Coherent Spectroscopy of Optically Gated Charged Single InGaAs Quantum Dots

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The excited states of neutral and charged single InGaAs/GaAs quantum dots are studied using a confocal microspectroscopy technique. Because of their different Coulomb energy shifts, the charged and neutral states of the same quantum dot can be selectively excited. The charge of the quantum dot is controlled by a photo-depletion mechanism. Time-resolved coherent spectroscopy shows that the dephasing time of the excited states is *longer* when the quantum dot is charged. Rabi oscillation of the excited state of a singly charged quantum dot is demonstrated.

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Since the development of local spectroscopy techniques, the atomiclike optical properties of semiconductor quantum dots (QDs) have been intensively studied. Discrete electronic states of single QDs can be considered as localized two levels systems. The increased interest in QDs has been related to the potential use of these two level systems as building blocks of prospective quantum logic gates [1,2]. The development of techniques to control Coulomb interactions [3,4] and to perform coherent manipulation of carrier wave functions in single QDs (i.e., on a time scale shorter than the dephasing time) [5–9] are key points for a successful implementation of quantum information processing in these systems and the development of QD based quantum information systems.

In this Letter we show how we can optically control the charge state of a single QD and coherently manipulate the confined wave function exploiting quantum interference and Rabi oscillation phenomena. We use microspectroscopy to study the optical properties of individual InGaAs/GaAs QDs. The excitonic species observed in the ground state of an individual QD are found to strongly depend on the laser excitation energy. Neutral and charged excitons are separately identified in the emission under resonant excitation. Wave packet interferometry is performed on the different excited states of these complexes. We find that the dephasing time of the first excited state of the QDs *increases* when an additional carrier is introduced in the ground state. These coherence times make possible the direct observation of Rabi flopping of the population of the excited state of a singly charged QD under resonant pulsed excitation.

The self-assembled QDs are grown using metalorganic vapor phase epitaxy. The deposition of a reduced amount of InAs, 1.8 ML (close to the critical thickness of the dot formation) is employed to produce a sparse QD density of about $1–5 \times 10^9$ cm^{-2}. The low temperature ($T \approx 5$ K) photoluminescence (PL) of individual QDs is excited and collected through large numerical-aperture microscope objectives and aluminum shadow masks with 0.2–1.0 μm apertures. In photoluminescence excitation spectroscopy (PLE), the QD is excited with a linearly polarized tunable Ti:sapphire laser and a cooled CCD camera is used for parallel detection of the complete PL of the s shell of the QD (exciton ground state emission region) as a function of the laser excitation energy.

PL and PLE spectra from four QDs are presented in Fig. 1. The s-shell emission of QD1 obtained under *nonresonant* excitation of the GaAs barriers is shown (expanded) in Fig. 2(a) for different excitation densities. At low excitation densities, a single narrow line (X) dominates the spectra. For higher powers, a second emission line (X_2) appears on the low-energy side of X. The linear and the quadratic power dependence of X and X_2, respectively [Fig. 2(a) (inset)] identify these lines as the recombination of the exciton and biexciton in the same QD. For high excitation densities [Fig. 2(a)] additional features, X_s, arising from the recombination of multiexciton complexes appear on the low-energy side of X_2 and in the vicinity of excited states absorption [Fig. 1, QD1] [10,11].

As the excitation energy is tuned below the wetting layer, the structure of the luminescence of the s shell changes drastically: a new emission line, labeled X^*, emerges on the high-energy side of X. To study in detail the properties of this new feature, the complete luminescence of the s shell is recorded as a function of the laser excitation energy [Fig. 1 (inset)]. The PLE spectra of each s-shell PL line is directly extracted from this map. The structure of the PLE shows a clear correlation between X and X^* detection, an observation consistent with both features arising from the *same* QD.

The X PLE spectra present sharp absorption lines on top of a continuum background related to spatially indirect transitions involving interface states of the wetting layer [12]. Even if the detailed structure of absorption resonances is slightly different from dot to dot, a general behavior can be extracted: as the excitation energy is increased, a sharp absorption (e_0) is first observed in the PLE spectra of X. A second sharp absorption (e_1) is then observed for X^* at slightly higher energy. In some of the QDs (e.g., QD3, QD4) e_1 presents a doublet structure. A signal, usually smaller, is also observed for X under excitation on e_1. Absorption resonances at higher energy

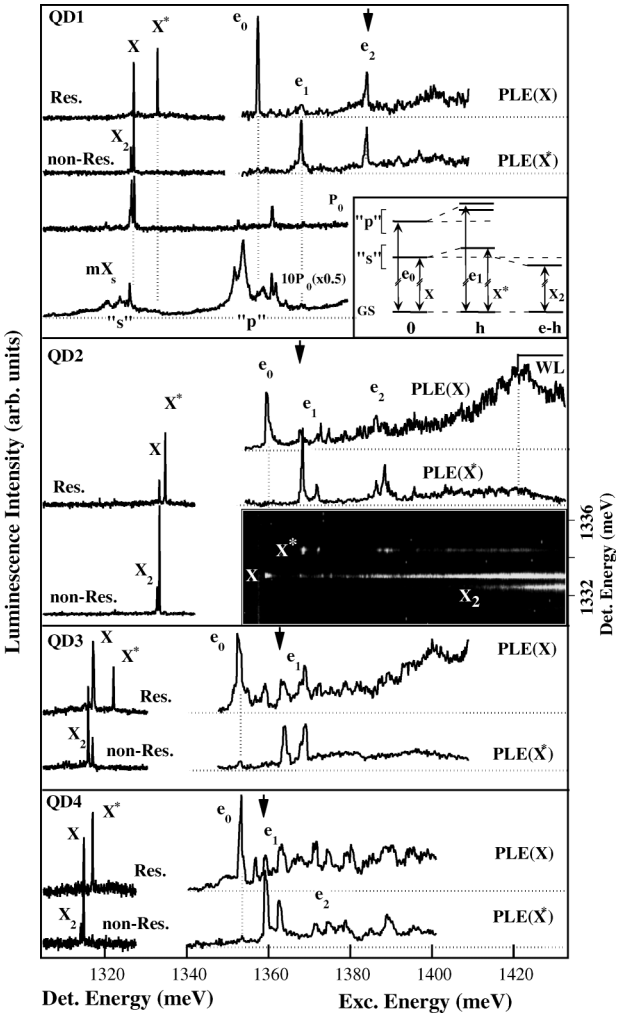

FIG. 1. PL and PLE spectra for four individual QDs (labeled 1,2,3,4). PL spectra are obtained under resonant (excitation energy indicated by arrows) or nonresonant ($\lambda_{exc} = 532$ nm) excitation. PLE spectra are obtained for detection at the energies of X and X^* lines observed in resonant PL. Inset (QD1): diagram presenting the optical transition discussed in the text. Inset (QD2): complete s-shell luminescence of QD2 as a function of detection and excitation energy.

give both a luminescence of X and X^*. The PLE background of X gradually increases until the threshold of the wetting layer absorption (≥ 1420 meV). A smaller absorption background is observed in the PLE of X^*.

The evolution with excitation intensity of the PL of QD1 under *resonant* excitation on e_1 is presented in Fig. 2(b). At low excitation intensity, X^* dominates the spectra. Both the intensity of X and X^* increase first linearly with excitation power [Fig. 2(b) (inset)] before X^* reaches a maximum and decreases to higher power as the emission of X_2 appears. Whereas excitation of e_0 selects the exciton, high excitation intensity on e_1 produces emission of both X and X^*.

To understand why X^* has not been identified in nonresonant spectroscopy, micro-PL experiments were carried out under *simultaneous* resonant and nonresonant

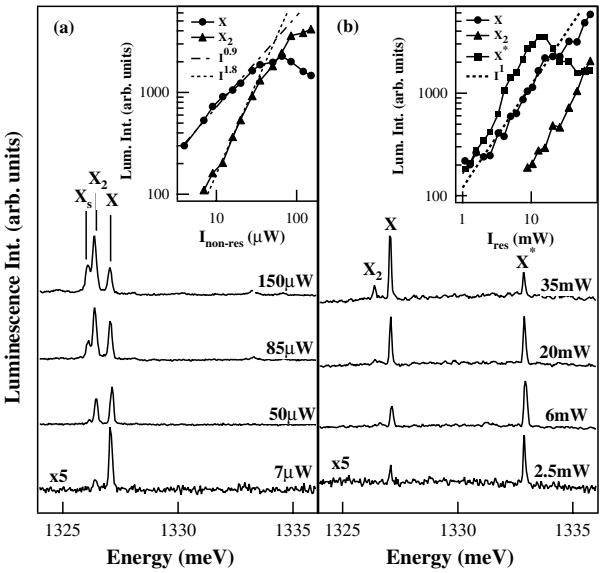

FIG. 2. QD1 s-shell PL spectra as a function of excitation intensity for (a) nonresonant and (b) resonant excitation at e_1. Inset: extracted intensity of X, X^2, and X^* as a function of excitation intensity.

excitation. The evolution of the PL of QD1 for a fixed excitation at e_2 while varying the nonresonant excitation is presented in Fig. 3. With resonant excitation, the transitions X and X^* dominate the spectra. The left inset of Fig. 3 directly shows that X^* can be produced only by resonant excitation: no traces of X^* are observed in nonresonant spectra. When carriers are also created in the GaAs barriers by nonresonant excitation, X^* progressively disappears as the contribution of X_2 increases. A similar exchange of intensity between X^* and X_2 is observed as the intensity of the p-state excitation increases.

This spectral evolution is characteristic of a photodepletion mechanism in modulation doped QDs [13]. High-energy photoexcited e-h pairs are dissociated in the space-charge region between ionized impurities and the charged QDs. Carriers attracted into the charged QD then recombine in the QD with the extra carrier. Thus photodepletion removes the charge from the QD at a rate proportional to the absorbed nonresonant excitation, and produces PL only at X. For excitation below the wetting layer and at low excitation densities, this photodepletion mechanism is turned off and the recombination of a charged exciton can be observed [14].

Thus, X^* likely corresponds to a charged exciton created by the transfer of carriers from a weak background doping in the barriers. With an extra charge carrier in the QD ground state, resonant p-shell excitation can occur only into the e_1 state, which relaxes into the s shell and emits at X^*. For the neutral QD, p-shell absorption is at e_0, relaxing to emit at X. Thus we show here that resonant optical excitation is capable of selectively pumping a single QD depending on its charge state.

Currently we are unable to directly prove whether X^* is positively or negatively charged, but in these kinds of

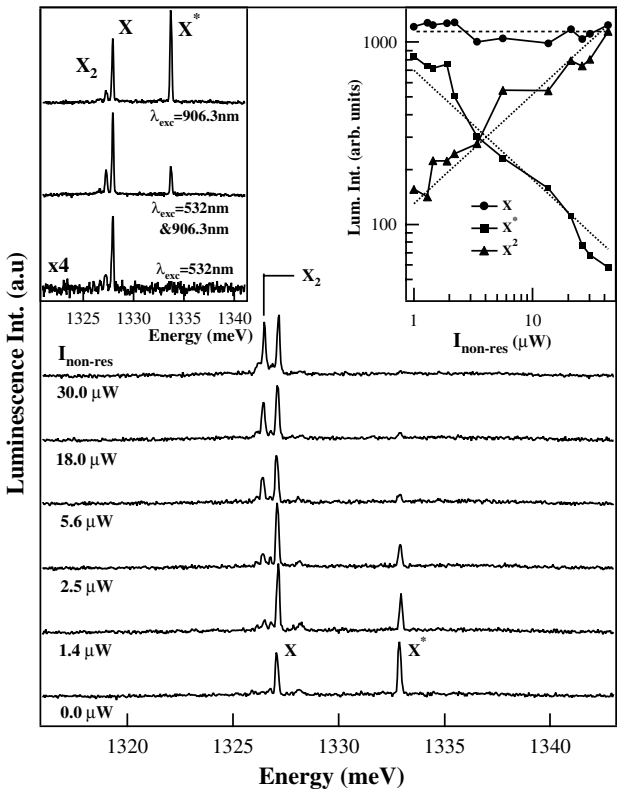

FIG. 3. QD1 PL for fixed resonant excitation intensity on e_2 ($\lambda_{\text{exc}} = 895.2$ nm) with varying nonresonant excitation intensity. Inset: (left) QD1 PL spectra for nonresonant ($\lambda_{\text{exc}} = 532$ nm), resonant on e_1 ($\lambda_{\text{exc}} = 906.3$ nm) and simultaneous resonant and nonresonant excitation. (right) Intensity variation of X, X_2, and X^* as a function of the nonresonant excitation intensity.

QDs the negatively charged exciton is invariably observed on the low-energy side of the exciton [3,4]. In fact, emission of the positively charged exciton has been tentatively observed a few meV above the exciton transition in mesa structures [17,18]. Thus we henceforth take X^* to be a positively charged exciton (although this is not crucial for our discussion below). The energy shift between X^* and X, $\Delta V_s = 1.5$–6 meV, is a measure of the relative energy of $2h + e$ and $h + e$ in the QD ground state. Since X^* is blueshifted above X, $h - h$ repulsion in the ground state dominates $e - h$ attraction [3,19,20].

The PLE spectra allow us to also measure the energy shift between p-state *absorptions* of X and X^*, $\Delta V_p = \hbar\omega(e_1) - \hbar\omega(e_0) = 5$–$12$ meV. In the simplest approximation, this energy difference arises from the Coulomb interaction between a p-state exciton and an s-state carrier. As in the case of the ground state charged exciton, the sign of ΔV_p shows that $h_p - h_s$ repulsion dominates $e_p - h_s$ attraction. What is less expected is that in all QDs, $\Delta V_p > \Delta V_s$ even though s and p holes are more spatially separated than two s holes. In addition both $\Delta V_{s,p}$ vary considerably in different QDs.

From a simple calculation, in a parabolic potential, we expect a splitting of the p states of the charged QDs due to the Coulomb exchange energy of the holes [20]. In some of the QDs (QD3,4), the absorption e_1 presents a doublet structure with a splitting $\Delta V_{ex} = 3$–5 meV. The amplitude of the components of the doublet can be different (QD2) and this doublet structure is not systematically observed (QD1). Possibly the lowest excited state does not have the simple symmetry of the p-states of a parabolic potential. Alternatively, as the energies of the studied excited states are in the vicinity of the energy of the bulk GaAs LO phonon (36 meV), the energies of these levels as well as their relaxation rate are strongly influenced by the coupling with LO phonon [21].

To study the influence of the long-lived charge carrier in the ground state, on the coherent optical properties of the excited states of the QDs, we performed single QD wave packet interferometry [5,6]. The excited state of the QD is resonantly pumped by a pair of phase-locked picosecond pulses obtained by sending a laser pulse through a Michelson interferometer with a coarse (τ_c) and a fine (τ_f) delay adjustment [Fig. 4(a) (inset)]. The quantum interference between the wave functions created by the two phase-locked pulses gives rise to sinusoidal oscillations at the optical frequency of the excited state population. The oscillation of the excited state population can be monitored through the PL of the ground state [5].

The map in Fig. 4(a) (inset) shows the resulting interferogram in the PL of QD2 resonantly excited at e_1 as a function of both the detection energy and the fine delay time τ_f, for a fixed delay $\tau_c = 10$ ps. Oscillations are observed in the intensity of X^* as the delay between pulses is changed in 0.3 fs steps. Oscillation at this time delay, where there is no overlap between the picosecond pulses, results from the interference of wave functions and has a purely quantum mechanical nature. The decrease of the oscillation amplitude as a function of pulse delay τ_c [Fig. 4(a)] shows that exciting e_1 results in an exponential decay with a fast dephasing time of $\sim$13 ps. For the same experiment carried out on the first excited state e_0, the decay of the oscillation in the X intensity roughly reproduces the 3 ps autocorrelation of the laser pulses. The dephasing time of this state is shorter than the current temporal resolution of our experimental setup.

These dephasing times are mainly controlled by the relaxation rate of the carrier to the ground state of the QD. A fast decay rate is expected due to efficient mixing of the first excited state and the one LO phonon sideband of the ground state [21]. If an additional carrier is introduced in the ground state of the QD, the relaxation rate is reduced due to the Pauli exclusion principle. Although a detailed theory is to be developed, this simple argument suggests the lifetime and hence the dephasing time of the p-state exciton increase for a charged QD.

As illustrated in Fig. 4(a) (inset), a QD under resonant excitation can be considered as a three level system. It is well known from atomic physics that the population inversion of a discrete transition under strong pulsed resonant excitation undergoes sinusoidal (Rabi) oscillations

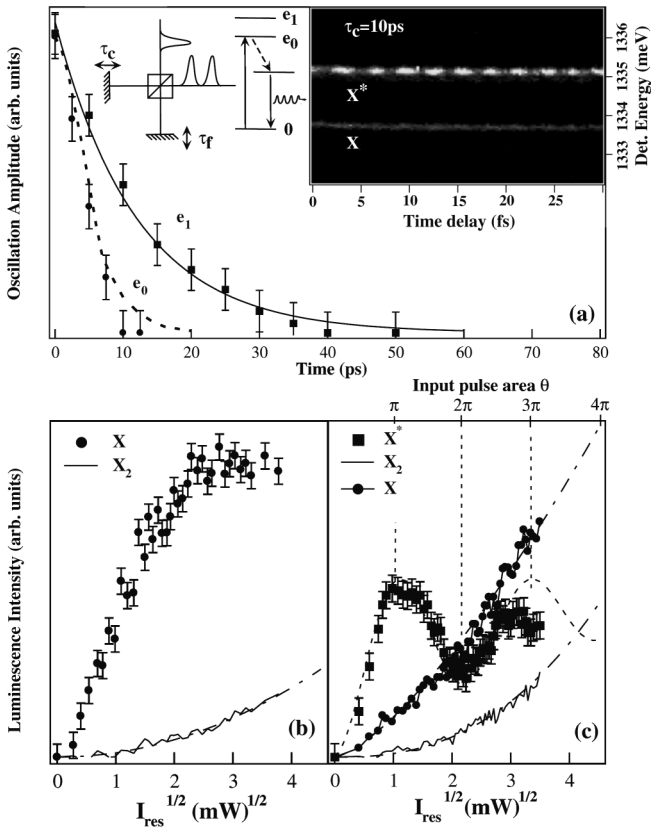

FIG. 4. (a) Amplitude of the oscillations in the PL of QD2 as a function of delay τ_c for excitation at e_0 (squares) and e_1 (circles). The dotted line corresponds to the autocorrelation of the excitation pulses. Inset: (left) schematic experimental setup; (right) quantum interference map for QD2 PL at $\tau_c = 10$ ps under resonant excitation of e_1, shown as a function of emission energy and pulse fine delay τ_f. (b),(c) Evolution of the QD2 PL intensity under pulsed resonant excitation of (b) e_0 and (c) e_1, as a function of the square root of the average power density.

as a function of input pulse area $\Theta(t) = 2\mu/\hbar \int_{-\infty}^{t} \epsilon(t')dt'$ [μ is the dipole moment of the transition and $\epsilon(t)$ the amplitude of pulses' electric field] [7,22]. This experiment provides a way of directly estimating the dipole moment of the excited transition [6,8].

The PL intensity of QD2 under pulsed resonant excitation on e_0 and e_1 (respectively) is plotted in Figs. 4(b) and 4(c) as a function of the square root of the average power density, which is proportional to the pulse input area. Exciting e_0 produces a saturation of the X PL at high excitation density together with the appearance of weak PL from X_2. Exciting e_1 is quite different: oscillations are observed in the power dependence of the PL of X^* [Fig. 4(c)]. Simultaneously, a monotonic increase of X and X_2 is observed. At this excitation energy, the contribution of X and X_2 mainly come from the absorption of the broadband pulses in the continuum background [15]. The oscillation of X^* is a direct manifestation of Rabi oscillations in the resonantly excited state e_1. The population of e_1 is maximum for a π pulse, decreasing as the excitation intensity is further increased. However, a portion of the excited state population loses its phase coherence or relaxes to the ground state during the pulse excitation and contributes to residual PL for a 2π pulse. From the estimated input pulse area, we find p-state dipole moments of about $\mu_{e_1} = 5$ Debye and $\mu_{e_0} < 2$ Debye, showing that the charged QD appears to have double the p-state oscillator strength. Such an increase has been theoretically predicted for negatively charged QDs [23]. Finally, we note that the intensity of ground state PL from X is roughly double that of X^* despite its smaller excited state oscillator strength, which suggests that a nonradiative process competes with the radiative recombination of X^*.

In summary, we applied coherent microspectroscopy to *selectively* study the excited states of neutral and charged individual InAs QDs. In addition to shifting the ground and excited state energies, an extra carrier in the QD is found to increase the excited state dephasing time. In the strong excitation regime, Rabi oscillations are observed in the excited states of charged single QDs, suggesting implementations of controlled qubit operations.

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[1] F. Troiani *et al.*, Phys. Rev. B **62**, R2 263 (2000).
[2] E. Biolatti *et al.*, Phys. Rev. B **65**, 075306 (2002).
[3] F. Findeis *et al.*, Phys. Rev. B **63**, 121309(R) (2001).
[4] J. J. Finley *et al.*, Phys. Rev. B **63**, 161305(R) (2001).
[5] N. H. Bonadeo *et al.*, Science **282**, 1473 (1998).
[6] H. Htoon *et al.*, Phys. Rev. Lett. **88**, 087401 (2002).
[7] T. H. Stievater *et al.*, Phys. Rev. Lett. **87**, 133603 (2001).
[8] H. Kamada *et al.*, Phys. Rev. Lett. **87**, 246401 (2001).
[9] A. Zrenner *et al.*, Nature (London) **418**, 612 (2002).
[10] E. Dekel *et al.*, Phys. Rev. Lett. **80**, 4991 (1998).
[11] M. Bayer *et al.*, Nature (London) **405**, 923 (2000).
[12] A. Vasanelli *et al.*, Phys. Rev. Lett. **89**, 216804 (2002).
[13] A. Hartmann *et al.*, Phys. Rev. Lett. **84**, 5648 (2000).
[14] At higher intensities, carriers are created in the wetting layer by absorption in the continuum background [15] and neutralize the charge complex. Alternatively, an excited charged exciton can decompose into an s exciton by ejecting the charge carrier from the QD through an Augerlike process [16], leaving X emission.
[15] C. Kammerer *et al.*, Phys. Rev. Lett. **87**, 207401 (2001).
[16] M. Brasken *et al.*, Phys. Rev. B **58**, R15 993 (1998).
[17] J. J. Finley *et al.*, Phys. Rev. B **63**, 073307 (2001).
[18] D. V. Regelman *et al.*, Phys. Rev. B **64**, 165301 (2001).
[19] Ph. Lelong *et al.*, Solid State Commun. **98**, 819 (1996).
[20] R. Warburton *et al.*, Phys. Rev. B **58**, 16 221 (1998).
[21] O. Verzelen *et al.*, Phys. Rev. Lett. **88**, 146803 (2002).
[22] H. M. Gibbs *et al.*, Phys. Rev. A **8**, 446 (1973).
[23] G. A. Narvaez *et al.*, Phys. Rev. B **61**, 13 753 (2000).