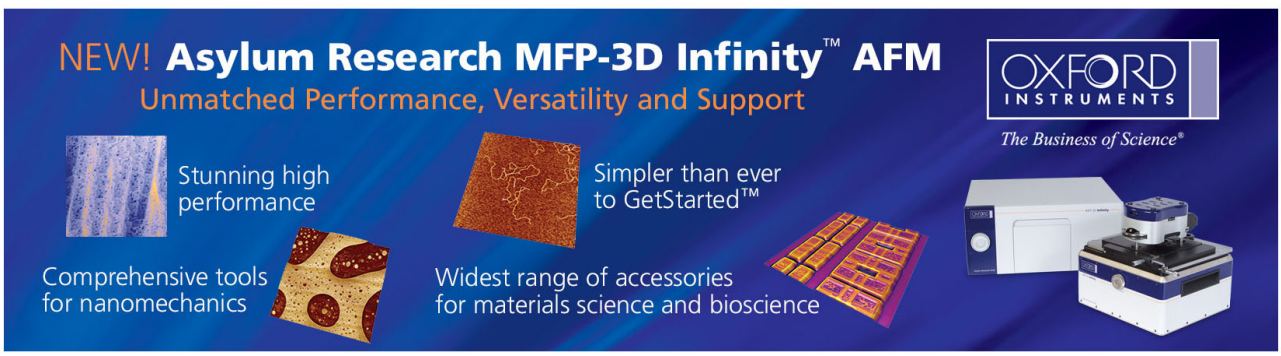
AIP Applied Physics Letters

Temperature dependence of time-resolved photoluminescence spectroscopy in InAs/GaAs quantum ring

C. H. Lin, H. S. Lin, C. C. Huang, S. K. Su, S. D. Lin, K. W. Sun, C. P. Lee, Y. K. Liu, M. D. Yang, and J. L. Shen

Citation: Applied Physics Letters **94**, 183101 (2009); doi: 10.1063/1.3130741
View online: http://dx.doi.org/10.1063/1.3130741
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/94/18?ver=pdfcov
Published by the AIP Publishing



Temperature dependence of time-resolved photoluminescence spectroscopy in InAs/GaAs quantum ring

C. H. Lin,[1,2] H. S. Lin,[2] C. C. Huang,[1] S. K. Su,[2] S. D. Lin,[2] K. W. Sun,[1,a)] C. P. Lee,[2] Y. K. Liu,[3] M. D. Yang,[3] and J. L. Shen[3]
[1]*Department of Applied Chemistry, National Chiao Tung University, Hsinchu 30010, Taiwan*
[2]*Department of Electronics Engineering and Institute of Electronics Engineering, National Chiao Tung University, Hsinchu 30010, Taiwan*
[3]*Department of Physics, Chung Yuan Christian University, Chung-Li 32023, Taiwan*

(Received 4 December 2008; accepted 13 April 2009; published online 4 May 2009)

We present detailed experimental results of the temperature dependence of continuous wave and time-resolved photoluminescence (PL) spectroscopy in self-assembled InAs/GaAs quantum dot and quantum ring nanostructures. A dramatic increase in PL decay time of the excited and ground states is observed in InAs quantum rings at high temperature. We speculate that the longer PL lifetime in quantum rings is due to the interplay among the dark states, ground states, and the reduced wave function overlapping between electrons and holes. A rate equation model is proposed to interpret the observed temperature dependence of the ground state exciton lifetime. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3130741]

Self-assembled semiconductor nanostructures with a size and shape that can be tailored and controlled to tune their electrical and optical properties have attracted substantial research attention. In particular, InAs on GaAs (001) self-assembled quantum structures is one of the most studied systems. For example, the morphological changes in quantum dot (QD) grown by molecular beam epitaxy (MBE) due to a thin GaAs cap have been reported.[1–7] Quantum rings (QRs) can be obtained by covering a layer of QDs with a thin cap, followed by subsequent annealing. Due to its unique rotational symmetry, the nanoring structure exhibits many interesting properties such as the Aharonov–Bohm effect,[8] large and negative excitonic permanent dipole moments,[9] and high oscillator strength of the ground state transition.[10] Ring-shaped nanostructures have been extensively studied experimentally[11–16] and theoretically[17–21] since their discovery. Lorke *et al.*[12] observed a ground state transition as an optical (noncontact) approach from angular momentum $\ell=0$ to $\ell=1$ when a magnetic field is applied perpendicular to the plane of the rings. The electronic structure in the QR complexes was analyzed using the microphotoluminescence (micro-PL) technique.[13] The interplay between the exciton radiative recombination and the electronic carrier tunneling in the presence of a stationary electric field was investigated and reported by Alén *et al.*[14] Recently, a shape-dependent electronic structure and exciton dynamics were reported by Gomis *et al.*[11] in quantum structures with different geometries such as a dot, dash, and camel hump. In their studies, an increase in radiative lifetime with temperature was observed and attributed to the thermalization between ground state and the first excited dark state. A three-dimensional confinement was also found to be very efficient in producing a strong emission band, and the radiation lifetime measured at a low temperature is below 1 ns in all cases. More recently, a PL decay time of less than 1 ns was also reported by Sanguinetti *et al.*[16] in concentric QRs. Both vertical and lateral confinements play an important role in the room temperature performance of optoelectronic devices based on these nanostructures. The understanding of carrier dynamics in these nanostructures is therefore of extreme relevance.

In this paper, we report on the cw and time-resolved PL spectroscopy of the excited and ground states of self-assembled InAs nanostructures with QD and QR shapes. We observe the strong dependence of PL decay time on the temperature in both the excited and ground states of QRs. A rate equation model is proposed and is found to agree well with the PL dynamics observed.

The GaAs QD samples studied in this work were grown on GaAs (001) substrates by MBE. The QDs were formed by depositing 2.6 ML of InAs with a growth rate of 0.056 μm/h at a growth temperature of 520 °C under As_2 atmosphere. QDs have an average base diameter of about 20 nm and a height of 2 nm. For the preparation of QR samples, after dot growth was completed, a thin partially capping layer of 2 nm was deposited on the dots with a 1 μm/h growth rate at 520 °C. Followed by an annealing process under As_2 flux from 5 to 30 s at the same temperature, the QD structures can be transformed into QR structures after the annealing processes. Figures 1(a) and 1(b) show the atomic force microscopy (AFM) images of samples with nanostructures of QDs and QRs. The QD and QR samples have areal densities of $\sim 3.4\times10^{10}$ and 2×10^{10} cm^{-2}, respectively. The final QR shape has a base width of ~60 nm, a height of ~1 nm, and an inner diameter of 30 nm. Figure 1(c) shows the cross-sectional transmission electron microscopy (XTEM) image of the QR sample. The QR appears as two dark-gray lobes (InAs rich) corresponding to a cut through the middle of the ring. The sizes determined by XTEM matched AFM results.

We performed time-resolved measurements for the above samples under nonresonant excitation in the GaAs barrier. The sample was placed in a closed-cycled helium dewar. A diode-pumped Ti:sapphire laser was used to excite steady-state PL. The signal was dispersed by a 0.18 m double spectrometer and detected by a TE-cooled InGaAs photodetector.

a)Author to whom correspondence should be addressed. Electronic mail: kwsun@mail.nctu.edu.tw.

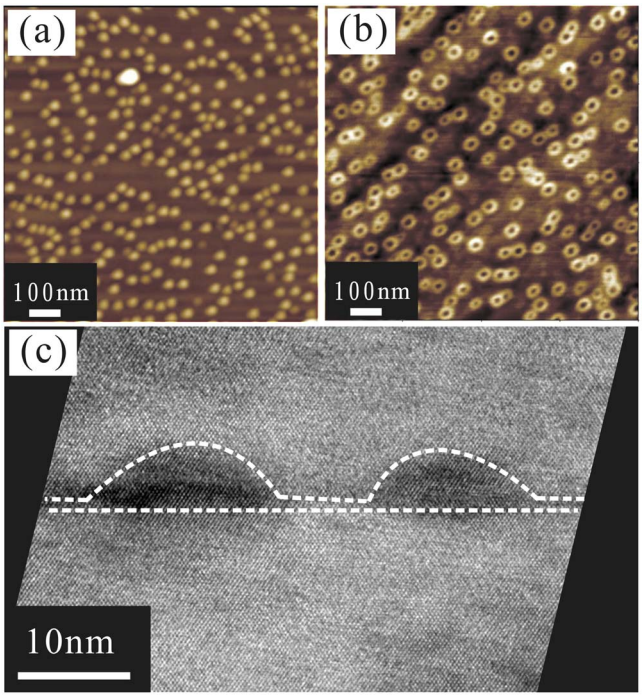


FIG. 1. (Color online) AFM images of (a) QD, (b) QR, and (c) XTEM image of QR.

For the time-resolved measurements, a pulsed diode laser was used as the excitation source at a wavelength of 635 nm. The pulse duration was 50 ps with a repetition rate of 5 MHz and an excitation density in the range of $1-100$ W/cm^2. The time-resolved PL signal was analyzed with a 0.55 m spectrometer and detected by a microchannel photomultiplier with a time correlated single photon counting setup. The overall spectral resolution and system response were 0.1 meV and 300 ps.

The ground state energies of the QD and QR are 1.21 and 1.25 eV at 15 K, respectively, as shown in Figs. 2(a) and 2(c). The barrier and wetting layer emission are observed clearly at around 1.51 and 1.43 eV for both samples. More information can be obtained following the PL excitation density dependence for each sample. The state filling of the excited state transition can be observed for the QRs by increasing the pumping power. We can identify the contributions to the PL of QR's first and second excited states at 51.5 and 94.6 meV above the ground state, respectively.

Figures 2(b) and 2(d) show the temperature evolution of the PL transient at ground state energy from $T=15$ K to $T=300$ K for the QD and QR samples, respectively. The detection energies were always fixed at the PL peaks at different temperatures, as indicated by dots shown in Figs. 2(a) and 2(c). A fast rise time of the order of instrumental resolution indicates that there is no phonon bottleneck effect in both samples. The experimental transient decay curves do not reveal saturation effects at a low excitation power as used here and can be described by single exponential functions. The decay time of the QD ground state was about 1.1 ns at a low temperature, which dropped to less than 0.5 ns when the temperature reached 300 K. Surprisingly, the temperature dependence of the decay time behaved quite differently for QR structures. For example, the decay lifetime of QRs became longer with the increasing temperature and reached 10.5 ns at room temperature. In Fig. 3(a), we plotted the temperature dependence of the decay time for both samples. It is clear that at temperature above 150 K, the exciton dynamics change dramatically in the QRs. In Fig. 3(b) the temperature evolution of the PL transient at the excited states of QRs also shows similar behavior.

The behavior of the excitons at a high temperature deserves greater attention. Its origin is possibly due to the thermal population of dark states (states that can neither be accessed by absorbing photons nor relax to other energy states nonradiatively), competing with the exciton radiative recombination ground state. It has been argued that due to the presence of piezoelectric/strain potential and a large asymmetry in the ring profiles, there is reduction in the overlap between the electron and hole wave functions.[18,21] Such

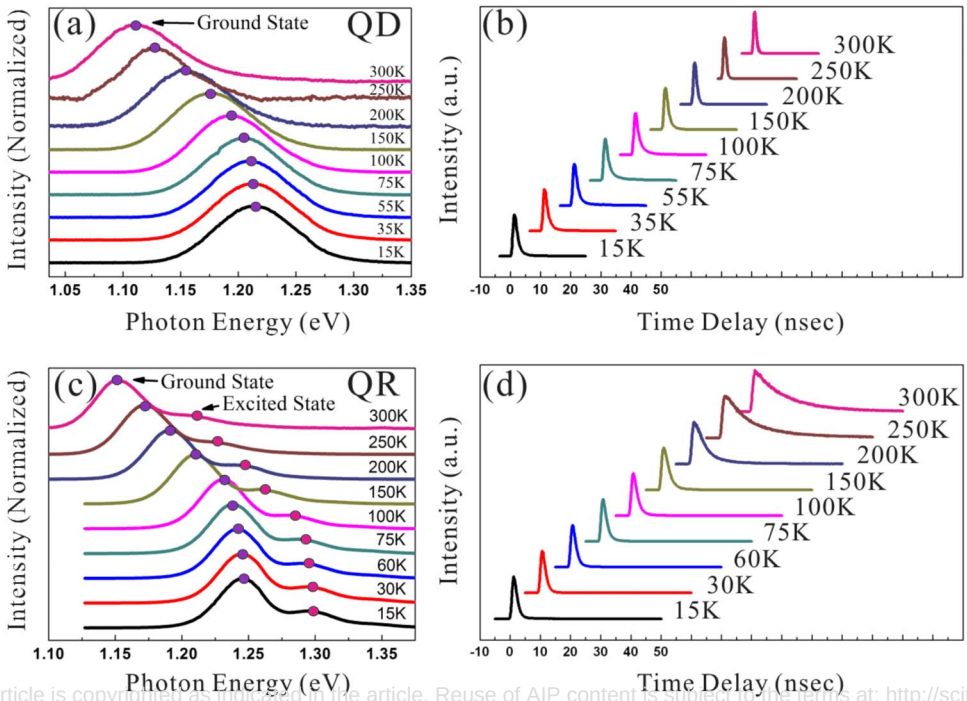


FIG. 2. (Color online) CW PL spectra of (a) QDs and (c) QRs from $T=15$ K to $T=300$ K. PL transient from $T=15$ to 300 K at ground state energy of (b) QDs and (d) QRs. Detection energies of time-resolved spectra were fixed at PL peaks indicated by the dots shown in (a) and (c) at all temperatures.

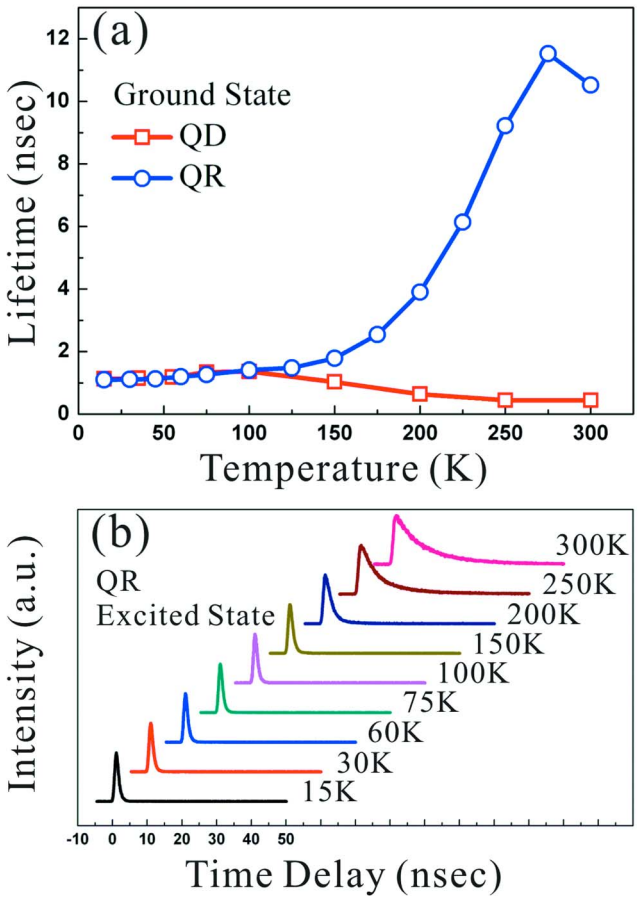


FIG. 3. (Color online) (a) Temperature dependence of the PL decay time of QD and QR ground states. (b) PL transients from $T=15$ to 300 K at first excited state energy of QRs. Detection energies were fixed at excited state peaks as indicated by the dots shown in Fig. 2(c) at all temperatures.

separation causes a reduction in exciton oscillator strength, and a longer decay time is expected for ring structures. We speculate that, at a temperature above 150 K, the electrons and holes tend to occupy states with reduced wave function overlapping, which leads to the slowing down of PL decay time in QRs.

The energy level scheme representative of our proposed model is depicted in Fig. 4. By using a simplified two-level model (as shown in the inset of Fig. 4), the temperature dependence of the decay time curve of QR can be fitted quite well with parameters $\Delta E=13.5$ meV and $\tau'=0.7$ ns (energy separation and relaxation time between the dark state and ground state, respectively). The temperature dependence of the PL lifetime agrees well with the proposed rate equation model of the dark state and exciton ground state. At present, an explanation for the dark excited states is still missing and more experiments and detail calculations are required to clarify the issue.

In conclusion, we have measured the temperature dependence of the PL transients for MBE-grown QD and QR nanostructures. The longer PL lifetime observed at a high temperature in QRs is attributed to the occupation of dark states and the reduced wave function overlapping between electrons and holes, which resulted in the suppression of radiative emission and longer exciton lifetime.

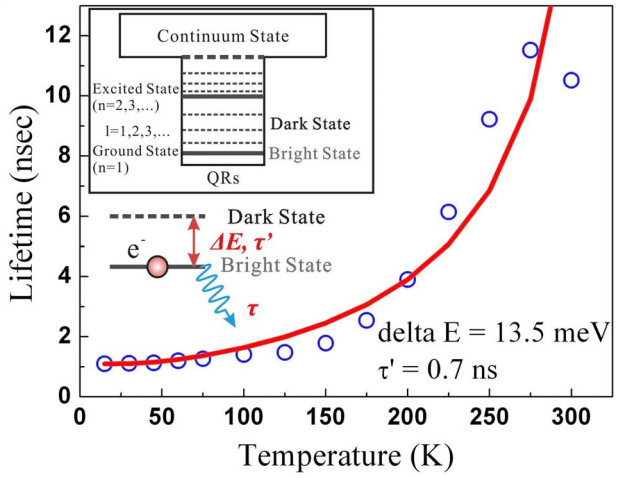


FIG. 4. (Color online) Schematic of the dark states and ground states model and the fitting of QR's decay time curve with a state energy separation of 13.5 meV and relaxation time of 0.7 ns.

This work was supported by the National Science Council of Republic of China under Contract No. NSC 96-2112-M-009-024-MY3 and the MOE ATU program.

[1]H.-S. Ling and C.-P. Lee, J. Appl. Phys. **102**, 024314 (2007).
[2]S. Suraprapapich, S. Panyakeow, and C. W. Tu, Appl. Phys. Lett. **90**, 183112 (2007).
[3]D. Granados, J. M. Garcia, T. Ben, and S. I. Molina, Appl. Phys. Lett. **86**, 071918 (2005).
[4]P. Offermans, P. M. Koenraad, J. H. Wolter, D. Granados, J. M. Garcia, V. M. Fomin, V. N. Gladilin, and J. T. Devreese, Appl. Phys. Lett. **87**, 131902 (2005).
[5]D. Granados and J. M. Garcia, Appl. Phys. Lett. **82**, 2401 (2003).
[6]T. Raz, D. Ritter, and G. Bahir, Appl. Phys. Lett. **82**, 1706 (2003).
[7]A. Lorke, R. J. Luyken, J. M. Garcia, and P. M. Petroff, Jpn. J. Appl. Phys., Part 1 **40**, 1857 (2001).
[8]M. Grochol, F. Grosse, and R. Zimmermann, Phys. Rev. B **74**, 115416 (2006).
[9]R. J. Warburton, C. Schulhauser, D. Haft, C. Schäflein, K. Karrai, J. M. Garcia, W. Schoenfeld, and P. M. Petroff, Phys. Rev. B **65**, 113303 (2002).
[10]H. Pettersson, R. J. Warburton, A. Lorke, K. Karrai, J. P. Kotthaus, J. M. Garcia, and P. M. Petroff, Physica E (Amsterdam) **6**, 510 (2000).
[11]J. Gomis, J. Martinez-Pastor, B. Alén, D. Granados, J. M. Garcia, and P. Roussignol, Eur. Phys. J. B **54**, 471 (2006).
[12]A. Lorke, R. J. Luyken, A. O. Govorov, J. P. Kotthaus, J. M. Garcia, and P. M. Petroff, Phys. Rev. Lett. **84**, 2223 (2000).
[13]T. Kuroda, T. Mano, T. Ochiai, S. Sanguinetti, K. Sakoda, G. Kido, and N. Koguchi, Phys. Rev. B **72**, 205301 (2005).
[14]B. Alén, J. Martinez-Pastor, D. Granados, and J. M. Garcia, Phys. Rev. B **72**, 155331 (2005).
[15]B. Alén, J. Bosch, D. Granados, J. Martinez-Pastor, J. M. Garcia, and L. Gonzalez, Phys. Rev. B **75**, 045319 (2007).
[16]S. Sanguinetti, M. Abbarchi, A. Vinattieri, M. Zamfirescu, M. Gurioli, T. Mano, T. Kuroda, and N. Koguchi, Phys. Rev. B **77**, 125404 (2008).
[17]N. A. J. M. Kleemans, I. M. A. Bominaar-Silkens, V. M. Fomin, V. N. Gladilin, D. Granados, A. G. Taboada, J. M. Garcia, P. Offermans, U. Zeitler, P. C. M. Christianen, J. C. Maan, J. T. Devreese, and P. M. Koenraad, Phys. Rev. Lett. **99**, 146808 (2007).
[18]A. O. Govorov, S. E. Ulloa, K. Karrai, and R. J. Warburton, Phys. Rev. B **66**, 081309 (2002).
[19]G. Piacente and G. Q. Hai, Phys. Rev. B **75**, 125324 (2007).
[20]J. Simonin, C. R. Proetto, Z. Barticevic, and G. Fuster, Phys. Rev. B **70**, 205305 (2004).
[21]J. A. Barker, R. J. Warburton, and E. P. O'Reilly, Phys. Rev. B **69**, 035327 (2004).