Optical Stark Effect in a Quantum Dot: Ultrafast Control of Single Exciton Polarizations

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We report the first experimental study of the optical Stark effect in single semiconductor quantum dots (QD). For below band gap excitation, two-color pump-probe spectra show dispersive line shapes caused by a light-induced blueshift of the excitonic resonance. The line shape depends strongly on the excitation field strength and is determined by the pump-induced phase shift of the coherent QD polarization. Transient spectral oscillations can be understood as rotations of the QD polarization phase with negligible population change. Ultrafast control of the QD polarization is demonstrated.

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Investigating single quantum systems, such as atoms, molecules, or semiconducting nanostructures, with ultrafast light pulses offers the possibility to probe and manipulate the quantum dynamics in isolated and coupled systems [1], an important prerequisite for ultrafast optical implementations of quantum information processing [2,3]. Many of such proposed implementations, e.g., with trapped ions [4] or semiconductor quantum dots [2,3], rely on controlling amplitude *and* phase of quantum bits with short laser fields. Thus there is considerable current interest in understanding and manipulating coherent light-matter interaction in such systems.

Here, a fundamental interaction is the optical Stark effect (OSE), describing the light-induced shift of the energy levels in the presence of nonresonant laser fields, i.e., the light-induced phase shift of the optical polarization. The OSE corresponds to an optical nonlinearity which in atoms [5,6] is described adequately by the optical Bloch equations for noninteracting two-level systems. In quasi-two-dimensional semiconductors, i.e., in quantum wells (QW), the dephasing times of optical excitations are much shorter than in atoms, and Stark shifts of excitonic lines have mainly been studied with ultrashort light pulses [7–10]. Theoretical treatments have demonstrated that, in contrast to atomic systems, Coulomb-mediated many-body effects, such as exciton-exciton interaction, biexciton formation, and higher order Coulomb correlations [9,11–13], affect the magnitude of the spectral shift and the excitonic oscillator strength and may even reverse the sign of the shift.

Semiconductor quantum dots (QDs) with electronic wave functions localized on a nanometer length scale find much current attention as model systems for artificial solid state atoms and for implementations of quantum logic. QDs possess atomiclike densities of states [14,15] and comparatively long dephasing times of excitonic excitations of up to several 100 ps [16]. The unavoidable inhomogeneous broadening in QD ensembles makes it important to study coherent excitations of *single* QDs and this has recently been achieved by applying nano-optical techniques [17–19]. Rabi oscillations, i.e., control of excitonic populations, have been demonstrated [18,20,21] and it was found that many-body interactions, e.g., excitation-induced dephasing due to exciton-exciton and/or exciton-free carrier interactions, contribute to the QD optical nonlinearity on a femtosecond time scale [18,19]. So far, however, phase control of the excitonic QD polarization, or more generally, an excitonic Stark shift induced by nonresonant light fields in a quasi-zero-dimensional semiconductor, has been studied to a very limited extent.

In this Letter, we report the first femtosecond study of the optical Stark effect in a single QD. The transient reflectivity measured with impulsive nonresonant excitation of the QD displays a spectral envelope depending on the strength of the excitation field: For small fields, we observe dispersive line shapes, which are indicative for spectral shifts of the exciton lines. At high external fields, one finds additional oscillatory contributions reflecting large amplitude pump-induced phase shifts of the exciton polarization. We demonstrate ultrafast control of the phase of the coherent QD polarization via the intensity dependence of these oscillations.

Our experiments are performed on interface QDs in a 5.1 nm thick GaAs QW layer grown by molecular beam epitaxy between two AlAs/GaAs superlattice barrier layers on a (100) GaAs substrate and buried 120 nm below the surface. Growth interruptions of 20 s lead to a large correlation length of the QW disorder potential and to the formation of interface QDs. Their confinement energies are typically 5–10 meV [22]. The QDs have large dipole moments of 50–100 D [18,19], consistent with a QD diameter of about 50 nm. The experiments were performed with a near-field optical microscope operating at 12 K and used in a pump-probe setup [19]. Collinearly polarized pump and probe pulses are derived from a Ti:sapphire oscillator by spectral and temporal shaping and coupled into an etched uncoated fiber tip. The photoluminescence (PL) signal and/or the probe light reflected from the sample is collected through the same fiber,

dispersed in a $f = 50$ cm spectrograph (resolution 100 μeV) and recorded with a charge-coupled device (CCD) camera. Pump-probe measurements are performed with 2 ps pump pulses centered at 1.647 eV (spectral bandwidth $\sigma = 0.8$ meV) or 200 fs pump pulses at 1.64 eV ($\sigma = 8$ meV), below the QD resonance. The 100 nW probe pulses of 150 fs duration ($\sigma = 10$ meV) are centered at 1.655 eV, around the QD resonance [Fig. 1(a)]. 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 frequency ω]. The measured signal is well described as the spectral interferogram $\Delta R(\omega, \Delta t) \propto \mathrm{Re}\{E_R^*(\omega)[E_{\mathrm{QD}}(\omega, \Delta t) - E_{\mathrm{QD},0}(\omega)]\}$ between the probe field E_R, reflected from the sample surface and the electric field E_{QD} emitted from the QD [19]. ΔR probes the effect of the pump laser on $\mathrm{Im}[P_{\mathrm{QD}}(\Delta t)]$, the imaginary part of the coherent QD polarization.

With high spatial and spectral resolution, the emission from single excitons localized in interface QDs can be isolated in the low energy region of the inhomogeneously broadened QW PL line [14,15,19]. This allows us to study the coherent polarization dynamics of a single quantum system, as opposed to those of an inhomogeneously broadened and possibly interacting ensemble of QDs.

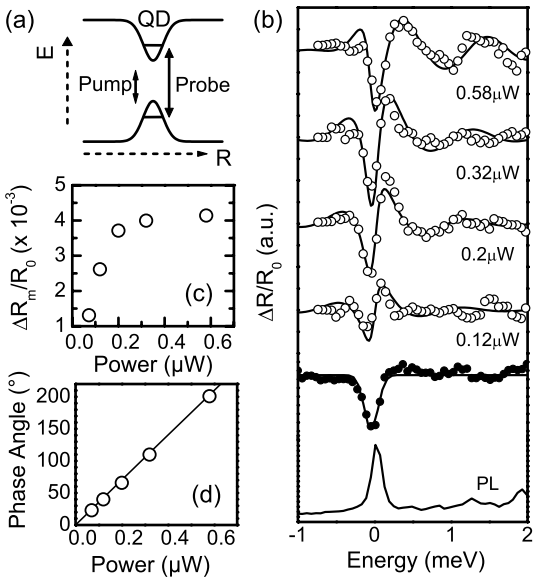

FIG. 1. (a) Schematic energy diagram. (b) PL spectrum of a single QD resonance $\omega_{\mathrm{QD}} = 1.6503$ eV and differential reflectivity $\Delta R(\omega)/R_0$ for above band gap excitation at $\Delta t = 50$ ps (solid circles). Open circles: $\Delta R(\omega)/R_0$ for below band gap excitation at $\Delta t = -4$ ps with 2 ps pulses at 1.647 eV ($\sigma = 0.8$ meV). The pump power is varied between 0.12 and 0.58 μW. Solid lines: Bloch equation model. (c) Variation of the signal magnitude $\Delta R_m(\omega_{\mathrm{QD}})/R_0$ with pump power. (d) Phase shift of the QD polarization versus pump power.

Figure 1(b) compares the PL (solid line) from a single QD at $\omega_{\mathrm{QD}} = 1.6503$ eV and the $\Delta R(\omega, \Delta t = 50\ \mathrm{ps})/R_0$ spectrum for excitation of QW continuum states by 200 fs pulses at 1.673 eV (solid circles). The ΔR spectrum shows a narrow resonance isoenergetic with the QD PL line. Its absorptive line shape reflects the bleaching of the QD resonance due to the relaxation of carriers generated in QW continuum states into the QD [19]. In addition, Fig. 1(b) shows $\Delta R(\omega)/R_0$ (open circles) measured with 2 ps pump pulses at 1.647 eV, i.e., 3 meV below the QD resonance for $\Delta t = -4$ ps (probe precedes pump). In these spectra, the pump power P_p is varied between 0.12 and 0.58 μW. In contrast to the signal for above band gap excitation, a dispersive line shape centered around ω_{QD} is observed for weak excitation ($P_p \lesssim 0.2$ μW). With increasing power, the signal strength ΔR_m, taken as the difference between minimum and maximum of $\Delta R(\omega)$, first increases and then saturates around 0.3 μW [Fig. 1(c)]. This saturation occurs together with a change in the line shape of $\Delta R(\omega)$: For strong excitation, the signal maximum shifts slightly towards higher energies and an increasing number of spectral oscillations is observed, in particular, on the high energy side of the QD resonance. In Fig. 1(d) we plot the phase shift $\Delta\phi$ of the QD polarization due to the interaction with the off-resonant pump laser, extracted from a Bloch equation modeling of the data in Fig. 1(b), as described below.

Figure 2 shows the time evolution of the QD nonlinearity as derived from $\Delta R(\omega)$ measurements with 200 fs pulses centered at 1.64 eV, i.e., 14 meV below the QD resonance at 1.654 eV. $\Delta R_m(\Delta t)/R_0$ is plotted as a function of delay time on a short 3 ps [Fig. 2(a)] and a longer 20 ps time scale [inset of Fig. 2(a)]. 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.

Figure 3 displays the time evolution of $\Delta R(\omega, \Delta t)/R_0$ spectra for an excitonic resonance at $\omega_{\mathrm{QD}} = 1.6531$ eV. The data are taken with 2 ps pump pulses centered 4.5 meV below the resonance ($P_p = 0.7$ μW). We find a nonlinear spectrum only at $\Delta t < 0$, showing pronounced *spectrally asymmetric* oscillations around ω_{QD}. The oscillation period decreases with increasing negative delay.

We now discuss these results providing the first direct evidence for an ultrafast optical Stark shift 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.

With pump pulses tuned to the transparent region below the QD resonance, the dynamics of $\Delta R_m(\Delta t)$ are different from those for above band gap excitation [19].

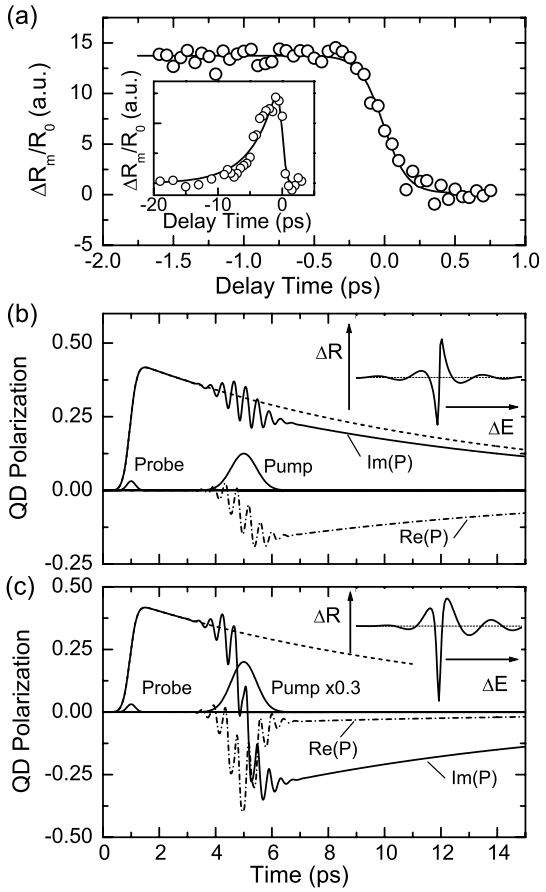

FIG. 2. (a) Time evolution of $\Delta R_m(\Delta t)/R_0$ for a single QD at $\omega_{\mathrm{QD}} = 1.6544$ eV. $\Delta R_m(\Delta t > 0)$ vanishes and the signal for $\Delta t < 0$ decays on a picosecond time scale. (b),(c) Bloch equation simulation of the QD polarization P_{QD} and ΔR spectrum (inset) in the weak and strong excitation regime.

No nonlinearity is detected for $\Delta t > 0$. This shows that we are sensitive to only the perturbation of the free-induction decay [19] of the probe-induced QD polarization via the off-resonant pump field. Signal contributions due to the generation of real carriers, e.g., through two-photon absorption [8] or direct absorption into neighboring quantum dots or QW continuum states, are negligible. The ΔR signals at $\Delta t < 0$ thus reflect a pure light-induced shift of the QD resonance. This is supported by finding that the spectrally integrated ΔR vanishes also for $\Delta t < 0$.

For low pump powers and small time delays, we observe a dispersive line shape, reflecting a blueshift of the QD exciton [Fig. 1(b)]. This is the signature of the OSE in the *weak excitation limit* [7]. In the presence of an ac electric field of frequency ω_p, the transition frequency of a two-level system shifts by $\Delta\omega_0(t) = \sqrt{[(\omega_0 - \omega_p)^2 + \Omega_R(t)^2]} + \omega_p - \omega_0$. Here, ω_0 is the transition frequency without external ac field, $\Omega_R(t) = \mu E_p(t)/\hbar$ is the Rabi frequency, μ the transition dipole moment, and $E_p(t)\cos(\omega_p t)$ the (pump) ac electric field.

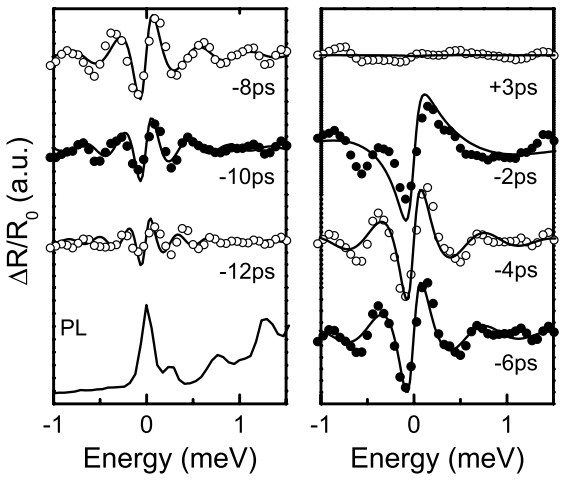

FIG. 3. Near-field $\Delta R(\omega)/R_0$ spectra (circles) measured with 2 ps pump pulses at different delay times Δt (exciton resonance at $\omega_{\mathrm{QD}} = 1.6575$ eV, $\omega_p = 1.6531$ eV, $P_p = 0.7$ μW). Solid lines: Bloch equation simulations assuming $T_2 = 8$ ps.

The blueshift $\Delta\omega_0(t)$ of the QD absorption resonance results in a dispersive $\Delta R(\omega)/R_0$ line shape, which can be approximated as $\Delta R(\omega)/R_0 \propto \Delta\omega_{0,\max}\partial\alpha(\omega)/\partial\omega$, where $\alpha(\omega)$ is the QD absorption spectrum and $\Delta\omega_{0,\max}$ is the maximum blueshift. Thus, in the weak excitation limit, the amplitude of the $\Delta R(\omega)/R$ signal is expected to increase linearly with increasing pump power, without change of the line shape. The spectra of Fig. 1(b) taken with pump powers ≤ 0.2 μW exactly display this behavior. For such pump powers, the Rabi frequency has a maximum value of $\Omega_{R,\max} = 1.75$ meV $\simeq 5\Delta\omega_{0,\max}$.

A theoretical description of the time-dependent $\Delta R(\omega)/R_0$ spectra requires a full integration of the optical Bloch equations. We describe the QD as a two-level system with a radiative lifetime of $T_1 = 100$ ps corresponding to a dipole moment $\mu = 50$ D [19]. An effective dephasing time of $T_2 = 8$ ps is assumed to account for the finite monochromator resolution [23]. Knowing the power and duration τ_p of the pump pulses and the spatial resolution of about 250 nm, the electric field of the pump laser is estimated to within a factor of 2 and no free parameters enter the simulation.

The calculated dynamics of the QD polarization in the weak excitation limit are displayed in the rotating frame in Fig. 2(b). The probe field resonant to the exciton line changes the QD population and drives a coherent polarization oscillating at the QD resonance frequency ω_{QD} that is 90° phase shifted with respect to the probe field ($\mathrm{Re}[P_{\mathrm{QD}}] = 0$), as recently verified experimentally for QW excitons [24]. During the pump pulse, the polarization is externally driven, leading to oscillations at the detuning frequency $\omega_{\mathrm{det}} = \omega_0 - \omega_p$. After the interaction, the polarization is phase shifted by $\Delta\phi \approx \int \Delta\omega_0(t)dt$. It is this shift $\Delta\phi$ of the QD polarization which changes the product $E_{pr}(\omega)E_{\mathrm{QD}}(\omega)$ of the complex

electric fields and, therefore, the line shape. Fourier transformation of the polarization dynamics gives directly the dispersive line shape of the $\Delta R(\omega)$ spectrum in the weak excitation limit, $\Delta\phi \lesssim 40°$, at early delay times [Fig. 1(b) and inset of Fig. 2(b)]. It also reproduces the time-dependent spectral oscillations at longer delay times (Fig. 3) [25]. Here, good agreement between the experimental data for a pump power of 0.7 μW and a Bloch equation simulation with a phase angle $\Delta\phi$ of 45° is evidenced.

For higher electric fields of the pump pulse, the weak excitation limit of the OSE nonlinearity is no longer valid. Experimentally one finds additional features in the transient reflectivity spectra [Fig. 1(b), traces for pump intensities of 0.32 and 0.58 μW]. The interaction of the QD polarization with a strong pump field gives rise to pronounced large amplitude oscillations of QD polarization at the detuning frequency during the presence of the pump laser. This is illustrated in Fig. 2(c) showing the solution of Bloch equations for strong excitation with $\Omega_R = 6$ meV ($\omega_{\mathrm{det}} = -10$ meV). A large phase shift $\Delta\phi$ of 172° of the QD polarization results from this interaction and the nonlinear ΔR spectrum shows additional oscillatory structures on the high energy side, as found in the experiment. This large amplitude phase rotation corresponds to the observation of *gain* on the resonance of a single QD. A comparison between experimental spectra and simulation [solid lines in Fig. 1(b)] allows us to quantify the phase shift $\Delta\phi$ experienced by the QD polarization. In Fig. 1(d) we plot $\Delta\phi$ obtained from the simulation of the data in Fig. 1(b) as a function of the pump power P_p. We find a linear increase in $\Delta\phi$ with P_p. This means that the light shift also increases linearly in our experiment, despite the saturation of ΔR_m. This linear increase in the polarization phase $\Delta\phi$ is in some analogy to the pulse area theorem for Rabi oscillations of the population of a two-level system when driven with a resonant pulse. Currently, we can quantitatively measure the phase shift with an accuracy of about 10° and achieve phase rotations of as much as 200°. Control of the exciton density through variation of the pulse area for resonant excitation has previously been established by the observation of Rabi oscillations [18,20,21]. Our present results show that a sequence of a resonant and an off-resonant laser pulse gives full control over amplitude *and* phase of the coherent excitonic polarization. In particular, we can switch the QD from absorption to gain within about 1 ps.

The quantitative agreement between experiment and a two-level system Bloch equation model indicates that many-body effects which are important in quasi-two-dimensional semiconductors are of minor relevance for the behavior observed in our present experiment. This seems surprising as biexcitonic transitions are known to have similar dipole moments as the excitonic resonance and may also be excited [26]. Simulations of the Bloch equations including biexcitons indicate that under our conditions the biexciton transition affects the excitonic nonlinearity only weakly.

In conclusion, we have presented the first experimental observation of the optical Stark effect in a single semiconductor quantum dot. Two-color pump-probe reflectivity spectra show intensity-dependent spectral oscillations caused by the light-induced shift of the quantum dot resonance. For weak driving fields, pure shifts of the excitonic resonance are found whereas spectral oscillations are observed for strong excitation. These oscillations reflect large amplitude phase shifts of the exciton polarization that may be quantitatively measured by nonlinear spectroscopy and controlled by variation of the pump laser intensity. This presents an additional step forward in using quantum dots for semiconductor-based implementations of quantum logical operations.

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