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Citation: Journal of Applied Physics **106**, 113522 (2009); doi: 10.1063/1.3267851
View online: http://dx.doi.org/10.1063/1.3267851
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/106/11?ver=pdfcov
Published by the AIP Publishing

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(Received 1 August 2009; accepted 30 October 2009; published online 9 December 2009)

We present systematic experimental studies of the temperature dependence of continuous wave and time-resolved photoluminescence spectroscopy in self-assembled InAs/GaAs nanostructures as the shape of quantum structures evolved from dot to ring. The carrier dynamics show strong dependence on the geometrical shape of the nanostructures under investigation. An increase in photoluminescence decay time of the excited and ground states is observed as the shape of nanostructures changed from dot, volcano, to ring. It is attributed to the carrier thermalization between the dark and ground states. The photoluminescence excitation spectra of the quantum rings reveal resonances related to the dark states. A rate equation model is proposed to interpret the observed carrier dynamics. © *2009 American Institute of Physics*. [doi:10.1063/1.3267851]

I. INTRODUCTION

Low-dimension nanostructures have recently attracted much attention due to their wide physical interests and their potential device applications. 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. Zero dimensional nanostructures, such as quantum dots, have already been widely studied for their applications in semiconductor laser and light emission diode because of the lower threshold current and less temperature sensitivity. In particular, InAs on GaAs (001) self-assembled quantum structures is one of the most studied systems. Several reports have provided experimental evidence for better understanding of the carrier capture and relaxation processes in InAs/GaAs QDs.[1–5] With the advance of the epitaxial growth technology, the quantum ring (QR) structure can now be readily fabricated.[6,7] The nanoscale ring structure, due to its unique geometry, exhibits many interesting physical properties.[8,9] 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,[10] large and negative excitonic permanent dipole moments,[11] and high oscillator strength of the ground state transition.[12] Lorke *et al.*[8] first 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 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] A shape-dependent electronic structure and exciton dynamics were reported by Gomis *et al.*[9] 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. Recently, a photoluminescence (PL) decay time of less than 1 ns was also reported by Sanguinetti *et al.*[15] in concentric QRs. More recently, a longer lifetime was observed at a high temperature in the PL transients for molecular beam epitaxy (MBE)-grown QRs.[16] However, a clear explanation for the suppression of radiative emission and longer exciton lifetime in the QRs is still missing.

In this paper, we report on the spectroscopy of the excited and ground states of self-assembled InAs nanostructures with their geometries evolving from dot, volcano, to ring shape. We observe the strong dependence of PL decay time on the shape of the nanostructures in both the excited and ground states, and attribute this to the thermalization between ground states and "dark" states. A rate equation model is proposed and is found to agree well with the exciton dynamics observed.

II. SAMPLES AND EXPERIMENTS

The GaAs QDs sample studied in this work were grown on GaAs (001) substrates by MBE. The QDs were formed by depositing 2.6 monolayers 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 volcanolike (QV) structures or ring (QR) structures after the annealing processes. Figure 1 shows the atomic force microscopy (AFM) images of the three samples with nanostructures of QV1, QV2, and QRs by performing different annealing times dur-

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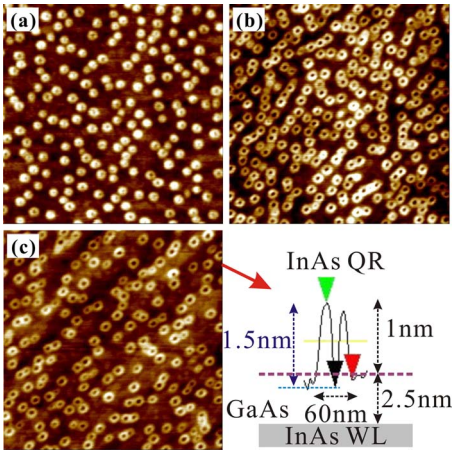

FIG. 1. (Color online) AFM images of (a) QV1, (b) QV2, and (c) QR. The inset shows the surface morphology of the ring, which gives an outer diameter of ~60 nm, a height of ~1 nm, and an inner diameter of 30 nm.

ing the growth procedures. All four samples have the same areal density of $\sim 2\times 10^{10}$ cm^{-2}. The volcanolike structures (QV1), shown in Fig. 1(a), have a base diameter of 50 nm, a height of ~2 nm, and a center crater depth of 0.8 nm. For the volcano structure (QV2) shown in Fig. 1(b), it has a base diameter of 55 nm, a height of ~1.7 nm, and a center crater depth of 2 nm. The final QR shape has a base width of ~60 nm, a height of ~1 nm, and an inner diameter of 30 nm. Figure 2 shows the cross-section 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 solid state diode-pumped (frequency doubled $Nd:YVO_4$) 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. 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

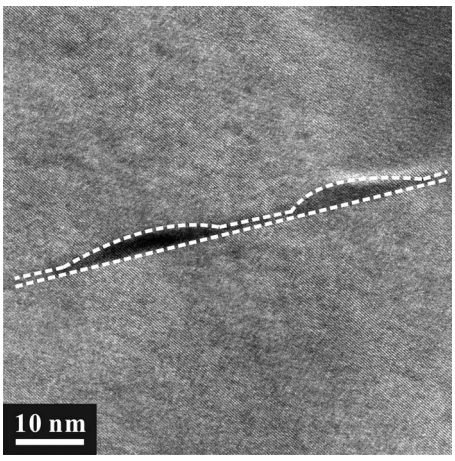

FIG. 2. XTEM image of QR.

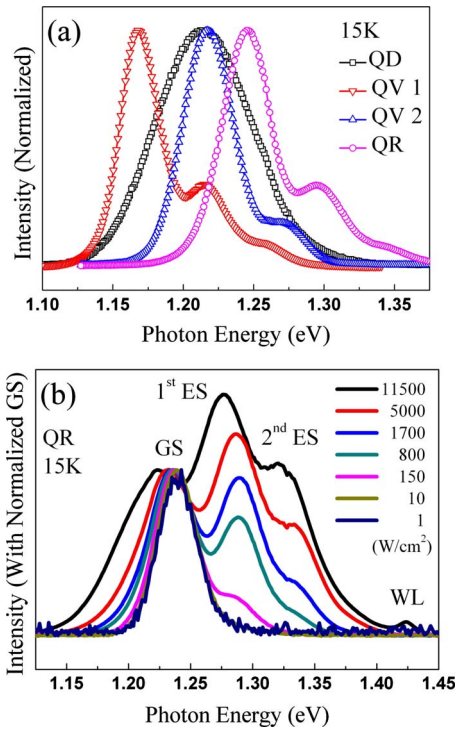

FIG. 3. (Color online) (a) Continuous wave PL spectra of QD, QV1, QV2, and QR. (b) Excitation density dependence of PL from QR sample.

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.

III. DISCUSSIONS

Figure 3(a) shows the continuous wave PL spectra of the four samples at 15 K. The ground state energies of the QD, QV1, QV2, and QR are 1.21, 1.17, 1.22, and 1.25 eV, respectively. The barrier and wetting layer emission are observed clearly at around 1.51 and 1.43 eV for all four samples. More information can be obtained following the PL excitation density dependence for each sample. As an example, the state filling of the excited state transition can be observed for the QRs by increasing the pumping power [as shown in Fig. 3(b)]. 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 4(a)–4(d) show the temperature evolution of the PL transient at ground state energy from $T=15$ K to $T=300$ K for the QD, QV1, QV2, and QR samples, respectively. The detection was fixed at the luminescence peaks at different temperatures. 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 dif-

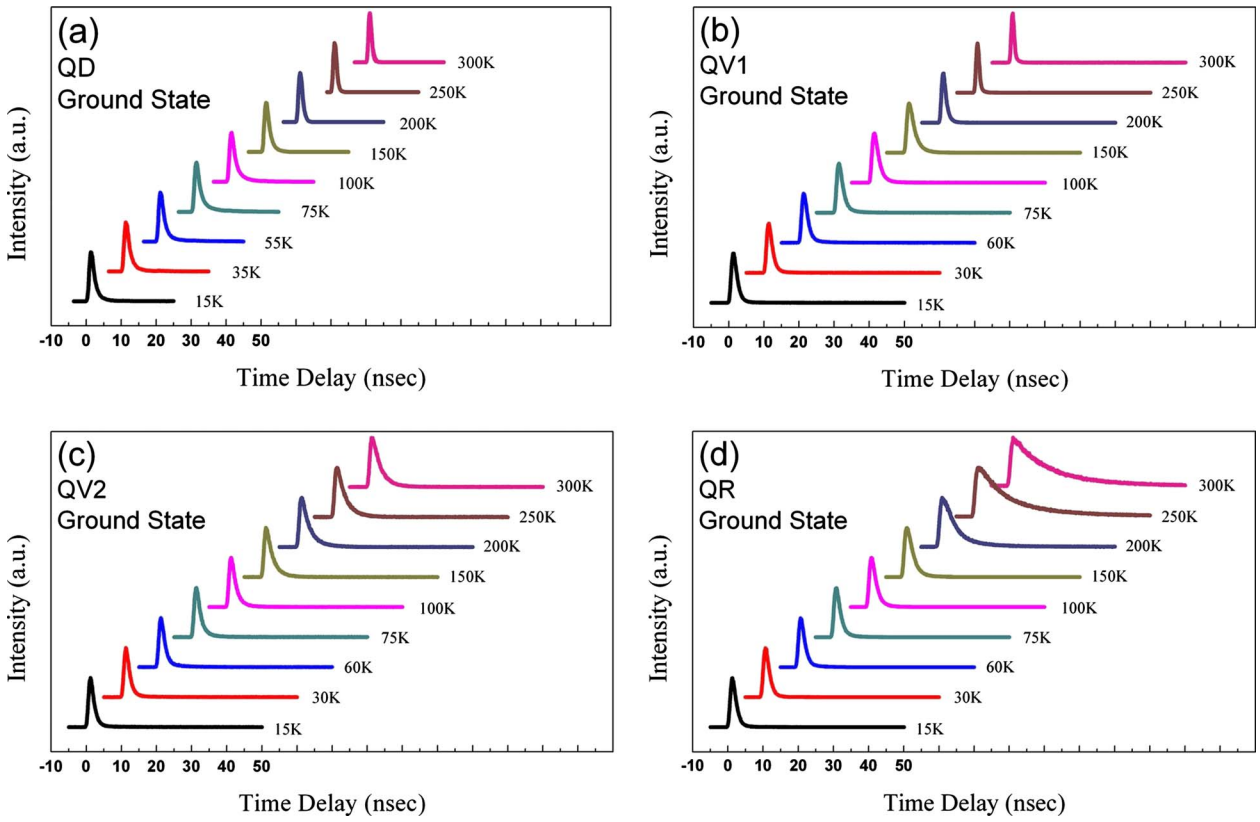

FIG. 4. (Color online) Temperature dependence of PL transients from $T=15$ K to 300 K at ground state energy of (a) QD, (b) QV1, (c) QV2, and (d) QR.

ferently for the QV and QR structures. For example, the decay lifetime of the QRs became longer with the increasing temperature and reached 10.5 ns at room temperature. In Fig. 5, we plotted the temperature dependence of the decay time for all four samples. It is clear that at temperature above 150 K, the exciton dynamics change dramatically as the structures evolve from QD to QR shape. The temperature evolution of the PL transient for QVs and QRs at the excited states also shows similar behavior, as shown in Fig. 6. It has been argued that due to the presence of a large asymmetry in the ring and volcano profiles, there is reduction in the overlap between the electron and hole wave functions.[17,18] Such separation causes a reduction in exciton oscillator strength, and a longer decay time is expected for the volcano and ring structures. However, at a temperature below 150 K, the decay times of all four samples were comparable. Therefore, the above factor could be ruled out. The behavior of the excitons at a high temperature deserves greater attention. Its origin is attributed here to the thermal population of nonradiative excited states (dark states), competing with the exciton radiative recombination states.

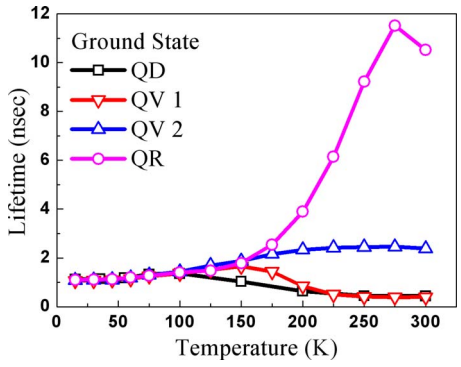

FIG. 5. (Color online) Temperature dependence of the PL decay time of QD, QV1, QV2, and QR samples and the fitting result of the two-band model.

In the following, we give estimates on the electron energy levels of QR and QV versus QD based on cylindrical geometry potential. Due to the cylindrical symmetry of the simulated potential, the three-dimensional problem can be reduced to a two-dimensional eigenvalue equation with two quantum numbers: n (radial part) and ℓ (angular part). The dimensions of the simulated quantum objects were determined from AFM measurements. In the calculations, both conduction and valence bands were modeled by using the $K\cdot P$ one band energy-dependent effective mass equation. The 2D differential equations for the given ℓ were solved by using the finite difference method. The semiconductor parameters (for example, effective mass, band alignment factor, etc.) used in the calculations were taken from Ref. 19.

The calculation results, as shown in Fig. 7, clearly show that there are quite a few ℓ states above the ground states in the ring geometry than in QD. Be noted that the QR also has a much smaller energy separation between the ℓ states than in the case of QD. As the temperature increased, the carriers in the ground state of QR were able to populate thermally the dark states (ℓ states). However, only excitons with zero total momentum can emit photons. This condition arises as the photons' angular momentum is exhausted by the atomic wave function involved in the optical transition. The optical selection rule $\Delta\ell=0$ means that the emission intensity of a

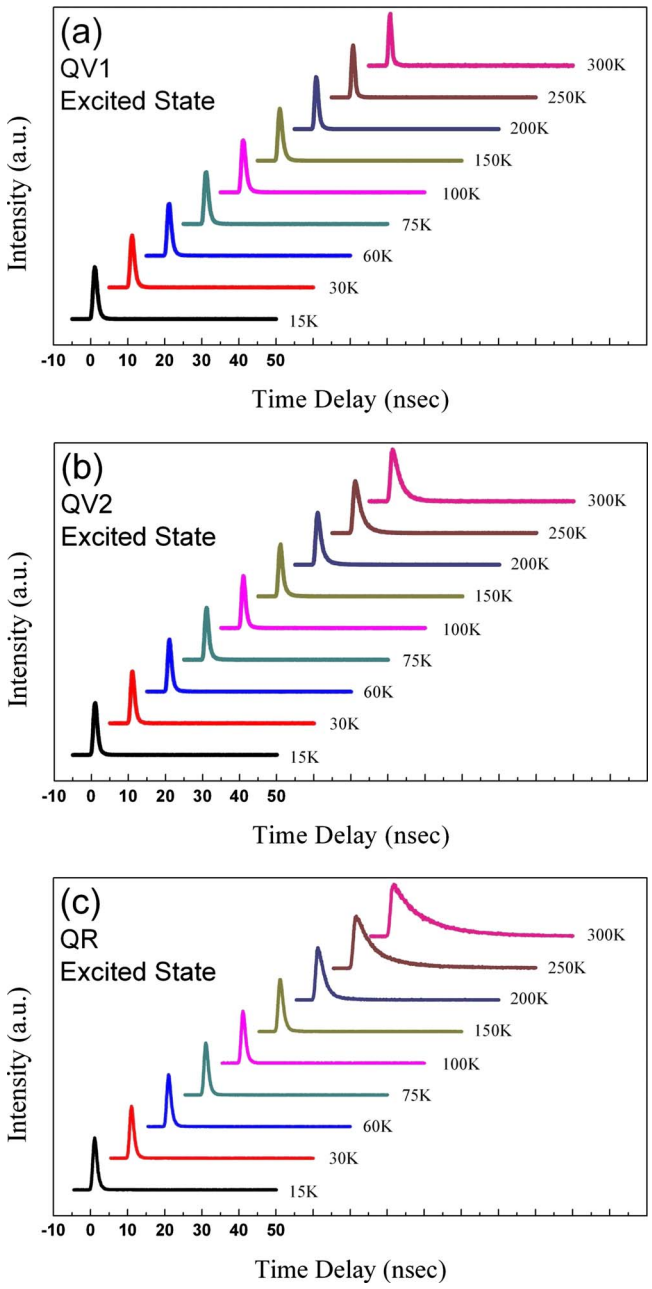

FIG. 6. (Color online) Temperature dependence of PL transients from $T=15$ K to 300 K at the first excited state energy of (a) QV1, (b) QV2, and (c) QR.

QR can be suppressed dramatically if the carriers were in favor of occupying the dark states ($\ell \neq 0$ states). This suppression implies that the exciton lifetime becomes significantly longer as its dipole-allowed decay is prohibited. On the contrary, for the quantum dots, the $\ell=0$ state plays a more important role due to the lesser ℓ states and larger energy separations. Therefore, in the QDs, the excitons in the $\ell=0$ states are more important.

It should be noticed that, in the calculations, the number of dark states in the ring structure is only a few times higher than the QD. However, the observed decay time of the QR at 300 K is ten times longer. In Fig. 3, the QR has the highest PL energy and is thus closer to the wetting layer. It is possibly due to the existence of a continuum of tail states near the

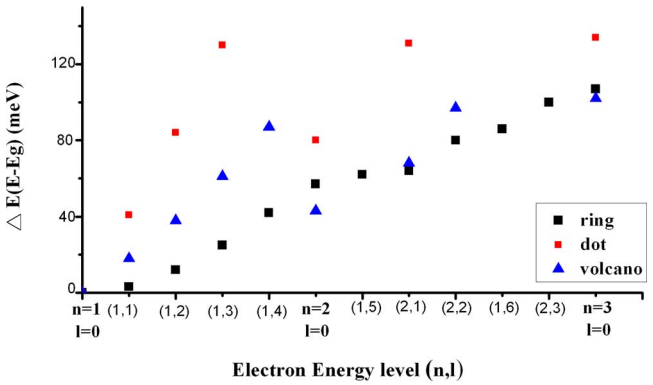

FIG. 7. (Color online) Calculated electron energy levels in QD, QV, and QR by solving the $K\cdot P$ one band energy-dependent effective mass equation.

wetting layer band edge,[20,21] which provides another origin of dark states for the excited carriers.

To interpret our experiment findings, we proposed a simplified two-level (dark and ground level) model in which the carrier relaxation can be described by the following rate equation: $dN/dt=-N/\tau_0+Ne^{-\Delta E/KT}/\tau'$, where N is the population of the ground state, ΔE is the energy difference between the dark and ground state, τ' is the decay time from the dark states to the ground states, and the τ_0 represents the lifetime of the exciton at the ground level at 0 K. The carrier lifetime as a function of temperature can be derived from the rate equation: $\tau=(1/\tau_0-e^{-\Delta E/KT}/\tau')^{-1}$. The temperature dependence of the decay time cure of QR (as shown in Fig. 5) can be fitted quite well by the above equation with parameters $\Delta E=13.5$ meV and $\tau'=0.7$ ns. Although our simple model will encounter an unphysical divergence of τ (for $\tau'/\tau_0=e^{-\Delta E/KT}$) at higher temperature, however, it should be noted that this simplified model has ignored mechanisms such as carriers escape through nonradiative recombination channels, retrapping or recapture of carriers, transitions from the ground state back to the dark states, and so forth. A thorough theoretical investigation including factors mentioned above should be able to solve the problem encountered in our model.

In Fig. 8, we show the photoluminescence excitation spectra recorded for the QR and QD samples with the detection energy fixed at the center of each sample's ground state energy. The PLE spectra for the QR sample reveal several

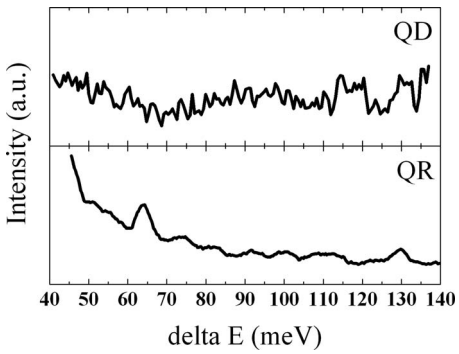

FIG. 8. PL excitation spectra of QD (upper curve) and QR (lower curve) above the first excited state. The detection energy is fixed at the center of ground state energy.

resonances above the detection energy. When the PLE spectra were detected on the low or high energy parts of the ground state PL spectrum, we found similar results. This was consistent with a smooth change in carrier confinement energy with the average QR dimensions. The average energy spacing between these resonances is about 10 meV, which is very close to the numbers used to fit the temperature dependence of the QRs' decay time curve in Fig. 5. Therefore, they must be related to the absorption at nonradiative states (the dark states). On the other hand, we could not identify any resolved contribution due to the dark state absorption in the QD PLE spectra, as shown in Fig. 8. Our interpretation of the exciton dynamics in the above quantum structures through the interplay between ground and dark states is further supported by the PLE results.

IV. CONCLUSIONS

In conclusion, we have measured the temperature dependence of the PL transients for MBE-grown QD, QV, and QR nanostructures. The longer PL lifetime observed at a high temperature as the structures evolved from dot to ring shape is attributed to the occupation of energy levels with different angular momentum in QRs. Those states with nonzero angular momentum in QRs became the dark states and were populated at a high temperature, which resulted in the suppression of radiative emission and longer exciton lifetime. The temperature dependence of the PL lifetime agrees well with the proposed rate equation model of the dark and bright states.

ACKNOWLEDGMENTS

This work was supported by the National Science Council of Republic of China under Contract Nos. NSC 96-2112-M-009-024–MY3 and NSC 96-2120-M-009-004-, and by the MOE ATU program.

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