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Nonlinear optical microscopy of a single self-assembled InGaAs quantum dot

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Carrier dynamics in InGaAs/GaAs quantum dots is analyzed with highly sensitive two-color femtosecond transmission spectroscopy. Especially, a single artificial atom is addressed in the optical near-field of a nanometer scale shadow mask. Resonantly exciting the wetting layer beneath the nanoisland, we detect transmission changes of the quantum dot with narrowband femtojoule probe pulses. We find bleaching signals in the order of 10^{-5} that arise from individual interband transitions. Moreover, the nonlinear optical response reveals a picosecond dynamics associated with carrier relaxation in the quantum dot. As a result, we have demonstrated an ultrafast optical tool for both manipulation and read-out of a single self-assembled quantum dot.

Semiconductor quantum dots (QDs) are attractive for studying the physics of quasi zero-dimensional solid state systems as well as for novel optoelectronic applications such as ultra-low threshold laser devices. Especially, self-organized III-V semiconductor QDs combine remarkably long coherence times of excitonic transitions with the potential for ultrafast optical switching [1]. As a result, coherently manipulating zero-dimensional nanostructures with femtosecond laser pulses is a promising concept for solid state-based quantum information processing [2]. However, most of these concepts require both ultrafast optical switching and all-optical read-out of a single QD [3] - a demanding experimental task. Until now, an analysis of the nonlinear optical response of a single QD has only been realized for the case of weakly confined excitons in disordered quantum wells [4, 5]. For isolated self-assembled QDs, Rabi oscillations have been studied extensively, thus proving the possibility of precise single qubit rotations [6–8]. However, the time-resolved all-optical read-out of a single self-organized QD remains to be demonstrated.

In this contribution, we present the first experimental realization of a two-color femtosecond transmission experiment of a single InGaAs/GaAs QD [9]. The nanostructure is isolated combining a low-density growth technique with nanometer-scale aluminum shadow masks. The transmission changes of one aperture with 450 nm diameter is analyzed after femtosecond photogeneration of electron-hole pairs in the wetting layer beneath 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}, thus being accessible in a shot noise limited experimental setup. The pulse energies necessary for both the manipulation and read-out of the single QD are as low as a few femtojoule. The transmission changes exhibit a pronounced picosecond dynamics as recently also demonstrated for the case of an ensemble of similar QDs [10].

The nanostructures of this study are self-organized InGaAs QDs grown on a GaAs. The excitonic ground state transitions of the InGaAs islands are centered around 1.35 eV. QD growth without rotation of the GaAs wafer allows to identify areas with a low QD density of a few artificial atoms per μm^2. In

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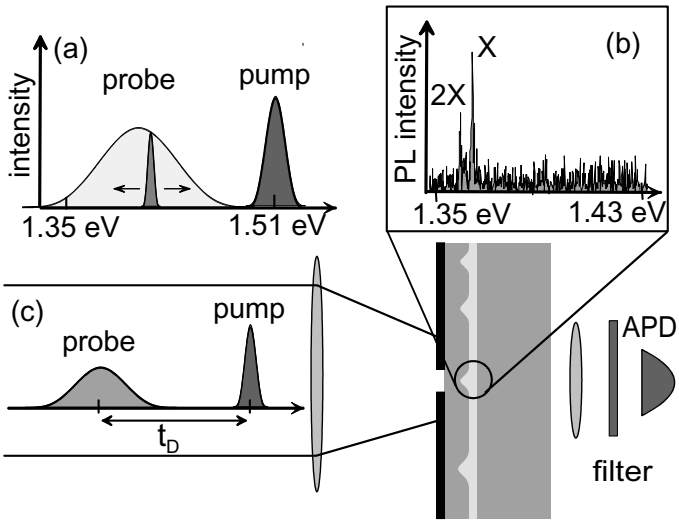

Fig. 1 (a) Excitation and probe pulses used in the experiment. The probe pulses of an energetic width of 1.2 meV are extracted from a broadband probe continuum using a pulse shaper (b) Photoluminescene of the nanoaperture for the case of a pulsed, low intensity excitation. (c) Scheme of the experiment: pump and probe pulse trains are focussed through the metallic shadow mask of 450 nm diameter. The transmitted light is detected with an avalanche photodiode (APD) after spectral filtering. The sample is held at $T_L = 4$ K in a microscope cryostate.

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 in a microscope cryostat ($T_L = 4$ K) for all present experiments. The photoluminescence 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.

The experimental scheme is sketched in Figs. 1(a) and (c). Two synchronized femtosecond pulse trains with 76 MHz repetition rate are derived from a two-color Ti:sapphire laser [11]. 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 low-dispersive pulse-shaping technique [12]. 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 with a numerical aperture of 0.4 that allows for a typical spot diameter of $1.5\,\mu$m. The light transmitted through the nanoaperture is collected with a microscope objective, filtered in order to reject transmitted pump photons and analyzed with an avalanche photo diode. Subsequently, the pump induced transmission changes are extracted using a lock-in technique with a 30 kHz modulation of the excitation pulse.

We now turn the analysis of the nonlinear optical response of this specific quantum dot characterized in Fig. 1(b). Figure 2(b) displays the relative transmission changes observed at a time delay of $t_D = 20$ ps after photoexcitation for two different intensities of the pump pulse. For comparison, the luminescence of the nanoaperture for these two excitation densities 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 under a shadow mask and the total ground state absorption in agreement with recent linear studies [13]. Additionally,

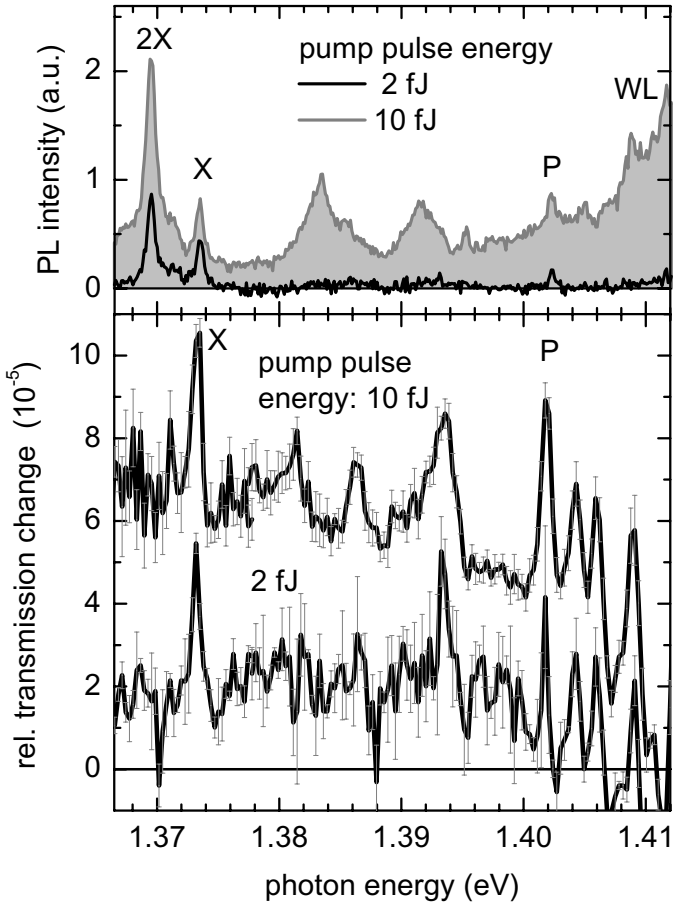

Fig. 2 (a) Photoluminescence (PL) spectrum for two different excitation intensities. The energy values indicated in the graph correspond to the pulse energy that is incident on the geometrical area of the shadow mask. (b) Transmission changes of the aperture detected at $t_D = 20$ ps after excitation of the sample with a 100 fs pulse centered at 1.51 eV. The spectral resolution of the experiment is determined by the energetic width of the tunable probe pulses of 1.2 meV. The data for the pump intensity of 10 fJ are shifted by 2 ordinate units for clarity.

a positive background signal appears especially for the higher excitation level of the study which might reflect a substrate induced optical nonlinearity. Surprisingly, we do not see a clear signature of the biexciton 2X in the transmission changes. For photon energies larger as compared to these s-shell transitions of the QD, the luminescence spectra in Fig. 2(a) show a series of broad emission peaks that are related to optical recombination from different multiexciton complexes. These excitonic transitions partially overlap with the emission of different localized states in the two-dimensional wetting layer with a band gap energy of 1.41 eV. Interestingly, most of these luminescence peaks have their direct counterpart in the nonlinear optical response. As an example, the luminescence peak P in Fig. 2(a) ($E_P = 1.4022$ eV) is paralleled by a remarkably sharp and pronounced nonlinear transmission peak in Fig. 2(b). For probe photon energies approaching the band edge of the wetting layer at 1.41 eV, we find a strongly structured nonlinear optical response that is related to different localized excitons in the wetting layer (only partially shown in Fig. 2). The amplitude of these transmission changes is one order of magnitude larger when compared to the ground state transition X. This finding is related to the giant transition dipole of these weakly confined excitonic complexes and is consistent with previous experiments [4].

We would like to note that our findings strongly depend on the delay time elapsed since photoexcitation (not shown in Fig. 2). As recently verified by ensemble measurements, we expect a filling of the excited QD states within a few picoseconds and a subsequent relaxation towards the excitonic ground state on a comparable timescale [10]. Especially, we find both the relative and the absolute intensitites of the different bleaching peaks in Fig. 2(b) to vary on the ps timescale. Moreover, also the spectral shape of

the transmission changes associated with some multiexciton configurations depends on the delay time. A deeper physical picture of these results remains to be developed.

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 is found to generate modifications of a probe photon flux in the order of 10^{-5} thus being accessible in a shot noise limited detection scheme. The transmission changes clearly reflect the optical bleaching induced by different multiexciton configurations. Extending the study to resonant QD pumping, we expect new insight into the physics of self-organized QDs and their potential applications in quantum information processing.

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References

[1] P. Borri et al., Phys. Rev. Lett. **87**, 157401 (2001).
[2] N. H. Bonadeo et al., Science **282**, 1473 (1998).
[3] F. Troiani, U. Hohenester, and E. Molinari, Phys. Rev. B **62**, R2263 (2000).
[4] T. Guenther et al., Phys. Rev. Lett. **89**, 057401 (2002).
[5] W. Langbein and B. Patton, Phys. Rev. Lett. **95**, 017403 (2005).
[6] T. H. Stievater et al., Phys. Rev. Lett. **87**, 133603 (2001).
[7] H. Kamada et al., Phys. Rev. Lett. **87**, 246401 (2001).
[8] A. Zrenner et al., Nature **418**, 612 (2002).
[9] M. Wesseli et al., Appl. Phys. Lett., in press (2006).
[10] S. Trumm et al., Appl. Phys. Lett. **87**, 153113 (2005).
[11] C. Fürst, A. Leitenstorfer, and A. Laubereau, IEEE J. Select. Topics Quantum Electron. **2**, 473 (1996).
[12] R. Grote and H. Fouckhardt, Opt. Expr. **4**, 328 (1999).
[13] B. Alen et al., Appl. Phys. Lett. **83**, 2235 (2003).