Selective manipulation of InAs quantum dot electronic states using a lateral potential confinement layer

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To further the objective of controlled manipulation of the electronic states in epitaxial island quantum dots (QDs), we introduce the notion of a lateral potential confinement layer (LPCL) whose judicious placement during island capping allows selective impact on ground and excited electron and hole states. The energy states of InAs/$In_{0.15}Ga_{0.85}As$ QDs are manipulated using 10-monolayer-thick $In_{0.15}Al_{0.25}Ga_{0.60}As$ LPCLs positioned at the bottom, upper, and top region of the QDs. The changes in the photoluminescence (PL) and PL excitation spectra reveal the nature of the electronic transitions impacted selectively through the spatial charge distributions of the states involved. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1517710]

Strain-driven In(Ga)As self-assembled quantum dots (QDs)[1] have been studied extensively for the physics and potential device applications of zero-dimensional semiconductor systems.[2] Understanding and manipulation of the electronic states of QDs are important issues for applications in electronics and optoelectronics. Tuning of the QD electronic structure can be achieved by varying QD size, QD capping layers,[3–5] or by thermal annealing.[6,7] Most such approaches affect the QD electronic structure as a whole and do not allow tuning primarily subsets of QD states. Calculations show that the charge centers of gravity of QD carrier wave functions can be located at different vertical positions inside the QD (i.e., in the growth direction).[8–10] Consequently, selective modification of the lateral confinement potential at various heights of the QDs offers a means of selectively tuning the energy levels and associated transitions. In this letter we report some results of the effect of inserting a 10 monolayer (ML)-thick $In_{0.15}Al_{0.25}Ga_{0.60}As$ layer as a lateral potential confinement layer (LPCL), positioned at different height of QDs, on the electronic states of otherwise $In_{0.15}Ga_{0.85}As$ capped InAs QDs.

All samples were grown on undoped GaAs (001) substrates by solid-source molecular beam epitaxy (MBE). InAs QDs were formed at substrate temperature of 500 °C using 2.5 ML InAs delivery at a rate of 0.054 ML/s under As_4 partial pressure of 7×10^6 Torr. Subsequently, the samples were cooled to 350 °C for the growth of capping layers deposited by migration enhanced epitaxy, which shows good optical characteristics. Such low temperature capping procedure minimizes both the degree of intermixing between InAs QDs and capping layers, and the decomposition of alloy In-(Al)GaAs capping layers. For photoluminescence (PL) and PL excitation (PLE) studies, the QD samples were excited by 514 nm Ar^+ laser and a quartz tungsten halogen lamp dispersed by 0.25 m monochromator with a triple grating turret, respectively. The emission was dispersed through 0.85 m double grating monochromator and detected with a liquid nitrogen cooled Ge detector.

In this study, InAs QDs capped by 30 ML $In_{0.15}Ga_{0.85}As$ + 140 ML GaAs (InGaAs capped sample) were used as a reference. To investigate the change in the electronic states of such QDs by LPCLs positioned at the bottom, upper, and top region of the QDs, 10 ML of the 30 ML $In_{0.15}Ga_{0.85}As$ were replaced by $In_{0.15}Al_{0.25}Ga_{0.60}As$, as shown in the inset of Fig. 1. Since the $In_{0.15}Al_{0.25}Ga_{0.60}As$ layer has the same In amount, it gives essentially the same strain relieving effect to QDs as does the $In_{0.15}Ga_{0.85}As$ layer. However, having a larger band-gap energy (~1.606 eV at 300 K)[11] than that of $In_{0.15}Ga_{0.85}As$ (~1.21 eV at 300 K),[12] it enhances the potential barrier in the lateral directions. Sample 1 has the LPCL located at the bottom of the InAs QDs. In samples 2 and 3, the LPCL is positioned 10 and 20 ML above the bottom of the QDs, respectively. Atomic force microscopy results obtained from uncapped 2.5 ML InAs QDs showed that QD density was ~400/μm^2, the average width was ~20 nm, and the average height was ~6 nm.

Figure 1 shows PL spectra of InAs QDs capped by a 170 ML GaAs layer (called GaAs capped sample), the InGaAs capped sample, and samples 1, 2, and 3. The GaAs capped

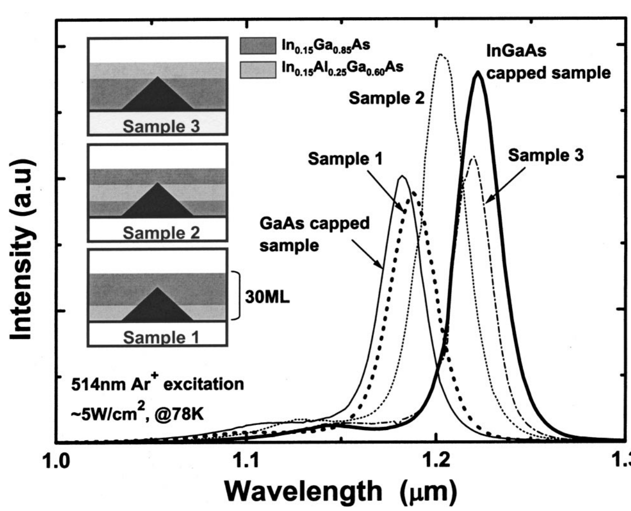

FIG. 1. PL spectra of the GaAs and the InGaAs capped reference samples, and samples 1, 2, and 3 at 78 K.

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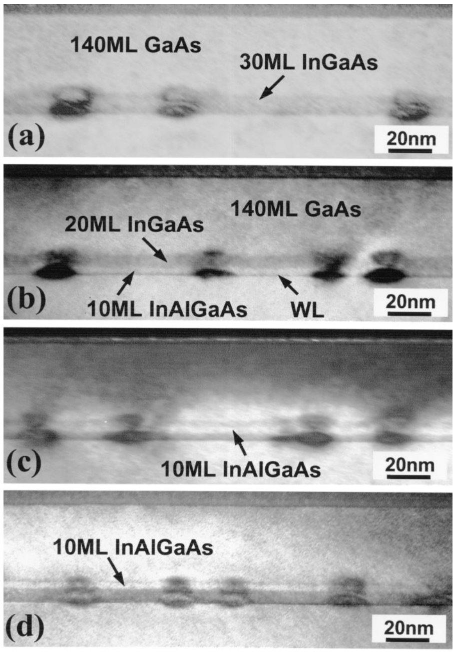


FIG. 2. Cross-sectional dark-field TEM images (**g**=002, [110] azimuth) of (a) InGaAs capped reference sample, (b) sample 1, (c) sample 2, and (d) sample 3.

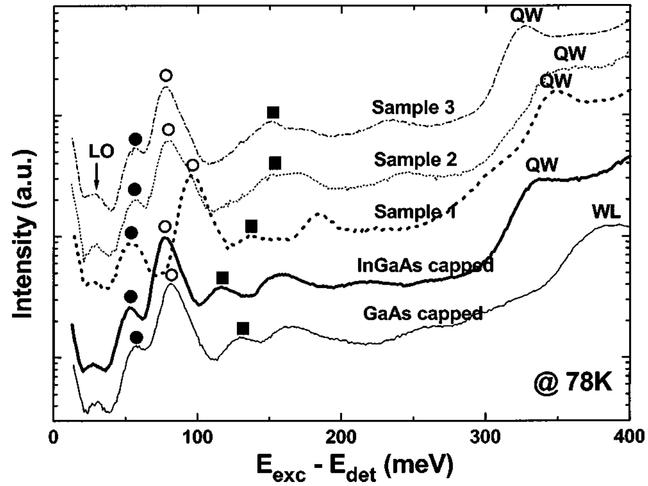
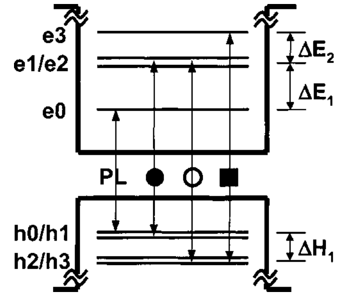


FIG. 3. PLE spectra of the GaAs and the InGaAs capped samples, and samples 1, 2, and 3 at 78 K. The detection is set at the peak value of the corresponding PL peak in each case.

sample shows a good PL spectrum peaked at 1.182 μm with high PL efficiency and a narrow full width at half maximum (FWHM) of 23 meV. As expected,[3,5] the PL peak of the InGaAs capped sample is redshifted (1.222 μm, FWHM of 23 meV) with respect to that of the GaAs capped sample because of an overall lower confinement potential in the case of InGaAs arising from lowering of the band edge discontinuities due to a combination of chemical and strain relieving effects. When the first 10 ML of $In_{0.15}Ga_{0.85}As$ layer of the InGaAs capped sample is replaced by 10 ML of $In_{0.15}Al_{0.25}Ga_{0.60}As$ (sample 1), the PL peak position (1.188 μm, FWHM of 25 meV) is almost back to that of the GaAs capped sample. Clearly, the replacement of Ga by Al has chemically enhanced the depth of the confinement potential without changing the overall strain too much, given the nearly identical atomic sizes of Al and Ga. When the 10-ML-thick $In_{0.15}Al_{0.25}Ga_{0.60}As$ layer is positioned at the upper region of QDs (sample 2), the PL spectrum is redshifted (1.203 μm, FWHM of 23 meV) with respect to that of sample 1. When the 10-ML-thick $In_{0.15}Al_{0.25}Ga_{0.60}As$ layer is located on top of the QDs (sample 3), the PL peak (1.220 μm, FWHM of 20 meV) reaches that of the InGaAs capped reference sample. It is evident that the ground state transition energy is most affected when the potential is made deeper in the base region of the QDs. This is expected since all calculations[8–10] reveal that the centers of gravity of the ground electron and hole state charge distributions are located in the lower region of the pyramidal QDs. We note parenthetically that the absence of a discernable change in the shape, intensity, and FWHM (20–25 meV) of the PL spectra suggests that the degree of change in QD size distribution is not significant among the studied samples. This is consistent with the lack of any significant change in QD size as indicated by cross-sectional transmission electron microscope (TEM) images of samples 1, 2, 3, and the InGaAs capped reference sample shown in Fig. 2. In each case, the $In_{0.15}Al_{0.25}Ga_{0.60}As$ LPCL and the corresponding $In_{0.15}Ga_{0.85}As$ layer is fairly flat, a consequence of the low temperature capping.[13] The change in electronic states of samples containing the LPCL layer can thus be discussed mainly in terms of potential confinement changes due to the chemical and strain relieving effects of $In_{0.15}Al_{0.25}Ga_{0.60}As$ and $In_{0.15}Ga_{0.85}As$ layers.

To gain insight into the behavior of the excited states of the LPCL containing QDs, systematic PLE studies were carried out. Figure 3 shows PLE spectra of the GaAs and the InGaAs capped samples, and samples 1, 2, and 3. The ~30 meV peak (marked LO) is a phonon-assisted peak, and the peaks observed above 300 meV are related to quantum wells formed by $In_{0.15}Ga_{0.85}As$ regions between the island QDs and the wetting layer formed by the ~1-ML-thick InAs regions between the island QDs in the GaAs capped sample. Compared with the GaAs capped sample, the InGaAs capped sample has smaller energy differences between the excited state transitions and smaller ground state transition energy because of lower confinement potential. Since the 30-ML-thick $In_{0.15}Ga_{0.85}As$ capping layer covers InAs QDs fully, the whole set of energy states of InGaAs capped sample are affected by the lower confinement potential, and most PLE peaks are redshifted (by less than 10 meV). Comparing the LPCL containing samples 1, 2, and 3 to the InGaAs capped sample, note that the first PLE peak (marked ●) is essentially unshifted. By contrast, when the LPCL is located at the bottom region of the QDs (sample 1), the second and third PLE peaks (marked ○ and ■) are blueshifted by ~20 meV while the energy differences between other peaks are not changed significantly compared to the InGaAs capped sample. When the LPCL is positioned at the upper and top regions of QDs (samples 2 and 3), the second PLE peak is, interestingly, back to that of the InGaAs capped sample, but the third PLE peak (marked ■) is blueshifted by ~34 meV with respect to that of the InGaAs capped sample. The large energy shifts of 20 to 34 meV of the specific PLE peaks depending on the LPCL position means that certain specific energy states are more perturbed than others. As discussed next, the observed invariance of the first PLE peak, the blueshift of the second peak in sample 1 only, and the blueshift of



(unit: meV)

	ΔH_1	ΔE_1	ΔE_2
GaAs capped	25	58	49
InGaAs capped	23	54	39
Sample 1	43	55	41
Sample 2	23	58	73
Sample 3	22	57	73

FIG. 4. Schematic of electron and hole energy levels and associated transitions of InAs QD.

the third PLE peaks in samples 1, 2, and 3 can be understood in terms of the varied impact of the LPCL ($In_{0.15}Al_{0.25}Ga_{0.60}As$ layer) on different electron and hole states depending upon the nature of their spatial charge distributions. Indeed, these shifts can be viewed as the first direct experimental probing and confirmation of the nature of the calculated charge distributions of higher (excited) hole and electron states in pyramidal QDs.

In Fig. 4, we schematically summarize the types of transitions involved suggested by the comparison of the observed PLE peak positions and the calculated transition energies available in the literature.[8,9,14,15] The first PLE peak energy (~54–58 meV) of Fig. 3 is well established to represent the energy difference of the ground electron state (e0) and the first subset of excited electron states (e1/e2).[14,15] The calculated e0 and the e1/e2 wave functions are s-like and p-like, respectively, but the heights of the charge density isosurfaces are reported to be very similar.[8–10] The observed insensitivity of the e0–e1/e2 transition (ΔE_1) to the different positions of the LPCL thus suggests that the e0 and e1/e2 energy levels are shifted almost in parallel. By contrast, the second PLE peak (marked ○) is blueshifted by ~20 meV only in sample 1 having the LPCL positioned at the bottom region of QDs. The energy difference of the first and the second PLE peaks represents the difference (ΔH_1) in excited hole energy levels (h2/h3) and the ground hole levels (h0/h1) (see Fig. 4). The hole states tend to be more confined inside dots than the electron states. A common finding[8,9] of the calculations is that the h2/h3 charge distributions have lobes located near the bottom corners of QDs. As a result, the h2/h3 states are more sensitive to the squeezing effect of the LPCL when positioned in the bottom region of QDs and contribute the ~20 meV blueshift in the e1/e2–h2/h3 transition energy. When the LPCL is located in the upper and top regions of QDs (samples 2 and 3), its effect on the h2/h3 states essentially disappears, and the second PLE peak is back to that of the InGaAs capped sample. However, in these cases, the third PLE peak (marked ■) is maximally blueshifted by ~34 meV. Attributing the third peak to the transitions involving the second subset of excited electron states (e3 or higher states) and h2/h3 hole states as indicated by (■) in Fig. 4 would be consistent with the blueshift arising from the increased lateral localization of the charge of the second subset of excited electron states if it were concentrated in the upper region of the QDs. The charge distribution of the potential electron states in the second subset calculated in Ref. 8 suggests that certain states have likely a significant part of their charge distribution in regions higher than the electron states of the first subset (e1/e2). A more quantitative explanation must await appropriate theoretical analysis.

In conclusion, we have systematically investigated the effect of a LPCL, positioned at different heights of QDs, on the electronic states of the $InAs/In_{0.15}Ga_{0.85}As$ QD system. The electron and hole ground states and the first subset of excited hole states are perturbed most effectively by the LPCL located in the bottom region of QDs, indicating that the charge center of gravity of these states resides near the QD bottom. Moreover, by positioning the LPCL in the upper region of the QDs, certain high excited electron energy levels could be tuned without significantly perturbing other states, thus indicating that these electron states have their charge distribution centers of gravity located in the upper regions of the QDs. Our findings open another path to manipulating intra- and interband transition energies for applications such as QD infrared photodetectors, lasers, and optical amplifiers.

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[1] S. Guha, A. Madhukar, and K. C. Rajkumar, Appl. Phys. Lett. **57**, 2110 (1990).
[2] D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, Chichester, 1999), and references therein.
[3] K. Nishi, H. Saito, S. Sugou, and J. S. Lee, Appl. Phys. Lett. **74**, 1111 (1999).
[4] F. Guffarth, R. Heitz, A. Schliwa, O. Stier, N. N. Ledentsov, A. R. Kovsh, V. M. Ustinov, and D. Bimberg, Phys. Rev. B **64**, 085305 (2001).
[5] E. T. Kim, Z. H. Chen, and A. Madhukar, Appl. Phys. Lett. **79**, 3341 (2001).
[6] S. Malik, C. Roberts, R. Murray, and M. Pate, Appl. Phys. Lett. **71**, 1987 (1997).
[7] R. Leon, S. Fafard, P. G. Piva, S. Ruvimov, and Z. Liliental-Weber, Phys. Rev. B **58**, R4262 (1998).
[8] O. Stier, M. Grundmann, and D. Bimberg, Phys. Rev. B **59**, 5688 (1999).
[9] S. J. Sun and Y. C. Chang, Phys. Rev. B **62**, 13631 (2000).
[10] L. W. Wang, J. Kim, and A. Zunger, Phys. Rev. B **59**, 5678 (1999).
[11] D. Olego, T. Y. Chang, E. Silberg, E. A. Caridi, and A. Pinczuk, Appl. Phys. Lett. **41**, 476 (1982).
[12] S. Adachi, J. Appl. Phys. **53**, 8775 (1982).
[13] Q. Xie, P. Chen, and A. Madhukar, Appl. Phys. Lett. **65**, 2051 (1994).
[14] I. Mukhametzhanov, Z. H. Chen, O. Baklenov, E. T. Kim, and A. Madhukar, Phys. Status Solidi B **224**, 697 (2001).
[15] R. Heitz, O. Stier, I. Mukhametzhanov, A. Madhukar, and D. Bimberg, Phys. Rev. B **62**, 11017 (2000).