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Long luminescence lifetime in self-assembled InGaAs/GaAs quantum dots at room temperature

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Time-resolved photoluminescence (PL) measurements of high-quality self-assembled small $In_{0.5}Ga_{0.5}As/GaAs$ quantum dots (QDs) show that the PL decay time of the QD ground state transition is nearly constant when the temperature is below 80 K and increases monotonously from 1.0 to 5.5 ns when the temperature increases from 80 to 300 K. The increased radiative lifetime of the QD ground state at higher temperatures is attributed to the thermal population of the subwetting-layer continuum states and could be one of the fundamental reasons for the low modal gain of the QD ground state transition in single-layer self-assembled QD lasers. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3021018]

Research on self-assembled semiconductor quantum dots (QDs) has received much attention due to their potential application in optoelectronics devices, such as QD lasers.[1] The radiative lifetime of the excitons in QDs at room temperature is one of the most important device parameters, being inversely proportional to the modal gain of QD lasers.[2–5] The radiative lifetime of strongly confined excitons in QDs, where the energy separation between the ground state and the first excited exciton state is larger than the thermal energy k_BT (k_B is the Boltzmann constant and T is the temperature), should be almost independent of T. However, in real QDs, the radiative lifetime of the ground state excitons is expected to increase with increasing temperature due to the thermal population of optically inactive or poorly active exciton states.[6–8] This phenomenon was first observed in InGaAs/GaAs QDs by Wang *et al.*[9] in 1994, in InAs/GaAs QDs by Yu *et al.*[10] in 1996, and by other groups later.[11] They found that the photoluminescence (PL) radiative lifetime increases first with increasing temperature and then decreases at high temperatures. In fact, Marcinkevičius and Leon[12] reported that the PL decay time of InAs QDs decreases monotonously with increasing temperature. These contradicting experimental results could be related to the different sample qualities. To investigate the intrinsic radiative lifetime of excitons in QDs at high temperatures, two important issues should be taken into account. First, the influence of the nonradiative recombination on the surface and in the substrate should be suppressed, which can be realized by growing AlAs confining layers around the QDs and their barriers. Second, the amount of defects in and around the QDs themselves should be minimized, which strongly depends on the growth temperature, the deposition amount, and the quality of the barrier layers. In this letter, we investigate the temperature dependence of the PL decay time in high-quality small Stranski–Krastanow-grown InGaAs/GaAs QDs. We find that the PL decay time increases monotonously with increasing temperature when the temperature is above 80 K and reaches a value of 5.5 ns at 300 K.

The sample was grown with a molecular beam epitaxy instrument on a (001)-oriented undoped GaAs substrate. The layout of the structure is as follows: GaAs substrate/500 nm GaAs/8 nm AlAs/80 nm GaAs/InGaAs QDs/20 nm GaAs/8 nm AlAs/20 nm GaAs Cap layer. The QDs were formed by depositing 5 monolayers (MLs) of $In_{0.5}Ga_{0.5}As$ at 500 °C. The details of the growth conditions and basic characterization of this sample can be found in Refs. 13 and 14. Transmission electron microscopy observation shows that the QDs are half-lens shaped with the lateral base of 5–8 nm and the height of around 2 nm. The area density of QDs is about 3×10^{11} cm^{-2}.

The power-dependent continuous-wave PL was excited via a Ti-sapphire laser with tunable wavelength. Shown in Fig. 1 is the PL spectrum measured at 10 K with the excitation wavelength of 790 nm. The focus spot is about 50 μm in diameter. At the excitation power of 0.02 mW, the PL spectrum is characterized by a peak located at 1.326 eV, with a full width at half maximum of 38 meV. As the excitation power increases from 0.02 to 36 mW, the low-energy transi-

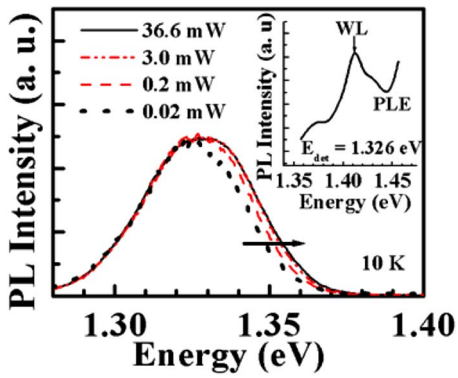

FIG. 1. (Color online) Continuous-wave PL spectra at different excitation powers. The maximum PL intensity is normalized for comparison. The inset shows the PLE spectrum.

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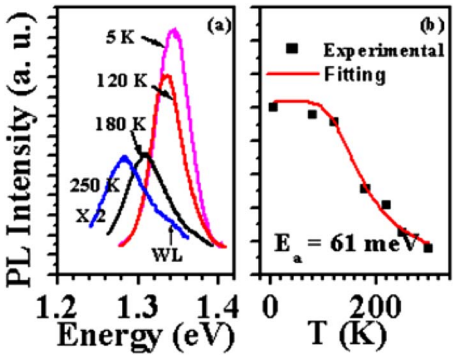

FIG. 2. (Color online) (a) PL spectra at different temperatures; (b) PL peak intensity vs temperature.

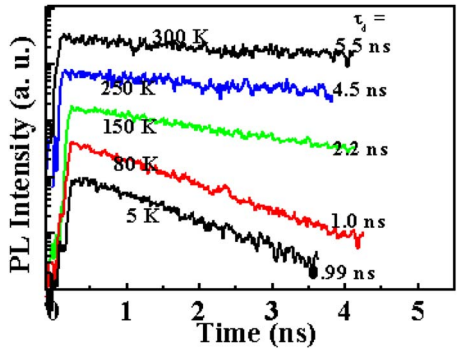

FIG. 3. (Color online) Time decay of PL detected at the peak energies at different temperatures.

tions saturate, while transitions at higher energy are activated, leading to a blueshift of the peak position. This is similar to what is previously observed in high-quality small InAs/GaAs QDs.[15] The high-energy tail at high excitation power results from an excited state transition involving ground state confined electrons and the two-dimensional hole continuum associated with the wetting layer (WL). The inset of Fig. 1 shows the PL excitation (PLE) spectrum with the detection energy of 1.326 eV. The peak at 1.41 eV is attributed to the electron to heavy-hole transitions of the WL. Note that the continuum background below the WL bandgap can be clearly seen. These sub-WL transitions could be related to the WL morphology and/or the coupling of the QDs with their surroundings and are believed to be responsible for the efficient carrier relaxation in self-assembled QDs.[16–20] The upconversion PL from the GaAs barrier layer can even be observed at room temperature when pumping in the continuum background region (not shown here), indicating the high crystal quality of the QD heterostructure.

The time-resolved PL was excited by a femtosecond Ti-sapphire laser with a pulse width of 120 fs and a repetition rate of 76 MHz at the wavelength of 800 nm. The PL signal was dispersed with a monochromator and detected with a streak camera. The overall time resolution of the system is 2.5 ps. The sample temperature can be tuned from 5 to 300 K in a helium cryostat. The excitation power is kept to be 4 mW. The time-integrated PL spectra at different temperatures are shown in Fig. 2(a). The peak intensity of the QD emission is plotted as a function of temperature in Fig. 2(b). The thermal activation energy E_a is determined to be 61 meV by fitting the experimental data with the formula $I(T)=I_0/[1+C\exp(-E_a/k_BT)]$, where $I(T)$ and I_0 are the intensities at temperatures T and 0 K, respectively. Clearly, the observed activation energy is much smaller than the energy difference between the QD peak energy and the WL bandgap, indicating that the drop of QD PL intensity at high temperatures cannot simply be ascribed to the thermal emission of carriers from QDs to the WL. Actually, not only the thermal emission of carriers from the QDs but also the carrier capture by the defects in the barrier matrix before capturing to the QDs, and the thermal population of the optically inactive states can lead to the drop of the PL intensity.

The time decays of the quantum-dot PL detected at the peak energies for different temperatures from 5 to 300 K are shown in Fig. 2. The change in the PL decay time as a function of the temperature is depicted in Fig. 3. It can be seen that the PL decay time is about 1 ns and constant from 5 to 80 K. As the temperature increases from 80 to 300 K, the PL decay time increases monotonously from 1 to 5.5 ns. Note that a decrease in the PL decay time at 300 K has been widely reported in the literature.[9–12,21–23] The observation of long PL decay times, even at 300 K, indicates a high quality of our QD sample. The two AlAs layers can suppress the influence of nonradiative centers on the surface and in the substrate. In addition, the high growth temperature and the small deposition amount facilitate the formation of defect-free QDs.

The thermal population of the higher exciton states, which are optically inactive or have lower oscillator strength, accounts for the prolonged net radiative recombination lifetime in the higher temperature region.[6–8] The radiative recombination rate $\Gamma_R(T)$ at temperature T is given by $\Gamma_R(T)=\Gamma_R(0)/[1+g\exp(-\Delta E/k_BT)]$, where ΔE is the energy difference between the ground state of the QD and some optically inactive excited states and g is the ratio between the degeneracy of the optically inactive states to that of the ground state. The total recombination rate of the QD ground state is given by[11]

$$\Gamma_{\mathrm{rec}}(T)=\frac{1}{\tau_{\mathrm{decay}}}=\Gamma_R(T)+\Gamma_{\mathrm{NR}}(T). \tag{1}$$

Here, τ_{decay} is the measured PL decay time. Γ_{NR} is the nonradiative recombination rate associated with carriers escaping out of the QDs and is given by $\Gamma_{\mathrm{NR}}=\Gamma_0\exp(-\delta/k_BT)$, where δ is the activation energy for thermal carrier escape and Γ_0 is an escape attempt frequency. The parameters in Eq. (1) can be determined to be $\Gamma_R(0)=1.0\times10^{-3}$ ps^{-1}, $g=27$, $\Delta E=36$ meV, and $\Gamma_0=7.0\times10^{-5}$ ps^{-1}, according to the nonlinear least-squares fitting of the experimental data in Fig. 3. The fitting result does not strongly depend on the value of δ. The radiative recombination rate is two orders of magnitude larger than the nonradiative recombination rate. Therefore, the measured PL decay time is dominated by the radiative recombination even at room temperature. The large value of 27 for the g factor (much larger than that determined in Ref. 11) rules out the possibility that the optically inactive states are related to the first excited hole states. The energy difference $\Delta E=36$ meV suggests that the optically inactive states are located in the continuum background determined in the PL spectrum. The values of ΔE and E_a are not the same because E_a contains more information on the complex mechanisms of the carrier loss in the QD heterostructure. For comparison, the typical data for small InAs or InGaAs QDs from Refs. 9, 10, and 23 are also fitted by using formula (1),

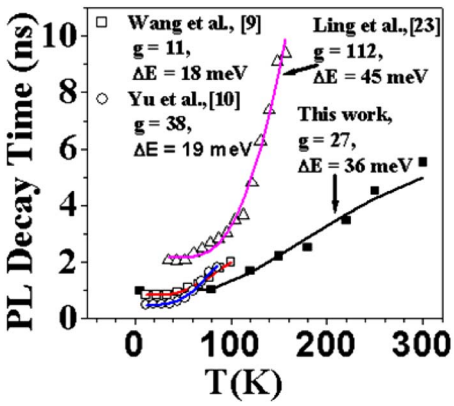

FIG. 4. (Color online) The PL decay times of InGaAs/GaAs QDs vs temperature (closed squares). Open symbols represent data from the literature. The solid lines show the fitting results. The fitting parameters are given in the figure.

as shown in Fig. 4. Note that the values of g ranges from 11 to 112. Therefore, the value of $g=27$ in our QD sample is reasonable.

It is widely believed that the sub-WL continuum background plays an important role in the carrier relaxation in self-assembled QDs.[16–20] The photoexcited carriers in the barrier are captured into the QD ground state through a continuum background relaxation.[19,20] The increase in the PL decay time of the QD ground state transition due to the feeding of carriers into the sub-WL states at high excitation levels and low temperatures has recently been reported.[19] As proposed by Vasanelli *et al.*,[17] the sub-WL continuum transition results from the intrinsic crossed transitions between the bound QD states and the delocalized states. Therefore, the sub-WL continuum transitions should have lower oscillation strength but higher degeneracy, compared with the QD ground state transitions. At high temperatures, the continuum states are thermally populated and behave like carrier reservoirs. Consequently, the PL decay time will be longer if the effect of the nonradiative recombination on the surface and in the substrate is suppressed enough.

In summary, the temperature dependent PL decay time in high-quality small InGaAs/GaAs QDs was investigated by using time-resolved PL experiments. We have observed that the PL decay time of the QD ground states increases with the increase in temperature from 80 to 300 K, and the increase is ascribed to the thermal population of the sub-WL continuum states. As the intrinsic radiative lifetime is at least as long as the observed PL decay time, our observation gives one of the fundamental reasons for the low modal gain of the QD ground states in single-layer self-assembled QD lasers.

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[1] D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, New York, 1998).
[2] L. V. Asryan, M. Grundmann, N. N. Ledentsov, O. Stier, R. A. Suris, and D. Bimberg, J. Appl. Phys. **90**, 1666 (2001).
[3] M. V. Maximov, Yu. M. Shernyakov, A. F. Tsatsul'nikov, A. V. Lunev, A. V. Sakharov, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, A. R. Kovsh, P. S. Kop'ev, L. V. Asryan, Zh. I. Alferov, N. N. Ledentsov, D. Bimberg, A. O. Kosogov, and P. Werner, J. Appl. Phys. **83**, 5561 (1998).
[4] Z. C. Xu, D. Birkedal, M. Juhl, and J. M. Hvam, Appl. Phys. Lett. **85**, 3259 (2004).
[5] Z. C. Xu, Y. T. Zhang, A. Tackeuchi, Y. Horikoshi, and J. M. Hvam, Appl. Phys. Lett. **92**, 063103 (2008).
[6] D. Citrin, Superlattices Microstruct. **13**, 303 (1993).
[7] H. Gotoh, H. Ando, and T. Kakagahara, J. Appl. Phys. **81**, 1785 (1997).
[8] G. Pistone, S. Savasta, O. Di Stefano, R. Girlanda, and S. Portolan, Phys. Status Solidi B **245**, 1067 (2008).
[9] G. Wang, S. Fafard, D. Leonard, J. E. Bowers, J. L. Merz, and P. M. Petroff, Appl. Phys. Lett. **64**, 2815 (1994).
[10] H. P. Yu, S. Lycett, C. Roberts, and R. Murray, Appl. Phys. Lett. **69**, 4087 (1996).
[11] M. Gurioli, A. Vinattieri, M. Zamfirescu, and M. Colocci, Phys. Rev. B **73**, 085302 (2006), and the references therein.
[12] S. Marcinkevičius and R. Leon, Phys. Rev. B **59**, 4630 (1999).
[13] Z. C. Xu, Y. T. Zhang, and J. M. Hvam, Nanotechnology **18**, 325401 (2007).
[14] Z. C. Xu, K. Leosson, D. Birkedal, J. M. Hvam, J. Sadowski, Z. Y. Zhao, X. S. Chen, Y. M. Liu, and K. T. Yang, J. Cryst. Growth **251**, 177 (2003).
[15] H. Lee, W. D. Yang, and P. C. Sercel, Phys. Rev. B **55**, 9757 (1997).
[16] Y. Toda, O. Moriwaki, M. Nishioka, and Y. Arakawa, Phys. Rev. Lett. **82**, 4114 (1999).
[17] A. Vasanelli, R. Ferreira, and G. Bastard, Phys. Rev. Lett. **89**, 216804 (2002).
[18] E. W. Bogaart, J. E. M. Haverkort, T. Mano, T. van Lippen, R. Nötzel, and J. H. Wolter, Phys. Rev. B **72**, 195301 (2005).
[19] 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).
[20] G. Rainò, G. Visimberga, A. Salhi, M. De Vittorio, A. Passaseo, R. Cingolani, and M. De Giorgi, Appl. Phys. Lett. **90**, 111907 (2007).
[21] A. Fiore, P. Borri, W. Langbein, J. M. Hvam, U. Oesterle, R. Houdré, R. P. Stanley, and M. Ilegems, Appl. Phys. Lett. **76**, 3430 (2000).
[22] 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).
[23] L. M. Kong, Z. C. Feng, Z. Y. Wu, and W. J. Lu, Semicond. Sci. Technol. **23**, 075044 (2008).