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Excitonic and biexcitonic effects in the coherent optical response of semiconductor quantum dots

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Abstract

We analyze few-particle effects in the coherent optical spectra of semiconductor quantum dots using a density-matrix approach that explicitly accounts for exciton-exciton interactions. A consistent description of additional peaks appearing at high photoexcitation density is given. \bigcirc 1999 Elsevier Science B.V. All rights reserved.

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In semiconductor quantum dots (QDs) photogenerated electron-hole pairs remain bound because of the strong three-dimensional quantum confinement. Thus, in the optical spectra of QDs a strong enhancement of excitonic effects is expected as compared to semiconductor systems of higher dimensionality, where carrier complexes can break up into spatially separated constituents. Indeed, clear experimental evidence of pronounced Coulombic few-particle effects was recently reported in the nonlinear optical spectra of single QDs [1–4]: Additional emission peaks appear with increasing excitation density, which then supersede the original lines when the density is further increased. Using a density-matrix approach at the level of two-particle correlations, in this paper we present a theoretical study of the excitonic properties of semiconductor QDs. We analyze single-exciton effects in the linear optical spectra and compute the nonlinear optical response by use of a time dependent pump probe setup. Our results allow us to discuss the nature of additional peaks appearing in the nonlinear optical spectra.

Within a density-matrix approach, at each instant of time the carrier system is characterized by its one-particle density matrix and its higher-order correlation functions. The time evolution of these dynamic variables is obtained from the Liovillevon Neumann equation, which couples through the Coulomb potential all higher-order correlations. Thus, as a central approximation of any densitymatrix approach one has – based on physical intuition – to truncate this hierarchy of equations. A particularly transparent truncation scheme

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follows for the coherent optical response, i.e., the optical response in absence of dephasing processes [5,6]. Classifying the coherent optical response in powers of the electric field of the laser light, it turns out that: (1) the linear optical response is described by the interband polarization p, which accounts for macroscopic phase coherence of electron-hole pairs introduced by the coherent laser field; (2) for the third-order optical response only one additional quantity b, which accounts for biexcitonic effects, is required [5–7].

In our calculations for the optical properties of QDs, the excitonic energies E_x and wave functions Ψ^x , which describe one interacting electron-hole pair confined in the QD, are obtained from the solutions of the excitonic eigenvalue problem [8–10].¹ Expanding the interband polarizations *p* and the biexcitonic transitions *b* in terms of Ψ^x , we obtain [5–7]

$$\begin{split} i\dot{p}_{x} &= E_{x}p_{x} + \sum_{x',\bar{x},\bar{x}'} \bar{V}_{xx',\bar{x}\bar{x}'} p_{x'}^{*} (p_{\bar{x}}p_{\bar{x}'} + b_{\bar{x}\bar{x}'}) \\ &- \mathscr{E} \bigg(M_{x} - \sum_{\bar{x}\bar{x}'} \bar{M}_{x,\bar{x}\bar{x}'} p_{\bar{x}}^{*} p_{\bar{x}'} \bigg), \end{split}$$
(1a)

$$i\dot{b}_{xx'} = (E_x + E_{x'})b_{xx'} + \sum_{\bar{x},\bar{x}'} \bar{V}_{xx',\bar{x}\bar{x}'}(p_{\bar{x}}p_{\bar{x}'} + b_{\bar{x}\bar{x}'}),$$
(1b)

where \overline{V} is the exciton-exciton Coulomb matrix elements, \mathscr{E} the electric field of the laser light (treated within the rotating-wave approximation), and M and \overline{M} are the optical matrix elements (treated within the dipole approximation). The different contributions to Eq. (1) can be interpreted as follows: First, the excitonic Coulomb interactions in Eq. (1a) consist of a mean-field contribution $\propto p^3$, and of a coupling between p and b; as will be shown further below, this coupling is responsible for the appearance of additional peaks in the nonlinear optical spectra. The coupling be-



Fig. 1. (a) Linear optical absorption spectra for QD; solid (dotted) line: Coulomb interaction included (neglected). (b) Nonlinear optical absorption spectra for the spin-parallel pump-probe setup as described in the text; the pump pulse creates excitons at the spectral position X_0 ; solid line: solutions computed from Eq. (1); dotted line: mean-field approximation; (c) Same as (b) but for spin-antiparallel pump-probe setup. Photon energy zero is given by the energy of the ground state exciton X_0 .

tween p and the light field is described by M where $\overline{M}p^*p$ accounts for phase-filling corrections due to photogenerated electron-hole pairs. In Eq. (1b) the term $\overline{V}p^2$ acts as a source term which describes how biexcitons build up in the presence of photogenerated carriers. Finally, $\overline{V}b$ couples the different biexcitonic transitions and leads to a renormalization of the biexcitonic spectrum (i.e., "bound" biexcitons vs. scattering states).

In our calculations for QDs, we consider a prototype confinement potential which is parabolic in the (x, y)-plane and box-like along z. The confinement energies due to the in-plane parabolic potential are $\omega_0^{\rm e} = 20$ meV for electrons and $\omega_0^{\rm h} = 3.5$ meV for holes (with this choice, electron and hole wave functions have the same lateral extension; we use material parameters for GaAs), and the quantum-well confinement along z is such that the intersubband splittings are much larger than $\omega_0^{\rm e,h}$. Fig. 1(a) shows the linear absorption spectrum. An inspection of the differences between the solid and dashed line indicates two pronounced excitonic

¹The single-particle states, which are needed for the calculations of the excitonic properties, are derived by numerically solving the three-dimensional single-particle Schrödinger equation within the envelope-function and effective-mass approximations [7].

effects in the optical spectra of QDs: First, an almost rigid redshift of the optical spectrum due to the attractive electron-hole interaction. Second, the appearance of additional peaks (X_0^*) , caused by the coupling of an optically allowed transition with an optically forbidden one.

The nonlinear coherent optical response is analyzed in the time domain in terms of a spin selective pump-probe setup: First, electron-hole pairs with a given spin orientation are generated by a strong pump pulse, where the density and shape of the photogenerated carrier distribution is tuned by the strength of the electric field and the photon energy of the laser. The response of the previously photoexcited system is then investigated by a very weak and short probe pule (10 fs, for spectral broadness); we directly compute the nonlinear optical response by Fourier-transforming the induced polarization of the probe pulse [8,9].²

Fig. 1(b) shows the nonlinear absorption spectra as computed from the solutions of Eq. (1) for a pump pulse which creates excitons X_0 with spin orientation σ_+ (with an exciton density $N_{X_0} \sim 0.3$), and for a probe pulse with spin orientation σ_{+} [σ_{-} in Fig. 1(c)]. Because of our neglect of dephasing processes, the nonlinear optical response for spinparallel pumping and probing strongly depends on the phase relation between the two pulses, as can be inferred in Fig. 1(b) from the resonance behavior at the spectral position X_0 . A comparison of the nonlinear optical spectra with the excitonic absorption spectrum of Fig. 1(a) shows that additional peaks $(e.g., B_0)$ appear when the QD is initially populated with photogenerated carriers. In contrast, the mean-field absorption spectra of Fig. 1 [dotted lines in Figs. 1(b) and (c)], which are obtained from Eq. (1a) by neglecting the coupling between p and b, show a small redshift of the exciton X_1 but, obviously, no appearance of additional emission peaks.

The absence of additional structures from the mean-field spectra clearly indicates that they are genuine few-particle effects. Recently, we have shown that with increasing power of the pump pulse the heights of these additional peaks increase, whereas the excitonic peaks (X_0, X_0^*, X_1) are quenched [11]. In this paper, we analyze the nature of these few-particle peaks in terms of excitonic interaction processes. We first note that within our present framework of Eq. (1) the term $\overline{V}b$ is responsible for the renormalization of the biexcitonic spectrum and, thus, for the appearance of additional peaks. A closer analysis of the exciton-exciton Coulomb interaction $\overline{V}_{xx',\bar{x}\bar{x}'}$ reveals that: (1) For excitons with opposite spin orientation, these matrix elements account for a classical dipoledipole coupling between two excitons, with the dipole moments corresponding to the excitonic transitions from x to x' and from \bar{x} to \bar{x}' (note that in ODs - because of the strong quantum confinement - all excitonic wavefunctions spatially strongly overlap). (2) For excitons with parallel spin orientation, in addition a proper antisymmetrization with respect to the electron (hole) coordinates has been performed. In general, due to the dipole character of the exciton-exciton coupling, the biexcitonic energy shift strongly depends on the polarizability of excitons, i.e., the dipole moment of excitons induced by another dipole. Roughly speaking, we expect a smaller polarizability for the strongly confined groundstate excitons than for the excited excitonic states, which are spatially more extended. Indeed, in Fig. 1(c) the binding energy of the biexcitonic groundstate B_0 , which – to a good approximation – consists of two excitons X_0 with opposite spin orientation, is smaller than the energy shifts of the biexcitonic resonances B_1 and B_2 .

We finally discuss the experimental results that have recently become available from luminescence spectroscopy of single QDs as a function of photoexcitation intensity. Although we shall not attempt a quantitative comparison with our data, from Fig. 1 one immediately observes that our calculations reproduce very well the main experimental trends concerning the appearance of additional lines, as well as their spectral positions. 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 [1–4]. We finally note that recently Bonadeo et al. [12] have presented results of coherent nonlinear optical spectroscopy

 $^{^{2}}$ In the calculation of the Fourier transform of the induced polarization we add a small dephasing rate of 0.2 meV.

measurements of single QDs. Although in their work the authors have been mainly concerned with the dephasing of single excitons, it is possible that some of the results could be also interpreted in terms of biexcitons (see, e.g., peak ω_{γ} in Fig. 1(b) of Ref. [12]).

In conclusion, we have presented a density-matrix approach for the description of photoexcited semiconductor QDs. We have shown that few-particle interactions play an important role in the appearance of additional lines with increasing pumping density, and we have analyzed their nature within the framework of biexcitonic interaction processes.

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