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Inter-sublevel dynamics in single InAs/GaAs quantum dots induced by strong terahertz excitation

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We combine micro-photoluminescence (PL) with terahertz excitation to investigate the response of single self-assembled InAs/GaAs quantum dots to intense terahertz pulses tuned to the s-to-p transition. Spectra and transients of single photoluminescence lines reveal the dynamics of electrons upon excitation and subsequent relaxation back into the initial state. Under certain circumstances, the terahertz pulse can release trapped charge carriers, which relax into the quantum dot. Furthermore, we demonstrate near-total depletion of the positive trion PL by an intense terahertz pulse. © *2016 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4942893]

Self-assembled III-V quantum dots (QDs) are a highly interesting material system for a variety of technological applications. In addition to achieving control over single exciton population[1–3] relevant for quantum cryptography and single photon and electron generation, the electrical and optical properties of QDs can be tuned over a wide range by changing their composition and size. In particular, inter-sublevel transitions in QDs have received much attention as a means for generating[4] and detecting[5] infrared light. However, these transitions have not been investigated as thoroughly as interband transitions—this imbalance caused in part by the experimental difficulties associated with the terahertz (THz) spectral range. Some efforts into this area have included the measurement and tuning of the energy spacing between s- and p-like states[6–8] and the observation of time-integrated photoluminescence (PL)[9] under THz excitation. The latter study included the observation of PL shifting away from the ground state towards the excited state transitions, as well as a broad resonance of this effect with respect to the THz photon energy. Two-color excitation experiments using the pump-probe technique[10] or streak-camera[11–13] have enabled time-resolved as well as spectrally resolved investigations, revealing the dynamics of inter-sublevel transitions. The aforementioned studies concerning inter-sublevel transitions have been performed with ensembles of QDs for which some degree of inhomogeneity is unavoidable, which can mask certain effects and introduce others like inter-dot tunneling.[12] In our present work, we investigate the response of a single QD to THz excitation, revealing the excitonic spectrum and allowing deeper insights into the various processes triggered by the terahertz pulses.

We have used a low-density self-assembled InAs/GaAs QD sample grown at 505 °C via molecular beam epitaxy (MBE), in which the average QD separation is larger than the spatial resolution of our setup of approximately 1 μm. A distributed Bragg reflector cavity for near-infrared wavelengths with 15 periods below the QDs and one period above was incorporated to improve the out-coupling of photoluminescence. The QDs were treated by a partial capping and annealing step[14] for 3 min at 550 °C in order to move the ground-state interband emission into the sensitivity range of a Si-CCD detector and to shift the s-p separation in the conduction band below the Reststrahlen band.[8,15] All measurements were performed at $T = 5$ K. The PL was excited with 3 ps pulses from a titanium sapphire laser. This beam was focused and the PL collected with a 100× microscope objective and a hemispherical high-index solid immersion lens placed directly on top of the sample. The energy resolved spectra were recorded with a spectrometer and a Si-CCD camera, whereas the time-correlated single photon counting measurements of single spectral lines were performed using a single-photon avalanche diode coupled to a secondary spectrometer output port, with a resolution of 50 ps full width at half maximum (FWHM). Tunable THz pulses in the spectral range of the QD inter-sublevel transition energies of the QDs were generated by the free-electron laser FELBE at Dresden-Rossendorf at a repetition rate of 13 MHz. The pulse trains of both lasers were synchronized with a phase-locked loop and a fast electro-optic modulator, and typically, the timing jitter of this scheme was 1–2 ps. The THz pulses were focused onto the substrate side of the QD sample by an off-axis parabolic mirror. This scheme enables probing the single-dot response to THz radiation in spite of the large diameter of the THz focus of approximately 320 μm (FWHM), since only the PL of a single QD is collected.

Figure 1(a) shows the spectrum of a typical single QD used in our measurements. We have identified the strongest line as the positive trion (X^+) using well-established methods,[16,17] the dominance of X^+ likely being due to unintentional carbon doping in the MBE growth chamber. We have not attempted to measure the THz absorption spectrum of

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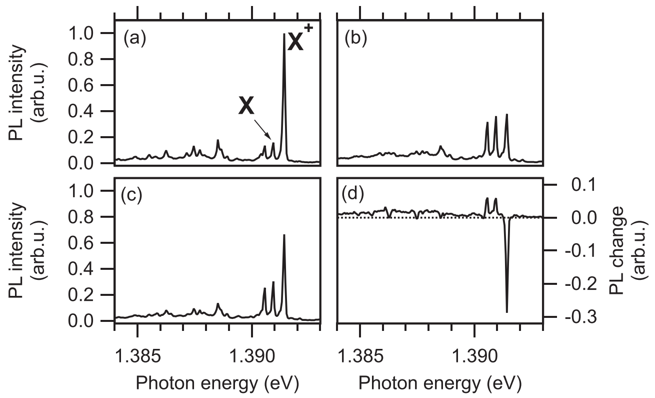

FIG. 1. (a) PL spectrum of a single QD under NIR excitation with $8.9\,\mathrm{MW/cm^2}$ at 1.54 eV, without THz excitation. (b) Same spectrum under additional THz excitation with $7.0\,\mathrm{kW/cm^2}$ at 19.2 meV, where the THz and NIR pulses arrive synchronously. (c) Same as (b), with the THz pulse arriving 13 ns before the NIR pulse. (d) The difference between (b) and (c). All spectra are normalized to panel (a).

our sample due to the low QD density. However, based on similar growth conditions as well as similar PL energies, we assume a similar behavior as described in the literature,[15] in particular, the s-p transition energy in the conduction band is expected to lie between 13 meV and 20 meV.

If the resonant pumping of the electron s-to-p transition is the dominant effect of THz irradiation, one would expect to see a decrease in the PL intensity of both the neutral exciton and the positive trion, since less electrons are available in the s-like state. However, in Figure 1(b), we observe a transfer from the dominant positive trion line towards the neutral exciton line (labeled X) when the QD is exposed to THz pulses. By comparing this spectrum with a temperature series without THz incidence, we can exclude sample heating by the THz beam as the cause, since the effect of increased temperature up to 30 K is the weakening of the X^+ line without a significant effect on the other lines (not shown). We also observe that the transfer is still present when the THz pulse is time-shifted up to 500 ps before the near-infrared (NIR) pulse and the onset of the PL transient, ruling out coherent phenomena. However, when the THz pulse is shifted 13 ns before the NIR pulse (Figure 1(c)), the population transfer is strongly reduced, but still present. We therefore conclude that this transfer is due to the trapped carriers freed by the THz pulses, which will have the tendency to neutralize the charge in the QD. Most likely, this effect has prevented the observation of interband spectral hole burning in QD ensembles in an earlier study.[9] Finally, we point out that this redistribution is not observed in every QD, which we attribute to variations in the local environments, such as thickness/composition fluctuations of the surrounding wetting layer and local electrostatic potentials.[18]

On the low-energy side of the spectrum in Figure 1(b), we observe several broader lines which have been previously attributed to multiexciton recombination, where an s-shell exciton recombination is perturbed by the excitons in higher states.[16] We observe that these broad lines (which start to merge into a continuum) gain intensity when the THz pulse temporally coincides with the PL emission (Figure 1(d)), and the increase is stronger for higher THz intensities (not shown). We attribute this to the increased population in the p-shell caused by THz pumping. When changing the THz pulse from the shifted to the synchronous position (thus excluding the effect of sample heating), we also observe an increase in the p-shell PL located 25–30 meV above the X^+ line (not shown), confirming a transfer from the s- to the p-shell.

Figure 2(a) shows the transient of the dominant X^+ PL line of a single QD for NIR excitation above the GaAs bandgap as well as THz excitation. For all measurements, we have used sufficiently low NIR intensities as to avoid significant population in the biexciton states. We have chosen to investigate the X^+ line in order to exclude spin-flip and fine-structure splitting effects. $t = 1.05\,\mathrm{ns}$ marks the arrival of the THz pulse, which leads to a significant quenching of the photoluminescence, followed by a strong recovery, which unexpectedly results in a net increase in the total number of emitted photons compared to the case without additional THz excitation. We use the phenomenological four-level decay model[12,19] shown in Figure 2(d) to fit our transients. For the promotion of electrons from the valence to the conduction band as well as for the excitation from $|s\rangle$ to $|p\rangle$, we assume an instantaneous population change, whereas for the relaxation from $|s\rangle$ to the crystal ground state $|0\rangle$, from $|\mathrm{CB}\rangle$ to $|s\rangle$ and from $|p\rangle$ to $|s\rangle$, we assume an exponential decay with time constants τ_1, τ_2, and τ_3, respectively. At the time of incidence of the THz pulse, a fraction q of the population in $|s\rangle$ is transferred to $|p\rangle$. Hence, larger values for q lead to larger drops in the PL transient. In order to take into account the observed extra carriers, a term $n_s(t_d) \times r$ is added to the

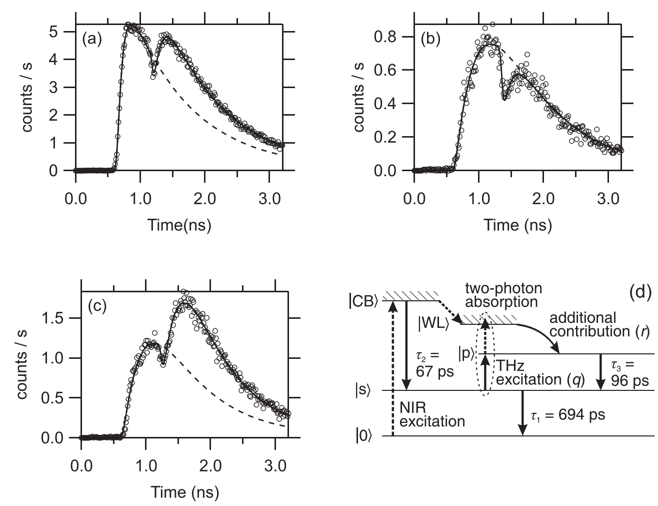

FIG. 2. Excitonic decay transient (single PL line) with different NIR excitation and additional THz excitation. (a) Above-band gap excitation with $169\,\mathrm{kW/cm^2}$ at 1.54 eV. (b) Quasi-resonant excitation with $23\,\mathrm{MW/cm^2}$ at 1.43 eV. (c) Wetting layer excitation with $2.5\,\mathrm{MW/cm^2}$ at 1.46 eV. The THz peak intensity was $167\,\mathrm{kW/cm^2}$ at 13.3 meV for all three transients. The solid line is a fit function, and the broken line is a reference function calculated from the fit parameters, where the effect of the THz pulse is omitted. (d) The phenomenological four-level model used to fit the PL transients involves transitions between the conduction band $|\mathrm{CB}\rangle$, the wetting layer $|\mathrm{WL}\rangle$, the s- and p-like QD states $|s\rangle$ and $|p\rangle$, and the crystal ground state $|0\rangle$. The fraction of electrons excited from $|s\rangle$ to $|p\rangle$ is expressed by the parameter q, and the ratio of additional electrons relative to the ones promoted from $|s\rangle$ to $|p\rangle$ is expressed by r. The lifetimes given are for the case of above-band gap excitation (Figure 2(a)), the initial population of the $|s\rangle$ level is slower for excitation in the wetting layer ($\tau_2 = 378\,\mathrm{ps}$) and quasi-resonant excitation ($\tau_2 = 479\,\mathrm{ps}$).

population of $|p\rangle$, where $n_s(t_d)$ is the population in $|s\rangle$ just before the THz pulse arrives. Hence, r, which we will call *carrier surplus*, is the relative increase in the level population as well as in the PL integral after the THz pulse. The population of $|s\rangle$ is assumed to be proportional to the PL signal, and the time resolution of our instrumentation is modeled via convolution with the measured detector response. The fitted curve produced by this model is the solid line in Figure 2(a), and the dashed curve uses the same parameters except for $q = r = 0$ and reproduces the behavior of the QD without THz excitation. The decay parameters extracted by the fit are given in Figure 2(d). The key findings from this fit are that $q = 48\,\%$ of the electrons are promoted from $|s\rangle$ to $|p\rangle$, there is a $r = 66\,\%$ increase in the $|p\rangle$-level, and the decay time for the relaxation back to $|s\rangle$ is $\tau_3 = 96 \pm 6$ ps (where q and r are dependent on the THz intensity). This time constant has about the same value as observed previously for polaron relaxation at similar energies,[15] even though the present experiment refers to a different elementary excitation, i.e., the formation of positive trions.

To further investigate the origin of the additional photoluminescence, we have repeated the measurement under quasi-resonant excitation with 1.43 eV NIR pulses (Figure 2(b)), bypassing the GaAs barrier as well as the wetting layer and generating electron-hole pairs directly inside the QD via phonon mediated relaxation.[1,20,21] We note that the population time constant τ_2 is larger for the quasi-resonant case, as generally expected for QD relaxations involving a smaller energy difference. Furthermore, there is a considerable difference to the previous case after the incidence of the THz pulse; the PL drops and then recovers slightly, but in contrast to Figure 2(a), we observe a net loss ($r = -12\,\%$) of photons. The behavior in Figure 2(a) is similar to the one previously reported for QD ensembles.[11] The explanation given (transfer of trapped carriers in quantum dots) is inconsistent with our measurements using quasi-resonant excitation, however, since the overall increase of photoluminescence is absent in this case. We have observed this strong excitation energy dependence of the transient for several different QDs.

In order to understand the cause of the carrier surplus, we have performed the same measurement with 1.46 eV excitation, generating electron-hole pairs in the wetting layer (Figure 2(c)). In this case, the fit reveals a large increase in PL intensity by $r = 160\%$ after the arrival of the THz pulse. We conclude that the additional PL for NIR excitation in the barrier is the result of trapped charge carriers in the wetting layer, which are freed by the THz pulse and relax into the QD, contributing to the PL. This picture is consistent with our aforementioned conclusions from the QD spectrum. The large r parameter in Figure 2(c) (compared to above-band-gap excitation) is most likely due to the larger NIR intensity necessary to produce an adequate count rate, in combination with less efficient relaxation into the QD, leading to a larger accumulation of charge carriers in the wetting layer. Additionally, the quasi-resonant measurement reveals a loss mechanism due to the THz pulse, which we will discuss in more detail below.

Figure 3(a) shows the dependence of the trion line transient on the peak intensity of the THz pulse. We have used our model to extract the fit parameters shown in Figure 3(b).

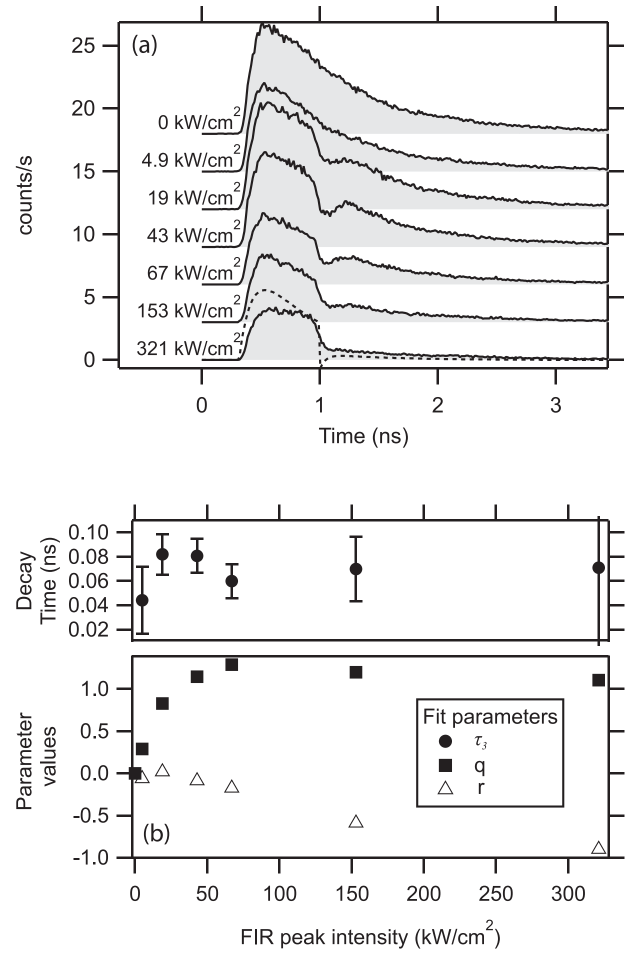

FIG. 3. Dependence of the PL transient on the THz peak intensity under quasi-resonant (1.43 eV) NIR excitation. (a) Transients vs THz peak intensity. The dashed line for 321 kW/cm^2 shows the behavior reconstructed from the fit parameters, removing the detector response. (b) Extracted fit parameters.

As the THz intensity is increased, we observe a fast increase and subsequent saturation of the fit parameter q towards $q \approx 1$, indicating that the entire population is transferred to the $|p\rangle$ level in our model. The corresponding threshold of approximately 50 kW/cm^2 is more than an order of magnitude smaller than previously reported for intersubband transitions in quantum wells.[19] We note that $q > 1$ should not occur according to our model, and ascribe this behavior to its simplistic nature (which is necessary to achieve an analytical solution) as well as to the noise in our data and to delayed THz pulses. The parameter r remains close to zero for low intensities and reaches -0.9 at 321 kW/cm^2, showing saturation as it approaches $r = -1$. Hence, we conclude that the THz pulse can deterministically suppress the positive trion PL from the QD. This is illustrated by the dotted line in 3a, which is the reconstructed transient at 321 kW/cm^2 from the fit parameters, without the detector response. It is obvious that the trion PL has been suppressed completely (within our measurement uncertainty), the slow decay seen after the THz pulse in the measured transient merely being a detector artifact. The negative overshoot in the reconstructed curves is an artifact due to the aforementioned overestimation of q. We point out that our method is different from absorption bleaching experiments, as bleaching merely implies that an equilibrium of unknown level populations has been achieved for the duration of the THz pulse. By contrast, our technique allows us to observe that the trion PL has been fully and permanently depleted.

We have repeated the measurement of Figure 3 for another QD and have obtained similar results (not shown)—the main difference being a slightly larger threshold intensity for the population transfer.

Concerning the nature of the transitions involved, we note that the relaxation time constant τ_3 extracted from the transients is consistent with the ones previously reported for InAs/GaAs QD inter-sublevel transitions, as is the THz energy.[15,22] However, contrary to our expectations, the exact THz energy turns out not to be crucial for the observation. We have scanned the THz energy from 13 to 20 meV in $\sim$0.3 meV steps and have found no signs of resonance in the transients or in the *p*-state PL.

We interpret the negative carrier surplus under non-resonant excitation as follows. We rule out tunneling from the excited states into the neighboring QDs due to the large QD separation. Similarly, tunneling from the excited states into the wetting layer or conduction band is extremely unlikely due to the relatively weak THz electric fields. It has been demonstrated that externally induced spin-flip processes can have a profound effect on the excitonic population;[2,3] however, spin-flip processes should have no impact on the trion PL. Based on the THz energy, we speculate that the observed loss is caused by electrons being promoted from $|s\rangle$ to the wetting layer via two-photon or sequential linear absorption, as sketched in Figure 2(d).

In conclusion, we have presented a detailed study on the response of single QD photoluminescence to strong terahertz pulses. Our results reveal the transfer of electrons to and recovery from the p-like QD state. In contrast to previous studies on QD ensembles, we find that the additional photoluminescence induced by a terahertz pulse is caused by charge carriers freed from the wetting layer. These findings are supported by changes in the relative intensity of different excitonic species in spectral measurements. Furthermore, we have demonstrated near-total depletion of the single QD trion PL by a strong THz pulse, which may prove useful for ultrafast gated luminescence experiments[23] without the need for external modulation.

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