Comparative study of InAs quantum dots with different InGaAs capping methods

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The authors have used cross-sectional scanning tunneling microscopy to examine strain relaxation profiles of InAs quantum dots with $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ layers overgrown by three distinct capping methods. A statistical analysis of strain relaxation profile allowed them to infer that the long wavelength emission ($>1.3\ \mu\text{m}$) of InAs quantum dots capped with sequential GaAs/InAs binary growth is mainly due to a weaker quantum confinement effect. This particular capping method is better than the traditional molecular beam epitaxy with simultaneous In/Ga/As deposition, and much better than a capping method with separated Ga deposition followed by As and InAs growth. © 2007 American Institute of Physics. [DOI: 10.1063/1.2454425]

Long wavelength III-V quantum dot (QD) lasers¹⁻³ operating at 1.3 or 1.55 μm hold great promise for future optoelectronic applications. Pure InAs QDs grown on GaAs(100) can hardly achieve long wavelength light emission due to their larger strain and smaller sizes. Consequently, several growth techniques are developed to resolve this limitation. For example, submonolayer migration enhanced deposition is used.⁴ InGaAs layers bordering quantum dots on one or both sides^{2,5,6} and metalorganic preparation of In(Ga)As (Ref. 7) have demonstrated light emission over $\sim 1.3\ \mu\text{m}$. With QDs embedded in $\text{In}_x\text{Ga}_{1-x}\text{As}$ layers, it appears to provide additional benefits of narrower photoluminescence (PL) linewidths^{3,5} and a good QD density up to $\sim 10^{11}\ \text{cm}^{-2}$. Furthermore, a recent study³ has shown that preparation sequence of the capping layer can have significant effects on the performance of QD lasers. In Ref. 3, Chang *et al.* use a high In content capped layer (9 ML $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$) and fabricate three types of QDs with the following capping sequences: (a) Type-A QDs: InAs QDs covered with sequential Ga, As, and InAs deposition; (b) type-B QDs: InAs QDs covered with alternating GaAs and InAs binary deposition; (c) type-C QDs: InAs QDs with capping layers prepared with conventional molecular beam epitaxy (MBE), i.e., simultaneous In, Ga, and As deposition. Although the 0.33 indium mole fraction used in Ref. 3 is significantly higher than typical fractions of 0.09–0.20 used in other studies,^{2,6} the type-B QDs are found to perform the best with a PL peak at $\lambda=1318\ \text{nm}$, whereas the type-A QDs are the worst with $\lambda=1267\ \text{nm}$. Since all InGaAs capping layers were formulated with identical chemical composition,³ growth kinetics of the InGaAs capping layers has critical influence on the postgrowth evolution and optoelectronic properties of InAs QDs. In this letter, we study the shape and strain profiles of the type-A, -B, and -C QDs with cross-sectional scanning tunneling microscopy (XSTM). Our study is useful as no XSTM studies have been conducted yet for InAs quantum dots with heavy In-content InGaAs capping

layers.⁸ By using an analysis procedure to separate strain profiles from the capping layers and the embedded QDs, we gathered strain profile statistics of hundreds of dots. We found that the long emission wavelength of the type-B QDs originates from a weaker confinement energy, and the inferior performance of the type-A QDs is due to both reduced In content and a stronger confinement effect.

The QDs were grown on Si-doped (100) GaAs substrates by gas-source MBE. A sample with sequentially grown type-A, type-C, and type-B QD layers was used. Each QD layer has 2.0 ML of InAs followed by a 9.0 ML $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ capping layer, grown at 485 °C. Details can be found in Ref. 3. Adjacent QD layers were separated by a 500 nm GaAs spacer grown at 485 °C for the first 6 nm, and 590 °C for the rest. The total annealing time at 590 °C for type-A, type-C, and type-B layers are thus 1.5, 1.0, and 0.5 h, respectively. To see if different thermal history alters the morphology and composition of type-A, -B, and -C QDs, we have verified that the 18 K PL spectra for separately grown QD PL samples (e.g., type-A, type-B, etc.) do not change after up to 1 h *ex situ* ultrahigh vacuum (UHV) annealing at 590 °C. Each QD PL sample has one InGaAs capped surface QD layer, a 500 nm GaAs spacer, and one buried InGaAs capped QD layer.³ Figure 1(a) inset shows that the PL peak positions, e.g., for a type-A QD PL sample, with a total 0.5 h (solid line) or 1.5 h (dashed line) annealing at 590 °C, remain unchanged. This is consistent with reports⁹ showing that QDs, once completely buried, hardly change by subsequent annealing. Thus, our stacked QD XSTM samples were valid and convenient for comparative studies, though each QD layer was subjected to different annealing time.

The XSTM was conducted with a commercial Omicron VTSPM housed in a home-built UHV system with a base pressure $<4 \times 10^{-11}$ torr. XSTM samples were first thinned down to $\sim 100\ \mu\text{m}$ and then were cleaved *in situ* at room temperature under UHV. Exceptional samples with flat terraces extending all the way to sample edges can be routinely obtained. For example, Fig. 1(a) shows a XSTM image of the three QD layers embedded in a step-free surface exceed-

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FIG. 1. (Color online) (a) XSTM image of the type-A, type-B, and type-C QD layers. Image size = $1.5 \times 1.0 \mu\text{m}^2$, sample bias $V_s = +2.3$ V, and tunneling current $I_t = 0.1$ nA. (a) Inset: 18 K PL spectra of a type-A QD PL sample before (solid line) and after (dashed line) 1 h UHV annealing at 590 °C. PL peak positions do not shift. XSTM images of individual (b) type-A, (c) type-B, and (d) type-C InAs QDs. Nanoscale inhomogeneous In distribution is more evident in the type-A and type-B dots.

ing $1.5 \mu\text{m}$ in width. This allowed us to evaluate strain profiles of many QDs without undesirable interference from steps. Closer inspection revealed that the uniformity of In distributions in the three types of QDs was different. Figures 1(b)–1(d) illustrate typical type-A, -B, and -C QDs, respectively. The type-C QD with a MBE capping layer generally shows a more uniform In distribution. This indicates alloy decomposition is enhanced when the growth of InAs and GaAs is separated.⁴

Due to the relatively high In concentration (33%) used in the capping layers, counting individual In atoms in each QD and determining its concentration was not practical. Instead, we analyzed outward strain relaxation profile of each QD. Our goal was to extract statistical average of dot density (ρ), dot width (w), dot height (h) and QD outward height (z), and volume (V) from these QD ensembles. Since the outward profile of a cleaved QD can contain both electronic and topographic contributions,¹⁰ a sample bias of +2.3 V was selected since it gave more stable long-term imaging and $\sim 90\%$ of the maximum topographic height. Figure 2 depicts a typical line profile crossing a QD (black solid line). The strain profile is due to both the QD and the InGaAs capping layer. The latter contribution is long ranged. Its relaxation profile can be obtained from contact mechanics and it writes as

$$z = C - \frac{2(1+\nu)\varepsilon_0}{\pi} \left[(x+a) \log \left| \frac{x+a}{a} \right| - (x-a) \log \left| \frac{x-a}{a} \right| \right], \quad (1)$$

where C is a base line constant, ν is Poisson ratio, ε_0 and a are strain and half width of the two-dimensional strained layer, respectively. Equation (1) is derived under normal

FIG. 2. (Color online) Line profile crossing the QD layer. The red line is the strain relaxation profile of a 5 nm $\text{In}_{0.17}\text{Ga}_{0.83}\text{As}$ quantum well, with the center position as a fitting parameter. The green line is the residual strain relaxation due to a QD. The vertical outward buckle z and the QD height h are deduced from a Lorentz fit. w and h are also depicted in Fig. 1(b).

plane stress with no shear traction. More details can be found in Ref. 11. We estimated the widths of the $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ layers directly from STM images and found them to be ~ 5 nm (2a) in all three QD layers. This was larger than the expected 2.6 nm, indicating that significant intermixing has occurred during the GaAs spacer growth. To keep total In content constant, we approximated the 2.6 nm thick $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ layer with a 5 nm thick $\text{In}_{0.17}\text{Ga}_{0.83}\text{As}$ layer. A typical fitted curve is shown as the red dashed line in Fig. 2. Subtracting this profile, we obtained a more accurate strain relaxation profile due to a QD as depicted by the lower green solid line. The QD profile usually appears symmetric along the growth direction, unlike the “inverted cone-shaped” In distribution¹² that occurs with higher temperature (500–550 °C) growth. Therefore, we fitted the QD profile by a phenomenological Lorentz function (lower blue dashed line). QD height h and outward relaxation z are taken as the full width at half maximum and peak height of the Lorentz fitting at the QD center, respectively. QD base width (w) of each QD is measured as the distance between the two points at which the QD line profile gives $\sim 5\%$ of the maximum height h . Finally, the outward volume (V) is obtained by integrating $dzdw dh$. In all, ~ 100 –200 QDs for each growth method were analyzed. Average structural parameters, $\langle h \rangle$ (nm), $\langle w \rangle$ (nm), $\langle V \rangle$ (nm³), and areal density ρ (10¹⁰/cm²), were measured. For XSTM, $\langle h \rangle$, $\langle w \rangle$, $\langle V \rangle$, ρ for type-A, -B, and -C QDs are (4.9, 30.5, 56, 1.56), (5.3, 21.7, 47.6, 4.90), and (5.0, 21.2, 50.8, 4.05), respectively. For scanning electron microscopy³ (SEM) of InGaAs-capped surface QDs, $\langle w \rangle$, ρ are (34, 2.3), (29, 7.6), and (27, 17.0) for type-A, -B, and -C QDs, respectively. Basically, XSTM and SEM give consistent results, though $\langle w \rangle$ and ρ of XSTM are significantly lower. This reflects that some QDs are not cleaved at the planes of maximum QD width. It also indicates that smaller QDs at the InGaAs/GaAs interface do not survive after the deposition of GaAs spacer (in particular, for the type-C QDs), presumably caused by Ga intermixing.

XSTM can further provide crucial insight on the vertical dot height and In concentration in QDs that is not accessible by SEM.^{10,12,13} Carrier confinement energy in a QD is dominated by its smallest dimension, i.e., its vertical height.¹⁴ Our

FIG. 3. (Color online) α plot for the type-A, -B, and -C QD ensembles [$\alpha = V/(w^2h)$, V , w , h defined in text]. The insets depict two different configurations of a QD under the cleaved plane.

data show that the average height, $\langle h \rangle$, is the largest for the type-B QDs and the smallest for the type-A QDs. This is consistent with the PL spectra in which the redshift of type-B QDs is the largest. We can also extract qualitative information, e.g., relative order of average In concentration, for the three QD types. A higher QD In concentration has two effects on its band gap ε_g , i.e., ε_g reduces due to chemical composition and increases due to higher compressive strain. The former effect is much larger than the latter¹⁵ and the net result leads to emission redshift. In Fig. 3, we define a dimensionless parameter, $\alpha = V/(w^2h)$, where V , w , h were defined earlier, and plot it for the three QD ensembles. In XSTM, some QDs have buried volume less [Fig. 3 inset (a)] or more [inset (b)] than half of their actual sizes. The trend of parameter α is that it decreases gradually as more QD volume is buried. Furthermore, α increases if the average strain of the QD ensemble increases. The above trends were confirmed in a finite element analysis (FEA) using ABAQUS.¹⁶ While we have not extracted quantitative values of the QD strain from the simplified FEA model,¹⁶ we emphasize that the order of In concentrations in the three QD types can be obtained with confidence.

In Fig. 3, the type-C QDs have the largest average value of α , and therefore the highest average In concentration. Note that the relative order of average In concentration alone is inconsistent with the PL measurements. To summarize, we found for confinement energy ε_1 , $\varepsilon_1(\text{type B}) < \varepsilon_1(\text{type C}) < \varepsilon_1(\text{type A})$; for band gap energy ε_g , $\varepsilon_g(\text{type C}) < \varepsilon_g(\text{type B}) < \varepsilon_g(\text{type A})$. Since the PL wavelength is determined by $\varepsilon = \varepsilon_g + \varepsilon_1$ and the measurements showed $\varepsilon(\text{type B}) < \varepsilon(\text{type C}) < \varepsilon(\text{type A})$, we concluded that the type-B QDs gain main PL redshift from a weaker carrier confinement, rather than from a reduction of QD band gap.

The type-A QDs are inferior in both aspects of reduced confinement and reduced band gap energy.

In summary, we have used XSTM to study InAs QDs overgrown with $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$ capping layers. We emphasized the importance of optimizing InGaAs capping layer growth sequence and found separation of GaAs and InAs growth beneficial for longer wavelength emission. By using a statistical approach to analyze strain relaxation profiles of many cleaved QDs, we clarified the relative importance of band gap reduction and confinement energy reduction associated with each particular InGaAs capping sequence.

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- ¹⁶ABAQUS/CAE, Version 6.4, <http://www.abaqus.com>, licensed by National Center for High-Performance Computing, Taiwan, R. O. C. In the FEA analysis, a square-based disk of uniform strain was embedded in a strain-free half plane and allowed to relax.