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Nonequilibrium carrier dynamics in self-assembled InGaAs quantum dots

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Carrier dynamics in InGaAs/GaAs quantum dots is analyzed with highly sensitive femtosecond transmission spectroscopy. In a first step, measurements on a large ensemble of nanoislands reveal the dynamical electronic filling of quantum dots from the surrounding wetting layer. Most interestingly, we find a spin-preserving phonon mediated scattering into fully localized states within a few picoseconds. Then, individual artificial atoms are isolated with metallic shadow masks. For the first time, a single self-assembled quantum dot is addressed in an ultrafast transmission experiment. We find bleaching signals in the order of 10^{-5} that arise from individual interband transitions of one quantum dot. As a result, we have developed an ultrafast optical tool for both manipulation and read-out of a single self-assembled quantum dot.

1 Introduction

Semiconductor quantum dots (QDs) are currently attracting much attention both as model systems to study the physics of quasi-zero dimensional systems and for their device applications [1]. For electronic devices, the discrete energy level structure of QDs offers significant advantages, e.g. in modern semiconductor lasers. Nonequilibrium carrier dynamics plays a central role in determining the performance limitations in these structures and has therefore been subject to considerable research efforts [2, 3]. Most of the studies of carrier relaxation in QDs rely on time-resolved photoluminescence spectroscopy, thus analyzing the scattering of carriers towards the lowest lying QD states [4–8]. Morover, the relaxation of carriers towards the QD ground states has been analyzed by differential transmission [9] and THz absorption spectroscopy [10].

In contrast, much less is known about the initial steps of the population transfer from the two dimensional wetting layer (WL) into the zero-dimensional structures. Theoretically, it has been argued that carrier-carrier interaction may be an efficient way to circumvent the "phonon bottleneck" [11, 12] that arises from the mismatch of the electronic level spacing with respect to the accessible phonon energies [13]. This result is supported by the recent experimental observation of ultrafast electron-capture in the presence of a p-modulation-doping within the QDs [14].

Semiconductor quantum dots are also attractive for studying the physics of quasi zero-dimensional solid state systems. Especially, self-organized III–V semiconductor QDs combine remarkably long coherence times of excitonic transitions with the potential for ultrafast optical switching [2]. As a result, coherently manipulating zero-dimensional nanostructures with femtosecond laser pulses is a promising concept for solid state-based quantum information processing [15]. However, most of these concepts

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require both ultrafast optical switching and all-optical read-out of a single QD [16] – a demanding experimental task. Until now, an analysis of the nonlinear optical response of a single QD has only been realized for the case of excitons in disordered quantum wells that typically exhibit enormous transition dipoles but only very weak confinement energies [17, 18]. For isolated self-assembled QDs, Rabi oscillations have been studied extensively, thus proving the possibility of precise single qubit rotations [19–21]. For the case of excitons in disordered quantum wells, even two qubit operations have been demonstrated [22]. Moreover, decoherence times approaching the nanosecond time scale have been extracted from four wave-mixing experiments [2]. However, the time-resolved all-optical read-out of a single self-organized QD remains to be demonstrated.

In this contribution, we report on two different aspects of nonequlibrium carrier dynamics in self-organized InGaAs quantum dots. In Section 2, we directly analyze the transfer of photoinjected carriers from the WL to fully quantized QD levels [23]. The experiment relies on the transient bleaching of the WL band edge and the QD interband transitions in a two-color femtosecond transmission experiment. We find a capture time of 3 ps indicating independent of the density of carriers photoinjected into the WL. Moreover, exploiting the selection rules for circularly polarized light, the carrier capture of electrons is shown to be spin-preserving. In Section 3, we present the first experimental realization of a two-color femtosecond transmission experiment ofa single InGaAs/GaAs QD. The main focus of this section is to demonstrate the experimental sensitivity to study one artificial atom in an ultrafast experiment. The nanostructure is isolated combining a low-density growth technique with nanometer-scale aluminum shadow masks. The transmission changes of one aperture with 450 nm diameter is analyzed after femtosecond photogeneration of electron–hole pairs in the wetting layer beneath the QD. Most strikingly, the nonlinear optical response clearly resembles several distinct excitonic transitions of the QD under investigation. The QD typically generates modifications of the probe photon flux in the order of 10^{-5}, thus being accessible in a shot noise limited experimental setup. The pulse energies necessary for both the manipulation and read-out of the single QD are as low as a few femtojoule.

2 Spin-preserving ultrafast carrier capture and relaxation in InGaAs quantum dots

The nanostructures of this study are self-organized InGaAs QDs grown on a GaAs. The ground state transitions of the inhomogeneously broadened ensemble are centered at 1.25 eV and the interband energetic spacing of the transitions from different QD shells is approximately 40 meV, i.e. in the order of the LO phonon energy $\hbar\omega_{\mathrm{LO}} = 36$ meV of the host material GaAs.

The scheme of our experimental approach is displayed in the inset of Fig. 1(a). A pump pulse with a duration of 100 fs is tuned to a central photon energy of 1.51 eV in the absorption continuum of the WL, thus resonantly creating electron–hole pairs in this two dimensional layer. After relaxation towards the band edge, the carriers are transfered into fully quantized QD levels. Correspondingly, a transient optical bleaching signal is established reflecting the occupation of the electronic states. These transmission changes of the QD monolayer are detected with a 20 fs probe pulse that is derived from the second branch of a two color-Ti:sapphire laser [24] and spectrally dispersed after transmission through the specimen. This second broadband laserpulse is typically one order of magnitude smaller as compared to the excitation pulse and is used as a test pulse to monitor the ultrafast dynamics of the sample.

Results for a moderate excitation density and a probe photon energy of 1.45 eV, near the band edge of the WL, are shown in Fig. 1(a). The transmission change rises within a few hundred femtoseconds reflecting the thermalization of nonequilibrium photogenerated carriers. More interestingly, the bleaching signal decays exponentially with a time constant of 3 ps. The negative signal detected for delay times $t_{\mathrm{D}} > 6$ ps arises from band gap renormalization (specifically, the nonthermal carriers lead to a small reduction of the band gap energy of the WL. As a result, the density of states for a given photon energy is enlarged leading to an induced absorption in the experiment). These dynamics are accompanied by the buildup of a transmission change probing the QD excited levels (see Fig. 1(b)). Thus, the present measurement directly traces the population transfer from the two dimensional WL into the QDs.

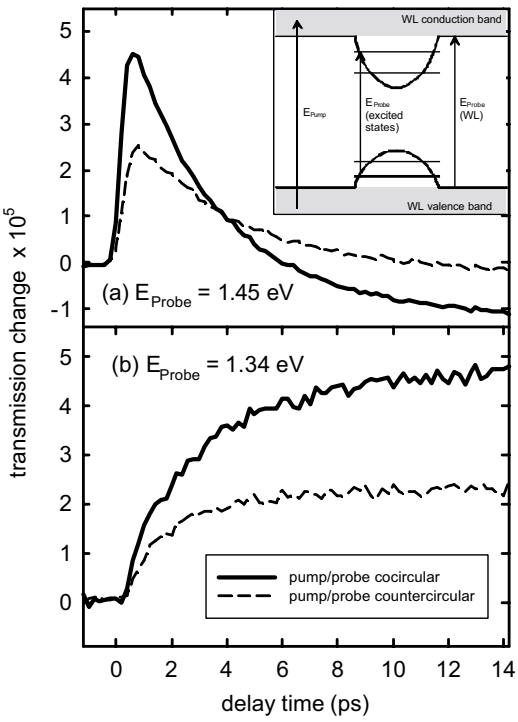

Fig. 1 (a) Transmission changes detected at the band adge of the WL at a probe photon energy of 1.45 eV after excitation with an 100 fs pulse centered at 1.51 eV. The solid (dashed) line corresponds to a co-circular (counter-circular) polarization of excitation and probe beams. The inset sketches the bandstructure with the vertical arrows indicating the interband transitions of the study. (b) Corresponding results detecting the transmission changes of the QD excited states at a probe photon energy of 1.34 eV.

More detailed insight into the carrier capture process is gained exploiting the spin selectivity of the interband transitions. Circularly polarized excitation yields a preferential spin orientation of the photogenerated carriers. While the spin orientation of holes is rapidly destroyed due to spin–orbit interaction, the electron spin in the two-dimensional WL is expected to be preserved over the timescale of the present study. As a consequence, detecting the transmission changes of the WL with a counter-circularly polarized probe pulse yields an overall reduction of the transmission change by a factor of two (see dashed line in Fig. 1(a)). The limited degree of polarization may be attributed to the strong disorder and band mixing in the WL. More surprisingly, the nonlinear optical response of the QD interband transitions depends on the polarization configuration in the same manner as observed for the WL (see Fig. 1(b)). These observations demonstrate that the carrier capture process is a predominantly spin-preserving process. Comparing the degree of polarization for the two probe photon energies, we even do not see evidence for *any* spin relaxation during the population transfer.

In order to identify the nature of the capture process, the QD filling is studied for various excitation densities. To this end, we analyze the transient transmission changes of the WL band edge at a probe photon energy of 1.45 eV as a function of the excitation power. Examples of the experimental data are displayed as symbols in Fig. 2(a). For weak excitation, we find that a exponential decay with a time constant of 3 ps provides a good fit to the experimental data (compare the lower gray line). Most interestingly, the decay times extracted from exponential fits (see filled circles in Fig. 2(b)) do not significantly depend on the excitation density indicating that carrier-carrier scattering has only a minor influence on the capture dynamics.

Increasing the excitation density to values comparable or larger than the QD density on the sample, the decay of the bleaching signal is effectively slowed down due to the emergence of a slowly varying signal component. This finding is readily understood from the onset of macroscopic state filling effects in the QDs. Modelling these dynamics with a bi-exponential fit, we identify a fast capture process with a time constant of again 3 ps and a relatively long-lived component (compare the filled triangles in Fig. 2(b)). For the highest excitation densities of the study, the signal is dominated by massive QD filling and decays only on a timescale approaching typical radiative multiexciton lifetimes in QDs.

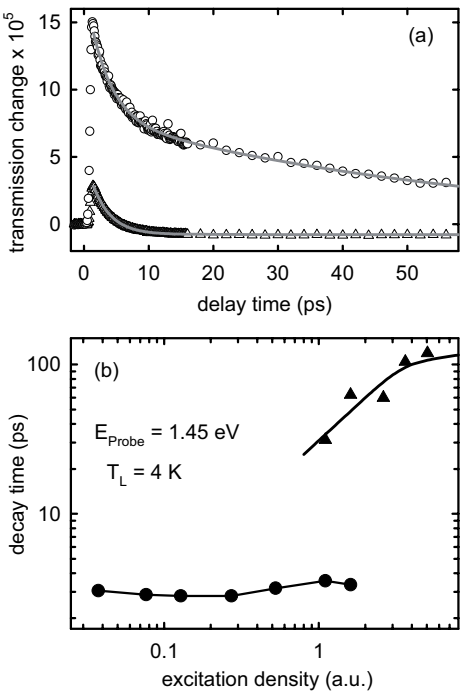

Fig. 2 (a) Transmission changes detected at the band edge of the WL at a probe photon energy of 1.45 eV for two different excitation densities. Symbols: experimental data, gray lines: exponential fits as discussed in the text. (b) Decay times ofthe WL population for various excitation densities. The time constants are extracted from the transmission changes of the WL band edge at 1.45 eV by single and double exponential fits as discussed in the text. The abscissa may be interpreted as an estimate for the number of photogenerated electron–hole pairs per QD.

The independence of the capture time on the excitation density clearly points towards a phonon mediated process that is most likely governed by LO phonon scattering in the polar host material. It is interesting to relate this finding to previous experimental results claiming the importance of carrier-carrier scattering for the relaxation dynamics. Time-resolved luminescence experiments [4, 6, 7] have demonstrated the importance of, e.g. Auger-type processes. However, this experimental approach is intrinsically sensitive to a combination of carrier capture and relaxation towards the QD ground state. Moreover, intraband spectroscopy [10] has shown a significant influence of electron–hole scattering for the electron dynamics in QDs with an electronic level spacing large as compared to optical phonon energies. In contrast, we study the capture into electronic states near the onset of the WL continuum in QDs with a level spacing comparable or smaller as the LO phonon energy. In this situation, model calculations [13] have predicted that phonon mediated processes may dominate the capture dynamics. Moreover, our results are supported by a time-resolved luminescence study on large InGaAs quantum dots [8].

In conclusion, we have analyzed the ultrafast, spin-preserving population transfer from a two dimensional WL to fully quantized QD levels in a femtosecond transmission experiment. The carrier capture time in our QDs with a level spacing comparable or smaller as the optical phonon energies amounts to 3 ps and is found to be independent of the excitation density. Taken together, these observations strongly indicate a phonon mediated process. As a result, we see no significant phonon bottleneck for the filling of the QD states. This finding may be an important ingredient for the optimization of modern QD based laser devices.

3 Nonlinear optical analysis of a single self-assembled InGaAs quantum dot

In this section, we study InGaAs/GaAs quantum dots that are nominally identical to the samples discussed above. In addition, aluminum shadow masks are deposited on a low-density area of the specimen with diameters between 200 nm to 450 nm. As a consequence, single QDs may be precisely addressed using standard free-space optics.

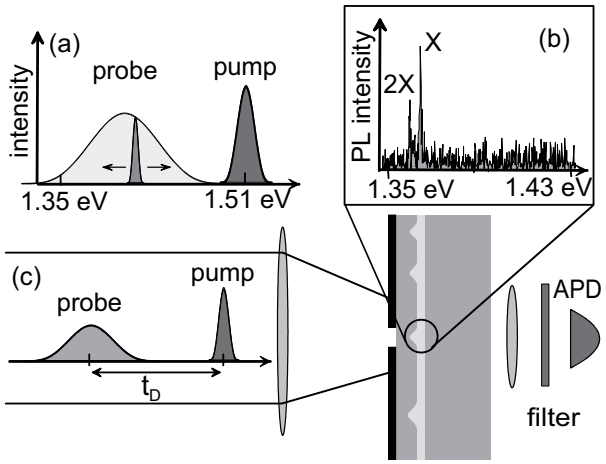

Fig. 3 (a) Excitation and probe pulses used in the experiment. The probe pulses of an energetic width of 1.2 meV are extracted from a broadband probe continuum using a pulse shaper. (b) Photoluminescene of the nanoaperture for the case of a pulsed, low intensity excitation. (c) Scheme of the experiment: pump and probe pulse trains are focussed through the metallic shadow mask of 450 nm diameter. The transmitted light is detected with an avalanche photodiode (APD) after spectral filtering. The sample is held at $T_L = 4\,\mathrm{K}$ in a microscope cryostate.

In this study, we focus on a single InGaAs QD characterized in Fig. 3(b). The specimen is mounted on the cold finger of a microscope cryostat ($T_L = 4\,\mathrm{K}$) for all present experiments. The photoluminescence spectrum observed for a pulsed low intensity excitation at $E = 1.51$ eV shows two pronounced, resolution limited peaks (the spectral resolution of the luminescence detection is 150 μeV). Varying the excitation power, these transitions are identified as excitonic (X, $E = 1.3735$ eV) and biexcitonic transition ($2X$, $E = 1.3696$ eV). The confinement energy in this QD is approximately 35 meV and 15 meV for electrons and holes, respectively, i.e. much larger than for localized excitons in disordered quantum wells. Interestingly, we do not see significant luminescence contributions from charged exciton configurations. We find this result to be related to exciting the quantum dot near the band edge of the WL.

The experimental scheme is sketched in Fig. 3(a) and (c). Two synchronized femtosecond pulse trains with 76 MHz repetition rate are derived from a two-color Ti:sapphire laser [24]. The 100 fs pump pulse is tuned to the absorption continuum of the wetting layer beneath the InGaAs QD thus efficiently filling the nanostructure with nonequilibrium carriers. The 30 fs probe pulse encompasses the entire spectral range from 1.36 eV to 1.44 eV giving access to both QD interband transitions and the wettinglayer band edge. Before transmission through the sample, probe pulses with a spectral width of 1.2 meV are extracted from this broad band pulse using a low-dispersive pulse-shaping technique [29]. As a result, transform limited pulses of 1.5 ps duration are used as test pulses for the ultrafast transmission experiment. As depicted in Fig. 3(c), both excitation and time-delayed probe beams are focussed onto the sample using a dispersion-free reflecting mirror objective with a numerical aperture of 0.4 that allows for a typical spot diameter of 1.5 μm. The light transmitted through the nanoaperture is collected with a microscope objective, filtered in order to reject transmitted pump photons and analyzed with an avalanche photo diode. Subsequently, the pump induced transmission changes are extracted using a lock-in technique with a 30 kHz modulation of the excitation pulse. The present scheme offers several distinct advantages over conventional pump–probe experiments that spectrally disperse the test pulse after transmission through the sample: (i) The probe photon flux per energy interval is kept high giving rise to an improved shot noise-limited sensitivity for the detection of ultrafast transmission changes. Simultaneously, the collection efficiency of probe light is very high. (ii) The total intensity of the test pulse can be restricted to a few fJ thus efficiently suppressing probe induced optical nonlinearities. (iii) The time resolution of 1.5 ps is sufficient to study typical relaxation phenomena in QDs.

We now turn the analysis of the nonlinear optical response of this specific quantum dot characterized in Fig 3(b). Figure 4(b) displays the relative transmission changes observed at a time delay of $t_D = 20$ ps after photoexcitation for two different intensities of the pump pulse. For comparison, the luminescence of the nanoaperture for these two excitation densities is depicted in Fig. 4(a). Most strikingly, we clearly identify a bleaching signal centered at the excitonic transition X even for a pump intensity as low as 2 fJ. Raising the pump power to 10 fJ, the amplitude of the ground state bleaching remains at a level of

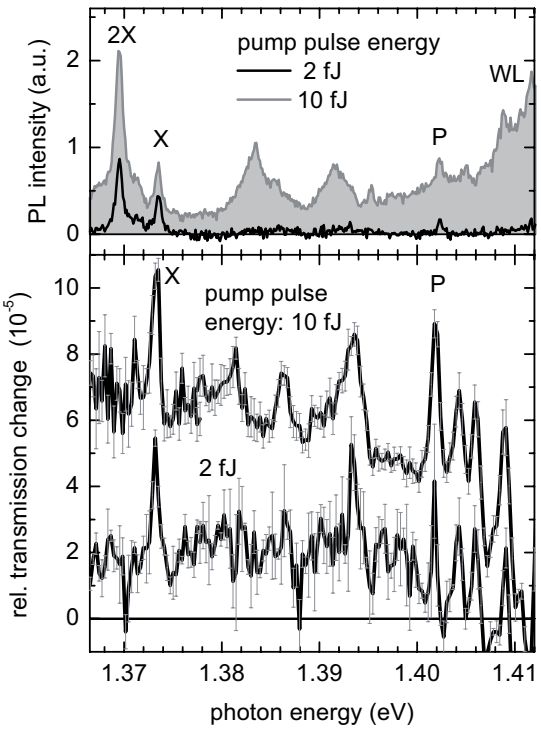

Fig. 4 (a) Photoluminescence (PL) spectrum for two different excitation intensities. The energy values indicated in the graph correspond to the pulse energy that is incident on the geometrical area of the shadow mask. (b) Transmission changes of the aperture detected at $t_D = 20$ ps after excitation of the sample with a 100 fs pulse centered at 1.51 eV. The spectral resolution of the experiment is determined by the energetic width of the tunable probe pulses of 1.2 meV. The data for the pump intensity of 10 fJ are shifted by 2 ordinate units for clarity.

3×10^{-5} indicating a saturation of this interband transition. As a consequence, this value may be seen as an estimate for both the maximum transmission changes associated with a single InGaAs QD under a shadow mask and the total ground state absorption in agreement with recent linear studies [30]. Unfortunately, it is not possible to relate the results to an absorptioncross section of the QD since we have no informationon the linewidth of the ground state transition. Additionally, a positive background signal appears especially for the higher excitation level of the study which might reflect a substrate induced optical nonlinearity (alternatively, this background could also be related to the background luminescence observed in Fig. 4(a)). Surprisingly, we do not see a clear signature of the biexciton $2X$ in the transmission changes (this finding might be related to a too strong excitation even for a 2 fJ pump pulse). For photon energies larger as compared to these s-shell transitions of the QD, the luminescence spectra in Fig. 4(a) show a series of broad emission peaks that are related to optical recombination from different multiexciton complexes. These excitonic transitions partially overlap with the emission of different localized states in the two-dimensional wetting layer with a band gap energy of 1.41 eV. Interestingly, some of these luminescence peaks have their direct counterpart in the nonlinear optical response. As an example, the luminescence peak P in Fig. 4(a) ($E_P = 1.4022$ eV) is paralleled by a remarkably sharp and pronounced nonlinear transmission peak in Fig. 4(b). However, a part of the luminescence peaks seems to be not visible in the transmission changes. This finding might be related to the luminescence of charged excitons in a highly excited dot which do not have an absorption cunterpart in an empty dot. For probe photon energies approaching the band edge of the wetting layer at 1.41 eV, we find a strongly structured nonlinear optical response that is related to different localized excitons in the wetting layer (only partially shown in Fig. 4). The amplitude of these transmission changes is one order of magnitude larger when compared to the ground state transition X. This finding is related to the giant transition dipole of these weakly confined excitonic complexes and is consistent with previous experiments [17].

In conclusion, we have demonstrated an experimental tool to both manipulate and read-out a single self-assembled quantum dot on ultrashort timescales. One artificial atom beneath a shadow mask of 450 nm diameter is found to generate modifications of a probe photon flux in the order of 10^{-5} thus being

accessible in a shot noise limited detection scheme. The transmission changes clearly reflect the optical bleaching induced by different multiexciton configurations. Extending the study to resonant QD pumping, we expect new insight into the physics of self-organized QDs and their potential applications in quantum information processing.

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