# Ultrafast Spectroscopy of QD Structures for Mid-Infrared Applications

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## 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. However, it has turned out that a number of scattering processes, including multiphonon emission, electron-electron scattering, and electron-hole scattering can circumvent the phonon bottleneck, leading to very different capture and relaxation times from approximately one to above hundred of picoseconds. This discrepancy is still a major problem when the suitability of QDs for optoelectronic applications as well as for applications in quantum information processing has to be evaluated. Therefore it is essential to study fundamental optical properties of self-assembled InAs quantum dots within a GaAs host matrix or within a GaAs/AlAs quantum well superlattice in all relevant spectral regions from the NIR to the THz.

## Experiments

For this purpose, we developed a multi-purpose spectroscopy system that can fulfill the special requirements for ultrafast time-domain spectroscopy in the near- and mid-infrared, while considering the dispersion properties of the short laser pulses as well as highly sensitive detection schemes in order to allow the study of isolated, single quantum dots. Further, this system offers all standard characterization methods for single quantum dot investigations and ensemble experiments such as micro-photoluminescence, photoluminescence excitation and photocurrent spectroscopy.

## QD Ensembles

The necessary fundamental studies on QDs are always accompanied by the investigation of quantum dot based mid-infrared photodetectors with and without additional AlAs quantum wells in the GaAs host matrix. Here we developed a suitable model to describe the effects of vertical dot alignment within these structures. From this model, we can correctly predict the spectral photoresponse of such devices based on the growth parameters of the GaAs/AlAs quantum well system and some basic quantum dot parameters such as its exciton ground state energy. Further, it allows predicting and optimizing the polarization characteristics of these devices and explaining the emission spectra of a quantum cascade structure with embedded quantum dots in its active region [1]. As expected, comparison between the model and the experiments indicated a strong influence of the quantum dot potential onto the superlattice structure and vice

versa, the dots energetic spectrum is modified due to the presence of the GaAs/AlAs superlattice [2].

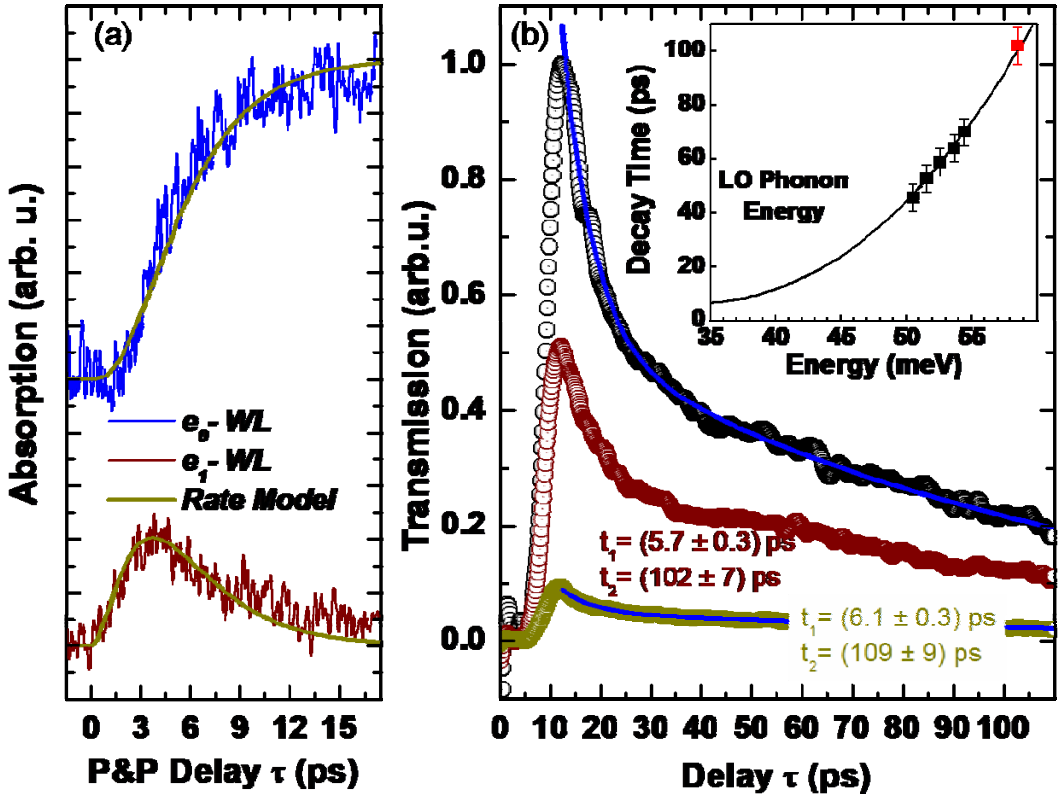


Fig. 1: Time-resolved differential transmission spectroscopy in a NIR pump, THz-probe configuration (a) and in a THz-pump, THz-probe configuration. Method (a) requires optical carrier injection into the QD via the NIR pump pulse, while method (b) traces the decay of excited doping electrons inside the dot. The inset in (b) relates the measured intersublevel decay to a polaronic decay mechanisms as described in [3].

In a parallel approach, we studied the depopulation dynamics of the excited quantum dot exciton and electron states by means of ultrafast interband pump and intersubband probe spectroscopy [4], or by single-color mid-infrared differential transmission spectroscopy on quantum dot ensembles. The related relaxation times vary between only a few and more than hundred picoseconds and strongly suggest extending our investigations to single quantum dots in order to exclude ensemble related effects such as Coulomb interactions within and between different dots [5]. Nevertheless, the obtained excited state lifetimes already point towards a significant influence of phonon related relaxation mechanisms within the quantum dots. Consequently, single quantum dot spectroscopy will also serve as a tool to further characterize these mechanisms. Within these experiments under resonant and non-resonant continuous wave laser excitation, we obtain important information about the energy level redistribution for multi-carrier complexes within a single quantum dot. Photoluminescence excitation studies further reveal and underline possible coupling mechanisms between the discrete quantum dot states and the surrounding matrix via phonons. Therefore, the quantum dot cannot be considered as isolated subsystem within the host matrix, but the variation of the coupling efficiency to different phonon modes for given intersublevel transition within the different dots suggests to further investigate those quantum dots by means of time-resolved spectroscopy for which longer relaxation times might be expected.

## QD Ensembles

In a first step we had to evaluate the influence of higher filling states of the single quantum dot intersublevel dynamics. Therefore, we performed emission saturation pump and probe spectroscopy (ESS) for intense pulsed excitation into the wetting layer region of the single quantum dot as shown in Fig. 2 (a) and (b). From a corresponding rate equation model (Fig.2 (c)) we deduced that the main effect of a high carriers density in and around the quantum dot is the shortening of the carrier capture time into the highest discrete quantum dot states, but no significant acceleration of the relaxation process from the excited dot states into the exciton ground state can be observed for these highly excited multi-carrier complexes.

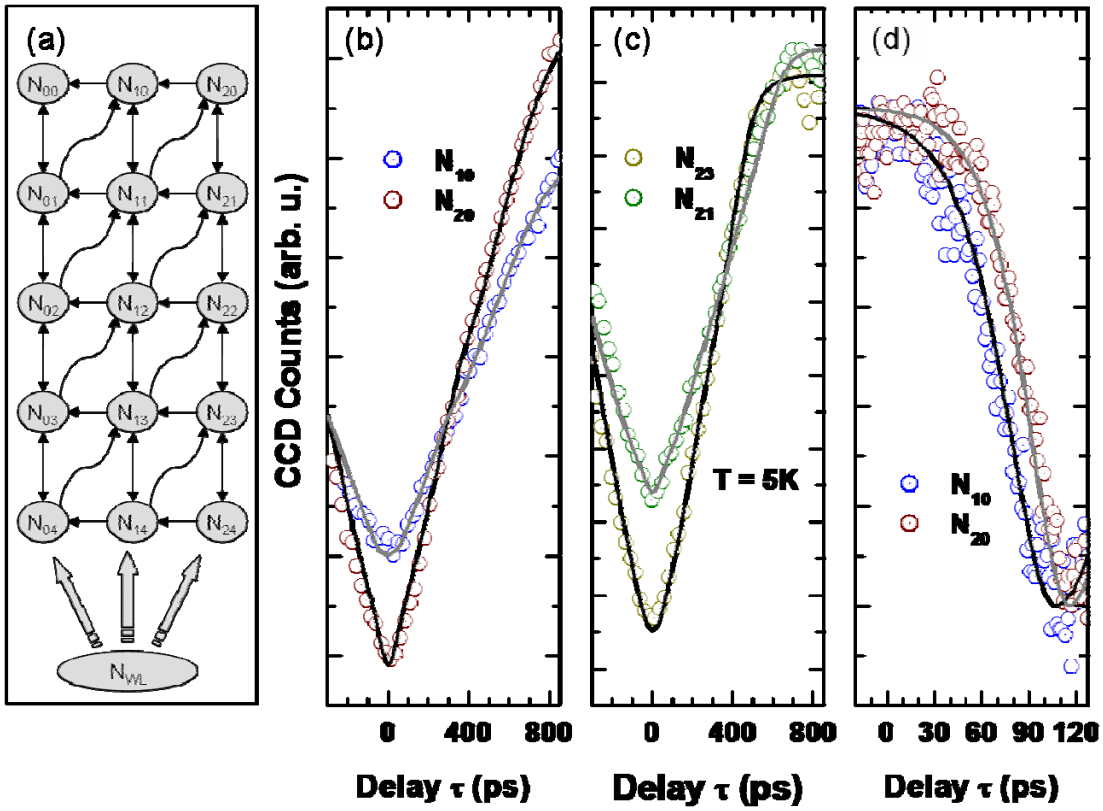


Fig. 2: (a) Scheme of the rate equation system describing the geminate capture and decay of excitons in a single QD. (b) and (c) show the ESS trace of the indicated S-states ($N_{10}$, $N_{20}$) and P-states ($N_{21}$, $N_{23}$) of a single QD. (d) Interband pump and intersubband probe spectroscopy on the same QD. All experiments lead to intersublevel relaxation times in the range of 60 to 80 ps.

Then we measured the relaxation time from the wetting layer into the exciton ground state by interband pump and intersubband probe spectroscopy, while monitoring the population of the exciton ground state via its electron-hole-pair recombination radiation. The result is shown in Fig. 2 (d). Again we can determine the intersublevel relaxation time between the S- and P-states of the QD to be in the range of several tens of picoseconds. Additionally, to our knowledge, this is also the first time that the influence of ultrafast mid-infrared excitation on the population state of a single self-assembled quantum dot is observed in the time-domain.

## Conclusion

As shown in the previous paragraphs, these experiments directly confirm the intersublevel relaxation time to be in the range of several tens of picoseconds, a period that is sufficiently long to use InAs quantum dots in THz related optoelectronic as well as in quantum information processing related applications. Further, the results underline the importance of considering ensemble related effects into the design of QD based devices, since these effects can affect the relaxation times inside the dots strongly. It has to be noted here that until today mostly interband and intraband properties of QDs have been exploited. Our results now strongly motivate to focus also on intersublevel related applications in QDs and QD molecules.

## Acknowledgements

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