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Space and time resolved coherent optical spectroscopy of single quantum dots

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Abstract

A novel experimental technique, combining near-field optics and femtosecond pump–probe spectroscopy, is demonstrated to analyse the coherent nonlinear optical response of single quantum dots on ultrafast time scales. The technique is used to study the effects of strong non-resonant light fields on the optical spectra of single excitons in interface quantum dots. Transient reflectivity spectra show dispersive line shapes reflecting the light-induced shift of the quantum dot resonance. The nonlinear spectra are governed by the phase shift of the coherent quantum dot polarization acquired during the interaction with the light field. The phase shift is measured and ultrafast control of the quantum dot polarization is demonstrated.

1. Introduction

Semiconductor quantum dots (QDs) with electronic wavefunctions localized on a nanometre length scale currently receive much attention as model systems for artificial solid state atoms. Discrete, atomic-like densities of states [1–3] and comparatively long dephasing times of excitonic excitations of up to several 100 ps [4] have been demonstrated.

During the last few years, several theoretical studies have suggested excitonic quantum dot excitations as possible bits of quantum information [5–7]. Coupling between these bits, necessary for implementing controlled quantum gates, may be provided, e.g., by dipole–dipole couplings [6]. Since exciton dephasing times are typically of the order of 10 ps to 1 ns, such implementations rely on using ultrafast optical pulses to control the exciton dynamics on a femtosecond time scale. Generally, control of both amplitude and phase of the exciton polarization is required. Control of the polarization amplitude can be achieved by varying the pulse area of a resonant pump laser, as has been demonstrated recently in studies of Rabi oscillations from excitons in single QDs [8–11]. Control of the polarization phase, on the other hand, can be achieved by using pairs of phase-locked excitation pulses, or by strong off-resonant laser fields, as is demonstrated in this paper by studying the optical Stark effect on a single QD.

As a model system, we investigate interface quantum dots in a thin semiconductor quantum well (QW). In such a QW, local minima of the QW disorder potential caused by interface roughness and alloy fluctuations lead to the localization of excitons in interface QDs with a confinement energy of about 10 meV. The large extension of the exciton centre-of-mass wavefunction of 30–50 nm gives rise to large single QD dipole moments of 50 to 75 Debye [12]. This strong coupling to light makes them interesting model systems for time resolved nonlinear spectroscopy of single QDs [8, 13].

In this paper, near-field optics and femtosecond pump–probe spectroscopy are combined to study the ultrafast nonlinear optical response of *single* quantum dots in the presence of strong non-resonant light fields. Two-colour pump–probe spectra in a reflection geometry show a dispersive line shape caused by the optical Stark effect on the excitonic QD resonance. This line shape critically depends on the pump laser power and is determined by the pump-induced phase shift of the QD polarization. This phase shift is quantitatively measured and ultrafast control of the QD polarization is demonstrated.

2. Experimental details

We investigate interface QDs in a sample consisting of 12 single QW layers of different thicknesses grown on a (100)

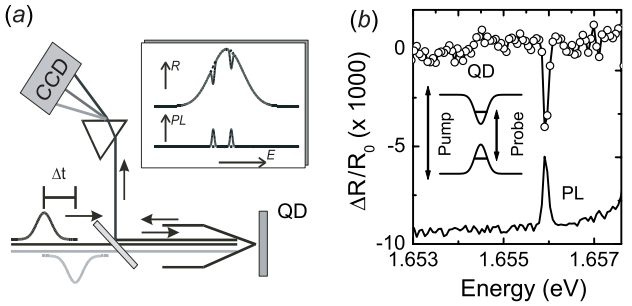

Figure 1. (*a*) Schematic illustration of the experimental setup and of near-field PL and reflectivity spectra of the QD sample. (*b*) Near-field PL spectrum of a single QD (solid line) and differential reflectivity spectrum $\Delta R/R_0$ at $\Delta t = 30$ ps. PL and ΔR are recorded with identical pump pulses centred at 1.675 eV, exciting electron–hole pairs in 2D continuum states. The 100 nW probe pulses of 19 meV bandwidth are centred at 1.655 eV, around the QD absorption resonance. Inset: schematic energy diagram.

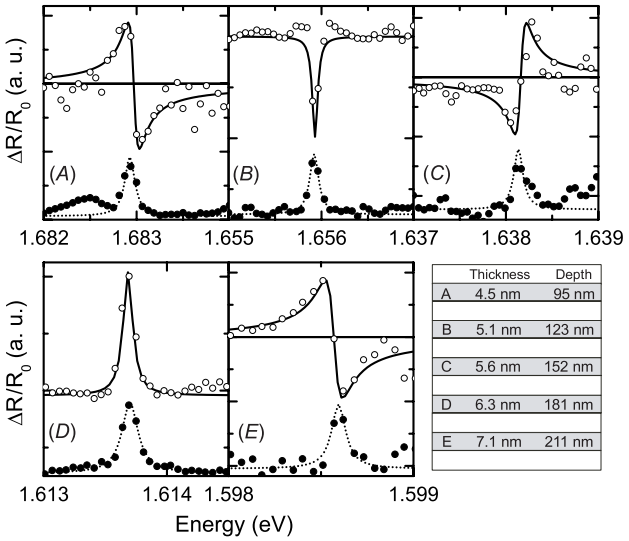

Figure 2. Differential reflectivity spectra (open circles) of five interface QDs located at different depths of 95 to 210 nm below the sample surface (see the inset). The differential reflectivity spectra are compared to simultaneously recorded PL spectra. Note the transition between dispersive and absorptive line shapes.

GaAs substrate. The QW layers are separated by AlAs/GaAs short period superlattice barriers, each formed by nine AlAs and GaAs layers with a total thickness of 23.8 nm. Here, we investigate the top seven QWs with thicknesses of 3.3 to 7.1 nm. The layers are buried at distances between 40 and 211 nm below the surface. Growth interruptions of 10 s at each interface lead to a large correlation length of the QW disorder potential and to the formation of interface quantum dots. The typical confinement energies of these QDs are 5–10 meV, as evidenced by near-field autocorrelation spectroscopy [14]. The QDs have large dipole moments of 50–100 Debye [13], consistent with a QD diameter of about 50 nm. The experiments were performed with a low-temperature near-field optical microscope operating at 12 K and used in a pump–probe setup [13]. Co-linearly polarized pump and probe pulses are obtained from a 80 fs Ti:sapphire oscillator and coupled into a chemically etched uncoated fibre tip (figure 1). Spectral and temporal shaping of the pulses leads to a combined temporal and spatial resolution of 250 fs and 200 nm, respectively. The photoluminescence (PL) signal and/or the reflection signal from the sample is collected through the same fibre, dispersed in a $f = 50$ cm spectrograph (resolution 100 μeV) and recorded with a liquid nitrogen cooled CCD camera. The 100 nW probe pulses of 150 fs duration and 10 meV bandwidth are centred at 1.655 eV, around the QW absorption resonance. We measure nonlinear differential probe reflectivity spectra $\Delta R(\omega, \Delta t)/R_0 = [R(\omega, \Delta t) - R_0(\omega)]/R_0(\omega)$ at a fixed spatial position of the near-field tip as a function of the time delay Δt between pump and probe pulses ($R_0(\omega)$: steady state reflectivity at a photon frequency ω). We have described the measured signal as the spectral interferogram

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

between the probe field $\tilde{E}_R(\omega)$, reflected from the sample surface and the electric field $\tilde{E}_{\mathrm{QD}}(\omega)$ emitted from the QD [13]. ΔR thus probes the effects of the pump laser on the coherent QD polarization dynamics.

3. Results and discussion

In this section, we first describe how QD nonlinearities are probed in near-field reflectivity measurements and then analyse the dynamics of the QD nonlinearities for below band-gap excitation.

The high spatial resolution of the near-field technique allows us to isolate the emission from single localized excitons in interface quantum dots. Figure 1(*b*) shows a representative near-field PL spectrum of the 5.1 nm QW in the low-energy region of the inhomogeneously broadened QW PL line. The peak at 1.6598 eV clearly reveals the emission from a single exciton localized in an interface QD. In this figure, the PL spectrum is compared to a simultaneously recorded differential reflectivity spectrum $\Delta R/R_0$. In this experiment the pump laser at 1.675 eV excites electron–hole pairs in QW continuum states. The spectrum is recorded at a time delay $\Delta t = 30$ ps (closed circles). It displays a single spectrally sharp resonance at exactly the same spectral position as in the PL spectrum. The large amplitude of the signal of 5×10^{-3} is consistent with a spatial resolution of the experiment of 200–250 nm. Here, the pump laser creates a non-equilibrium distribution in QW continuum states. Subsequent trapping of carriers gives rise to a bleaching of the QD absorption. The absorptive line shape of the $\Delta R/R_0$ spectrum can be understood by inspecting equation (1). The spectral shape of this interferogram evidently depends not only on the QD polarization dynamics but also on the phase delay between QD emission and the probe field $E_R(t)$ reflected from the sample surface. In a simplified local oscillator model, treating the QD as a point dipole and the near-field tip as a point-like emitter, the phase delay is given by the distance d between the QD and the near-field tip, and an absorptive line shape is expected for an optical path $4\pi nd/\lambda$ which is a multiple of π. This model suggests that the reflectivity spectrum should show a transition from an absorptive to a dispersive line shape if the optical path is changed by $\pi/2$. This interference effect is nicely seen in figure 2, comparing ΔR spectra of five single localized excitons in five different QWs buried at distances of 95 nm to 211 nm below the surface. We clearly observe the

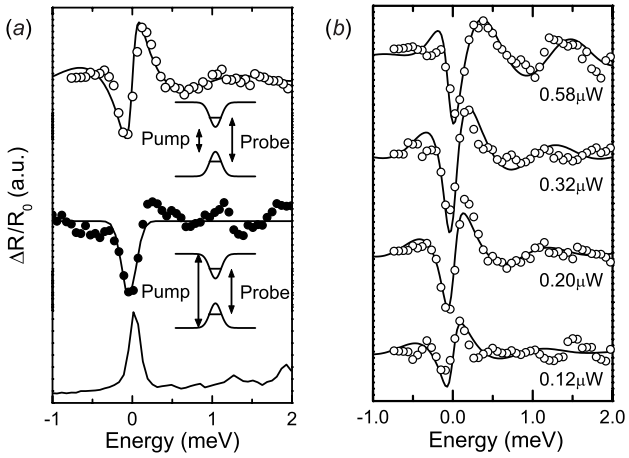

Figure 3. (*a*) Near-field PL spectrum of a single QD resonance $E_{\mathrm{QD}} = 1.6503$ eV (solid line) and differential reflectivity spectrum $\Delta R(E_{\mathrm{QD}})/R_0$ for above band gap excitation at $\Delta t = 50$ ps (solid circles). Open circles: differential reflectivity spectra for the same QD for below band gap excitation at $\Delta t = -4$ ps with 2 ps laser pulses (power 0.12 μW) centred at 1.647 eV. (*b*) Pump dependence of the differential reflectivity spectra of the same QD. The pump laser is again centred at 1.647 eV ($\Delta t = -4$ ps) and the pump power is varied between 0.12 μW and 0.58 μW. The solid lines display simulated spectra based on the optical Bloch equations.

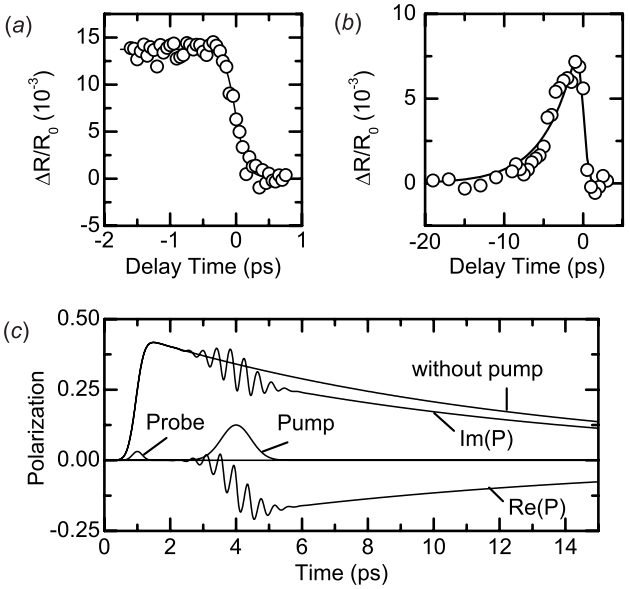

Figure 4. (*a*) Time evolution of $\Delta R(\Delta t)/R_0$ for a single QD resonance at $E_{\mathrm{QD}} = 1.6544$ eV. Here, the pump laser with a pump power of 58 μW and a pulse duration of $\tau_p = 200$ fs was centred at 1.640 eV. (*b*) Time dynamics on a 20 ps time scale. (*c*) Bloch equation simulation of the QD polarization p in the rotating frame. In the presence of the non-resonant pump pulse a light shift is induced that gives rise to a persistent phase change $\Delta\phi$ of the QD polarization.

transition between dispersive and absorptive line shapes as the QD to surface distance is varied. A phase change of $\pi/2$ for a change in QD–surface distance of 28 nm is estimated from the local oscillator model, which is in quite good agreement with figure 2. We consider this as convincing evidence for the validity of the phenomenological local oscillator model described above. Clearly a more detailed analysis of the data using, e.g., a Green function solution of Maxwell's equations for a realistic experimental geometry is desirable for a more quantitative comparison.

In figure 3(*a*), pump–probe spectra taken on the same QD in the 5.1 nm QW for above and below band gap excitation are compared. The data for above band gap excitation are recorded under similar experimental conditions as those in figure 1, while the data for below band gap data are taken at $\Delta t = -4$ ps with pump pulses detuned by 3 meV below the QD resonance. In contrast to the signal for above band gap excitation, a dispersive line shape centred around the exciton resonance is observed. This line shape is strongly affected by variations of the pump power. Figure 3(*b*) compares $\Delta R/R_0$ spectra taken on the same QD at pump powers between 0.12 μW and 0.58 μW. A clear saturation of the signal magnitude is observed. The maximum of ΔR shifts towards higher energies and an increasing number of spectral oscillations is observed.

Figure 4 shows the time dynamics of the QD nonlinearity for below band gap excitation. $\Delta R(\omega)$ spectra are measured around a single QD resonance at different delay times and the signal magnitude $\Delta R_m(\Delta t)/R_0$, taken as the difference between minimum and maximum in $\Delta R(\omega)/R_0$ is plotted on a short 3 ps time scale in figure 4(*a*) and on a longer 20 ps scale in figure 4(*b*). The signal vanishes completely for positive delay times $\Delta t > 0$ (pump precedes probe) and rises around $\Delta t = 0$ within the time resolution of our experiment of 250 fs. For $\Delta t < 0$, $\Delta R_m(\Delta t)$ decays with a time constant of $\tau_d = 8$ ps. For these experiments, 200 fs pump pulses tuned 14 meV below the QD resonance are used.

We now discuss these results, which provide the first direct evidence for an ultrafast optical Stark effect on the excitonic polarization of a *single* QD. The spectrally sharp $\Delta R/R$ resonance for above band gap excitation, which is isoenergetic with the QD PL peak ensures that we are indeed probing optical nonlinearities of *single* QD excitons. Its absorptive line shape due to exciton bleaching indicates that $\Delta R/R$ probes the imaginary part of the QD polarization.

Within our signal-to-noise ratio, no nonlinearity is detected for $\Delta t > 0$. Thus, generation of real carriers, e.g. through two-photon absorption or direct absorption into neighbouring quantum dots or QW continuum states is negligible and does not affect our signals. The ΔR signals at $\Delta t < 0$ reflect a pure light-induced shift of the QD resonance.

The dispersive line shape observed in figure 3(*a*) for low pump powers and small time delays is the well-known signature of the optical Stark effect. This can be understood by analysing the time dynamics of coherent exciton polarization, displayed in a rotating frame in figure 4(*c*). We describe the QD as an effective two-level system with a dephasing time $T_2 = 8$ ps, taken to account for our finite monochromator resolution. The dynamics of the QD polarization are described by optical Bloch equations. The resonant probe laser changes the QD population and drives a coherent polarization oscillation at the QD transition frequency, phase-shifted by $\pi/2$ with respect to the probe field. In the presence of the pump laser, the polarization is externally driven at the detuning frequency $\nu = (\omega_{\mathrm{QD}} - \omega_{\mathrm{pu}})$, with ω_{QD} and ω_{pu} being the QD resonance frequency and the pump laser frequency, respectively. After the interaction with the pump laser, the

polarization is phase-shifted by $\Delta\phi$. This phase shift is approximately given as the product of the light-induced shift of the quantum dot resonance and the duration of the pump pulse. A phase shift of the polarization changes the product of the $\tilde{E} \cdot \tilde{E}_{\mathrm{QD}}$ in equation (1), and this gives rise to the dispersive spectral line shape.

Such a dispersive line shape has previously been observed [15, 16] and theoretically analysed [17] in experiments probing the optical Stark effect in higher dimensional semiconductors. An effect of the pump intensity of the spectral line shape, however, has not been reported before. This effect is clearly evident from the data in figure 3(*b*). A Bloch equation model qualitatively accounts for this behaviour. The interaction of the QD polarization with a strong pump laser gives rise to pronounced, large amplitude oscillations of the QD polarization and population at the detuning frequency in the presence of the pump laser. A large phase shift $\Delta\phi$ of the QD polarization results from this interaction. A comparison between experimental spectra and Bloch equation model allows us to quantify the phase shift $\Delta\phi$ experienced by the QD polarization, which may reach values of up to 200° under our experimental conditions (figure 3(*b*), $P_{\mathrm{pu}} = 0.58\ \mu$W).

In summary, we have introduced a novel experimental concept, combining near-field microscopy and femtosecond pump–probe spectroscopy to probe the nonlinear optical response of single semiconductor quantum dots. We give first experimental evidence for the optical Stark effect on a single QD exciton and demonstrate control over the phase of the exciton polarization. This is an important step forward in using such excitations for semiconductor-based implementations of quantum information processing.

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