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Carrier dynamics in particle-irradiated InGaAs/GaAs quantum dots

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The influence of high-energy (2.4 MeV) proton irradiation on the photoluminescence (PL), PL excitation (PLE) and time-resolved PL (TRPL) spectra of InGaAs/GaAs quantum dot (QD) structures is investigated. A pronounced impact of the Fermi level, shifting towards the centre of the gap with increasing irradiation dose, is observed. The results evidence carrier tunnelling out of excited QD states to adjacent defects. It is suggested that for the dose range investigated in the study ($\leq 10^{14}$ p/cm^2) the dots do not contain radiation defects.

1 Introduction Recently, an enhanced hardness of the QD epitaxial structures and fully processed laser diodes against particle irradiation as compared to bulk and 2D counterparts has been found [1–3]. The effect has been attributed to the strong localization of the carriers inside the QDs, preventing them from reaching irradiation-induced non-radiative recombination centres. Nevertheless, the existence of an interaction of the QDs with radiation defects, which leads to a reduction of resonantly excited QD PL intensity, has been concluded [2]. The influence of the radiation damage on the recombination processes in bulk semiconductors has been widely studied during the past decades. On the contrary, almost nothing is known about the microscopic details of the impact of defects on the carrier relaxation and recombination in radiation-damaged QD structures. In this paper, we report investigations of the photoluminescence (PL), PL excitation (PLE) and time-resolved PL (TRPL) spectra in proton-irradiated InGaAs/GaAs QD heterostructures.

2 Experimental The samples used (sample A and sample B) were grown by metalorganic chemical vapour deposition and have the following structure: a GaAs buffer layer was grown on top of a GaAs substrate, the active layer together with GaAs cladding layers were placed between two AlGaAs barriers, with a GaAs capping layer on top of the whole structure. The sample A has an active layer composed by the QD layer, with a dot density of $\approx(3–5) \times 10^{10}$ cm^{-2}, overgrown by a 2 nm $In_{0.25}Ga_{0.75}As$ QW. The sample B has a dot density of $\approx 10^9$ cm^{-2} in an active layer composed by a single QD layer. The irradiation by 2.4 MeV protons with fluences in the range from 1×10^{12} to 1×10^{14} cm^{-2} was performed at room temperature using a Van de Graaff accelerator.

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The TRPL measurements were carried out at 2 K using single photon counting techniques. The excitation was performed by a Ti:sapphire laser system including an OPO, with spectrally narrow (<1 meV) 2 ps pulses, either above the GaAs band gap or resonantly via excited states transitions. The emitted light 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 time resolution of the detecting system was about 20 ps.

The cw PL and PLE experiments were performed in a continuous flow He cryostat between 7 K and 300 K. A tungsten lamp dispersed by a 0.27 m double-grating monochromator served as a low density, tunable light source. The emission was spectrally dispersed by a 0.3 m double-grating monochromator and detected with a cooled Ge diode using lock-in techniques.

3 Results and discussion The irradiation creates defects with deep levels that act as non-radiative recombination centres. The existence of stable point defects created by atomic displacements at room temperature (RT) inside the In(Ga)As QDs or ultra-thin QWs has never been proven. Since the primary defects (vacancies and interstitial atoms) are mobile at RT in GaAs (see, e.g., [4–6]) and, certainly, in InAs, it is very likely that they are captured at the interfaces (cf. Ref. [7]). Moreover, the defects raise the free energy of the crystal, so that it is only natural that the QDs expulse mobile defect components into the matrix. On the other hand, it has been shown that some leakage of the wavefunction into the barrier leads to a carrier escape to adjacent defects, thus deteriorating the QW and QD luminescence intensity [8, 9].

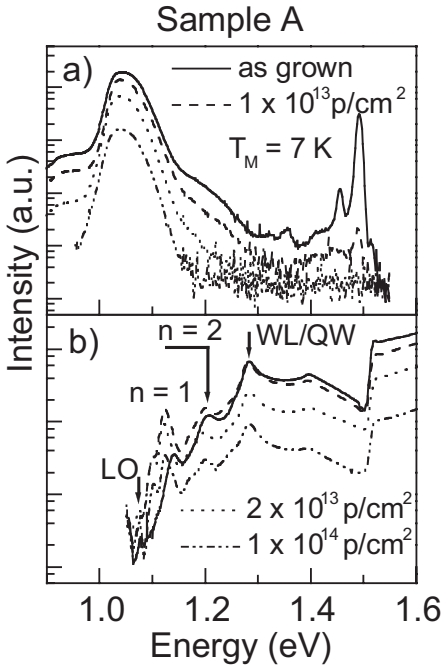

Fig. 1 a) PL and b) PLE spectra of sample A for various proton irradiation doses, measured at 7 K. PLE spectra were recorded at the QD PL maximum.

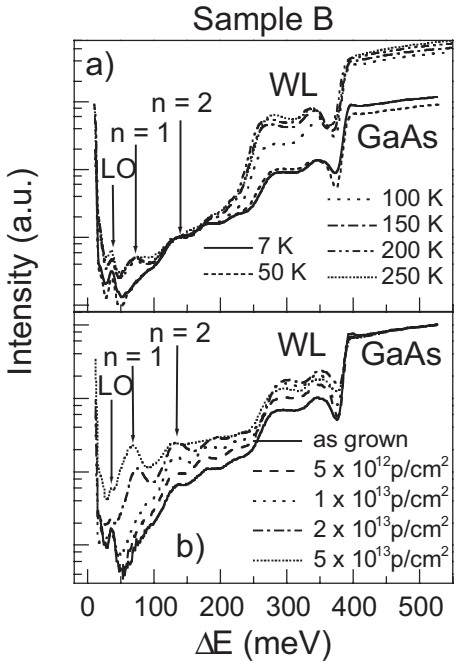

Fig. 2 a) PLE spectra vs. the excess excitation energy $\Delta E = E_{exc} - E_{det}$ of the as-grown sample B taken at different temperatures. Detection was performed at the QD PL maximum. The spectra are normalized to the detection energy and to the second excited state intensity. b) PLE spectra of sample B for various proton doses, recorded at the QD PL maximum at 7 K. The spectra are normalized to the intensity above the GaAs bandgap.

Usually the defects reduce the lifetime of non-equilibrium carriers and, consequently, their diffusion length, thus limiting the carrier supply to the radiative recombination centres and killing the PL intensity. However, because of the presence of the closely spaced AlGaAs barriers, the carrier capture by the QDs in our samples is not diffusion-limited. That is why we do not observe any difference in the quenching factor of the PL intensity at a given irradiation dose for the above-bandgap and below-bandgap excitation for all energies above the $n = 2$ QD excited state (Fig. 1b). Thus, the loss of carriers occurs mainly in the dots themselves. A probable reason of this effect is a tunnelling out of the dots to adjacent radiation-induced non-radiative recombination centres.

Another striking feature of the PLE spectra is the *increase* upon irradiation of the PLE intensity at energies corresponding to the low-lying QD excited states (Fig. 1b and Fig. 2b). The effect is particularly well seen in Fig. 2b (sample B), where the PLE intensities for all doses have been normalized to that of the as-grown sample at energies exceeding the GaAs bandgap, and in the contour plots (Fig. 3). Qualitatively similar effects are observed when the temperature is increased (Fig. 2a) or a reverse bias is applied to a diode structure with the dots located in the space charge region [10, 11]. The radiation defects shift the Fermi level towards the middle of the bandgap, thus emptying the low-lying QD states from spectator carriers and permitting the resonant absorption. In the case of a temperature increase, another important effect is the growth of the phonon density, which accelerates the relaxation and helps overcome the phonon bottleneck [12].

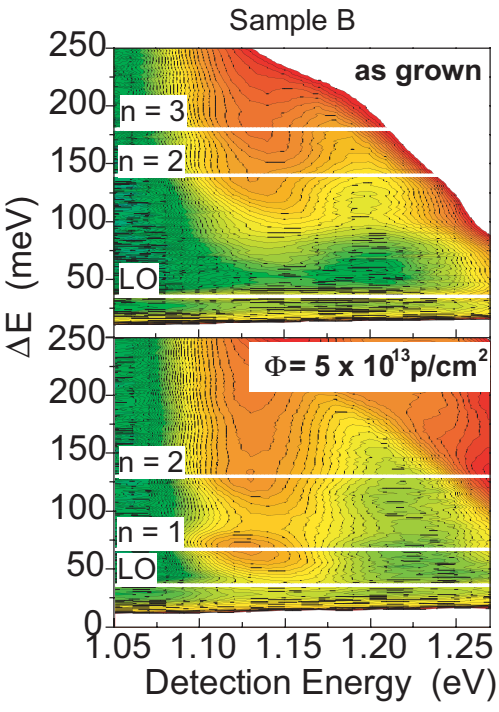

Fig. 3 (online colour at www.interscience.wiley.com) Contour plots of the QD PL intensity as a function of the detection energy and the excess excitation energy $\Delta E = E_{exc} - E_{det}$ for sample B at 7 K. The white lines denote PLE resonances from the excited states and LO phonons.

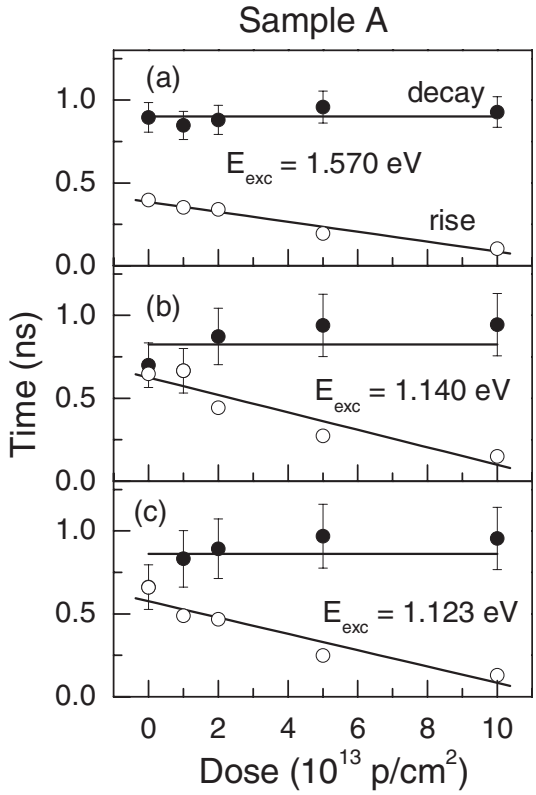

Fig. 4 Rise (open circles) and decay (solid circles) times from TRPL measurements of sample A. The detection energy was at the ground state transition. a) E_{exc} = 1.570 eV (above the GaAs bandgap); b) E_{exc} = 1.140 eV (a sublevel of the 1st excited state); c) E_{exc} = 1.123 eV (another sublevel of the 1st excited state).

The results of the time-resolved PL (TRPL) measurements performed on sample A as a function of the irradiation dose, for resonant and non-resonant excitation, corroborate the PLE data. No influence of the irradiation on the PL decay kinetics from the ground state is observed (Fig. 4). However, the rise time shortens by about a factor of four for the maximum dose used (1×10^{14} p/cm^2), both for the excitation into the barrier (Fig. 4a) and into the sublevels of the first excited state at 1.140 and 1.123 eV (Figs. 4b, c). This means that the rise time shortening upon above-bandgap excitation is caused by a carrier (exciton) loss in the QDs and not by any reduction of the diffusion length in the barrier or the WL. The effect can be explained by the tunnel escape of the carriers to adjacent defects. The ground state, having a more localized wavefunction than the excited ones, remains essentially "undamaged". On the contrary to earlier measurements on electron irradiated QDs of another type [13], no development of a second, shorter decay time has been observed. We attribute this difference to the heavier damage caused by the electron doses in the range from 10^{16}–10^{17} e/cm^2 used in Ref. [13].

4 Conclusions In conclusion, we have made PLE and TRPL measurements on proton-irradiated InGaAs/GaAs QD heterostructures. For the sample configuration used (two AlGaAs diffusion barriers surrounding the QD plane), the main part of the carrier loss due to introduced radiation damage occurs in the dots, even for the excitation of the GaAs barrier. An increase of the ground state PL yield upon resonant excitation into the low-lying QD states has been observed upon irradiation and attributed to a lowering of the Fermi level, causing a "recharging" of the dots due to electrical compensation of the barrier material. The PL rise time gets shorter with increasing irradiation dose owing to carrier escape out of the excited QD states to adjacent radiation-induced defects. For the doses used in the study, the decay time of the ground state PL remains unaffected, proving a high radiation hardness of the dots. Along with the high localization of the wavefunction in the dots, another important reason of this hardness may be a probable expulsion of the mobile defect components out of the dots.

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