Dr Thomas Moldaschl

Ultrafast Spectroscopy of Self-Assembled Quantum Dots

Supervisor: Karl Unterrainer

ABSTRACT

The scope of this thesis is to show the limitations of ensemble spectroscopic attempts with high excitation intensities in self-assembled Indiumarsenide-Galliumarsenide quantum dots, as well as to provide a solution to circumvent these problems through the construction of an optical spectroscopy setup that is capable of accessing individual quantum dots.

Fundamental quantum optics experiments, which can be realized with quantum dots, require the application of strong coherent radiation fields. Time-resolved spectral hole burning spectroscopy is employed on a self-assembled quantum dot sample, to coherently control the excitonic ground state. The corresponding high intensity optical field results in the observation of parasitic effects. The most significant manifestations are the emergence of a broadened absorption line and luminescence from the quantum dots at energies higher than the excitation energy. These effects are explained through a two-photon absorption mediated carrier relaxation. The carriers, generated through two-photon absorption in the Galliumarsenide host matrix, relax back into the quantum dots, where they either decay radiative (causing luminescence) or supplement resonantly created excitons and thereby shift their energies (absorption line broadening), which destructs any coherence. Another observation is the two-photon biexciton creation. Optical selection rules permit the simultaneous absorption of two photons with subsequent creation of a biexciton. This is also portrayed through the emergence of two additional spectral holes. Unlike the twophoton mediated charging, however, it does not influence the resonantly created excitons.

The second part is dealing with the development and characterization of a time- and energy-resolved pump-probe setup which is capable of measuring single quantum dot absorption. This setup uses acousto-optic modulators as amplitude modulators with frequencies in the Megahertz region and spectral shapers for both, pump and probe. A reduction in the number of quantum dots is equal to the reduction in the necessary excitation energy, thereby reducing the parasitic effects. Furthermore, the direct observation of the physical properties of single quantum dots is possible, not masked by the huge ensemble size. The proof of principle of this setup is demonstrated through the observation of quantum dot excitonic transitions as well as two-photon absorption in the Galliumarsenide host matrix. It bears great prospects for further measurements.