

Induced absorption dynamics in quantum dot based waveguide electroabsorbers

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Two-color pump-probe measurements are used to study the carrier dynamics of InAs/GaAs quantum dots in a waveguide structure under reverse bias conditions. For the case of initially populating the ground state (GS), we find relaxation dynamics that include both absorptive and bleaching components in the excited state (ES) wavelength range. We reproduce the main features of this induced absorption dynamics using a simple model with an additional term for induced absorption at the ES due to carriers injected at the GS. The induced absorption dynamics includes multiple recovery timescales which can be attributed to phonon-assisted processes of GS/ES interaction.

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The carrier dynamics of semiconductor quantum dot (QD) absorbers have recently been investigated using time-resolved pump-probe spectroscopy. In particular, these techniques were applied to QD structures to explain the nature of tunneling processes at high reverse bias voltages¹ and to demonstrate the electroabsorption properties of a bilayer QD waveguide.² More recently, single-color measurements revealed fundamental differences in intradot relaxation dynamics which were attributed to phonon-assisted or Auger processes being dominant for initially populated ground states (GSs) or excited states (ESs), respectively,³ and explained multiple recovery stages by nonlinear energy exchange between the GS and ES.⁴

Two-color experiments showing pump induced absorption in the probe differential transmission near the first excited-state transition have previously been reported for InGaAs QDs at 1300 nm and revealed the role of Coulomb renormalization of the first ES in the presence of one electron-hole pair in the GS.⁵ Similar experiments with InAs/GaAs QDs in the range of 970 nm attributed induced absorption to the formation of a nondegenerate biexciton state.⁶ More recently, experiments at 1100 nm reported on anomalous induced absorption at the InAs QD ES at high current densities and time-dependent energy level shifts due to Coulomb carrier-carrier interactions.⁷

In this letter, the nonlinear recovery of QD based reversed-biased waveguide absorbers is analyzed using a two-color heterodyne pump-probe technique. Either the dots' GS or ES is initially populated using a strong pump pulse. The probe pulse, usually at a different wavelength, samples the recovery dynamics of initially empty states. In the ES pump GS probe case, the dynamics consist of an initial absorption bleaching followed by absorption recovery dynamics similar to the single color case and related to the electron

dynamics. The recovery dynamics in GS pump ES probe experiments is more complicated and includes both absorptive and bleaching features.

In our experiments, we study intradot relaxation processes as a function of reverse bias using time-resolved spectroscopy. The QD waveguide absorber was 1 mm long, had 4 μm width ridges together with tilted, antireflection coated facets. It was fabricated from material that included six stacks of InAs/GaAs QDs in a dots-in-a-well structure, grown by Zia Inc. (see Ref. 8 for further details of the material and experimental technique). In forward bias, the ground and ESs appear clearly in the amplified spontaneous emission around 1320 and 1250 nm, respectively. The pump-probe differential transmission was measured using a heterodyne detection technique. Pulses of about 600 fs width at either 1320 or 1250 nm were obtained from a titanium-sapphire laser pumped, optical parametric oscillator and split into the following three beams: reference, pump, and probe. After propagation through the room temperature stabilized waveguide absorber with suitable delays, the probe and reference beams were overlapped on a detector. The amplitude of the difference frequency was detected using a high frequency lock-in amplifier. This signal is proportional to the transmission T of a probe pulse. The resulting data are normalized to the transmission at zero pump T_0 .

Figure 1 displays the absorption spectra under reversed biased conditions, measured using a continuous wave tunable source. The inhomogeneously broadened GS transition occurs around 1320 nm at 1 V while the ES transition occurs around 1250 nm. As the reverse voltage is increased, the peak is shifted to longer wavelengths due to the quantum confined Stark effect while the absorption reduces due to increased spatial separation of electron and hole wave functions with increasing electric field (see Ref. 9 for further explanation of a similar structure).

Figure 2 displays the normalized GS and ES relaxations for single color and two-color measurements under the same

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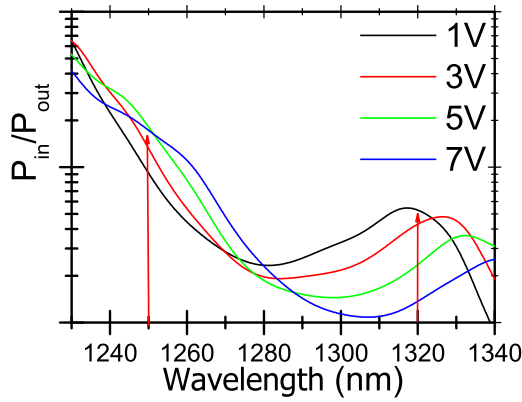


FIG. 1. (Color online) Absorption spectra at various reverse bias voltages. Pump and probe central wavelengths are denoted by arrows.

conditions. The single-color timetraces [Fig. 2(a), GS] and [Fig. 2(b), ES] are very similar, both exhibiting pump induced bleaching of the absorption and subsequent recovery. In addition, the recovery of the ES absorption occurs over a faster timescale compared to that of the GS. This difference originates from a change in the dominant intradot relaxation process, i.e., phonon-assisted or Auger processes being dominant for initially populated GS or ES states, respectively.³ The carrier relaxation of the two-color ES pump/GS probe measurement [Fig. 2(c)] also displays pump induced bleaching and is very similar to the single color measurements. The two-color GS pump/ES probe measurement [Fig. 2(d)] features pump induced absorption in contrast to the pump induced bleaching seen in the other three cases.

The dynamics of this pump induced absorption can be classified into two groups depending on the reverse bias. The pump induced absorption dynamics in the low voltage (1–6 V) group has two distinct stages of recovery, a fast stage (≈ 1 ps) and a slow stage (10–100 ps), similar to the pump induced transmission previously reported.³ For higher (7–10 V) reverse voltages both induced absorption and absorption bleaching occur over similar timescales. This complicated behavior is difficult to analyze due to the onset of tunneling processes at these voltage levels.¹

In the low voltage range, the slow recovery times τ_{slow} are extracted from the experimental data and shown, as a function of voltage, on the inset of Fig. 2(d). The slow time

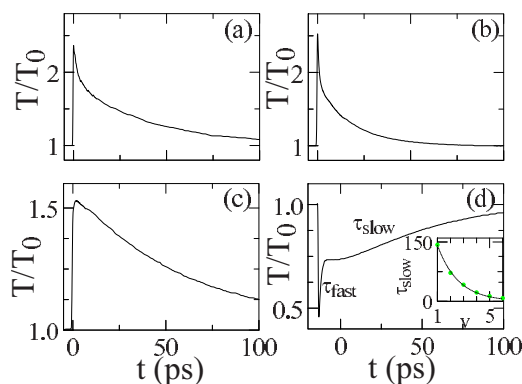


FIG. 2. (Color online) Typical pump probe dynamics for various pump/probe combinations at 1 V: (a) GS pump/GS probe; (b) ES pump/ES probe; (c) ES pump/GS probe; (d) GS pump/ES probe. (Inset) Exponential behavior of slow timescale in GS pump/ES probe case.

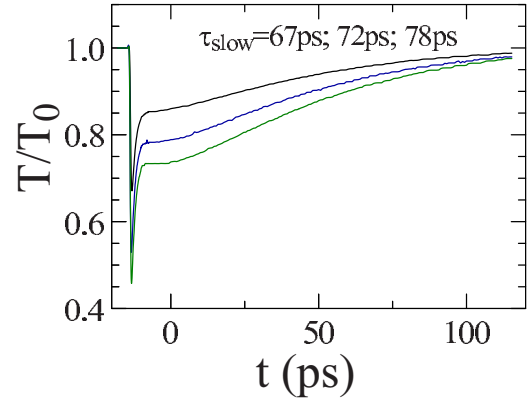


FIG. 3. (Color online) GS pump/ES probe traces at -2 V for pump powers of $100 \mu\text{W}$ (black, upper), $190 \mu\text{W}$ (blue, middle), and $260 \mu\text{W}$ (green, lower). The slow timescale shows a very weak dependence on pump power and can be neglected.

can be described by $\tau_{\text{slow}} = \tau_{s0} \exp(-V/V_0)$, where $\tau_{s0} \approx 250$ ps and $V_0 \approx -1.6$ V. This exponential dependence on voltage is similar to that found in single-color experiments in Figs. 1(a) and 1(b) and is a result of thermionic depopulation of the ES.³ A very weak dependence of the slow recovery on the GS-pump power can be seen in Fig. 3, this dependence is neglected in the analysis.

To reproduce the experimental behavior, we follow the model of QD absorbers with phonon-assisted GS–ES interactions described in Refs. 3 and 4 which reads,

$$\partial_t \rho_g = -\tau^{-1} \rho_g + 2F(\rho_g, \rho_e), \quad (1)$$

$$\partial_t \rho_e = -\tau_w^{-1} \rho_e - F(\rho_g, \rho_e), \quad (2)$$

$$F(\rho_g, \rho_e) = \tau_{\text{cap}}^{-1} \rho_e (1 - \rho_g) - \tau_{\text{esc}}^{-1} \rho_g (1 - \rho_e). \quad (3)$$

The factor 2 in Eq. (1) accounts for the degeneracy of the QD's ES energy level, $\tau \approx 1$ ns is the carrier recombination time in the dots. The function $F(\rho_g, \rho_e)$ is associated with the carrier exchange rate between the GS and ES in the dot, $1 - \rho_{g,e}$ is the Pauli blocking factor and τ_{cap}^{-1} , τ_{esc}^{-1} are the carrier capture and escape rates, respectively. The term $\tau_{\text{cap}}^{-1} \rho_e (1 - \rho_g)$ describes phonon-assisted recapturing by the GS and is linearly proportional to the population of the ES. The parameter τ_w^{-1} is the carrier escape rate from the ES to the wetting layer and depends on the reverse bias.

As mentioned earlier, nonresonant pump induced absorptive features in QDs have been previously attributed to Coulomb renormalization effects at room temperature^{5,7} and nondegenerate biexciton formation at low temperature.⁶ In addition, below band gap absorption measurements have suggested that absorptive bound to continuum transitions play an important role in QD laser structures.¹⁰ In order to reproduce the experimental timescales with our model, we note that each of these effects can be simply represented as an increased ES probe absorption which is proportional to the occupational probability $\rho_g(t)$ of the pumped GS.

Thus, we assume that the transmission of a probe pulse at the ES wavelength can be described by two terms and formulate the dynamical change in the overall transmission as $\Delta T(t) = -W_g(V) \rho_g(t) + W_e(V) \rho_e(t)$, where $W_{g,e}(V)$ are weight functions related to the change in transmission at the GS and ES with reverse bias voltage. Here, the product

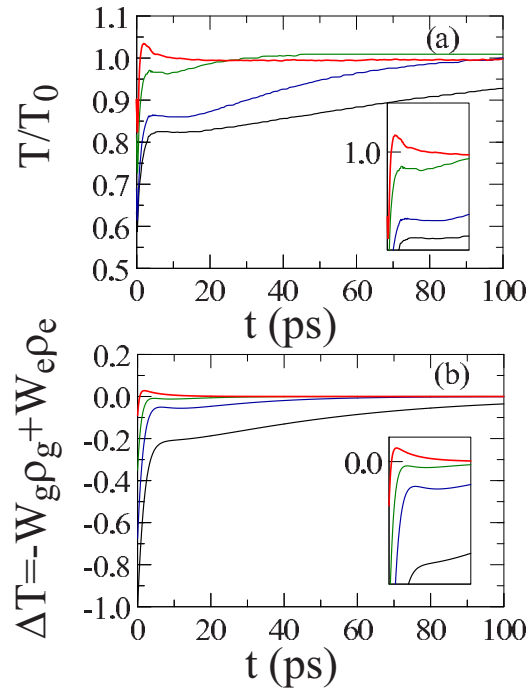


FIG. 4. (Color online) GS pump/ES probe experimental traces (top) and theory (bottom) for a variety of reverse voltages (from top to bottom, 7V, 5V, 3V, 1V). In the experimental case, T_0 changes with voltage according to Fig. 1 and the ratio T/T_0 emphasizes the magnitude of the induced absorption effect.

$W_e(V)\rho_e(t)$ describes the usual transmission due to carriers at the ES while $W_g(V)\rho_g(t)$ is the amount of ES absorption induced by the GS.

In the simulations, we assume $\rho_e(0)=0$, $\rho_g(0)=\rho_0(V) \leq 1$ and use $W_{g,e}(V)$ as fitting parameters. The resulting simulations are presented in Fig. 4 with the corresponding experimental timetraces for different reverse voltages. The simulations clearly reproduce the fast and slow recovery stages that possess similar timescales to the experiment. The values for $\rho_0(V)$ and $W_{g,e}(V)$ are given on Table I with the other parameters remaining the same as previously used.^{3,4} These values can be related to the decrease in absorption in the GS and the increase in absorption in the ES with voltage, experimentally shown on Fig. 1.

The effect of the ES population $\rho_e(t)$ in the transmission dynamics is twofold. First, it leads to a nearly flat recovery stage which connects the fast and slow recovery stages at low voltages, and is a result of compensation between in-

TABLE I. Parameters used in the simulation at various voltage levels.

V_{rb}	$\rho_0(V)$	$W_g(V)$	$W_e(V)$
1	1	1	2
3	0.9	0.75	3
5	0.5	0.5	4
7	0.6	0.15	4

duced absorption and bleaching terms. Second, it results in a net weak bleaching at high voltage. The exact mechanism for the appearance of instantaneous induced absorption remains unresolved and will require an extensive nonresonant pump probe study together with a more detailed theoretical description to elucidate further. However, as we demonstrate, the essential features of the dynamics can be captured using a remarkably simple treatment. From an applications point of view, the induced absorption may play a considerable role in the regime of dual modelocking,^{11,12} which is currently attracting a lot of attention due to the appealing technological possibilities of simultaneous lasing at both the GS and ES. In particular, control of the quantum-confined Stark effect in the absorption section may allow switching between the modelocked GS and ES emissions.¹³

In conclusion, we use a two-color heterodyne technique to measure the absorption dynamics in a InAs based QD structure and show that pumping the GS leads to induced absorption at the ES. We discuss the various possible mechanisms and find that a simple model accounting for absorptive and bleaching features of the GS and ES yields the recovery timescales in good agreement with the experiment.

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¹D. B. Malins, A. Gomez-Iglesias, S. J. White, W. Sibbett, A. Miller, and E. U. Rafailov, *Appl. Phys. Lett.* **89**, 171111 (2006).

²D. Malins, A. Gomez-Iglesias, P. Spencer, E. Clarke, R. Murray, and A. Miller, *Electron. Lett.* **43**, 686 (2007).

³T. Piwonski, J. Pulka, G. Madden, G. Huyet, J. Houlihan, E. A. Viktorov, T. Erneux, and P. Mandel, *Appl. Phys. Lett.* **94**, 123504 (2009).

⁴E. A. Viktorov, T. Erneux, P. Mandel, T. Piwonski, G. Madden, J. Pulka, G. Huyet, and J. Houlihan, *Appl. Phys. Lett.* **94**, 263502 (2009).

⁵F. Quochi, M. Dinu, L. N. Pfeiffer, K. W. West, C. Kerbage, R. S. Windeler, and B. J. Eggleton, *Phys. Rev. B* **67**, 235323 (2003).

⁶K. Kim, T. B. Norris, and U. Hohenester, *J. Appl. Phys.* **103**, 113702 (2008).

⁷R. P. Prasankumar, W. W. Chow, J. Urayama, R. S. Ataluri, R. V. Shenoi, S. Krishna, and A. J. Taylor, *Appl. Phys. Lett.* **96**, 031110 (2010).

⁸I. O'Driscoll, T. Piwonski, C.-F. Schleussner, J. Houlihan, G. Huyet, and R. J. Manning, *Appl. Phys. Lett.* **91**, 071111 (2007).

⁹G. Visimberga, G. Rainò, A. Salhi, V. Tasco, M. T. Todaro, L. Martiradonna, M. De Giorgi, A. Passaseo, R. Cingolani, and M. De Vittorio, *Appl. Phys. Lett.* **93**, 151112 (2008).

¹⁰P. Moreno, M. Richard, M. Rossetti, M. Portella-Oberli, L. H. Li, B. Deveaud-Plédran, and A. Fiore, *Nano Lett.* **8**, 881 (2008).

¹¹M. A. Cataluna, D. I. Nikitichev, S. Mikroulis, H. Simos, C. Simos, C. Mesaritis, D. Syvridis, I. Krestnikov, D. Livshits, and E. U. Rafailov, *Opt. Express* **18**, 12832 (2010).

¹²M. A. Cataluna, W. Sibbett, D. A. Livshits, J. Weimert, A. R. Kovsh, and E. U. Rafailov, *Appl. Phys. Lett.* **89**, 081124 (2006).

¹³S. Breuer, W. Elsasser, and M. Hopkinson, *Electron. Lett.* **46**, 161 (2010).