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Electron dynamics in modulation p-doped InGaAs/GaAs quantum dots

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Abstract. We investigate the electron dynamics of p-type modulation doped and undoped InGaAs/GaAs quantum dots using up-conversion photoluminescence at low temperature and room temperature. The rise time of the p-doped sample is significantly shorter than that of the undoped at low temperature. With increasing to room temperature the undoped sample exhibits a decreased rise time whilst that of the doped sample does not change. A relaxation mechanism of electron-hole scattering is proposed in which the doped quantum dots exhibit an enhanced and temperature independent relaxation due to excess built-in holes in the valence band of the quantum dots. In contrast, the rise time of the undoped quantum dots decreases significantly at room temperature due to the large availability of holes in the ground state of the valence band. Furthermore, modulation p-doping results in a shorter lifetime due to the presence of excess defects.

PACS. 78.47.Cd Time resolved luminescence – 78.67.Hc Quantum dots

1 Introduction

InGaAs (InAs) is one of the most important semiconductor quantum dots (QDs), providing tunable wavelengths to cover the near-infrared range for the optical communications industry. The three-dimensional confinement results in unique discrete levels and energy relaxation in quantum dots. Carrier capture and subsequent relaxation are the most important characteristics and have been the subject of extensive research for the past decade because they are relevant to both applications and fundamental research. The slow inter-sublevel relaxation rate in a quantum dot has been confirmed as evidence of a phonon bottleneck in many observations [1,2]. On the other hand, other studies have indicated that relaxation within the quantum dots proceeds at a rate greatly exceeding that originally thought possible in the presence of a phonon bottleneck. Several explanations have been proposed for this fast relaxation, including Auger processes [3,4], electron-hole scattering [5,6] and multiphonon emission [7,8]. The wide range of energy relaxation times is likely to result from a number of factors, for instance, variations in experimental conditions, such as lattice temperature, excitation intensity and excitation energy, and various self-assembled quantum dot features resulting from different growth conditions such as different dot sizes, shapes, ground state energies and electron and hole potential depths.

P-doped self-assembled QDs can greatly improve the performance of QD lasers, such as stable temperature characteristics and promising high-speed properties [9,10]. The stable temperature characteristics are attributed to thermal smearing of the holes in closely spaced hole levels. The high-speed characteristics are known to be strongly affected by carrier capture and subsequent relaxation. In doped QDs, the presence of built-in carriers could alter the carrier dynamics and result in an enhancement of the relaxation rate. A few studies of carrier dynamics in modulation doped QD structures have been reported [6,10,11]. Gündoğdu et al. proposed that electron-hole scattering is the main mechanism of carrier capture from the barrier or wetting layer (WL) into the ground state of the QDs and their ultrafast experiment revealed a significantly enhanced relaxation in p-doped InAs QDs due to efficient electron-hole scattering involving a built-in carrier population [6]. An increased rise time was found in both undoped and p-doped InAs QDs with increasing temperature, which was interpreted as being due to an increased

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filling of the excited hole states due to thermal spreading. Siegert et al. suggested that photoexcited electrons were first captured into the excited states of the QDs from the barrier or wetting layer by phonon emission for both undoped and p-doped InAs QDs, and the electrons subsequently relaxed into the ground state of the QDs by different mechanisms, i.e., phonon-assisted relaxation for the undoped and scattering between electrons and holes built-in the valence band for the p-doped InAs QDs [11]. The reported experimental results do not show a consistent mechanism for carrier capture and relaxation in InAs or InGaAs QDs. It is likely to be due to a sensitively dependent relaxation mechanism on the features of the QDs, such as the composition, dot size or doping density. To date most of the dynamics investigations have been carried out for InAs QDs and a detailed investigation on the carrier dynamics in p-doped InGaAs QDs is still lacking. Thus a clear understanding of the physical mechanism involved in carrier capture and subsequent relaxation in p-doped InGaAs QDs is required [7,12].

In this paper, we investigate the electron dynamics in self-assembled InGaAs quantum dots with modulation p-type doping and without doping using up-conversion photoluminescence at low and room temperatures.

2 Experiment

The InGaAs self-assembled QD samples include undoped and modulation p-doped samples. Each sample was grown on a semi-insulating (100) GaAs substrate by low-pressure metalorganic chemical vapour deposition for structures with doping and without doping. For the undoped sample, a 200 nm GaAs buffer layer was first grown at 650 °C on a semi-isolated GaAs substrate, followed by $In_{0.5}Ga_{0.5}As$ dots grown and capped with 11 nm GaAs at 550 °C. After a 1 min interruption, the sample was capped with 200 nm of GaAs as the temperature was ramped to 650 °C. For the modulation p-doped sample, the same procedure was used: a 200 nm GaAs buffer layer, undoped $In_{0.5}Ga_{0.5}As$ dots and an 11 nm capping layer were grown followed by a 1 min. interruption, then 10 nm GaAs was grown as the temperature was ramped to 650 °C, and a 20 nm thick doped GaAs (III/V ratio of 0.70) was grown at 650 °C with a CCl_4 flow at 5×10^{-7} Torr. Finally, the sample was capped with 170 nm of undoped GaAs at 650 °C.

The time-resolved photoluminescence (TRPL) experiments were performed with the PL up-conversion technique similar to that described previously [13]. The excitation pulses originate from a Ti: sapphire regenerative amplifier and have duration 80 fs, energy 1–10 μJ and repetition rate 1 kHz. The luminescence from the sample was collected in a backward geometry and mixed in a BBO crystal with a variable delay gating pulse (800 nm, 80 fs, 20 μJ) to generate a sum-frequency signal. The up-conversion system has a time resolution and spectral resolution of 150 fs and 2 nm, respectively. The time integrated PL (CWPL) was performed at 77 K and room temperature using two kinds of laser excitation, a 532 nm laser for stable and low intensity excitation and 800 nm femtosecond pulses from the same laser source as the TRPL experiment for high intensity excitation. A cooled InGaAs photodetector was used at the output slit of a 0.25 m monochromator.

3 Experimental results

Figure 1 shows the PL spectra of the undoped and doped samples excited by the 532 nm laser at a low intensity of 2 mW at 77 K. Each spectrum can be fitted well with a single Gaussian function, which is attributed to the ground state transition. No multiple peaks are observed that can be regarded as evidence of a phonon bottleneck [14,15]. Multiple PL peaks from the ground state and excited state transitions should be observed either at high excitation or low excitation intensity if the intersubband relaxation rate in the QD is comparable to the electron-hole recombination rate. A clear blue shift of 29 meV is observed for the doped sample compared to the undoped sample (centre wavelength 1070 nm), which suggests substantial carrier accumulation inside the QDs. This spectral shift has been observed and studied theoretically for charged QDs [16,17]. Regelman et al. showed that, unlike higher dimensionality charged collective states, positively charged dots show an increase in emission energy because the energy associated with hole-hole repulsion is larger than the energy associated with electron-hole attraction, which in turn is larger than the energy associated with electron-electron repulsion. Therefore, the emission due to radiative recombination of an electron-hole pair in p-doped QDs is shifted to the blue relative to the undoped QDs [17].

At low excitation intensity many-body effects, such as band-gap renormalization and state filling, can be neglected. Basically, the dot size is an important effect for the shift of the emission energy. Each sample has a very similar dot size and dot density because of the very similar growth process (the only difference is the modulation doping layer). Thus the observed blue shift should be due to extra holes in the QDs.

With increasing laser excitation intensity, a strong asymmetry appears on the high energy side of the PL spectrum (inset of Fig. 1), which can be attributed to a state filling effect and not a phonon bottleneck. At 52 mW laser excitation the CWPL spectrum for each sample is well characterized by a two Gaussian fit, where the two Gaussian components can be attributed to transitions from the ground state and the first excited state, respectively, as shown in the inset of Figure 1 for the undoped sample. Energy separations between the ground state and excited states of 62 meV and 57 meV are deduced for the undoped and doped samples, respectively. Under these excitation conditions, we do not find any components for transitions from the higher excited states or the WL.

With femtosecond laser excitation at 800 nm it is possible to observe transitions from higher excited states and the wetting layer and even from the GaAs barrier, due to the much higher excitation intensity. With increasing excitation intensity, transitions from the ground state, the

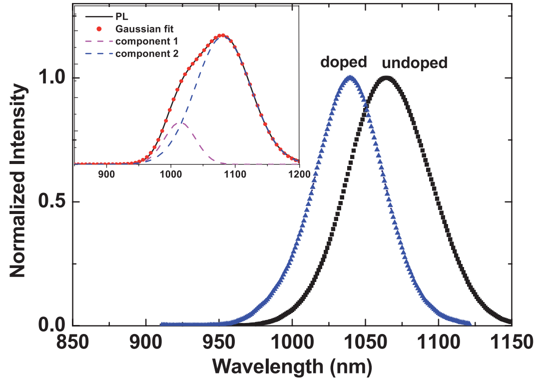

Fig. 1. Integrated photoluminescence of InGaAs/GaAs QDs for undoped and modulation p-doped at 77 K at low excitation at 532 nm. A blue shift of 29 meV is observed for the doped QDs. The inset is the integrated photoluminescence of the undoped sample with a high excitation. The dotted line is a two-Gaussian fit showing the two components.

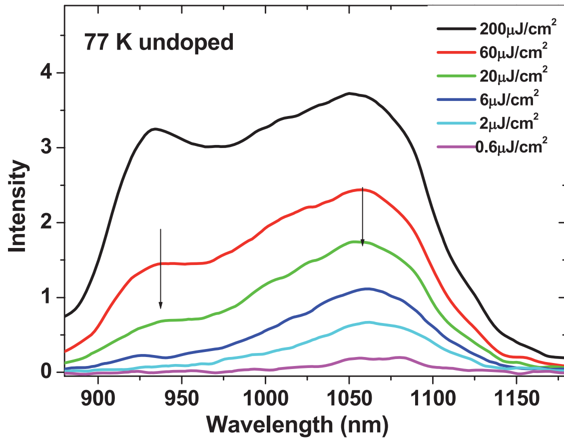

Fig. 2. Variation of the PL spectrum excited at 77 K by a femtosecond laser at 800 nm for the undoped samples. Arrows indicate the positions of the ground state and wetting layer transitions.

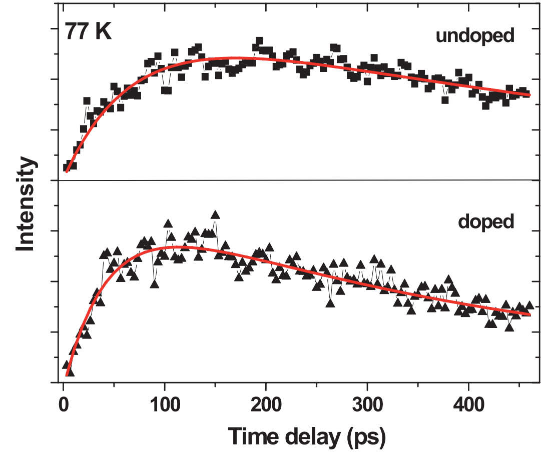

Fig. 3. Evolution of PL for the ground state transition for undoped and modulation doped samples with low intensity excitation at 800 nm and 77 K.

Table 1. Rise times and decay times for the ground state and the first excited state transitions of the undoped and doped samples at 77 K and 293 K.

Sample	Undoped		Doped	
	τ_R(ps)	τ_D(ps)	τ_R(ps)	τ_D(ps)
		77 K		
Ground state transition	65 ± 3.5	750 ± 28	35 ± 2.7	400 ± 25
Excited state transition	55 ± 3.1	730 ± 22	25 ± 2.2	400 ± 22
		293 K		
Ground state transition	23 ± 3.1	455 ± 39	30 ± 2.8	395 ± 36
Excited state transition	18 ± 2.7	430 ± 34	23 ± 2.5	350 ± 33

excited states and the WL appear sequentially, as shown in Figure 2 for the undoped sample. The doped sample gives a similar spectrum with the same wetting layer position but a different wavelength for the ground state transition (not shown here). The undoped and doped samples have the same transition wavelength from the WL located at 925 nm (1.338 eV) at 77 K, which results from the identical conditions of the dot growth. This observation also shows that modulation doping does not affect the growth of the quantum dots and the formation of the wetting layer.

Figure 3 compares the PL evolution from the ground state transition of undoped and doped samples excited at 800 nm at 77 K at an excitation intensity of 6 μJ/cm^2, which corresponds to less than one photoexcited electron-hole pair per pulse per dot. Each PL evolution can be fitted with an function of the form $A[\exp(-t/\tau_D) - \exp(-t/\tau_R)]/(\tau_D - \tau_R)$, with decay time τ_D and rise time τ_R. The rise times and decay times for the transitions from the ground state and the first excited state are deduced from the fits and are summarized in Table 1. At low temperature, the doped sample has significantly shorter rise times for both the ground state and excited state transitions than those of the undoped sample and the doped sample exhibits a shorter lifetime than the undoped sample. A moderately shorter rise time is observed for the excited state transition for each sample. At the same time, the decay times for both the ground state and excited state transitions do not show a significant difference for each sample.

The PL evolutions were also observed at room temperature under the same excitation conditions, as shown in Figure 4 for the ground state transitions of undoped and doped samples. The deduced rise times and decay times are included in Table 1. At room temperature, the rise times for the undoped sample decrease significantly but those for the doped sample remain essentially unchanged.

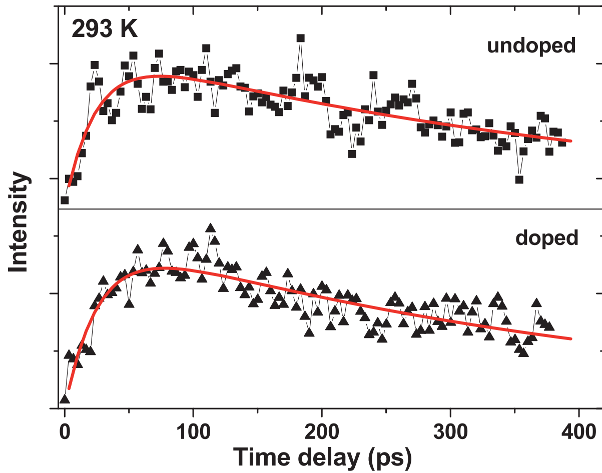

Fig. 4. Evolution of PL for the ground state transition for undoped and modulation doped samples with low intensity excitation at 800 nm and 293 K.

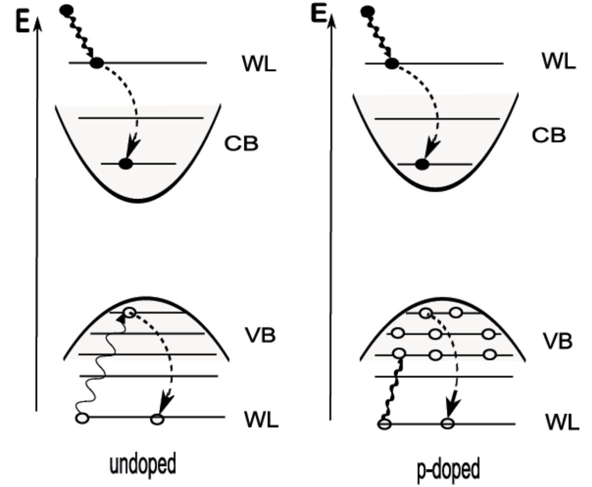

Fig. 5. Electron-hole scattering in undoped and p-doped InGaAs QDs.

4 Discussion

For InGaAs/GaAs QDs after 800 nm laser excitation, hot electrons and holes are generated in the GaAs barrier layer. A rapid longitudinal optical (LO) phonon emission via Fröhlich interaction is expected due to the continuum band in the GaAs barrier [18,19]. In this way electrons can rapidly relax into the ground state of the barrier and the wetting layer. Then two relaxation channels are expected, through which electrons relax into the ground state of the QDs. In the first channel electrons are captured directly into the ground state of the QDs from the WL, through either electron-electron scattering or electron-hole scattering. In the second channel electrons are first captured into the excited states of the QDs and then relax into the ground state through intersubband relaxation, electron-hole scattering or phonon scattering.

For a three-dimensional confined quantum dot, the LO relaxation process is forbidden because it is difficult to satisfy energy conservation in a discrete state system. The deformation potential interaction with longitudinal acoustic (LA) phonons, which is already weaker in the bulk, becomes even weaker as the dot size is reduced due to a decreasing form factor [19]. The relaxation via LA phonon emission in quantum dots is slower than in the bulk by many orders of magnitude [18]. Efficient phonon relaxation can be achieved only when the energy separation between the discrete states of the QDs is equal to the LO phonon energy or is within the narrow window of LO±LA [19]. The energy separation of the QD states typically does not match the LO phonon energy and therefore a slow intersubband relaxation rate is generally observed in a QD system, which has been called a phonon bottleneck [2,15]. Considering that the PL peaks correspond to transitions between an electron and a hole in individual quantum dots with the same quantum number [20] and taking into account the ratio between the intraband electron and hole level spacings, $\Delta E_C/\Delta E_V$, is roughly equal to 2 [21], we estimate the interlevel energy spacing for the conduction band and the valence band to be about 40 meV and 20 meV, respectively. These energy interlevel spacings cannot match the LO energy of 32 meV for $In_{0.5}Ga_{0.5}As$ [22] and thus effective LO relaxation is not possible.

The PL experiment shows there is no so-called phonon bottleneck in these InGaAs QDs. At the same time, both the PL and the TRPL experiments show that the intersubband relaxation is not very effective, because for each sample the rise time and decay time of the excited state transitions do not show obvious differences from those of the ground state [23]. For the ground state transitions, the decay time is determined by the rate of recombination of electrons and holes, while for the excited state transitions it is determined not only by the recombination rate but also by the intersubband relaxation rate. Thus in the case of effective intersubband relaxation, a markably different decay time is observed between transitions from the ground state and the excited states [23].

Capture processes of carriers from the WL to the QD states are often possible for holes since they have a shallower confinement energy than electrons [24]. Thus electron-hole scattering becomes a possible relaxation channel, in which electrons transfer their extra energy to holes and relax into the ground state of the QDs, while the holes jump into the wetting layer [25].

The primary difference between the undoped and p-doped samples is that the doped sample has excess holes in the valence band of the QDs. The scattering between electrons and holes built-in the valence band is likely to be the dominant relaxation mechanism [6,11,26]. Figure 5 illustrates schematically electron-hole scattering in both the undoped and p-doped system. In this process a photo-excited electron first relaxes into the wetting layer and then scatters with a hole in the ground state of the valence band, which results in the electron relaxing into the ground state of the QDs and the hole jumping into the wetting layer of the valence band. This scattering process is determined by the availability of holes in the

ground state of the valence band. For undoped QDs the holes in the wetting layer cannot effectively relax into the ground state at low temperature because the thermal energy is less than the separation of the hole energy states and a LO or LA phonon cannot match the separation. A hindered relaxation of holes results in a low hole availability in the ground state and thus a long rise time is observed. At room temperature holes can effectively relax into the ground state due to the large thermal energy and the relatively small interlevel separation in the valence band; so there is a large availability of holes in the ground state and thus effective electron-hole scattering is expected. Therefore a short rise time is observed at room temperature. For p-doped QDs the availability of holes does not depend on the hole thermal relaxation because there are excess holes in the ground state before laser excitation due to the doping. At both low and room temperatures an effective electron-hole scattering is expected and thus a short rise time is observed. However, according to this mechanism the rise times of the undoped QDs should not be shorter than those of the doped even at room temperature. We observe a slightly shorter rise time for the undoped QDs at room temperature. Possible reasons include, firstly, a tiny difference between the undoped and p-doped QDs may result in slightly different features in the QDs because of the individual fabrications, and therefore a slight wave-function overlap giving rise to the observed rise times. We also observed a slight difference in the energy separation between the undoped and p-doped QDs. Furthermore the relatively lower signal-to-noise ratio at room temperature may result in a relatively larger error and thus an affected rise time.

A different relaxation mechanism was observed by Gündoğdu et al. for the undoped and p-doped InAs QDs where thermal emission leads to a reduction of hole population in the ground hole state and thus a reduction of the electron-hole scattering rate due to increased filling of the excited hole states. Therefore a longer rise time was observed at higher temperatures [6]. This may result from the difference in composition, dot growth and doping process. At the same time our model is different from the dynamics of the InAs QDs in reference [11] where the photo-excited electrons are first captured into the excited electron state by phonon scattering, then relax into the ground state of the QDs by phonon scattering for the undoped QDs and by electron-hole scattering for the p-doped QDs, respectively.

From the time evolution of the PL, we deduce decay times of 750 ps and 400 ps for the undoped and modulation doped samples, respectively. Generally, the decay time of the ground state of the QDs is determined by the recombination between electrons and holes, including radiative and non-radiative recombination. Because the structures of all samples are similar except for the doping layer, the lifetime should be related to the doping. The experiment shows that the lifetime of the doped samples decreases, which should result from a small number of dopant-related defects acting as nonradiative recombination centres, as observed in other doped QDs [27]. It has been shown that the presence of doping-induced excess carriers can increase the number of certain kinds of defects because of the free carrier influence on inter-diffusion occurring during the dot overgrowth [28]. For example, in a p-type doping structure, the concentration of interstitials will increase, which leads to a positive charge. Thus a small number of point defects could result from p- or n-doping [29]. However, further investigation is required to determine the exact nature of the defects.

5 Conclusions

We have studied the electron dynamics in undoped and modulation p-doped InGaAs/GaAs quantum dots at low temperature and room temperature. For the p-doped sample the doping provides sufficient holes in the ground state of the valence band and thus electron-hole scattering is significantly enhanced. Thermal activation does not affect the availability of holes in the ground state and electron-hole scattering, and therefore the rise time is basically independent of the temperature. For the undoped QDs thermal relaxation of holes is the key mechanism for electron-hole scattering in the quantum dots. At low temperature the long rise time is due to an ineffective thermal hole relaxation into the ground state of the valence band and thus inefficient electron-hole scattering. Furthermore, modulation p-doping also leads to a shorter lifetime due to the presence of doping induced excess defects.

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