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Structure and composition profile of InAs/GaAs quantum dots capped by an InGaAs and InAlAs combination layer

Jun He[1,3], Yuan Wu[2] and Kang L Wang[1]

[1] Electrical Engineering Department, University of California at Los Angeles, Los Angeles, CA 90095, USA
[2] Material Department, University of California at Santa Barbara, Santa Barbara, CA 93117, USA

E-mail: hejun@ee.ucla.edu

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Abstract
Cross-sectional transmission electron microscopy (X-TEM) and scan-TEM are performed to study how the structural properties of InAs/GaAs quantum dots (QDs) are affected when capped by an InGaAs and InAlAs combination layer (CBL), which currently is one of the most promising active regions for a 1.3 μm QD laser. GaAs capping causes leveling of the QDs, which is suppressed by the introduction of an InGaAs and InAlAs CBL. Scan-TEM results show that the CBL significantly suppresses In segregation and In–Ga intermixing during the capping process. Therefore, the height and In concentration in the buried QDs remain larger than that of GaAs capped QDs, leading to the enhanced optical properties of InAs QDs.

(Some figures in this article are in colour only in the electronic version)

Low cost optical components that operate in the near infrared windows of fiber optic transmission (1.3 and 1.55 μm) are in high demand due to the rapid development of optical communications. GaAs-based lasers emitting at 1.3 and 1.55 μm are one of the mainstreams in semiconductor optoelectronics research, which may offer a low cost alternative to InP-based lasers operating at the same wavelength [1–4]. To date, three kinds of gain materials have been demonstrated in edge-emitting GaAs-based telecom lasers operating at 1.3 and 1.55 μm: In(Ga)As/GaAs QDs [5–7], InGaAsN quantum wells (QWs) [8, 9], and InAsSb QWs [10]. Among them, GaAs-based lasers with QD active regions are theoretically expected to have a lower threshold current density and a higher characteristic temperature T_0 because the state density of this zero-dimensional structure is a delta function. Thus tremendous work has been focused on QDs gain materials.

Typically, self-assembled InAs/GaAs QDs emit at 1 μm with a linewidth of a few tens of millielectronvolt [11–14]. Nishi *et al* in 1999, proposed the use of a combination of self-assembled QDs with a strain reducing InGaAs layer that covers the QDs being used for extending the InAs QDs wavelength emission from 1 to 1.3 μm and narrowing the linewidth photoluminescence (PL) emission to 21 meV [5]. This discovery led to an extensive study on different capping materials for InAs/GaAs QDs for 1.3 and 1.55 μm emissions, such as thin InGaAs and InAlAs CBLs [15–17] and a GaAsSb capping layer [18]. In the last few years, InAs/GaAs QDs capped with thin InAlAs and InGaAs layer(s) have been shown to be the most promising candidate for a GaAs-based laser active region, due to their optical properties of narrower linewidth and larger energy separation between the ground state and the first excited state transition, which is crucial to improve the characteristic temperature of QD lasers. In these studies [15–17, 19], the authors attributed these merits to the suppression of In and Ga atoms intermixing between the InAs/GaAs QDs and the overgrowth of the GaAs barrier layer and/or to the reduction of the strain of the dots by the introduction of the InAlAs and InGaAs strain reducing layers, as the lattice constant of the strain reducing layer is larger than that of GaAs, hence, there is a smaller lattice mismatch with InAs QDs. However, these experiments lack information about the shape and the residual height of the QDs, in particular, the suppression of In segregation, In–Ga intermixing between

[3] Author to whom any correspondence should be addressed.

Table 1. Layer details of samples A and B.

Sample	Layer	Capping	Temperature
A	1	InAs capped by CBL	470 °C
	2	InAs capped by CBL	470 °C
	3	InAs capped by CBL	470 °C
B	Reference	InAs capped by GaAs	470 °C
	1	InAs capped by GaAs	470 °C
	2	InAs capped by CBL	470 °C
	3	InAs capped by GaAs	350 °C

the thin InGaAs and InAlAs CBL capped InAs QDs and the subsequently grown GaAs layer. Therefore, the mechanism for the observed enhanced optical properties with a thin InAlAs and InGaAs CBL is still a topic of considerable debate. In this paper, we provide an insight into the structural properties and composition profiles of the InAlAs/InGaAs/InAs/GaAs QDs by using X-TEM and scanning-TEM (STEM). The effects of inserting an InGaAs and InAlAs CBL between InAs QDs and the subsequent GaAs layer on the intermixing of In–Ga are presented.

The samples were grown by solid source molecular beam epitaxy (MBE) on semi-insulating GaAs(100) substrates. After native oxide desorption at 580 °C, a buffer layer made of a short-period GaAs/AlAs superlattice followed by 1500 Å GaAs was grown at a substrate temperature of 580 °C. The temperature was then lowered to 470 °C for the deposition of InAs QDs. For PL measurement (sample A), three stacked InAs QD layers are grown at 470 °C capped by 5 nm $In_{0.2}Ga_{0.8}As$ and 3 nm $In_{0.2}Al_{0.8}As$, with a 60 nm GaAs space layer grown at 580 °C, the nominal thickness of InAs is 2.8 monolayers (MLs). For X-TEM (sample B), the structure consisted of four stacked InAs QD layers with the same nominal thickness of 2.8 ML, the QD layers are capped by 10 nm GaAs at 470 °C (reference layer and layer 1), 5 nm $In_{0.2}Ga_{0.8}As$, 3 nm $In_{0.2}Al_{0.8}As$ at 470 °C (layer 2), and 10 nm GaAs at 350 °C (layer 3), respectively, with a 60 nm GaAs space layer grown at 580 °C. Structures of sample A and B are summarized in table 1. During growth, the QD evolution was monitored by reflection high energy electron diffraction (RHEED). Formation of InAs QDs was verified by the sharp transition from streaky to spotty of the RHEED pattern. The structural and optical properties of the QDs were investigated by X-TEM, STEM, and PL spectroscopy.

The lateral size of the uncapped InAs/GaAs QDs was found to be about 30 nm and the height about 7 nm from atomic force microscopy (AFM) measurement (not shown). In figure 1, the PL spectrum of sample A is shown taken at room temperature (RT) with an excitation power of 100 mW. The emission wavelength is extended to about 1.34 μm as compared with that of the InAs/GaAs QDs capped by GaAs [7, 12]. The full width at half maximum is about 34 meV for the ground state [5, 15]. The emission of the excited states indicates a more efficient quantum confinement of the carriers in the InAs QDs capped with InAlAs and InGaAs CBL. The energy separation between the ground and the first excited radiative transition is about 95 meV. This relatively larger energy separation, together with the extended emission

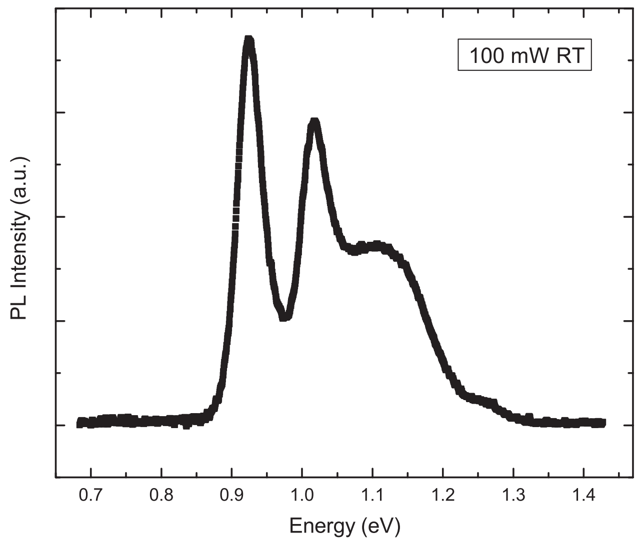

Figure 1. PL spectrum of sample A taken at RT under the pump power of 100 mW.

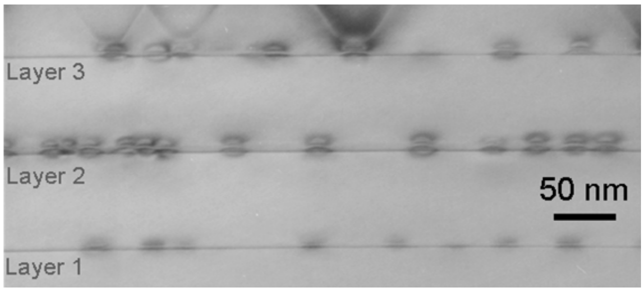

Figure 2. The bright field images recorded under $g = 002$ of sample B showing the QD layers. The growth direction is vertically upwards for the images.

wavelength compared with that of GaAs capped InAs QDs is attributed to the introduction of CBL, which is of great significance to the performance of GaAs-based QD lasers, for example by decreasing the temperature sensitivity of the lasing threshold.

The above-mentioned optical properties of capped InAs QDs with an InAlAs and InGaAs CBL could have resulted from a number of effects, such as an increased confinement potential due to the introduction of the InAlAs layer, increasing height of the QDs and/or decreasing of In–Ga intermixing. To further investigate the mechanisms governing the enhanced optical properties, the structural properties of the TEM sample were carefully studied by cross-sectional TEM. A scattering contrast study was conducted on sample B in a Tecnai T20 operated at 200 kV, while high-resolution imaging and EDX analysis were performed in a Tecnai F30U operated at 300 kV, equipped with a field emission gun, an EDX detector and a high angle annular detector. TEM specimens were prepared by conventional mechanical grinding followed by ion milling.

Figure 2 shows typical cross-sectional bright field images of samples B (reference layer not shown), recorded under $g = 002$. The QDs are revealed mainly by the strain contrast. The lateral size of the InAs QDs is between 25 and 30 nm, which is

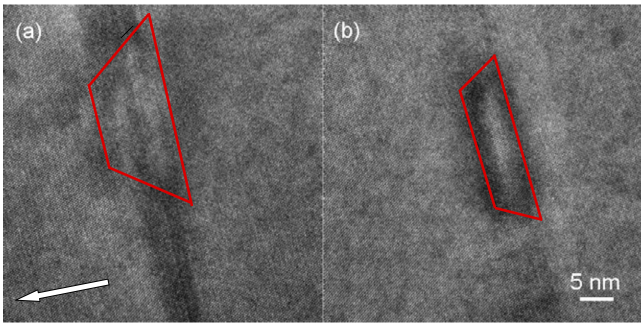

Figure 3. The HRTEM images recorded from an InAs quantum dot in layer 1 in (a) and layer 2 in (b). The arrow indicates the growth direction.

in good agreement with the AFM results. It is well known that InAs QDs capped by GaAs in the conventional temperature of 470–520 °C exhibit a reduced height compared to free-standing ones due to QD leveling or In detachment from the apex of QDs (QD collapse) during the capping process [20]. It is clearly shown in figure 2 that for QDs in layer 3 with capping of GaAs at 350 °C, the heights of the QDs are larger than those in layer 1 capped by GaAs at conventional 470 °C, which may indicate that In segregation and diffusion in the growth direction are strongly suppressed for GaAs overgrowth at 350 °C due to small diffusion length of In and Ga adatoms at such low substrate temperatures. Our TEM results agree well with the report by Gong *et al* [20], using cross-sectional scanning tunneling microscopy. Interestingly, in layer 2, QDs capped with an InAlAs and InGaAs CBL, the height of the quantum dots remains similar to those of QDs in layer 3, which implies that a InAlAs and InGaAs CBL can significantly suppress In segregation and In–Ga intermixing during overgrowth at 470 °C like capping the QDs with GaAs at low temperatures of 350 °C in layer 1. However, since there is strain contrast in the scattering contrast TEM images, it is difficult to tell the shape of the QDs, and most importantly, the In composition profile, which is direct proof of suppression of In segregation and In–Ga intermixing.

To reveal the structure of QDs clearly, high-resolution electron microscopy (HREM) images of the QDs are shown in figures 3(a) and (b) recorded in the [110] zone-axis from the QDs in layer 1 and layer 2, respectively. The arrow indicates the growth direction. The HREM images clearly show that the QDs are coherent with the surrounding GaAs matrix. When capped by GaAs, strain is increased and drives In atoms to detach from the top InAs QDs, which preferentially migrate to the wetting layer and/or the side of the QDs. As a consequence, a truncated pyramidal QD is formed as shown in figure 3(b), and hence the height of the QD is reduced during the capping process of GaAs. This is in accordance with the previous report [20]. It is interesting that when capping with InAlAs and InGaAs CBL, the strain enhancement at the apex of the QDs is smaller as compared to that when capped by GaAs. This is because of the smaller lattice mismatch with InAs QDs, so the probability for In atom detachment from the apex is reduced, and thus InAs QDs remain higher, as shown in figure 3(a).

High angle annular dark field (HAADF) STEM incoherent imaging is well known as Z-contrast imaging, which is sensitive to compositional difference. To provide a detailed profile of the In compositional variation across the InAs QDs along the growth direction and to further study the effect that an InAlAs and InGaAs CBL significantly reduces In segregation, diffusion, and In–Ga intermixing, the composition of the InAs QDs in layer 1 and layer 2 has been probed by energy dispersive x-ray (EDX) analysis. Figures 4(a) and (b) are the STEM images recorded from a QD in layer 1 and layer 2, respectively. The brighter contrast corresponds to heavier atoms, here, the In-rich area. The squares in the STEM images were used by the image analysis software to make sample drift correction when performing an EDX line scan. The electron beam probe diameter is about 0.5 nm. The scanned EDX signal is acquired with a low-temperature x-ray fluorescence detector with a resolution of 0.8 nm. By comparing figures 4(a) and (b), again, it is clear that the height of the QD in layer 2 is larger than that of QD in layer 1. The corresponding EDX line scans of In composition as a function of position are shown in figures 4(c) and (d), respectively. We can see that the In distribution along the growth direction in the line scans across the InAs QD in layer 1 and the InAs QD in layer 2 shows the expected sharp composition change as the probe moves from the bottom GaAs cladding layer into the InAs QD. It is obvious that, for the QD in layer 1, the peak

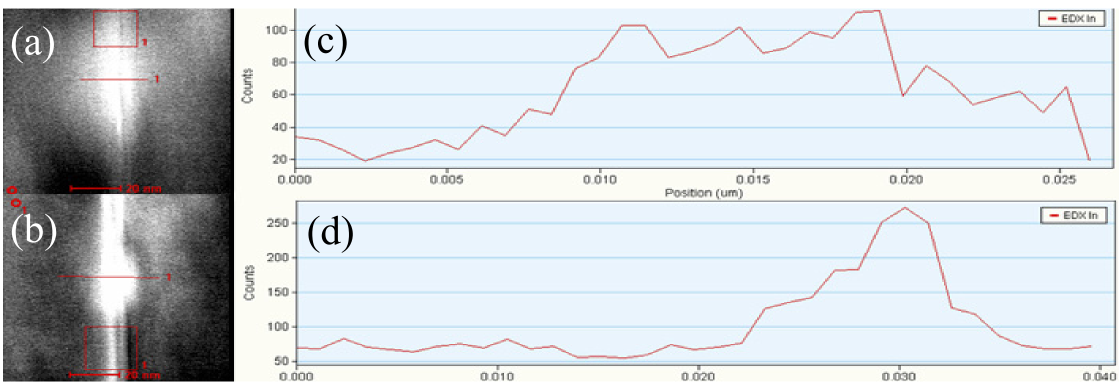

Figure 4. The STEM images recorded from the quantum dot in layer 1 in (a) and layer 2 in (b). The growth direction is horizontally leftwards for the two images. The corresponding EDX line profile of indium for the QD in layer 1 is shown in (c) and layer 2 in (d).

is broad across about 19 nm, indicating the In segregation, diffusion, and In–Ga intermixing broadly along the growth direction when the QD is capped by GaAs at conventional 470 °C. On the contrary, the peak is rather narrower for the QD in layer 2 capped by an InAlAs and InGaAs CBL, ranging over only about 7–9 nm. Moreover, the intensity of the peak of the QD in layer 2 is much higher than that in layer 1, and the fact indicates that more In atoms are confined in the InAs QDs, thus there is higher In concentration in the QDs with the introduction of the CBL. Reports of these direct quantitative experimental results have not been found in the literature previously. As we know, the QDs height and In composition profile of the self-assembled InAs/GaAs QDs strongly depend on In segregation, diffusion, and In–Ga intermixing during the capping process, STEM results provide a direct proof of these effects by insertion of an InAlAs and InGaAs CBL before capping of the consequent GaAs layers. We believe that the InAlAs and InGaAs CBL plays an important role in drastically decreasing the In segregation and In–Ga intermixing mainly due to the inactivity of Al adatoms at the capping temperature of 470 °C, and hence much less In–Al intermixing. These, together with the above-mentioned strain reducing effect (hence the reduction of In detached from the apex of the QDs), lead to the higher residual InAs QDs and In concentration within self-assembled InAs QDs after the capping process by the InAlAs and InGaAs CBL as compared with that capped by a simple GaAs layer. Such results provide a clear understanding and further insight into the mechanism of the above-observed optical properties of the capping of InAs QDs with an InGaAs and InAlAs CBL.

In summary, we have used X-TEM to study how an InAlAs and InGaAs CBL could affect the structural and optical properties of InAs/GaAs self-assembled QDs grown by MBE. STEM result revealed that the introduction of a thin InAlAs and InGaAs CBL strongly suppresses In segregation, diffusion, and In–Ga intermixing along the growth direction during the capping process of the InAs QDs, leading to the increased height of the nanostructures and a higher In concentration in InAs QDs after capping. Our structural information provides a deeper insight into the mechanism of the enhanced properties of InGaA and InAlAs CBL capped InAs QDs, which so far are one of the most promising candidates for the gain materials in GaAs-based 1.3 and 1.55 μm lasers.

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