Carrier dynamics of LT InAs/GaAs QDs using time resolved differential reflectivity

Carrier dynamics of LT InAs/GaAs QDs using Time Resolved Differential Reflectivity

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Abstract: We present a Time Resolved Differential Reflectivity (TRDR) study of LT (low temperature grown) Stransky - Krastanov InAs/GaAs Quantum Dots (QDs) grown using molecular beam epitaxy. The photoluminescence (PL) spectrum shows a QD-peak around 1200nm. In the TRDR measurements we observe an initial fast decay (80ps) followed by a much slower decay of about 800ps. The strong temperature dependence of the PL-signal is not observed in the reflectivity signal. This leads us to conclude that the electrons are trapped at a fast rate by As antisite defects while the hole decay dynamics take place at a slower rate, which is also monitored in TRDR.

Introduction

Stransky–Krastanov Quantum Dots (QDs) have enhanced optical non–linearity due to their delta function like density of states which results in a sharp absorption line with a high peak absorption. In addition, the presence of a single electron hole pair within a QD is able to completely bleach the absorption line, while two electron hole pairs generate optical gain, indicating a large optical nonlinearity, which occurs already at very low pump power. The time response of the optical nonlinearity is determined by the combination of the radiative and nonradiative decay times and the carrier escape out of the QD. In low-temperature (LT) grown QDs an ultrafast response is expected due to trapping of carriers into Arsenic antisite defects. In this paper we present a study of LT InAs/GaAs QDs grown using Molecular Beam Epitaxy (MBE) as an attempt to combine large QD-nonlinearity with an ultrafast response time. We have performed both photoluminescence (PL) and time resolved differential reflectivity (TRDR) measurements on these samples to study the carrier dynamics.

Sample details

We have studied a sample with LT-grown (250°C) InAs QDs on top of a 500 nm LT-grown (250°C) GaAs lower barrier[1,2]. The QDs have been annealed at 480°C directly after QD-growth. The QDs are subsequently capped with a high temperature grown (480°C) 4 nm GaAs layer before growing the 250 nm LT-GaAs upper barrier. Finally, the sample is given a post growth anneal at 580°C. We first performed photoluminescence (PL) measurements on the sample. The details of the PL measurements are given elsewhere[3]. The PL spectrum is broad with a QD photoluminescence peak around 1200nm on top of a slowly rising background resulting from recombination due to EL2 and EL6 centers. The PL spectra also show strong quenching of the PL-intensity for increasing temperature between 4K and 40K. The strong PL-quenching is also observed for the high temperature QDs grown within an LT-GaAs barrier[3]. So the PL-quenching is attributed to trapping of photo-generated carriers in the LT barrier before being captured into the QDs. We also excited the sample with different wavelengths, both above and below the LT-GaAs barrier bandgap,

which is at 816 nm. We observe that the PL efficiency is much higher for excitation directly in the quantum dot structure (>840nm).

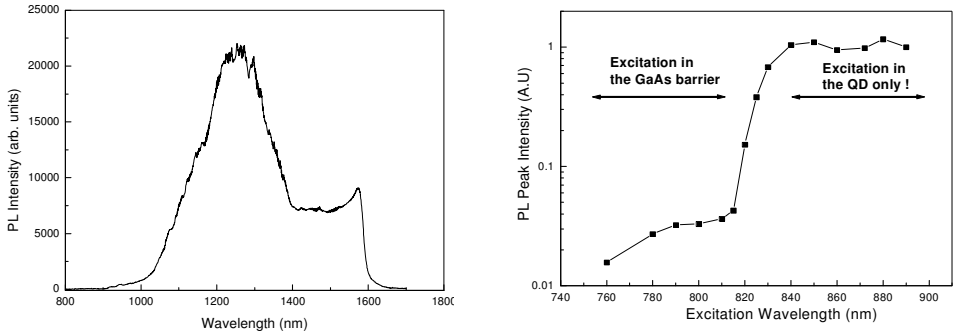

Fig 1: (left) PL spectra of LT-grown QD sample, indicating a QD emission peak around 1200 nm. (right)Excitation wavelength dependence of the PL-intensity, showing a strong increase of the PL-intensity for excitation directly into the QDs.

Time resolved differential reflectivity experiments

To explain the carrier dynamics we performed two-color pump-probe time resolved differential reflectivity measurements. The sample is excited using a mechanically chopped pump beam from a mode locked Ti:Sapphire laser at a wavelength of 785nm (above GaAs barrier band gap). The capture and subsequent decay of the carriers in the dots are probed using probe pulses from an OPO, which are synchronized with the pump pulses. The probe laser is tuned in resonance with the QDs. A time delay is introduced between the two beams by introducing a movable optical delay in the path of the pump beam. Both the beams are focused on the sample with pump beam diameter of 40μm and probe diameter of 30μm.

The carriers generated by the pump beam in the GaAs barrier diffuse to the QDs and are subsequently captured into the QDs thereby bleaching the QD transition and maximizing the reflected probe signal. TRDR signal has an exponentially rising part representing carrier capture and relaxation time[4], followed by a decaying signal representing radiative and nonradiative carrier recombination. Since the pump beam is mechanically chopped, we measure a modulated reflectivity signal ΔR/R. Total reflectivity of the signal is given by the equation[5]

$$\left|r_{tot}\right|^2 = r_s^2 - 2r_s(1-r_s)^2 \frac{(\Gamma+\gamma)\cos(2k_z D) + (\omega_0-\omega)\sin(2k_z D)}{(\omega-\omega_0)^2+(\Gamma+\gamma)^2}\,\Gamma$$

where Γ is the oscillator strength which is equal to inverse of radiative lifetime, and γ represent additional i.e. nonradiative relaxation. Due to inhomogeneous broadening resulting from QD-size distribution, the $\sin(2k_zD)$ term vanishes and the peak QD reflectivity can be considered as an absorption bleaching signal. Then absorption - bleaching spectrum reflects QD density of state.

Fig. 2a shows a typical TRDR signal from the LT-QD sample at 5K. The probe wavelength is 1167nm. An exponential fit gives a capture time of ~12ps and the decay has a bi- exponential nature with an initial faster portion of ~80ps followed by a longer decay of 800ps. Fig. 2b shows the dependence of peak reflectivity with pump power. It is clear that peak reflectivity approaches a saturation limit around a power of 70mW. So

all subsequent measurements are done in the linear region of pump power to ensure that only ground states are filled.

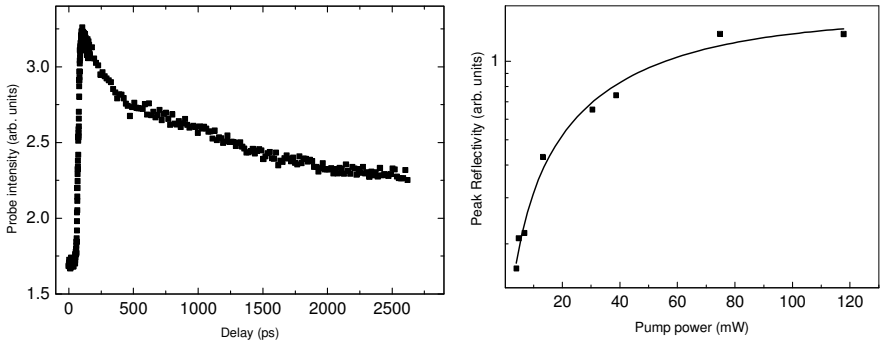

Fig 2a: (left)TRDR signal at probe wavelength of 1167nm at a temperature 5K.
Fig 2b: (right) Peak reflectivity versus pump power

Reflectivity spectrum is obtained by taking the measured peak reflectivity in each of the TRDR versus time curves. This peak reflectivity is subsequently plotted versus probe wavelength in Fig. 3, together with PL-spectrum at 5K. For this reflectivity spectrum, pump wavelength is kept constant (785nm) in the barrier region. The excitation power is 21mW, well in the linear region. The incident probe power is also kept steady at ~580μW. It is observed that the reflectivity spectrum has a first derivative shape. The signal changes phase by 180° when the wavelength is tuned below 1122nm. The PL spectrum is clearly broadened and red-shifted with respect to the reflectivity spectrum by an amount of 79meV. This result clearly indicates that the PL-spectrum also probes defect-related transitions below the QD-bandgap.

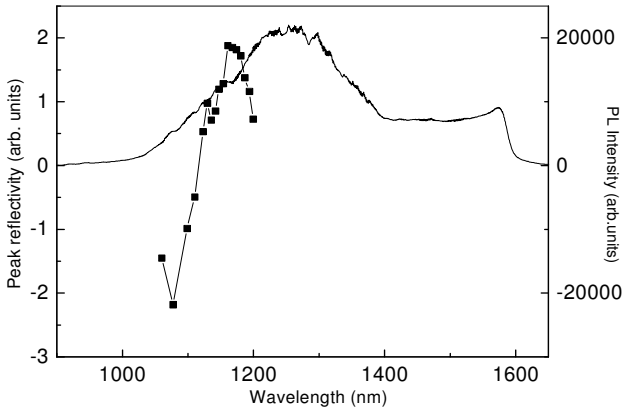

Fig3: Peak reflectivity spectrum at 5K (■) in comparison with PL at 5K

Fig 4 shows TRDR measurements at different temperatures. The inset also shows the PL-results at different temperatures. The PL shows a very strong dependence with temperature with a strong quenching of the PL-intensity at around 30K. Surprisingly, the same behavior is not reflected in TRDR. In TRDR, there is only a very small temperature dependence of decay time, with initial fast decay part becoming even faster at room temperature.

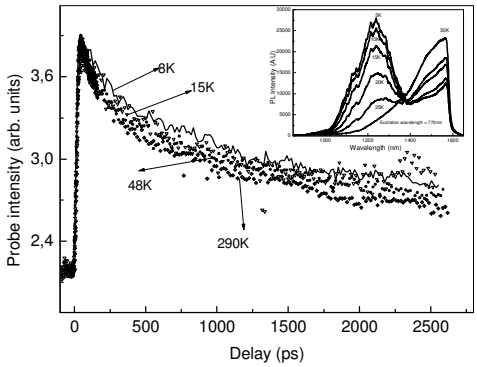

Fig 4: TRDR and PL at different temperature.

Low temperature growth is done in an excess arsenic environment leading to the formation of As antisite (As_{Ga}) defects which are mid gap energy levels[6]. The neutral antisite level is ionized by the Ga vacancy acceptor states (V_{Ga}). The first ionized state of As antisite (As_{Ga}^{+}) act as a fast electron trap. So when the sample is excited in the GaAs barrier, the photo-excited electrons in the GaAs barrier get trapped by the As_{Ga} traps while diffusing to the quantum dots, thereby leading to a strongly reduced PL efficiency. When the temperature is increased, the confined electrons in the QDs will transfer[7] faster towards the As_{Ga} traps in which they remain trapped. Sercel has calculated a multi-phonon assisted electron tunneling from the QD to a trap at a rate of 10^{10}/s at 4K and 10^{12}/s at 300K for traps which are separated between 0 and 10 nm from the QD. We attribute the strong decrease of the PL efficiency with increasing temperature to this tunneling mechanism out of the QD.

The TRDR does not show the same type of temperature dependence. This leads us to believe that the faster part of the decay reflects the electron trapping out of the QDs into the anti-site defects, while the slower decay reflects the trapping of holes, which is expected to be much slower since the hole trapping probability into EL2 is much slower than the electron trapping probability.

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