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Tuned exciton kinetics in self-organized InGaAs/GaAs quantum dots

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Abstract

The exciton dynamics in self-organized InGaAs/GaAs quantum dots with systematically varying structural properties is investigated by time-resolved photoluminescence spectroscopy. Decreasing the lateral confinement by ex situ annealing increases the exciton lifetime and accelerates intradot relaxation. The altered exciton dynamics is attributed to the decreasing electron–hole overlap in the annealed QDs, resulting from the smoother confinement potential. The results demonstrate the effect of the actual confinement potential on the exciton–photon and exciton–phonon interactions, pointing out possible pathways to device optimization. © 2002 Published by Elsevier Science B.V.

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Within the last decade quantum dots (QDs) attracted continuously increasing interest due to their unique optical properties. The coherent nature of defect-free self-organized QDs suppresses the interaction with surface and defect states, which often mask the intrinsic properties [1]. This is particularly crucial for the investigation of the carrier/exciton dynamics. The oscillator strength of excitons in QDs is predicted to show a characteristic size dependence, scaling with the localization (i.e. the QD) volume and saturating for small QDs [2]. Nevertheless, realistic calculations based on pseudo-potential [3] and 8 band *kp* [4] theory revealed a strong shape dependence. Additionally, the composition of the QD is attributed to alter the excitonic carrier wavefunctions in small QDs. Thus, annealing induced systematic changes of the confinement potential reveal new insights into the exciton dynamics in zero-dimensional structures.

Self-organized QDs formed in the In(Ga)As/GaAs system provide strong quantum confinement, i.e. the size of the QDs (typically 20 nm width by 3–10 nm height) is sufficiently small for confinement energies to exceed Coulomb energies. For such QD structures a variety of time-resolved photoluminescence (TRPL) measurements are reported [5,6]. The observed spread of the decay time ranging from some hundred picosecond up to several nanosecond is not understood yet. Up to now, quantitative descriptions of the dynamics are limited to idealized dots, assuming a complete overlap of the electron and hole wave functions within the structure [2]. The exciton lifetime is expected to scale reciprocal to the QD volume $V/(\pi a_B^3)$ [7]. Thereby, the effect of inequivalent electron and hole wavefunctions in low-symmetry strained QDs and, thus, the actual structural properties have been neglected.

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In this paper, we present resonantly excited TRPL experiments on a series of annealed self-organized InGaAs/GaAs QDs with systematically changing structural properties. The investigated samples were derived from a single sample by ex situ annealing as described in detail in Ref. [8]. The original ('as-grown') dots were fabricated by MOCVD on a GaAs (0 0 1) substrate depositing five monolayers of $In_{0.5}Ga_{0.5}As$ and capping them with GaAs at 485°C. Transmission electron microscope images revealed $\sim 10^9$ QDs/cm^2 with a flat, truncated shape. Different pieces of the sample were annealed at 580°C, 610°C, 640°C, and 700°C. The sharp potential steps, especially in the growth direction, are smoothed by the thermal treatment and the localization decreases for both electrons and holes. The averaged Ga content of the QDs increases leading to an increasing ground state transition energy [8,9]. Simultaneously the effective QD size increases [10]. With decreasing localization an increasing influence of the respective effective carrier mass is expected, leading to a decreasing overlap of the electron and hole wavefunctions, with an extension reciprocal on the respective effective mass.

Photoluminescence (PL) spectra (Fig. 1) excited by an cw Ar^+-laser at 2.4 eV with an excitation density of ~ 5 kW/cm^2 reveal ground (I0) and excited state transitions (I1, I2). The transitions exhibit a blueshift with annealing, which is attributed to the increasing Ga content inside the QDs. Simultaneously, the energy spacing of adjacent transitions decreases mirroring the decreasing quantization due to the increasing effective QD size.

The downwards pointing arrows in Fig. 1 denote the excitation energies used for the TRPL measurements whereas the upwards pointing arrows mark the corresponding detection energies. Fig. 2 shows typical transients excited resonantly to I1 and recorded at the denoted transitions. The dots represent the measurements while the solid lines are fits using an exponential rise and decay, respectively, folded with the system response (dubbed laser). The investigated flat QDs exhibit a fairly fast (~ 350 ps) recombination of the ground and first excited state transition. Additionally, the decay of the first excited state transition is mirrored by the rise of the ground state transition. The

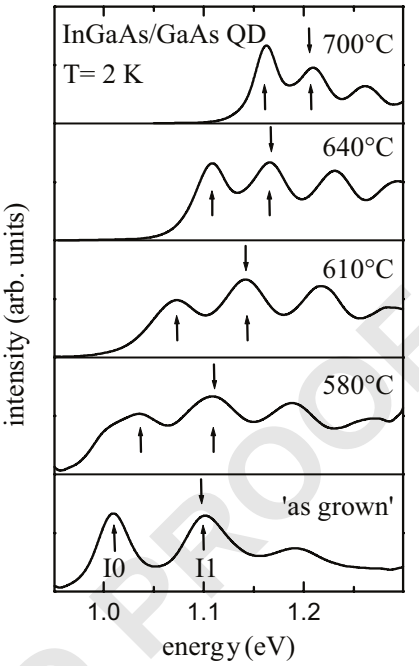

Fig. 1. High excitation density PL spectra of the annealed quantum dot samples. The ground (I0) and first excited (I1) state transitions are indicated by upward arrows, denoting the detection energies for the TRPL measurements, while downward arrows mark the respective excitation energies.

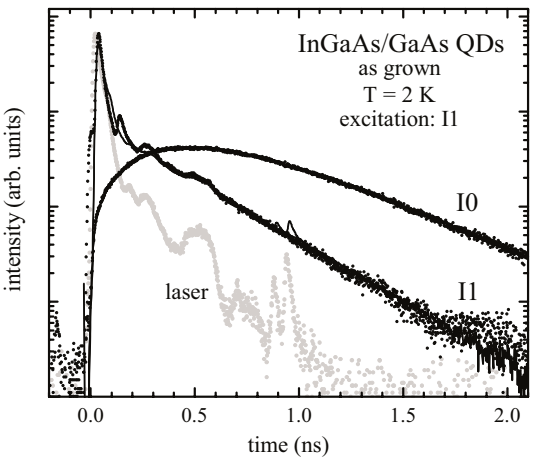

Fig. 2. TRPL transients of I0 and I1 for the 'as grown' sample excited resonantly to the first excited state transition I1. Dots represent the measured transients while solid lines denote fits.

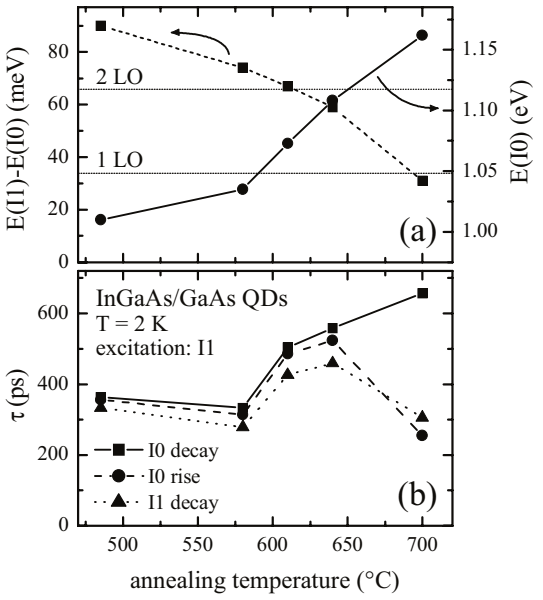

Fig. 3. The annealing-induced structural changes lead to an increase of the ground state transition energy while the substate splitting decreases (panel (a)). The lines 1LO and 2LO correspond to the energy of 1 and 2 phonons compared to the substate splitting (left scale). Contrary to a predicted decrease of the lifetime (increasing QD size) the heat treatment induced In/Ga interdiffusion leads to an increase of the lifetime by a factor of 2.

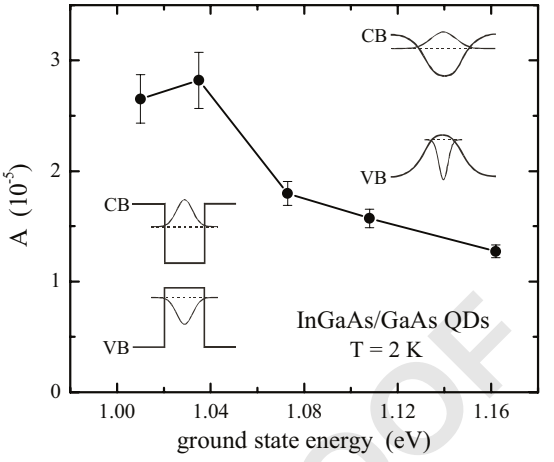

Fig. 4. Dimensionless quantity A, proportional to the oscillator strength, derived from the measured ground state lifetime. The insets indicate qualitatively the decreasing overlap of electron and hole wavefunctions caused by the annealing-induced smoothing of the confinement potential.

slow rise time of I0 indicates a suppressed relaxation due to a negligible phonon coupling as reported recently [6].

The time constants shown in Fig. 3(b) were derived from transients excited resonantly in I1 while detecting at I0 and I1, respectively, whereas the corresponding ground state transition energy and substate splitting are depicted in Fig. 3(a). Obviously, the exciton dynamics is dominated by the actual confining potential. The lifetime of I0 increases in the annealed samples. That of I1 behaves similar up to 640°C. At higher annealing temperatures the decay of I1 accelerates becoming finally twice as fast as that of I0. Interestingly, the radiative lifetime increases with increasing effective QD size, whereas commonly the opposite trend is expected [2]. Furthermore, we find the rise of I0 to mirror decay of I1 in all samples indicating an excitonic relaxation [6].

The bright exciton lifetime in the strong confinement limit is proportional to $1/(EP_{cv}^2 I)$ [5] where E is the transition energy, P_{cv} the optical interband matrix element representing the contribution of the Bloch-functions and I the overlap integral of electron and hole envelope functions. As shown in Fig. 4 the product $A \sim (P_{cv}^2 I)$, being proportional to the oscillator strength, decreases significantly upon annealing. Due to the small influence of P_{cv} (for $In_xGa_{1-x}As$ P_{cv} is expected to vary by only 10%) the major effect can be attributed to a decreasing electron–hole overlap. The overlap of excitonic electron and hole wavefunctions is sensitive to the actual confining potential [4,5]. Changes in shape and composition induced, for instance, by annealing allow to tune the overlap of the wavefunctions and, thus, the oscillator strength of strongly confined excitons in self-organized QDs. In consequence an effective pathway for tuning the dynamics in self-organized QDs is provided by optimizing the confining potential.

In conclusion, a significant influence of the electron–hole overlap integral on the exciton dynamics in self-organized InGaAs/GaAs QDs was demonstrated. Ex situ annealing slows down radiative recombination and accelerates relaxation in qualitative agreement with the expected change of the electron–hole overlap caused by In/Ga interdiffusion. The results unambiguously evidence 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.

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