Radiation hardness of InGaAs/GaAs quantum dots

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The interaction between point defects in the matrix and excitons localized in self-organized InGaAs/GaAs quantum dots is investigated for structures irradiated by protons. The exciton ground state is demonstrated to be unaffected by radiation doses up to 10^{14} p/cm^2. The close proximity of radiation-induced defects leads to a strong nonmonotonous temperature dependence of the luminescence yield: Carriers are lost via tunneling from excited quantum dot states to irradiation-induced defects below ~100 K, whereas at higher temperatures, carriers escape to the barrier and are captured by defects.
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Self-organized quantum dots[1] (QDs) have evolved in recent years as promising active media for devices, such as diode lasers,[2] detectors,[3,4] and storage applications.[5,6] The performance advantages result mainly from the confinement-induced quantization of electronic states. Another important advantage results from the localization of the carriers, minimizing the influence of defects in such structures. The direct capture of carriers from the barrier might be strongly reduced by defects, but once localized, tunneling through the potential barrier separating the localized QD states from the defect states would be necessary. Such tunneling processes have been invoked to explain the observation of multiphonon assisted relaxation[7] or to predict fast defect-mediated relaxation.[8] Although enhanced defect formation in the vicinity of self-organized QDs was observed in capacitance measurements,[9,10] the influence of defects on the optical properties of QDs has not been analyzed in detail, yet.

Irradiation with, for example, protons provides a systematic *ex situ* means to generate defects in self-organized InGaAs/GaAs QD structures. The proton-induced defects generate deep levels with a volume density approximately proportional to the dose, allowing to control the average separation between the QDs and defects. Proton irradiation of GaAs generates various deep electron and hole traps associated with vacancies and interstitials as well as complexes thereof.[11–13] For In(Ga)As/GaAs QDs irradiation with electrons or protons is found to decrease the photoluminescence (PL) intensity, although the QD PL withstands an about one order of magnitude higher dose than comparable quantum wells.[14,15] The improved radiation hardness is reflected in a superior performance of irradiated QD lasers.[16]

Here, we use PL excitation (PLE) and time-resolved PL spectroscopy to investigate the effect of defects on excitons already localized in QDs. The previously observed reduction of the PL efficiency by irradiation-induced defects is demonstrated to result from enhanced nonradiative recombination in the barrier and tunnel-escape from *excited* QD states, that is, a reduced excitation efficiency. The ground state is completely unaffected by doses up to $\Phi = 10^{14}$ p/cm^2, reflecting its radiation hardness.

The samples were grown by metalorganic chemical vapor deposition on GaAs(001) substrates as described in detail in Ref. 17. The InGaAs QDs have an area density of $\sim(3-5)\times10^{10}$ cm^{-2}.[17] The main part of the paper concentrates on sample "A," containing InGaAs QDs covered by an InGaAs QW (see Ref. 18). The QD layer is separated by two AlGaAs diffusion barriers from the surface and the substrate to minimize the impact of carrier diffusion in the matrix. In samples B and C the InGaAs QD layer was incorporated in a p^+n- and a n^+p-diode structure, respectively, for electrical characterization.[19]

The samples were irradiated by high-energy (2.4 MeV) protons with doses between 1×10^{12} p/cm^2 and 1×10^{14} p/cm^2 in a van de Graaff accelerator. Figure 1 shows deep-level transient spectroscopy (DLTS) spectra of the p^+n-diode (sample B) for $\Phi = 1\times10^{12}$ p/cm^2 and 1×10^{13} p/cm^2 and the nonirradiated reference. Proton radiation results in well-known defect levels, marked in Fig. 1. The activation energies obtained by DLTS ($E_A > 280$ meV) are consistent with previous reports,[11,12] and, more important, lie energetically below the electron ground state of the

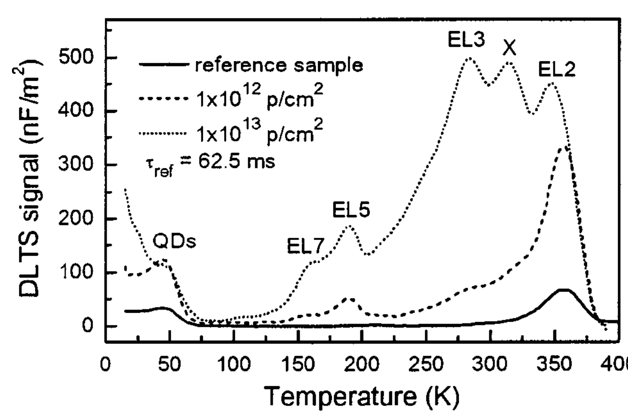

FIG. 1. DLTS spectra of sample B for different radiation doses (peak X could not unambiguously be assigned).

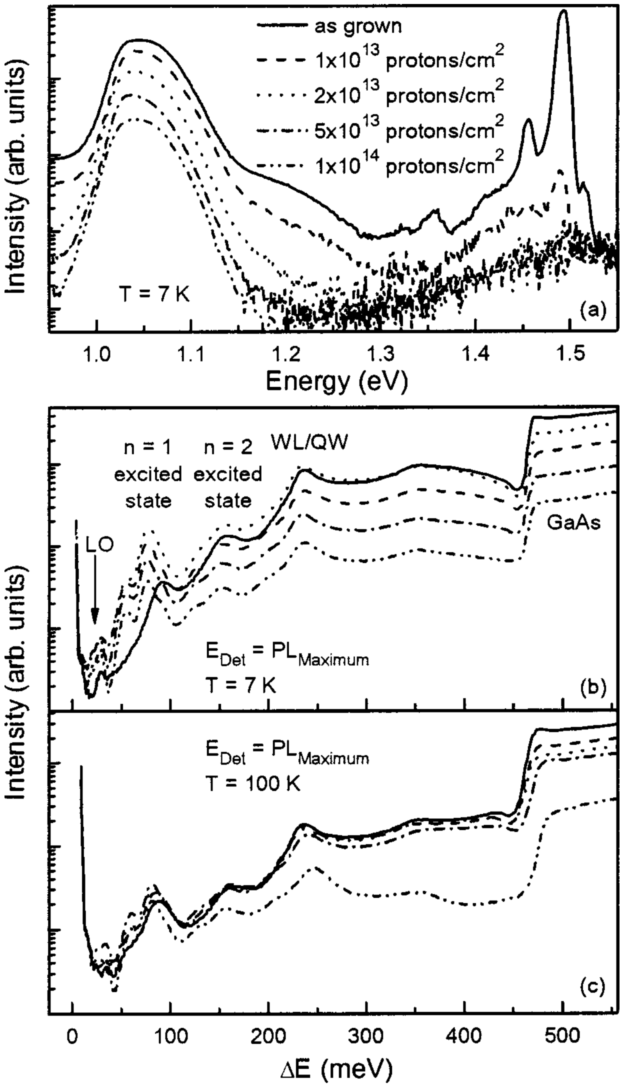

FIG. 2. (a) PL spectra of sample A as function of the irradiation dose. Corresponding PLE spectra of the QD PL are shown for 7 K (panel b) and 100 K (panel c).

QDs. The defect concentrations N_D were estimated probing the GaAs matrix below the QDs to avoid interference with the QD states. Protons with an energy of 2.4 MeV produce in GaAs a homogeneous defect density up to a depth of a few 10 μm. The average defect introduction rate $dN_D/d\Phi$ was extracted from DLTS measurements as $\sim$100 cm^{-1}. The defect concentration for the sample irradiated with $\Phi = 1 \times 10^{13}$ p/cm^2 is already close to the doping concentration of $\sim 4 \times 10^{15}$ cm^{-3} as extracted from $C-V$ measurements and, hence, significantly lowers the free carrier concentration, hampering the electrical characterization at higher doses.

The PL and PLE experiments were performed as described in Ref. 18. Figure 2(a) shows for sample A the effect of proton irradiation on the QD PL, exciting the GaAs barrier nonresonantly. The GaAs PL is almost completely quenched for the lowest applied dose, whereas even at the highest dose of 10^{14} p/cm^2, 10% of the QD PL survives. The observation of the QD ground state PL upon nonresonant excitation requires diffusion of carriers/excitons to the QDs as well as capture and relaxation to the QD ground state. Thus, *a priori*, it is not clear to which extent the various processes involved in the excitation/recombination cascade are affected by the defects, contributing to the intensity loss upon irradiation.

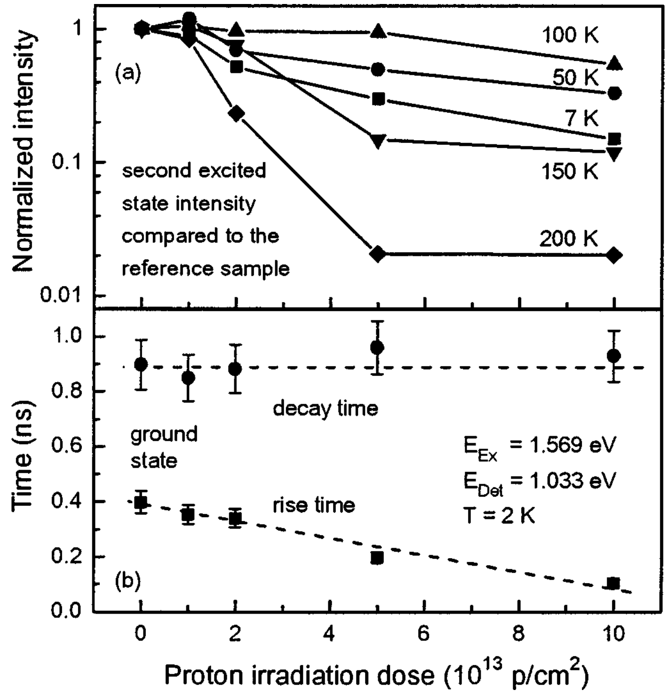

FIG. 3. (a) Intensity evolution in dependence of the irradiation dose for excitation of the second excited state with respect to the reference sample for different temperatures. Panel (b) shows an irradiation dose independent decay time of the ground state transition.

Time-resolved measurements allow probing the excitation and recombination processes of the QD PL separately [see Fig. 3(a)]. The samples were mounted in a He-bath cryostat at 2 K and excited by a ps-Ti:sapphire laser system either above the GaAs band gap or resonantly via excited state transitions. The PL was detected by a multichannel plate photomultiplier with a S1 cathode. Surprisingly, the increasing defect concentration with increasing dose does not affect the lifetime of the ground state exciton, always showing a single-exponential decay. The data suggest tunneling from the QD ground state to defect states to be negligible at the proton irradiation dose used here. However, the generated defects cause the rise time to decrease from 400 to 100 ps. The long rise time in the reference sample is attributed to slowed down intradot relaxation.[20] Carrier migration in the GaAs barrier is negligible for the rise time due to the AlGaAs diffusion barriers reducing the average diffusion length to below ~50 nm. The conclusion is supported by the observation of practically identical rise (and decay) times for resonant excitation into the excited state at 1.140 eV (not shown). The reduced PL rise time and the decreased PL efficiency in the irradiated samples suggest tunneling from excited QD states to defects. The increasing tunneling probability with increasing defect concentration quenches the excitation efficiency and the rise time [Figs. 2(a) and 3(a)].

The crucial role of the slowed-down intradot relaxation is supported by PLE spectra as a function of temperature. At 7 K [Fig. 2(b)] the irradiation-induced defects cause the PL to decrease almost independently of the excitation process, demonstrating that either the first excited state at 1.126 eV or the ground state suffer nonradiative recombination. The situ-

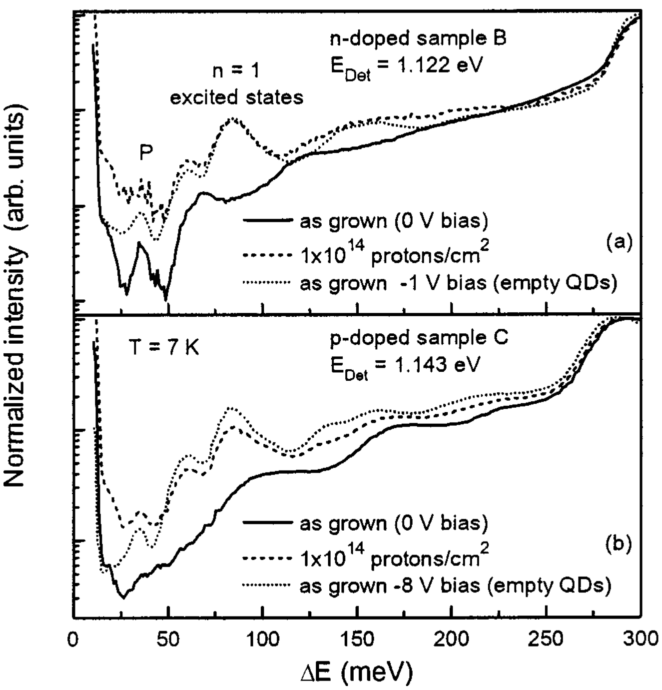

FIG. 4. PLE spectra normalized to the wetting layer peak for sample B [*n*-doped, panel (a)] and sample C [*p*-doped, panel (b)]. The solid/dotted lines show PLE spectra of charged and neutral QDs in the reference sample without and with bias, respectively. The dashed lines show the PLE spectra of the QDs in the irradiated samples.

ation changes dramatically, when the temperature is increased to ~100 K [Fig. 2(c)]. The PL intensity in the irradiated samples is recovered (within ~10%) to the undamaged case. The channel leading to nonradiative recombination is obviously not relevant at elevated temperatures. We attribute this observation to the temperature-dependent competition of intradot relaxation and nonradiative recombination in the first excited state.[7] With increasing temperature intradot relaxation becomes more efficient.[20] Consequently, the probability of relaxation to the unperturbed ground state accelerates, bypassing tunneling to defects. The ground state luminescence intensity upon exciting into the second excited state is shown in Fig. 3(a) as a function of the dose for temperatures from 7 to 200 K. Upon increasing the temperature above 100 K, the intensity decreases dramatically with increasing dose. We suggest that thermal activation into the barrier allows the carriers to reach defect states without tunneling.

Note that, based on typical structural properties of QDs grown under the used condition[21] and the estimated defect introduction rate ~10% of the QDs in the sample with the highest irradiation dose (1×10^{14} p/cm^2) should contain a defect. Those QDs should show no luminescence and, thus, not contribute in our experiment. Unfortunately a 10% decrease of the PL efficiency cannot be excluded or verified from our results.

Finally, we would like to note that an important effect of the irradiation-induced defect formation is a lowering of the free carrier concentration. We demonstrated recently that the charge state of the QDs has a pronounced impact on the PLE spectra, changing the excitation efficiency and renormalizing the excited state transition energies.[21] The effect is shown in Fig. 4 for the diode structures B and C, comparing PLE spectra for zero bias, where the QDs are multiply charged with electrons [Fig. 4(a)] and holes [Fig. 4(b)], respectively, and at a reverse bias where the QDs are neutral. Irradiating the samples with a dose of 1×10^{14} p/cm^2 has the same effect as the reverse bias in the reference samples. The defects lower the Fermi-level compensating the doping and, thus, neutralize the QDs. A similar effect can be observed for the nominally semi-insulating sample A [Fig. 2(b)]. Here, carbon contamination of the AlGaAs diffusion barriers might serve as hole source.

In summary, we have investigated the impact of defect levels generated by proton irradiation up to doses of 10^{14} p/cm^2 on the optical properties of InGaAs/GaAs QDs. The ground state is found to be unaffected by nearby deep defects. The decreasing PL intensity upon irradiation reflects the decreasing excitation efficiency originating from (i) tunneling out of excited QD states due to slow relaxation at low temperatures and (ii) efficient nonradiative recombination in the barrier, when carriers escape thermally from the QDs at elevated temperatures.

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