Nonlinear Optical Microscopy of a Single Self-Assembled InGaAs Quantum Dot

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Abstract. A single InGaAs/GaAs quantum dot is addressed in a two-color femtosecond transmission experiment. We find bleaching signals arising from individual excitonic interband transitions thus providing a scheme for ultrafast optical read-out of one artificial atom.

Semiconductor quantum dots (QDs) are promising building blocks for optoelectronic devices. Especially, several schemes have been proposed to implement quantum-coherent operations in QDs [1]. These schemes rely on both ultrafast optical switching and optical read-out of a single self-assembled QD – an experimental requirement that has not been realized so far.

In this contribution, we present the first experimental realization of a two-color femtosecond transmission experiment of a single InGaAs/GaAs QD [2]. The transmission changes of one aperture with 450 nm diameter are analyzed after femtosecond photoexcitation of the QD. Most strikingly, the nonlinear optical response clearly resembles several distinct excitonic transitions of the QD under investigation. The QD typically generates modifications of the probe photon flux in the order of 10^{-5}. The pulse energies necessary for both the manipulation and read-out of the single QD are as low as a few femtojoule.

The nanostructures are InGaAs/GaAs QDs with excitonic ground state transitions centered at 1.35 eV. In addition, aluminum shadow masks are deposited on the specimen with diameters between 200 nm to 450 nm. As a consequence, single QDs may be precisely addressed using standard free-space optics.

In this study, we focus on a single InGaAs QD characterized in Fig. 1(b). The specimen is mounted on the cold finger of a microscope cryostat (T = 4 K) for all present experiments. The photoluminescence (PL) spectrum observed for a pulsed low intensity excitation at E = 1.51 eV shows two pronounced, resolution limited peaks. Varying the excitation power, these transitions are identified as excitonic (X, E = 1.3735 eV) and biexcitonic transition (2X, E = 1.3696 eV). The confinement energy in this QD is approximately 35 meV and 15 meV for electrons and holes, respectively, i.e. much larger than for localized excitons in disordered quantum wells.

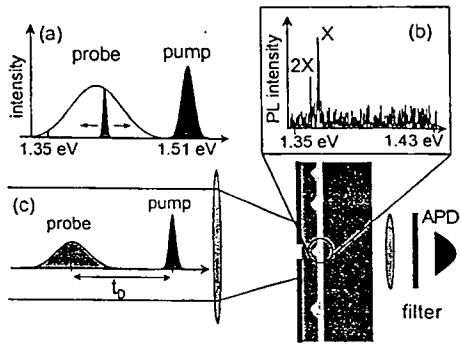

Fig. 1: Pump and probe pulses of the experiment (b): PL of the nanoaperture for the case of a pulsed, low intensity excitation. (c) Optical setup: the pulse strains are focussed through the metallic shadow mask. The transmitted light is detected with an avalanche photodiode (APD) after spectral filtering. The sample is held at T = 4 K in a microscope cryostate.

The experimental scheme is sketched in Fig. 1(a) and (c). Two synchronized femtosecond pulse trains are derived from a two-color Ti:sapphire laser. The 100 fs pump pulse is tuned to the absorption continuum of the wetting layer beneath the InGaAs QD thus efficiently filling the nanostructure with nonequilibrium carriers. The 30 fs probe pulse encompasses the entire spectral range from 1.36 eV to 1.44 eV giving access to both QD interband transitions and the wetting layer band edge. Before transmission through the sample, probe pulses with a spectral width of 1.2 meV are extracted from this broad band pulse using a pulse-shaper. As a result, transform limited pulses of 1.5 ps duration are used as test pulses for the ultrafast transmission experiment. As depicted in Fig. 1(c), both excitation and time-delayed probe beams are focussed onto the sample using a dispersion-free reflecting mirror objective that allows for a typical spot diameter of 1.5 μm. The light transmitted through the nanoaperture is collected with a microscope objective, filtered and analyzed with an avalanche photo diode.

We now turn the analysis of the nonlinear optical response of this specific quantum dot. Fig. 2(b) displays the relative transmission changes observed at a time delay of t_D = 20 ps for two different intensities of the pump pulse. For comparison, the corresponding luminescence is depicted in Fig. 2(a). Most strikingly, we clearly identify a bleaching signal centered at the excitonic transition X even for a pump intensity as low as 2 fJ. Raising the pump power to 10 fJ, the amplitude of the ground state bleaching remains at a level of 3×10^{-5} indicating a saturation of this interband transition. As a consequence, this value may be seen as an estimate for both the maximum transmission changes associated with a single InGaAs QD in agreement with recent electromodulation studies [3].

For photon energies larger as compared to these s-shell transitions of the QD, the PL spectra in Fig. 2(a) show a series of emission peaks related to recombination from different multiexciton complexes. These excitonic transitions partially overlap with the emission of localized states in the wetting layer at 1.41 eV. Most of these luminescence peaks have their direct counterpart in the

nonlinear optical response. For probe photon energies approaching the band edge of the wetting layer at 1.41 eV, we find a strongly structured nonlinear optical response The amplitude of these transmission changes is much larger when compared to the ground state transition X is consistent with previous experiments [4].

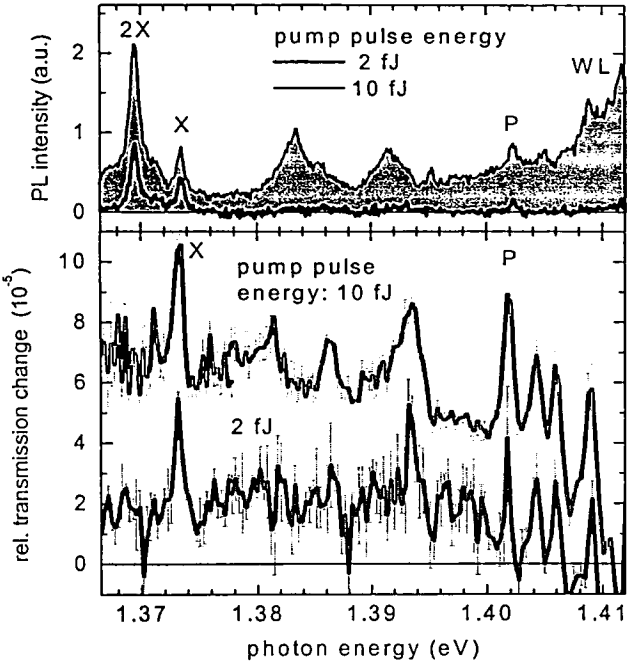

Fig. 2(a): PL spectra for two different excitation intensities. (b) Transmission changes detected at $t_D = 20$ ps after excitation of the sample with a 100 fs pulse centered at 1.51 eV.

In conclusion, we have demonstrated an experimental tool to both manipulate and read-out a single self-assembled quantum dot on ultrashort timescales. One artificial atom beneath a shadow mask of 450 nm diameter generates modifications of a probe photon flux in the order of 10^{-5}. These transmission changes clearly reflect the optical bleaching of different multiexciton configurations. This work is supported by the DFG in the framework of the Sonderforschungsbereich SFB631.

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