Femtosecond probing of few-fermion dynamics and deterministic single-photon gain in a single semiconductor quantum dot

R Bratschitsch1, T Thomay1, F Sotier1, J Korger1, T Hanke1, S Mahapatra2, A Frey2, K Brunner2 and A Leitenstorfer1

1Department of Physics and Center for Applied Photonics, University of Konstanz, D-78457 Konstanz, Germany
2Institute of Physics, EP3, University of Würzburg, D-97074 Würzburg, Germany

Email: Rudolf.Bratschitsch@uni-konstanz.de

Abstract. We report on femtosecond readout of the optical properties of a single CdSe/ZnSe quantum dot. Owing to the uncertainty principle, this timescale represents the ultimate limit for coherent quantum manipulation of such an artificial atom. After resonant excitation of a hot electron-hole pair the absorption of the fundamental exciton resonance is switched off via instantaneous Coulomb renormalization. Subsequently, optical gain builds up after ultrafast intraband relaxation. The speed of thermalization is dominated by the electron spin, since our system is charged permanently with one excess electron. When operating in the nonlinear regime, the number of quanta in a femtosecond light pulse may be changed by exactly ±1. We demonstrate that this deterministic single photon amplifier is characterized by a flat gain spectrum.

1. Introduction

Semiconductor quantum dots exhibit a discrete electronic level structure due to three-dimensional quantum confinement. Optically [1] and electrically [2] pumped single-photon emitters, as well as non-classical current sources [3, 4] have been implemented. Spin and charge degrees of freedom are hot candidates for solid-state quantum information processing. Since individual semiconductor nanocrystals typically consist of $10^4$ atoms, ensembles of quantum dots suffer inhomogeneous broadening. Therefore, the best insight into their physical properties and functionalities is gained studying single specimens.

Most optical studies on single quantum dots are based on the detection of spontaneous emission of photons. This process typically occurs on time scales longer than 100 ps. In contrast, transient absorption experiments with ultrashort laser pulses provide high temporal resolution. However, carrying out such experiments on single quantum systems is difficult since single-photon signals have to be filtered from an intense background. Up to now, experiments with femtosecond resolution were limited to single excitons in local minima of narrow GaAs/AlGaAs quantum wells with small confinement potential and giant transition dipole moments [5, 6], or four wave mixing studies of the coherent interband polarization [7].
2. Two-color femtosecond pump-probe experiment on a single semiconductor quantum dot

Here, we present the first femtosecond transient transmission measurements on a single quantum dot with large confinement potential [8]. We use a 700 fs pump pulse to resonantly excite the self-organized CdSe/ZnSe quantum system via a transition in the p-shell at a photon energy of 2.21 eV (Fig. 1). One electron-hole pair is generated by a pump with pulse area $\pi$.

![Figure 1](image)

**Figure 1.** Stationary light emission from the singly-charged CdSe/ZnSe quantum dot. The black line shows the luminescence emission intensity ($X^-$ PL) of the quantum system as a function of photon energy. A photoluminescence excitation spectrum ($X^-$ PLE) is given by the blue graph and provides information on electron-hole pair excitations with higher excess energy. The spectral envelopes of the pump and probe pulses provided by the two-color femtosecond fiber laser are depicted by the green and red lines, respectively.

We investigate a quantum dot that is charged with one electron from a donor in the slightly n-doped ZnSe matrix. Therefore, an excited electron-hole pair forms a three-particle state. The transmission change induced by the pump excitation is then measured as a function of delay time $t_D$ with a 180 fs probe pulse resonant to the fundamental trion transition $X^-$ at 2.11 eV. Our experiments are based on a fs-Er:fiber laser system that emits two independently tunable pulse trains with a relative timing jitter as low as 43 attoseconds at a repetition rate of 100 MHz [9,10]. In order to optimize the interaction of the probe photons with the single-electron quantum system, the CdSe/ZnSe dot is located in the optical near-field of a metallic nanoaperture. Energy information is obtained by spectrally dispersing the broadband probe beam transmitted through the sample with a resolution set to 200 $\mu$eV. Time-averaged parallel readout of $10^5$ spectral channels for the differential transmission is achieved by operating a CCD array in a lock-in mode at a modulation frequency of 30 Hz.

3. Results and discussion

The differential transmission change $\Delta T/T$ at the fundamental trion resonance $X^-$ is depicted as a function of delay time $t_D$ and probe photon energy in Fig. 2. At negative $t_D$, a weak signal is discernible starting considerably before zero time delay. A step-like increase in differential transmission occurs around $t_D = 0$, i.e. when pump and probe electric fields are arriving...
simultaneously at the sample. A constant region in $\Delta T/T$ follows, which persists until $t_D = +10$ ps. After an additional increase in transmission change, the signal remains stationary again for delay times $t_D$ beyond 20 ps.

![Figure 2](image.png)

**Figure 2.** Experimental differential transmission change $\Delta T/T$ at the fundamental trion resonance X as a function of pump-probe delay time and probe photon energy. The probe pulse energy is kept in the linear regime while the pump pulse excites one electron-hole pair. The perturbed free induction decay at negative time delays is marked by two red lines as a guide to the eye. Vertical black lines denote different spectral cuts at delay times $t_D = -2$ ps, $+2$ ps, and $+20$ ps (see figure 3).

To explain the phenomena underlying the dynamics found in Fig. 2, we focus on differential spectra for several fixed time delays $t_D$ (Fig. 3). At $t_D = -2$ ps, the probe pulse precedes the pump. In this situation, the test beam first creates a coherent polarization at the s-shell transition which is damped with the dephasing time corresponding to the sharp X resonance. The pump-induced perturbation of this free induction decay results in oscillations in the differential transmission spectrum which are symmetric to a positive peak centered at the fundamental trion resonance. This coherent phenomenon is known from spectrally resolved pump-probe measurements on narrow resonances [6,11,12]. It serves as a benchmark to prove that we are readily detecting femtosecond transmission signals from a strongly confined quantum dot. The lines in the upper panel of Fig. 3 represent fits to the data based on the optical Bloch equations [11]. In our model, the duration of the laser pulses is taken from the experiment, the onset of perturbation in the free induction decay is given by the time integral of the pump intensity, and the dephasing time is set to 5 ps to match the finite spectral resolution of our setup.

The differential spectrum shortly after excitation is sampled at a delay time of $+2$ ps (Fig. 3). In this case, we find a transmission increase with a Lorentzian line shape. The instantaneous onset of this feature around $t_D = 0$ is explained as follows: When the pump pulse creates an electron-hole pair in the p-shell (lower part of Fig. 3), the charge distribution in the dot is altered. Due to the small dielectric constant of II-VI semiconductors, the resulting Coulomb force strongly renormalizes the electronic states. As a consequence, the original trion resonance vanishes and the probe beam suffers no more absorption. Since our experiment measures the differential transmission change $\Delta T/T = (T_{\text{pump on}} - T_{\text{pump off}})/T$, we observe a maximum at the fundamental trion transition.
The signal at the beginning of the second plateau in $\Delta T/T$ is probed at $t_D = +20$ ps (Fig. 3). The delayed increase in differential transmission after the instantaneous Coulomb bleaching is due to energy relaxation of the hot electron-hole pair into the s-shell of the quantum dot: If the carriers excited into the p-shell have reached the lowest states in the potential wells (lower part of Fig. 3), the fundamental trion resonance $X^-$ is restored, but now under inversion conditions. When the probe pulse arrives at the system, it experiences optical gain due to stimulated emission.

The absolute amplitude of the differential transmission signal that we measure in the plateau at $t_D > 20$ ps is intriguing: Assuming complete intraband relaxation and negligible interband recombination, one would expect an increase of $\Delta T/T$ by exactly a factor of 2, as compared to the pure bleaching signal immediately after excitation. Instead, the observed increase saturates at a factor of 1.5 for delay times between $t_D = 20$ ps and 50 ps (upper part of Fig. 3). This finding indicates that inversion at the trion resonance and optical gain are established on a 15 ps time scale only after 50%
of the pump pulses. This effect is caused by the electron spin: There is equal probability for parallel and antiparallel relative alignment of the spins of the electron resident in the dot due to doping and the photoexcited electron since no magnetic field is applied to the sample. If the two electron spins form an antiparallel singlet pair, they are allowed to occupy the same spatial wave function in the s-shell of the potential well (lower part of Fig. 3). Hence relaxation into the trion ground state proceeds quickly. If the two spins form a parallel triplet pair (lower part of Fig. 3) relaxation of the photoexcited electron into the s-shell is forbidden by the Pauli exclusion principle. In our system, the only degree of freedom available to flip the electron spin from $s_z = -1/2$ to $s_z = +1/2$ is a Coulomb collision promoting the hole from its heavy hole ground state with total angular momentum projection $j_z = \pm 3/2$ into a light hole state with $j_z = \pm 1/2$. In order to fulfill energy and momentum conservation, a phonon scattering event of at least second order has to occur simultaneously. Obviously, this process operates on a time scale much longer than 50 ps. Indeed, only after $t_0 = 100$ ps, $\Delta T/T$ starts to decrease slowly because of the interband recombination time of the fundamental trion state of $\tau_{rad} = 480$ ps as measured by a time-resolved photoluminescence experiment.

The data presented in Figs. 2 and 3 have been measured with weak probe pulses in the linear regime. If we increase the probe pulse area to $\pi$, exactly one photon should be absorbed from the 180 femtosecond probe pulse if it passes the quantum dot in its ground state. Even more excitingly, exactly one photon is added to the femtosecond pulse if it hits the system in the fundamental trion state. As explained before, we can switch between both situations on a time scale as fast as 20 ps, while no interaction with the system would occur in the intermediate time interval due to Coulomb renormalization. To start exploring the physics of coherent single photon gain triggered by ultrashort light pulses, we have carried out the following experiment: The differential transmission at $t_D = 20$ ps is measured for increasing average powers of the probe beam starting at $P_{probe} = 75$ $\mu$W (Fig. 4(a)). In this case, the pulse area amounts to a value of 0.3 $\pi$. We assume that the quantum dot is initially in the inverted state $|X\rangle$. The Bloch vector of the two-level system is then turned downwards by an angle of 0.3 $\pi$, creating a considerable amount of interband polarization. We plot the temporal envelope of the probe electric field (blue line) and of the polarization amplitude (green line) derived from the optical Bloch equations (Fig. 4(a)). When starting at $|X\rangle$, the electric field reemitted by the polarization of the inverted transition interferes constructively with the probe and creates a positive peak in the differential transmission spectrum. This feature appears in both the experimental data (circles in the l.h.s. of Fig. 4(a)), as well as in the simulated differential transmission spectrum (magenta line). In Fig. 4(b), we further increase $P_{probe}$ to 400 $\mu$W, which corresponds to a pulse area of 0.7 $\pi$. The polarization (green line) now experiences a maximum during the probe pulse. Ending at the complementary latitude of the Bloch sphere, the polarization exhibits a tail which is similar in amplitude as after the 0.3 $\pi$ pulse of Fig. 4(a). However, the probe electric field has been increased substantially at 0.7 $\pi$ compared to 0.3 $\pi$ (blue line in Fig. 4(b)). For this reason, the coherent two-level model yields a decrease in peak amplitude for the relative transmission change $\Delta T/T$ (magenta line in Fig. 4(b)). In addition, a positive background signal extending over the entire probe bandwidth starts to develop, corresponding to the Fourier transform of the fast initial peak in the polarization response.

The most interesting situation is encountered when $P_{probe}$ is further enhanced to 800 $\mu$W, yielding a probe pulse area of $\pi$ (see Fig. 4(c)). Such a light pulse coherently drives the quantum system from the eigenstate $|X\rangle$ back to its ground state $|\phi\rangle$. Accordingly, the polarization induced by the probe pulse lacks a tail and follows closely the temporal envelope of the probe electric field (see green and blue graphs on the r.h.s. of Fig. 4(c)). In this situation exactly one photon is added to the probe pulse by a narrow resonance, but it does not change the shape of the temporal and spectral envelope of the femtosecond light pulse that gets amplified. As a result, the model predicts a flat but slightly positive differential transmission spectrum which is in good agreement with the $\Delta T/T$ we record experimentally (see magenta graph and circles in Fig. 4(c)).
Figure 4. Spectral transmission change $\Delta T/T$ measured at a delay time of $t_D = 20$ ps and for average powers of the probe beam $P_{\text{probe}}$ increasing beyond the linear regime. Experimental data are represented by hollow circles on the left hand side of a-c. The magenta lines represent differential transmission spectra calculated via the optical Bloch equations. The temporal envelopes of the probe electric field (blue lines) and the electron-hole polarization (green lines) are depicted on the right hand side of a-c for increasing probe pulse area $A_{\text{probe}}$.

We would like to stress that due to the inherent nonlinearity of the two-level system its polarization response to a short and broadband pulse is different to the coherent sum of the isolated responses induced by its narrow-band Fourier components. Spectral analysis of the probe after transmission through the quantum system yields information about the temporal structure of the single-photon gain: An isolated peak at $X^{-}$ indicates that radiation is triggered linearly into the picosecond polarization decay (Fig. 4(a)). The positive broadband signal in Fig. 4(c) directly corresponds to deterministic single-photon emission within a time window set by the 180 fs probe. Finally, the ratio between the spectral weight of the narrowband resonance and the broadband background found together in Fig. 4(b) measures absolutely the admixture of both limiting cases. We note that the features we observe in the nonlinear probing regime are intrinsic to coherent light-matter interaction and beyond the validity of rate equation models for stimulated emission.

In our present sample, a mesoscopic number of approximately $10^4$ transmitted photons per pulse are necessary to read out the $X^-$ resonance. Therefore, the influence of our femtosecond single photon absorber and amplifier on the quantum statistics of the probe field is limited. Nevertheless, the enhancement of the interaction between quantum dot and electromagnetic field towards the strong coupling regime might lead to an extreme nonlinearity, where single photons can be deterministically added to or subtracted from transients containing single or few quanta of light. Such ultrafast manipulation of pulses with well-defined photon number (so called Fock states) could enable a new
class of quantum optics experiments and offer possibilities for quantum information processing at terahertz clock rates. An enhanced interaction of the light field and the single quantum dot may be envisioned by positioning a single quantum dot in the gap of a broadband optical antenna [13] and/or inside a dielectric microcavity [14].

References
[1] Michler P, Kiraz A, Becher C, Schoenfeld W V, Petroff P M, Zhang L, Hu E and Imamoglu A 2000 A quantum dot single-photon turnstile device Science 290 2282-2285
[2] Yuan Z, Kardynal B E, Stevenson R M, Shields A J, Lobo C J, Cooper K, Beattie N S, Ritchie D A and Pepper M 2002 Electrically driven single photon source Science 295 102-105
[3] Zrenner A, Beham E, Stufler S, Findeis F, Bichler M and Abstreiter G 2002 Coherent properties of a two-level system based on a quantum-dot photodiode Nature 418 612-614
[4] Ramsay A J, Boyle S J, Kolodka R S, Oliveira J B, Liu H Y, Hopkinson M, Fox A M and Skolnick M S 2008 Fast Optical Preparation, Control, and Readout of a Single Quantum Dot Spin Phys. Rev. Lett. 100 197401
[5] Guenther T, Lienau C, Elsaesser T, Glanemann M, Axt V M, Kuhn T, Eshlaghi S. and Wieck A D 2002 Coherent nonlinear optical response of single quantum dots studied by ultrafast near-field spectroscopy Phys. Rev. Lett. 89 057401
[6] Unold T, Mueller K, Lienau C, Elsaesser T and Wieck A D 2004 Optical Stark effect in a quantum dot: ultrafast control of single exciton polarizations Phys. Rev. Lett. 92 157401
[7] Patton B, Langbein W, Woggon U, Maingault L and Mariette H 2006 Time- and spectrally-resolved four-wave mixing in single CdTe/ZnTe quantum dots Phys. Rev. B 73 235354
[8] Sotier F, Thomay T, Hanke T, Mahapatra S, Frey A, Brunner K, Bratschitsch R and Leitenstorfer A 2009 Femtosecond probing of few-fermion dynamics and deterministic single photon gain in a single CdSe quantum dot Nature Physics 5 352-356
[9] Moutzouris K, Adler F, Sotier F, Träutlein D and Leitenstorfer 2006 A Multimilliwatt ultrashort pulses continuously tunable in the visible from a compact fiber source Opt. Lett. 31 1148-1150
[10] Adler F, Sell A, Sotier F, Huber R and Leitenstorfer A 2007 Attosecond relative timing jitter and 13 fs tunable pulses from a two-branch Er:fiber laser Opt. Lett. 32 3504-3506
[11] Brito Cruz C H, Gordon J P, Becker P C, Fork R L and Shank C V 1988 Dynamics of spectral hole burning IEEE J. of Quant. El. 24 261-269
[12] Joffre M, Hulin D, Migus A, Antonetti A, Benoit a la Guillaume C, Peyghambarian N, Lindberg M and Koch S W 1988 Coherent effects in pump-probe spectroscopy of excitons. Opt. Lett. 13 276-278
[13] Merleijn J, Kahl M, Zuschlag A, Sell A, Halm A, Boneberg J, Leiderer P, Leitenstorfer A and Bratschitsch R 2008 Nanomechanical control of an optical antenna Nature Phot. 2 230-233
[14] Kahl M, Thomay T, Kohnle V, Beha K, Merleijn J, Hagner M, Halm A, Ziegler J, Nann T, Fedutik Y, Woggon U, Artemyev M, Pérez-Willard F, Leitenstorfer A and Bratschitsch R 2007 Colloidal quantum dots in all-dielectric high-Q pillar microcavities Nano Lett. 7 2897-2900