Widely tunable intersubband energy spacing of self-assembled InAs/GaAs quantum dots due to interface intermixing

Wang, X. C.; Xu, S. J.; Chua, S. J.; Zhang, Zi-Hui; Fan, Weijun; Wang, C. H.; Jiang, J.; Xie, X. G.

1999

Wang, X. C., Xu, S. J., Chua, S. J., Zhang, Z.-H., Fan, W., Wang, C. H., Jiang, J., & Xie, X. G. (1999). Widely tunable intersubband energy spacing of self-assembled InAs/GaAs quantum dots due to interface intermixing. Journal of applied physics, 86(5), 2687.

https://hdl.handle.net/10356/99690

https://doi.org/10.1063/1.371111

Widely tunable intersubband energy spacing of self-assembled InAs/GaAs quantum dots due to interface intermixing

X. C. Wang
Department of Electrical Engineering, Centre for Optoelectronics, National University of Singapore, Singapore 119260, Singapore

S. J. Xu[a] and S. J. Chua[b]
Institute of Materials Research and Engineering, National University of Singapore, Singapore 119260, Singapore

Z. H. Zhang
Department of Electrical Engineering, Centre for Optoelectronics, National University of Singapore, Singapore 119260, Singapore

W. J. Fan, C. H. Wang, J. Jiang, and X. G. Xie
MBE Technology Pte. Ltd., 14 Science Park Drive, No. 04-03 The Maxwell, Singapore Science Park, Singapore 118226, Singapore

(Received 22 December 1998; accepted for publication 22 May 1999)

In this article, we showed the significant reduction of the energy spacing between ground state and excited state emissions from InAs/GaAs quantum dots (QDs) due to interface interdiffusion induced by thermal treatment. In addition, the strong narrowing of the luminescence linewidth of the ground state and excited state emissions from the InAs dot layers for the annealed samples indicates an improvement of the size distribution of the QDs. Large blueshift of the energy positions of both emissions was also observed. High resolution x-ray diffraction experiments give strong evidence of the interface atom interdiffusion in the annealed samples. This work shows the ability to tune the wavelength for applications like infrared detectors and lasers based on intrasubband transitions of self-assembled QDs. © *1999 American Institute of Physics.* [S0021-8979(99)05017-3]

I. INTRODUCTION

Self-assembled quantum dots (QDs) directly obtained via the so-called Stranski–Krastanow growth mode are of great interest due to their fundamental physics and potential device applications. Among them, InAs/GaAs QD is a representative system, which has been extensively investigated both experimentally[1–6] and theoretically.[6–10] However, the electronic structure of InAs dots still remains an open problem. Indeed, the shape and size of the QDs as well as the strain distribution in and around them, which strongly affect the electronic structure, are very difficult to be accurately determined. Currently, most studies show that there are several bound levels rather than one bound state in the conduction band of InAs dots. Recently, intersubband transitions within the conduction band of the QDs have been demonstrated with different techniques,[11–16] and the QD infrared photodetectors based on such transitions have been reported.[17–19]

Although the defect-free dots can be formed in lattice mismatched heterostructures by the SK growth mode, inevitable size distribution in the dots results in inhomogeneous broadening both for the photoluminescence (PL) emission from the interband transitions and infrared absorption due to the intersubband transitions in the dots. Therefore, further developments in the use of QD structures for devices such as lasers and detectors depend on the achievement of tunability of dot shape and sizes in order to both control the wavelength and reduce the linewidth of the luminescence. Recently, the powerful capability of postgrowth thermal annealing in adjusting the shape and size and thus optical properties of self-assembled QDs has been shown by Leon *et al.*,[20] Malik *et al.*,[21] and the authors.[22] The main achievements of the above work are the significant blueshift and narrowing of the PL peak due to interband transition between electron and hole ground states of the dots. Large blueshift of the PL peak is attributed to strong intermixing at the interface between dot and barrier layers. Significant narrowing of the PL peak is the indicative of great improvement of the dot size dispersion. Very recently, Leon *et al.*[23] reported the first demonstration of the tunability for the intersubband spacings of the InGaAs/GaAs QDs through the interfacial composition disordering induced by thermal treatment. They showed that the energy spacing ΔE_{2-1} between the ground level and the first excited level of electrons could be reduced from 49 to 25 meV. This important finding gives a range in tunability for applications such as QD infrared photodetectors and QD lasers based on intersubband transitions. Here we show that the energy spacing between the ground state and excited state emissions of the InAs/GaAs QDs could be widely tuned from 90.9 to 14.6 meV by interdiffusion. Furthermore, a large blueshift and significant narrowing of all PL peaks for annealed samples were observed. Besides the practical applications, these findings allow us to investigate effects of the

[a]Current address: Department of Physics, The University of Hong Kong, Pokfulam Road, Hong Kong, China, Electronic mail: sjxu@hkucc.hku.hk
[b]Also with: Department of Electrical Engineering, Center for Optoelectronics, National University of Singapore, Singapore; electronic mail: elecsj@nus.edu.sg

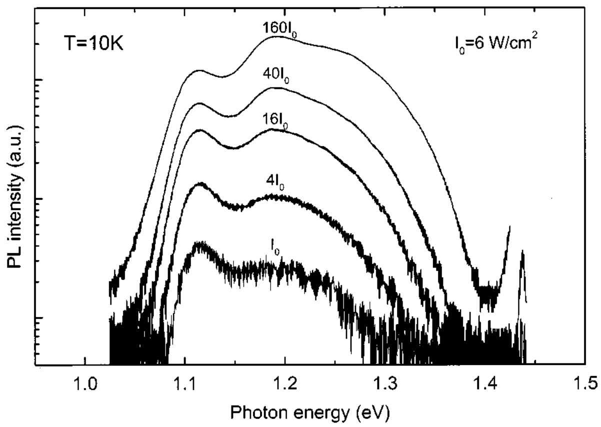

FIG. 1. 10 K PL spectra of InAs/GaAs QDs under different excitation intensities.

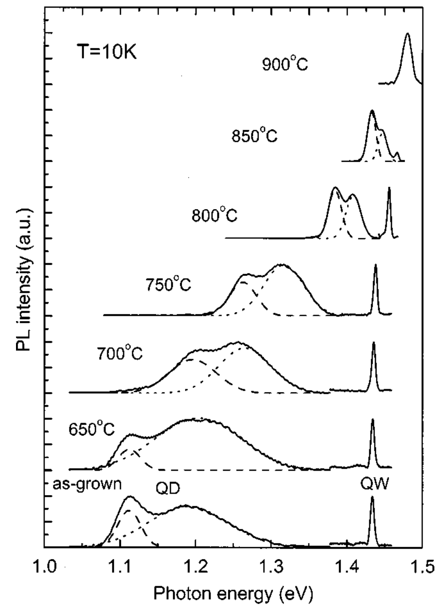

FIG. 2. 10 K PL spectra for as-grown and annealed InAs/GaAs QDs samples at different annealing temperatures. Dash-line and dot-line are Gaussian fitted curves for the ground state emission and for the excited emission, respectively.

shape and size of the InAs QDs and the strain distribution in them on their electronic structure which still remain an open problem. In particular, the wide tunability of the energy spacings between the electronic ground and excited levels of the self-assembled QDs provide a good opportunity to study some fundamental physical problems such as electron–phonon interaction in semiconductor QDs.

II. EXPERIMENT

The InAs/GaAs QDs studied in the present work were grown by MBE on semi-insulating GaAs (100) substrate. The growth order of the whole structure is a 300 nm thick GaAs buffer layer, a 3 nm $In_{0.17}Ga_{0.83}As$ layer as a reference quantum well (QW), a 100 nm GaAs barrier layer, a 10 cycled stack of 6 Å (about 1.7 monolayers thick) InAs QDs layer and 50 nm GaAs barrier layer, and finally a 20 nm thick GaAs cap layer. About 200 nm thick SiO_x films were deposited on the samples, cut from the central region of the wafer, by the plasma enhanced chemical vapor deposition technique. After deposition of the SiO_x films, the samples were subjected to rapid thermal annealing (RTA) in nitrogen ambient at temperatures ranging from 650 to 900 °C for 50 s. Before PL measurement, the SiO_x films were removed by using buffered HF solution. The PL measurement system was described previously.[22]

III. RESULTS AND DISCUSSION

Figure 1 shows the semilogarithmic PL spectra from the as-grown InAs/GaAs QD sample as a function of excitation power. There are at least two emission peaks from the sample under higher excitation power. The peak at higher energy becomes more prominent with increasing excitation intensity. These two peaks correspond to the ground state emission and the first excited state emission of the QDs.[3,4,8–10]

Figure 2 shows the 10 K PL spectra measured from the samples annealed at different temperatures. The broad and doublet luminescence peak at lower energy is from the InAs dots, while the narrow one at the higher energy is from the reference $In_{0.17}Ga_{0.83}As$ QW. For clarity, all spectra in Fig. 2 were normalized. Figure 2 also shows a nearly perfect line shape fit with two Gaussians for each spectrum except for the sample annealed at 900 °C where the two peaks have merged into a single peak. The dash line peak is for ground state emission and the dot line peak is for excited state emission. From Fig. 2 it can be seen that with increasing annealing temperature the energy spacing between the ground state and first excited state emissions gradually reduces from 90.9 to 14.6 meV. For the 900 °C-annealed sample, the PL spectrum no longer has a doublet-peak feature that implies the disappearance of the island structure in the dot layer. Our transmission electron microscopy (TEM) observation demonstrated that RTA at too high temperatures could transform the dot layer into a QW-like structure due to strong interface atom intermixing.[22] Besides the significant shrinking of the energy spacing between the ground state and the excited state emissions, a large blueshift in energy position was observed for both emissions with annealing temperature, which is strong evidence of the interface atom interdiffusion. For the reference QW case, the PL peak displays much smaller blueshift with temperature. Obviously, there is great difference between the effect of RTA for QD and QW structures. It should be noted that unlike the case of the QW structure, there are three ''contact'' interfaces between dot and barrier materials. The interface atoms between dot and barrier, therefore, can interdiffuse along both directions, parallel and vertical to the growth direction. Consequently, RTA has a much more significant effect for QD than for QW. Another prominent effect of RTA for self-assembled QDs is the significant narrowing of the PL linewidth with increasing annealing temperatures. This can be attributed to great improvement of the natural size distribution of self-assembled QDs.[20–23]

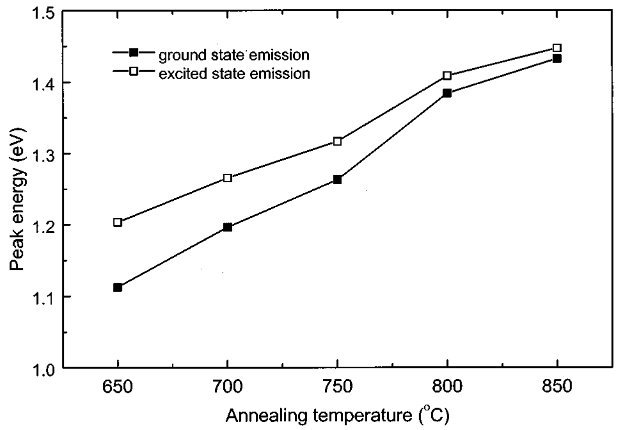

FIG. 3. Annealing temperature dependence of the PL peak position energy of the ground state and excited state emissions. The peak energetic positions of the two emissions were determined through Gaussian fitting.

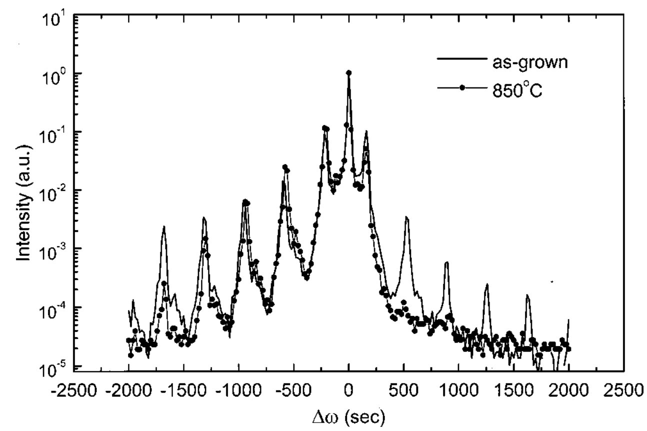

FIG. 4. Measured HRXRD rocking curves of the as-grown and 850 °C annealed InAs/GaAs QDs samples.

We next discuss the change of the energy spacing between the ground state emission and the first excited state emission of the InAs/GaAs QDs with annealing temperature. Figure 3 shows the annealing temperature dependence of the PL peak position of the PL spectra for the ground state and excited state emissions. It can be seen that the energy separation between the ground and excited state emissions reduces significantly with increasing RTA temperature. Generally, both the size and compositional changes of the QDs are thought to be responsible for the blueshift and the linewidth narrowing of their PL peaks.[20–23] The interdiffusion of the In and Ga atoms at the interface between the QD and the GaAs barrier results in a change in the size and the composition of the QDs during annealing. Although there are several different views on the shape of InAs self-assembled QDs, in particular, for the buried InAs dots, almost all available theoretical calculations[6–10] are based on the assumption of the square-based pyramidal-shaped InAs QDs. Due to existence of the strong strain around the boundary between dots and capping layer, it is difficult to accurately determine the shape of the dots even using the most advanced (TEM) technique.[24] Available experimental results show that the buried islands may adopt different shapes such as lens, pyramids with different facets, and truncated pyramids. For these geometries, an aspect ratio between the height h and the base length b can be used to characterize them. When the InAs/GaAs dot samples were subjected to RTA, In–Ga interdiffusion may make the base length of the InAs dots become larger due to anisotropic strain distribution at the interface.[8–10] The aspect ratio of the InAs dots becomes smaller with increasing RTA temperature. That is, the InAs islands after annealing become ''flatter.'' According to the valence-force-field theory of Keating[25] and Martin,[26] the strain tensor strongly depends on the aspect ratio of InAs dots. Several different models have been developed to calculate the electronic structure of the square-based pyramidal-shaped InAs-GaAs dot.[6–10] In particular, using a pseudopotential plane-wave approach, Zunger *et al.* have very recently calculated the electronic structure of pyramidal InAs dots embedded in a GaAs matrix, for a few aspect ratios. They found that the energy difference between the electron ground state and the first excited state decreases as the base length b increases. Their theoretical finding is qualitatively in agreement with our experimental observation. Other theoretical work[8,9] also predicted the same tendency. Therefore, we attribute the reduction of the energy spacing between the ground state and excited state emissions of the InAs/GaAs QDs with annealing temperature to the aspect ratio change of the dots.

The high resolution x-ray diffraction (HRXRD) technique has been developed into a powerful tool for the nondestructive investigation of epilayers of heterostructures and superlattices. Information on interface interdiffusion and intermixing in heterostructures and superlattices can be also obtained from HRXRD measurements. Figure 4 shows HRXRD rocking curves of the as-grown sample and samples annealed at 850 °C. The HRXRD experiments were performed with a Bede Scientific D^3 system. The clear satellite peaks can be seen in both curves, which indicate good periodicity of the samples. However, there is an obvious difference between both curves. The intensities of the higher order satellite peaks are strongly reduced for the annealed samples. This is also strong evidence of the interface interdiffusion in periodic heterostructure systems.[27] Unfortunately, so far a theoretical model considering interface atoms interdiffusion and strain distribution is not available to simulate x-ray diffraction curves of self-assembled QDs.

IV. SUMMARY

In summary, we have studied the effect of RTA on the optical properties of the self-assembled InAs/GaAs QDs by using PL techniques. Significant reduction of the energy spacing between ground state and excited state emissions was observed. It is attributed to the enhanced interdiffusion of the atoms at the interfaces between InAs dots and GaAs barrier layers and hence changes in aspect ratio of the QDs. HRXRD experiments show strong evidence of the interface atoms interdiffusion. Our results show that postgrowth RTA can be used to widely tune the intersubband energy spacing between the ground state and the excited state of the InAs/

GaAs QDs. This gives a range in wavelength tunability for applications like infrared detectors and lasers based on intersubband transitions.

[1] D. Leonard, M. Krishnamurthy, C. M. Reaves, S. P. Denbaars, and P. M. Petroff, Appl. Phys. Lett. **63**, 3203 (1993).
[2] D. Leonard, K. Pond, and P. M. Petroff, Phys. Rev. B **50**, 11687 (1994).
[3] F. Adler, M. Geiger, A. Bauknecht, F. Scholz, H. Schweizer, M. H. Pilkuhn, B. Ohnesorge, and A. Forchel, J. Appl. Phys. **80**, 4019 (1996).
[4] K. H. Schmidt, G. Medeiros-Ribeiro, M. Oestreich, P. M. Petroff, and G. H. Döhler, Phys. Rev. B **54**, 11346 (1996).
[5] M. Grundmann, N. N. Ledentsov, O. Stier, D. Bimberg, V. M. Ustinov, P. S. Kopev, and Zh. I. Alferov, Appl. Phys. Lett. **68**, 979 (1996).
[6] M. Grundmann, O. Stier, and D. Bimberg, Phys. Rev. B **52**, 11969 (1995).
[7] J.-Y. Marzin and G. Bastard, Solid State Commun. **92**, 437 (1994).
[8] M. A. Cusack, P. R. Briddon, and M. Jaros, Phys. Rev. B **54**, R2300 (1996); **56**, 4047 (1997).
[9] H. Jiang and J. Singh, Phys. Rev. B **56**, 4696 (1997); IEEE J. Quantum Electron. **34**, 1188 (1998).
[10] J. Kim, L.-W. Wang, and A. Zunger, Phys. Rev. B **57**, R9408 (1998).
[11] H. Drexler, D. Leonard, W. Hansen, J. P. Kotthaus, and P. M. Petroff, Phys. Rev. Lett. **73**, 2252 (1994).
[12] K. W. Berryman, S. A. Lyon, and M. Segev, Appl. Phys. Lett. **70**, 1861 (1997).
[13] J. Phillips, K. Kamath, X. Zhou, N. Chervela, and P. Bhattacharya, Appl. Phys. Lett. **71**, 2079 (1997).
[14] S. Sauvage, P. Boucaud, F. H. Julien, J.-M. Gérard, and J.-Y. Marzin, J. Appl. Phys. **82**, 3396 (1997).
[15] S. Sauvage, P. Boucaud, F. H. Julien, J.-M. Gérard, and V. Thierry-Mieg, Appl. Phys. Lett. **71**, 2785 (1997).
[16] S. J. Chua, S. J. Xu, X. H. Zhang, X. C. Wang, T. Mei, W. J. Fan, C. H. Wang, J. Jiang, and X. G. Xie, Appl. Phys. Lett. **73**, 1997 (1998).
[17] D. Pan, E. Towe, and S. Kennerly, Appl. Phys. Lett. **73**, 1937 (1998).
[18] S. Maimon, E. Finkman, G. Bahir, S. E. Schacham, J. M. Garcia, and P. M. Petroff, Appl. Phys. Lett. **73**, 2003 (1998).
[19] S. J. Xu *et al.*, Appl. Phys. Lett. **73**, 3153 (1998).
[20] R. Leon, Y. Kim, C. Jagadish, M. Gal, J. Zou, and D. J. H. Cockayne, Appl. Phys. Lett. **69**, 1888 (1996); R. Leon, D. R. M. Williams, J. Krueger, E. R. Weber, and M. R. Melloch, Phys. Rev. B **56**, R4336 (1997).
[21] S. Malik, C. Roberts, R. Murray, and M. Pate, Appl. Phys. Lett. **71**, 1987 (1997).
[22] S. J. Xu, X. C. Wang, S. J. Chua, C. H. Wang, W. J. Fan, J. Jiang, and X. G. Xie, Appl. Phys. Lett. **72**, 3335 (1998).
[23] R. Leon, S. Fafard, P. G. Piva, S. Ruvimov, and Z. Liliental-Weber, Phys. Rev. B **58**, R4262 (1998).
[24] X. Z. Liao, J. Zou, X. F. Duan, J. J. H. Cockayne, R. Leon, and C. Lobo, Phys. Rev. B **58**, R4235 (1998).
[25] P. N. Keating, Phys. Rev. **145**, 637 (1966).
[26] R. M. Martin, Phys. Rev. B **1**, 4005 (1969).
[27] A. Krost, G. Bauer, and J. Woitok, in *Optical Characterization of Epitaxial Semiconductor Layers*, edited by G. Bauer and W. Richter (Springer, Berlin, 1996), Chap. 6, p. 346.