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Shape-Dependent Exciton Dynamics in InGaAs/GaAs Quantum Dots

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The influence of local charge separation in self-organized In(Ga)As/GaAs quantum dots (QDs) on the exciton dynamics under resonant excitation of the confined exciton in the first excited state is investigated by time-resolved photoluminescence spectroscopy. The oscillator strength and the relaxation rate are shown to depend oppositely on the charge separation, which again depends on the shape and composition profile. A pronounced phonon bottleneck effect is demonstrated for excitons in flat, truncated QDs, whereas recombination-limited dynamics are observed for QDs favouring local charge separation. The results point out possible pathways for optimizing such QDs for device applications.

Introduction The coherent, defect-free nature of self-organized In(Ga)As/GaAs quantum dots (QDs), for a recent review, see [1] enables investigations of the *intrinsic* electronic properties of QD structures, which is particularly crucial for studies of the carrier/exciton dynamics. Nevertheless, neither the radiative recombination probability nor the relaxation processes are unambiguously understood yet. On the one hand, widely varying photoluminescence (PL) decay times ranging from a few hundred picoseconds to ~2 ns have been reported [2–5] and, on the other hand, fast population of the ground state by non-resonantly excited carriers is observed in contradiction to the phonon bottleneck based on the week coupling assumption [6]. Recently, the polar exciton–LO-phonon interaction was proposed to be sufficient to make the formation of vibronic or polaron states important [7], accounting for intradot relaxation on a time scale of a few picoseconds [8].

In this paper we demonstrate that the structural properties, in particular the shape and composition profile, of self-organized In(Ga)As/GaAs QDs strongly affect the *exciton* dynamics. The low symmetry and inhomogeneous strain provides in general for distinctively different electron and hole wave functions [9] and enables one to engineer the electron–hole overlap, i.e. the oscillator strength, as well as the local charge density, i.e. the polar exciton–LO-phonon coupling strength. The ability to modify the structural properties via the growth conditions promises control of the electronic and optical properties of excitons confined in such QDs. For example, the excited state spectrum [10], the permanent dipole moment [11], and the polar exciton–LO-phonon interaction [12] of the confined excitons could be shown to depend on the actual structural properties.

Experimental To investigate the effect of the structural properties on the exciton dynamics, distinctly different samples have been investigated: pyramid-like InAs/GaAs QDs grown by molecular beam epitaxy [10, 12, 13], which have a base length of about

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17 nm and a ground state transition energy of 1.067 eV; and In-rich flat, truncated In(Ga)As/GaAs QDs [14, 15] grown by metalorganic chemical vapour-phase epitaxy, which have typically a width and height of 20 and 3 nm, respectively, and a ground state transition energy of about 1.0 eV, and an annealing series characterized by a systematically changing In distribution [14, 16]. Time-integrated PL spectra were recorded with a tungsten lamp dispersed by a double-grating monochromator as excitation source and a cooled Ge diode for detection. Time-resolved PL (TRPL) was excited in resonance with the first excited state transition I1 by spectrally narrow (1 meV) picosecond pulses of an optical parametric oscillator pumped by a picosecond Ti:sapphire laser. The luminescence was dispersed by a subtractive double-grating monochromator and detected with a multi-channel plate photomultiplier with an S1 cathode in photon-counting mode. The system response with a full width at half maximum (FWHM) of ~20 ps was taken into account in the analysis of the transients using convolution techniques.

Results and Discussion Figure 1 depicts PL transients of the as-grown InAs and InGaAs QDs excited resonantly at I1, showing distinctively different exciton dynamics. The pyramid-like InAs QDs show a fast rise (150 ps) and a slow decay (920 ps) as reported previously [17]. Detailed optical studies indicate recombination-limited exciton dynamics [10], i.e. relaxation faster than recombination, allowing the excitons to reach a thermal equilibrium distribution. The PL of the truncated InGaAs QDs decays much faster (375 ps) and peaks with a delay of ~500 ps, corresponding to a rise time of 370 ps. Note that the rise time of the ground state PL presents only a lower limit for the actual relaxation time due to the competition with direct radiative recombination of the excited state. Further optical investigations suggest that the observed rise time is indeed close to the excited state lifetime and that the relaxation time is of the order of several nanoseconds.

The time-integrated optical properties of a similar sample are shown in Fig. 2a. In low-temperature PL the ground state transition peak at 0.999 eV is accompanied by a weak (~16%) high-energy shoulder at 1.064 eV, which is attributed to the recombination of hot excitons in the first excited state. Indeed, the PL excitation (PLE) spectrum has a minimum at the energy of the first excited state transition I1, demonstrating unambiguously suppressed exciton relaxation, i.e. a phonon bottleneck. At 200 K the PL and PLE spectra are redshifted by 38 meV and the I1 absorption

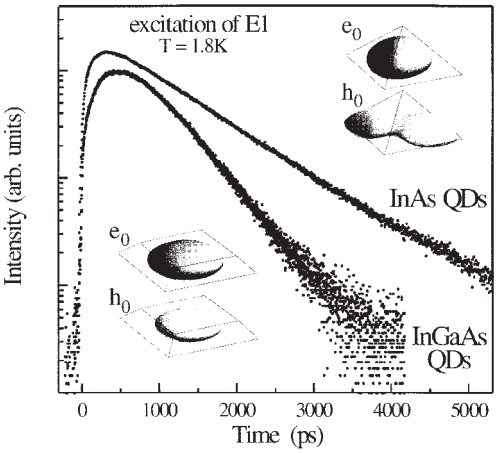

Fig. 1. PL transients of pyramid-like InAs/GaAs and flat, truncated InGaAs/GaAs QDs exciting resonantly the first excited state I1. The insets depict the electron and hole components of the exciton ground state in full and truncated pyramidal InAs/GaAs QDs calculated according to Ref. [9]

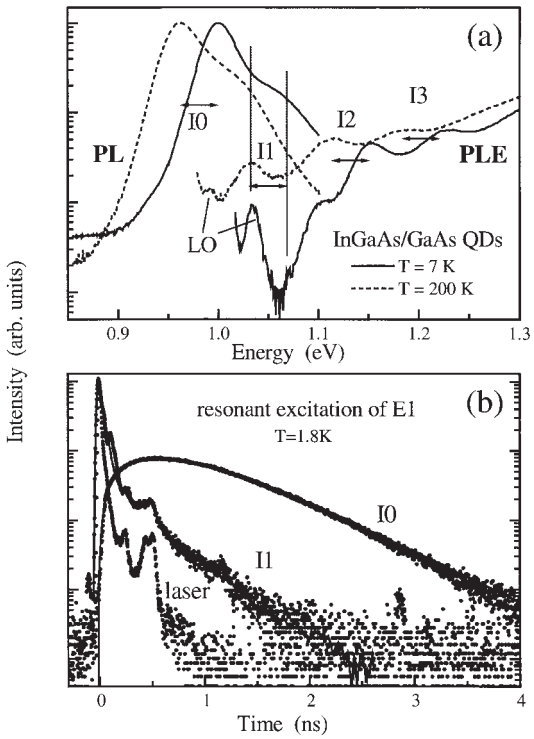

Fig. 2. a) PL and PLE spectra of a typical InGaAs QD sample. The solid and dotted lines represent spectra recorded at 7 and 200 K, respectively. b) Transients of the ground (I0) and first excited (I1) state PL upon resonant excitation of excitons in the first excited state

leads to a prominent excitation resonance 71 meV above the ground state transition (dotted line). Note that the transition spectrum might be more complex owing to the actual structural properties of the strained QDs [9, 10]. The fine structure is, however, masked by the inhomogeneous broadening and will be neglected in the following. These results point to suppressed relaxation in the QDs at low temperatures, which prevents population of the ground state exciting resonantly the first excited exciton state. As shown in Fig. 2b, time-resolved experiments allow one to measure the resonant PL of I1. Note that the decay time of 430 ps corresponds, within experimental uncertainty, to the rise time (480 ps) of the ground state PL indicating the resonantly excited I1 state to be the bottleneck in the relaxation to the ground state [5]. The previous observation of multi-LO-phonon resonances in PLE spectra of the ground state PL suggests indeed a single-step exciton relaxation process [4]. A fit of the temperature-dependent relaxation yield (from PLE) as well as the I1 lifetime in a three-level exciton model yields a low-temperature relaxation time of $\sim$8 ns and an activation energy of 33 $\pm$ 4 meV, corresponding to the LO-phonon energy [18]. The results indicate that inelastic phonon scattering is the dominant relaxation process between the confined exciton states and they place an upper limit on the probabilities of competing Auger or Coulomb scattering processes.

Figures 1 and 2 give experimental evidence for a pronounced influence of the QD shape on the interaction of the confined excitons with photons and phonons. The insets of Fig. 1 show the electron and hole components of the ground state exciton in pyramidal and flat, truncated pyramidal QDs, respectively, calculated based on the eight-band $\mathbf{k} \cdot \mathbf{p}$ model described in Ref. [9]. The shape asymmetry and piezoelectric potential spatially separate the electron and hole in such strained QDs, where the extent depends strongly on the shape: the effect is pronounced for a pyramidal but strongly quenched for a flat shape. Thus, the ground state lifetime is expected to be shorter (large overlap integral) in the flat InGaAs QDs, whereas the stronger polar exciton–LO-phonon interaction (large local charge density) should provide for faster relaxation in the pyramidal InAs QDs, as indeed is observed in the experiments. Depending on the QD shape, relaxation- or recombination-limited exciton dynamics are observed.

To support the interpretation given, parts of the InGaAs/GaAs sample of Fig. 1 were treated to ex-situ annealing, altering the confinement potential in a somewhat controlled way by In/Ga interdiffusion. The interdiffusion leads to a blue shift of the ground state transition and decreases the excited state splitting as observed in high-density PL experiments (Fig. 3a) [14, 16]. The increasing Ga content increases the potential within the QDs and the increasing effective size of the QDs decreases the quantum confinement. TRPL measurements under resonant excitation of I1 show a systematic variation of the relaxation and recombination dynamics with the annealing temperature, i.e. the degree of In/Ga interdiffusion (Fig. 3b). The radiative decay time of the ground state transition I0 increases with the annealing temperature, becoming twice as long for annealing for 30 min at 700 °C.

The radiative lifetime decreases with increasing effective QD size, which is opposite to the size dependence predicted for highly symmetric QDs [19, 20]: the oscillator strength scales with the localization (i.e. the QD) volume in the weak confinement regime and becomes size independent only for strong confinement, when the quantization energies dominate the Coulomb interaction, resulting in an atomic-like situation.

A comparison of the single-particle and exciton states in pyramidal InAs QDs [10] suggests that the self-organized In(Ga)As QDs investigated are in the strong confinement regime. In this case, the bright exciton lifetime is given by

$$\frac{1}{\tau_{\mathrm{rad}}} = \frac{8}{3}\,\alpha\,\sqrt{\varepsilon}\,\omega\left(\frac{P_{\mathrm{CV}}}{\hbar c}\right)^2 I\,, \qquad (1)$$

where α is the fine structure constant, ε the dielectric constant of the matrix, ω the frequency of the emitted photon, P_{CV} the optical interband matrix element describing the contribution of the Bloch functions, and I the overlap integral defined by the envelope functions of the confined states. Since in the energy range investigated $\sqrt{\varepsilon}$ is practically constant (~3.5 for GaAs), the variation of the radiative lifetime is caused by

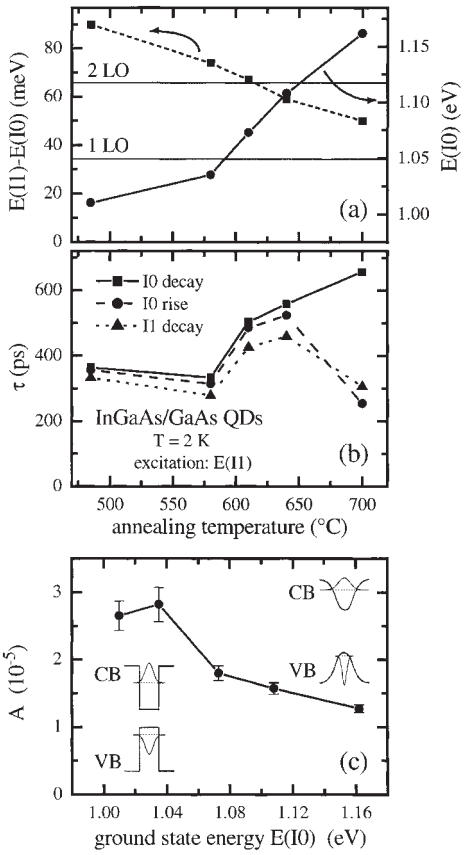

Fig. 3. Properties of ex-situ annealed InGaAs/GaAs QDs: a) the ground state transition energy and the I1/I0 splitting derived from high-density PL spectra; b) the rise and decay times of I0 and I1 upon resonant excitation of I1; c) the dimensionless factor $A = (P_{\mathrm{CV}}/\hbar c)^2 I$ being proportional to the oscillator strength. The insets illustrate schematically the effect of the annealing-induced smoothing of the confining potential on the electron and hole localization

changes of the transition energy, of the optical interband matrix element P_{CV}, and of the electron–hole overlap integral I. The annealing-induced blue shift of the ground state transition (Fig. 3a) contributes to a decrease in the radiative lifetime of ~20%. Thus, the observed increase of the radiative lifetime is the result of P_{CV} (being almost independent of the composition) and I changing with the structural properties of the QDs. Figure 3c shows that the dimensionless quantity $A = (P_{CV}/\hbar c)^2 I$ is proportional to the oscillator strength. Obviously, the increase of the radiative lifetime upon annealing is caused by a decreasing overlap of the electron and hole wave functions.

For the annealing series the smoothing of the QD potential by In/Ga interdiffusion during ex-situ annealing provides a qualitative explanation for the observed trend, as shown schematically in the insets of Fig. 3c. For the truncated as-grown QDs the sharp chemical potential step in the growth direction leads to similar electron and hole localization (left inset of Fig. 3c) and, thus, a large overlap integral I, consistent with the observed short lifetime of 370 ps. The extension of the ground state wave function in a confinement potential softened by In/Ga interdiffusion depends on the effective mass of the confined carrier and, thus, is different for electrons and holes. For the samples investigated annealing is expected to decrease the electron–hole overlap and to enhance the local charge density, i.e. the polar exciton–LO-phonon coupling. Indeed, for the highest annealing temperature (700 °C) the I1 lifetime becomes significantly shorter than that of I0, suggesting the transition from relaxation- to recombination-limited dynamics. Obviously, the recombination and relaxation (between I1 and I0) rates show opposite dependencies on the electron–hole overlap integral.

Conclusions A significant influence of the shape- and composition-dependent electron–hole overlap on the exciton dynamics is demonstrated for self-organized In(Ga)As/GaAs QDs. Local charge separation in pyramidal QDs favours recombination-limited dynamics, whereas the quenched exciton–LO-phonon coupling in flat, truncated QDs suppresses exciton relaxation between I1 and I0, leading to a pronounced phonon bottleneck effect. The results indicate unambiguously the effect of the complex confining potential of strained self-organized QDs on the exciton dynamics, pointing out possible pathways to optimize such QDs for device applications, e.g. in view of a large saturation gain and modulation band width.

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