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Few-particle effects in the optical spectra of semiconductor quantum dots

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Abstract

We analyze few-particle effects in the optical spectra of semiconductor quantum dots using a density-matrix approach that explicitly accounts for exciton–exciton, as well as exciton–carrier interactions. We give a consistent description of additional peaks appearing at high photoexcitation density, and predict that a strong polarization dependence should be characteristic of few-particle features.

Keywords: A. Semiconductors; A. Nanostructures; D. Optical properties; D. Electron–electron interaction

In semiconductor quantum dots (QDs), carriers are confined in all space directions on a length scale comparable to their de Broglie wavelength. Due to the strong quantum confinement and the resulting discrete energy spectra, QDs have been referred to as artificial atoms. Here Coulomb interactions are enhanced with respect to semiconductors of higher dimensionality, and can be tuned by tailoring confinement potentials. Therefore, QDs can be considered a unique laboratory for the investigation of few-particle effects. Indeed, clear evidence of these effects was detected by transport experiments in the addition spectra of different classes of QD nanostructures [1,2]. Very recently, fingerprints of such few-particle interactions have been observed also in the nonlinear photoluminescence spectra of single QDs. Additional emission peaks appear with increasing excitation density, which then supersede the original lines at even higher densities [3–6]. Besides its fundamental interest, a detailed understanding of these strong optical nonlinearities could help to manipulate correlations between few particles in a controlled manner; an issue of crucial importance in the future implementation of coherent-control techniques [7,8], or quantum computation in QD systems.

From a theoretical point of view, the optical response of various semiconductor structures can be successfully explained by analyzing the propagation of a single photogenerated electron–hole pair in the mean-field of all other carriers. This task is usually accomplished by the semiconductor Bloch equations (SBE), which describe the nonequilibrium carrier dynamics in terms of distribution functions and interband polarizations, the latter quantities accounting for the macroscopic phase coherence of electron–hole pairs introduced by the coherent laser field [9–12]. However, it is clear that the usual mean-field approximation is *not* adequate to account for the above experimental findings in quantum dots, as will be discussed below.

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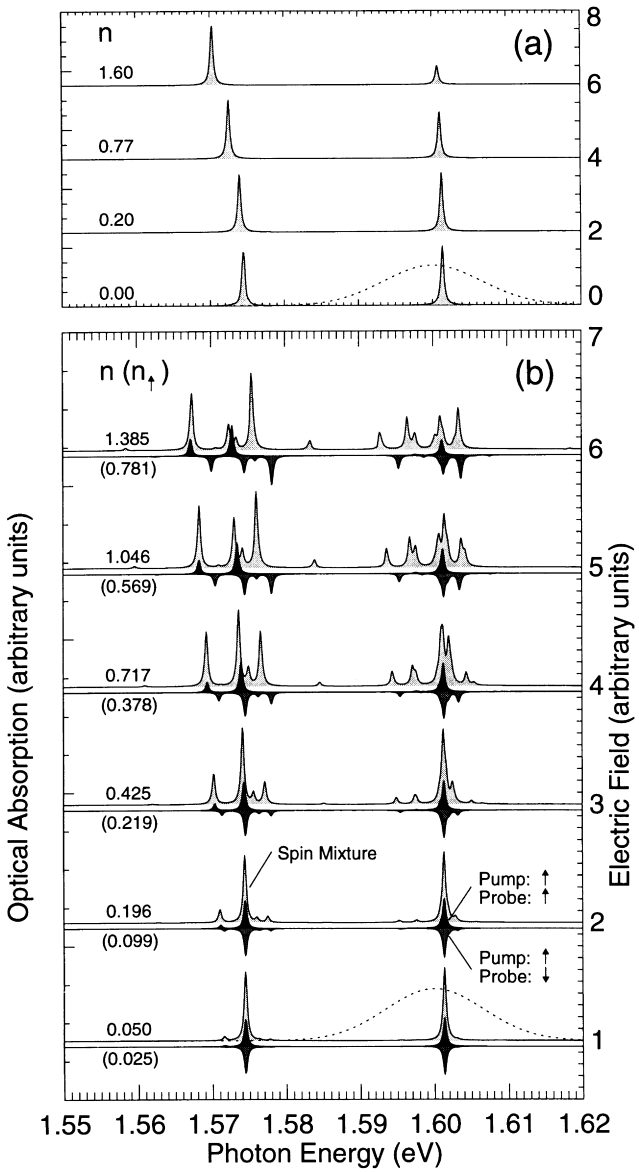

Fig. 1. Optical spectra of QD for different excitation densities, *n*, as obtained from (a) the mean-field SBE approach and (b) from calculations including two-particle correlations. The dephasing rate is 0.5 meV and the photon energy corresponds to the energetic separation between the first excited electron and hole states (dashed line: photoexciting laser pulse). The spectra corresponding to different intensities of the pumping laser are offset for clarity, and the mean number of electrons, *n*, is shown on the left-hand side. In (b), we also show below each curve the corresponding polarized spectra (curves for opposite probing polarization are displayed with opposite sign).

To solve this problem, in this paper we analyze the optical properties of semiconductor QDs within a density-matrix approach at the level of two-particle correlations. Here, the carrier dynamics is described in terms of the one-particle density matrix, ρ, and of two-particle correlations, c. Indeed, from the analysis of semiconductor systems of higher dimensionality it is well known that: (1) in the linear optical response, ρ properly accounts for excitonic effects [9,12]; (2) in the nonlinear optical response higher-order

correlations are required for the description of exciton–exciton and exciton–carrier interactions (e.g. biexcitons and charged excitons) [13,14].

If we describe the problems in terms of the Fermionic field operators $\psi_n^\dagger$, that either create an electron in the conduction ($n = c$) or valence band ($n = v$), we can write $\rho_{nn'} = \langle \psi_{n'}^\dagger \psi_n \rangle_t$, where the brackets denote the statistical ensemble average at time t. The matrices ρ_{cc} and ρ_{vv} contain the distribution functions together with the intraband polarizations; ρ_{cv} accounts for the interband polarizations, which play a central role in the description of the coherent optical response of semiconductors. Likewise, c is given by the expectation value of four Fermionic field operators ψ_n; according to the different combinations of ψ_c and ψ_v, the two-particle correlations then account for the various exciton–exciton, exciton–carrier and carrier–carrier correlations. The time evolution of ρ and c follow from the Liouville–von Neumann equation [15]:

$$\hbar\dot{\rho} = -i[h_o + h_{op}(t) + \bar{V}[\rho], \rho] - iF_2^{(1)}[c] + \hbar\dot{\rho}\,|_{\text{env}} \tag{1}$$

$$\hbar\dot{c} = -i[h_o + h_{op}(t) + \bar{V}[\rho], c] - iF_2^{(2)}[\rho, c] - iF_3^{(2)}[\rho, c] + \hbar\dot{c}\,|_{\text{env}}. \tag{2}$$

Here, h_o accounts for the single-particle states of carriers confined in the QD; $h_{op}(t)$ accounts for the interaction with an external light field; $\bar{V}[\rho]$ is the Hartree–Fock potential that describes the mutual Coulomb interactions among photoexcited electrons and holes on a mean-field level; $\dot{\rho}|_{\text{env}}$ and $\dot{c}|_{\text{env}}$ account for interaction processes with the environment of the QD (e.g. phonons, electrons in the wetting), which in this paper are treated within the relaxation-time approximation; finally, the different Fs account for genuine Coulombic few-particle effects. More specifically, $F_2^{(1)}$ describes the coupling between ρ and c; $F_2^{(2)}$ describes interaction processes between two carriers (beyond mean-field); and $F_3^{(2)}$ describes interactions among the three particles. Because of our restriction of using only ρ and c (i.e. neglecting three-particle correlations), the latter processes are treated within the approximation of two carriers moving in the mean field of the rest. Given the statistical nature of our approach, interactions with the environment of the QD can be incorporated in a natural manner—in contrast to direct-diagonalization techniques [16]. Such an environment coupling has been reported to become of crucial importance under high photoexcitation [3–6].

In our calculations for QDs, the single-particle properties are derived by solving the single-particle Schrödinger equation within the envelope-function and effective-mass approximations, by means of a plane-wave expansion with periodic boundary conditions [10,11]. External electric and/or magnetic fields, as well as arbitrary confinement potential profiles, can be easily treated within this scheme. The nonlinear optical response of few-particle QDs is analyzed in the time domain in terms of a pump–probe set-up: first, electron–hole pairs are generated by a short pump pulse, where the density and shape of photogenerated carrier distributions is tuned by the strength of the electric field and the photon energy of the laser (spectral shape according to the dashed line in Fig. 1); when all phase memory from the first pulse has been damped out by QD–environment interactions[†], the response of the previously photoexcited system is investigated by a very weak and short probe pulse (10 fs, for spectral width); the nonlinear absorption spectra are then directly computed by Fourier-transforming the induced polarization of the probe pulse [10,11].

Fig. 1 shows the optical absorption spectra for a prototype QD (parabolic confinement potential in the (x,y)-plane and box-like along z)[‡], as obtained from the conventional mean-field SBE approach (Fig. 1(a)) from the solution of Eqs. (1) and (2), which include the one-particle density matrix ρ together with the two-particle correlation functions c (Fig. 1(b)). Let us first consider Fig. 1(b) and concentrate on the calculations performed for a spin mixture, i.e. assuming that the light-field creates with equal probability electron–hole pairs with both spin-orientations. With increasing excitation density, additional

[†] In our calculations we use a dephasing rate of 0.5 meV, and the time delay between the pump and probe pulse is 10 ps.

[‡] In our cylindrical QD, the confinement energies due to the in-plane parabolic potential are $\omega_0^{(e)} = 20$ meV for electrons, and $\omega_0^{(h)} = 3.5$ meV for holes; with this choice, electron and hole wavefunctions have the same lateral extension. The quantum-well confinement along z is such that the intersubband splittings are much larger than $\omega_0^{(e,h)}$. Material parameters for GaAs are used.

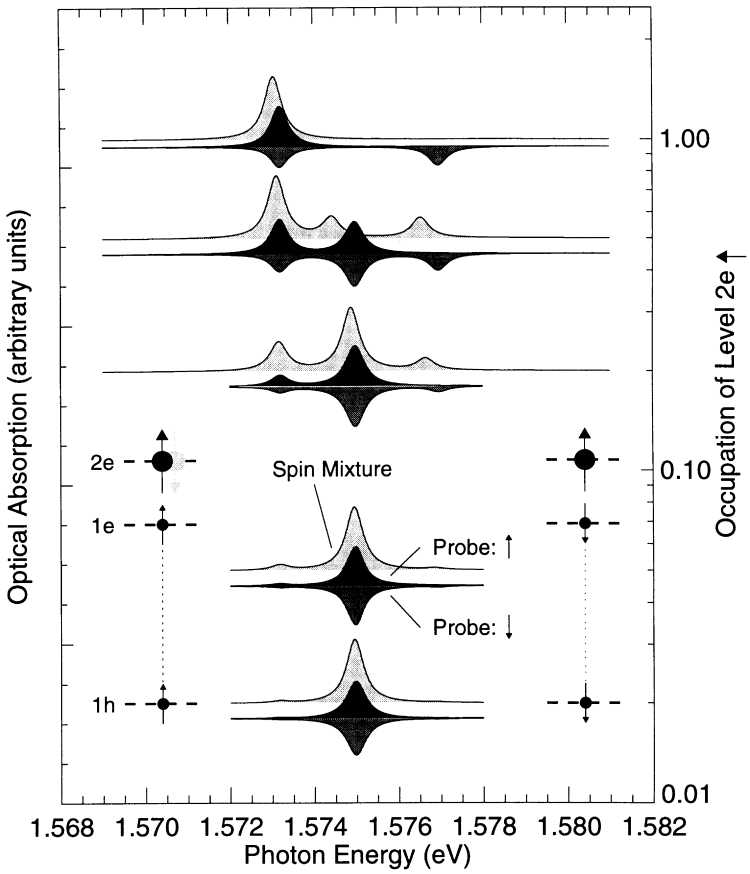

Fig. 2. Optical spectra of a simplified three-level QD system as described in the text, obtained within our approach including two-particle correlations. Notations as in Fig. 1.

lines appear at both higher and lower photon energies, which then supersede the original peaks when the density is increased further[†]. In contrast, the mean-field absorption spectra of Fig. 1(a) shows a small red-shift of the ground-state transition and a bleaching of the second peak (~1.60 eV) but, obviously, the appearance of additional emission peaks with increasing excitation density cannot be reproduced.

The absence of additional structures from the mean-field SBE spectra clearly indicates that they are genuine few-particle effects. To gain further insight into the mechanisms responsible for the appearance of additional peaks, in the following, we focus on a simplified model system. Fig. 2 shows the density dependence of the optical spectra for a three-level system consisting of two electron states and a single hole state. Because transitions between the hole state (1*h*) and the excited electron state (2*e*) are optically forbidden (this selection rule is exact due to our assumption of the same extensions for electron and hole wavefunctions), we initially prepared the excited electron state, 2*e*, with a given electron distribution. To further simplify the problem, we next consider the case that the level 2*e* is populated with an electron with spin up, and the ground-state transition is either probed with equal or opposite spin orientation (Fig. 2, panels on the left- and right-hand side, respectively). At this level of simplification, it is easy to grasp the

[†] Note that the absorption spectra in Fig. 1 are normalized to their maximum. By analyzing the absolute peak heights, we find that the intensity of the additional peaks first increases with the photogenerated carrier density, and then quench when the excitation density is further increased.

form of the many-particle wavefunctions from basic considerations: for parallel orientation of the electron spins, the spin-part of the electron wavefunction is symmetric and, thus, the remaining part antisymmetric; for antiparallel spin orientation, on the other hand, there exist two different possible configurations with either symmetric or antisymmetric spin part. The corresponding energies then consist of the single-particle energies reduced by the potential energy between electron 1*e* and hole 1*h* (direct term). Moreover, the exchange interaction results for parallel spin orientation in an additional decrease, and for antiparallel spin orientation in a symmetric energy splitting. In view of these considerations, we trace back the appearance of additional peaks in the spin-selective spectra of Fig. 2 to the exchange interaction (note that for clarity, the absorption spectra for spin-opposite probing are shown with opposite sign in the figure). The coexistence of the ground-state transition (1.575 eV) and the additional peaks, originating from few-particle interactions, is due to our statistical approach, which averages over the two possible configurations of empty or filled level 2*e*, respectively.

From the analysis of this simplified three-level system, we are finally led to conclusions that hold also for the more complicated case of realistic QDs. First, since the exchange interaction plays a prominent role in the appearance of additional peaks in the optical spectra, a strong spin dependence is expected also in this case. Indeed, Fig. 1 shows the nonlinear optical spectra for a spin-selective pump–probe set-up: electron–hole pairs with a given spin orientation are generated by the first pump pulse and the response of the system is then probed by carriers with either equal or opposite spin orientation. A clear difference between the two configurations is observed. As in most semiconductors, electron–hole pairs with a given spin orientation can be selectively created by photons with a well-defined circular polarization, we propose to study in future experiments the polarization dependence of the nonlinear optical spectra.

Our second conclusion concerns the interpretation of optical spectra in terms of excitons and exciton–exciton or exciton–carrier complexes. Because of the strongly reduced phase space, the QD quasiparticle spectrum undergoes a number of distinct transitions with increasing charging. Although features of configurations with different numbers of electron–hole pairs are always present in the optical spectra due to configuration averages, at each instant in time, the QD contains a well-defined number of photogenerated carriers, and quasiparticles can only be redistributed among the available states. Therefore, exciton complexes cannot break up into single excitons since single-exciton excitations do not exist in QDs populated with more than one electron–hole pair. Thus, quasiparticles in strongly confined electron–hole systems are expected to behave in a distinctly different way (e.g. in their temperature or density dependence) as compared to higher-dimensional systems (e.g. biexcitons or charged excitons).

We finally discuss the experimental results that have recently become available from luminescence spectroscopy of single QDs as a function of photoexcitation intensity. We shall not attempt a quantitative comparison with our data, due to the differences that are expected between luminescence and absorption. Nevertheless, from Fig. 1 one can immediately observe that our calculations reproduce very well the main experimental trends concerning the appearance of additional lines, as well as the dependence of their energy and amplitude on photoexcitation intensity [3]. The results of our calculations thus further support the conclusion that interactions of few carriers within a single QD can give rise to a number of additional peaks in the optical spectra [3–6].

In conclusion, we have studied the nonequilibrium carrier dynamics of semiconductor QDs by the use of a density-matrix approach at the level of two-particle correlations, and we have analyzed few-particle effects in the nonlinear optical absorption spectra. We have shown that exchange interactions play an important role in the appearance of additional lines with increasing pumping density, and predict a strong polarization dependence for these features.

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