Capture and release of carriers in InGaAs/GaAs quantum dots

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Abstract. We observe the ultrafast capture and release of charge carriers in InGaAs/GaAs quantum dots (QDs) at room-temperature with time-resolved terahertz spectroscopy. For excitation into the barrier states, a decay of the photoinduced conductivity, due to capture of carriers into the nonconducting QD states is observed. The increase of the decay time constant with increasing pump fluence is attributed to filling of the QD states. In the case of resonantly excitation into the QD ground state a maximum conductivity is reached 35 ps after photoexcitation, which is assigned to the release of carriers from the QDs into the wetting layer and barrier states.

1. Introduction
Ultrafast carrier dynamics in semiconductor quantum dots (QDs) has been widely studied for better understanding of QD-based optical devices, such as QD-lasers [1], QD-infrared photodetectors [2] and QD semiconductor saturable absorber mirrors (SESAM) [3]. Several ultrafast optical techniques, like time-resolved photoluminescence [4], optical pump-probe spectroscopy [5, 6] and time-resolved THz spectroscopy [6, 7, 8, 9] have been used. Previous THz-studies on QD systems have concentrated on the carrier capture into the QDs. In this work we have studied both the carrier capture into solid state QDs as well as the release of carriers from the QDs into the wetting layers (WLs) and barrier states (BSs) with an optical pump - terahertz probe technique.

2. Experiments and Results
The capture and release of carriers is measured in a QD SESAM structure. The sample is grown by molecular beam epitaxy. A schematic of the sample structure is shown in Fig. 1(a). It consists of 15 layers of In₀₅Ga₀₅As/GaAs QDs, separated by GaAs barrier layers with a total thickness of 611 nm and a 33-layer Al₀₉Ga₀₁As Bragg reflector. The Bragg reflector has a 100-nm-wide stopband centered around 1060 nm (Fig. 1(b)). Besides the small signal reflectivity of the QD SESAM, the room-temperature photoemission spectra of the QD ground state (GS) is shown in Fig. 1(b). The emission spectrum has a central wavelength around 1017 nm, which is within the stopband of the Bragg reflector. The Bragg reflector is important in our experiment in the
Figure 1. (a) Schematic of the studied InGaAs/GaAs quantum dot semiconductor saturable absorber mirror. (b) Small signal reflectivity (black curve) and room temperature optical emission spectra (blue curve) from the QD ground state. (c) Carrier dynamics processes in photoexcited quantum dots. CS: conducting state and GS: QD ground state.

case of resonantly excitation into the QD GS. The optical pump beam is reflected at the Bragg grating, preventing carrier excitation in the thick GaAs substrate by two-photon absorption, which could easily obscure the signal from the release of carriers from the QDs.

To measure ultrafast carrier dynamics in InGaAs/GaAs QDs on a picosecond timescale, time-resolved THz spectroscopy is used. For the experiment, the output of a regenerative Ti:sapphire amplifier with a central wavelength of 800 nm is used. Part of the beam is used as a pump, to excite carriers either directly into the GaAs BS or with 1024 nm central wavelength pulses, generated via frequency conversion in an optical parametric amplifier, into the QD GS. The 1024-nm pulses have a full-width-at-half-maximum (FWHM) of 45 nm, ensuring resonant excitation into the QD GS. The other part of the 800 nm laser beam is used to generate and detect the THz probe pulses. The THz probe pulses, generated by optical rectification in a nonlinear crystal, are transmitted through the sample, thereby probing the changes in conductivity, and then detected by free-space electrooptic sampling. The changes in conductivity of the photoexcited sample is measured by monitoring the relative differential transmission of the peak of the THz pulse, $-\Delta T/T_0$, for different pump-probe delay times [10]. The relative differential transmission can then be related to the change in conductivity.

The relevant ultrafast processes in QDs after optical excitation in the QD GS and the BS are shown in Fig. 1(c). Only carriers in the BS and WL, here referred to as the conducting state (CS) (marked in blue in Fig. 1(c)), will contribute to the conductivity. For simplicity only the conduction band with electron capture into the QDs, the release of electrons into the CS and carrier recombination in both QDs and CS are shown. Also important for the ultrafast carrier dynamics in QDs are intradot carrier relaxation, carrier-carrier and carrier-phonon scattering. All these process are temperature and carrier density dependent [5, 11].

To measure the carrier capture dynamics into the QDs, carriers were excited into the BS with 800 nm excitation. The time dependencies of the frequency integrated change in sheet conductivity, $\Delta \sigma_{\text{sheet}}$, for pump fluences in the range 0.04-1.18 $\mu$J/cm$^2$ are shown in Fig. 2(a). A near instantaneous rise of the photoinduced conductivity is observed, followed by a fast decay and a long-lived contribution. The fast decay is attributed to carrier capture into the nonconducting QD states. To support this interpretation, the change in photoinduced conductivity of bulk GaAs is measured (black curve in Fig. 2(a)), which shows a much slower initial decay. The
Figure 2. (a) Color lines: Dynamics of photoinduced sheet conductivity, $\Delta \sigma_{\text{sheet}}$, of a InGaAs/GaAs QD SESAM at 800 nm optical excitation into the barrier layers, with pump fluences of 0.04-1.18 $\mu$J/cm$^2$. Black line: Dynamics of $\Delta \sigma_{\text{sheet}}$ in bulk GaAs, with 800 nm excitation with a pump fluence of 0.9 $\mu$J/cm$^2$. (b) 10% decay time of the traces in (a), as a function of pump fluence.

The long-lived component is most likely caused by carriers excited in the Bragg reflector, which is accessible for 800 nm light. The Bragg reflector is a type-II Al$_{0.9}$Ga$_{0.1}$As semiconductor, causing separation in space of electrons and holes, via $\Gamma$-X transfer [12]. The little bump at around 22 ps (Fig. 2(a)) is an experimental artifact due to double reflection in a 2-mm thick beam attenuator.

The initial rise in conductivity depends linearly on excitation fluence (not shown here), as expected for one-photon absorption. From the fast decay of the conductivity, the trapping time into the QDs can be estimated. In Fig. 2(b), the time required for the conductivity to drop by 10% from its maximum, as function of pump fluence is plotted. A near-linear growth of the 10% decay time from 1.2 to 5.7 ps is observed, with increasing pump fluence. This growth in trapping time with increasing pump fluence is in agreement with observation made in Refs. [7, 8, 9, 10] and is attributed to filling of the QD trap states at higher excitation fluences, which decreases the trap efficiency. However, these observation are in contrast to the observed decrease in trapping time in Refs. [5, 6]. The different technique used in Ref. [5], where only the population of the QD GS is measured may explain the different observation. The suggested trapping mechanism electron-electron scattering may also provide carriers back into CS from the higher-lying QD states, thereby slowing down the decay dynamics of the conductivity. It is not clear at this point what causes the difference between Ref. [6] and our observations.

To observe the release of carriers from the QDs into the CS, carriers are resonantly excited into the QD GS at a wavelength of 1024 nm, with pump fluences in the range 2.1-22.8 $\mu$J/cm$^2$ (Fig. 3(a)). A non-instantaneous rise of the conductivity, which peaks after approximately 35 ps after photoexcitation, followed by a slow decay is observed. We interpret the slow rise time of the photoconductivity as release of carriers into the CS. After 35 ps the recombination in the BSs and WLs becomes the dominant process, causing the decay of the conductivity. The peak conductivity has a sub-linear dependence on pump fluence (Fig. 3(b)), suggesting that the rise in conductivity is caused by a saturated one-photon process, expected for resonantly excitation into the QD GS. From the fits to the saturable absorption [13] function, $\propto F_p \exp[-F_p/F_s]$, where $F_p$ is the pump fluence and $F_s$ is the saturation fluence, a saturation fluence of 42 $\pm$ 6 $\mu$J/cm$^2$ is obtained. This is in reasonable agreement with a value of $F_s = 25 \mu$J/cm$^2$, observed in a similar QD-based structure [3].
Figure 3. (a) Dynamics of photoinduced sheet conductivity, $\Delta \sigma_{\text{sheet}}$, at resonant optical excitation of the QD GS at 1024 nm, with pump fluence of 2.1-22.8 $\mu$J/cm$^2$. (b) Maximum of $\Delta \sigma_{\text{sheet}}$ traces in (a), as a function of pump fluence. Solid line: Fit to the saturable absorption function with saturation fluence $F_s = 42 \pm 6$ $\mu$J/cm$^2$. Inset: Normalized $\Delta \sigma_{\text{sheet}}$ traces for excitation at 800 nm into the barrier state with a fluence of 1.18 $\mu$J/cm$^2$ (red line) and at 1024 nm into the QD GS with a fluence of 22.8 $\mu$J/cm$^2$.

3. Conclusion
We have demonstrated the difference in photoconductivity dynamics of an InGaAs/GaAs SESAM, depending on whether carriers were optically excited resonantly into the QD GS or into the BS (inset of Fig. 3(b)). The dynamics can be explained by the dominating process, which is either carrier capture into the QDs or release of carriers into the CS.

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References
[1] Rafailov E U, Cataluna M A and Sibbett W 2007 Nat. Photonics 1 395
[2] Liu H C, Gao M, McCaffrey J, Wasilewski Z R and Fafard S 2001 Appl. Phys. Lett. 78 79
[3] Lagatsky A A, Bain F M, Brown C T A, Sibbett W, Livshits D A, Erbert G and Rafailov E U 2007 Appl. Phys. Lett. 91 231111
[4] Morris D, Perret N and Fafard S 1999 Appl. Phys. Lett. 75 3593
[5] Müller T, Schrey F F, Strasser G and Unterrainer K 2003 Appl. Phys. Lett. 83 3572
[6] Yarotski D A, Averitt R D, Negre N, Crooker S A, Taylor A J, Donati G P, Stintz A, Lester L F and Malloy K J 2002 J. Opt. Soc. Am. B 19 1480
[7] Turchinovich D, Pierz K and Jepsen P U 2003 phys. stat. sol. (c) 0 1556
[8] Cooke D G, Hegmann F A, Mazur Y I, Ma W Q, Wang X, Wang Z M, Salamo G J, Xiao M, Mishima T D and Johnson M B 2004 Appl. Phys. Lett. 85 3839
[9] Prasankumar R P, Scopatz A, Hilton D J, Taylor A J, Averitt R D, Zide J M and Gossard A C 2005 Appl. Phys. Lett. 86 201107
[10] Lui K P H and Hegmann F A 2001 Appl. Phys. Lett. 78 3478
[11] Nielsen T R, Gartner P and Jahnke F 2004 Phys. Rev. B 69 235314
[12] Feldmann J, Nunneke J, Peter G, Göbel E, Kuhl J, Ploog K, Dawson P and Foxon C T 1990 Phys. Rev. B 42 5809
[13] Haiml M, Grange R and Keller U 2004 Appl. Phys. B: Lasers Opt. 79 331