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Excited States in optically-gated charged single InAs quantum dots

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Using a micro-spectroscopy technique, resonant excitation of the excited states of single neutral and charged InAs quantum dots are observed. Photo-depletion is used to efficiently control the presence of excess carriers in the quantum dot. Due to their different Coulomb energy shifts, the charged and neutral states of the same quantum dot can be selectively excited. Photoluminescence excitation spectroscopy allows to compare the structure of the excited sates in neutral and charged quantum dots. Time-resolved coherent spectroscopy shows that the dephasing time of the excited states is longer when the quantum dot is charged.

The atomic-like optical properties of semiconductor quantum dots (QDs) make these structures attractive for the realization of quantum devices such as solid-state quantum logic gates [1, 2]. The development of techniques to control Coulomb interactions [3, 4] and to perform coherent manipulation of carrier wavefunctions [5, 6] (i.e. on a time scale shorter than the dephasing time) are key points for a successful implementation of quantum information processing in these solid state systems and the development of QD based quantum information systems.

In this paper we show how we can optically control the charge state of a single QD and coherently manipulate the confined wave function exploiting quantum interferences. We use confocal micro-spectroscopy to study the optical properties of single self-assembled InAs/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 ground state emission under resonant excitation. Wavepacket interferometry is performed on the different excited states of these complexes.

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. For the 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 different individual QDs are presented in Fig. 1. The *s*-shell emission of QD1 obtained under *non-resonant* excitation of the GaAs barriers and *resonant* excitation below the wetting layer are shown (expanded) in Fig. 2. The linear and the quadratic power dependence

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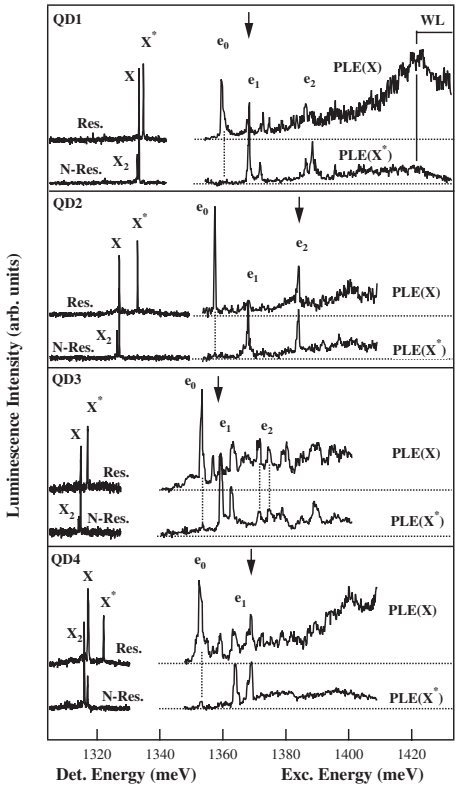

Fig. 1 PL and PLE spectra for four single QDs (labeled 1, 2, 3, 4). PL spectra are obtained under resonant (excitation energy indicated by arrows) or non-resonant ($\lambda_{exc} = 532$ nm) excitation. PLE spectra are obtained for detection at the energies of X and X^* lines observed in resonant PL.

of X and X_2 under non-resonant excitation [Fig. 2 (left inset)] identify these lines as the recombination of the exciton and biexciton in the same QD. 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. The PLE spectra of each *s*-shell PL line is then extracted from the obtained two dimensional data. The structure of the PLE [Fig. 1] shows a clear correlation between X and X^* detection, an observation consistent with both features arising from the *same* QD. Many QDs were investigated, all of which show rather similar behavior.

The X PLE spectra present different sharp absorption lines on top of a continuum background related to spatially-indirect transitions involving interface states of the wetting layer usually observed in this type of QDs [7]. 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 (labeled e_0) is first observed in the PLE spectra of X. PL of X^* is not observed at this excitation wavelength. A second sharp absorption (e_1) is then observed for X^* at slightly higher energy. e_1 presents a doublet structure in some of the QDs (e.g. QD3, QD4). A signal, usually smaller, is also observed for X at this excitation energy e_1. Absorption resonances at higher energy give both a luminescence of X and X^*. The

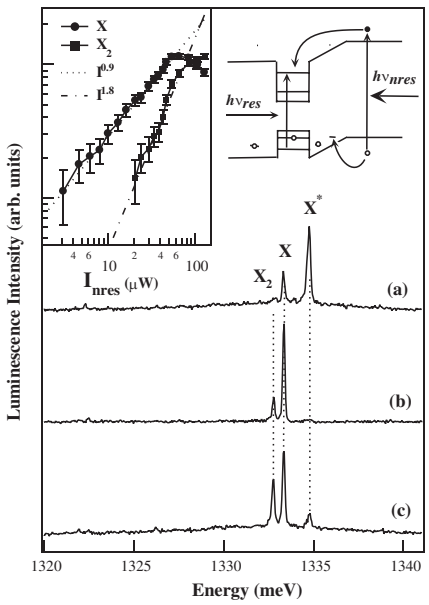

Fig. 2 QD1 PL for resonant excitation (a) on e_1 ($\lambda_{exc} = 906.5$ nm), non-resonant exciation (b) ($\lambda_{exc} = 532$ nm), and simultaneous resonant and non-resonant excitation (c). Inset: (left) Intensity of X and X_2 as a function of non-resonant excitation intensity. (right) Schematic illustration of the photo-depletion mechanism.

PLE background of the exciton gradually increases till the threshold of the wetting layer absorption ($\approx$1420 meV). Only a small absorption background is observed in the PLE of X^*.

The exciton and the biexciton are clearly identified in the non-resonant PL spectra. The additional line X^* arising from the *s*-shell of the QD comes from the recombination of a charged excitonic complex. Even if the sample is not intentionally doped, the transfer of carriers from a residual background doping impurities can introduce excess charges in the QDs [8]. In order to understand why X^* is not observed in the non-resonant spectra, micro-PL experiments were carried out under *simultaneous* resonant and non-resonant excitation. Spectra obtained on QD1 under resonant, non-resonant and both resonant and non-resonant excitations are compared in Fig. 2. No trace of X^* is observed in the non-resonant spectra (Fig. 2b) showing that X^* is exclusively produced by resonant excitation on an excited state of the QD. With resonant excitation on e_1, the transitions X and X^* dominate the spectra (Fig. 2a). However, when carriers are simultaneously created in the GaAs barriers by non-resonant excitation, X^* progressively disappears as the contribution of the biexciton increases (Fig. 2c).

This spectral evolution is characteristic of photo-depletion [8]. This mechanism is illustrated in the right inset of Fig. 2 in the case of a positively charged QD. High-energy photo-excited $e-h$ pairs are dissociated in the space-charge region between ionized acceptors and the charged QDs. Electrons are attracted by the positively-charged QD and recombine with the QD hole. Thus photo-depletion removes holes from the QD at a rate proportional to the absorbed non-resonant excitation, and produces PL only at X. As long as singly charged QDs are considered, this depletion mechanism prevents us from observing the charged exciton under non-resonant excitation. The first captured carrier neutralizes the QD and recombine with the excess charge: the emission of the exciton is observed. The recharging process (transfer of a carrier from an impurity to the QD) is expected to be very slow compare to the radiative lifetime of the exciton. Consequently, only neutral species can be generated. For excitation below the wetting layer and at low excitation densities, this photo-depletion mechanism is turned off and a charged exciton can be created in the QD. We show here that resonant optical excitation is capable of selectively pumping a single QD depending on its charge state, a necessary component to quantum gates. With an extra carrier in the QD ground state, resonant *p*-shell excitation can only occur 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.

Currently we are unable to directly prove whether X^* is positively- or negatively-charged, but in these kind of 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 [9]. Thus we henceforth take X^* to be recombination of a positively-charged exciton (although this is not crucial for our discussion). 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 blue-shifted above X, $h - h$ repulsion in the ground state dominates $e - h$ attraction [3, 10].

The PLE spectra allow us to directly measure the energy shift between *p*-state *absorptions* of X and X^*, $\Delta V_p = \omega(e_1) - \omega(e_0) = 5 - 12$ meV. In the simplest approximation, this energy difference arises from the Coulomb interaction (direct and exchange) between a *p*-state exciton and an *s*-state charge 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. From a simple calculation, in a symmetric parabolic potential we also expect a splitting of the *p*-states of the charged QDs due to the Coulomb exchange energy of the holes [10]. In some of the QDs, the PLE absorption e_1 presents a doublet structure with a splitting $\Delta V_{ex} = 3 - 5$ meV. The amplitude of the two components of the doublet can be different (see QD1) and this doublet structure is not systematically observed (see QD2). The PLE signal not only depends on the oscillator strength of the excited state but also on the relaxation rate to the ground state. As the energies of the studied excited states are in the vicinity of the energy of the LO phonon (36 meV), the energies of these levels as well as their relaxation rate are strongly influenced by the coupling with LO phonons [11, 12]. Different coupling with LO phonon could be responsible for the difference in the amplitude of the components of the exchange doublet.

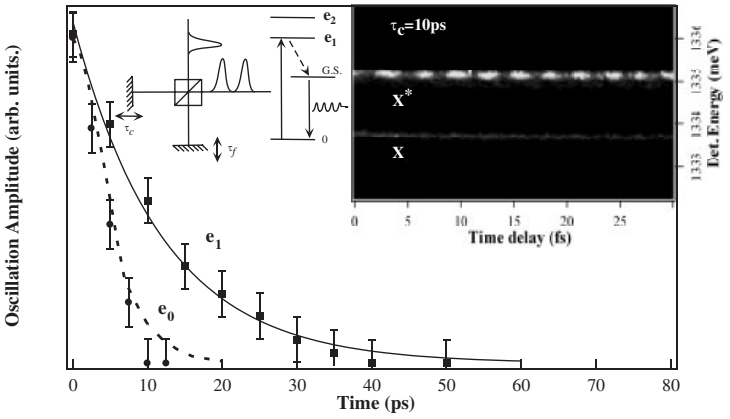

Fig. 3 Amplitude of the oscillations in the PL of QD1 as a function of delay τ_c for excitation at e_0 (circles) and e_1 (squares). The dotted line corresponds to the autocorrelation of the excitation pulses. Inset: (left) schematic experimental set-up, (right) quantum interference map for QD1 PL at $\tau_c = 10$ ps under resonant excitation of e_1, shown as a function of emission energy and pulse fine delay τ_f.

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 wavepacket interferometry. Similar experiments were first reported for the coherent manipulation of the excited states of interface-localized excitons in narrow GaAs QWs [5]. 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 sub-wavelength stable Michelson interferometer with a coarse (τ_c) and a fine (τ_f) delay adjustment [Fig. 3 (inset)]. The quantum interference between the wavefunctions created by the two phase-locked pulses gives rise to sinusoidal oscillations at the optical frequency of the excited state population as a function of time delay. The oscillation of the excited state population can be monitored through the PL of the ground state of the QD [5, 6].

The map in Fig. 3 (inset) shows the resulting interferogram in the PL of QD1 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 amplitude of the oscillations as a function of pulse delay τ_c [Fig. 3] show that exciting e_1 results in an exponential decay with a fast dephasing time of $\sim$12 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 set-up.

These dephasing times are mainly controlled by the relaxation rate of the carrier to the ground state of the QD. With an energy difference between the ground state and the first excited state in the energy range of the bulk GaAs LO phonon (36 meV), 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 [13]. If an additional carrier is introduced in the ground state of the QD, the relaxation rate is reduced due to the Pauli exclusion principle, as well as Coulomb-induced perturbations in the wavefunction overlap of *s* and *p* excitons. Although more detailed theory is to be developed, these simple arguments suggest the lifetime and hence the dephasing time of the *p*-state exciton increase for a charged QD, in agreement with our measurements.

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