Continuous Absorption Background and Decoherence in Quantum Dots

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We consider theoretically the role of crossed transitions on the interband optical properties of quantum dots. These transitions, which involve one bound state and one delocalized state, are inherent to the joint nature of the valence-to-conduction density of states in quantum dots. We show that they play a crucial role both on the interband absorption and on the broadening of the quantum dot lines.

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Semiconductor quantum dots are nanometric embeddings of a semiconductor in a similar host (such as, e.g., InAs dots on GaAs). These islands (typically 10 nm in size), which float on a thin layer of dot material (the wetting layer), correspond to attractive regions for both the electrons and the holes. Single carriers display correspondingly an atomiclike energy spectrum, characterized by a discrete sequence of low lying bound levels, followed by a continuum of spatially delocalized states associated either to the InAs wetting layer or to the GaAs barrier. Owing primarily to these spectral characteristics, semiconductor quantum dots have been advocated as potential candidates for the implementation of solid-state nanodevices [1]. In fact, elementary q-bit processing asks for simple systems, with a few energy levels well isolated spectrally and insensitive to external perturbations, in order to minimize decoherence effects. In a quantum dot, the role of environment was expected to be largely inhibited, owing to the localized nature of its eigenstates. However, this simple picture fails for actual dots: we have already shown that electrons, holes, and electron-hole pairs in *bound* dot orbits are nevertheless coupled to phonons, which involve the motion of the whole crystal lattice [2]. Thus, because of the intrinsic crystalline nature of actual samples, spatial localization does not prevent bound carriers from the influence of the dot environment, and couplings to phonons result in an intrinsic broadening of the dot eigenstates. In this Letter, we discuss another source of *intrinsic* broadening for the dot eigenstates, which reveals a different way the dot environment affects the bound dot levels. It arises in the situation (much exploited experimentally) of interband optical excitation, when electron-hole pairs are photoinjected in the dots, and is related to the existence of *crossed transitions* involving one bound state and one state delocalized near the dot. We show in the following that these transitions are inherent to the joint nature of the valence-to-conduction density of states in quantum dots and play a crucial role both on the interband absorption and on the broadening of the excited quantum dot lines.

The fluctuations (in size, composition, etc.) within an ensemble of quantum dots impose stringent limitations to the study of the intrinsic properties of a single dot. In this way, interband optical experiments performed on a collection of dots are characterized by inhomogeneously broadened lines. This has motivated the use of local probe techniques, which provide both spatial and spectral resolutions. These powerful filtering techniques have allowed the study of a few (ideally one) dots. Micro-optical spectra have shown series of narrow lines [3–6]. Surprisingly, optical excitation experiments done on single dots have also demonstrated the existence of an absorption "background," rising smoothly up to the edge of the wetting layer (WL) [3–5]. In their pioneering article, Toda *et al.* broadly suggest that it might be related to a kind of transition between zero and three-dimensional density of states [3]. Kammerer *et al.*'s experiments have shown that this continuum background is somehow related to the WL [5]. These authors tentatively attributed it to the WL tail, by arguing that the WL is by far not a flat layer, but most probably presents many local defects. The difficulty with this explanation is the fact that the continuous feature experimentally extends downwards very deeply in energy, much more than one can reasonably associate to fluctuations of the WL thickness near the dot. We provide in this Letter a theoretical interpretation of the observations. We show that quantum dots present an *intrinsic* deep (extending far below the WL edge) continuous density of electron-hole states, and that these states provide a natural explanation of the continuous background measured in excitation experiments.

We calculate the interband optical absorption of a single InAs/GaAs dot. The dots are approximated by truncated cones (basis radius R, top radius r, height h) floating on a WL of thickness d. We use the one-band effective mass model with position-independent conduction (isotropic mass $m_c = 0.067m_0$) and heavy-hole (anisotropic mass $m_{hz} = 0.337m_0$ and $m_{h\perp} = 0.112m_0$) masses. Because of the cylindrical symmetry, the envelope eigenstates possess well defined projections of the angular momentum along the growth (z) axis: $l = 0$ for S-like states, $l = \pm 1$ for P-like states, etc. The dot potentials are approximated by steplike potentials: $V_0 = 0$ in the InAs regions and $V_0 = V_{0e}$ and V_{0h} in the GaAs regions for the electrons and heavy holes, respectively (see below). The dot is placed at the center of a large

cylindrical box and we use a finite-difference method to obtain the eigenstates. The solutions for the electrons $[e_M, \phi_M(\mathbf{r})]$ and heavy holes $[h_N, \chi_N(\mathbf{r})]$ are then used to evaluate the band-to-band optical absorption probability:

$$\alpha(\omega) \propto \sum_{M,N} |\langle \phi_M(\mathbf{r}) \mid \chi_N(\mathbf{r}) \rangle|^2 \delta(\hbar\omega - e_M - h_N), \quad (1)$$

where $M = (m_e, l_e)$ represents the m_eth electron state of symmetry l_e and equivalently for $N = (n_h, l_h)$; $\hbar\omega$ is the photon energy and the summation is over all the hole and electron states. Of course, the cylindrical symmetry imposes l to be conserved during the excitation ($l_e + l_h = 0$), leading correspondingly to the S, P, etc. contributions to the absorption. In the results discussed below, the delta is approximated by a Gaussian with FWHM $\sigma_{G(M,N)}$.

Before discussing the interband absorption, let us consider the single-particle states. Bound dot states are present, since the dot confinement is strong enough. The dot spectrum contains correspondingly a discrete series of bound states followed by a continuum of unbound levels, corresponding either to the WL or barrier continua. When evaluating the interband transitions, one deals with different kinds of transitions [see Fig. 1(a)]: (i) the bound-to-bound transitions; (ii) the continuum-to-continuum transitions related to the WL and barrier delocalized states; (iii) the bound-to-continuum and the continuum-to-bound "crossed" transitions. These different transitions follow naturally from the composition ("addition") of the two single-particle spectra and are ultimately related to the fact that one photon creates two particles, one electron in the conduction band and one hole in the valence band.

Figure 2 shows the absorption spectrum (continuous line) for the following parameters: $R = 110$ Å, $r = 100$ Å, $h = 25$ Å, and $d = 20$ Å; $V_{0e} = 248$ meV and $V_{0h} = 102$ meV for a $In_{0.27}Ga_{0.73}As/GaAs$ system; $\sigma_G = 0.25$ meV for the ground S transition, 0.5 meV for the first P transition, and 2.5 meV otherwise. The spectrum for the WL without dot is also presented for comparison (dashed line). The two peaks at 1325 and 1360 meV correspond to the bound-to-bound $1S$ and $1P$ transitions, respectively. The edge of the WL two-dimensional transition is at 1405 meV. The broad feature with onset at around 1340 meV corresponds to the free-to-bound absorption involving the delocalized WL hole levels and the localized S-like electron levels of the dot. Different crossed (free-to-bound and bound-to-free) transitions contribute to the smooth increase of the absorption for energies below the WL edge.

The dot in Fig. 2 presents only a few bound states. Figure 3 shows the absorption spectrum for a more confining dot (of roughly identical size: $R = 100$ Å, $r = 80$ Å, $h = 30$ Å, $d = 10$ Å; $V_{0e} = 413$ meV and $V_{0h} = 288$ meV for a InAs/GaAs system). Many more transitions are present in this case and crossed transitions lead to a much deeper tail of the absorption. It is worth stressing that the first *excited* bound-to-bound transition is now isolated from the crossed continua. This provides a plausible coherent explanation to the results of the single dot experiments of Bayer and co-workers, which, in contradiction with other findings, showed a photoluminescence excitation spectrum dominated by thin lines and free of any residual background in the energy range around the S, P, and D transitions [6]. To make clear this point, note that for increasing electron and hole potential depths (at fixed geometry), the energy of a bound-to-bound transition decreases roughly twice as fast as the crossed ones, and thus various electron-hole states can fall below the first crossed continuum in very confining dots. This trend also appears when considering the size variation at constant confinement potentials, as illustrated in Fig. 4, where we show the variations of different bound-to-bound transitions and of the edges of two crossed transitions as a function of the dot radius (basis angle $\alpha = 12°$, $h = 15$ Å, $r = R - h/\tan\alpha$, $d = 6.66$ Å; V_{0e} and V_{0h} as in Fig. 3; the energies are

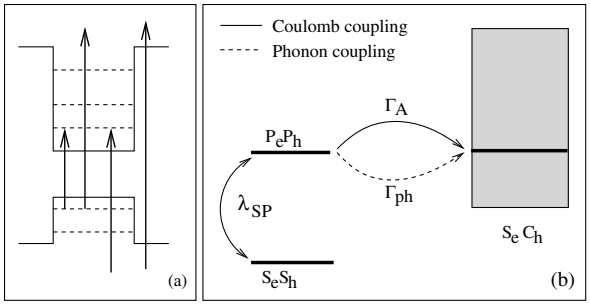

FIG. 1. (a) Schematic representation of the different contributions to the joint density of states of a quantum dot. (b) Schematic representation of different couplings affecting the bound P transition (see text).

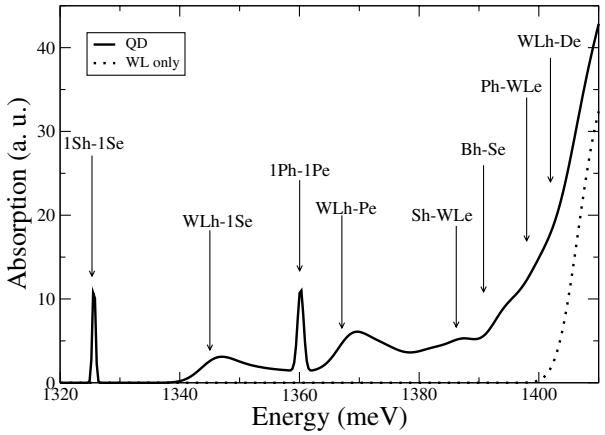

FIG. 2. Interband absorption probability for a quantum dot (solid line) structure and for the sole WL (dashed line). The arrows indicate either the energies of the different bound-to-bound transitions or the edges of the crossed ones.

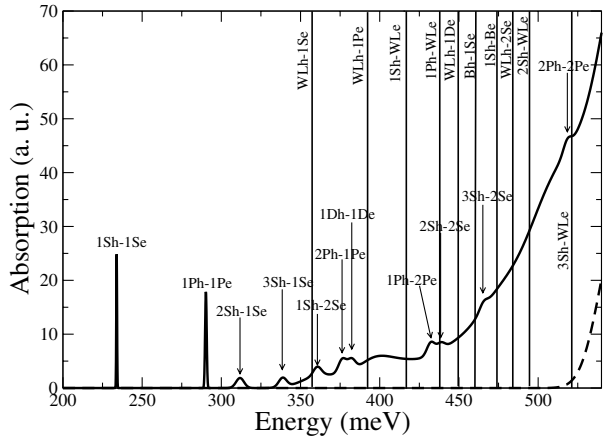

FIG. 3. Same as Fig. 2 for a more confining quantum dot (see text).

measured from the WL edge). The ground interband transition is necessarily located below the first crossed continuum. Large (and/or very confining) dots may present in addition a few *excited* radiative transitions below the crossed edges. The importance of having a bound transition resonant or not with a continuum is discussed below in connection with decoherence effects.

Let us first comment on some aspects of our calculations. First, the calculations were done for a single dot within a large numerical box of area S and height H ($\sqrt{S}/R > 10$, $H/h > 20$). For an ensemble, the crudest approximation is to assume that the dots are strictly independent. In this case, the spectrum for $N_d = N_S S$ dots (N_S is the dot areal density) is obtained from the single dot one by multiplying both the bound-to-bound and crossed oscillator strengths by the number of dots N_d, while leaving the free-to-free contributions unchanged. Finally, increasing the surface S at constant dot concentration leaves unchanged the optical absorption *profile* (the whole spectrum is proportional to S). In the same way, increasing H should not affect the whole spectrum for sufficiently large H (the number of barrier states increases and their squared wave functions decrease at the same rate). Another point is related to the valence states. Even though light holes may not possess bound dot levels, they bring additional contributions to the interband spectrum and should reinforce the high energy continuum (WL- and barrier-related) part of the absorption as well as the free-to-bound transitions. This point deserves further consideration. The last point is related to the role of the electron-hole Coulombic coupling on the dot absorption. We have estimated this effect within a simple model: the bound and delocalized hole (electron) levels in the presence of Coulombic interactions are numerically calculated by assuming the electron (hole) on a bound state [this amounts to averaging the Coulomb potential by the fixed electron (hole) distribution]. The Coulombic coupling shifts downwards the bound-to-bound lines but does not significantly alter their strengths. The crossed absorption results are only weakly modified: the energy positions of the different crossed edges are slightly affected, but not the background strength. Thus, the Coulombic term does not modify the existence of a smooth background absorption related to crossed electron-hole levels.

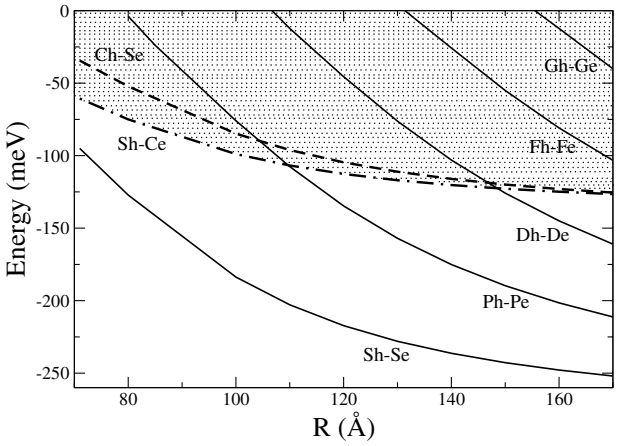

FIG. 4. Variation of the energies of some bound-to-bound transitions (full lines) and of the edges of the first free-to-bound (dashed line) and first bound-to-free (dashed-dotted line) transitions with the dot radius.

The existence of crossed valence-to-conduction joint density of states has far-reaching consequences. First, it provides a long awaited natural explanation of the continuous absorption background observed experimentally, as shown above. Second, it leads to novel insights in the understanding of decoherence effects in quantum dots, as discussed in the following. Whenever an excited bound-to-bound transition is located inside a crossed continuum, it autoionizes: any potential coupling the "discrete" and continuum levels leads to a disintegration process and, thus, to an *intrinsic broadening* of the bound transition. This coupling has been partly explored in the past, in connection with the Auger process for pairs of carriers (two electrons, two holes, or one electron-hole pair) placed in excited dot orbits (such as the P levels) [7]. In this case, the coupling is ensured by the Coulomb potential and leads to an efficient temperature independent intradot relaxation process. We now show that phonons can also couple the same states and lead to a temperature dependent broadening of the excited bound transitions. We discuss the case of longitudinal acoustical (LA) phonons, for the particular configuration of an electron-hole pair photoexcited in the P shell of a dot. In this case, the initial bound state (energy E_i) reads:

$$\Psi_i(\mathbf{r}_e, \mathbf{r}_h) = aP(\mathbf{r}_e, \mathbf{r}_h) + bS_e(\mathbf{r}_e)S_h(\mathbf{r}_h), \quad (2)$$

$$P(\mathbf{r}_e, \mathbf{r}_h) = \frac{P_{e(+)}(\mathbf{r}_e)P_{h(-)}(\mathbf{r}_h) + P_{e(-)}(\mathbf{r}_e)P_{h(+)}(\mathbf{r}_h)}{\sqrt{2}}, \quad (3)$$

where $T_\alpha(\mathbf{r}_\alpha)$ corresponds to the envelopes of the $T = S$ and $T = P_{\pm 1}$ bound orbitals for electrons ($\alpha = e$) and holes ($\alpha = h$). The presence of S contributions in the wave functions of the P levels results from Coulombic couplings within the different pair configurations of bound levels. The excited state is, however, mainly P-like, since $b \ll a$ ($b \simeq \lambda_{SP}/\Delta E_{SP}$, with λ_{SP} the intershell Coulombic term [see Fig. 1(b)] and ΔE_{SP} the intershell energy detuning). The final state is a crossed one, approximated by $S_e(\mathbf{r}_e)C_h(\mathbf{r}_h)$ in the case of a bound electron and a delocalized hole (see Fig. 2). The deformation potential coupling acts separately on the electron and hole. The scattering rate reads:

$$\frac{\hbar}{\tau} \simeq 4\pi |b|^2 C_0 k_B T \rho_P(E_i), \tag{4}$$

$$\rho_P(E_i) = \sum_{n,l} \left[\int d\mathbf{r} |C_{h(n,l)}(\mathbf{r})|^2 |S_h(\mathbf{r})|^2 \right] \delta(E_i - E_{nl}), \tag{5}$$

where we have used $n_{ph} \simeq \frac{k_B T}{\hbar\omega}$ for the phonon occupancy and added the emission and absorption processes. The summation is over different symmetries of the hole continuum ($n = 1, 2, \ldots$; $l = 0, \pm 1, \ldots$) with energy E_{nl}. $C_0 = D_h^2/2\rho c_s^2$ is a material parameter (for GaAs we have the potential deformation for holes $D_h = 6.7$ eV, the density $\rho = 5.3\ \mathrm{g\,cm^{-3}}$, and the sound velocity $c_s = 5000\ \mathrm{m\,s^{-1}}$). The total broadening of the P transition is finally written as:

$$\Gamma(T) = \Gamma(0) + a_{LA} T, \tag{6}$$

where $\Gamma(0)$ includes Auger contribution [Γ_A in Fig. 1(b)] and couplings to LO phonons via polaron effects (at low temperature) [2], and the linear coefficient a_{LA} is related to scattering by LA phonons [$\Gamma_{ph} = a_{LA} T$ in Fig. 1(b)]. The latter depends on the projected (as measured by the localized weighting function $|S_h(\mathbf{r})|^2$) density of states ρ_P of the final crossed continuum. We have obtained numerically $a_{LA} \simeq 20\ \mu\mathrm{eV/K}$ for the dot parameters of Fig. 2. This value is in good agreement with the measurements of Kammerer *et al.* [8] on dots of similar characteristics to the ones in Fig. 2. Note that a_{LA} is proportional to the *projected* density of states ρ_P, which is not exactly the same as the *optical* one governing the crossed absorption (although both increase with increasing density of crossed states). In addition, we stress that for single particles (electron or hole) the scattering is very inelastic, in spite of being roughly elastic for the photocreated *pair* (the energy of the involved acoustical phonons is typically a few meV).

Crossed states naturally arise from the joint nature of the dot density of states, involving valence and conduction bands with a discrete + continuous series of levels. Crossed electron-hole levels should contribute to any interband property of quantum dots. We have discussed here their role both on the interband optical absorption and on the broadening of a discrete (resonant) electron/hole state. The first effect provides an answer to the unsettled (to date) problem of the origin of the continuous absorption background of single dots, which was previously associated with sample imperfections but that we have shown to be an intrinsic effect. The second effect brings a clear explanation on the way low energetic LA phonons can contribute to the broadening of electron/hole transitions involving strongly localized conduction and valence states (bypassing in this way any kind of scattering inhibition resulting from the discrete nature of such states). Finally, let us briefly mention different effects where crossed transitions are also expected to play a major role: (i) crossed levels could provide an intrinsic mechanism for the nonlinear up-conversion measurements after excitation below the WL edge [5]: two photons are used to photocreate one dot and one WL pair via a free-to-bound and a bound-to-free absorption; (ii) the presence of mixed continua should help significantly the reaching of an even population of excited pair states among the dots (such recycling should be important especially on macro-optics experiments involving dots of different sizes); (iii) Fano effects could result for the resonant bound dot states. These points deserve, of course, further theoretical and experimental studies, but highlight even more the prominent role of the crossed transitions on the interband optical properties of quantum dots.

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