Two-color Femtosecond Spectroscopy of Blue-Shifted InAs/AlGaAs Quantum Dots

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Ultrafast relaxation phenomena in self-organized InAs/AlGaAs quantum dots with ground state transition energies of 1.42 eV are analyzed with different methods of femtosecond spectroscopy. A time-correlated luminescence experiment of an ensemble of InAs islands reveals a picosecond relaxation mechanism of carriers within the quantum dots. Moreover, modifications of the quantum dot properties due to excitation of the surrounding wetting layer is found. Employing a shadow-mask technique, a structure containing only a few zero-dimensional objects is studied. Selectively creating electron–hole pairs in excited states of approximately ten quantum dots, we observe characteristic line shifts of their ground state absorption in a two-color femtosecond transmission experiment. This finding demonstrates the feasibility of ultrafast studies of single self-assembled quantum dots.

1. Introduction In semiconductor quantum dots, material of low band gap energy is embedded in a wide-gap matrix. Due to the typical length scale of 10 nm, the electronic states are fully quantized. As a consequence, these quasi-zero dimensional systems exhibit properties comparable to those of atoms. Especially, single quantum dots show optical line spectra and are often referred to as artificial atoms [1–3]. Extraordinary long polarization dephasing times of several hundred picoseconds have been reported [4].

Due to their unique properties, quantum dots are discussed for potential implementations of solid-state based quantum information processing. Specifically, isolated quantum dots may be manipulated coherently with ultrashort laser pulses. Especially, basic ingredients of quantum computation such as the quantum-controlled NOT operation may be implemented in a single quantum dot [5].

In this paper, we report on the current status towards the experimental realization of quantum-coherent manipulations on single self-assembled InAs/AlGaAs quantum dots. Sections 2 and 3 introduce the material system and the experimental concepts that pave the way to sensitive femtosecond experiments on these zero-dimensional objects. Section 4 presents experimental results on the dynamical properties of a large ensemble of quantum dots. On this stage of the experiment, interesting insight into the ultrafast relaxation processes of electronic excitations within the quantum dots is given. Section 5 comprises the first experimental results of a two-color femtosecond experiment on a system consisting of only a few quantum dots. Our findings demonstrate the realizability of quantum-coherent operations via this experimental scheme.

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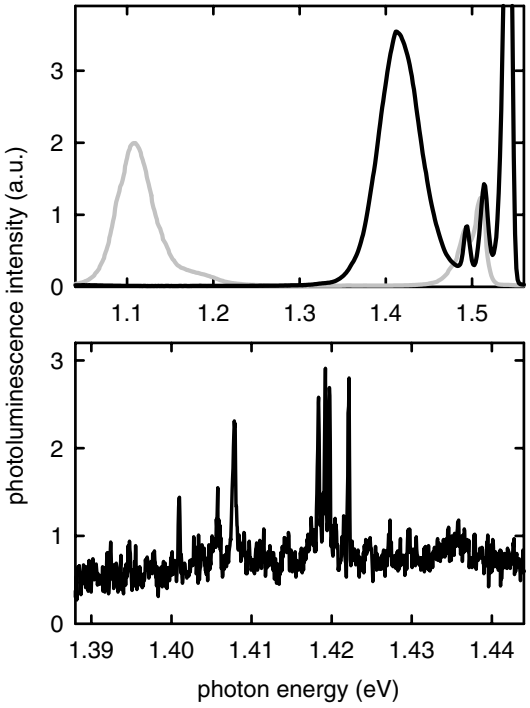

Fig. 1. Upper part: photoluminescence of a large ensemble of InAs/AlGaAs quantum dots at a lattice temperature of $T_L = 4.2$ K: as-grown (grey line) and after the annealing procedure described in the text (black line). The sample is excited with a cw laser at 1.96 eV; lower part: photoluminescence observed through a 350 nm shadow mask at the same experimental conditions. Note the different scale of the abscissa

2. Blue-shifted InAs/AlGaAs Quantum Dots We study self-organized InAs quantum dots grown on an $Al_{0.3}Ga_{0.7}As$ layer capped with two monolayers of GaAs [6]. This procedure allows for the growth of high-quality quantum dots with a large confinement potential. As depicted in Fig. 1, the as-grown sample exhibits a ground state luminescence around 1.1 eV (grey line), slightly blue-shifted as compared to the standard InAs islands embedded in GaAs. After an annealing step of 160 s at a temperature of $T = 1000$ K, the low temperature luminescence is found at photon energies around 1.42 eV (black line in the upper part of Fig. 1). This drastic blue-shift is attributed to an interdiffusion of aluminum into the InAs islands. The luminescence linewidth of 55 meV points towards a broad distribution of sizes and material compositions within the quantum dot ensemble. An additional prominent luminescence peak is found at 1.515 eV indicating the energetic position of both the band edge of the GaAs substrate and the wetting layer surrounding the InAs islands.

To overcome the problem of inhomogeneous broadening of the quantum dot ensemble, single zero-dimensional structures are to be isolated on the sample. To this end, the InAs layer is deposited without rotating the wafer during the growth procedure. Therefore, regions with a low quantum dot density may be found on the sample. Additionally, nanometer scale aluminum shadow masks are deposited on the wafer. The lower part of Fig. 1 depicts the photoluminescence detected through a 350 nm aperture. Narrow emission lines between photon energies of 1.40 eV and 1.43 eV are clearly identified and correspond to the ground state luminescence of a few quantum dots. As a consequence, this material system allows for optical experiments in a spectral window where femtosecond Ti:sapphire lasers and sensitive Si-detectors may be operated.

3. Experimental Concept Our experimental setup relies on a special two-color femtosecond Ti:sapphire laser that provides two independently tunable, perfectly synchronized laser pulses with durations between 13 fs and 120 fs [7]. The additional implemen-

tation of dispersion-free pulse shapers [8] allows for the generation of laser pulses with a smaller bandwidth of $\Delta E \geq 1.5$ meV and a corresponding transform-limited duration of up to 1 ps. The temporal separation of the pulses is adjusted via a variable delay line. The sample is mounted on the cold finger of a liquid helium microscope cryostate operated at lattice temperatures of $T_L \geq 3$ K. The laser light is focussed onto the sample employing a dispersion-free reflecting mirror objective with a high numerical aperture of NA = 0.4 mounted on piezo transducers outside the cryostate. An aspheric lens on the rear side of the sample is used to collect both the transmitted laser light and the sample luminescence. After passing a monochromator (typical spectral resolution: 1 meV) the light is analyzed with a single photon counting system or a silicon avalanche diode for luminescence or transmission experiments, respectively.

4. Results for an Ensemble of Quantum Dots First, the properties of blue-shifted InAs quantum dots at a lattice temperature of $T_L = 3$ K are studied with the experimental method of time-correlated luminescence. Taking into account the size of the laser spots on the sample and the density of quantum dots on the wafer, the number of zero-dimensional structures contributing to the observations is estimated to be $N \approx 1000$.

A first 100 fs laser pulse centered at a photon energy of 1.47 eV resonantly generates localized electron–hole pairs within the quantum dots (interband transitions (1) in Fig. 2. After a certain delay time t_D, a second pulse (probe) with a duration of 200 fs and a variable central photon energy is used to analyze the transient modifications of the sample. To this end, information on the absorption of the probe laser pulse is obtained via the emission changes at the ground state luminescence of the InAs islands at 1.355 eV.

First, we discuss the case of resonant carrier generation in the wetting layer or the GaAs substrate via the interband transitions (2) and (3) in Fig. 2. For this case, the experiment is sensitive espe-

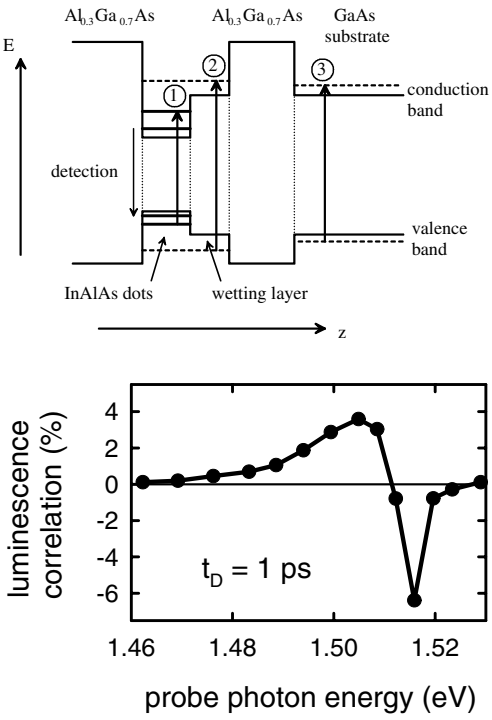

Fig. 2. Upper part: schematic band diagram of the quantum dot sample. The interband transitions employed in the experiment are denoted with (1), (2), and (3) and explained in the text; lower part: time-correlated luminescence for various probe photon energies observed at a delay time of $t_D = 1$ ps after exciting the quantum dots with a 100 fs pulse centered at 1.47 eV. The lattice temperature is $T_L = 3$ K and the sample luminescence is detected at 1.355 eV with an energy bandwidth of 10 meV

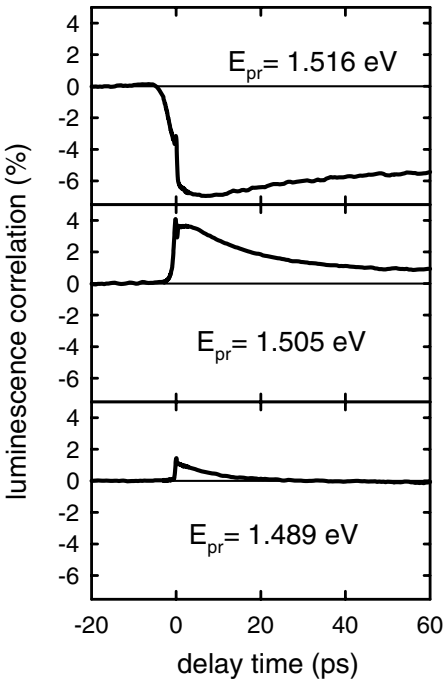

Fig. 3. Time-correlated luminescence as a function of delay time for various probe photon energies E_{pr} given in the figure. The sample is excited with a 100 fs pulse at a photon energy of 1.47 eV. The lattice temperature is $T_{\mathrm{L}} = 3$ K

cially to the occupation of the quantum dots. Specifically, electrons and holes generated in the wetting layer may scatter into the quantum dots efficiently only if the energy levels are unoccupied. Correspondingly, a negative luminescence change is found at probe photon energies of 1.516 eV (compare lower part of Fig. 2) after excitation with the pump pulse. As may be seen from the topmost curve in Fig. 3, this transient modification decays on a timescale of 100 ps related to the carrier lifetime in the quantum dots.

Surprisingly, the time-correlated luminescence signal decreases rapidly for larger probe photon energies in Fig. 2. Apparently, the capture process of uncorrelated electron–hole pairs from the wetting layer into the quantum dots differs qualitatively from the case of excitonic states near the band edge.

A distinctl different behavior is found when the probe pulse generates electron–hole pairs within the quantum dots (generation of localized carriers via the interband transitions (1) in Fig. 2). As may be seen from the time-correlated luminescence for the probe photon energies of 1.505 eV and 1.489 eV in Fig. 3, an enhanced probe-induced luminescence is found after excitation with the pump pulse. This finding reflects the modifications of interband transitions (1) in Fig. 2 due to the carrier generation in the dots: occupying self-organized quantum dots with electron–hole pairs generally results in an energetic red-shift of interband transitions of several meV due to Coulomb renormalization [9, 10]. As a consequence, a larger number of interband transitions is resonant with the probe pulse after carrier injection. The picosecond dynamics of the time-correlated luminescence signal is related to an intra-quantum dot relaxation mechanism, e.g. by polar optical scattering of carriers into energetically lower quantum dot states. However, it is difficult to obtain a more precise understanding of the relaxation mechanisms since a large, inhomogeneously broadened ensemble of quantum dots contributes to the signal.

5. Results on Isolated Quantum Dots In order to obtain a more detailed picture of the dynamical properties of single self-assembled quantum dots, we now turn to the investigation of shadow masks that isolate systems of a few zero-dimensional InAs islands on the specimen. Figure 4 depicts the photoluminescence spectra of a 350 nm aperture detected with an energetic resolution of 2 meV for various excitation powers.

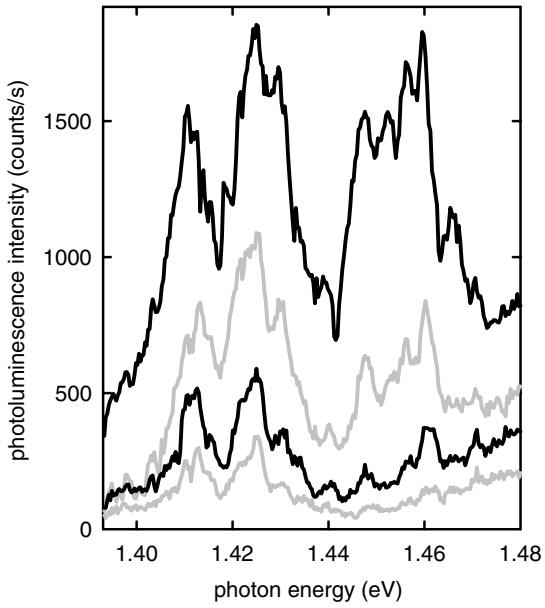

Fig. 4. Photoluminescence detected through a 350 nm aperture at a lattice temperature of $T_L = 4.5$ K. The specimen is excited with a cw laser at 1.96 eV. The different curves correspond to various excitation powers: 50 μW (upper black line), 25 μW (upper grey line), 12 μW (lower black line), and 6 μW (lower grey line). The spectral resolution is set to 2 meV

Free carriers are generated with a cw laser at 1.96 eV at a lattice temperature of $T_L = 4.5$ K. As may be seen from the spectra for low excitation powers, the emission from the nanoaperture is dominated by the ground state luminescence of approximately five quantum dots around 1.42 eV. The intensity of these lines is found to increase slightly sublinearly with the excitation power indicating a beginning saturation of the quantum dot occupation. In contrast, the intensity of the luminescence lines found at photon energies between 1.446 eV and 1.462 eV exhibit a clearly superlinear increase with the excitation power. Apparently, these features correpond to the emission of excited states of the quantum dots under the nanoaperture. Our findings indicate an energetic separation of the first excited state transition in the order of 40 meV which is in agreement with studies on conventional self-organized quantum dots [11].

A comparable aperture with a diameter of 2 μm and a corresponding number of quantum dots in the order of $N \approx 100$ is now studied in a two-color femtosecond transmission experiment. In contrast to the results of the time-correlated luminescence experiment described above, very low signal levels are expected due to the weakly absorbing quantum dot monolayer. However, a transmission experiment utilizing broadband probe pulses allows for studies with optimum energy–time uncertainty.

The specimen is excited with a 300 fs pump pulse of a narrow bandwidth of 5 meV centered at 1.462 eV. As a consequence of this choice, only a subensemble of $N \approx 10$ quantum dots is expected to be excited via resonant absorption into the first excited excitonic states. The broadband 50 fs probe pulse is centered at 1.43 eV and encompasses the interesting spectral window around the ground state transition energies of the selectively excited quantum dots. The transmitted probe light is spectrally dispersed with a resolution of 1.5 meV. Figure 5 depicts the spectrally resolved transmission changes obtained for a delay time of $t_D = 1$ ps. As the most striking feature, a dispersive structure is well resolved at 1.424 eV, red-shifted by 38 meV with respect to the excitation. This observation points towards a slight redshift of the ground state transition energies of the occupied quantum dots due to Coulomb renormalization. Comparable energetic shifts have been found in photoluminescence experiments [9, 10] as well

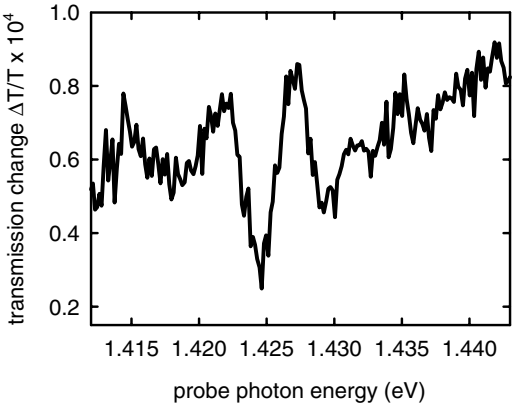

Fig. 5. Spectrally resolved transmission changes detected through a 2 μm aperture for a delay time of $t_D = 1$ ps after excitation with a 300 fs pulse centered at 1.462 eV. The lattice temperature is $T_L = 4.5$ K

as in theoretical simulations [12, 13]. In future experiments, these energetic shifts of an excitonic state may be exploited in quantum-correlated operations with two interband transitions serving as qubits [5].

6. Conclusion In conclusion, we have studied self-organized InGaAs/AlGaAs quantum dots with different methods of femtosecond spectroscopy. Investigating a large ensemble of InAs islands in a time-correlated luminescence experiment, a picosecond relaxation mechanism within the quantum dots is found. The additional implementation of nanoapertures on the sample allows to study systems containing only a few zero-dimensional objects. The energetic separation of the first excited excitonic transition and the ground state is found to be approximately 40 meV paving the way for ultrafast switching operations, e.g. implementations of a quantum-controlled NOT gate which may be addressed on a femtosecond timescale. In the first realization of a two-color femtosecond transmission experiment on isolated quantum dots, we observe an energetic red-shift of the ground state transition energy after occupation of the first excited state. As a consequence, this technique may soon demonstrate ultrafast studies on single self-assembled quantum dots and coherent manipulations in a system of two excitonic qubits.

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