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The exciton radiative lifetime in submonolayer (SML) InGaAs/GaAs quantum dots (QDs) grown at 500 °C was measured by using time-resolved photoluminescence from 10 to 260 K. The radiative lifetime is around 90 ps and is independent of temperature below 50 K. The observed short radiative lifetime is a key reason for the high performance of SML QD devices and can be explained by the theory of Andreani *et al.* [Phys. Rev. B **60**, 13276 (1999)] calculating the radiative lifetime of QDs formed at the interface fluctuations of a quantum well, as the SML QDs are 20–30 nm in diameter and embedded within the lateral InGaAs QW. © *2008 American Institute of Physics*.
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The stacked submonolayer (SML) growth of quantum dots (QDs) presents an alternative approach to Stranski-Krastanow (SK) grown QDs.[1] SML quantum dots have been used to fabricate high-power edge-emitting lasers[2–4] and single-mode vertical-cavity surface-emitting lasers (VCSELs) with high modulation bandwidth.[5,6] Self-sustained pulsation in the oxide-confined VCSELs based on SML InGaAs QDs has recently been observed.[7] The high performance of SML QD lasers has usually been ascribed to the high density and uniformity of SML QDs.

As a matter of fact, another important parameter determining the maximum modal gain (g_{mod}) for transition in a QD is the exciton radiative lifetime (τ),[8] i.e., $g_{mod} \propto 1/\tau$. In our previous work,[9] we investigated the carrier dynamics of SML InGaAs QDs grown at 480 °C. The exciton radiation lifetime at 10 K was found to be around 700 ps; however, the QD photoluminescence (PL) signal quenches at room temperature.[10] The PL signal from the SML QD ground state can be observed even at room temperature if we increase the growth temperature of SML QDs to 500 °C.[11] Lasers using high-temperature grown (HTG) SML QDs show very high modal gain and zero linewidth enhancement factor.[11] In this letter, we report the exciton radiative lifetime of HTG SML QDs from 10 to 260 K, via time-resolved photoluminescence (TRPL). We found that the exciton radiative lifetime in SML QDs is around 90 ps and is almost independent of temperature below 50 K.

The sample was grown on a semi-insulating GaAs (0 0 1) substrate via molecular beam epitaxy. The layer structure of the sample is depicted in Fig. 1. The SML QD layer was formed with ten cycles of alternate deposition of 0.5 ML InAs and 2.5 ML GaAs, at 500 °C, the same as in Ref. 11. The inset of Fig. 1 shows the in-plane transmission electron microscope (TEM) image of SML QDs. The characteristic size of the SML QDs is 20–30 nm in diameter, which lies between the reported value in Refs. 6 and 12.

In the TRPL setup, the sample was cooled in a closed-cycle He cryostat and excited in the GaAs barriers with 130 fs pulses from a Ti:sapphire laser at the wavelength of 400 nm. The PL signal was collected, dispersed, and synchronously detected using a streak camera with overall time resolution of 15 ps. The excitation spot in TRPL is about 100 μm in diameter.

The time-integrated PL spectrum of the HTG SML QDs at from 10 to 260 K is shown in Fig. 2(a). At 10 K, the PL band shows a peak at 1.355 eV with a full width at the half maximum of 19 meV and the wavelength-integrated PL in-
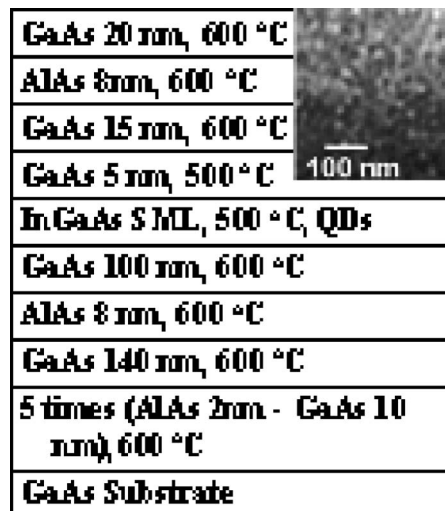

FIG. 1. The layer structure of InGaAs SML QD sample. The inset shows the in-plane TEM image of SML QDs.

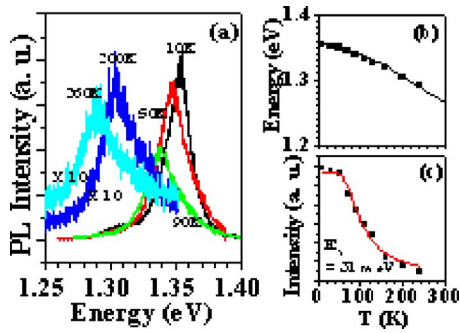a)Electronic mail: zcxu@nankai.edu.cn.

FIG. 2. (Color online) (a) The time-integrated PL spectrum from SML InGaAs/GaAs QDs at different temperatures. (b) The energy shift of the PL peak with respect to the temperature. (c) The variation of the integrated PL intensity with respect to the temperature. The dots and the solid lines in (b) and (c) represent the experimental and the fitting results, respectively.

FIG. 4. (Color online) (a) The PL transients of HTG SML InGaAs QDs detected at different emission energies at 10 K. (b) The PL decay time as a function of emission energies.

tensity increases linearly with the increase of the excitation power, indicating very low defect density in our sample. As shown in Fig. 2(b), the peak energies of the PL band redshifts as the temperature increases, and the trend can be well fitted by using Varshini formula,[13] $E(T) = E(0 \text{ K}) - \alpha T^2/(\beta + T)$. Here, $E(T)$ is the peak energy at temperature T, $E(0 \text{ K}) = 1.355$ eV, $\alpha = 3.56 \times 10^{-4}$ eV/K, $\beta = 83$ K. Figure 2(c) shows that the integrated PL intensity is kept constant below 50 K and then decreases as the temperature increases, with a thermal activation energy of $E_a = 31$ meV. E_a is determined by fitting the experimental data with the formula $I(T) = I(10 \text{ K})/(1 + A \exp(-E_a/kT))$, where $I(T)$ is the integrated PL intensity at temperature T, k is the Boltzmann constant, and A is a fitting parameter.

In order to clarify the origin of the activation energy, we measured the surface photovoltaic (SPV) signal from the SML QD laser structure of Ref. 11. The experiments were carried out at room temperature, under the condition of normal incidence. As shown in Fig. 3, three peaks can be clearly identified. The peak located at 1.276 eV is due to the QD ground states. The broad peak from 1.334 to 1.352 eV is due to the lateral quantum well (QW) states.[10,12] The third broad peak located at 1.43 eV is ascribed to the doped GaAs barrier. Assuming the conduction (valence) bandoffset to be 60 (40%) of the bandgap offset between the QDs and the lateral QW, the conduction bandoffset will be 34.8 meV. This value is very close to the measured thermal activation energy of 31 meV, suggesting that the PL quenching at high temperature is mainly due to the thermal escape of carriers from the QDs to the lateral QWs.

Figure 4(a) shows the PL transients at different emission energies at 10 K. The PL decay time τ_d can be obtained by fitting the curve with a single exponential process. Figure 4(b) shows τ_d across the PL band. It can be seen that τ_d at the
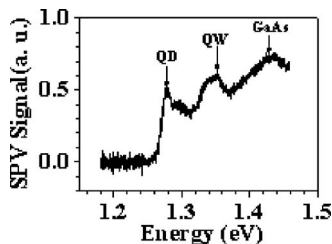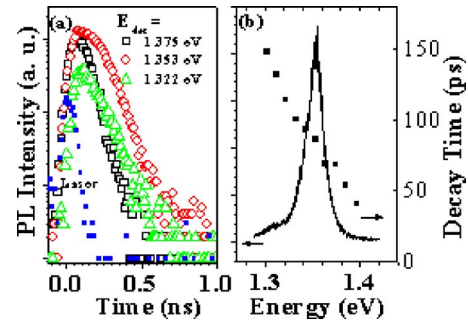
PL peak energy is as short as 90 ps. The trend for the size dependence of τ_d for HTG SML InGaAs QDs is the same with that for low-temperature grown SML QDs.[9] Figure 5(a) shows the PL transient detected at PL peak energies at different temperatures. The corresponding τ_d from 10 to 260 K is plotted in the inset of Fig. 5(a). It can be seen that τ_d is almost independent of temperature.

Now let us discuss the PL decay time τ(K) for HTG SML QDs. Below 50 K, the integrated PL intensity is nearly independent of the temperature, so the measured PL decay time is dominated by the exciton radiative lifetime. The nonradiative process plays an important role above 50 K, as the emission efficiency decreases as the temperature increases. Using the relations $\tau_{\text{rad}}(T) = \tau(T) \times I(10 \text{ K})/I(T)$ and $\tau_{\text{nrad}}(T) = \tau(T) \times I(10 \text{ K})/[I(10 \text{ K}) - I(T)]$ given in Ref. 14, the radiative (τ_{rad}) and nonradiative (τ_{nrad}) lifetimes at different temperatures, can be evaluated, as shown in Fig. 5(b). It can be seen that the radiative lifetime is around 90 ps, nearly independent of temperature below 50 K and then increases up to 800 ps at 260 K. The nonradiative lifetime decreases with the increase of temperature due to the thermal escape of carriers from the QDs to the lateral QWs. The nonradiative lifetime decreases to 100 ps at 260 K, which is very close to the reported value for SK InGaAs/GaAs QDs.[15]

In 2000, Fiore *et al.* reported that the PL decay time for SK InAs/InGaAs QDs varies from 1.2 ns (at 50 K) to 1.8 ns (at 300 K).[16] The decrease of the radiative efficiency from 5 to 300 K by a factor of 13 was attributed to either the long radiative lifetime of 18 ns or the existence of the nonradiative traps in the In(Ga)As matrix around the QDs. The ob-

FIG. 3. The surface photovoltaic signal from a SML InGaAs/GaAs QD laser structure at room temperature.
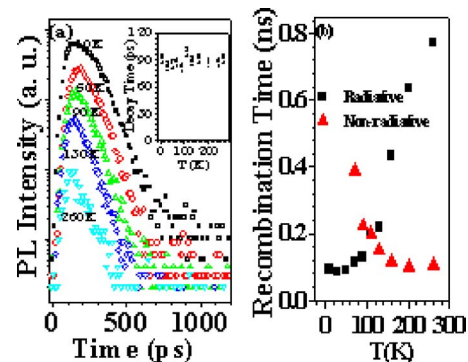

FIG. 5. (Color online) (a) The PL transient of HTG SML InGaAs QDs detected at the PL peak energies at different temperatures. (b) The temperature dependence of radiative lifetimes and nonradiative lifetimes of SML InGaAs/GaAs QDs.

served long radiative lifetime for the SK QDs is normally attributed to the small overlap integral of the electron and hole wave functions due to the piezoelectric effect,[8,17] and can be modified by thermal annealing.[18] On the other hand, the low-temperature radiative lifetime of InGaAs/GaAs quantum wells is around 400 ps,[19] much longer than the present case. Therefore, the observed short radiative lifetime for the HTG SML InGaAs QDs cannot be simply explained by the enhancement of the overlap integral of the electron and hole wave functions.

Very short radiative lifetime of 100 ps for GaAs QDs formed at the interface fluctuation of a GaAs/AlGaAs QW has recently been observed by Hours *et al.*[20] Actually, the SML InGaAs/GaAs quantum dots are disklike and embedded within the lateral quantum well, according to Refs. [10] and [12]. They are very similar to the InGaAs QDs formed at the interface fluctuation of an InGaAs/GaAs QW. As predicted in the theory of Andreani *et al.*, the oscillator strength should be very large and depends on the QD radius.[21] Our previous results show that the radiative lifetime for SML InGaAs QDs of 7.5 nm in diameter is around 700 ps, due to the strong confinement.[9] In the present work, the average lateral size of SML QDs is 25 nm and very close to the Bohr radius of excitons.[22] According to the theory of Andreani *et al.*, the carriers are confined within the SML QDs and the center of mass of the exciton is quantized with the lateral QW. The oscillator strength should be larger due to the larger area of the center of mass of the exciton wavefunction.

In summary, by using TRPL at various temperatures, we found that the exciton radiative lifetime of SML InGaAs QDs grown at high temperature is as short as 90 ps, and is almost independent of temperature below 50 K. Above 50 K, as the temperature increases, the radiative lifetime increases and reaches to 800 ps at 260 K. The observed short radiative lifetime can be explained using the theory of Andreani *et al.* calculating the radiative lifetime of the QDs formed at the interface fluctuations of a QW. The large oscillator strength of SML QDs is one of the key reasons for the high modal gain of the SML QD lasers and the high modulation bandwidth of SML QD VCSELs.

[1] I. L. Krestnikov, N. N. Ledentsov1, A. Hoffmann, and D. Bimberg, Phys. Status Solidi A **183**, 207 (2001).

[2] S. S. Mikhrin, A. E. Zhukov, A. R. Kovsh, N. A. Marleev, V. M. Ustinov, Y. M. Shernyakov, I. P. Soshnikov, D. A. Livshits, I. S. Tarasov, D. A. Bedarev, B. V. Volovik, M. V. Maximov, A. F. Tsatsulnikov, N. N. Ledenstov, P. S. Kopev, D. Bimberg, and Z. I. Alferov, Semicond. Sci. Technol. **15**, 1061 (2000).

[3] A. F. Zhukov, A. R. Kovsh, S. S. Mikrin, N. A. Maleev, V. M. Ustinov, D. A. Livshits, I. S. Tarasov, D. A. Bedarev, M. V. Maximov, A. F. Tsatsulnikov, I. P. Soshnikov, P. S. Kopev, Z. I. Alferov, N. N. Ledentsov, and D. Bimberg, Electron. Lett. **35**, 1845 (1999).

[4] A. R. Kovsh, A. E. Zhukov, N. A. Maleev, S. S. Mikhrin, D. A. Livshits, Y. M. Shernyakov, M. V. Maximov, N. A. Pihtin, I. S. Tarasov, V. M. Ustinov, Z. I. Alferov, J. S. Wang, L. Wei, G. Lin, J. Y. Chi, N. N. Ledentsov, and D. Bimberg, Microelectron. J. **34**, 491 (2003).

[5] S. A. Blokhin, N. A. Maleev, A. G. Kuzmenkov, A. V. Sakharov, M. M. Kulagina, Y. M. Shernyakov, I. I. Novikov, M. V. Maximov, V. M. Ustinov, A. R. Kovsh, S. S. Mikhrin, N. N. Ledentsov, G. Lin, and J. Y. Chi, IEEE J. Quantum Electron. **42**, 851 (2006).

[6] F. Hopfer, A. Mutig, M. Kuntz, G. Fiol, D. Bimberg, N. N. Ledentsov, V. A. Shchukin, S. S. Mikhrin, D. L. Livshits, I. L. Krestnikov, A. R. Kovsh, N. D. Zakharov, and P. Werner, Appl. Phys. Lett. **89**, 141106 (2006).

[7] A. G. Kuzmenkov, V. M. Ustinov, G. S. Sokolovskii, N. A. Maleev, S. A. Blokhin, A. G. Deryagin, S. V. Chumak, A. S. Shulenkov, S. S. Mikhrin, A. R. Kovsh, A. D. McRobbie, W. Sibbett, M. A. Cataluna, and E. U. Rafailov, Appl. Phys. Lett. **91**, 121106 (2007).

[8] L. V. Asryan, M. Grundmann, N. N. Ledentsov, O. Stier, R. A. Suris, and D. Bimberg, J. Appl. Phys. **90**, 1666 (2001).

[9] Z. C. Xu, Y. T. Zhang, J. M. Hvam, J. J. Xu, X. S. Chen, and W. Lu, Appl. Phys. Lett. **89**, 013113 (2006).

[10] Z. C. Xu, K. Leosson, D. Birkedal, V. Lyssenko, J. M. Hvam, and J. Sadowski, Nanotechnology **14**, 1259 (2003).

[11] Z. C. Xu, D. Birkedal, M. Juhl, and J. M. Hvam, Appl. Phys. Lett. **85**, 3259 (2004).

[12] Z. C. Xu, D. Birkedal, J. M. Hvam, Z. Y. Zhao, Y. M. Liu, K. T. Yang, A. Kanjilal, and J. Sadowski, Appl. Phys. Lett. **82**, 3859 (2003).

[13] Y. P. Varshini, Physica (Amsterdam) **34**, 149 (1967).

[14] G. Bacher, C. Hartmann, H. Schweizer, T. Held, G. Mahler, and H. Nickel, Phys. Rev. B **47**, 9545 (1993).

[15] S. Marcinkevicius and R. Leon, Phys. Rev. B **59**, 4630 (1999).

[16] 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).

[17] D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, New York, 1999).

[18] Z. C. Xu, Y. T. Zhang, and J. M. Hvam, Nanotechnology **18**, 325401 (2007).

[19] G. Bacher, H. Scheizer, J. Kovac, A. Forchel, H. Nickel, W. Schlapp, and R. Losch, Phys. Rev. B **52**, 1493 (1995).

[20] J. Hours, P. Senellart, E. Peter, A. Cavanna, and J. Bloch, Phys. Rev. B **71**, 161306 (2005).

[21] L. C. Andreani, G. Panzarini, and J.-M. Gérard, Phys. Rev. B **60**, 13276 (1999).

[22] H. Fu, L.-W. Wang, and A. Zunger, Phys. Rev. B **59**, 5568 (1999).