Dephasing and energy relaxation processes in self-assembled In(Ga)As/GaAs quantum dots

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Abstract. Time-resolved two-beam experiments were performed on self-assembled In(Ga)As/GaAs quantum dots (QD's). In contrast to previous two-beam transmission experiments on stacked QD's we investigated the photoluminescence properties of single QD layers. The dynamics of the ground state saturation and the state filling processes of the excited states were found to depend on the excitation intensity. As a result the exciton life-times of several hundred ps for the ground state and the excited states were determined. Under resonant excitation a short decay of about 25 ps was observed for specific circular polarization.

Since long dephasing times in quantum dots (QDs) are predicted the dynamic of the phase relaxation is extensively studied by several groups. Transient two-beam experiments were performed. Detecting the differential transmission or the four-wave mixing signal the life time T_1 and the dephasing time T_2, respectively, can be determined. This technique was successfully used for the investigation of bound excitons in bulk crystals [1]. Its application on QD's is limited. Single layers of QD's neither change the transmission above the detection limit nor provide sufficient scattering volumes. Hence, stacked QD's with up to 30 QD layers were used. [2-5]

We present an alternative approach. The photoluminescence (PL) excited by the probe beam is measured under the influence of the pump beam. Since the PL is the optical property which is dominated by the QD's, it allows us to investigate single QD layers.

The investigated structures consists of pyramidal InAs/GaAs QD's and In-rich flat, truncated In(Ga)As/GaAs QD's. The growth processes, structural investigations, and previous optical studies on these samples are described elsewhere [6, 7].

In our experiment the QD ensemble is excited nonresonantly by a mode-locked Ti:sapphire laser (1.60v eV) and resonantly by an optical parametric oscillator.

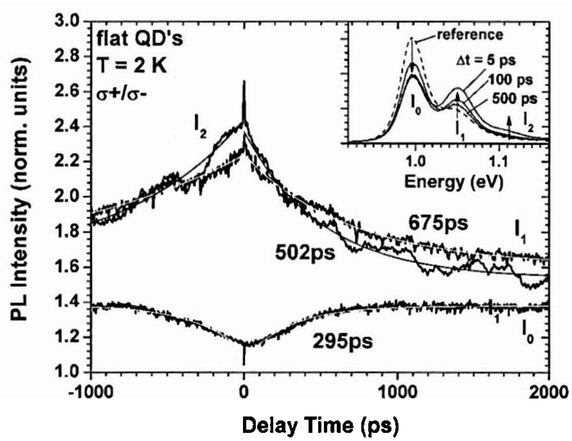

FIGURE 1. Low excitation intensity: normalized PL intensity as a function of Δt, pulse energies: pump: 190 pJ, probe: 25 pJ.

The luminescence light was detected by an InGaAs photo-detector attached on a subtractive 0.35 m double-grating monochromator. Modulating the probe beam and using lock-in technique the PL generated by the probe could be separated from the pump PL. The temporal delay between pump and probe pulse can be varied in 0.1 ps steps. The temporal resolution of the experiment is limited by the autocorrelation of both pulses having a full width at half maximum (FWHM) of 2 ps.

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Under low nonresonant excitation the intensity of the QD ground state I_0 decreases with decreasing Δt due to the saturation by the pump pulse in favor of the increasing PL intensity of the excited states I_1 and I_2 (inset Fig. 1). At Δt = 0 ps the autocorrelation of the pump and the probe pulse occurs (Fig. 1). Around this coherence peak the saturation of I_0 is strongest. The population of I_0 by the pump prevents a further excitation of this state by the probe. The transient behavior shows a exponential decay of the saturation. This describes the depopulation of I_0 which can now be refilled by carriers excited by the probe. Hence the time constant of 295 ps for the flat QD's, respectively, corresponds to the life time of I_0. For the pyramidal QD's a longer life time of 940 ps was determined due to the smaller electron-hole wavefunction overlap in these QD's [6]. The PL intensity of I_1 and I_2 increases with decreasing delay. It evidences that the charges excited by the probe recombine in I_1 as long as I_0 is filled up by the pump pulse.

At higher excitation density the picture changes drastically (Fig. 2). I_0 and I_1 are saturated which leads to an increase of the I_0 saturation time. With increasing excitation intensity the I_0 saturation time increases from 295 ps to 716 ps. This time constant consists of the life times of I_0 and I_1. The I_1 saturation time (270 ps) and the I_2 rise time (355 ps) correspond to the I_1 life time of 350 ps [6].

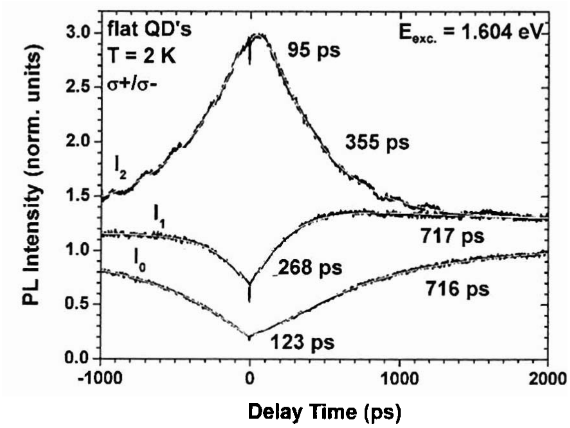

FIGURE 2. High excitation intensity: normalized PL intensity as a function of Δt, pulse energies: pump: 500 pJ, probe: 75 pJ.

Concluding this work transient pump-probe experiments on single layers of pyramidal InAs/GaAs QD's and flat In(Ga)As/GaAs QD's were performed. In contrast to previous two-beam experiments the QD luminescence was detected. This allows us to study the influence of the state filling processes on the relaxation and recombination dynamics and the involvement of the excited QD states on the optical properties, respectively. The interaction of the QD states especially at high excitation densities are relevant for laser applications. As a result the exciton life times of the QD ground state and its excited states were determined. An additional polarization-dependent short time constant was observed. Further investigations shall clarify its origin and the applicability of this technique for the investigation of dephasing processes.

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