. In general, in an inhomogeneous ensemble, there are no constants of motion in the N-dot dynamics and the system is described by the 2^{2N} elements of the general density matrix, which is beyond the simulation capabilities already for several dots. Here, we will restrict the discussion to the low excitation case, with at most one exciton present in the QD ensemble (in this case, collectivity results from delocalization of this single excitation over many emitters jointly interacting with the field). Then, the normally ordered higher order terms vanish and one obtains a closed system of equations of motion in the form

$$\dot{x}_{\alpha\beta} = \frac{i}{\hbar}(\epsilon_\alpha - \epsilon_\beta)x_{\alpha\beta} + i\sum_\gamma(\Omega_{\gamma\alpha}x_{\gamma\beta} - \Omega_{\beta\gamma}x_{\alpha\gamma}) - \frac{1}{2}\sum_\gamma(\Gamma_{\gamma\alpha}x_{\gamma\beta} + \Gamma_{\beta\gamma}x_{\alpha\gamma}), \tag{3}$$

where it is assumed that $\Omega_{\gamma\gamma} = 0$.

The photon detection rate for a hypothetical ideal detector collecting from a solid angle $d\Omega$ around a point $\boldsymbol{R}$ at a large distance from the sample is proportional to the field correlation function,[21]

$$J(\hat{\boldsymbol{R}})d\Omega = \frac{2cR^2}{nE\varepsilon_0\varepsilon_\mathrm{r}}\langle\boldsymbol{D}^{(-)}(\boldsymbol{R},t)\cdot\boldsymbol{D}^{(+)}(\boldsymbol{R},t)\rangle d\Omega, \tag{4}$$

where $\boldsymbol{D}^{(\pm)}$ are the positive and negative frequency parts of the field. The field originates from the interband dipoles and can be related to the transition operators σ_α. In the Markov limit one finds[19]

$$J(\hat{\boldsymbol{R}}) = \frac{3\Gamma(1-|\hat{\boldsymbol{d}}_0\cdot\hat{\boldsymbol{R}}|)}{8\pi}\sum_{\alpha\beta}e^{ik\boldsymbol{r}_{\alpha\beta}\cdot\hat{\boldsymbol{R}}}\langle\sigma_\alpha^\dagger(t)\sigma_\beta(t)\rangle. \tag{5}$$

For a spectrally resolved detection, one has to replace the field operators in Eq. (4) by spectrally filtered operators $\tilde{\boldsymbol{D}}^{(\pm)}(t) = \int dt' f(t-t')\boldsymbol{D}^{(\pm)}(t')$, where $f(t)$ is the inverse Fourier transform of the filter function $f(\omega)$. This results in an analogous replacement for the σ_α,σ_β operators in Eq. (5). The two-time correlation functions are calculated by means of Eq. (1) and the quantum regression theorem,[21] starting from the equal time average known from the solution to Eq. (3). For a milli-electron-volt filter width, the relevant time differences are on the picosecond scale, much shorter than the exciton lifetime, so that the dissipative part of Eq. (1) is neglected.

To model the experimental situation,[8] we consider ensembles of quantum dots placed on square mesas. The spectral properties of the dots are modeled by a Gaussian distribution of their transition energies with the mean E and variance σ^2. The dots are located at random in the sample plane, with the restriction that the interdot distance cannot be smaller than 10 nm. The mesas are "cut out" from a larger sample, so that the number of dots fluctuates. At the initial time, the QD ensemble is supposed to be coherently excited into the delocalized state $|\psi(0)\rangle = (1/N)^{1/2}\sum_\alpha\sigma_\alpha^\dagger|0\rangle$, where $|0\rangle$ is the system ground state. For each set of parameters, the results are averaged over many repetitions with different, randomly generated QD distributions. In the simulations presented here, we assume $1/\Gamma = 390$ ps, $n = 2.6$, $E = 2.59$ eV, and the QD surface density $\nu = 10^{11}$ QDs/cm^2. For the SR coupling between the QDs we choose the simplest exponential model, $\Omega_{\alpha\beta}^{(\mathrm{T})} = V_0\exp[-r_{\alpha\beta}/r_0]$, with the amplitude $V_0 = 5$ meV and the spatial range $r_0 = 15$ nm, chosen to fit the experimental data.

The decay of the exciton population in a system of 30 QDs on the average with various degrees of inhomogeneity and coupled by interactions of different kinds is shown in Fig. 1. If only LR dipole couplings are included [Fig. 1(a)] the evolution

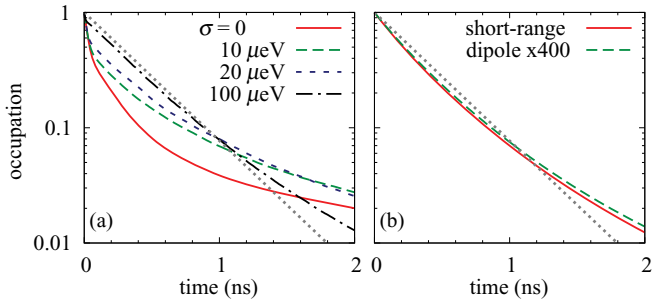

FIG. 1. (Color online) The time dependence of the exciton occupation for a QD ensemble on a 0.03 μm^2 mesa (30 QDs on the average). (a) With LR dipole couplings only. Solid (red) line: Homogeneous system. Dashed lines: Three different inhomogeneous systems with the standard deviations of the fundamental transition energies σ as indicated. (b) With additional SR couplings (solid red line) and with the dipole coupling artificially enhanced by a factor of 400 (dashed green line) for $\sigma = 28$ meV. In both figures, the dotted (gray) line shows the exponential decay with the rate Γ characteristic of a single dot. The results are averaged over 20 random QD distributions (the same for each case).

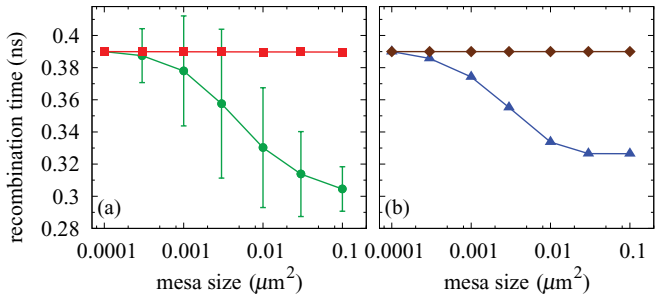

FIG. 2. (Color online) (a) Dependence of the average exciton life time on the "mesa" size used in the simulation for an ensemble of dipole-coupled dots (red squares) and SR-coupled dots (green circles). The error bars show formally the values of the standard deviation of the results obtained for various QD distributions; however, the actual distribution of results is strongly asymmetric with a tail towards shorter lifetimes and no results above the single dot exciton lifetime of 0.39 ns. (b) Average exciton lifetime as a function of the mesa size for two hypothetical QD ensembles: With dipole forces scaled up by a factor of 400 (blue triangles) and with SR couplings but emitting to separate reservoirs (brown diamonds). The results are averaged over a number of repetitions ranging from 10^4 for the small systems to 100 for the largest system studied.

of a system of identical dots is markedly nonexponential with a long stage of strongly accelerated emission (red solid line). However, this effect vanishes very quickly as soon as the inhomogeneity of the fundamental transition energies comes into play. As can be seen in Fig. 1 (black dashed-dotted line), already for $\sigma = 0.1$ meV, the decay almost exactly follows the exponential evolution of a single dot, with only a very short initial period of enhanced emission.

The results are completely different if a sufficiently strong SR coupling is taken into account [Fig. 1(b), red solid line]. Now, the exciton occupation decays with an enhanced rate even for the realistic value of $\sigma = 28$ meV.[8] Moreover, although the time dependence is not strictly exponential, the emission rate is much more constant over the two decades of occupation change shown.

Although one could expect that the LR dipole interaction should lead to qualitatively different dynamics than the SR coupling, this turns out not to be the case for the inhomogeneous QD system. As shown in Fig. 1(b) (green dashed line), the evolution of the exciton occupation in the hypothetical case of dipole couplings magnified by a factor of 400 is almost identical to that observed for SR couplings for the parameters of these couplings chosen here. The reason is that in any case the coupling is important for the spontaneous emission only when its magnitude is at least comparable to the energy mismatch between the emitters.[9] For a realistic, rather strongly inhomogeneous ensemble of QDs, only coupling between relatively close QDs is strong enough to have any impact on the system dynamics. Hence, the properties of the interactions on longer distances are irrelevant.

As pointed out in Ref. 8, a fingerprint of collective spontaneous emission is the dependence of the exciton decay rate on the number of emitters. In Fig. 2(a), we show the simulation results for this dependence in the case of a QD sample coupled only by the dipole forces (red squares) and in the presence of the SR coupling (green circles). As expected from the discussion presented above, there is no noticeable decrease of the exciton lifetime in the case of LR EM coupling. However, including the SR coupling leads to a considerable acceleration of the spontaneous emission to a degree comparable to that observed in the experiment.[8] A similar result is obtained in the hypothetical case of artificially magnified dipole couplings, as shown by blue triangles in Fig. 2(b).

Interestingly, for both kinds of couplings, the effect of a variation in the QD surface density is quite similar: We have checked that decreasing the density by a factor of 4 reduces the increase of the decay rate for a 0.03 μm^2 mesa to 23% and 25% of the original value for the SR and enhanced LR coupling models, respectively. Increasing the density by the same factor enhances this collective effect by similar factors of 3.3% and 2.6%, respectively. At the same time, even in a sample with increased density, the normal dipole coupling is still not sufficient to induce a noticeable enhancement of the spontaneous emission.

The results of simulations show that strong SR coupling between neighboring QDs is crucial for the enhancement of the spontaneous emission, while the LR dipole coupling encompassing the whole sample is irrelevant. It must be noted, however, that the collective nature of the interaction between the QDs and the EM field is still essential for the observed effect. As follows from Eq. (1), collective EM coupling affects the system dynamics in two ways: It not only mediates the dipole interactions described by the coupling constants $\Omega_{\alpha\beta}^{(\mathrm{rad})}$ but also leads to the appearance of interference terms $\Gamma_{\alpha\beta}$ for $\alpha \neq \beta$ in the dissipative part. These terms are absent in the hypothetical case of QDs emitting to separate reservoirs (but still coupled by interactions).[10] The brown diamonds in Fig. 2 show the exciton lifetimes for such an artificial system of QDs with additional SR interactions and coupled to separate reservoirs. Clearly, no enhancement of the spontaneous emission is observed.

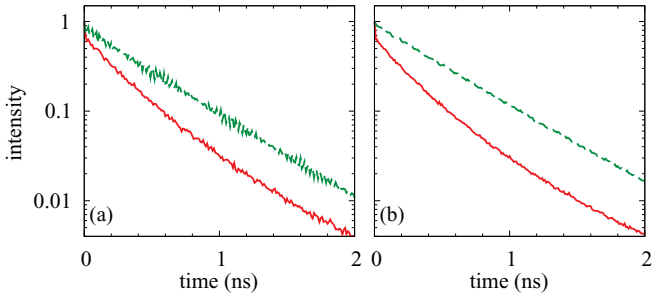

FIG. 3. (Color online) Time dependence of the luminescence intensity (normalized to the initial value) for detection at the photoluminescence maximum (solid red lines) and 40 meV away from the maximum (dashed green lines): (a) For a 0.01 μm^2 mesa; (b) for a 0.1 μm^2 mesa. The results are averaged over 100 different QD distributions.

Another experimental fingerprint of collective emission is the dependence of the luminescence decay time on the detection energy:[8] Since in the spectral range around the maximum of the ensemble photoluminescence (PL) more dots contribute to the emission, the collective effects in this energy sector are stronger and the decay is faster. Our simulations, shown in Fig. 3, show that also this feature is reproduced in a model with SR coupling. Here, we have simulated the measured signal from an ensemble with $\sigma = 28$ meV, using a Gaussian spectral filter $f(\omega) \sim \exp\{-(1/2)[(\omega - \omega_0)/\Delta\omega]^2\}$ with $\hbar\Delta\omega = 3$ meV, and $\hbar\omega_0$ at the PL maximum or 40 meV away, for two sizes of the simulated mesas. As can be seen, in both cases the PL decay at the PL maximum is indeed faster than in the tail of the inhomogeneous distribution.

Our simulations of spontaneous emission from ensembles of dipole- and SR-coupled QDs show that the emission rate in such a system can indeed be increased due to collective coupling of the emitters to the EM field. However, in view of relatively large inhomogeneity of the QD transition energies, sufficiently strong coupling between the dots is needed to stabilize the collective nature of the emission. For typical interdot separations, fundamental dipole interactions are too weak to play an important role in the emission kinetics. However, the presence of SR interactions, which may be due to some kind of electronic coupling between the dots in combination with Coulomb correlations, leads to enhanced emission in quantitative agreement with the experimental results.[8] While such a coupling is likely to exist in QD samples,[13] its exact microscopic nature is neither important for the emission dynamics nor can be inferred from it. In any case, the presence of collective effects in the emission means that QDs are not necessarily independent emitters, as usually assumed when modeling their optical properties. Whether the increase of emission rate at low excitation is a signature of the true superradiance that could result in a delayed outburst of radiation under strongly inverted initial conditions[22] remains an open question. Also the evolution of interdot coherence in the process of carrier trapping and relaxation, which would account for the experimentally observed differences between emission under resonant and nonresonant conditions, remains an interesting problem for further study. Finally, in the course of further research it may be interesting to clarify whether this kind of cooperative behavior can be responsible for radiative transfer of excitation between remote dots.[23]

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