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T. Piwonski
Department of Physical Sciences, Cork Institute of Technology

Jaroslaw Pulka
Department of Physical Sciences, Cork Institute of Technology

Guillaume Huyet
Department of Physical Sciences, Cork Institute of Technology

Et. al.

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APPLIED PHYSICS LETTERS **100**, 241108 (2012)

Pump dependence of the dynamics of quantum dot based waveguide absorbers

Evgeny A. Viktorov,[1] Thomas Erneux,[1] Tomasz Piwonski,[2] Jaroslaw Pulka,[2] Guillaume Huyet,[2] and John Houlihan[3]
[1]*Universite Libre de Bruxelles, Optique Nonlineaire Theorique, Campus Plaine, Code Postal 231, 1050 Bruxelles, Belgium*
[2]*Tyndall National Institute, Cork, Ireland, and Department of Applied Physics, Cork Institute of Technology, Ireland*
[3]*Department of Computing, Maths and Physics, Waterford Institute of Technology, Waterford, Ireland*

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The nonlinear two stage recovery of quantum dot based reverse-biased waveguide absorbers is investigated experimentally and analytically as a function of the initial ground state occupation probability of the dot. The latter is controlled experimentally by the pump pulse power. The slow stage of the recovery is exponential and its basic timescale is independent of pump power. The fast stage of the recovery is a logistic function which we analyze in detail. The relative strength of slow to fast components is highlighted and the importance of higher order absorption processes at the highest pump level is demonstrated. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4729155]

The performance of high speed electro-optical devices is strongly influenced by the properties of their constituent materials. For many of these devices, a semiconductor saturable absorber is the most important element and the recovery time of the absorption usually determines the properties of the resulting ultrafast pulse train, e.g., in monolithic two-section semiconductor lasers and passively modelocked fiber lasers with semiconductor saturable absorber mirrors (SESAMs).[1,2] While the role of the saturable absorber in monolithic devices is relatively well understood, recent studies of SESAMs reveal a more complicated picture. It was shown that a two stage recovery of SESAMs is beneficial for a harmonic passively mode-locked soliton laser. The fast recovery stage influences the pulse width and the slow stage determines the start-up of modelocked operation.[3] Experimentally, the slow component was recently found to play a dominant role in laser soliton bunch formation.[4] The two stage recovery is an important property of quantum dot (QD) based semiconductor materials. Specifically, it consists of a fast decrease of the occupation probability of a dot at the ground state followed by a slow exponential decrease. SESAMs using QD absorbers show better design flexibility to control the saturation fluence and the relative modulation depth.[5] The role of intradot relaxations in the formation of a two stage recovery in QD absorbers has been experimentally clarified[6,7] and described using a rate equation approach.[8]

The dependence of the absorber timescales with respect to the initial QD population is an important issue for such devices. In forward bias, a dependence of these timescales on the initial population of the dots has been outlined both theoretically[9] and illustrated experimentally[10] where the gain recovery dynamics was faster after optical QD depletion by a prepump pulse. In this letter, we investigate the reversed-biased situation both theoretically, by analyzing a rate equation model, and experimentally, by using time-resolved pump probe spectroscopy. We also find a dramatic change of the unbiased absorber's recovery for higher pump pulse powers and relate this effect to higher order absorption processes.

The model for the QD absorber carrier dynamics has been examined analytically in Ref. 8 where the carrier dynamics are described by occupational probabilities ρ_g and ρ_e of the ground state (GS) and the first excited state (ES) of a dot, respectively. The model accounts for a phonon-assisted interaction between these states, limited by Pauli blocking factors $(1-\rho_{g,e})$.

The speed of the intradot interaction is determined by the ES to GS carrier capture rate τ_{cap}^{-1} and GS to ES carrier escape rate τ_{esc}^{-1}. The model assumes that reverse bias (RB) conditions preclude capture of carriers from the barrier or wetting layer and that escape from the ES to the wetting layer can be described by a linear term $\tau_w^{-1}\rho_e$, which strongly depends on the RB.

The parameters τ_{cap}, τ_{esc}, and τ_w determine the time-dependent recovery of the QD absorber, which consists of a fast stage or initial layer $(\Delta t \sim \tau_{cap})$ followed by a slow $(>\tau_w)$ almost exponential decay. In a pump probe comparison, the initial condition $\rho_g(0)=\rho_0 \leq 1$ depends on the pump pulse power. We also assume that there are no free carriers in the ES initially, $\rho_e(0)=0$. To consider the effect of initial condition ρ_0 (pump power in the experiment) on the two-stage recovery of the dot, it is useful to examine the fast stage (time scale τ_f) and the slow stage (time scale τ_s) separately.

As outlined in Ref. 8, the exponential form of the slow recovery stage can be written as $\rho_g(t) \sim \exp(-t/\tau_s)$ where $\tau_s \equiv \tau_w\left(1+\frac{\tau_{esc}}{2\tau_{cap}}\right)$. The time scale τ_s does not depend on initial condition ρ_0 and so it will remain constant under changes of the pump power.

The fast recovery time τ_f does not depend on τ_w, in the first approximation but depends on the intradot transition properties as well as the initial occupational probability ρ_0.[8] It can be written as

$$\rho_g(t) = \rho_0 - 2\frac{\rho_+(1-\exp(-t/\tau_f))}{1-\rho_+/\rho_-\exp(-t/\tau_f)} \tag{1}$$

and is a logistic function of t starting at $\rho = \rho_0$ and saturating at $\rho_0 - 2\rho_+ < \rho_0$. The fast time scale is defined by $\tau_f \equiv \tau_{cap}/\alpha$, where α and $\rho_\pm$ are given by

$$\alpha \equiv \sqrt{[1-\rho_0+\varepsilon(2+\rho_0)]^2 + 8(1-\varepsilon)\varepsilon\rho_0}, \tag{2}$$

$$\rho_\pm \equiv \frac{1}{4(1-\varepsilon)}[-(1-\rho_0+\varepsilon(2+\rho_0))\pm\alpha] \tag{3}$$

and

$$\varepsilon \equiv \tau_{cap}/\tau_{esc} \ll 1. \tag{4}$$

The fast recovery depend on the three parameters τ_{cap}, ε, and ρ_0 and the direct fitting of the nonlinear solution (1) with experimental data is delicate. To simplify our comparison, we consider two limiting cases of the parameter ρ_0. Our goal is to relate τ_f to the decay time obtained by exponential fitting of the data. Specifically, we examine the limits $\rho_0 \to 0$ and $\rho_0 \to 1$ corresponding to low pump and high pump powers, respectively.

In the low pump limit, we scale ρ_0 as $\rho_0 = \delta x$ where δ is defined as a small parameter $(x = \mathrm{O}(1))$ and note from Eq. (3) with $\epsilon = O(\delta)$ that ρ_+ is $O(\delta^2)$ small while ρ_- is $O(1)$ large. Assuming $t/\tau_f = O(1)$, Eq. (1) then simplifies as $\rho_g - \rho_0 \simeq 2\rho_+\exp(-t/\tau_f)$ where $\tau_f \simeq \tau_{cap}/(1+4\varepsilon-2\rho_0)$. We conclude that for low initial populations (or pump powers), the fast recovery time scale τ_f increases with the initial population (pump power).

In the high pump limit, we introduce $\rho_0 = 1 - \delta x$ where δ is defined as a small parameter $(x = O(1))$ and note from Eq. (3) that $\rho_+/\rho_- = -1 + O(\delta^{1/2})$. Assuming $t/\tau_f = O(1)$, Eq. (1) now simplifies as

$$\rho_g - 1 \simeq \sqrt{2\varepsilon}\tanh(-t/(2\tau_f)), \tag{5}$$

where $\tau_f \simeq \tau_{cap}/\sqrt{8\varepsilon}$. The solution (5) is independent of the initial condition ρ_0, in the first approximation.[11] The short and long time behaviors of Eq. (5) are $\rho_g - 1 = -\sqrt{\varepsilon/2}(1-\exp(-t/\tau_f))$ and $\rho_g - 1 = -\sqrt{2\varepsilon}(1-2\exp(-t/\tau_f))$, respectively, and so the time evolution of Eq. (5) for small and large times is determined by the same exponential function $\exp(-t/\tau_f)$. In summary, we conclude that the fast recovery time increases for low initial populations and will show much less population dependence at higher powers.

An important property of an absorber needed for its design is the ratio of the slow to fast recovery stages $\Delta R_{slow}/\Delta R_{fast}$, where ΔR_{slow} (or ΔR_{fast}) represents the change in GS occupancy that occurs over the slow (or fast) regime of the dynamics. From Ref. 8, we have $\Delta R_{slow}/\Delta R_{fast} = \frac{\rho_0}{2\rho_+} - 1$ and this ratio is independent of the RB as confirmed experimentally in Ref. 14. In the limit of small pump pulse power, ρ_0 is $O(\delta)$ small and ρ_+ is $O(\delta^2)$ small meaning that $\Delta R_{slow}/\Delta R_{fast} \gg 1$. In the limit of high pump pulse power $\rho_0 \to 1$, and $\Delta R_{slow}/\Delta R_{fast} = 1/\sqrt{2\tau_{cap}/\tau_{esc}} - 1$ which is less than unity if $\tau_{cap}/\tau_{esc} \approx 0.2$. As a result, higher pump powers lead to a reduction of the ratio of slow to fast components.

To summarize, the model predicts no power dependence for the slow stage of the recovery timescale, a complicated power dependence for the fast stage, and a reduction in the

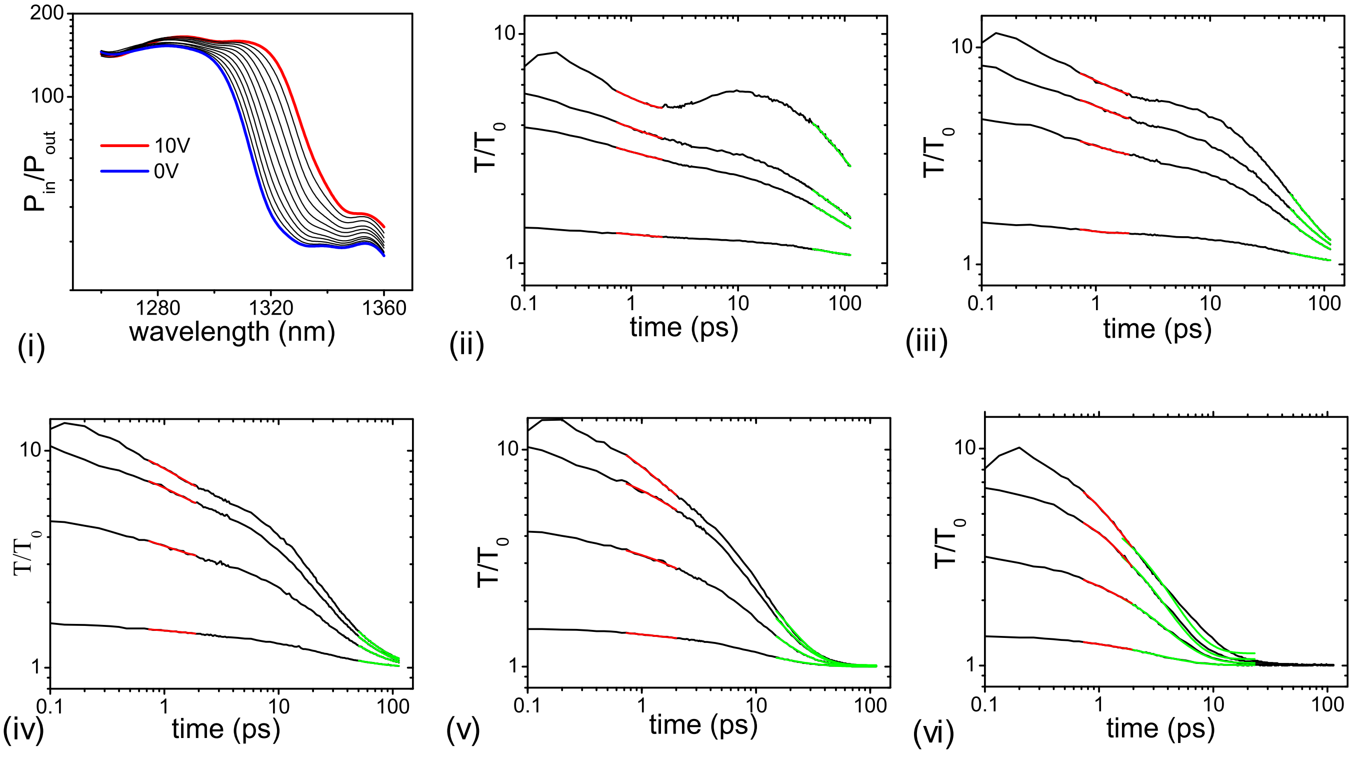

FIG. 1. Measured absorption spectra (i) and GS dynamics for 0 V, (ii) 3 V, (iii) 6 V, (iv) 9 V, (v) and 12 V (vi) at 1300 nm. Panels (ii)–(vi) contain data at pump levels (from bottom to top) of 0.05 mW, 0.2 mW, 1 mW, and 5 mW and the fast/slow time exponential fits are shown in red/green.

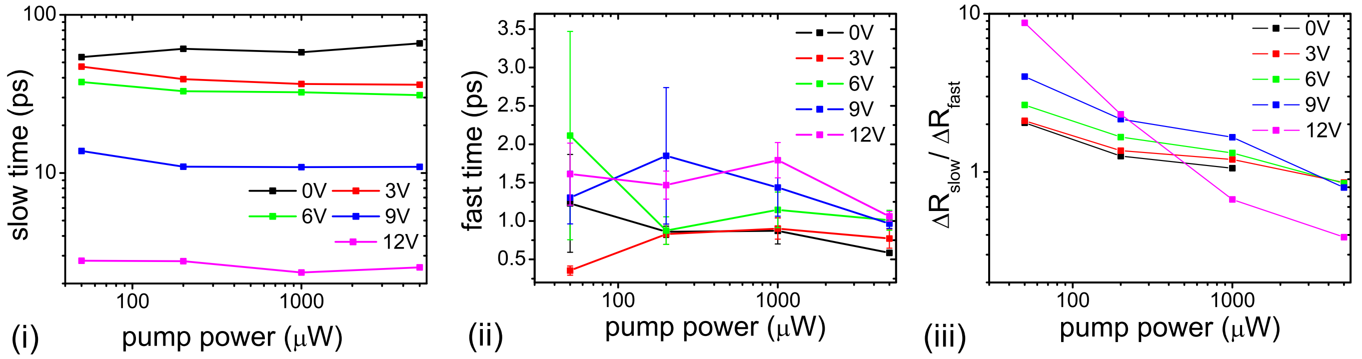

FIG. 2. Slow (i) and fast (ii) times extracted from single exponential fits as shown on Figure 1. Panel (iii) contains the modulation strength as a function of pump power and voltage.

ratio of slow to fast components with increasing pump power.

In order to test these predictions, heterodyne pump probe spectroscopy[15] was carried out on a QD sample as function of pump power and RB. The QD waveguide absorber was a 1 mm long, ridge waveguide device with tilted, anti-reflection coated facets. It was fabricated from material that included 10 stacks of InAs based quantum dots, grown by Innolume GmbH. The absorption spectrum of the structure is shown on Figure 1(i) as a function of RB and exhibits the usual Stark red-shift and changes in absorption due to varying spatial separation of electron and hole wavefunctions (see Ref. 16 for explanation of a similar structure).

To track the GS dynamics, 20 nm bandwidth pulses at 1300 nm of about 360 fs duration were obtained from an optical parametric oscillator and split into three beams: reference, pump, and probe. After propagation through the waveguide absorber with suitable delays, the frequency shifted probe and reference beams were overlapped on a detector and the amplitude of the difference frequency was detected using a high frequency lock-in amplifier. Panels (ii)–(vi) on Figure 1 display the differential transmission of the device at various pump powers and RB voltages together with corresponding fits to extract the fast and slow timescales. Single exponential fitting was performed between 0.8 ps and 2 ps in order to extract the fast time component of the signal and at the trailing end of the signal to extract the slow component. The particular interval for fitting the fast time was varied and the range chosen to avoid pump dependent features not captured in the theory but discussed later in the text.

The extracted slow and fast timescales as a function of pump power and reverse bias are shown in Figure 2. Physically, the slow time has been connected with thermalization from the dot into the WL/barrier and has been shown to depend exponentially on the RB voltage.[6,7] The variation of the slow time with pump level can be seen on panel (i) for each RB voltage. The long time appears insensitive to the pump level as predicted by the previous analysis. Different behavior at 0 V is not unexpected as carrier recapture from the WL/barrier may be possible in the absence of a RB voltage to sweep out photogenerated carriers. This process was neglected in the analysis and may modify the trend. Nonetheless, at higher RB voltages the long time behavior with pump is consistent with prediction.

Panel (ii) contains the extracted fast time as a function of pump level and reverse voltage. The data is somewhat scattered and a clear trend with pump power is difficult to discern. Fitting of the data over different time intervals results in a similar behavior. There does appear to be shortening of the timescale at the highest pump power level, however, we associate this regime with the onset of additional higher order absorption process that are not considered in the model. Higher order processes such as two-photon absorption have recently been reported as a significant feature of the dynamics of quantum dash based devices.[12,13]

The impact of such absorption processes can be seen in the anomalous increase in the transmission, clearly visible at 0 V around 10 ps at the highest pump level (Figure 1(ii)). This anomalous increase in the transmission is linked to carrier capture to the dot from the WL/barrier that was mentioned previously. This effect is particularly noticeable at high pump levels due to the additional carriers present in WL/barrier states due to higher order processes such as two-photon or bound to continuum absorption. A similar signature is present in the 3 V RB case for the highest pump, albeit at a much reduced level due to increased sweepout of carriers. Due to further increased sweepout at increased RB voltage this feature is no longer noticeable. An additional signature of the onset of nonlinear absorption is present for each RB voltage at the highest pump over 100-200 fs where the differential transmission displays a small anomalous increase. This increase is an indication of the presence of pump dependent absorption of the probe (e.g., two photon absorption) that reduces as the temporal overlap between pump and probe pulses decreases.

In order to evaluate the relative strength of the slow to fast components, the data was fitted to a biexponential function. As a consequence, the anomalous carrier capture feature that occurs at around 10 ps will be incorporated into the fitting and since it is so strong at the highest pump at 0 V, this trace is not used. We do not expect this feature to have a large effect on the ratio for the other traces. The results of this calculation are shown in Figure 2(iii), where the corresponding times are similar to those obtained using single exponential fitting. The trend to a lower ratio with increased pump is evident for each voltage level and is very similar for each voltage level with the exception of 12 V where the trend is stronger, possibly due to the dominance of tunnelling processes at such increased voltage levels.[6,7]

In summary, a theoretical and experimental analysis of the two-stage recovery of a reversed bias waveguide QD absorber as a function of the pump pulse power is presented. For the slower stage, we predict and confirm that its timescale is independent of pump power. For the faster stage, we predict a more complicated behavior. The experimental results are inconclusive for lower pump levels but reveal the importance of higher order absorption processes at the highest pump level. In addition, the power dependence of the relative strength of slow to fast components was predicted and confirmed. These results will provide important information for device designers incorporating such absorbing structures.

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