

## Near-Field Spectroscopy of Disordered Nanostructures

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Dedicated to Professor Dr. Roland Zimmermann on the occasion of his 60th birthday

For many years, Roland Zimmermann and his research group have devoted much of their attention to the effects of disorder and exciton localization on the optical properties of semiconductor nanostructures. With the recent development of spectroscopic techniques providing subwavelength spatial resolution, new experiments became possible to reveal some of the rich physics linked to exciton localization. This paper briefly reviews recent work of the authors' group using near-field nano-spectroscopy. Specifically, it is shown that near-field autocorrelation spectra give strong evidence for quantum mechanical level repulsion of localized exciton states and allow for a quantitative estimate of the underlying microscopic disorder features. Femtosecond non-linear near-field spectroscopy allows to probe the transient optical nonlinearity from a *single* localized exciton in a thin quantum film on ultrashort time scales.

Semiconductor nanostructures are never perfect [1]. Despite the high standard of modern growth techniques, local monolayer height fluctuations at the interfaces (interface roughness) and fluctuations of the alloy composition (alloy disorder) are unavoidable. In quantum wells (QW), this structural disorder gives rise to inhomogeneous broadening of far-field optical spectra, as is well known since more than two decades [2]. Inhomogeneous broadening may be of little importance for thick, high quality quantum wells (Fig. 1). It dominates, however, the linear and nonlinear properties of thin quantum wells, which are often the basis for growth of low-dimensional nanostructures, such as quantum wires and dots. Inhomogeneous broadening is intimately linked to a random spatial localization of electron and hole wave functions within the disordered quantum well and the resulting fluctuations in optical transition energies. This is directly evidenced by recording luminescence spectra with high spatial ( $<1\ \mu\text{m}$ ) and spectral ( $<0.1\ \text{meV}$ ) resolution [3–6]. In such experiments, the smooth far-field PL spectrum breaks up into a series of sharp emission spikes from single localized excitons (Fig. 1).

Detailed theoretical investigations in R.Z.'s group [7] suggest that as long as the exciton binding energy is larger or similar to the disorder-induced broadening, the optical properties of such thin quantum wells should follow from solutions of a stationary two-dimensional single-exciton Schrödinger equation. This equation describes the 1s excitonic center-of-mass motion (COM) with an effective disorder potential  $V(\mathbf{R})$ .  $V(\mathbf{R})$  is given by the three-dimensional spatial average of the exciton relative wave function over the local band edge fluctuations. This factorization ansatz reduces the numerical analysis of the optical properties of disordered QW to solving of a two-dimensional

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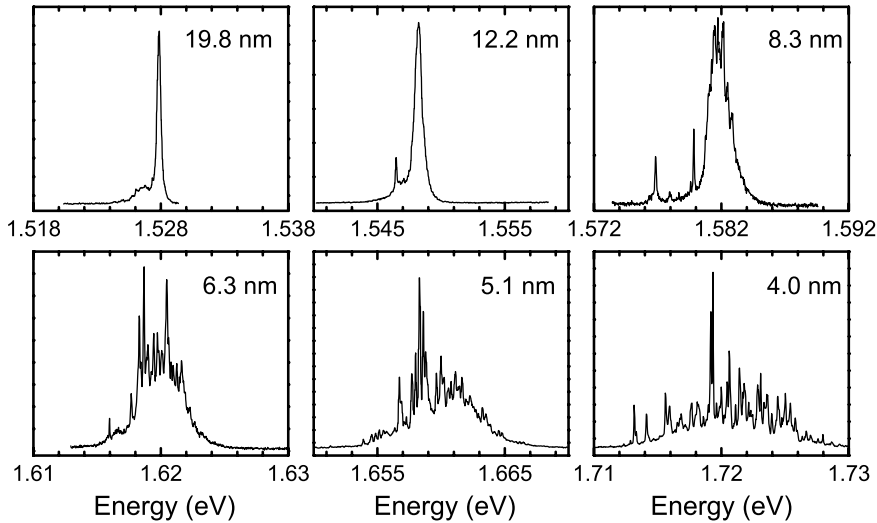


Fig. 1. Near-field PL spectra ( $T = 12$  K) of a GaAs multiple QW sample containing QW with different layer thicknesses varying between 19.8 and 4.0 nm. The QW are clad by AlAs/GaAs short period superlattice barriers. The increase in inhomogeneous broadening with decreasing well width and the break up of the spectrum into sharp single exciton emission peaks is clearly visible

eigenvalue equation. Based on such solutions, R. Z. and his collaborators made quite a number of predictions about the effects of disorder on optical QW spectra. For instance, an anomalous Stokes shift [8, 9] due to nonthermal distribution functions [7], the characteristic spectral shape of the absorption line [7], the complex shape of the COM wave functions [10] and excitonic level repulsion [10–12] were discussed.

Our group became interested in these theories when attempting to study exciton transport along single (311)A GaAs quantum wires (QWR) [13] in novel coupled wire-dot structures [14]. At low temperatures, exciton diffusion was strongly suppressed and it was clear that we needed to learn about disorder in order to understand the transport properties. We performed low-temperature [15] near-field imaging and spectroscopy<sup>2)</sup> on those quantum wires [6]. The high spatial (150 nm [16]) and spectral (100  $\mu$ eV) resolution and the excellent sensitivity gained by using uncoated near-field probes in an illumination/collection geometry allows to well resolve single exciton emission (Fig. 2). For nonresonant excitation at 1.96 eV, the PL spectra are surprisingly insensitive to a variation in excitation power by almost three orders of magnitude. In such spectra, we observed even at the lowest power of only 2 nW, a characteristic spectrally broad background emission in addition to the sharp localized exciton lines [5, 17]. The origin of this background can be understood by analyzing a two-dimensional array of such spectra, recorded by raster-scanning the tip across a  $5 \times 5 \mu\text{m}^2$  area with a pixel

<sup>2)</sup> At this point, it may be interesting to note that R. Z. seemed always to be quite skeptical about the usefulness of near-field techniques. C. L. remembers well a discussion at ICPS24 – we had just published our first results on quantum wires [36] – in which R. Z. tried to convince him that in particular the metal coating and its interaction with the semiconductor would make it difficult to interpret any of these near-field measurements. This was in fact not too encouraging for someone just trying to enter this field.

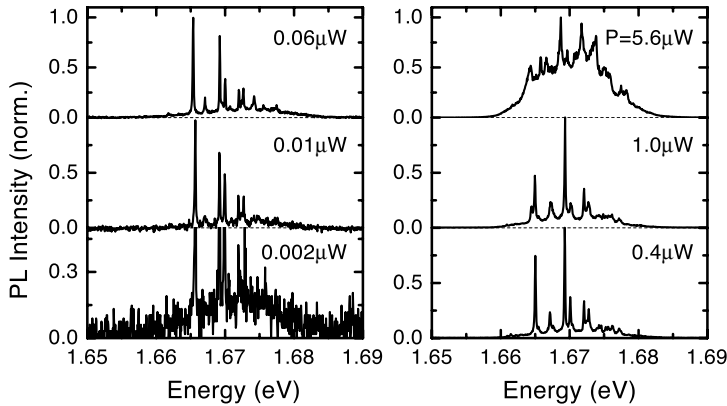


Fig. 2. Power dependent near-field QWR PL spectra taken at 10 K. The excitation power is varied between  $P = 0.002$  and  $5.6 \mu\text{W}$ . Note that the broad continuum assigned to the emission of delocalized excitons contributes significantly to the PL spectra even at the lowest excitation density

of 100 nm. The PL images recorded at the spectral position of the sharp exciton lines show indeed that this emission stems from regions resolution-limited in size (Fig. 3b). In the low-energy side of the broad continuum (out of the sharp resonances), however, the PL extends to regions of 400–600 nm in diameter (Fig. 3c). For detection in the high energy part of the background PL band, we find a spatially homogeneous emission, extending over mesoscopic distances of more than  $2 \mu\text{m}$  along the QWR (Fig. 3d). This behavior is consistent with results from simulations of excitonic absorption spectra of a disordered QWR [6]. In these simulations, we followed Ref. [10] and described the effective COM potential  $V(\mathbf{R})$  by adding a Gaussian-correlated disorder potential with an amplitude  $\sigma = \text{meV}$  and a correlation length  $\xi = 20 \text{ nm}$  to the average QWR confinement potential [18]. The simulations show clearly the contributions from localized excitons with COM wave functions of nearly Gaussian shape and extensions of tens of nm. These localized excitons couple strongly to light and give rise to sharp emission peaks in the near-field spectra. They also show a large density of states that are delocalized along the QWR axis. “Delocalized” is defined from the perspective of an experimentalist as larger than the extension of a single potential minimum and the spatial resolution of the experiment. Strictly speaking, all wave functions in a disorder potential are localized in two or lower dimensions, see, e.g. [19]. Their wave functions are highly complex and can be described as the disorder-induced interference of a broad distribution of plane waves [20]. Since their spectral linewidth is smaller than the energy separation between neighboring resonances, individual resonances are not resolved in the experimental spectra. The spectrally broad and spatially homogeneous emission in Fig. 3 is attributed to those delocalized excitons. This coexistence of wave functions with different degrees of localization and their interplay in the optical is a general feature of disordered systems [21, 22].

In these simulations, we found that a rather short correlation length of  $\xi = 20 \text{ nm}$ , only slightly larger than the excitonic Bohr radius, gave a good agreement with our experimental results. This short correlation length is partly due to the inherent corrugation

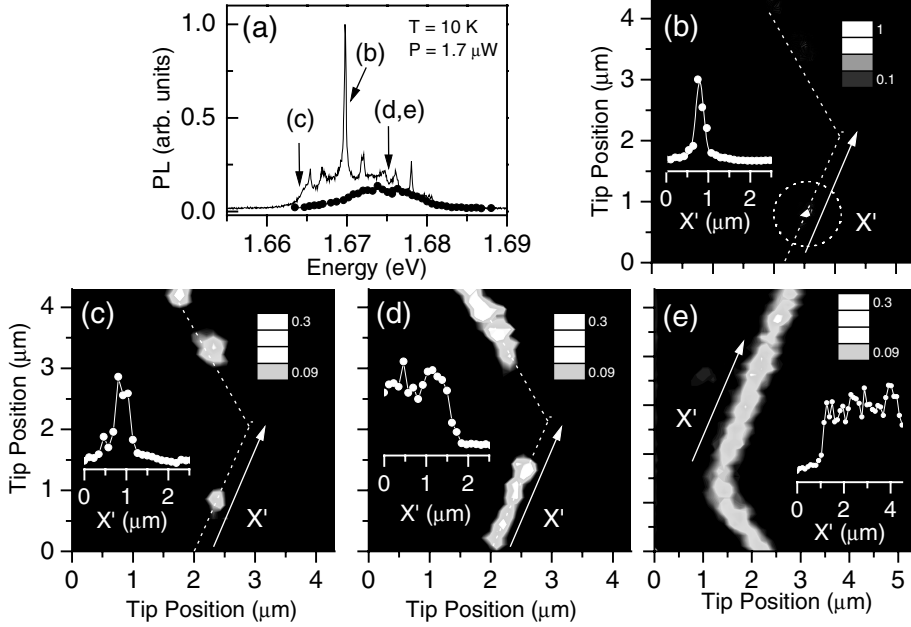


Fig. 3. a) Near-field PL spectrum of the QWR (solid line) taken at 10 K at a fixed tip position corresponding to (2.5 and 0.5  $\mu\text{m}$ ) in b) to d). Spectral distribution of the spatially delocalized PL contribution (circles). b) Two-dimensional near-field PL image taken at a fixed detection energy of  $E_{\text{det}} = 1.6698$  eV (sharp emission peak in a), excitation 1.96 eV) with a spectral resolution of 100  $\mu\text{eV}$ . The spatial orientation of the QWR-dot structure is indicated as a dotted line. Inset: cross section through the 2D image along the QWR axis  $X'$ ; c) same as b), with  $E_{\text{det}} = 1.6643$  eV. d) same as b) with  $E_{\text{det}} = 1.674$  eV (delocalized PL component). e) Near-field PL image taken at  $E_{\text{det}} = 1.674$  eV (delocalized PL component) for excitation at 1.70 eV at a different spatial position on the sample covering a corner where two QWRs are not interrupted by dots

of the (311)A GaAs surface [23]. This makes the sample interesting to investigate another key concept in the theory of localization, the correlations between energy levels of localized exciton states [24]. Theory indicates that a robust correlation found in all disordered systems is the so-called level repulsion. If the wave functions of two eigenstates with the same spin quantum numbers are spatially overlapping, their eigenenergies cannot be degenerate but split into two levels of different energy. On the contrary, states which are localized in non-overlapping spatial regions are allowed to be quasi-degenerate. Runge and R. Z. [10] pointed out that the level repulsion effect should be verifiable by a statistical analysis of the two-energy autocorrelation function of local optical spectra  $R_c(\Delta E) = R(\Delta E) - R_0(\Delta E)$ , where

$$R(\Delta E) = \langle \int dE' I_n(E') I_n(E' - \Delta E) \rangle, \quad (1)$$

$$R_0(\Delta E) = \int dE' \langle I_n(E') \rangle \langle I_n(E' - \Delta E) \rangle. \quad (2)$$

Here,  $I_n(E)$  represents the  $n$ -th local optical spectrum taken on the sample under investigation, and  $\langle \dots \rangle$  denotes the ensemble average over many individual measurement spots.

Figure 4 shows experimental autocorrelation functions measured at three different temperatures. The first discussion with R. Z. about these data, this time at ICPS25, was

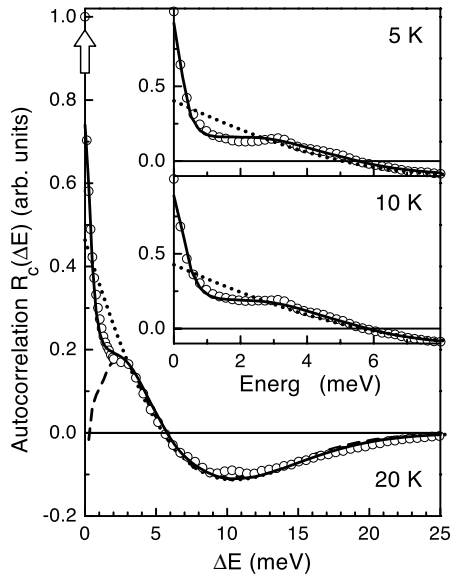


Fig. 4. Autocorrelation function  $R_c(\Delta E)$  at  $T = 20$  K, shown for the experimental data (circles), the numerical simulation (solid line), the classical limit (dotted line) and the numerical simulation with the self-correlation peak (dashed line). Level repulsion is evident from the shoulder below 3 meV. The arrow denotes the  $\delta$ -function part (self-correlation) of the dashed curve. Insets: level repulsion of autocorrelation functions recorded at  $T = 5$  and 10 K, respectively

actually quite controversial and ended with R. Z. stating that “experimentalists always measure something”. It took a few more rounds of discussion and the calculations by Vincenzo Savona and Erich Runge to reach a certain agreement between experimentalists and theoreticians. The individual curves are extracted from a set of over

400 near-field photoluminescence spectra recorded on a 3 nm (311)A GaAs QW region in the coupled wire-dot structure discussed above [6]. The interpretation of these data requires a careful lineshape analysis of  $R_c(\Delta E)$ , as  $R_c(\Delta E)$  is not only sensitive to “quantum mechanical” correlations due to level repulsion but also to “classical” correlations of the underlying disorder potential  $V(\mathbf{R})$ . Even in the “classical” limit,  $\hbar \rightarrow 0$ , where the eigenvalues of the Schrödinger equation are simply given by the local values of  $V(\mathbf{R})$ ,  $R_c(\Delta E)$  takes on finite values. In potentials with finite correlation lengths, it is more likely to find similar values of  $V(\mathbf{R})$  in the immediate vicinity and hence  $R_c^{(\text{class})}(\Delta E)$  is positive at small energy values. This positive correlation is compensated for by negative values of  $R_c^{(\text{class})}(\Delta E)$  at large energy differences, and the integral  $\int R_c^{(\text{class})}(\Delta E) d\Delta E$  vanishes. For a given form of the potential correlation, an analytic expression for  $R_c^{(\text{class})}(\Delta E)$  can be obtained, which is shown in Fig. 4 as a dotted line for a potential with a single correlation length of  $\xi = 17$  nm. This classical autocorrelation is in good agreement with experiment for all values  $\Delta E > \text{meV}$  (Fig. 5). The effects of the exciton kinetic energy are twofold. First, a spectrally narrow positive self-correlation peak exists around  $\Delta E = 0$ . It arises from the self-convolution of the individual spectrally sharp emission spikes in the near-field PL spectra (Fig. 2). Its width is roughly twice the linewidth of the localized exciton PL spikes. Second, and most important here, the level repulsion effect shows up as a broad dip in  $R_c(\Delta E) - R_c^{(\text{class})}(\Delta E)$  for small values of  $\Delta E$ . Due to level repulsion, the values of  $R_c(\Delta E)$  are distinctly smaller than those of the classical autocorrelation  $R_c^{(\text{class})}$  in this energy range. This dip is clearly observed in our experimental data (Fig. 5). The data are in good agreement with theoretical simulations based on solutions of a two-dimensional one-particle Schrödinger equation [25, 26] for a statistical disorder potential with a correlation length  $\xi = 17$  nm. We consider this very strong evidence for quantum mechanical level repulsion in disordered semiconductor nanostructures. Similar conclusions are drawn in a recent investigation of autocorrelation spectra of V-groove QWR [27].

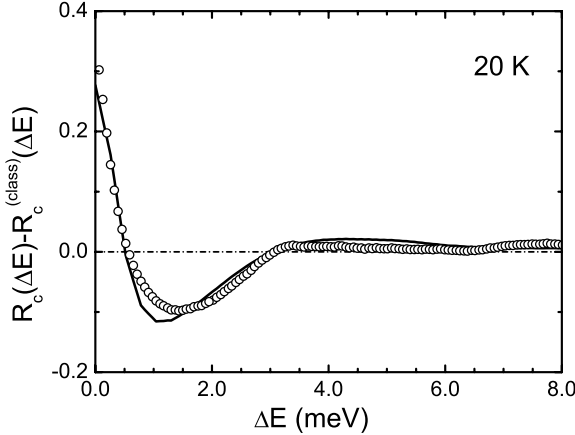


Fig. 5. Difference between  $R_c(\Delta E)$  at  $T = 20$  K and the classical autocorrelation  $R_c^{(\text{class})}(\Delta E)$ . The experimental data are shown as circles and the numerical result as a solid line. The level repulsion effect gives rise to *negative* values of  $R_c(\Delta E) - R_c^{(\text{class})}(\Delta E)$

One should note that, at present, PL experiments are compared to theoretical simulations of absorption spectra. This seems valid only if the effective temperature of the exciton gas is larger than the energy separation between neighboring localized states. The experimental results in Ref. [6] and kinetic simulations [7] seem to indicate that this assumption is reasonably valid for the investigated sample. As discussed above, the autocorrelation function contains contributions from both “classical” potential correlations and from quantum-mechanical level repulsion. It may a priori be non-trivial to disentangle these two contributions without independent information about the disorder potential. In our case, the excellent agreement between  $R_c(\Delta E)$  and the simulations for a statistically fluctuating disorder potential with a finite, rather short correlation length is striking and the evidence for the level repulsion effect is clear. In samples, where the disorder strength is much larger than the exciton binding energy [28] or where the disorder potential is not well described by a single correlation length (e.g. because of the existence of large monolayer islands [5, 29]), the shape of  $R_c(\Delta E)$  may be more complex than described in [10, 12] and observed in [25]. Also in such cases the analysis of  $R_c(\Delta E)$  promises to give valuable information about the microscopic properties of the underlying disorder potential [29]. This can be seen, e.g., in autocorrelation functions  $R_c(\Delta E)$  recorded on thin, high quality (100) GaAs QW (Fig. 6). These results show an additional correlation peak at energies around 3–4 meV. The peak energy shifts to the blue with decreasing QW size. Such a peak is not reproduced by statistical models assuming a single correlation length. It is a signature of correlations between excitonic eigenstates localized within a single potential minimum and these minima obviously have similar shape and size. This will be discussed in more detail in a forthcoming publication.

The success of these experiments mainly relied on improvements in sensitivity and stability of low-temperature near-field spectroscopic techniques. These improvements made it possible to probe ultrafast optical nonlinearities of *single* quantum dots. There are two main driving forces behind such studies. First, it is of fundamental interest to probe the interactions of QD optical excitations with charge or spin excitations in their environment, e.g., of neighboring quantum dots. Second, optical excitations of single quantum dots have recently been theoretically proposed by different groups as building blocks (qubits) for semiconductor-based implementations of quantum logic [30, 31].

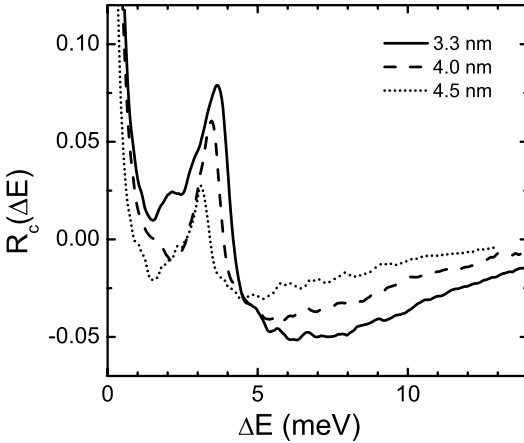


Fig. 6. Autocorrelation function of  $R_c(\Delta E)$  three different growth-interrupted (100) GaAs QW at  $T = 10$  K. The QW thicknesses are 3.3 nm (thick solid line), 4.0 nm (thin solid line), and 4.5 nm (dotted line). The correlation peak around 3–4 meV most likely reflects the correlation between ground and excited state PL of excitons localized in the same island

Since the dephasing times of excitonic polarizations in QDs are comparatively short (10–1000 ps), such implementations rely on the use of ultrafast optical pulses to control and probe coherent polarizations in such systems. We have recently demonstrated a new experimental concept [32], based on ultrafast near-field pump–probe spectroscopy, that allows us to probe single QD optical nonlinearities on ultrafast time scales. These experiments were performed on interface QD in a 5.1 nm thick (100) GaAs QW layer, buried 120 nm below the sample surface. Briefly, ultrafast probe pulses, spectrally resonant with the excitonic QD transition at 1.6558 eV, are transmitted through an uncoated near-field fiber probe and the reflected probe light is locally collected through the same fiber. The collected light is dispersed in a monochromator and detected with a high spectral resolution CCD camera (Fig. 7a). A second, off-resonant pump pulse at 1.675 eV creates about five electron–hole pairs in QW continuum states. The pump pulse modifies the probe reflectivity and differential reflectivity spectra  $\Delta R(E_{\text{det}}, \Delta t)/R_0 = [R(E_{\text{det}}, \Delta t) - R_0(E_{\text{det}})]/R_0(E_{\text{det}})$  are recorded at a fixed spatial

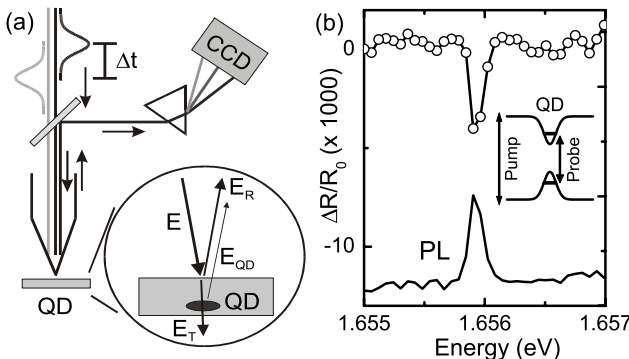


Fig. 7. a) Schematic of the experimental setup. b) Near-field PL spectrum of a single QD (solid line) and differential reflectivity  $\Delta R/R_0$  at  $\Delta t = 30$  ps. PL and  $\Delta R$  are recorded with identical pump pulses centered at 1.675 eV, exciting electron–hole pairs in 2D continuum states. Inset: schematic energy diagram

position of the near-field tip as a function of the time delay  $\Delta t$  between pump and probe pulses ( $R_0(E_{\text{det}})$ : steady state reflectivity at a photon energy  $E_{\text{det}}$ ). For delay times  $\Delta t > 0$ , the differential reflectivity spectra display spectrally sharp resonances at the same spectral position,  $E_{\text{QD}}$ , as the simultaneously recorded PL lines (Fig. 7b). The linewidth of PL and  $\Delta R$  is mainly limited by the monochromator resolution. The time evolution of  $\Delta R(E_{\text{QD}})$  at the QD resonance (Fig. 8) shows an 8 ps picosecond rise at negative delay times, much slower than the 150 fs cross correlation of pump and probe pulses. A biexponential decay is found for  $\Delta t > 0$  with a fast decay time of 6 ps, followed by a much slower decay. The fast signal components are very similar for different QD's investigated, whereas the decay time of the slow component varies between 30 and 150 ps. The spectral characteristics of  $\Delta R/R_0$  are markedly different at positive and negative  $\Delta t$  [32]. At  $\Delta t < 0$ , pronounced *spectrally symmetric* oscillations around the excitonic resonance are observed. Their oscillation period decreases with increasing negative delay. At  $\Delta t \gg 0$ , the spectra show a bleaching of the QD resonance.

The spectral characteristics of the reflectivity spectra are well described within a model that assumes that the reflected probe field represents a coherent superposition of the field  $E_R(t)$  reflected from the sample surface, and the field  $E_{\text{QD}}(t)$ , radiated in back-direction from the probe-induced excitonic polarization of the QD,  $P_{\text{QD}}(t) = \int dt' \chi_{\text{QD}}(t') E_T(t - t')$ , located at a distance  $d$  below the sample surface. Here,  $E_T(t)$  and  $\chi_{\text{QD}}$  denote the probe field interacting with the QD and the QD susceptibility, respectively. The time-integrated reflectivity  $R_0(\omega)$  detected behind the monochromator is proportional to  $|\tilde{E}_{\text{QD}}(\omega) + \tilde{E}_R(\omega)|^2$ , where a tilde denotes the Fourier transform. The differential reflectivity  $\Delta R(\omega, \Delta t)$  then represents the spectral interferogram of  $\tilde{E}_R(\omega)$  and  $\tilde{E}_{\text{QD}}(\omega, \Delta t)$ .

From a detailed analysis of the transient spectra, the following conclusions can be drawn: (i) At negative delay times, the probe laser induces a coherent QD polarization, decaying with the dephasing time  $T_2$ . Due to the finite monochromator resolution, we do not probe  $T_2$  directly and can only give a lower limit of  $T_2 > 22$  ps. The off-resonant

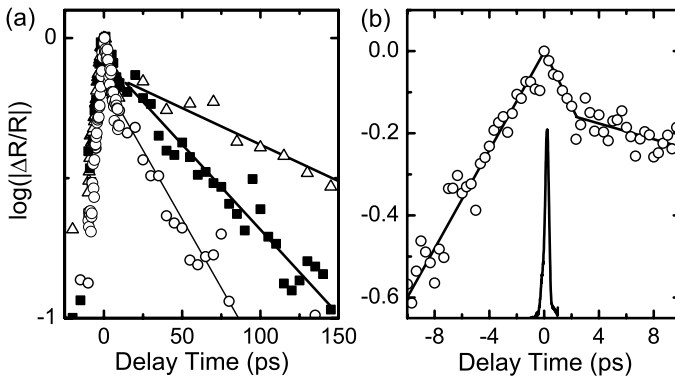


Fig. 8. a) Time evolution of  $\Delta R(E_{\text{QD}})/R_0$  for three different QD resonances (logarithmic ordinate scale). All decays at  $\Delta t > 0$  are biexponential with slow decay times between 30 and 150 ps. b) Early time dynamics of  $\Delta R(E_{\text{QD}})/R_0$  of a single QD resonance. A slow rise of  $\Delta R/R_0$  is observed at negative delay times. Solid line around delay zero: cross-correlation of pump and probe pulses indicating a time resolution of 150 fs



pump laser creates electron–hole pairs in QW states well above the QD resonance. The Coulomb scattering between the QD polarization and this initial *nonequilibrium* carrier distribution perturbs the free induction decay polarization by causing an additional fast damping. In the frequency domain, this excitation-induced dephasing [33] leads to oscillatory features in the spectrum  $\Delta R(\omega, \Delta t)$  with a period determined by the time delay between probe and pump, similar to oscillations that have been observed before in studies of transient QW nonlinearities [34]. This pump-induced damping of the QD polarization after the arrival of the pump pulse is the leading contribution to the QD nonlinearity at early times. Under our experimental conditions, the transient spectral oscillations are phenomenologically well described by assuming that the presence of the pump laser increases the QD dephasing rate from its initial value  $T_2 > 22$  ps to  $T_{\text{EID}} = 3$  ps (excitation-induced dephasing). A theoretical analysis of the data was performed on the basis of the semiconductor Bloch equations in mean-field approximation [32]. Excellent agreement between experiment and theory was obtained by assuming a phenomenological dephasing rate  $\gamma = 1/T_2 + \gamma_1 n$  depending on the excitation density  $n$ . The theoretical simulations show clearly the importance of EID contributions to the perturbed free induction decay. (ii) This assumption of a density-dependent dephasing rate can also explain the fast decay of  $\Delta R(E_{\text{QD}}, \Delta t)$  at early positive delay times. We have to assume that  $n$  reflects the *nonequilibrium* carrier distribution in QW continuum states and that this nonequilibrium distribution decays on a time scale of about 3 ps. This decay is most likely due to carrier trapping into QD states. Then the initial fast decay of  $\Delta R(E_{\text{QD}}, \Delta t)$  reflects the transition from a QD nonlinearity that is dominated by EID to a nonlinearity dominated by exciton bleaching due to population relaxation into the QD. (iii) The slow decay of  $\Delta R(E_{\text{QD}}, \Delta t)$  is determined by radiative recombination [5] and inversely proportional to the QD dipole moment [35]. We estimate dipole moments of 60 to 110 Debye for between 30 and 100 ps. These values exceed those of atomic systems by more than an order of magnitude and reflect the strong light-matter coupling of these QD excitons. These values are in good agreement with the very large, experimentally measured amplitudes of the reflectivity changes of up to several percent.

These results show quite clearly that although single QDs resemble in many respect atomic systems, Coulomb many-body interactions can contribute significantly to optical nonlinearities on ultrashort time scales and should be considered as an additional dephasing mechanism. On the other hand, the strong Coulomb and light-matter interactions in excitonic systems opens up new and interesting ways for a controlled ultrafast manipulation of coherent QD polarizations, e.g. via dipole–dipole interactions of neighboring quantum dots. This may eventually be relevant for semiconductor-based implementations of quantum information processing.

In summary, this review gives some examples of the intense and fruitful collaboration between theory – often linked to the work of R. Z. and his group – and experiment that has developed over the last few years in an attempt to unravel the rich physics behind the inhomogeneously broadened far-field spectra of thin disordered quantum films. We are convinced that R. Z. will continue to be such a stimulus for his experimental and theoretical colleagues and wish him all the best for the years to come.

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