Carrier Dynamics in Quantum Dots

T. Müller, F.F. Schrey, G. Fasching, L. Rebohle, G. Strasser, K. Unterrainer

Institut für Photonik & Zentrum für Mikro- und Nanostrukturen, TU-Wien
Floragasse 7, A-1040 Wien, Austria

The electron capture and relaxation dynamics in self-assembled InAs/GaAs quantum dots is investigated by means of interband pump – intraband probe spectroscopy. By tuning femtosecond infrared pulses into resonance with intraband transitions between confined quantum dot states and the wetting layer continuum, the electron population of the quantum dot ground state is determined as a function of time-delay after the interband pump.

Introduction

The capture and relaxation of carriers in semiconductor quantum dots (QDs) has attracted much attention during the last decade, since a profound understanding of these processes is essential for the development of novel optoelectronic devices. A drastic slow-down of the relaxation compared to higher-dimensional structures has been predicted because of the so-called phonon bottleneck effect [1], [2]. However, it has turned out that a number of scattering processes, including multi-phonon emission [3] – [7], electron-electron scattering [8] – [10] and electron-hole scattering [11], [12], can circumvent the phonon bottleneck, leading to capture and relaxation times from approximately one to several tens of picoseconds. Most of the experiments have been performed by using interband techniques, such as time-resolved photoluminescence (PL) spectroscopy [4], [5], [7], [9] and differential transmission spectroscopy [6], [11], where the signal reflects the combined electron-hole dynamics. In this letter we report an interband pump – intraband probe experiment, which is sensitive to the capture and relaxation of electrons only. The pump excites electrons and holes in the GaAs matrix surrounding the QDs, while the infrared (IR) probe is tuned into resonance with electronic intraband transitions [13] between the bound QD states and continuum states in the wetting layer (WL).

Results and Discussion

The investigated sample was grown by molecular beam epitaxy on a semi-insulating GaAs substrate. It 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.

Room-temperature PL spectra for two different excitation densities were recorded using a continuous-wave Ti:sapphire laser (740 nm excitation wavelength) and are presented in Fig. 1. At low excitation density (25 W/cm^2) we observe two transitions corresponding to *e*1*h*1 luminescence at 1.081 eV and *e*2*h*2 luminescence at 1.137 eV. At higher excitation (1 kW/cm^2) the QD states are filled up and luminescence corresponding to the *e*3*h*3 transition at 1.194 eV is also observable, as well as luminescence at 1.333 eV from the underlying InAs WL. The inhomogeneous broadening of the transitions (~50 meV full width at half maximum (FWHM)) mainly reflects the size distribution

of the InAs QDs. At T = 5 K the PL shifts by ~90 meV towards higher energy which is due to the temperature-dependence of the GaAs and InAs bandgaps. The energy differences between the QD states, however, are not affected. From the PL we determine an energetic difference of 252 meV between the QD ground state *e*1*h*1 and the WL. Approximately two thirds of this energetic difference occur between the conduction band offsets [6]. Thus, we estimate the intraband transition energy between the QD ground state *e*1 and the WL to be ~160 meV.

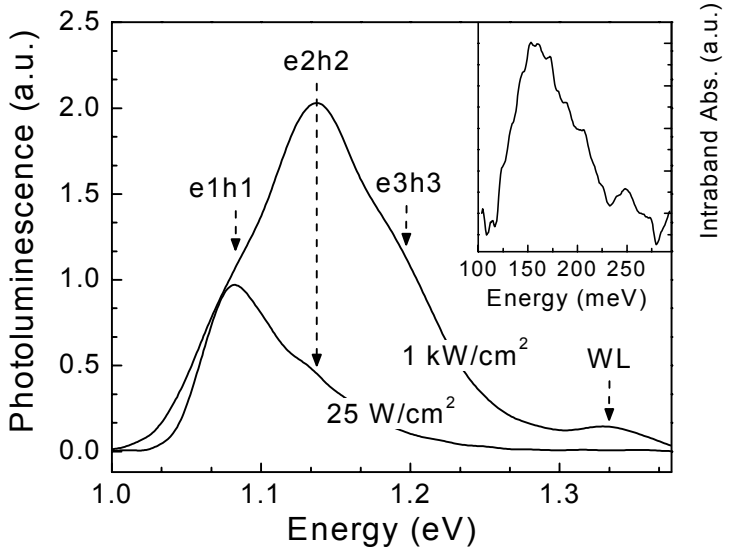

Fig. 1: Room-temperature photoluminescence spectra at two different excitation densities. Inset: Photoinduced intraband absorption spectrum at T = 5 K.

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 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 [14]. The probe pulses are tunable in the E_{pr} = 75 – 155 meV range (10 – 20 meV FWHM) with pulse durations between ~100 and ~200 fs.

Figure 2 (a) shows typical pump-probe signals (E_{pr} = 155 meV, I_p = 25 W/cm^2) at room-temperature when the probe is tuned into resonance with the *e*1–WL transition. 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.

The inset of Fig. 2 (a) displays the excitation density dependence of the capture time τ_c at room-temperature. We observe two regimes exhibiting a different density dependence of the capture time. τ_c decreases with increasing excitation density above 25 W/cm^2. In contrast, the capture time changes only slightly at low excitation densities. The high-power dependence can be explained by electron-electron scattering: An elec-

tron is scattered into the QD ground state by transferring its energy to a hot electron in the barrier or the WL. A sequence of scattering processes where the electron relaxes through excited QD states is even more probable [8]. Assuming that the electron-electron scattering rate is proportional to the excitation density and taking another density-independent scattering process (described by the scattering rate τ_i^{-1}) into account we can write for the overall capture rate [9] $\tau_c^{-1} = \tau_i^{-1} + \sigma \cdot I_p$, where σ is a fitting parameter. Using this simple expression we can fit the experimental results, as shown by the solid line in Fig. 2 (a).

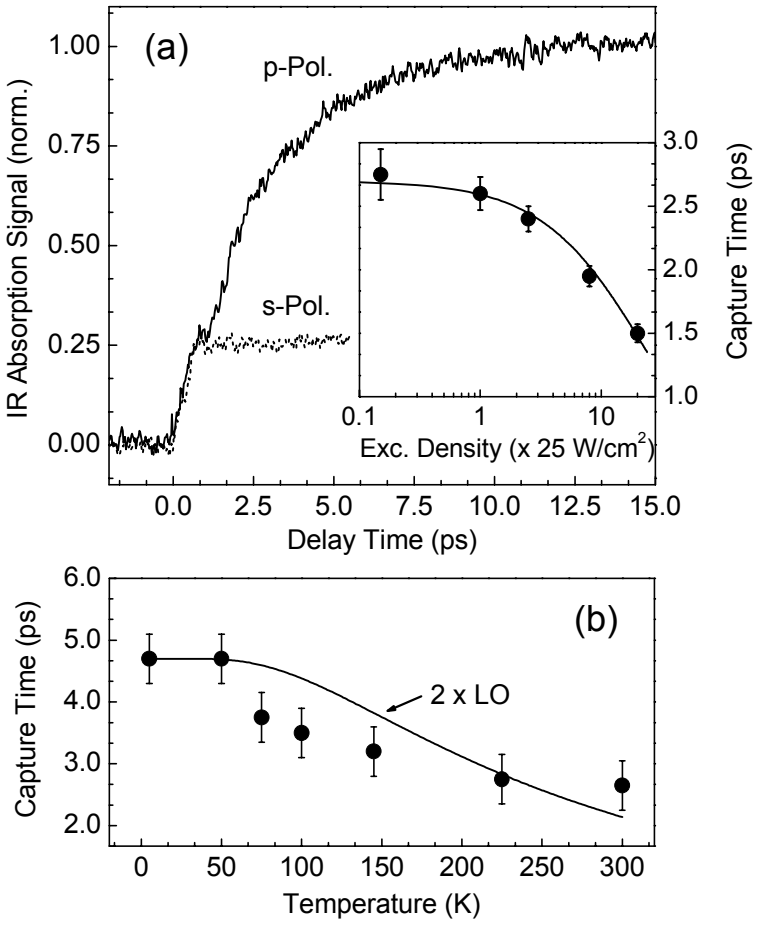

Fig. 2: (a) Photoinduced IR absorption signals of the QD sample as a function of pump-probe delay for a probe energy of 155 meV. Inset: excitation density dependence of the capture time at room-temperature. (b) Temperature-dependence of the capture time at an excitation density of 25 W/cm^2.

Let us now turn our attention to the low-density regime where electron-electron scattering can be ruled out as a relaxation mechanism. Figure 2 (b) shows 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 at low excitation density:

(i) Multi-phonon emission: 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. The corresponding two-phonon scattering rate for this process can be written as [7] $\tau_c^{-1} = \Gamma_0 \times [N_{LO}(T) + 1]^2$, where Γ_0 is the scattering rate at

T = 0 K and $N_{LO}(T)$ is the Bose-Einstein distribution function for LO phonons. The calculated curve can roughly account for the experimental temperature dependence, as shown in Fig. 2 (b) by the solid line. 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 [14].

(ii) Electron-hole scattering: Another fast relaxation mechanism in QDs is 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 [11]. 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.

Acknowledgements

This work was sponsored by “Gesellschaft für Mikroelektronik (GMe)” and “Fonds zur Förderung der wissenschaftlichen Forschung (SFB-ADLIS)”.

References

[1] U. Bockelmann and G. Bastard, Phys Rev. B **42**, 8947 (1990).

[2] H. Benisty, C. M. Sotomayor-Torres, and C. Weisbuch, Phys Rev. B **44**, 10945 (1991).

[3] T. Inoshita and H. Sakaki, Phys. Rev. B **46**, 7260 (1992).

[4] B. Ohnesorge, M. Albrecht, J. Oshinowo, A. Forchel, and Y. Arakawa, Phys. Rev. B **54**, 11532 (1996).

[5] R. Heitz, M. Veit, N. N. Ledentsov, A. Hoffmann, D. Bimberg, V. M. Ustinov, P. S. Kopev, and Zh. I. Alferov, Phys. Rev. B **56**, 10435 (1997).

[6] J. Feldmann, S. T. Cundiff, M. Arzberger, G. Böhm, and G. Abstreiter, J. Appl. Phys. **89**, 1180 (2001).

[7] M. De Giorgi, C. Lingk, G. von Plessen, J. Feldmann, S. De Rinaldis, A. Passaseo, M. De Vittorio, R. Cingolani, and M. Lomascolo, Appl. Phys. Lett. **79**, 3968 (2001).

[8] U. Bockelmann and T. Egeler, Phys. Rev. B **46**, 15574 (1992).

[9] D. Morris, N. Perret, and S. Fafard, Appl. Phys. Lett. **75**, 3593 (1999).

[10] S. Sauvage, P. Boucaud, F. Glotin, R. Prazeres, J.-M. Ortega, A. Lemaître, J.-M. Gérard, and V. Thierry-Flieg, Appl. Phys. Lett. **73**, 3818 (1998).

[11] T. S. Sosnowski, T. B. Norris, H. Jiang, J. Singh, K. Kamath, and B. Bhattacharya, Phys. Rev. B **57**, R9423 (1998).

[12] R. Ferreira and G. Bastard, Appl. Phys. Lett. **74**, 2818 (1999).

[13] S. Sauvage, P. Boucaud, F. H. Julien, J.-M. Gérard, and V. Thierry-Mieg, Appl. Phys. Lett. **71**, 2785 (1997).

[14] R. A. Kaindl, D. C. Smith, and T. Elsaesser, Opt. Lett. **23**, 861 (1998).