Ultrafast Intersublevel Spectroscopy of Quantum Dot Ensembles and Single Quantum Dots

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Based on ultrafast interband pump intraband probe spectroscopy we measure electron capture and relaxation times into the discrete dot energy levels within a few picoseconds and discuss possible relaxation mechanisms. Further we present an approach to rule out dot ensemble related effects within the time resolved measurements by combining interband micro-photoluminescence measurements with the intersublevel pump and probe spectroscopy. We show preliminary results for time-resolved, mid infrared induced direct electron transfer between electron intersublevels, which is monitored by changes in the interband luminescence of a single quantum dot.

Introduction

The strong interest in semiconductor quantum dots (QD) is based on their macroscopic quantum mechanical properties. Their low dimensionality within the surrounding 3D semiconductor matrix reduces phase scattering within the quantum mechanical wavefunctions of the QDs and leads to atomlike eigenstates in their energetic structure. Within the last decade, many attempts were made to exploit these properties in order to optimize the performance of optoelectronic devices like mid- and far-infrared photodetectors [1] or QD based near-infrared band-gap lasers [2]. Furthermore, it became possible to directly address single QDs, which led to optically and electrically driven single photon turnstile devices [3]. QDs can even be regarded as candidates for quantum-bit operations since its eigenstates could be entangled and basic q-bit operations have been carried out [4].

However many questions concerning carrier dynamics in QDs still remain open. Especially where only the electrons within the dots contribute to the device performance, the existing interband techniques to study exciton dynamics within these nanostructures deliver only ambiguous results, which complicate or even prevent the correct design of the device. Hence, getting access to the electron properties separated from hole contributions and ensemble related effects will allow accurate predictions how far QDs can improve devices working in the MIR and FIR regions.

We will demonstrate how ultrafast interband pump intraband probe spectroscopy will access these informations.

Experiments

Both samples were grown by MBE on semi-insulating GaAs substrates. The first one, for QD ensemble experiments, consists of 30 layers of InAs QDs separated by 50 nm thick GaAs barriers and a 50 nm GaAs cap layer. The dot density was estimated from an atomic force microscopy study to be around 2×10^{10} cm^{-2} per layer. For IR absorption measurements, the sample was polished at 58° to the growth axis in order to form a single-pass waveguide for the IR radiation and to enable a considerable electric field

component along the growth axis. The second sample for single dot (SQD) experiments contains only one QD layer with a density gradient between 10^7 dots/cm^2 and 5×10^{10} cm^{-2}. In the low-density area, the sample was processed into 8.5 µm tall micropillars with diameters between 1 and 10 µm. This assures the illumination and detection of one QD only in the micro-photoluminescence (µPL) experiments below.

For the first sample room-temperature photoluminescence (PL) spectra for two different excitation densities were recorded using a continuous-wave Ti:sapphire laser (740 nm excitation wavelength). From the PL we determine an energetic difference of 252 meV between the QD ground state e1h1 and the wetting layer (WL). Approximately two thirds of this energetic difference occur between the conduction band offsets [5]. Thus, we estimate the intraband transition energy between the QD ground state e1 and the WL to be ~160 meV. Accordingly, we expect intraband transitions from the excited states e2 and e3 to the WL at energies of ~105 meV and ~50 meV, respectively. Thus, the intraband absorption into the WL at probe energies of 160, 105 and 50 meV can be interpreted in terms of e1, e2 and e3 QD level electron populations, respectively.

For time-resolved probing of the intraband transitions, we used a mode-locked Ti:sapphire laser that delivers 12 fs pulses centered at a wavelength of 780 nm. Half of the laser intensity I_p served as an interband pump to inject electrons and holes in the GaAs barriers. The other part was used to generate the linear polarized IR probe pulses by phase-matched difference frequency mixing in a 0.5 mm thick GaSe crystal [6]. The probe pulses are tunable in the 75 – 155 meV range with pulse durations between ~100 and ~200 fs.

Absorption signals were measured at two different IR probe beam polarizations: When the probe is polarized perpendicular to the growth direction (s-polarization) we observe a step-like increase of the absorption, which rises within the time resolution of the experiment and decays within several hundred picoseconds. This signal is attributed to free-carrier absorption in the substrate and barriers and its decay to free-carrier recombination. When the probe is polarized in growth direction (p-polarization) a slowly rising absorption superimposed on the free-carrier signal is observed. Relaxation and thermalization in the GaAs barriers and the InAs WL occur on a time scale <1 ps. Thus, the slow rise of the absorption reflects the effective electron capture into the QD, more precisely into the QD ground state. From the absorption data, we deduce the capture time τ_c by an exponential fitting procedure.

Let us turn our attention to an excitation regime where electron-electron scattering can be ruled out as a relaxation mechanism [7]. Recording the temperature-dependence of the capture time at I_p = 25 W/cm^2 we find an increase of τ_c from 2.7 ps to 4.7 ps upon decreasing the temperature from 300 K to 5 K. Two scattering mechanisms could explain such short capture times: (i) multi-phonon emission and (ii) electron-hole scattering.

(i) Multi-phonon emission: The most efficient capture pathway in our sample is the step-wise relaxation through the excited QD states. The longitudinal optical (LO) phonon energy in InAs amounts to E_{LO} = 29 meV and the average electron level separation in our QDs is ~55 meV. Thus, one possible explanation for the observed short capture times could be that electrons scatter between subsequent QD states via repeated emission of two LO phonons [7], [8]. Although LO phonons can relax electrons in a QD only for a narrow range of dot sizes (because of their weak dispersion), emission of energetically different LO phonons (from the GaAs barriers, the WL, the QDs, and the respective interfaces) and also broadening of the phonons (due to strain and alloy inhomogeneities) could enlarge this energetic window [6]. This “continuum” of LO phonons could relax the entire distribution very efficiently.

(ii) Electron-hole scattering: since this process involves only a single electron and a single hole in a QD, it can occur even at very low excitation densities [9]. Due to the high effective hole mass, the QD level separation in the valence band is in the range of

a few meV. This allows holes to thermalize within several hundred femtoseconds. Electrons in the conduction band can transfer their energy to holes, which then lose their energy via phonons. Holes can be scattered depending on their initial energy either into higher QD hole states or into the WL.

Our understanding of the capture process is the following: Electrons are generated by the pump pulse in the GaAs barrier and are then transferred very fast into the WL. This process, which is mediated by LO phonons, occurs on a sub-picosecond timescale [10]. Afterwards the electrons relax between subsequent QD states either via repeated emission of two LO phonons or via subsequent electron-hole scattering processes.

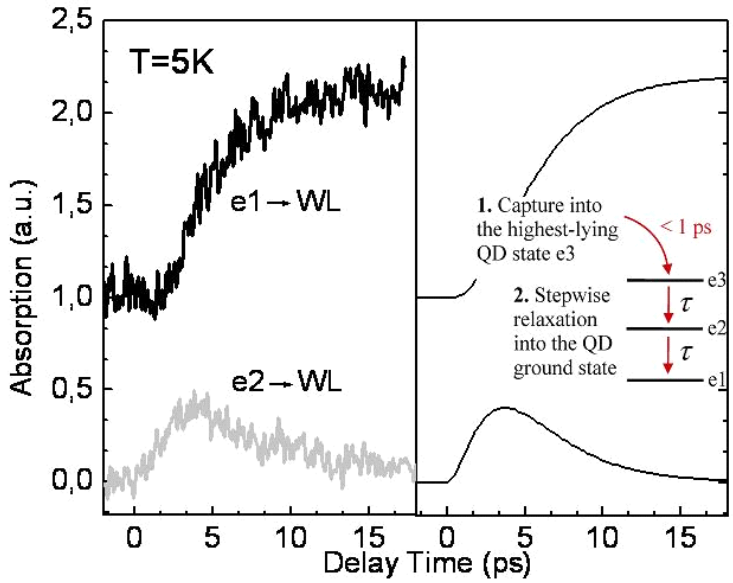

Fig. 1: (a) Photoinduced IR absorption signals for the E1-WL transition at 155 meV and the E2-WL transition at 105 meV (lower curve) as a function of pump-probe delay at 5K. (b) Results from the three level model sketched between the curves.

In order to support this interpretation we compare in Fig. 1 (a) the IR absorption at energies of 155 meV and 105 meV, corresponding to the e1-WL and e2-WL intraband transitions, respectively (T = 5 K). Note that in this figure the free-carrier absorption signals have been subtracted for better clarity. Since the absorption signals can be interpreted in terms of QD level populations it is clear that the QD ground state e1 gets populated via the excited state e2, because the electrons leave e2 with the same time constant as they arrive in e1. In Fig. 1 (b) we present results obtained by solving the rate equations for a simple three-level model, shown in the inset. In this model it is assumed that electrons populate the highest-lying QD state e3 within 1 ps after the pump and relax afterwards via e2 to the ground state e1. Good agreement with the experimental data is obtained for τ = 2.5 ps. These results suggest that not only the capture, but also the relaxation of electrons in our QDs occurs on a picosecond timescale [7].

However, these experiments cannot completely satisfy question concerning the relaxation processes. The ensemble broadening in PL as well as in the absorption signal might cover important aspects within the dynamic behavior of the dots. To rule out ensemble related processes or contributions from the surrounding material these time resolved studies have to be extended into the SQD regime. Since direct MIR pump and probe measurements on SQDs are impossible due to the small sensitivity of absorption measurements, another radiative process has to be used to monitor the intersublevel dynamic. We suggest using the recombination NIR radiation of excitons and excited

excitons within the SQD for this purpose. By combining P&P with the µPL setup, we obtain all necessary tools for our time resolved investigation.

Preliminary results from NIR excitation of a SQD within a micromesa structure superimposed with cw MIR radiation indicate a direct electron transfer between the s- and p-levels of the SQD [11], [12]. Corresponding to the electron occupation the photon count rate of the exciton lines is changed if MIR excitation is superimposed on the NIR excitation.

In case of a time resolved experiment we expect changes in the photon count rate in dependence of the NIR-pump-MIR-probe delay. Such an experiment is depicted in Fig. 2 (a) where we recorded the delay dependent count rate for an exciton ground state in a SQD within a 5 µm micropillar.

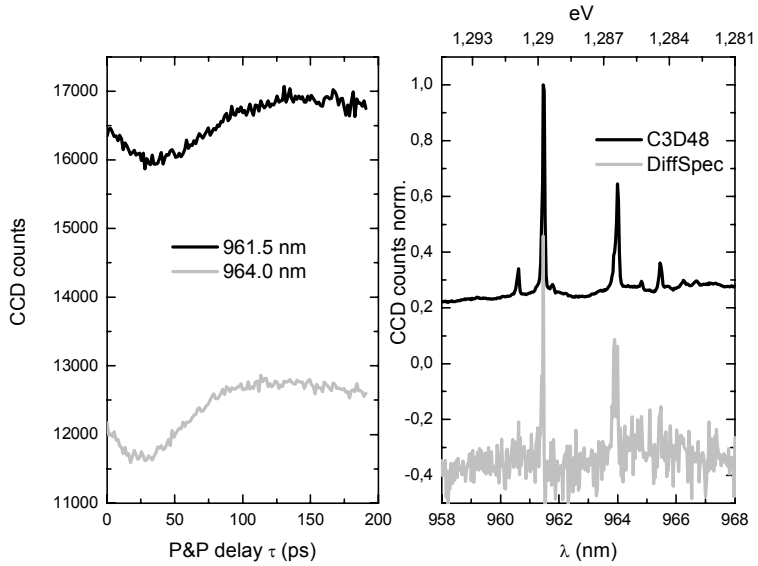

Fig. 2: Intersublevel dynamics in a SQD: (a) Photon count rate in dependence of P&P time delay τ. (b) Spectrum of QD ground state (upper curve) and difference spectrum after NIR/MIR superposition at τ = 30 ps.

In this case, the electron is transferred via the 95 meV MIR probe pulse from the ground state into the WL continuum and thus removed from the recombination channel of the SQD. Consequently, the PL count rate decreases for the observed transitions at 961.5 nm and 964 nm. As control experiment we recorded the SQD spectrum for NIR excitation with and without superimposed MIR radiation at delay τ = 30 ps. Afterwards we subtracted both spectra from each other as depicted in the lower curve of Fig. 2 (b). Again the count rate was decreased if an appropriate delay interval was chosen while no effect could be monitored if a very large time delay (>400 ps) was chosen. Further investigations of these MIR induced electron transfers will follow and new insights into the relaxation effects within SQDs are expected in the near future.

Conclusions

We suggest exploiting the capacity of ultrafast interband pump intraband probe spectroscopy combined with µPL tools for the investigation of QD dynamics. We could monitor the electron capture and relaxation dynamics of a QD ensemble on a low picosecond timescale. In addition, we could demonstrate a P&P procedure that allows studying electron dynamics in a single QD.

Acknowledgements

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