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Terahertz activated luminescence of trapped carriers in InGaAs/GaAs quantum dots

J. Bhattacharyya,[1,a)] M. Wagner,[1] M. Helm,[1] M. Hopkinson,[2] L. R. Wilson,[3] and H. Schneider[1]
[1]*Institute of Ion Beam Physics and Materials Research, FZD-Rossendorf, D-01314 Dresden, Germany*
[2]*EPSRC National Centre for III-V Technology, University of Sheffield, Sheffield S1 3JD, United Kingdom*
[3]*Department of Physics and Astronomy, University of Sheffield, Sheffield S3 7RH, United Kingdom*

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Optical properties and interdot transfer dynamics of trapped carriers in InGaAs quantum dots (QDs) are investigated. Time resolved photoluminescence (PL) was measured for time-delayed interband and intraband excitations. Terahertz activated luminescence (TAL) from trapped carriers having lifetimes of ~250 ns at 8 K, was observed. Spectral shift of the TAL with respect to the PL showed the trionic nature of the PL in the n-doped QDs. With increasing terahertz excitation intensity, the TAL increased and reached saturation. The activation energy associated with the trapped carrier decay was quite close to the intersublevel transition energy (~20 meV) indicating trapping in the QDs. © *2010 American Institute of Physics*. [doi:10.1063/1.3464163]

Study of carrier dynamics in semiconductor quantum dots (QDs) is essential for the understanding and improvement of their performances as optoelectronic devices. Time resolved photoluminescence (PL) measurements have been extensively used to investigate the lifetimes and relaxation mechanisms of carriers in the QDs.[1–4] In these experiments, the initial carrier distribution depends on the excitation energies. For nonresonant excitation, the electron-hole pairs are generated in the barrier or the wetting layer (WL) of the self-assembled QD systems. These carriers rapidly relax into the QDs by phonon scattering and then recombine giving PL signals corresponding to the QD transitions. During the relaxation process, the carriers can be trapped in QDs with no appropriate partners to recombine.[5] Trapping of carriers in the potential fluctuations in the WL has also been reported earlier.[6] These long-lived trapped carriers are found to have significant influence on the PL transients, resulting in modification of PL decay rates[5] and increased rise times.[7] The dependences of the PL transients on the temperature and excitation intensity[7–9] have been used to investigate the influence of trapped carriers which relax by interdot transfer via the WL (Ref. 10) or tunneling to the adjacent QDs.[7,11] Study of the optical properties of these long-lived carriers from the PL transients requires measurements with long time window and high sensitivity. We propose a technique to probe these carriers using a streak-camera based detection enabling simultaneous time- and energy-resolved measurements.

In this report we present our investigation of trapped carriers where we isolated the luminescence from the trapped carriers from the regular PL, enabling a direct study of their distribution and dynamics. We studied the trapped carriers in InGaAs/GaAs QD ensembles using terahertz induced PL measurements. Using temporally separated laser pulses for interband and intraband excitations, we observed terahertz activated luminescence (TAL) from the trapped carriers.

The self-assembled InGaAs QD samples were grown on (100) GaAs using molecular beam epitaxy, in the Stranski–Krastranow growth mode. The sample studied consisted of 80 layers of QDs separated by 50 nm wide GaAs barriers to ensure structural and electrical isolation between the layers. Using postgrowth annealing at 850 °C, the intersublevel transition energy in the conduction band was redshifted from 55 meV (for the as-grown sample) to 20 meV and the intradot relaxation time was increased from a few picoseconds to 60 ps.[12] From atomic force microscope measurements of an uncapped reference sample, the QD density was estimated to be about 4×10^{10} cm^{-2} with an average base diameter of 20 nm and height of about 5 nm, before capping. The QDs were n-doped by a Si-layer grown ~2 nm below the QD layers, providing an average doping of one electron per dot. Time-resolved PL quenching measurements were performed using a tunable mode-locked Ti:sapphire laser (TSL), emitting ~4 ps long pulses, for interband excitation. Emission from a free-electron laser (FEL) with a pulse width of ~10 ps was synchronized to the TSL and used to induce intradot transitions resulting in quenching of the PL and other effects as will be discussed shortly. The TSL spot size on the sample was ~80 μm in diameter (full width at half maximum) with an average power of 1 mW. Both the laser pulses were incident at an angle of ~15° on the sample. A pulse picker, triggered by the FEL pulses, was used to reduce the repetition rate of the TSL (78 MHz) to match with that of the FEL (13 MHz). By tuning the pulse picker, any of the six pulses from the TSL could be chosen which enabled us to introduce a relative time delay (Δt) up to 76.9 ns between the TSL and FEL pulses. Finer adjustments in the time delay were made using a mechanical delay unit. Time- and wavelength-resolved detection of the luminescence was done by a Hamamatsu streak camera coupled to a spectrometer. The sample was cooled in a He cryostat enabling measurements at different temperatures.

A schematic of the sequence of the laser pulses incident on the sample is shown in Fig. 1(a). Using the mechanical delay unit, the FEL pulse was set at 1.2 ns before the TSL pulse. Therefore the effective delay of the FEL pulse with respect to the previous TSL pulse, i.e., $\Delta t=75.2$ ns. Figure 1(b) shows the measured PL transient at 5 K corresponding to the ground state recombination in the QDs, for interband

[a)]Electronic mail: j.bhattacharyya@fzd.de.

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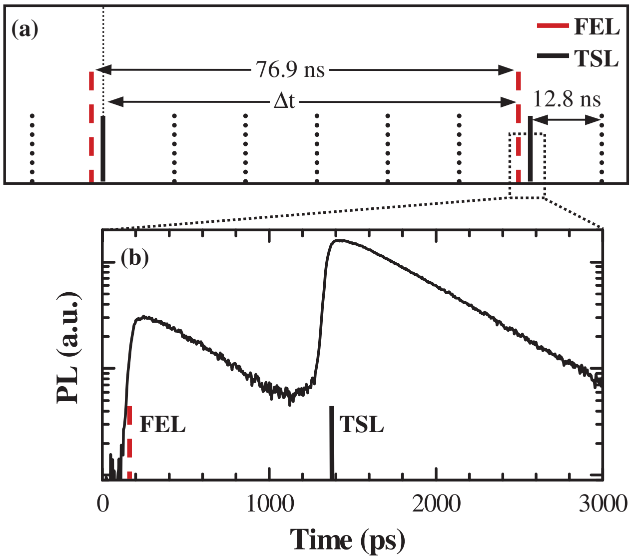

FIG. 1. (Color online) (a) Schematic showing the pulse sequence of the TSL (black lines) and the FEL (red dashed lines). The dotted black lines correspond to the TSL pulses suppressed by the pulse picker. (b) Luminescence transient at 5 K as measured by the streak camera for TSL and FEL excitation wavelengths of 802 nm at 1 mW and 61.4 μm at 74 mW, respectively, showing PL and TAL.

excitation in the GaAs barrier. The signal around 1400 ps is the PL emission excited by the TSL pulse. The FEL was tuned to the conduction band $s-p_x$ intersublevel transition energy of the QDs at 20 meV (61.4 μm). The feature at 200 ps corresponds to the TAL due to the FEL pulse. For measurements where larger Δt was required, different TSL pulses [shown by dotted lines in Fig. 1(a)] were chosen by the pulse picker. However, the streak camera being triggered at 78 MHz repetition rate of the TSL, the temporal position of the laser pulses in the streak camera image appeared unchanged. Measurements were done for different interband excitation energies. TAL was observed only for TSL excitation in the barrier or the WL. This suggested that the TAL originated either from carriers trapped in the WL interfaces or from the lone carriers trapped in the QDs. For interband excitation below the WL energy, the carriers were generated in pairs within the same dots. Therefore the existence of lone trapped carriers in the dots or interfaces was negligible, leading to the absence of TAL signal. The measured rise times of the PL and TAL were limited by the resolution of our experiment which was ~35 ps. Monoexponential fits to the luminescence decay (τ_{PL}) gave similar decay times of 500 ps for both signals which are associated with the ground state recombination rate.

Isolation of the trapped carrier luminescence from the PL enabled us to investigate the spectral response of the TAL. The PL spectrum in Fig. 2 shows two peaks which arise from the ground state and first excited state transitions in the QD ensemble. The spectrum for the TAL is similar to that of the PL except that it is blueshifted with respect to the PL emission and is weaker in intensity. We performed measurements for different intensities of the FEL and fitted the luminescence spectra with two Gaussians each (as shown in Fig. 2) to get an estimate of the emission energies and intensities. Figure 3(a) shows the variation in the peak energies of the PL and TAL with the FEL power at 10 K. The blueshift of the TAL decreases with increasing FEL power. We observed a maximum ground state energy shift of ~4 meV which corresponds well with the trion renormalization energy in InAs.[13,14] With an average doping of one electron per dot, the emitted PL is expected to be trionic. Since the FEL energy is resonant with the electron $s-p_x$ transition, the interdot transfer occurs primarily for electrons. The transferred electron recombines when it arrives in a QD having a hole which makes the TAL signal predominantly excitonic. With increasing FEL intensity, the number of detrapped carriers increases, making the transition more trionic. The TAL intensity increases and reaches saturation and this is accompanied by an equivalent decrease in the PL intensity such that the total remains nearly constant. This demonstrates the influence of long-lived carriers on the PL. The excited state tran-

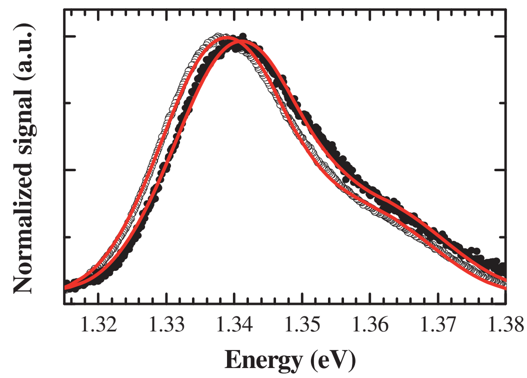

FIG. 2. (Color online) Normalized PL (open circles) and TAL (solid circles) spectra measured at 5 K for TSL and FEL excitation wavelengths of 802 nm (in the barrier) at 1 mW and 61.4 μm at 74 mW, respectively. The solid lines are the Gaussian fits.

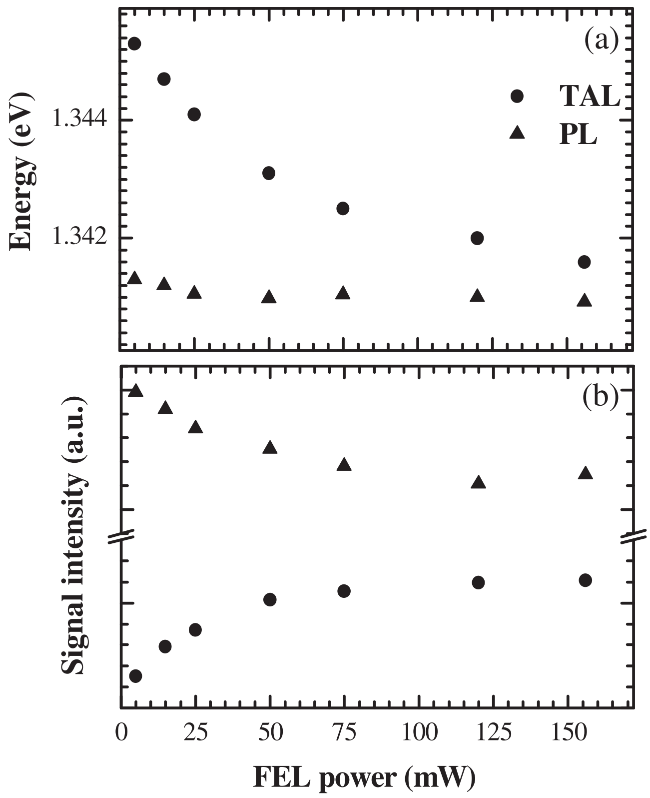

FIG. 3. The ground state transition (a) energy and (b) intensity of the QD ensemble at 10 K plotted as a function of FEL excitation power. The triangles and circles correspond to PL and TAL, respectively.

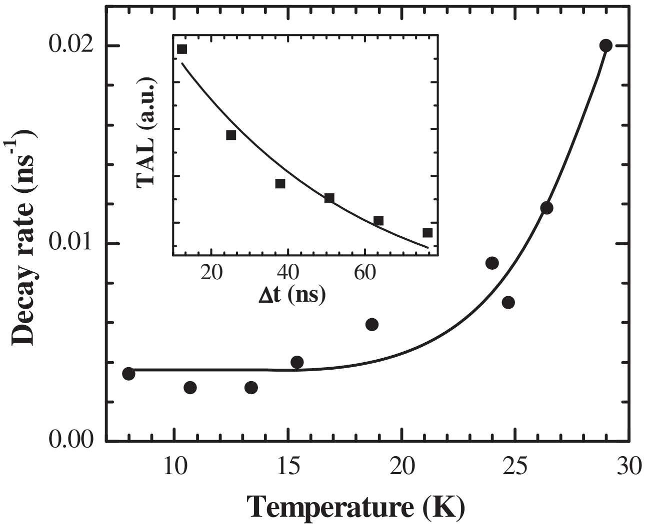

FIG. 4. Temperature dependence of the trapped carrier decay rates $(1/\tau_{tc})$. The line represents an Arrhenius fit giving an activation energy of 17 ± 3 meV. The inset shows a typical TAL decay with Δt at 29 K, fitted with a monoexponential function.

sition also showed similar behavior but with weaker magnitudes (not shown here).

As mentioned earlier, the carriers can be trapped in the WL interfaces or the QDs. To distinguish between these two possibilities we performed PL quenching measurements using FEL pulses detuned from the intersublevel transition energy. For FEL energy of 29 meV (42 μm) we found that the TAL intensity was drastically reduced. If the majority of the carriers were trapped in the WL interfaces, the FEL photons with higher energy would still be able to release the carriers into the continuum, which would relax into the QDs and emit TAL signals. However, for carriers trapped in the QDs, due to the delta-like density of states of the QDs, the FEL energy is required to match the intersublevel transition energy to excite the trapped carriers from s to p_x states in the QDs. The excited carriers have appreciable tunneling probability into adjacent QDs. When the carrier reaches a QD with another appropriate carrier, it can recombine radiatively. This emitted luminescence gives rise to the TAL signal.

As a further verification of our understanding we performed measurements to estimate the activation energy of the transfer mechanism of the trapped carriers. Apart from the TAL decay time (τ_{PL}) as mentioned earlier, another relevant lifetime is the trapped carrier lifetime (τ_{tc}) which results in a change in the TAL peak intensity as a function of the time delay between FEL and TSL pulses (Δt). Monoexponential fits to the TAL decay curves (see inset of Fig. 4) gave us an estimate of τ_{tc} at a given temperature. Measurements were done at different sample temperatures and the corresponding decay rates $(1/\tau_{tc})$ are plotted in Fig. 4. The temperature range was limited to 30 K, since the temperature dependence of the PL intensity (without FEL pulses) initially increased and reached saturation at around 30 K and later decreased. Similar behavior for PL have been reported for QDs (Ref. 15) where the initial increase was attributed to the trapped carrier luminescence, which fits to our description. Beyond 30 K, the trapped carriers were depleted and other thermal effects dominated. An Arrhenius fit of the decay rates gave an activation energy of 17 ± 3 meV. This value is quite close to the $s-p_x$ intersublevel transition energy, confirming that a majority of the carriers were trapped in the QDs. The background decay rate $1/\tau_{tc}\sim0.004$ ns^{-1} $(\tau_{tc}\sim250$ ns) is attributed to the weak interdot tunneling,[16] which is independent of the temperature.

In conclusion, we showed that time-resolved PL measurements with two-color pulsed laser excitations can be used to probe long lived carriers trapped in QD systems. The decrease in the TAL signal with increasing delay between the FEL and TSL pulses enabled us to directly measure the decay of the trapped carrier population. This gave an estimate of the trapped carrier lifetime without any influence of the radiative recombination lifetimes. Comparison of the spectral responses of the TAL and PL showed the excitonic and trionic nature of the transitions, respectively. From the temperature and FEL intensity dependent measurements we found that the carriers were trapped in the QDs.

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[1]M. Paillard, X. Marie, E. Vanelle, T. Amand, V. K. Kalevich, A. R. Kovsh, A. E. Zhukov, and V. M. Ustinov, Appl. Phys. Lett. **76**, 76 (2000).
[2]H. Yu, S. Lycett, C. Roberts, and R. Murray, Appl. Phys. Lett. **69**, 4087 (1996).
[3]S. Malik, E. C. Le Ru, D. Childs, and R. Murray, Phys. Rev. B **63**, 155313 (2001).
[4]Yu. I. Mazur, B. L. Liang, Zh. M. Wang, D. Guzun, G. J. Salamo, G. G. Tarasov, and Z. Ya. Zhuchenko, J. Appl. Phys. **100**, 054316 (2006).
[5]A. S. Shkolnik, L. Ya. Karachinsky, N. Yu. Gordeev, G. G. Zegrya, V. P. Evtikhiev, S. Pellegrini, and G. S. Buller, Appl. Phys. Lett. **86**, 211112 (2005).
[6]C. Lobo, R. Leon, S. Marcinkevicius, W. Yang, P. C. Sercel, X. Z. Liao, J. Zou, and D. J. H. Cockayne, Phys. Rev. B **60**, 16647 (1999).
[7]J. W. Tomm, T. Elsaesser, Y. I. Mazur, H. Kissel, G. G. Tarasov, Z. Y. Zhuchenko, and W. T. Masselink, Phys. Rev. B **67**, 045326 (2003).
[8]L. Brusaferri, S. Sanguinetti, E. Grilli, M. Guzzi, A. Bignazzi, F. Bogani, L. Carraresi, M. Colocci, A. Bosacchi, P. Frigeri, and S. Franchi, Appl. Phys. Lett. **69**, 3354 (1996).
[9]R. Chen, H. Y. Liu, and H. D. Sun, J. Appl. Phys. **107**, 013513 (2010).
[10]Z. Ma, K. Pierz, J. Hübner, and W. W. Rühle, Physica E (Amsterdam) **28**, 203 (2005).
[11]A. Tackeuchi, Y. Nakata, s. Muto, Y. Sugiyama, T. Usuki, Y. Nishikawa, N. Yokoyama, and O. Wada, Jpn. J. Appl. Phys., Part 2 **34**, L1439 (1995).
[12]E. A. Zibik, T. Grange, B. A. Carpenter, N. E. Porter, R. Ferreira, G. Bastard, D. Stehr, S. Winnerl, M. Helm, H. Y. Liu, M. S. Skolnick, and L. R. Wilson, Nature Mater. **8**, 803 (2009).
[13]A. D. Ashmore, J. J. Finley, R. Oulton, P. W. Fry, A. Lemaître, D. J. Mowbray, M. S. Skolnick, M. Hopkinson, P. D. Buckle, and P. A. Maksym, Physica E (Amsterdam) **13**, 127 (2002).
[14]P. A. Dalgarno, J. M. Smith, J. McFarlane, B. D. Gerardot, K. Karrai, A. Badolato, P. M. Petroff, and R. J. Warburton, Phys. Rev. B **77**, 245311 (2008).
[15]W. H. Jiang, X. L. Ye, B. Xu, H. Z. Xu, D. Ding, J. B. Liang, and Z. G. Wang, J. Appl. Phys. **88**, 2529 (2000).
[16]J. Rihani, V. Sallet, N. Yahyaoui, J. C. Harmand, M. Oueslati, and R. Chtourou, J. Lumin. **129**, 251 (2009).