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Monitoring single scattering events in single quantum dots

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Abstract

The quantum-jump approach is used for a theoretical description of resonance luminescence from a *single* semiconductor quantum dot in contact with its solid-state environment. For continuous excitation of the single-exciton groundstate the luminescence exhibits bright periods, where photons are spontaneously emitted from the exciton decay, which are interrupted by dark periods when one electron or hole suffers a spin flip. It is shown that continuously monitored resonance luminescence provides a very sensitive measure of such rare single scattering events in quantum dots. © 2001 Elsevier Science Ltd. All rights reserved.

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Semiconductor quantum dots (QDs) consist of a small island of lower-bandgap material embedded in a solid-state matrix of higher-bandgap material [1,2]. Proper choice of the material and dot parameters, thus can give rise to confinement of a few carrier states within this lower-bandgap region. Because of the resulting discrete atomic-like density-of-states, semiconductor QDs have been referred to as *artificial atoms*. When optical experiments are performed on large QD ensembles, all the available samples still suffer from the effects of inhomogeneity and dispersion in dot size. A major advancement in the field has come from different types of local optical experiments that allow the investigation of individual QDs: extremely narrow linewidths have been reported from single-dot spectroscopy (for a review, see Ref. [3]) and first time-resolved measurements indicate carrier lifetimes a few orders of magnitude larger than typically observed in semiconductors of higher dimension [4].

Theoretically, the system *dot interacting with environment* is conveniently described within a density-matrix description [5–8]. Adopting the approximation of Fermi's golden rule, at each instant of time the dot system is then

completely determined by its density matrix ρ (with the diagonal and off-diagonal elements providing information about state occupation and coherence, respectively), and environment interactions are accounted for by instantaneous scattering events. Because of its statistical nature, the elements of ρ have to be interpreted either as the ensemble average over a large number of identical dots, or, for ergodic systems, as the average over a sufficiently long measurement time. Apparently, when performing time-resolved measurements on single dots the situation is substantially different. Thus, the most interesting questions arise: does the interpretation of optical experiments of *single* dots require theoretical concepts beyond the usual density-matrix description for the carrier dynamics in semiconductor nanostructures [5–8], and can time-resolved single-dot measurements provide additional information as opposed to ensemble-dot measurements?

As we will show in the following, the answer to both the questions is: yes. It is worth noting that related questions first arose almost a decade ago, when it became possible to store single ions in a Paul trap and to continuously monitor their resonance fluorescence (see Ref. [9] for a review). In the seminal work of Dalibard et al. [10], the authors showed for a V-scheme, where the groundstate 0 of an atom is coupled to a short-lived state 1 and to a metastable state 2, that the fluorescence of the laser-driven system exhibits long

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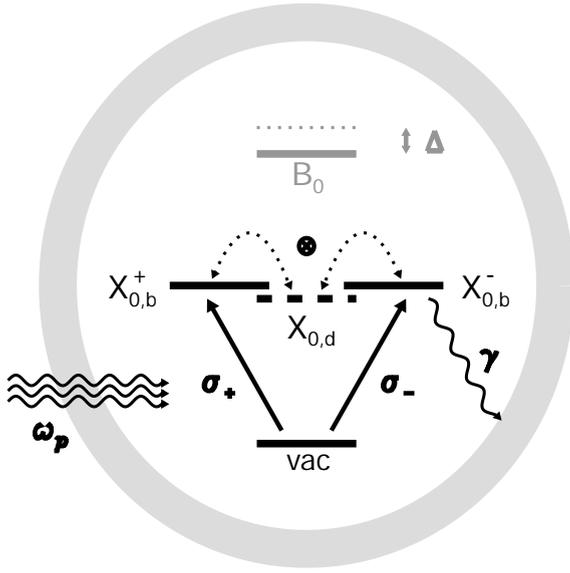


Fig. 1. Schematic illustration of the proposed experimental setup, where a single QD is surrounded by a 4π photon detector (with unit efficiency) which detects all spontaneously emitted photons γ . The linearly polarized pump pulse (ω_p) couples the vacuum state to the optically allowed groundstate excitons $X_{0,b}^\pm$; B_0 denotes the bi-exciton groundstate, whose energy is by an amount of Δ smaller than $2E_{X_{0,b}^\pm}$ due to correlation effects. Spin-flip processes (indicated by \otimes) couple the optically allowed states $X_{0,b}^\pm$ with the optically forbidden $X_{0,d}$ ones, which, because of the electron–hole exchange interaction, have a slightly smaller energy.

dark periods, associated to the excitation of the extremely weak $0 \leftrightarrow 2$ transition, which are followed by bright periods with many photon emissions from the decay of the short-lived state 1.

In this article, we propose to use continuous laser excitation and continuous monitoring of luminescence for the observation of *single* spin-flip processes in semiconductor QDs. Contrary to the proposal of Dalibard et al., within our scheme the rare scattering events originate from the coupling of the QD carriers to the elementary excitations of their solid-state environment. Thus, the dot serves as a *sensor* of its environment, which makes possible the observation of the otherwise almost inaccessible rare scattering events. Besides the most challenging prospect of measuring single scatterings in solid state,¹ the long-lived spin excitations in QDs have recently attracted strong interest [12–15] because of their potential utilization for quantum-information processing [16,17].

Fig. 1 sketches the proposed scheme: a single QD is located inside an ideal broadband counter with unit effi-

¹ Another striking example of single scatterings in semiconductor QDs is provided by the random-telegraph noise in photoluminescence due to electric-field fluctuations. For a semiclassical analysis, see Ref. [11].

ciency and coverage of all angles (grey circle).² Initially, all valence-band states of the dot are occupied and all conduction-band states are empty. When the pump laser ω_p is turned on, an electron is promoted from the valence to the conduction band, where the photogenerated electron and hole (i.e. the missing valence-band electron) have opposite spin orientations. Because of the coherence of the driving laser the system starts to undergo Rabi-type oscillations, i.e. the electron is transferred back and forth between the valence and conduction band. These oscillations are interrupted by spontaneous emissions of photons, γ , from the decay of electron–hole (exciton) states $X_{0,b}^\pm$, where after detection of γ , the system is reset back to the vacuum state, and the Rabi-type oscillations start again. After some time, however, due to environment coupling the spin of either the electron or hole is flipped (see \otimes in Fig. 1); since each QD state can be occupied at most by two carriers with *opposite* spin orientations, after such a spin flip the system becomes optically inactive (i.e. no further electron–hole pair can be excited by the pump laser). This can be also inferred from Fig. 2(a) which shows results of a simulation of resonance luminescence from a single QD (for details see further below): After turning on the pump pulse at time $t = 0$, photons are emitted by spontaneous decay of $X_{0,b}^\pm$ and are detected as single photon counts. These bright periods are then interrupted by dark periods with zero photon count, which indicate the occurrence of a spin-flip process. It finally requires a second spin flip to bring back the system to the bright sector, and to turn on luminescence again.

The theoretical framework accounting for the coherent laser excitation and the detection of single photon emissions is conveniently provided by the *quantum-jump approach*[9]. Here, the central idea is that the broad-band photon counting can be approximately described by a series of repeated *gedanken* measurements at times $t_n = n\Delta_t$ ($n = 0, 1, 2, \dots$) [18], where as observable the projector on the photon vacuum $\mathbb{P}_0 = |0_\gamma\rangle\langle 0_\gamma|_{\text{dot}}$ indicates whether a photon has been emitted or not. If at time t_0 the system is described by the density matrix ρ_0 , the density matrix at time t_n provided that no photons have been emitted in the interval $[t_0; t_n]$ is given by [9,18]:

$$\rho_{t_n}^{(0)} = \mathbb{P}_0 U(t_n, t_{n-1}) \dots \mathbb{P}_0 U(t_1, t_0) \rho_0 \times U^\dagger(t_1, t_0) \mathbb{P}_0 \dots U^\dagger(t_n, t_{n-1}) \mathbb{P}_0, \quad (1)$$

where $U(t_n, t_{n-1})$ is the time evolution operator from t_{n-1} to t_n , and the superscript on $\rho^{(0)}$ indicates the requirement of no-photon emission. The quantity $P_0(t) = \text{tr}(\rho^{(0)}(t))$ then gives the probability that, starting at time t_0 with ρ_0 , no photons are emitted up to time t ; apparently, $P_0(t)$ is a monotonically decreasing function since the probability of photon emission

² Experimentally, it is certainly impossible to achieve complete 4π -coverage. However, as discussed in Ref. [9], smaller detector efficiency does not cause changes of the qualitative behavior.

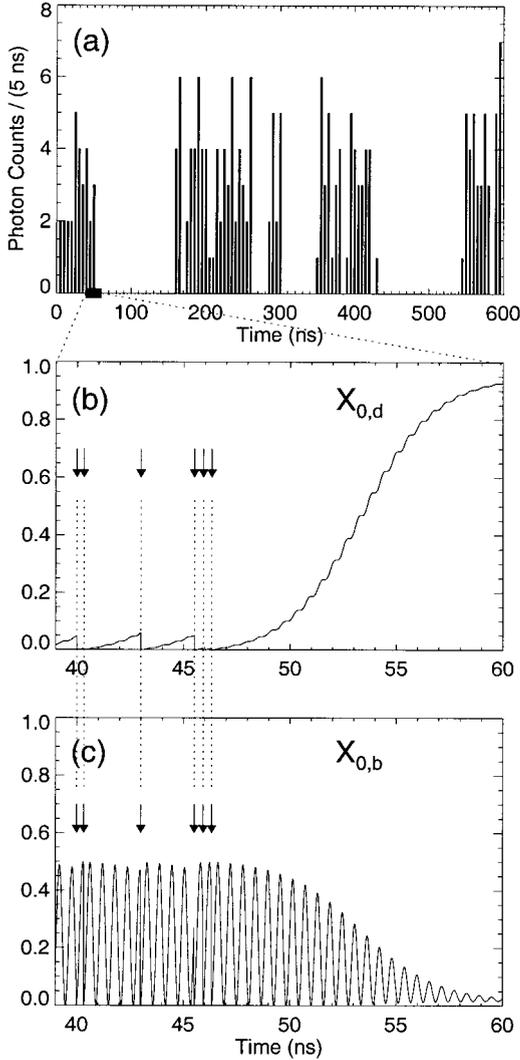


Fig. 2. Results of our Monte-Carlo simulations for resonance luminescence from a single QD: (a) histogram of photon emission (with bin width of 5 ns); (b,c) trajectory of $\rho_{xx}^{(0)}/\text{tr}(\rho)$ for $x = X_{0,d}, X_{0,b}^{\pm}$. The arrows indicate times of photon detection. Prototypical dot and laser parameters of $\Delta = 2$ meV and $\Omega_p^{\pm} = 10$ μ eV are used. For the energy splitting δ between $X_{0,b}^{\pm}$ and $X_{0,d}$ we assume $\delta \ll T$, thus obtaining the same scattering rates for $X_{0,b}^{\pm} \rightarrow X_{0,d}$ and $X_{0,d} \rightarrow X_{0,b}^{\pm}$; we use $(2\Gamma^s)^{-1} = 50$ ns and $(2\Gamma^{\gamma})^{-1} = 0.86$ ns for the radiative decay, which corresponds to the bulk-GaAs value.

will increase with time. If finally at time t_n a photon is detected, we know that the system has changed to the one-photon subspace $\rho^{(1)}$ (we assume that Δ_t is short enough and only *one* photon is emitted per interval). What the photon detector does, however, is to absorb the photon and to reset the density matrix to [18]:

$$|0_{\gamma}\rangle \text{tr}_{\gamma}(\mathbb{P}_1 U(t_n, t_{n-1}) \rho_{n-1}^{(0)} U^{\dagger}(t_n, t_{n-1}) \mathbb{P}_1) \langle 0_{\gamma} | \text{tr}(\rho), \quad (2)$$

with $\mathbb{P}_1 = 1 - \mathbb{P}_0$, and the denominator ensures that $\text{tr}(\rho) = 1$ after detection of γ .

In this article we apply the above scheme to the resonance luminescence of exciton states in a single QD. Quite generally, the driving laser cannot only excite single excitons but also multi-exciton states (e.g. biexcitons). Thus, we use x to denote generic multi-exciton states, with $N_x = 0, 1, 2, \dots$ the number of electron-hole pairs; the corresponding dot Hamiltonian is of the form $\sum_x E_x |x\rangle\langle x|$, with E_x representing the multi-exciton energies. The coupling to the pump laser with electric field $\mathcal{E}_p^{\sigma}(t) = \mathcal{E}_p^{\sigma} \cos(\omega_p t)$ is described within the usual rotating wave and dipole approximations, $-\frac{1}{2} \sum_{\sigma, xx'} \Omega_p^{\sigma} (e^{i\omega_p t} \mathcal{P}_{x'x}^{\sigma} + e^{-i\omega_p t} \mathcal{P}_{xx'}^{\sigma*}) |x'\rangle\langle x|$, where $\sigma = \pm 1$ denotes left- and right-hand circular polarization, respectively (we assume that, as in most semiconductors, electron-hole pairs with given spin orientation can be selectively created by circular polarized light); $\Omega_p^{\sigma} = \mu_0 \mathcal{E}_p^{\sigma}$ is the Rabi frequency with μ_0 the dipole element of the bulk semiconductor; $\mathcal{P}_{x'x}^{\sigma} = \int d\mathbf{r} \langle x' | \hat{\psi}_{h,-\sigma}^{\dagger}(\mathbf{r}) \hat{\psi}_{e,\sigma}(\mathbf{r}) | x \rangle$, with $\hat{\psi}_{e,\sigma}^{\dagger}$ ($\hat{\psi}_{h,\sigma}^{\dagger}$) creating an electron (hole) with spin orientation σ . The spontaneous radiative decay of dot states is accounted for by the Hamiltonian $\sum_{\mathbf{k}\sigma, xx'} i g_{\mathbf{k}}^{\gamma} (a_{\mathbf{k}\sigma}^{\dagger} \mathcal{P}_{x'x}^{\sigma} - a_{\mathbf{k}\sigma} \mathcal{P}_{xx'}^{\sigma*}) |x'\rangle\langle x|$, where $g_{\mathbf{k}}^{\gamma} = \mu_0 (2\pi\omega_{\mathbf{k}})^{\frac{1}{2}}$, $a_{\mathbf{k}\sigma}^{\dagger}$ creates a photon with wavevector \mathbf{k} and circular polarization σ , and $\omega_{\mathbf{k}} = ck/n$ is the photon energy (with c the speed of light and n the semiconductor refraction index).

Spin-flip scatterings in III-V semiconductors are conveniently described as a two-step process [14,20]: First, spin-orbit coupling introduces a small mixing of electron (hole) states with different spin orientations; secondly, environment coupling (e.g. to phonons) mediates transitions between states with different spin orientations. While in semiconductors of higher dimensionality all different types of elastic and quasi-elastic scatterings contribute to such spin-flip processes [20,21], in semiconductor QDs severe phase-space restrictions lead to a strong suppression, and first experimental results indicate extremely long spin coherence times [12–14]. As a genuine model accounting for spin flips in QDs we consider, within the spirit of Ref. [16], a Caldeira-Leggett-type model where a set of harmonic oscillators (assumed to be in thermal equilibrium) is coupled linearly to the system spins by $\sum_j g_j^s \mathbf{S} \cdot (\mathbf{b}_j + \mathbf{b}_j^{\dagger}) / \sqrt{2}$; here \mathbf{b}_j are bosonic operators whose free motion is governed by $\sum_j \omega_j \mathbf{b}_j^{\dagger} \cdot \mathbf{b}_j$, and g_j^s is the coupling constant to the carriers in the dot.

Next, we assume that the time interval Δ_t of successive *gedanken* experiments is short as compared to the time evolution of multi-exciton states due to the driving pump laser, but long as compared to ω_p^{-1} ; thus, in the evaluation of U of Eq. (1) we perform the Markov and adiabatic approximations. Using the interaction representation according to $\sum_x \Delta_x |x\rangle\langle x|$ (with $\Delta_x = E_x - N_x \cdot \omega_p$), we then find within second order perturbation theory for the time evolution of the conditional density matrix $\rho^{(0)}$ [18,22]:

$$\dot{\rho}^{(0)} = -i(h_0 \rho^{(0)} - \rho^{(0)} h_0^{\dagger}) + \hat{\mathbf{J}}^s \rho^{(0)}, \quad (3)$$

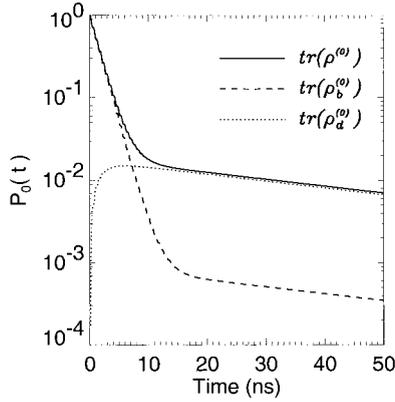


Fig. 3. Time-dependent probability $P_0(t)$ of no-photon emission for a coherently driven system which is in the groundstate at time 0 (same parameters as in Fig. 2). The solid line corresponds to $\text{tr}(\rho^{(0)})$, whereas the dotted and dashed lines show the contributions of $\langle X_{0,d} | \rho^{(0)} | X_{0,d} \rangle$ and remainder.

with $\hat{\rho}_n^{(0)} \equiv (\rho_n^{(0)} - \rho_{n-1}^{(0)})/\Delta_t$, the effective Hamiltonian $(h_0)_{xx'} = \delta_{xx'}\Delta_x - (1/2)\sum_{\sigma}\Omega_{\text{p}}^{\sigma}(\mathcal{P}_{xx'}^{\sigma} + \mathcal{P}_{x'x}^{\sigma*}) - i(\Gamma_{xx'}^{\gamma} + \Gamma_{xx'}^s)$, where $\Gamma^{\gamma,s}$ are out-scattering contributions due to photon and spin interactions, and $\hat{\mathbf{J}}^s$ accounts for spin-flip induced in-scatterings. More specifically, we obtain:

$$\Gamma_{x'x}^{\gamma} = \sum_{\sigma,\bar{x}} \alpha^{\gamma}(\omega_{x\bar{x}})\mathcal{P}_{\bar{x}x'}^{\sigma*}\mathcal{P}_{\bar{x}x}^{\sigma} \quad (4a)$$

$$\Gamma_{x'x}^s = \sum_{\sigma,\bar{x}} \alpha_{\text{T}}^s(\omega_{x\bar{x}})\mathcal{S}_{\bar{x}x'}^{\sigma*}\mathcal{S}_{\bar{x}x}^{\sigma} \quad (4b)$$

$$\mathbf{J}_{x'x',\bar{x}\bar{x}'}^{\gamma} = \sum_{\sigma} (\alpha^{\gamma}(\omega_{\bar{x}x}) + \alpha^{\gamma}(\omega_{\bar{x}'x'}))\mathcal{P}_{\bar{x}\bar{x}'}^{\sigma}\mathcal{P}_{x'x}^{\sigma*}, \quad (4c)$$

$$\mathbf{J}_{x'x',\bar{x}\bar{x}'}^s = \sum_{\sigma} (\alpha_{\text{T}}^s(\omega_{\bar{x}x}) + \alpha_{\text{T}}^s(\omega_{\bar{x}'x'}))\mathcal{S}_{\bar{x}\bar{x}'}^{\sigma}\mathcal{S}_{x'x}^{\sigma*}, \quad (4d)$$

with the spectral distribution function $\alpha^{\lambda}(\omega) = \pi\sum_{\ell}(g_{\ell}^{\lambda})^2\delta(\omega - \omega_{\ell}^{\lambda})$ ($\lambda = \gamma, s$ and $\ell = \mathbf{k}, j$), and the thermally broadened function $\alpha_{\text{T}}^{\lambda}(\omega) = \theta(\omega)n(\omega) + 1\alpha^{\lambda}(\omega) + \theta(-\omega)n(-\omega)\alpha^{\lambda}(-\omega)$, where $n(\omega)$ is the Bose–Einstein distribution at temperature T . Finally, $\mathcal{S}_{x'x} = \langle x'|S^{-\sigma}|x\rangle$ is the overlap between states x and x' when one electron or hole flips spin. (Note that because of our restriction to the zero-photon subspace there are no corresponding in-scatterings due to photon decay). Finally, whenever a photon is detected the density matrix is reset to $\rho^{(0)} \rightarrow \hat{\mathbf{J}}^{\gamma}\rho^{(0)}/\text{tr}(\cdot)$, with the reset matrix $\hat{\mathbf{J}}^{\gamma}$.

In our calculations we use prototypical dot parameters, which correspond to the model structure presented in Ref. [19] (see Fig. 2), and for the pump laser we assume linear polarization (i.e. $\mathcal{E}_{\text{p}}^{+} = \mathcal{E}_{\text{p}}^{-}$) and photon energy ω_{p} tuned to the groundstate-exciton energy (see Fig. 1). Because excited single-exciton states are energetically separated from $E_{X_{0,b}^{\pm}}$ by the order of the confinement energy (~ 10 – 50 meV for most dots [1,2]), for moderate Rabi frequencies they do not

couple to the driving laser and are therefore neglected in our analysis. As the only multi-exciton state, we consider the biexciton groundstate B_0 , which approximately consists of $X_{0,b}^{+} + X_{0,b}^{-}$ (with other multi-exciton states again energetically well separated). In Refs. [12,13] spin dephasing times $\tau_s \sim 3$ ns have been reported, which, however, have been dominated by inhomogenous broadening. Thus, as a representative value within a single dot we assume a value of 50 ns. Smaller values of τ_s would not change the qualitative behavior of our results, as long as they would be significantly longer than the radiative lifetime ~ 1 ns. Fig. 3 shows the probability $P_0(t)$ that upto time t no photon has been emitted. At times below ~ 10 ns the time evolution is governed by the optical decay of the laser-excited exciton states $X_{0,b}^{\pm}$; with increasing time, however, the probability of a spin-flip process increases and becomes dominant at later times. Because spin-forbidden transitions do not couple to the light, in this regime $P_0(t)$ decreases only very slowly and the decay is determined by additional spin-flip processes bringing back the system to the bright sector. We used these results to perform Monte-Carlo simulations of resonance luminescence from a single QD (Fig. 2) [9]: At time t_0 , we start with a density matrix ρ_0 ; the time of the next-photon emission is then determined by use of a random number $r \in [0, 1)$ from $r = 1 - P_0(t)$ (in the numerical solution of Eq. (3) we neglect the time discretization Δ_t); finally, after a photon count the density matrix is set back to $\hat{\mathbf{J}}^{\gamma}\rho^{(0)}/\text{tr}(\cdot)$, and the simulation starts again. Fig. 2(c) shows a single history of the occupation of $X_{0,b}^{\pm}$: Rabi-type oscillations are interrupted by sudden jumps (arrows), associated with the detection of emitted photons; correspondingly, the dark-exciton contributions (Fig. 2(b)) slowly increase in time, and are reset back to zero after photon detection (note the beginning of a dark period at later times in Fig. 2(b) and (c)). Finally, Fig. 2(a) shows the histogram of emitted photons: one clearly observes bright and dark sectors, whose lengths are determined by τ_s . Experimental observation of single spin flips requires coherent excitation of single QDs and efficient continuous broadband counting of photons on a timescale of nanoseconds.

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References

- [1] L. Jacak, P. Hawrylak, A. Wojs, Quantum Dots, Springer, Berlin, 1998.

- [2] D. Bimberg, M. Grundmann, N. Ledentsov, *Quantum Dot Heterostructures*, Wiley, New York, 1998.
- [3] A. Zrenner, *J. Chem. Phys.* 112 (2000) 7790.
- [4] N.H. Bonadeo, J. Erland, D. Gammon, D.S. Katzer, D. Park, D.G. Steel, *Science* 282 (1998) 1473.
- [5] F. Rossi, *Semicond. Sci. Technol.* 13 (1998) 147.
- [6] V.M. Axt, S. Mukamel, *Rev. Mod. Phys.* 70 (1998) 145.
- [7] J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures*, Springer, Berlin, 1996.
- [8] M.O. Scully, M.S. Zubairy, *Quantum Optics*, Cambridge University Press, Cambridge, 1997.
- [9] M.B. Plenio, P.L. Knight, *Rev. Mod. Phys.* 70 (1998) 101.
- [10] J. Dalibard, Y. Castin, K. Mølmer, *Phys. Rev. Lett.* 68 (1992) 580.
- [11] Al. L. Efros, M. Rosen, *Phys. Rev. Lett.* 78 (1997) 1110.
- [12] J.A. Gupta, D.D. Awschalom, X.A. Peng, A.P. Alivisatos, *Phys. Rev. B* 59 (1999) R10241.
- [13] J.A. Gupta, D.D. Awschalom, X.A. Peng, A.P. Alivisatos, *Phys. Rev. B* 60 (1999) 8394.
- [14] H. Kamada, H. Gotoh, H. Ando, J. Temmyo, T. Tamamura, *Phys. Rev. B* 60 (1999) 5791.
- [15] A.V. Khaetskii, Y.V. Nazarov, *Phys. Rev. B* 61 (2000) 12639.
- [16] D. Loss, D.P. DiVincenzo, *Phys. Rev. A* 57 (1998) 120.
- [17] A. Imamoglu, D.D. Awschalom, G. Burkard, D.P. DiVincenzo, D. Loss, M. Sherwin, A. Small, *Phys. Rev. Lett.* 83 (1999) 4204.
- [18] G.C. Hegerfeldt, *Phys. Rev. A* 47 (1993) 449.
- [19] F. Troiani, U. Hohenester, E. Molinari, *Phys. Rev. B* 62 (2000) R2263.
- [20] M.Z. Maialle, A. de Andrada e Silva, L.J. Sham, *Phys. Rev. B* 47 (1993) 15776.
- [21] M. Potemski, et al., *Solid State Commun.* 110 (1999) 163.
- [22] U. Hohenester, to be published.