Multiple stacking of self-assembled InAs quantum dots embedded by GaNAs strain compensating layers

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(Received 6 February 2006; accepted 18 July 2006; published online 26 October 2006)

We have investigated a growth technique to realize high-quality multiple stacking of self-assembled InAs quantum dots (QDs) on GaAs (001) substrates, in which GaN_xAs_{1-x} dilute nitride material was used as a strain compensation layer (SCL). The growth was achieved by atomic hydrogen-assisted rf molecular beam epitaxy, and the effect of strain compensation was systematically investigated by using high-resolution x-ray diffraction measurements. By controlling the net average lattice strain to a minimum by covering each QD layer with a 40-nm-thick $GaN_{0.005}As_{0.995}$ SCL, we obtained a superior QD structure with no degradation in size homogeneity. Further, no dislocations were generated even after 30 layers of stacking, and the area density of QDs amounted to as high as 3×10^{12} cm^{-2}. The photoluminescence peak linewidth was improved by about 22% for QDs embedded in GaNAs SCLs as the accumulation of lattice strain with increasing growth of QD layers was avoided, which would otherwise commonly lead to degradation of size homogeneity and generation of dislocations. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2359623]

I. INTRODUCTION

Recently, a variety of optoelectronic device applications of quantum dots (QDs) has been demonstrated such as QD lasers,[1] optical amplifiers,[2] and high-speed optical switches.[3] In particular, most important device characteristics such as low threshold currents,[4,5] low chirping,[6] and high characteristic temperatures[7] have been demonstrated in QD lasers. Moreover, the integration of GaAs-based long-wavelength optoelectronics is also desirable in cost-effective optical communication systems as well as in the next-generation photovoltaic devices.[8]

In the case of InAs QDs/GaAs system, one of the challenging technological issues today is to develop a multiple stacking technique in order to increase the total QD density, because the effective active volume of QDs per layer is much smaller than an active layer of quantum well (QW). To this end, if the host material used for burying the QDs is the same as the substrate, then the QDs in a stacked configuration would be, in general, accompanied by degradation of size homogeneity due to a buildup of internal strain with increasing number of QD layers.[9,10] To overcome these problems, several techniques, such as In-flush method[11] and columnar QD structures[12] have been reported. More recently, a strain compensation growth technique was demonstrated with InP-based system,[13] and stacking of up to ten QD layers has been achieved with GaAs-based system.[14,15] However, a further increase of stacked QD layers is required in order to realize a more practical QD superlattice structure. Here, we propose a fabrication method to increase the number of stacked InAs QD layers on GaAs (001) substrates without any degradation in the size uniformity and optical properties. Here, the strain compensation scheme was achieved by using GaNAs as a strain compensation layer abbreviated as SCL in the following.

II. EXPERIMENTS

For the fabrication of multiple layers of self-assembled InAs QDs on GaAs (001) substrates, we employed atomic hydrogen-assisted molecular beam epitaxy (H-MBE) with a radio frequency (rf) nitrogen plasma source.[16,17] After oxide removal and surface cleaning of each GaAs substrate by atomic H irradiation at 500 °C, a 250-nm-thick GaAs buffer layer was grown at a growth rate of 1 μm/h at 580 °C. Then, 2.0 ML (monolayer) of InAs QD layer and a 40-nm-thick GaN_xAs_{1-x} SCL were consecutively grown in pairs up to 20 or 30 multiple cycles at 480 °C. No growth interruption was employed during the stacking process. The growth rate of QD layer and spacer layer were 0.1 and 1.2 μm/h, respectively. In a separate experiment, we determined that in the first InAs QD layer prior to stacking, the average QD size was 23.8 nm in diameter, 3.1 nm in height, 13.4% in diameter dispersion, and $\sim 1.0 \times 10^{11}$/cm^2 in area density. The arsenic (As_4), hydrogen, and nitrogen back pressures during growth were kept constant at 1.2×10^{-6}, 5.0×10^{-6}, and 1.4×10^{-4} Torr, respectively. The rf power was varied from 150 to 200 W in order to control the N composition in GaN_xAs_{1-x} SCLs. The growth process and surface morphology were studied *in situ* by reflection high-energy electron diffraction (RHEED), *ex situ* by atomic force microscope (AFM), and scanning transmission electron microscope (STEM). The strained state of the QD superlattice structure in each sample was determined by using high-resolution x-ray diffraction (HR-XRD), and the optical properties were investigated by photoluminescence (PL) mea-

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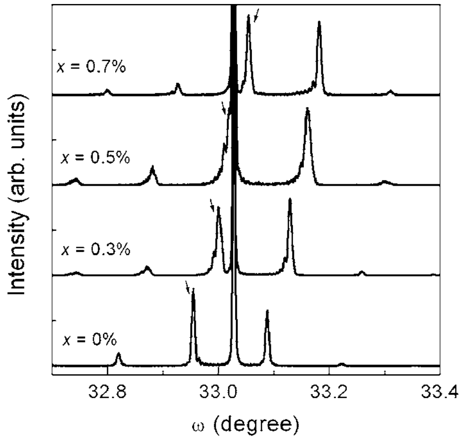

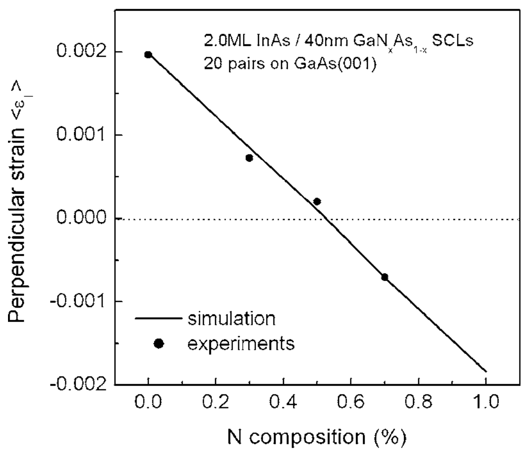

FIG. 1. (a) Symmetric (004) x-ray diffraction patterns for samples with 20 stacked layers of InAs QDs structure embedded in GaN_xAs_{1-x} SCLs with x=0%, 0.3%, 0.5%, and 0.7%. (b) Simulated (solid line) and measured (closed circle) strains in InAs/GaNAs system perpendicular to the growth axis on GaAs (001) substrate.

surements by using a standard lock-in technique at room temperature (RT) with a cw 532 nm second harmonic generation (SHG) Nd:YVO_4 laser as an excitation source and a liquid nitrogen cooled InGaAs photodetector.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the measured XRD spectra of $\omega/2\theta$ (004) scans for samples with 20 stacked pairs of InAs QDs with x=0% (GaAs), 0.3%, 0.5%, and 0.7% of GaN_xAs_{1-x} SCLs. Satellite peaks originating from the periodic superlattice structure can be clearly observed in each sample and these spectra were characterized by the zeroth-order peaks as indicated by arrows in the figure, which were located at 32.9544° for x=0, 33.0004° for x=0.3%, 33.0196° for x=0.5%, and 33.053 77° for x=0.7%.

The perpendicular lattice strain $\langle\varepsilon_\perp\rangle$ accumulated around the superlattice layer can be calculated by

$$\langle\varepsilon_\perp\rangle = \frac{\sin\theta_{\mathrm{GaAs}}}{\sin(\theta_{\mathrm{GaAs}}+\Delta\theta)} - 1,$$

where θ_{GaAs} is the Bragg angle for GaAs substrate and $\Delta\theta$ is the angle difference between the reflection peak of GaAs substrate and that of the zeroth-order peak. The XRD results and other QD parameters are summarized in Table I. It was clearly seen that the perpendicular strain $\langle\varepsilon_\perp\rangle$ gradually reduced with increasing N composition, and most importantly, the zeroth-order peak for x=0.5% showed a nearly perfect lattice match with GaAs. In this case, the averaged lattice constant of $InAs/GaN_{0.005}As_{0.995}$ almost matched that of GaAs, i.e., the tensile strain with respect to GaAs substrate accumulated around each QD layer has been canceled out by introducing a compressive strain generated by a 40-nm-thick $GaN_{0.005}As_{0.995}$ SCL. Furthermore, the small oscillation fringes observed indicated that abrupt heterointerfaces between QD and spacer layers have been obtained. The dependence of $\langle\varepsilon_\perp\rangle$ on N composition for samples with 20 pairs of $InAs/GaN_xAs_{1-x}$ structure are shown in Fig. 1(b). The simulated strain curve as drawn by a solid line was calculated by using the lattice constant of 5.6536 Å for GaAs, 6.0590 Å for InAs, and 4.503 Å for cubic-GaN. Here, the QD layer was approximated by 2.0 ML of uniform InAs layer for simplicity. From the simulation results, it can be seen that the perpendicular strain $\langle\varepsilon_\perp\rangle$ became almost zero at $x\sim0.5\%$, and all the experimental data were in good agreement with the simple simulation result. In other words, the N composition in GaNAs SCLs has been precisely controlled in our growth system.

Figure 2 shows the cross-sectional STEM image for a sample with 20 stacked layers of InAs QDs embedded in (a) GaAs spacer layers and (b) $GaN_{0.005}As_{0.995}$ SCLs. Only the top portions of the stacked structures are shown. A larger QD diameter of ~25 nm in the top layers of sample (a) was observed compared to ~20 nm for (b). This can be possibly explained by a larger residual internal strain in (a) than in (b). However, in STEM measurements, it was difficult to obtain accurate information on the strained state of QDs, and thus the compositional changes due to possible interdiffusion at the heterointerface between QD and spacer layers were studied. A significant outdiffusion of In and/or N atoms and hence intermixing would be commonly expected if the growth was performed at high growth temperatures. In such a case, the highly strained structure would also cause a generation of dislocations,[18] which in turn accelerates further intermixing of constituent atoms.[19] Though the actual lattice mismatch at the heterointerface between QD layer and GaNAs SCL was larger than that for InAs/GaAs heterointerface, we observed no strain-induced defects or dislocations as in Fig. 2(b), which would otherwise have led to an enhanced intermixing at the interface. Therefore, we believe that at least under our growth conditions, the outdiffusion of In and/or N atoms was minimal though further analysis will be required.

In a stacking of QD layers in self-assembled growth, QDs are usually vertically aligned if the spacer layer thickness is relatively thin, typically 10–20 nm in InAs/GaAs system.[10] However, it is also known that the strain field ex-

TABLE I. The QD parameters for samples with 20 stacked layers of InAs QDs embedded in GaN_xAs_{1-x} spacer layers. The mean diameter, height, density, and size fluctuation are quoted, and also HR-XRD results including the difference $\Delta\theta$ of diffraction angle between GaAs substrate peak and zeroth order satellite peak and net perpendicular strain $\langle\varepsilon_\perp\rangle$ are summarized.

	Single InAs QDs	20 stacked pairs of InAs QDs/GaN_xAs_{1-x}			
		x=0%	x=0.3%	x=0.5%	x=0.7%
Diameter (nm)	23.8	26.7	25.6	24.7	25.4
Height (nm)	3.1	3.8	2.8	2.3	3.2
Sheet density (cm^{-2})	1.0×10^{11}	1.3×10^{11}	1.0×10^{11}	1.0×10^{11}	9.0×10^{10}
Total density (cm^{-2})	⋯	2.5×10^{12}	2.0×10^{12}	2.0×10^{12}	1.8×10^{12}
Fluctuation(%)	13.4	25.6	16.4	14.2	15.7
$\Delta\theta$ (deg)	⋯	0.0731	0.0271	0.0079	−0.0263
$\Delta\varepsilon$	⋯	0.001 967	0.000 728	0.000 210	−0.000 704

tends over an even longer length. The pairing probability of the two neighboring InAs QDs in the vertical direction is still as high as ~64% as in Fig. 2(a), while the pairing probability was smaller and ~48% for strain controlled sample as in Fig. 2(b).

Figure 3 shows the AFM images measured for the topmost QD layer of 30 layer stacked structures embedded in (a) GaAs spacer layers and (b) $GaN_{0.005}As_{0.995}$ SCLs. The sample (a) with no strain compensation (uncontrolled) showed coalesced and large islanded structures, while sample (b) showed a significantly improved size dispersion and homogeneity. In a separate experiment, we confirmed that the density of large coalesced islands in a stacked QD structure increased with increasing stacked layers and became strongly apparent after about 30 stacks or more in InAs/GaAs system. The QD parameters in the topmost QD layer for 20 layer stacked samples are also summarized in Table I. The QD size became smaller with increasing N composition and reached a minimum at x=0.5% and was almost the same as that obtained for the single layer QD sample. Furthermore, the size fluctuation was minimal under the

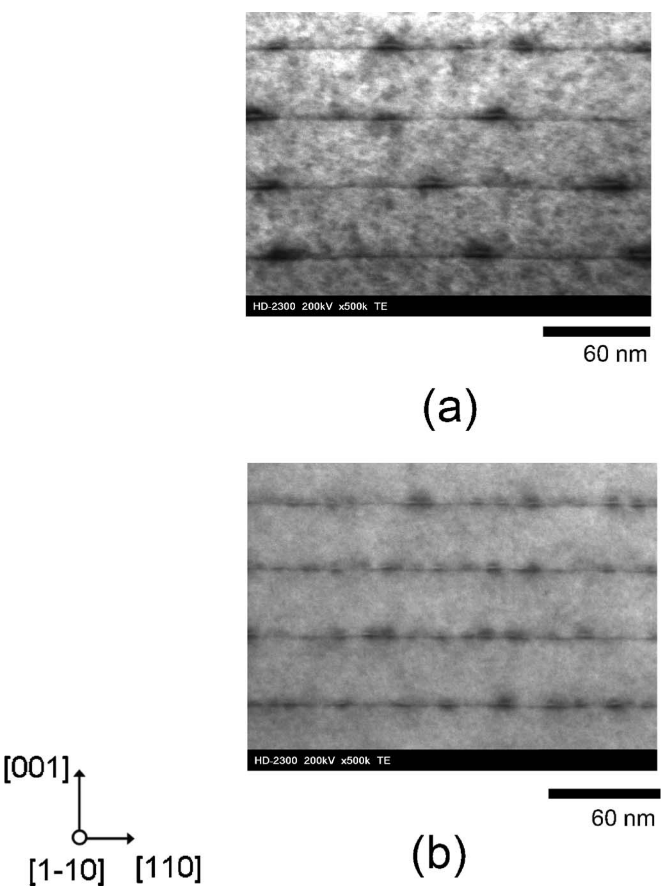

FIG. 2. Cross-sectional STEM image of top portion of 20 stacked layers of InAs QDs sample embedded in (a) GaAs spacer layers and (b) $GaN_{0.005}As_{0.995}$ SCLs.

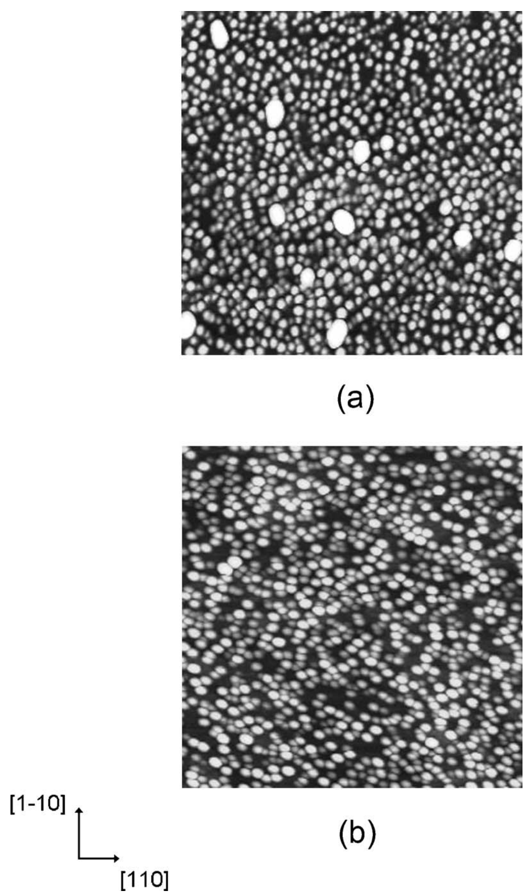

FIG. 3. AFM images of the top QD layer of 30 layer stacked InAs QDs embedded in (a) GaAs spacer layers and (b) $GaN_{0.005}As_{0.995}$ SCLs. Scan size is 500×500 nm^2 in both cases.

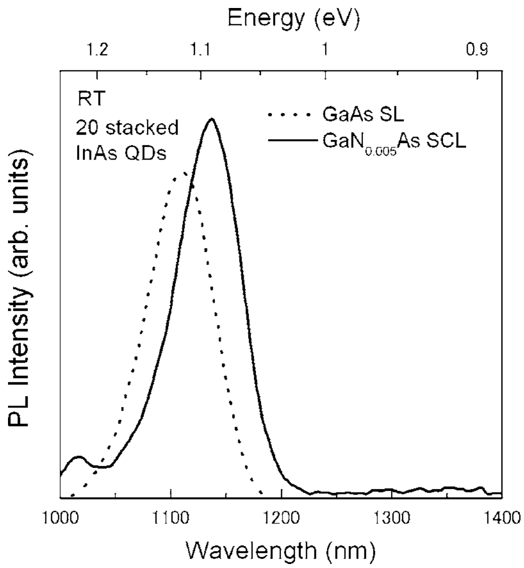

FIG. 4. PL spectra measured at room temperature from 20 stacked layers of InAs QDs embedded in GaAs spacer layers (dashed line) and $GaN_{0.005}As_{0.995}$ SCLs (solid line).

strain compensated condition. In Fig. 3(b), we calculated a total QD density of as high as $\sim 3.0 \times 10^{12}$ cm^{-2} after 30 pairs of stacking.

Finally, Fig. 4 shows the room temperature PL spectra from a sample with 20 pairs of stacked QDs with minimized overall strain by using $GaN_{0.005}As_{0.995}$ SCLs (solid line) and from an uncontrolled sample with GaAs spacer layers (dashed line). We observed that a single emission peak and a strong PL intensity was clearly observed in the stacked sample, while an InAs QD sample with a single layer of dots showed a weak PL emission at room temperature. On the other hand, by burying the QDs with GaNAs SCLs, we observed several important features such as (1) PL emission wavelength from InAs QDs redshifted from 1120.8 to 1144.9 nm, and (2) full width at half maximum (FWHM) of the emission peak was reduced from 65.2 to 53.0 meV. The redshift of the emission wavelength can be explained by the effect of strain compensation and reduction of potential barrier of InAs QDs embedded in GaNAs SCLs compared to GaAs spacer layers.[20] The observed reduction of FWHM was attributed to the improvement of size homogeneity in the growth direction.

IV. CONCLUSION

In conclusion, we have characterized the effect of strain compensation in multiple stacked InAs QD structures grown by atomic H-assisted rf-MBE. The samples were fabricated with 40-nm-thick GaN_xAs_{1-x} which was used as a strain compensation layer (SCL), and 20 or 30 layers of InAs QDs were fabricated on GaAs(001) substrates. From XRD measurements, 40-nm-thick $GaN_{0.005}As_{0.995}$ SCLs provided sufficient compressive strain to compensate for tensile strain induced by 2.0 ML of InAs QDs. The reduced overall strain not only resulted in suppression of generation of coalesced islands and dislocations, but also improved the PL linewidth. Though more studies on the optical properties of multistacked QD samples are required and currently under investigation, the results reported herein would suggest that the use of GaNAs SCLs is a promising way of realizing high-quality optoelectronic devices on cost-effective GaAs substrates, which can be applied to long-haul optical communication lasers as well as high efficiency multijunction tandem solar cells.

ACKNOWLEDGMENTS

This work was supported in part by the Incorporated Administrative Agency New Energy and Industrial Technology Development Organization (NEDO) under Ministry of Economy, Trade and Industry (METI), Japan, and the Thermal and Electric Energy Technology Foundation. One of the authors (R. O.) is supported by Research Fellowships of the Japan Society for the Promotion of Science (JSPS) for young scientists. The authors also acknowledge Dr. K. Akahane of National Institute of Information and Communication Technology, Tokyo, for valuable discussions.

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