Self-induced transparency in semiconductor quantum dots

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Abstract.

We theoretically analyze the propagation of strong laser pulses in a sample of inhomogeneously broadened quantum dots. Because of the long exciton coherence times above a given threshold the laser pulse can propagate without suffering significant losses (*self-induced transparency*). At the highest field strengths the light pulse is shown to create within a self-modulation process massive exciton entanglement.

1. Introduction

In semiconductor quantum dots the three-dimensional carrier confinement results in discrete, atomic-like spectra and strongly enhanced carrier lifetimes. Such remarkable features make quantum dots similar to atoms under many respects, and suggest the exploitation of optically induced quantum coherence and correlations to drastically alter their nonlinear optical properties. Indeed, much recent work has been devoted to optically induced effects in quantum dots reminiscent of few-level systems: experimentally, coherent-carrier control of excitons [1] or photon antibunching [2, 3] have been demonstrated, while theoretically coherent population transfer [4], quantum-information processing [5, 6], or the measurement of single scatterings [7] have been proposed.

Observation of optical coherence effects in ensembles of quantum dots is usually spoiled by inhomogeneous line broadening due to dot size fluctuations, with typical broadenings comparable to the level splittings themselves. To overcome this problem, within the last couple of years a number of experimental techniques were developed to allow the observation of single quantum dots, hereby establishing the rapidly growing field of *single-dot spectroscopy* [8]. It should be emphasized, however, that inhomogeneous line broadening leads to decoherence effects which are substantially different from those induced by homogeneous broadening: a light pulse propagating in a medium of inhomogeneously broadened dots excites the (multi)excitons *in phase,* where —in contradistinction to homogeneous broadening— each quantum dot has a coherent time evolution. However, the phase varies from dot to dot, thus leading to interference effects, which in most cases prevent the observation of the coherent radiation–matter interaction. A striking exception is the phenomenon of *self-induced transparency* (SIT) [9, 10, 11], a highly nonlinear optical coherence effect which directly exploits inhomogeneous level broadening.

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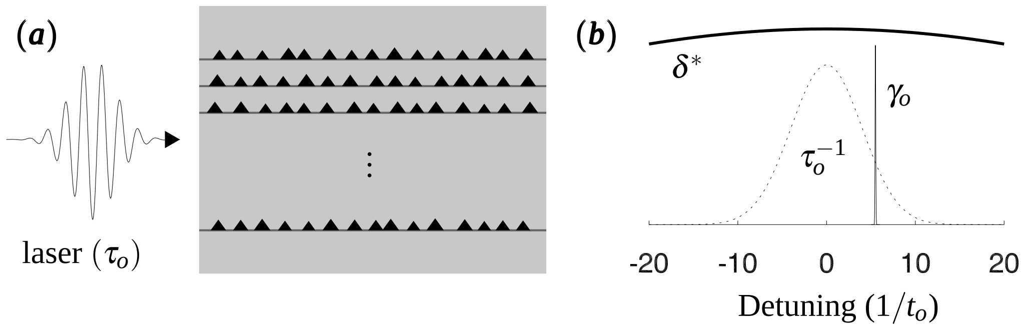

Figure 1. Schematic representation of: (a) the setup where an intense laser pulse with a temporal width τ_o enters from the left-hand side into the dot sample; (b) the groundstate exciton absorption spectrum for the inhomogeneously broadened dots (δ^*) and for a single dot (γ_o); the dashed line shows the spectral width of the laser pulse.

In their pioneering work McCall and Hahn [9, 10] first demonstrated that there exists a specific temporal pulse shape for which the light pulse entering a material of inhomogeneously broadened two-level systems propagates without suffering significant losses (*self-induced transparency*); this was indeed observed experimentally for atomic ensembles. SIT is important not only as a demonstration of quantum-coherence-induced modifications of optical properties, but also as a prototypical example of a cooperative phenomenon: its theoretical analysis not only requires to consider the material response in the presence of the driving light pulse, but also the back-action of the macroscopic material polarization on the light propagation (through Maxwell's equations). In fact, it turns out that this light-matter interplay leads above a given threshold to a self-modulation of a laser pulse with arbitrary pulse shape.

In this contribution we theoretically investigate self-induced transparency for an ensemble of semiconductor quantum dots. Here, the situation is more involved as compared to atoms because a strong light pulse can not only excite single electron-hole pairs (excitons) but also multiple pairs (multi-excitons). Using a general theoretical framework suited for the description of light propagation in dot samples, we will demonstrate that SIT should be observable in this class of material. Finally, we will show that the additional biexciton channel leads above a given threshold to massive exciton entanglement.

2. Theory

Our theoretical approach is based on the simulation of the coupled light-matter system, which requires the solution of both the material and Maxwell equations: here, the laser pulse (described through its electric field $\mathscr{E}$) creates an interband polarization in the quantum dots, which, on its part, serves again as a source term in Maxwell's equation and thus acts back on $\mathscr{E}$. Let us first discuss the time dynamics of a single dot (introducing an appropriate ensemble average later) which we describe within a common master-equation framework [12, 13]. Following Refs. [14, 15] we characterize the quantum dot system through its density-matrix ρ, whose diagonal elements ρ_{xx} describe the occupation of the few-particle states x (groundstate, single- and multi-excitons), and the off-diagonal terms $\rho_{xx'}$ account for the coherence between states x and x'. The time dynamics of ρ is then governed by [14, 15] ($\hbar = 1$ throughout):

$$\dot{\rho} = -i(h_{\text{eff}}\rho - \rho h_{\text{eff}}^{\dagger}) + \mathscr{J}\rho, \tag{1}$$

with $h_{\text{eff}} = h_o + h_{op} - i\Gamma$ accounting for: h_o, the Coulomb-renormalized few-particle states x; h_{op}, the light-coupling described within the usual rotating-wave and dipole approximations

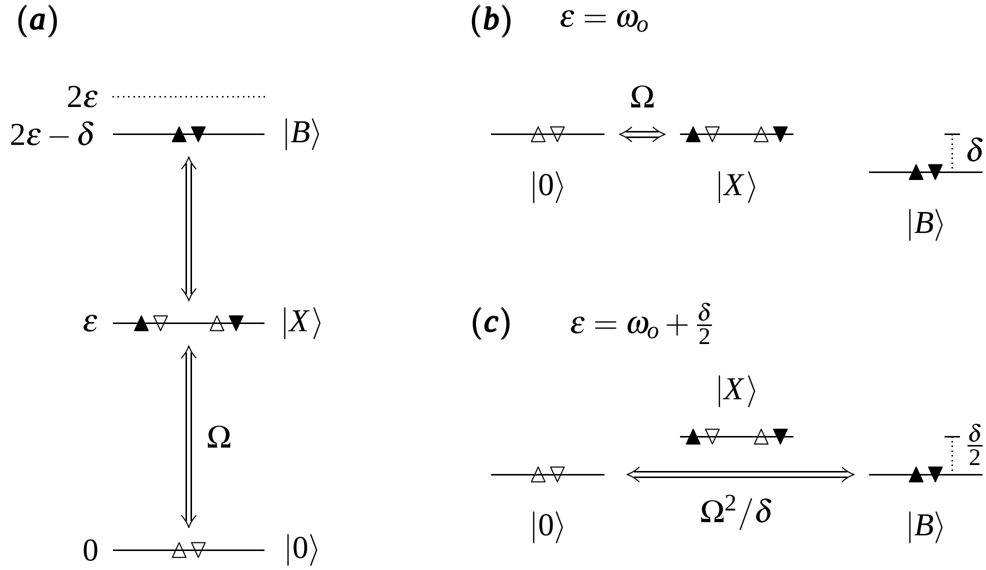

Figure 2. (a) Prototypical exciton-level scheme used in our calculations: $|0\rangle$ is the vacuum state; $|X\rangle$ are the spin-degenerate single-exciton states, and $|B\rangle$ is the biexciton groundstate whose energy is reduced as compared to 2ε because of correlation effects [8]; optical selection rules for linear polarization apply as indicated in the figure; (b,c) see discussion in text.

[13]; and $i\Gamma$, dephasing and relaxation due to environment interactions. While, within the spirit of Boltzmann's equation, the latter processes can be described as generalized *out-scatterings*, $\mathscr{J}$ accounts for *in-scatterings* which guarantee that the trace of ρ is preserved at all times. In this paper we shall consider low temperatures throughout, and thus take spontaneous photon emissions as the only source of dephasing and relaxation [16].

As regarding the time evolution of the light pulse, we assume a geometry (Fig. 1a) where a laser pulse enters from a dot-free region into the sample of inhomogeneously broadened quantum dots. Denoting the pulse propagation direction z and assuming an electric-field profile $\mathscr{E}_o \cos\omega_o t$, with envelope $\mathscr{E}_o$ and central frequency ω_o, we describe the light propagation in the slowly-varying envelope approximation [14, 11]:

$$\left(\partial_z + \frac{n}{c}\partial_t\right)\mathscr{E}_o(z,t) \cong -\frac{2\pi\omega_o}{nc}\,\mathrm{Im}\mathscr{P}(z,t), \tag{2}$$

where n is the semiconductor refraction index and c the speed of light. Most importantly, the term on the right-hand side describes the back-action of the material polarization $\mathscr{P}(z,t)$ on the light propagation. Here [14, 15]:

$$\mathscr{P}(z,t) = \mathscr{N}\int g(\varepsilon)d\varepsilon \sum_{xx'} \boldsymbol{M}_{x'x}(\varepsilon)\rho_{xx'}(\varepsilon,z;t), \tag{3}$$

with $\mathscr{N}$ the uniform dot density, ε the exciton energies, $g(\varepsilon)$ a normalized distribution characterized through the full-width of half maximum δ^* of the inhomogeneously broadened ensemble, and $\boldsymbol{M}_{x'x}$ the optical dipole matrix elements. Note that for each z and ε the time evolution of $\rho_{xx'}(\varepsilon,z;t)$ is given by Eq. (1). Computationally, we solve the coupled set of Eqs. (1,2,3) on a sufficiently dense real-space grid z_i, i.e., typically thousand points, where $\mathscr{P}(z,t)$ is calculated for each z_i from Eq. (3).

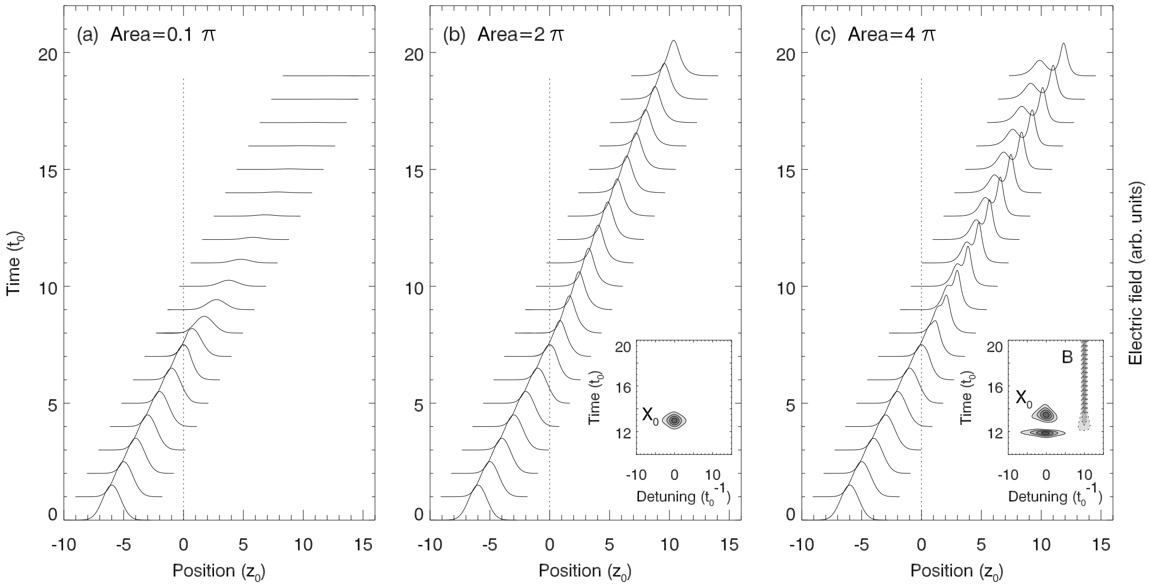

Figure 3. Results of our simulations of pulse propagation in a sample of inhomogeneously broadened quantum dots and for different pulse areas $A = \int_{-\infty}^{\infty} dt\, \Omega(t)$, with $\Omega(t)$ the usual Rabi frequency [11, 14]; we use linear polarization and assume a setup where the pulse enters from a dot-free region (negative z-values) into the dot region. The insets show contour plots of the time evolution of $\rho(\varepsilon, z; t)$ for $z = 5z_o$ (with detuning $\varepsilon - \omega_o$). We use a prototypical biexciton binding of 20 t_o^{-1}.

3. Results

In our calculations we assume a laser frequency ω_o tuned to the maximum of the inhomogeneously broadened exciton-groundstate transitions, Fig. 1b, and a typical exciton energy splitting of the order of several tens of meV, thus restricting our analysis to the generic level scheme of Fig. 2a, which consists of: the groundstate $|0(\varepsilon)\rangle$ (no electron-hole pairs present); the (spin-degenerate) excitons of lowest energy, $|X(\varepsilon)\rangle$; and the biexciton groundstate $|B(\varepsilon)\rangle$, whose energy $2\varepsilon - \delta$ is reduced because of Coulomb renormalizations [8, 17, 15].

3.1. Self-induced transparency

Before discussing the results of our simulations, Fig. 3, let us briefly address the basic characteristics expected for such a pulse propagation. Quite generally, we shall assume that:

$$\gamma_o \ll \tau_o^{-1} \ll \delta^*, \tag{4}$$

with γ_o the scattering rate for excitons and τ_o the temporal width of the laser pulse; in other words, the homogeneous level broadening should be much smaller than the spectral width of the laser pulse, and both broadenings should be much smaller than the inhomogeneous broadening δ^* (Fig. 1b). Apparently, for typical dot and laser parameters $\gamma_o^{-1} \sim 1$ ns, $\tau_o \sim 1$–10 ps, and $\delta^* \sim 50$ meV, Eq. (4) is easily fulfilled. As regarding the general trends of pulse propagation, we first concentrate on the case of a weak laser pulse entering the dot region: apparently, the laser will excite excitons and hereby suffer attenuation; a more detailed analysis reveals an exponential damping (Beer's law of linear absorption) with

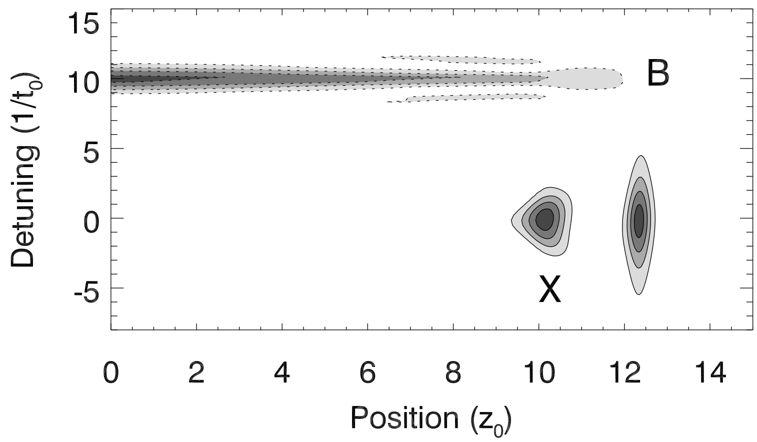

Figure 4. Exciton and biexciton population at $t = 20t_o$ and for a pulse area of 4π (see also Fig. 3c); contour lines of 0.25, 0.45, 0.65, and 0.85, respectively.

$z_o = nc/(2\pi^2 \mathscr{N} \omega_o \mu_o^2 g(\omega_o))$ providing a characteristic length scale [9, 10, 11] (henceforth we shall measure length in units of z_o, times in units of $t_o = nz_o/c$, and energy in units of t_o^{-1}, with $z_o \sim 0.25$ mm, $t_o \sim 3$ ps, and $t_o^{-1} \sim 0.2$ meV for typical InGaAs dot samples [14]). Because of the weak dephasing the laser-induced coherence keeps stored in the material even after attenuation of the laser pulse. Indeed, in the pioneering work of McCall and Hahn [9, 10] the authors showed for a proto-typical two-level system that above a given threshold this stored energy can again be fully extracted from the material and given back to the laser pulse. To come to this conclusion the authors made two important observations: first, there exists a specific pulse shape, i.e., hyperbolic secant, for which all two-level systems are driven through a sequence of exited states back to their groundstates irrespective of the detuning ε, provided that the pulse area is a multiple of 2π; second, a laser pulse of arbitrary shape entering a region of inhomogeneously broadened two-level systems achieves in a self-modulation-like process this hyperbolic secant shape. Consequently, it can propagate without suffering significant losses such that at each instant of time the pulse gives and receives the same amount of energy from the material; thus, this striking phenomenon has been given the name *self-induced transparency*.

Indeed, such behavior is observed in Fig. 3 which shows results of simulations using the level scheme of Fig. 2a: for small field strengths, Fig. 3a, the pulse becomes attenuated quickly; however, if the pulse area exceeds a certain value, Fig. 3b, self modulation occurs and the pulse propagates without suffering significant losses; the inset of fig. 3b shows a contour plot of the exciton population as a function of ε and time, where one clearly observes the above-mentioned excitation and de-excitation of exciton states; finally, at the highest pulse area, Fig. 3c, we observe pulse breakup [11, 9, 10]; the inset shows a 4π-rotation of the exciton states and an additional population of the biexciton channel, which, however, does not spoil the general pulse propagation properties.

3.2. Exciton entanglement

Let us concentrate in the following on this remaining biexciton population (inset of Fig. 3c and Fig. 4): here, the underlying states are *entangled exciton states*. To see that, we first note that because of the negligible dephasing the time dynamics can be considered as almost coherent (thus allowing for a wavefunction description); introducing furthermore the suggestive notations $|00\rangle$ for the groundstate, $|10\rangle$ and $|01\rangle$ for the spin-degenerate excitons $|X^{\pm}\rangle$, and $|11\rangle$ for the biexciton, whereby we have assumed that in the strong-confinement regime $|B\rangle$ is approximately given by the product state $|X^{+}\rangle \otimes |X^{-}\rangle$ [17], we observe that within each dot the wavefunction is of the form:

$$|\Psi\rangle = const \times (|00\rangle + \xi |11\rangle), \tag{5}$$

with ξ a complex number)—this wavefunction is exactly an *entangled* one (we checked that our results do not depend decisively on the specific values of τ_o and δ, and thus reflect a general behavior). Two important points should be addressed. Firstly, it is somewhat surprising that the pulse in Fig. 3c does not suffer attenuation despite continously creating entangled states. One must, however, note that it is the area of the pulse (and not its intensity) which contributes to self-induced transparency: thus, although continously loosing intensity at each instant of time the pulse reshapes to conserve area and to compensate for the losses suffered (note that for this to happen the mutual light-matter coupling is essential; see Ref. [10] for a related discussion). Secondly, we comment on the mechanism for the formation of entangled states. As apparent from the figures, the spectral width of the laser pulse, τ_o^{-1}, is insufficient to allow for a biexciton population through intermediate excitation of off-resonant excitons. To understand the origin of this entanglement-creation, we first note that the transition occurs at the photon energy where the biexciton is in two-photon resonance, i.e., $2\varepsilon - \delta = 2\omega_o$; introducing furthermore a rotating-frame representation according to $\omega_o|X\rangle\langle X| + 2\omega_o|B\rangle\langle B|$ [14], with Ω the usual Rabi frequency, the effective Hamiltonian of Eq. (1) reduces to:

$$h_o + h_{op} = -\frac{1}{2}\begin{pmatrix} 0 & \Omega & 0 \\ \Omega & \Delta & \Omega \\ 0 & \Omega & 2\Delta - \delta \end{pmatrix}. \tag{6}$$

where $\Delta = \varepsilon - \omega_o$ is the exciton-photon detuning. Figure 1 shows the corresponding level schemes for: (b) $\Delta = 0$, i.e., exciton in resonance with laser pulse; (c) $2\Delta - \delta = 0$, i.e., biexciton in two-photon resonance—here, the two states $|0\rangle$ and $|B\rangle$ are coupled by Ω through an off-resonant level $|X\rangle$. For constant Ω and assuming $\Omega \ll \delta$ one can analytically obtain the eigenstates of Eq. (6) [18], which consist of: the bare exciton state $|X\rangle$; two mixed states of $|0\rangle$ and $|B\rangle$. More specifically, if the system is initially in state $|0\rangle$ its time evolution is governed by the Hamiltonian [18]:

$$h_o + h_{op} \cong \frac{\Omega^2}{2\delta}\left(|B\rangle\langle 0| + |0\rangle\langle B|\right). \tag{7}$$

This two-photon coupling is completely analogous to the usual light-matter coupling [13], with the only difference that the coupling strength is given by Ω^2/δ rather than Ω. Apparently, as time goes on the system will oscillate between $|0\rangle$ and $|B\rangle$; consequently, the final biexciton population in Figs. 3 and 4 is governed by the pulse intensity and can be controlled through variation of τ_o. Elsewhere, we will show that this massive exciton entanglement leads to a clear-cut signature in the non-linear optical response [19].

4. Summary

In conclusion, we have discussed the phenomenon of self-induced transparency in a semiconductor quantum dot sample with inhomogeneous level broadening. Using a generic theoretical framework accounting for the mutual light-matter system in presence of single- and multi-exciton transitions, we have shown that for typical dot and material parameters

SIT should be observable in state-of-the-art quantum dot samples. Numerical simulations have revealed that the qualitative behavior of SIT is not changed in presence of biexciton transitions. However, the additional biexciton channel has been shown to lead to massive exciton entanglement; an intuitive explanation for the underlying mechanism has been given. SIT in dot samples constitutes a prototypical example for quantum coherence and for mutual light-matter interactions. It is expected to be of importance for the implementation of optically controllable switches or the propagation of intense laser pulses in high-density quantum dot samples, which might be of relevance for laser devices utilizing dots.

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