Selective control of self-organized $In_{0.5}Ga_{0.5}As/GaAs$ quantum dot properties: Quantum dot intermixing

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Selective postgrowth control of the photoluminescence (PL) wavelength has been demonstrated for a single layer self-organized $In_{0.5}Ga_{0.5}As/GaAs$ quantum dot (QD) structure. This was achieved by rapid thermal processing of dots using different dielectric caps. Selective band gap shifts of over 100 meV were obtained between samples capped with sputtered and plasma enhanced silica deposition, with the band gap shift under regions covered with plasma enhanced chemical vapor deposition SiO_2 less than 70 meV. The effects of different caps on the PL linewidth were also observed. The differential band gap shift will enable the integration of passive and active devices in QD systems. © *2000 American Institute of Physics.* [S0021-8979(00)01721-7]

I. INTRODUCTION

Quantum dots (QDs) are currently a topic of extensive research[1] due to interest in the fundamental physics of three-dimensional (3D) quantum confinement, together with the novel device functionality that they can provide. Self-organized growth following the Stranski–Krastanov mechanism is, at present, the most developed technique of realizing QDs in semiconductors.[2] Recently, substantial reduction in the threshold current density of QD lasers and lower temperature sensitivity of the threshold current have been achieved,[3] setting the scene for fabrication of practical devices containing QD structures.[3,4] Integrating QD laser diodes with other functional devices on the same chip is highly desirable, and allows further benefit from 3D confinement functionality to be realized in devices such as modulators and amplifiers. A simple and low-cost technology, such as compositional intermixing, can be an attractive means of achieving integration in quantum-confined heterostructures.[5] The initial studies of the thermal treatment of QD structures were undertaken to improve the subsequent growth quality of GaAs cap layers and AlGaAs cladding layers on top of the QD layers.[6,7] These articles also revealed that it is possible to retain the 3D confinement of dots after high temperature annealing, suggesting that postgrowth control of the band gap can be achieved in QD structures. To our knowledge, no demonstration has yet been reported for spatial control over the band gap of the QD layers. Selective control of the band gap of laser heterostructures is necessary for useful monolithic photonic integrated circuits (PICs), therefore techniques originally developed for quantum well intermixing (QWI) are investigated in this work as means of achieving QD intermixing (QDI).

Impurity free vacancy disordering (IFVD) utilizes Ga outdiffusion into dielectric caps at elevated temperatures to create group III point defects.[5,8] Since its discovery,[8] IFVD has been widely used to fabricate optoelectronic integrated circuits and PICs.[9] The temperatures at which IFVD occurs in GaAs/AlGaAs QWs are around 900 °C. Although reasonable for unstrained QW structures, this temperature range may cause relaxation of highly strained QD layers, and hence, jeopardize device performance. Recently, we reported a technique that enables selective QWI to be realized at substantially lower temperatures than those used in IFVD. In this technique the silica is deposited by sputtering, during which point defects are created at the surface of the semiconductor.[10] It is believed that the defects, as well as the dielectric cap, play a role in the QWI that takes place underneath the regions where the sputtered SiO_2 cap is in direct contact with the semiconductor surface. It has been shown that QWI can be inhibited using photoresist to protect the surface of semiconductor from the sputtered silica induced disordering. Typical annealing temperatures for QWI using this technique are around 700 °C. In this article we present the results of QDI using the IFVD and sputtered silica techniques.

II. EXPERIMENTS

The structure used in this work was a *p-i-n* separate confinement laser heterostructure grown by molecular beam epitaxy on a (001) Si-doped GaAs substrate. It contained a single layer of self-organized $In_{0.5}As_{0.5}As/GaAs$ QDs, sandwiched between two 1.2 μm $Al_{0.3}Ga_{0.7}As$ cladding layers as shown in Table I. The structure was grown at 600 °C, except for the 4 monolayers (ML) of $In_{0.5}As_{0.5}As$ QDs and the subsequent 10 nm GaAs cap, which were grown at 485 °C with an arsenic pressure of $2-3\times10^{-6}$ Torr. A 0.6-μm-thick GaAs *p*-contact layer was grown on top of the laser structure. Samples undergoing the IFVD process were capped with 200 nm of plasma enhanced chemical vapor deposition (PECVD) SiO_2,[5] while those undergoing the sputtering process were capped with 200 nm of sputtered SiO_2.[10] Samples annealed without any dielectric caps were also studied for comparison. The samples were annealed epilayer down on a fresh piece of GaAs at different temperatures for 60 s by rapid thermal processing (RTP). After annealing, the SiO_2 cap was removed by wet etching in buffered HF for 60 s. The heavily

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TABLE I. Schematic of single layer self-organized $In_{0.5}Ga_{0.5}As/GaAs$ QD laser heterostructure.

Contact	p^+ GaAs	0.6 μm
Graded	p-$Al_{0.3\rightarrow 0}$	0.1 μm
Upper cladding	p-$Al_{0.3}Ga_{0.7}As$	1.2 μm
Barrier	GaAs	95 nm
Single layer QDs	$In_{0.5}Ga_{0.5}As$	4 ML
Barrier	GaAs	95 nm
Lower cladding	n-$Al_{0.3}Ga_{0.7}As$	1.2 μm
Graded	n-$Al_{0\rightarrow 0.3}$	0.1 μm
Buffer	n-GaAs	0.3 μm
Substrate	n-GaAs (100)	

doped thick GaAs p-contact layer and part of the upper AlGaAs cladding layer were removed using a 1:8:40 mixture of H_2SO_4:H_2O_2:H_2O at room temperature. Photoluminescence (PL) measurements were carried out at the temperature of 10 K using an Ar^+ laser ($\lambda = 514$ nm) and liquid nitrogen cooled germanium detector.

III. RESULTS AND DISCUSSION

Figure 1 shows the band gap shifts obtained by different intermixing techniques as a function of temperature to demonstrate the possibility of selective intermixing in QDs. A blueshift, of up to 190 meV, in the PL peak emission was observed accompanied by narrowing of inhomogeneous linewidth from 80 meV for the as-grown QD sample to 37 meV when annealed with a sputtered SiO_2 cap at 725 °C. Samples annealed with PECVD SiO_2 caps exhibited a maximum blueshift of 228 meV and a minimum linewidth of 27 meV at 950 °C, as shown in Fig. 2. Similar results on QDI by IFVD have also been reported by other authors.[11] QDs annealed without any dielectric cap showed band gap shifts of up to 75 meV at 950 °C, which are smaller than the band gap shifts obtained using the caps. In contrast to the caps, a broadening of the PL linewidth was observed upon annealing (Fig. 2). In all cases, the energy shift was approximately linear over a wide range of annealing temperatures, as can be seen in Fig. 1.

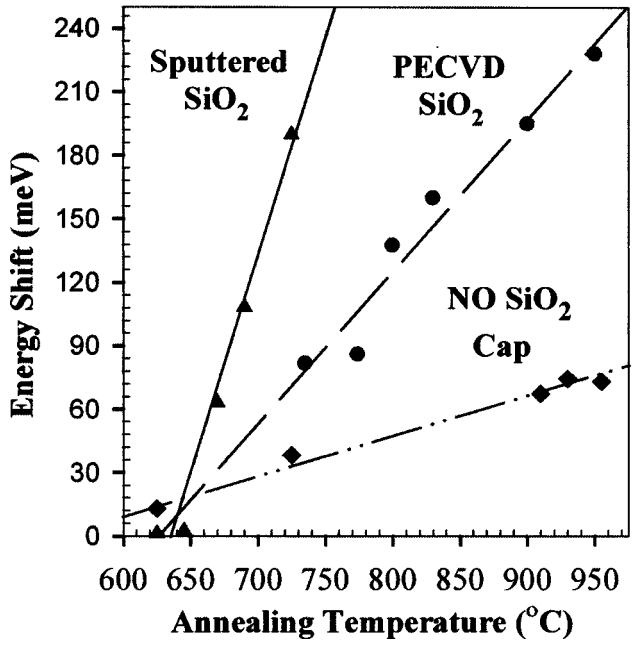

FIG. 1. QD band gap shift as a function of annealing temperature for the different intermixing techniques.

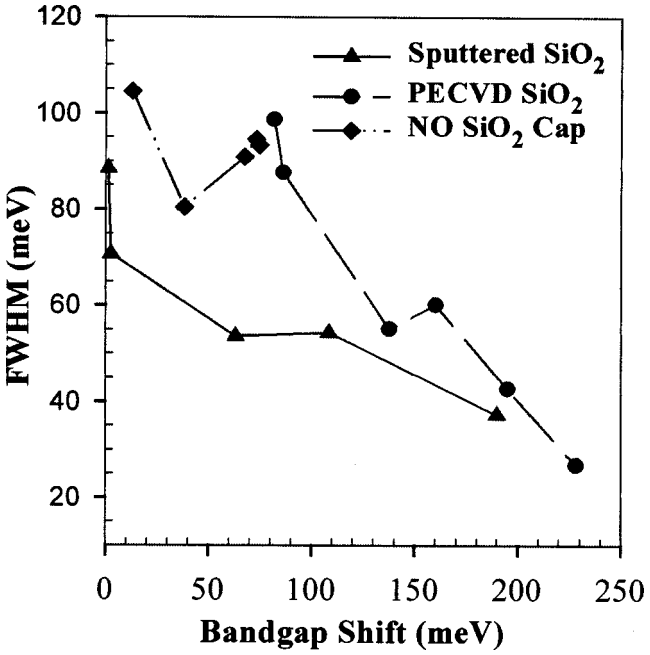

FIG. 2. QD emission linewidth vs band gap shift using different intermixing techniques.

Figure 2 shows the PL linewidth versus band gap shift in the QDs obtained by different intermixing techniques. The inhomogeneous broadening of the QD PL spectrum is reduced significantly due to an effective increase in the dot size for both the IFVD and sputtered SiO_2 processes for annealing temperature ranges where In–Ga interdiffusion across the dot-barrier interface promotes a large blueshift induced by group III point defects.[7,11] However at relatively lower annealing temperatures, where no substantial intermixing is taking place,[6,12] the PL linewidth of annealed dots increases compared to as-grown QDs for both the techniques. An interesting effect seen in the samples annealed with no silica cap is that the emission linewidth increases as a function of the extent of intermixing (Fig. 2). The reasons for such behavior are not yet understood. Thermal interdiffusion could be the reason for broadening of the PL linewidth in QDs without any cap as predicted recently utilizing a simplified model based on Fickian interdiffusion of the dot and barrier material.[12] It is also known that annealing without dielectric caps results in the loss of As from the sample surface, hence, generating a large number of group V vacancies.[13] These observations suggest that, although As outdiffusion (and hence, the generation of group V point defects) does not have a great impact on intermixing, it may cause broadening of the PL linewidth from the dots. Conversely, when Ga outdiffusion, and hence, group III point defect generation, is used to control the intermixing of the QDs, narrowing in the emission linewidth from the dots is achieved. Although further systematic studies are being car-

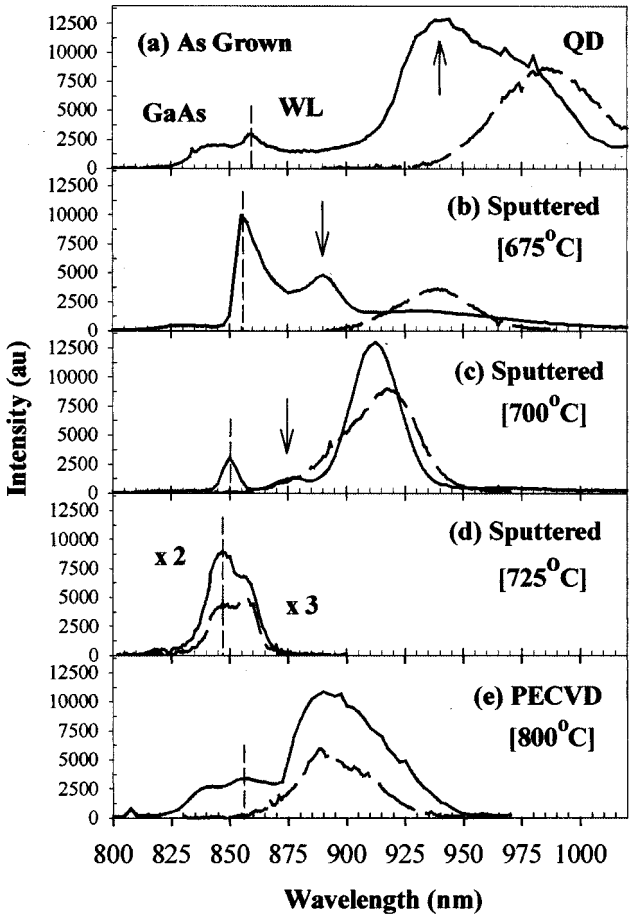

FIG. 3. 10 K PL spectra of $In_{0.5}Ga_{0.5}As/GaAs$ QDs under different operating conditions (a) as-grown dots; (b), (c) and (d) QDs capped with sputtered SiO_2 after annealing at temperatures of 675, 700, and 725 °C, respectively, for 60 s; and (e) QDs capped with PECVD SiO_2 and annealed at a temperature of 800 °C. The dashed and solid curves are for low and high excitation power density, respectively. The transitions from higher subbands are indicated by arrows and the WL is marked by a dotted line.

ried out to explain the experimental results, comprehensive modeling tools for the effect of strain and compositional change on the QD energy levels are needed to fully analyze the behavior.

The band filling effect[11,14] was investigated by pumping the samples at low and high optical power densities to understand the nature of the QD confinement potential after high temperature annealing. Figures 3(a)–3(d) shows the PL spectra of QDs capped with sputtered SiO_2 and annealed at different temperatures as well as those of the as-grown sample for comparison. The PL spectra of dots annealed at 800 °C using the IFVD technique are also depicted in Fig. 3(e) for comparison. The dashed curves represent luminescence measured from as-grown and annealed dots utilizing different intermixing techniques at low power excitation and mainly indicate ground state recombination. The solid curves show the PL signals from as-grown and annealed dots at high excitation power density. The PL peaks shown by arrows are identified as the excited dot states and the peaks marked by dotted lines are attributed to the InGaAs wetting layer (WL). The as-grown dots had a ground state PL peak at a wavelength of 985 nm, with a full width at half maximum (FWHM) of about 80 meV. At high excitation, the PL spectrum of the as-grown sample contains three distinct peaks at 940, 859, and 840 nm, respectively. The PL peak at 940 nm is due to emission from an excited state with an energy 60 meV above the QD ground state. The peak at 859 nm, which is 185 meV above the dot ground state, could originate from the WL. It is known from recent theoretical and experimental observations[14,15] that the energy difference between the QD ground state and WL is in the range of 175 meV for the type of dot structures studied here. The third peak at 840 nm is from the GaAs barrier or substrate.

A similar pattern of luminescence lines was also observed from the annealed dots, with a blueshift including a reduction of the PL peak separations. The WL transitions in the dots annealed at higher temperatures [see, e.g., Fig. 3(d)] were not well resolved in the PL spectra, as reported earlier by Leon *et al.*[7] The WL peak only shifts slightly, from 859 nm in as-grown QDs to 847 nm (i.e., around 20 meV) in samples capped with sputtered silica and annealed at 725 °C. The linewidth of the WL emission broadens at higher excitation as can be seen from Fig. 3(d)–3(e). In addition, at higher annealing temperatures, the PL intensity is greatly reduced implying degradation in material quality.

The shallower confining potential due to compositional disordering in dot layers and an effective increase in the dot size after thermal annealing induce a large blueshift in the transitions and reduce the separation of the energy subbands of QDs.[11,12] The rapid refilling of higher subbands as well as the WL is, therefore, expected to be enhanced in QDs due to the decrease in the carrier confinement energy. The composition of the WL is not well known and previous work[11] suggested that the WL could be an InGaAs alloy. Our results show that the compositional change in the WL is relatively small resulting in a small energy shift, in agreement with other reports.[6,11] Further investigations (such as photoluminescence excitation (PLE) measurements to identify clearly the different transition levels) are necessary to find out the degree of 3D confinement in annealed QDs.

By annealing without dielectric caps it is possible to obtain differential band gap shifts, when combined with PECVD SiO_2 at 950 °C. However, RTP of highly strained structures such as self-organized QDs (~3.5% lattice mismatch in our case) in this temperature range degrades the material quality and even destroys the dots, as has been reported earlier.[6,16] QDs can be selectively intermixed in the lower temperature range of 700–725 °C, using either a combination of sputtered SiO_2 cap and no cap or PECVD SiO_2 and sputtered SiO_2 caps. The surface morphology of annealed dots without any dielectric cap is poor and so the former technique may not be suitable for active device fabrication.[11] With sputtered and PECVD SiO_2 caps, a differential shift in excess of 100 meV can be achieved by annealing at 725 °C, as can be seen in Fig. 1. The use of such a low annealing temperature would ensure good material quality for device integration, combining active QD regions and passive intermixed regions on a single wafer.

IV. SUMMARY

Selective control of the PL wavelength of single layer self-organized $In_{0.5}As_{0.5}As/GaAs$ QD laser structures has been demonstrated using IFVD and sputtered silica induced disordering techniques. A reduction in the carrier confinement energy within the dots accompanied by narrowing of the emission linewidth has been observed. The results indicate that it is possible to realize differential band gap shifts of over 100 meV, which will allow passive and active device integration in QD systems.

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[1] Y. Arakawa and H. Sakaki, Appl. Phys. Lett. **40**, 939 (1982).
[2] D. Leonard, M. Krishnamurthy, C. M. Reaves, S. P. Denbaars, and P. M. Petroff, Appl. Phys. Lett. **63**, 3203 (1993).
[3] D. Bimberg, N. Kirstaedter, N. N. Ledentsov, Zh. I. Alferov, P. S. Kop'ev, and V. M. Ustinov, IEEE J. Sel. Top. Quantum Electron. **3**, 196 (1997).
[4] P. Bhattacharya *et al.*, IEEE Trans. Electron Devices **46**, 871 (1999).
[5] A. C. Bryce, F. Camacho, P. Cusumano, and J. H. Marsh, IEEE J. Sel. Top. Quantum Electron. **3**, 885 (1997).
[6] A. O. Kosogov *et al.*, Appl. Phys. Lett. **69**, 3072 (1996).
[7] R. Leon, Y. Kim, C. Jagadish, M. Gal, J. Zou, and D. J. H. Cockayne, Appl. Phys. Lett. **69**, 1888 (1996).
[8] D. G. Deppe, L. J. Guido, N. Holonyak, Jr., K. C. Hsieh, R. D. Burnham, R. L. Thorton, and T. L. Paoli, Appl. Phys. Lett. **49**, 510 (1986).
[9] *Selected Papers on Quantum Well Intermixing* edited by E. H. Li (SPIE, Bellingham, WA, 1998).
[10] O. P. Kowalski, C. J. Hamilton, S. D. McDougall, J. H. Marsh, A. C. Bryce, C. C. Button, and J. S. Roberts, Appl. Phys. Lett. **72**, 581 (1998).
[11] S. Malik, C. Roberts, R. Murray, and M. Pate, Appl. Phys. Lett. **71**, 1987 (1997).
[12] F. Heinrichsdorff, M. Grundmann, O. Stier, A. Krost, and D. Bimberg, J. Cryst. Growth **195**, 540 (1998).
[13] K. V. Vaidyanathan, M. J. Helix, D. J. Wolford, B. G. Streetman, R. J. Blattner, and C. A. Evans, J. Electrochem. Soc. **124**, 1781 (1977).
[14] M. Grundmann, N. N. Ledentsov, O. Stier, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, and Zh. I. Alferov, Appl. Phys. Lett. **68**, 979 (1996).
[15] H. Jiang and J. Singh, Phys. Rev. B **56**, 4696 (1997).
[16] 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).