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MBE growth of InAs self-assembled quantum dots embedded in GaNAs strain-compensating layers

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Abstract

We fabricated self-assembled InAs quantum dots (QDs) embedded with a combined set of a GaNAs strain-compensating layer (SCL) and an InGaAs strain-reducing layer (SRL) by using atomic hydrogen-assisted molecular beam epitaxy (H-MBE) with a RF plasma nitrogen source. By inserting an InGaAs SRL between the InAs QD layer and GaNAs SCL, we were able to achieve a significant improvement of optical properties. A 1.3 μm-range photoluminescence (PL) emission was clearly obtained at 300 K from QDs embedded in a 5 nm-thick $In_{0.15}Ga_{0.85}As$ SRL followed by a 20 nm-thick $GaN_{0.008}As_{0.992}$ SCL. Further, PL emission from the first excited state of QDs was also observed under a high excitation intensity of ~20.0 W/cm^2.

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1. Introduction

Self-assembled quantum dots (QDs) have been of great interest because of their potential applications to high-performance lasers [1], single electron transistors [2], and high-efficiency solar cells [3]. Recently, InAs/GaAs QDs have been intensively investigated for the low-cost and highly efficient QD laser diodes operating at the optical fiber communication wavelength. However, the optical emission from InAs/GaAs QDs often appears at a wavelength of ~1.0 μm, due to the strong three-dimensional quantum confinement as well as compressive strain accommodated around the QDs. In order to realize a long-wavelength emission, several methods to partially reduce the effect of compressive strain have been reported, where InAs QD layer was embedded in InGaAs [4], and GaAsSb [5] strain-reducing layers (SRLs). In the case of using a high In or Sb composition in SRL, however, one has to take into account of the effect of critical thickness, which would lead to the generation of dislocations and a significant degradation of optical properties.

In our previous work, we proposed InAs self-assembled QDs structures embedded in GaNAs strain-compensating layers (SCLs) [6]. Since GaN_xAs_{1-x} alloy has a smaller lattice constant than GaAs, it is possible to reduce the net average strain by inducing a tensile strain by using GaNAs. Further, by replacing the more common GaAs embedding layer with GaNAs SCL of a suitable N composition, we were able to observe, (1) photoluminescence (PL) peak shifting toward longer wavelengths, (2) a stronger PL intensity compared to the sample embedded in GaAs, and (3) a decreased density of non-radiative recombination centers. However, increase of N composition would generally result in a degradation of overall crystal quality of GaNAs, and also increases the lattice mismatch at the heterointerface. In this work, we propose to insert an InGaAs SRL between the InAs QD layer and GaNAs SCL in order to decrease the lattice mismatch at the heterointerface. We show remarkably improved PL characteristics, and a peak emission at 1.3 μm was observed at RT.

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2. Experimental procedure

Self-assembled InAs QD structures were grown on GaAs (0 0 1) substrates by atomic hydrogen-assisted molecular beam epitaxy (H-MBE) with a RF plasma nitrogen source. Atomic H was generated in passing the hydrogen gas by using a cracking cell with tungsten filament heated at 1500 °C by direct current. After the oxide removal and surface cleaning of GaAs substrate by atomic H irradiation at 500 °C for 30 min, a 300 nm-thick GaAs buffer layer was first grown at a growth rate of 1 μm/h at 580 °C. Next, a single layer of 2.0 monolayers (MLs) of InAs QD was deposited at a growth rate of 0.1 μm/h at 480 °C. Then, four different types of embedding layers were used to bury the QDs as shown in Fig. 1 in order to investigate the effect of strained state of QD layer on the optical properties, (a) a 40 nm-thick GaAs layer as a reference, (b) a single layer of 20 nm-thick $GaN_{0.016}As_{0.984}$ SCL, (c) a single layer of 5 nm-thick $In_{0.25}Ga_{0.75}As$ SRL, and (d) a combined set of a 5 nm-thick $In_{0.15}Ga_{0.85}As$ SRL and a 20 nm-thick $GaN_{0.008}As_{0.992}$ SCL. Finally, a 40 nm-thick GaAs capping layer was grown in all samples. Atomic H was irradiated during the entire growth sequence in order to obtain an atomically smooth surface and passivation of defect centers in GaAs and GaNAs [7]. The RF plasma power was set at 200 W. The growth rate of GaNAs layer was changed from 1.0 μm/h to 0.5 μm/h in order to control the N composition, since N composition is known to vary in inverse proportion to the growth rate [8]. No growth interruption was employed during growth, and the arsenic (As_4), hydrogen and nitrogen back pressures were set to 1.4×10^{-6}, 4.5×10^{-6} and 1.5×10^{-4} Torr, respectively. The growth mode was monitored in situ by reflection high-energy electron diffraction (RHEED) and the surface morphology was studied ex situ by atomic force microscope (AFM). The PL measurements were performed within a closed cycle helium cryostat, and the signals were detected by using a standard lock-in technique with a cw 532 nm second harmonic generation (SHG) Nd:YVO_4 laser as an excitation source and a liquid nitrogen-cooled InGaAs photodetector.

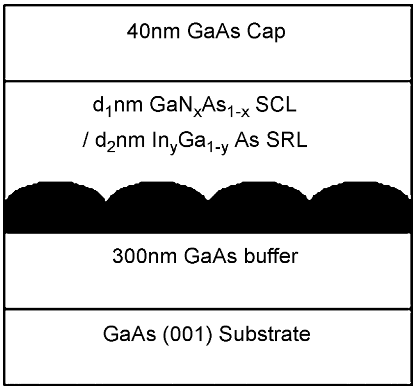

Fig. 1. Schematic structure of QD samples fabricated in this work.

3. Results and discussion

Prior to covering the QDs with a capping layer, we confirmed that the average size of InAs QDs was 35.3 nm in diameter and 7.3 nm in height with an area density of $3.7 \times 10^{10}/cm^2$. A good size uniformly in the diameter with a dispersion of only ~15% was obtained in our sample [6].

Fig. 2 shows the PL spectra measured at 77 K for InAs QDs embedded with a (a) GaAs, (b) $GaN_{0.016}As_{0.984}$ SCL, (c) $In_{0.25}Ga_{0.75}As$ SRL, and (d) combined set of $In_{0.15}Ga_{0.85}As$ SRL and $GaN_{0.008}As_{0.992}$ SCL, respectively. The excitation intensity was 2 W/cm^2. In Fig. 2(b)–(d), we observed a long wavelength emission at around 1.20 μm compared with 1.03 μm in (a). However, the PL intensities were strongly dependent on the actual structure of embedding layers. A redshift of PL peak for the sample with GaNAs SCL as of Fig. 2(b) can be explained by a reduction of the net strain and potential barrier height around QDs. However, a decrease of PL intensity was possibly due to the effect of both a larger lattice mismatch in the heterointerface between InAs QDs and GaNAs SCL and a poor crystalline quality of GaNAs material. We previously reported that increasing the N composition above 0.8% in GaNAs SCL would result in a monotonous degradation of PL properties [6]. In the case of Fig. 2(c), the observed redshift was likely due to a reduction of the local strain accommodated around QDs by use of an $In_{0.25}Ga_{0.75}As$ SRL. By using a combined set of $In_{0.15}Ga_{0.85}As$ SRL and $GaN_{0.008}As_{0.992}$ SCL, a strong PL emission at 1.2 μm was achieved as shown in Fig. 2(d). The shoulder peak observed at a higher energy was from the contribution of an excited state as will be discussed later. The narrower full width at half maximum (FWHM) of

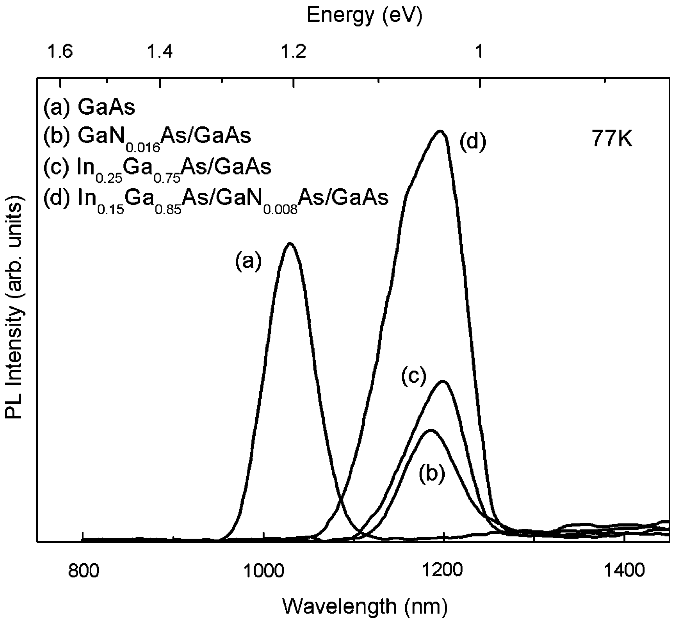

Fig. 2. PL spectra measured at 77 K for InAs QDs embedded with (a) GaAs (reference), (b) $GaN_{0.016}As_{0.984}$ SCL, (c) $In_{0.25}Ga_{0.75}As$ SRL, and (d) $In_{0.15}Ga_{0.85}As/GaN_{0.008}As_{0.992}$, respectively.

45.5 meV for (d) was obtained by a Gaussian fitting for the ground state emission compared to 57.1 meV for (b) and 64.4 meV for (c), respectively. Furthermore, a 1.3 μm PL emission at 300 K with a FWHM of 41.1 meV was observed for the sample (d) as shown in Fig. 3, while the other three showed much weaker PL emission.

Next, we evaluated the effect of embedding layer structure on radiative emission efficiency of InAs QDs. Fig. 4 shows the temperature dependence of PL integral intensity for the sample with (a) GaAs, (b) $GaN_{0.016}As_{0.984}$ SCL, (c) $In_{0.25}Ga_{0.75}As$ SRL, and (d) $In_{0.15}Ga_{0.85}As/GaN_{0.008}As_{0.992}$, respectively. The experimental data can be fitted with the following equation:

$$I = I_0/\{1 + C_1 \exp(-E_1/k_B T) + C_2 \exp(-E_2/k_B T)\} \quad (1)$$

which is characterized by the two thermally activated processes. k_B is Boltzmann constant, and I_0, C_1 and C_2 are the constants independent of temperature. The activation energy E_1 shows the thermal escape of carriers from the QDs to the potential barrier, while E_2 shows the escape of carriers from the QDs to the shallow levels induced by defects and dislocations at the heterointerface. C is known to be proportional to the density of non-radiative recombination centers [9]. It can be seen that good fitting curves were obtained and Table 1 summarizes the fitting parameters E_1, E_2, C_1 and C_2 for each sample, respectively. It can be seen that the use of GaNAs SCL was effective for reducing the value of C_1 compared to the case of GaAs capping as of (a), which was due to the effect of strain-compensation. However, a lower E_1 and higher C_2 were observed as a result of the reduction of potential barrier and larger lattice-mismatch at the heterointerface. On the other, the use of InGaAs SRL showed a marginal reduction of C_1 and E_1 compared to GaAs capping. By using the combined set of InGaAs/GaNAs layer, we were able to obtain a higher E_1 value of 140 meV and lower C_1. Therefore, we confirmed that a combination of InGaAs SRL/GaNAs SCL significantly improves the optical properties of InAs QDs. The optical properties of QDs depend strongly on the factors such as the barrier height, strain effect, and strain-induced interdiffusion.

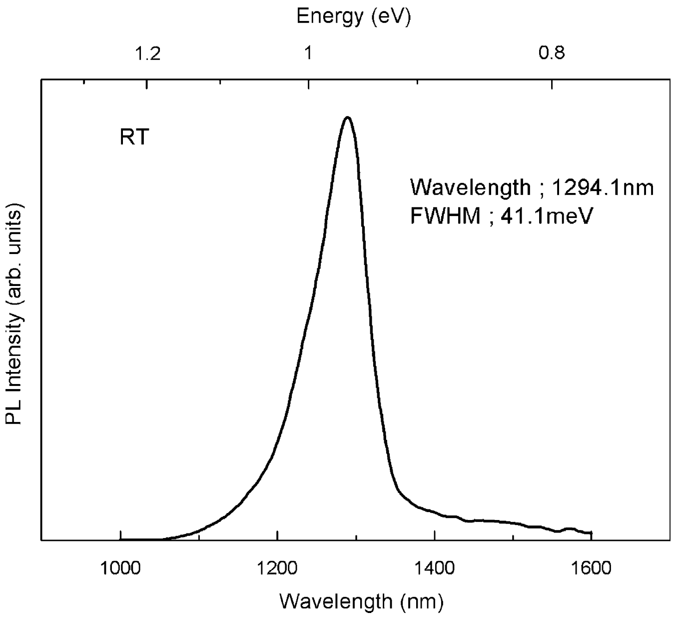

Fig. 3. PL spectrum measured at 300 K for InAs QDs embedded with a combined set of $In_{0.15}Ga_{0.85}As/GaN_{0.008}As_{0.992}$ layers.

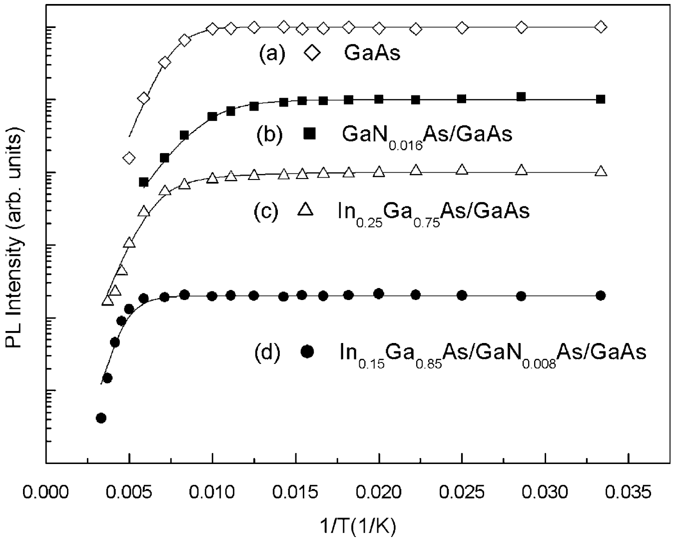

Fig. 4. Temperatures dependence of PL integral intensity for sample with (a) GaAs, (b) $GaN_{0.016}As_{0.984}$ SCL, (c) $In_{0.25}Ga_{0.75}As$ SRL, and (d) $In_{0.15}Ga_{0.85}As/GaN_{0.008}As_{0.992}$, respectively. The fitted curves are shown as solid lines.

Here we should also consider the effect of both the interdiffusion at the interface and the segregation of nitrogen leading to a high quantity of nitrogen being accumulated toward the surface [10]. However, we think these effects are much smaller than the effect of local and net average strain, and the interdiffusion should be minimal because no growth interruption was employed during the growth of QDs and embedding layers. A partial reduction of compressive strain around QDs by use of InGaAs SRL as well as a reduction of net average strain by using GaNAs SCL are both important in improving the optical properties of InAs QDs.

Finally, we investigated the excitation intensity dependence of PL characteristics for the sample embedded with

Table 1
Activation energies E_1, E_2 and C_1, C_2 constants obtained from the fitting by using Eq. (1) for each sample

Cap layer structure	E_1 (meV)	C_1	E_2 (meV)	C_2
(a) GaAs	115	25 000	20	1
(b) $GaN_{0.016}As_{0.984}/GaAs$	70	1200	20	5
(c) $In_{0.25}Ga_{0.75}As/GaAs$	110	10 000	25	1
(d) $In_{0.15}Ga_{0.85}As/GaN_{0.008}As_{0.992}/GaAs$	140	2500	25	0.3

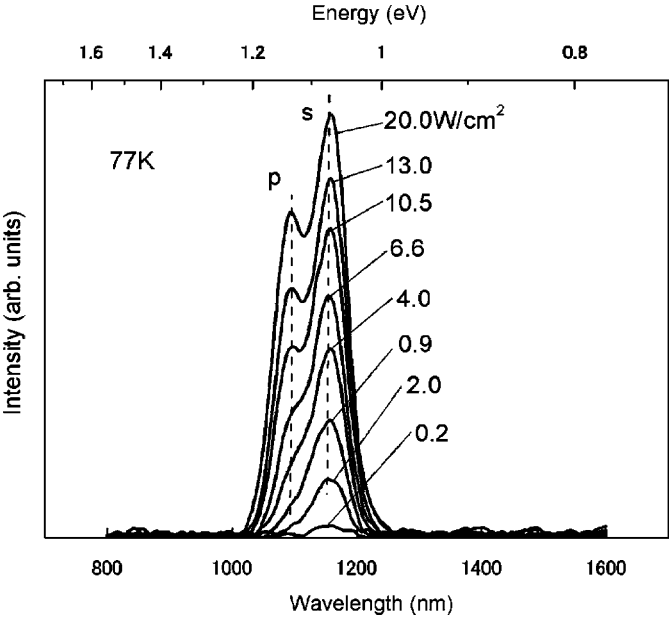

Fig. 5. Dependence of PL characteristics on excitation intensity for sample embedded with a combined set of $In_{0.15}Ga_{0.85}As/GaN_{0.008}As_{0.992}$ layers. The excitation intensity was varied from 0.2 to 20.0 W/cm^2. S-peak indicates an emission from the ground state, while p-peak shows that from the first excited state of QDs, respectively.

InGaAs/GaNAs combined layer and the results are shown in Fig. 5. The excitation intensity was varied from 0.2 to 20.0 W/cm^2. We clearly observed an emission from the first excited state (p) as the excitation was increased. Though not shown, the emission from the excited state was not observed in the sample with a single layer of GaNAs SCL or InGaAs SRL even at a high excitation intensity of 20.0 W/cm^2. The energy spacing between the ground state (s) and p-state was about 66.6 meV, which agreed well with the reported value for a typical InAs/GaAs system [11]. The appearance of a p-state indicates that the use of $In_{0.15}Ga_{0.85}As/GaN_{0.008}As_{0.992}$ embedding layer increases the recombination lifetime and hence eases the state filling, which can be attributed to an improved quality of the heterointerface and reduced non-radiative recombination. Thus, the use of InGaAs SRL/GaNAs SCL is considered a powerful way to improve the optical properties of InAs QDs.

4. Summary

In summary, we investigated the optical properties of InAs self-assembled QDs embedded in InGaAs/GaNAs layers on GaAs (0 0 1) substrates by atomic H-assisted RF-MBE. From PL measurements, a strong emission at around 1.3 μm with a narrow linewidth of 41.1 meV at 300 K was achieved by using InGaAs/GaNAs combined layer. We attribute these improvements to the reduction of non-radiative recombination due to reduction of both local and net strain. Though more studies are required such as the effect of interdiffusion, the results found here may be very useful in realizing high-quality long-wavelength lasers on cost effective GaAs substrates.

References

[1] Y. Arakawa, H. Sakaki, Appl. Phys. Lett. 40 (1982) 939.
[2] U. Meirav, E.B. Foxman, Semicond. Sci. Technol. 11 (1996) 255.
[3] A.J. Nozik, Physica E 14 (2002) 115.
[4] K. Mukai, M. Sugawara, Appl. Phys. Lett. 74 (1999) 3963.
[5] K. Akahane, N. Yamamoto, N. Ohtani, Physica E 21 (2004) 295.
[6] R. Oshima, T. Hashimoto, H. Shigekawa, Y. Okada, Physica E 32 (2006) 77.
[7] Y. Shimizu, N. Kobayashi, A. Uedono, Y. Okada, J. Crystal Growth 278 (2005) 553.
[8] S.G. Spruytte, M.C. Larson, W. Wampler, C.W. Coldren, H.E. Petersen, J.S. Harris, J. Crystal Growth 227 (2001) 506.
[9] J.I. Pankove, Optical Processes in Semiconductors, Dover Publications, Inc., New York.
[10] J.A. Gupta, Z.R. Wasilewski, B.J. Riel, J. Ramsey, G.C. Aers, R.L. Williams, G.I. Sproule, A. Perovic, D.D. Perovic, T. Garanzotis, A.J. Springthorpe, J. Crystal Growth 242 (2002) 141.
[11] R. Heitz, A. Kalburge, Q. Xie, M. Grundmann, P. Chen, A. Hoffmann, A. Madhukar, D. Bimberg, Phys. Rev. B 57 (1998) 9050.