Ultrafast coherent spectroscopy of a single quantum dot

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Abstract. The nonlinear optical response of a single quantum dot is studied for the first time with femtosecond resolution. We find pronounced spectral oscillations in the differential reflectivity reflecting the perturbed free induction decay of the coherent polarization of a single exciton.

1. Introduction

Quantum dots currently attract much interest as model systems for artificial solid state atoms and as probes of the physics of wavefunction localization in disordered media [1]. The potential of using their optical excitations as bits for semiconductor-based implementations of quantum information processing is under intense investigation [2]. Although recently dephasing times of up to 600 ps have been demonstrated, such implementations are intimately linked to the ability to probe and control optical nonlinearities in single and coupled quantum dots on ultrafast time scales [3], a research field that remains to be explored.

In this paper, we describe and demonstrate a novel technique, combining near-field optics and femtosecond pump-probe spectroscopy, to analyze the nonlinear optical response of single quantum dots on ultrafast time scales. We resolve, in differential reflectivity spectra, transient oscillatory structures around the quantum dot exciton resonance. These oscillations persist up to negative time delays of more than 10 ps and reflect the perturbation of the free induction decay of the coherent QD polarization by transient manybody interactions.

2. Experimental Methods

We investigate excitons localized in interface quantum dots in a single 5.1 nm thick (100) GaAs QW layer buried at d = 120 nm below the surface. Differential probe reflectivity spectra $\Delta R(E_{\mathrm{det}},\Delta t)/R_0 = \left[R(E_{\mathrm{det}},\Delta t) - R_0(E_{\mathrm{det}})\right]/R_0(E_{\mathrm{det}})$ [$R_0(E_{\mathrm{det}})$: steady state reflectivity at photon energy E_{det}] are recorded as a function of pump-probe delay Δt in a near-field pump-probe setup at 12 K with a combined spatial and temporal resolution of 200 nm and 100 fs (Fig. 1a). The 100

nW, 18meV broad probe laser is centered at 1.66 eV, around the QW absorption resonance. Photoluminescence (PL) from the sample and/or the reflected probe light is collected through the same fiber, spectrally dispersed and detected with a CCD camera. The pump-laser is centered at 1.675 eV to create about 5 electron-hole pairs per pulse in 2D continuum states.

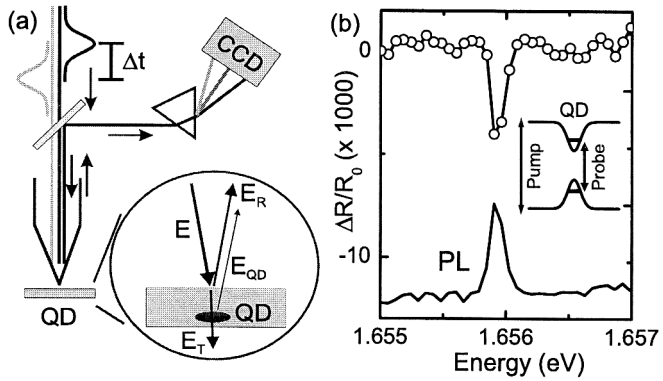

Fig. 1. (a) Schematic of the experimental setup. (b) Near-field PL spectrum of a single QD (solid line) and differential reflectivity $\Delta R/R_0$ at Δt = 30 ps. PL and ΔR are recorded with identical pump pulses centered at 1.675 eV, exciting electron-hole pairs in 2D continuum states. Inset: Schematic energy diagram.

3. Results and Discussion

For $\Delta t>0$, the $\Delta R/R_0$ spectra for above bandgap excitation at 1.675 eV show a few spectrally ultrasharp resonances, that are isoenergetic with single exciton emission peaks in simultaneously recorded PL spectra [Fig. 1(b)]. The reflectivity change ΔR displays a slow decay with time constants of 30 – 100 ps, depending on the size of the QD (Fig. 2a). Such time constants are determined by radiative recombination in the QDs.

The early time dynamics of $\Delta R\left(E_{QD}\right)/R_0$ (Fig. 2b) show striking differences from those expected for an isolated two-level system. The signal displays an 8 ps rise at negative delay times that occurs much slower than 150 fs cross correlation of pump and probe pulses and is followed by a fast partial decay with a time constant of about 6 ps. The spectral characteristics of $\Delta R/R_0$ at $\Delta t>0$ and $\Delta t<0$ are markedly different. At $\Delta t>0$, the spectra show the expected bleaching of the dot resonance, while at $\Delta t<0$, (Fig. 2c), we find pronounced *spectrally symmetric* oscillations around the excitonic resonance.

As shown in Fig. 1(a). the reflected probe light, locally collected by the near-field tip, represents a superposition of the field $E_R(t)$ reflected from the sample surface, and the field $E_{QD}(t)$, radiated in back-direction from the probe-induced excitonic QD polarization $P_{QD}(t)=\int dt' \chi_{QD}(t)E_T(t-t')$. Here, $E_T(t)$ and $\chi_{QD}(t)$ denote the probe field interacting with the QD and the QD susceptibility,

respectively. Excitation by the pump affects $\chi_{QD}(t)$ and – thus – results in a change of reflectivity. For $\Delta t < 0$, the pump perturbs the free induction decay of P_{QD} created by the probe (inset of Fig. 3), whereas for $\Delta t > 0$ the probe interacts with the system modified by the pump, resulting in a probe polarization different from the unexcited system. Then, the spectrally resolved differential reflectivity represents the spectral interferogram of $\tilde{E}_R$ and $\tilde{E}_{QD}$ (~: Fourier transform):

$$\Delta R(\omega,\Delta t) \propto \mathrm{Re}\left\{\tilde{E}_R^*(\omega)\left[\tilde{E}_{QD}(\omega,\Delta t) - \tilde{E}_{QD,0}(\omega)\right]\right\} \tag{1}$$

The line shape of ΔR evidently depends on the phase delay between E_R and E_{QD}. As a function of distance d it oscillates between absorptive and dispersive shape. In our measurements with d = 120 nm, this results in an absorptive (Lorentzian) shape at $\Delta t \gg 0$ and ΔR probes the imaginary part of the QD polarization, $\mathrm{Im}(P_{QD})$.

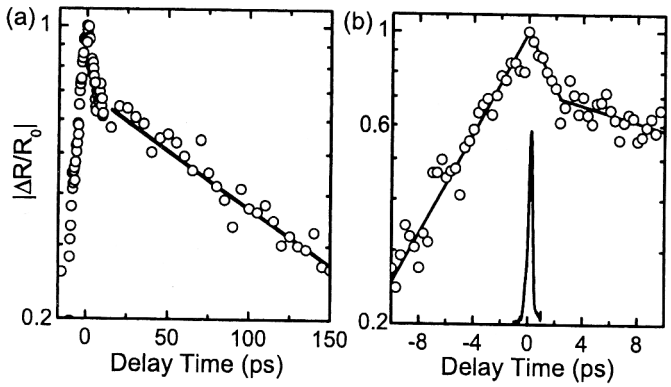

Fig. 2. (a) Time evolution of $\Delta R(E_{QD})/R_0$ for a single QD resonance at E_{QD} = 1.6598 eV (logarithmic ordinate scale). The decay is biexponential with a slow decay time 150 ps. (b) Early time dynamics of $\Delta R(E_{QD})/R_0$. A slow rise of $\Delta R/R_0$ is observed at negative delay times. Solid line around delay zero: 150 fs cross-correlation of pump and probe pulses.

In general, the free induction decay of $P_{QD}(t)$ can be perturbed by pump-induced many-body interactions changing the resonance energy, oscillator strength and/or dephasing rate of the QD polarization. Energy renormalizations give rise to spectrally asymmetric nonlinearities, as, e.g., for the dynamic Stark shift of exciton lines in quantum wells. Such energy shifts are absent in our data. Also, the spectrally integrated reflectivity change vanishes for each of the spectra recorded at negative time delays and – thus – changes of the oscillator strength are small. We conclude that the pump-induced damping of the QD polarization is the leading contribution to the QD nonlinearity at early times. Such excitation induced dephasing (EID) of the QD polarization arises from Coulomb scattering with the initial *nonequilibrium* carrier distribution in QW continuum states. The damping time of the QD polarization after the arrival of the pump pulse determines the energy range over which the reflectivity spectra display oscillations. In Fig. 3, spectral oscillations are observed in a window of about 0.5 meV around the

QD resonance, corresponding to a damping on a time scale of a few ps. The solid lines in Fig. 3 are calculated from a simple model in which the probe-induced QD polarization $P_{QD}(t)$ decays initially with an effective dephasing time T_2 = 15ps decreasing to T_{EID} = 3ps after the arrival of the pump laser (inset of Fig. 3). Evidently, this model is in quite good agreement with the experimental results. Due to the finite monochromator resolution T_2 = 15ps is a lower limit of the intrinsic QD dephasing time.

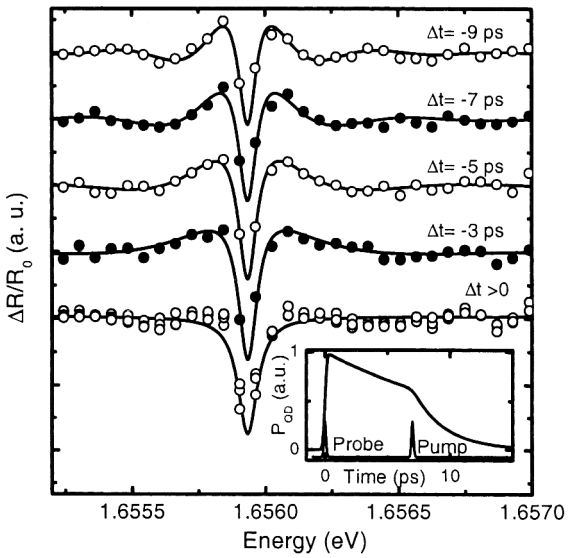

Fig. 3. Near-field $\Delta R/R_0$ spectra (circles) at different delay times Δt. The solid solid shows simulated spectra for the perturbed free induction decay of the coherent QD polarization assuming T_2 = 15 ps. Inset: Simulated dynamics of $P_{QD}(\Delta t)$.

To confirm our qualitative interpretation, we analyze the nonlinear optical QD response on the basis of the semiconductor Bloch equations (SBE) in mean-field approximation, including both disorder and Coulomb interaction. Nonlinear absorption spectra $\Delta A(\omega) \propto \mathrm{Im}\left[\tilde{E}_T^*(\omega)\tilde{P}(\omega)\right]$ are calculated, probing the imaginary part of $P_{QD}(\Delta t)$, as in the experiment. We chose a one-dimensional Gaussian-correlated two-band disorder potential with a correlation length of 10 nm and a disorder strength of 5 meV [Fig. 4(a), inset]. Excitation-induced dephasing is treated phenomenologically by a dephasing rate $\gamma = 1/T_2 + \gamma_1 n$ depending on the excitation density n. To reduce the numerical complexity, the relevant dephasing rates are scaled to T_2=2 ps and T_{EID}=0.4 ps. Pump and probe pulses are localized to 100 nm. The linear absorption spectra show resonances from single localized excitons at low energies. Transient, spectral symmetric oscillations govern the optical nonlinearity at negative delay times. The calculations clearly confirm the importance of EID contributions on the perturbed free induction decay and the minor role of coherent renormalizations of the QD resonance energy. Although

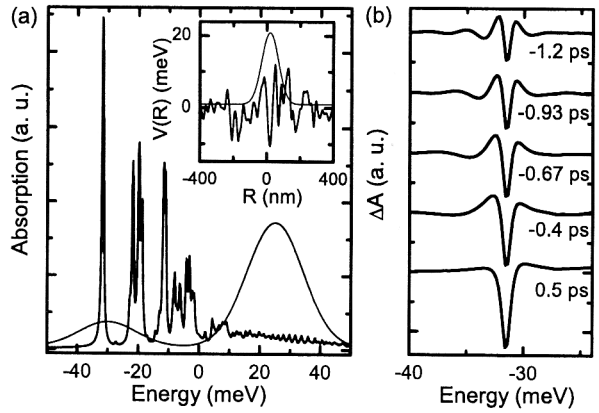

Fig. 4. (a) Spatially resolved linear absorption spectra of a QD ensemble calculated from the semiconductor Bloch equations. A Gaussian-correlated one-dimensional disorder potential is assumed (inset). Dashed lines: Spectra of pump (centered at 25 meV) and probe pulses (at -30 meV) with spot sizes of 100 nm. (b) Nonlinear absorption spectra for different pump-probe delays showing the perturbed free induction decay on the lowest resonance of Fig. (a).

single QDs resemble in many respects atomic systems, Coulomb interactions give rise to pronounced optical nonlinearities at negative delay times, which have been reported so far only for higher-dimensional systems

In conclusion, we have presented the first observation of the femtosecond excitonic nonlinearity of a single quantum dot. The free induction decay of the coherent excitonic polarization is shown to be perturbed by manybody interactions with continuum excitations on picosecond time scales. The exquisite sensitvity of the here demonstrated nano-optical pump-probe technique opens a new way to an ultrafast probing and manipulation of coherent quantum dot polarizations by external electromagnetic fields and/or material polarizations which is important for semiconductor-based implementation of quantum information processing.

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References

1 F. Intonti, V. Emiliani, C. Lienau, T. Elsaesser, V. Savona, E. Runge, R. Zimmermann, R. Nötzel and K. H. Ploog, Phys. Rev. Lett. 87, 076801 (2001).
2 G. Chen, N. H. Bonadeo, D. G. Steel, D. Gammon, D. S. Katzer, D. Park and L. J. Sham, Science, 289, 1906 (2000)
3 E. Biolatti, R. C. Iotti, P. Zanardi, and F. Rossi, Phys. Rev. Lett. 85, 5647 (2000).