PHYSICAL REVIEW B **66**, 081306(R) (2002)

Rabi oscillations in the excitonic ground-state transition of InGaAs quantum dots

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(Received 19 April 2002; revised manuscript received 28 May 2002; published 15 August 2002)

We present measurements and calculations of optical Rabi oscillations in the excitonic ground-state transition of an InGaAs quantum dot ensemble at low temperature. Rabi oscillations which are damped versus pulse area and change period when changing pulse duration are observed. Comparisons with calculations show that the observed damping is not intrinsic to a single dot. Dephasing processes and the biexciton resonance change the amplitude and the period of the oscillations, respectively, while the damping versus pulse area is due to a distribution of transition dipole moments in the ensemble.

DOI: 10.1103/PhysRevB.66.081306 PACS number(s): 78.67.Hc, 42.50.Hz, 78.47.+p

With the recent achievements in fabrication of epitaxially grown semiconductor quantum dots (QD's) with high crystalline and optical quality,[1] light-matter interaction with atomlike objects having a large transition dipole moment can be explored. One fundamental example of coherent nonlinear light-matter interaction in a discrete level system is the well-known phenomenon of optical Rabi oscillations,[2,3] which is presently addressed in QD's both theoretically[4] and experimentally[5–7] for its application as one-qubit rotation in a QD-based quantum computer.

Optical Rabi oscillations are temporal oscillations of the population inversion in a two-level system driven by a strong resonant optical field on a time scale shorter than the dephasing time,[2] with an oscillation frequency proportional to the transition dipole moment μ and the electric-field amplitude ε. Population flopping over many periods is possible in systems with long dephasing time and large dipole moments. Under pulsed excitation, Rabi oscillations also manifest themselves as a sinusoidal dependence of the population inversion on the pulse area $\int_{-\infty}^{\infty} dt \mu \varepsilon(t)/\hbar$ (the time-integrated Rabi frequency). They have recently been observed on single exciton states weakly confined by the lateral disorder in narrow GaAs quantum wells.[5] However, many-body effects, which complicate the occurrence of Rabi oscillations in higher-dimensional semiconductor structures,[8] are still present in these weakly confined systems, resulting in a strongly damped oscillation versus pulse area.

Self-assembled InGaAs/GaAs QD's are ideal candidates for Rabi oscillations since they exhibit strong confinement energies, large transition dipole moments compared to atoms, and ultralong dephasing times of the excitonic ground-state transition at low temperatures.[9] First experiments on InGaAs QD's have been focused on Rabi oscillations in the excited-state transitions[6,7] that have, however, dephasing times five to ten times shorter than the excitonic ground state.[9] Moreover, the experiments were focused on single QD's. On the other hand, a QD ensemble is relevant, e.g., in the realization of quantum devices where Rabi oscillations can be implemented with light propagation to give rise to self-induced transparency solitons, similar to what has been reported in doped fiber waveguides.[10]

In this work we present experiments and calculations of optical Rabi oscillations versus pulse area in the excitonic *ground-state transition* of an InGaAs QD ensemble. We show that mechanisms intrinsic to a single dot such as biexcitonic effects and dephasing time, although influencing the amplitude and the period of the oscillation, respectively, do not damp the oscillations versus pulse area. Conversely, uniformity of the dot ensemble appears to be crucial and a distribution of transition dipole moments is shown to introduce a strong damping of the observed oscillations. The sample consists of three layers of self-organized $In_{0.7}Ga_{0.3}As$ QD's grown in the intrinsic region of a p-i-n ridge waveguide structure of 5-μm width and 500-μm length (details can be found also in Ref. 9). The excitonic ground-state transition (labeled 0-X) shows a Gaussian inhomogeneous broadening of the transition energies of 60-meV full width at half maximum (FWHM) attributed to fluctuations in dot size and indium concentration. We measure 65-meV energy separation between the excitonic ground state and first excited-state transition, and a wetting-layer transition ~210-meV above the excitonic ground state, demonstrating the strong confinement.[9] A pump-probe experiment is performed using a heterodyne technique[9] where copropagating and copolarized Fourier-limited optical pulses, resonant to the 0-X transition, are coupled into and out of the waveguide held in a high numerical aperture cryostat at a temperature of 10 K.

An intense pump pulse creates a ground-state exciton population which modifies the absorption properties of the dots. An absorption coefficient proportional to the population inversion of the 0-X transition is probed by a weak probe pulse after a delay time longer than the pump-pulse duration but shorter than the exciton lifetime. For a pump-pulse duration shorter than the dephasing time, the change of the time-integrated transmitted probe intensity induced by the pump should exhibit sinusoidal oscillations versus the pump-pulse area reflecting the Rabi oscillations of the population inversion.[5] In Fig. 1(a) the time-integrated differential transmission probe intensity ($\triangle T/T$) is shown versus pump-pulse area for Gaussian pump and probe pulses of 1.2-ps FWHM of the intensity (1.5-meV FWHM spectral width) and 20-ps pump-probe delay time (we measured an exciton lifetime of

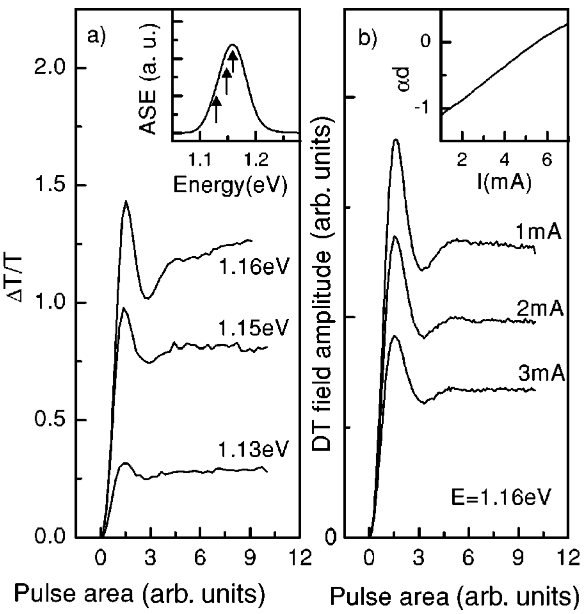

FIG. 1. (a) Time-integrated differential transmission probe intensity versus pump-pulse area measured for 1.2-ps pulses of different spectral positions in the inhomogeneous distribution, as indicated. In the inset the amplified spontaneous emission at low current injection, which evidences the inhomogeneous broadening, is shown. (b) Differential transmission probe field amplitude measured at different injection currents, as indicated. In the inset, the optical density versus injection current is shown.

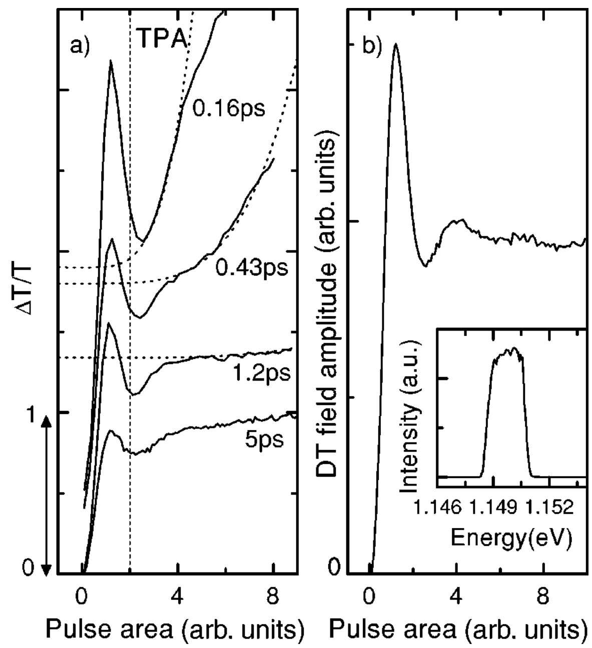

FIG. 2. (a) Time-integrated differential transmission probe intensity versus pump-pulse area measured for different pulse durations at 1.16-eV spectral position and 1-mA injection current. Curves are vertically displaced for clarity. The vertical dashed line is a guide for the eyes. Dotted lines show the effect of two-photon absorption (TPA). (b) Differential transmission probe field amplitude for a spectrally square-shaped pulse. The pulse spectrum is shown in the inset.

1 ns and a dephasing time of the zero-phonon line of 500 ps at 10 K in these dots[9]). We observe Rabi oscillations which are damped after the first maximum and minimum.

To understand the origin of the damping we first consider the influence of propagation. At the spectral position resonant with the center of the inhomogeneous distribution we estimate a small-signal absorption corresponding to ~ 1.5 optical density. To reduce propagation effects, we have performed measurements with varying optical density below 1. They are shown in Fig. 1 where the wavelength of the optical pulses is tuned to the tail of the inhomogeneous distribution [Fig. 1(a)] or part of the dots is bleached by electrically injected carriers [Fig. 1(b)].[11] In all cases, the oscillations exhibit strong damping and their visibility eventually deteriorates in the tail of the absorption. Propagation effects are thus not responsible for the observed damping.

Let us now consider biexcitonic effects. A QD excitonic ground-state transition is not a two-level system but a four-level system with two differently spin-polarized exciton states and an exciton-biexciton transition (X-XX) nearly degenerate to the 0-X transition.[12] The biexciton binding energy (E_{XX}) is 3 meV in the investigated dots.[9] We have experimentally studied the effect of the biexcitonic resonance in the Rabi oscillations by using different pulse durations. The results are shown in Fig. 2(a) for Gaussian pulses from 0.16 ps to 5 ps, having spectral widths from 11.5 meV to 0.36 meV. Intuitively (but also confirmed by the numerical simulations discussed later), for a pulse spectral width much smaller than E_{XX} the X-XX transition is out of resonance and cannot be relevant, thus the Rabi oscillations are basically those of a two-level system involving only the 0-X transition. However, for all pulses the damping of the oscillations remains, excluding biexcitonic effects as the origin. The effect of the biexciton is instead a change in the oscillation period, which is also reproduced by the calculations discussed later (Figs. 3 and 4).

Dephasing processes are relevant when the pulse duration is comparable or longer than the dephasing time. At 10 K the 0-X polarization decay is dominated by a 500-ps dephasing time of the zero-phonon line,[9] well above the used pulse durations (also for small injection currents[13]). However, a fast initial dephasing of about 1.5 ps due to exciton-acoustic-phonon interactions is also present. A pump-pulse duration well below 1 ps reduces the effect of this initial dephasing. In fact, in Fig. 2 we observe a quenching of the *amplitude* of the oscillations with increasing pulse duration. At high peak pump-pulse intensities, i.e., for short pulse duration, a sizeable two-photon absorption (TPA) occurs in the waveguide which covers the observation of Rabi oscillations. The estimated effect of the TPA is shown in Fig. 2(a) (dotted lines). It was taken to be proportional to the nonresonantly excited density given by the absorbed pump power integrated over the pulse duration [$\propto (A/t_0)^4 t_0$ with A pulse area and t_0 pulse duration].[14] Note that TPA was evidenced also in single pulse transmission measurements[15] and pump-probe experiments at transparency.[16] In Fig. 2(b) the Rabi oscillations are shown for a square-shaped spectrum of pump and probe pulses of 2-meV spectral width corresponding to ~ 1-ps pulse duration. For this specific case, we can distinguish a

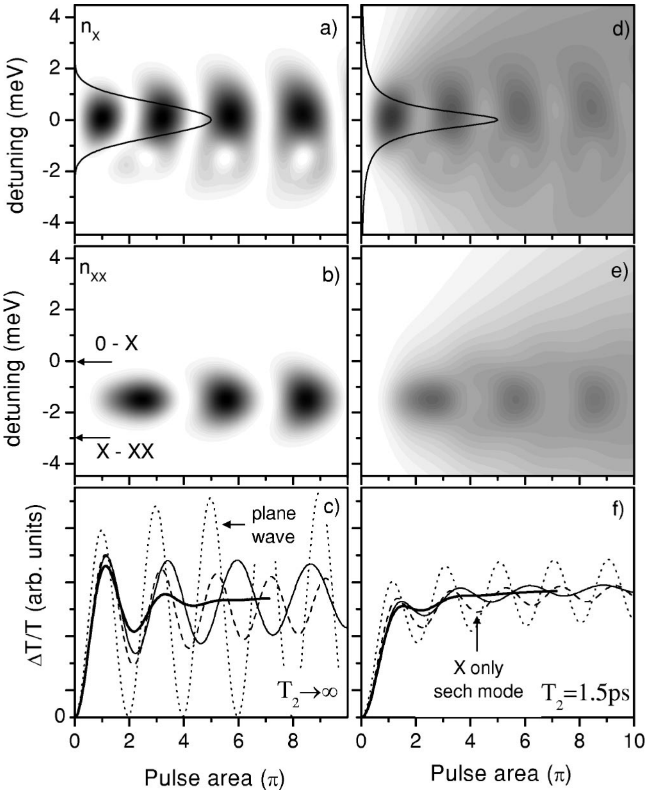

FIG. 3. Exciton (a) and biexciton (b) occupation probabilities calculated versus the detuning between the pulse carrier frequency and the 0-*X* transition and as a function of the pulse area. The contour plot is in gray linear scale (black=1). The pulse spectrum is also shown as solid line in (a), and the arrows in (b) indicate the energy positions of the 0-*X* and *X*-*XX* transitions. (d) and (e) as in (a) and (b), respectively, but using a finite dephasing time of 1.5 ps. The Lorentzian absorption cross section is also shown in (d) as a solid line. (c) Differential transmission intensity calculated: without biexciton and with a uniform plane-wave field (dotted line), with an hyperbolic secant field profile (dashed line), including the biexciton (solid line) and after averaging over a distribution (20% standard deviation) of dipole moments (thick solid line). (f) as in (c) for 1.5-ps dephasing time.

second oscillation maximum, indicating that a sharpened distribution of the spectral intensity improves the visibility of the oscillations.

To obtain more insight into the origin of the observed damping, we compare the experimental results with calculations. We have numerically solved the optical Bloch equations of a four-level system that includes the *X*-*XX* transition with the same oscillator strength as the 0-*X* transition,[12] using 3-meV biexciton binding energy and neglecting fine-structure polarization splittings of the exciton ground state.[17] Results are shown in Fig. 3 for a Gaussian pulse of 1.2-ps duration. Figures 3(a) and 3(b) are two-dimensional plots showing the occupation probability after the pulse of the exciton state [Fig. 3(a)] and of the biexciton state [Fig. 3(b)] for an infinitely long dephasing time calculated versus pulse area and detuning between the 0-*X* transition and the pulse carrier frequency. The inclusion of the biexciton resonance in the calculations creates a biexciton density which is maximum when the pulse carrier frequency is in resonance with half the 0-*XX* transition energy. Such a biexciton density oscillates versus pulse area with a *different period* compared to the exciton density. In Fig. 3(c) we show the calculated $\triangle T/T$ versus pump-pulse area, to directly compare with the experiments. Integration over the inhomogeneous broadening of the transition energies and spatial averaging over an hyperbolic secant field profile of the transverse electric waveguide mode in the QD plane have been included in the calculation of the spectral absorption experienced by the probe pulse. Then, the total transmitted probe intensity is evaluated by integrating over the probe spectrum. In Fig. 3(c) the calculation is shown (solid thin line) and compared with calculations neglecting the biexciton resonance (dashed line) and with a uniform plane-wave field (dotted line).[18] Averaging over the spatial mode profile quenches the amplitude of the oscillations, while the effect of the biexciton is a change in the period of the Rabi oscillations versus pulse area. However, *many oscillation periods are still present*. The introduction of a nonexponential dephasing, according to the measured polarization decay,[9] is nontrivial. Calculations that overestimate the effect of the initial fast dephasing are shown in Figs. 3(d)–3(f) using an exponential dephasing of 1.5 ps for both the 0-*X* and *X*-*XX* transitions. This fast dephasing results in a reduced amplitude of the oscillations which, however, are not additionally damped versus pulse area. Only when including a Gaussian *distribution of transition dipole moments* (20±5% standard deviation) was a strong damping similar to the experiment found, as shown by the curves with thick solid lines in Figs. 3(c) and 3(f). The damping is therefore an ensemble effect, not intrinsic to the Rabi oscillations of each single QD.

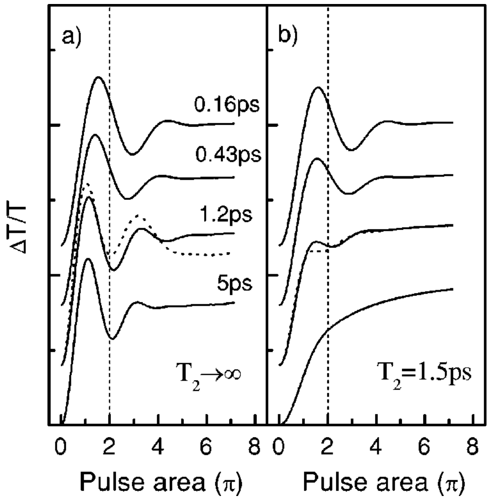

FIG. 4. Differential transmission intensity calculated for different pulse durations, as indicated, for an infinitely long dephasing time (a) and 1.5-ps dephasing time (b), vertically displaced for clarity. Dotted lines are calculations for a spectrally square-shaped pulse of ~1-ps duration. The 2π pulse area is marked with a dashed line, for reference.

As mentioned before, the pulse duration affects the period of the oscillations due to the *X*-*XX* transition. This is calculated in Fig. 4 for an infinitely long dephasing time [Fig.

4(a)] and 1.5-ps dephasing time [Fig. 4(b)], including all the previously mentioned averaging. With decreasing pulse duration, corresponding to a spectral width comparable to or larger than the biexciton binding energy, the period of the Rabi oscillation increases, as also experimentally observed [see Fig. 2(a)]. This implies that biexcitonic effects are important in the estimation of the transition dipole moment from the period of Rabi oscillations. Finally, results using a spectrally square-shaped pulse, similar to the experiment in Fig. 2(b), are shown as dotted lines in Fig. 4 and confirm the stronger visibility of the second maximum in this case.

Let us now estimate the mean value of the 0-*X* transition dipole moment. From the measured 1-ns exciton radiative lifetime and with 3.5 refractive index we calculate[19] $\mu=34$ Debye. This is consistent with $\mu=31$ D deduced from the measured small-signal absorption coefficient ($\alpha=30\ \mathrm{cm}^{-1}$), with a dot areal density of $2\times10^{10}\ \mathrm{cm}^{-2}$ and a size of the waveguide mode in the growth axis of 0.37-μm intensity FWHM, as estimated experimentally from an image of the far-field mode. The dipole moment can be also estimated using the Rabi oscillations. This estimate depends both on the size of waveguide mode and on the intensity of the field effectively coupled into the sample and it is thus the most inaccurate. We deduce $\mu=18$ D from 71-fJ pulse energy at 2π pulse area of a Gaussian pulse of 1.2-ps duration with a waveguide mode of 0.7-μm intensity FWHM in the lateral direction.

In conclusion, we have reported measurements and calculations of optical Rabi oscillations in the excitonic ground-state transition of an inhomogeneously broadened InGaAs quantum dot ensemble. We found that a distribution with 20% standard deviation of transition dipole moments results in a strong damping of the oscillations versus pulse area. These results show *quantitatively* how uniformity in dot size and shape is important for any application based on a coherent optical control of excitonic transitions in a dot ensemble. Conversely, Rabi oscillations in a single dot are expected to be quite "robust" as a function of pulse area, even when dephasing processes and biexcitonic effects are included. Remarkably, the latter are found to change the period of the Rabi oscillations both in the experiments and in the calculations.

This work was supported by DFG in the frame of Grant Nos. Wo477/17-1 and SFB296. P.B. is supported by the European Union with the Marie Curie Individual Fellowship No. MCFI-2000-01365.

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