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Photoluminescence study of InAs quantum dots embedded in GaNAs strain compensating layer grown by metalorganic-molecular-beam epitaxy

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Self-assembled InAs quantum dots (QDs) embedded in GaN_{0.007}As_{0.993} strain compensating layers have been grown by metalorganic-molecular-beam epitaxy on a GaAs (001) substrate with a high density of $1 \times 10^{11} \text{ cm}^{-2}$. The photoluminescence properties have been studied for two periods of InAs quantum dots layers embedded in GaN_{0.007}As_{0.993} strain compensating layers. Four well-resolved excited-state peaks in the photoluminescence spectra have been observed from these highly packed InAs QDs embedded in the GaN_{0.007}As_{0.993} strain compensating layers. This indicates that the InAs QDs are uniformly formed and that the excited states in QDs due to the quantum confinement effect are well defined. This is explained by tensile strain in GaNAs layers instead of the usual GaAs layers to relieve the compressive strain formed in InAs QDs to keep the total strain of the system at a minimum. © 2002 American Institute of Physics. [DOI: 10.1063/1.1516873]

I. INTRODUCTION

Quantum dots (QDs) have been an important topic of research for more than 10 years. Great effort has been devoted to the fabrication and exploitation of self-assembled QDs, due to their novel physical properties and potential applications.¹⁻³ QD lasers have been extensively studied. The performance of QD lasers has been improved significantly in the past few years. However, the emission wavelength from self-assembled In(Ga)As QDs on GaAs is typically around $1.0 \mu\text{m}$ and is mostly limited to wavelengths shorter than $1.3 \mu\text{m}$. In order to be applied to fiber optical communications, extension of its optical emission wavelength to $1.55 \mu\text{m}$ and beyond is necessary.⁴

In order to realize longer wavelength emission from InAs QDs, several groups have achieved 1.3 or $1.55 \mu\text{m}$ emission for InAs QDs with InGaAs strain reducing layers (SRLs).⁴⁻⁷ Using the InGaAs SRL instead of GaAs as the capping layer, the band gap energy of InAs QDs is reduced due to the relief of compressive strain in InAs QDs. As a result, the photoluminescence (PL) peak of InAs QDs shifts toward a longer wavelength.⁵ But the higher indium composition in the SRL further accumulates the amount of overall compressive strain and the PL efficiency tends to be degraded due to the nonradiative recombination centers produced in the SRL due to the excess strain.⁵

In our previous work,⁸ we proposed an InAs QDs structure embedded in GaNAs strain compensating layers (SCLs). Tensile strain in GaNAs layers grown on GaAs, instead of InGaAs or GaAs layers, will release the compressive strain formed in InAs QDs and keep the total strain of the system at a minimum. This will reduce the strain effect on the InAs energy gap, i.e., the increase of the energy gap with compressive strain will be lowered by the relief of strain inside the InAs QDs. Emissions of 1.3 and $1.55 \mu\text{m}$ have recently been

observed from InAs QDs embedded in GaNAs SCLs.

QD lasers are expected to have low threshold current densities, high gain, high quantum efficiencies, and ultrahigh characteristic temperatures.⁹⁻¹¹ Significant progress on the performance of QD lasers has been made in the past few years. Up to now, however, their performance in devices has not necessarily satisfied theoretical predictions for low threshold current densities and high optical gains because of limitations on the dot densities and size uniformity of QDs. Thus, improvement of QD lasers will depend on further investigations of increasing the QD density and controlling the size distribution.¹²

It has been extensively studied on the dot density and size uniformity of QDs. The well-resolved excited-state peaks of quantum dots have been observed in PL spectra with improvement of the dot size homogeneity for lower QD density. High QD density up to $10^{11}/\text{cm}^2$ has been reported but well defined PL from QDs states has not been observed under such high QD packing density. It has been difficult to identify QD energy states for so high a dot density due to the remaining inhomogeneity of the QDs.

In this work, it will be demonstrated that self-assembled InAs quantum dots embedded in GaNAs SCLs show excellent uniformity and clear PL subpeaks originating from QD energy states are observed in spite of their very high dot density of $1 \times 10^{11}/\text{cm}^2$. The temperature and excitation dependence of PL spectra will be investigated in detail for two stacks of InAs quantum dots layers embedded in a GaN_{0.007}As_{0.993} SCL. The slight redshift of the PL peaks for increasing excitation intensity as well as the unusual dependence of the PL linewidth on the temperature will be discussed. The thermal activation energy estimated from the temperature dependence of the PL intensity correlates well with the separation in energy of the PL peak energy position, and this suggests that the energy barrier height in the present InAs QDs embedded in GaNAs SCLs is larger than that of conventional InAs QDs embedded in GaAs barrier layers.

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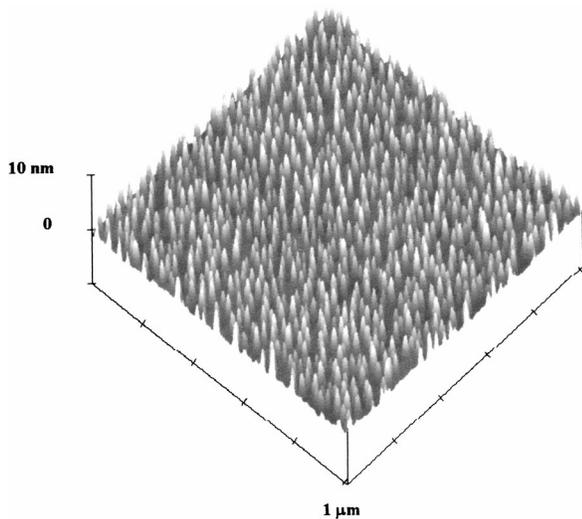


FIG. 1. Atomic force microscopy image of InAs QDs on a GaAs (001) substrate.

II. EXPERIMENT

Self-assembled InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL were grown on GaAs (001) substrates by metalorganic-molecular-beam epitaxy (MOMBE). The metalorganic precursors used were triethylgallium (TEGa), trisdimethylaminoarsenic (TDMAAs), and Trimethylindium (TMIn). The GaAs buffer layer was first grown on a GaAs substrate with thickness of 300 nm at 550 °C. The QDs were formed under Stranski–Krastanov growth mode, and a wetting layer (WL) with thickness of 2 monolayers (ML) was formed under the InAs QDs at 450 °C. Then a 10 nm GaNAs strain compensating layer was grown at 550 °C. The nitrogen composition was about 0.7%. Similar growth conditions were used for growth of the second stacked layers. The three-dimensional growth was monitored by reflection high-energy electron diffraction (RHEED) and QD nucleation was observed directly via the onset of a spotty RHEED pattern. Details of the growth are reported elsewhere.⁸ For an atomic force microscopy investigation, growth was stopped and the samples were quickly cooled down to room temperature.

The 532 nm line from the second harmonic generation of a YAG laser was used as the excitation source for the PL measurements, which were performed between 18 K and room temperature in a closed-cycle He cryostat. A LN_2 -cooled Ge detector was used to measure the signal dispersed by a 0.5 m monochromator via a lock-in mode detection scheme.

III. RESULTS AND DISCUSSION

Figure 1 shows the $1\ \mu\text{m} \times 1\ \mu\text{m}$ atomic force microscopy image of self-assembled InAs QDs grown on GaAs (001). The characteristic diameter and the mean height of the QDs are about 30 and 3 nm, respectively. Very high area density of $1 \times 10^{11}/\text{cm}^2$ was observed. The surface coverage is about 78%, and it is close to the maximum coverage. In our sample, the average spacing of two neighboring dots in the lateral direction is 8 nm.

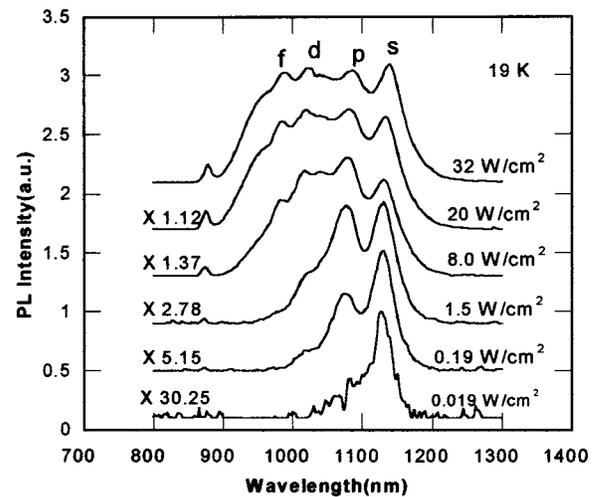


FIG. 2. Excitation intensity dependence of normalized PL spectra of two-period stacked InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL. The QD shells are labeled with atomic notations *s*, *p*, *d* and *f*.

Every dot from the upper layer is not necessarily vertically aligned with each other in the two stacks of InAs QDs with the 10 nm $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL. The dots in the upper and lower layers are almost equal in size. The lattice distortion due to the strain field was very small around the dots, as measured by a cross-sectional transmission electron microscope (TEM). This shows the strain compensation effect of the GaNAs SCL. Their crystal quality shown TEM was reported in detail elsewhere.⁸

The excitation intensity dependence of the normalized PL spectra for the two stacks of InAs QDs embedded in the $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL measured at 19 K is shown in Fig. 2, which displays four well-resolved excited-state peaks. It indicates that the InAs QDs are uniformly formed and that the excited states in QDs due to the quantum confinement effect are well defined. At low excitation intensity ($0.019\ \text{W}/\text{cm}^2$) we observe only a single peak (labeled *s*) in the PL spectrum. When the excitation intensity is increased, the emergence of the second and third peaks (labeled *p* and *d*) was observed in the PL spectra. The fourth peak (labeled *f*) gradually appeared as the excitation intensity was increased to over $8\ \text{W}/\text{cm}^2$. These results indicate that the excitation intensity dependence of the PL spectra are due to the state filling process in the QDs,^{13–16} namely, the shells of the QDs are progressively populated by a number of photoexcited carriers with an increase in excitation intensity. When the excitation intensity is increased, the saturation of the *s*, *p*, *d* and *f* peak intensities was also observed as shown in Fig. 2. According to the state filling effect, lower energy states will become saturated and a higher energy state will then begin to be populated if the excitation intensity is high enough. Strong photoluminescence was observed even for the low excitation intensity of $0.019\ \text{W}/\text{cm}^2$, which indicates that the InAs QDs embedded in the $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL have excellent quality and a low density of defects. The origin of the PL peak that appeared at 873 nm (1.42 eV) will be discussed later.

Usually for high QD density it is difficult to identify the zero-dimensional (0D) states due to lateral coupling or strain interaction. In this regard, high QD density up to 1

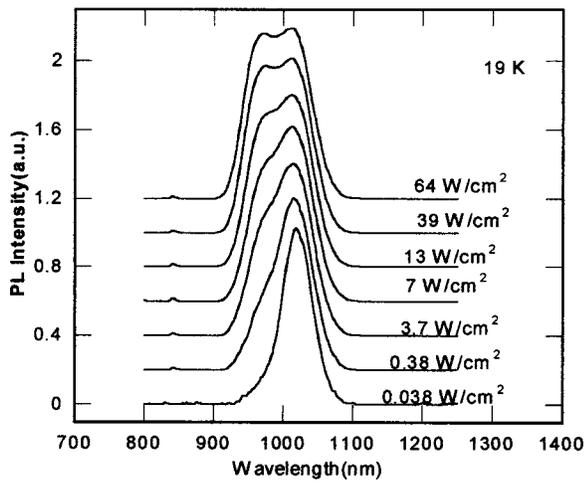


FIG. 3. Excitation intensity dependence of normalized PL spectra of two-period stacked InAs QDs embedded in GaAs spacer layers at 19 K.

$\times 10^{11}/\text{cm}^2$ has been reported but the well defined PL from QDs states has not been observed under such a high packing density of QDs.^{17,18} These problems are usually even worse when multistacked layers are used to try to increase the optical gain in the active region of QD lasers. But observation of the well-resolved excited states in this work clearly demonstrates that the QDs are quite uniform from one layer to the next although the area density of the QDs is very high. The observation of a number of well-resolved excited states is, therefore, the principal figure of merit for QDs. On the other hand, we could not observe such well defined excited-state peaks for the samples grown with 10 nm GaAs spacer layers with two stacks of InAs QDs grown under the same optimized growth condition. As is seen by the excitation intensity dependence of the normalized PL spectra shown in Fig. 3, only the broader first excited state was observed with much higher excitations. The PL intensity of this sample was also weaker than that shown in Fig. 2. This comparison demonstrates that the tensile strain in the GaNAs layers grown on GaAs, instead of InGaAs or GaAs layers, will relieve the compressive strain formed in InAs QDs and keep the total strain of the system at a minimum. This helps the InAs QDs embedded in the $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL to be quite uniform and almost equal in size from one layer to the next although the area density of the QDs is very high, and the InAs QDs embedded in the $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL have a low density of defects on the InAs/GaNAs interface.

We can see in Figs. 2 and 3 also that the PL peak energy of the ground state of InAs QDs embedded in $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL is redshifted with respect to that grown with GaAs spacer layers even if the InAs QDs were prepared under the same growth condition. This can also be explained by the strain effect. Using the GaNAs SCL instead of GaAs as the capping layer, the band gap energy of InAs QDs is reduced due to the relief of compressive strain in InAs QDs. As a result, the PL peak of InAs QDs shifts toward a longer wavelength.⁸

Figure 4 shows the excitation intensity dependence of the PL peak energy for two-period stacked InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL. The horizontal line is added

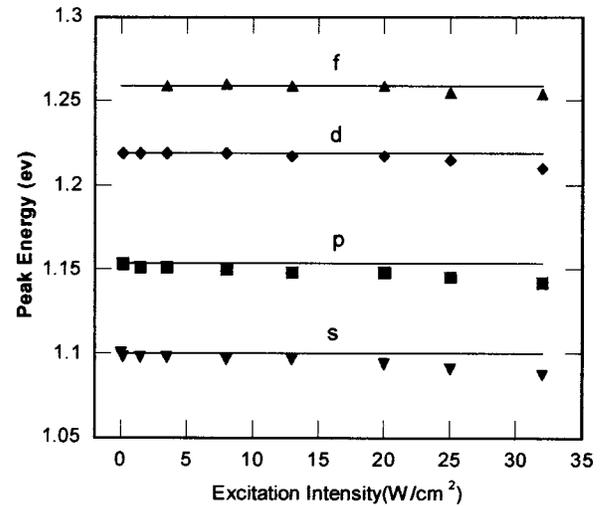


FIG. 4. Excitation intensity dependence of PL peak energy of two-period stacked InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL. The energy positions of the ground state and excited states are denoted by *s*, *p*, *d* and *f* curves, respectively.

for easier comparison with the experimental data. We can see in Fig. 4, the *s*, *p*, *d*, and *f* peaks showed a slight redshift with an increase in excitation intensity. When the excitation intensity was increased, the density of the photogenerated carriers increased, and the exchange interaction between carriers of the same charge increased. We attribute the redshift of the *s*, *p*, *d*, and *f* peaks with an increase in excitation intensity to band gap renormalization due to carrier-carrier exchange interaction.¹⁹⁻²¹

The temperature dependence of normalized PL spectra of InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL with excitation intensity of 9.5 W/cm^2 is shown in Fig. 5. We observed four well-resolved excited-state peaks labeled *s*, *p*, *d*, and *f* from the QDs and one additional peak at 873 nm measured at 18 K. When the temperature was increased to 70 K, the emission peak at 873 nm disappeared, the origin of which will be discussed later. The *f* and *d* peaks were gradually

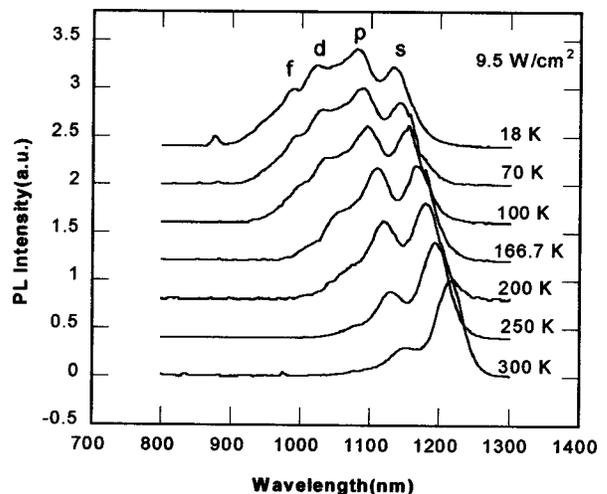


FIG. 5. Temperature dependence of normalized PL spectra of two-period stacked InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL. The QD shells are labeled with atomic notations: *s*, *p*, *d* and *f*.

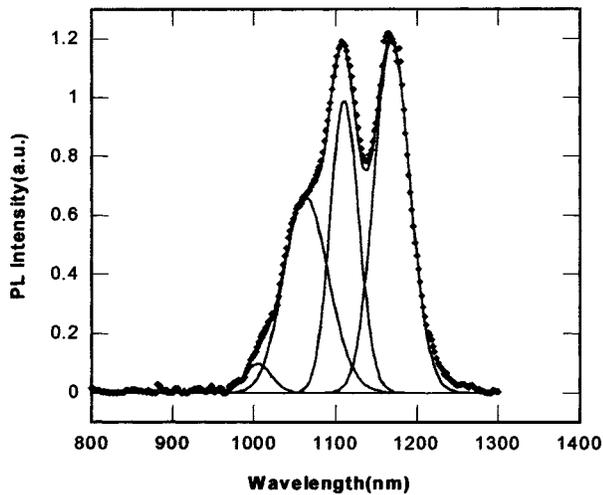


FIG. 6. PL spectrum and multiple fitted Gaussian curves of two-period stacked InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL at 166.7 K.

reduced and finally disappeared as the temperature was increased. At room temperature, the *s* peak was predominantly observed. The decrease of the PL intensity is attributed to thermal quenching due to carrier escape from the QDs via thermionic emission into the GaNAs barriers followed by nonradiative recombination which will be discussed below.

For each temperature, the PL spectra can be fitted by multiple Gaussian line shapes as is shown in Fig. 6, where the solid lines represent multiple fitted Gaussian curves, and triangles represent experimental data. It is obvious that the fit is in fairly good agreement with the experimental result.

Figure 7 shows the temperature dependent full width at half maximum (FWHM) of PL spectra of InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL. In the temperature range from 19 to 300 K, the linewidth shows unusual dependence on the temperature. This is explained by the tunneling effect.^{22,23} Due to the difference in depth of the QD potentials, even at low temperatures, photogenerated carriers in a high-density

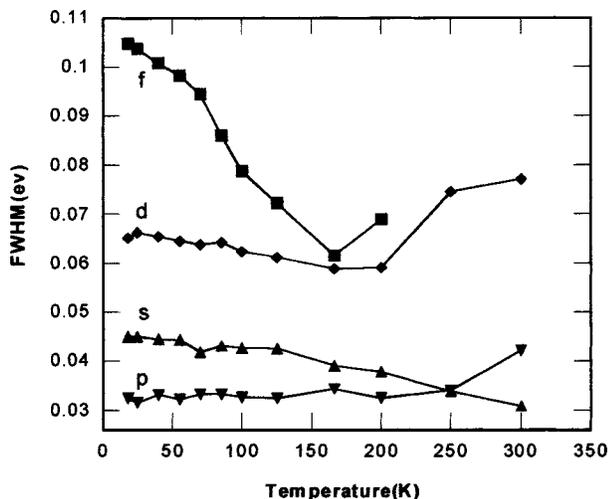


FIG. 7. Temperature dependence of the FWHM of PL spectra of two-period stacked InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL. The FWHMs of the ground state and excited state are denoted by *s*, *p*, *d* and *f* curves, respectively.

QD system move from smaller dots to larger ones via tunneling. The carriers recombine radiatively at nearby larger dots based on the transfer and relaxation processes of the carriers from smaller dots to larger ones. It is clear in Fig. 7 that the FWHM decreases and then increases as the temperature increases. At low temperature, the PL linewidth of quantum dots is determined by the inhomogeneous broadening coming from size fluctuation of the dot size. That is, PL arising from all of the emission from many dots of different size is observed. For the higher temperature, more carriers will become thermalized in the quantum dots and the carriers will tunnel and redistribute among nearby QDs and find a local-energy minimum in each energy shell. Thus, the PL linewidth will be reduced for the higher temperature. When the temperature is higher than -200 K, electron-phonon scattering becomes a dominant contribution. Then the PL linewidth starts to increase for the higher temperature. The two-stage behavior of the temperature dependence of the FWHM of PL spectra will thus be understood.^{22,23}

In Fig. 7, it is obvious that the behavior is strongly dependent on the energy levels. The FWHM of the PL spectra for the *f* energy level decreases for the higher temperature up to ~ 170 K and then increases for the higher temperature. For the *D* and *P* energy levels, the FWHM starts to decrease until the temperature is 200 and 250 K, respectively. However, the FWHM of the *S* energy level does not show the increasing behavior up to 300 K. We define the temperature of the minimal FWHM as T_f . The temperature T_f is the turning point of the dominant factor to determine the PL linewidth changing from the repopulation process to electron-phonon scattering for the higher temperature. It is found that temperature T_f is higher for the lower shell energy levels. This behavior can be explained by the fact that the carriers at high levels (excited states) can easily become thermalized due to the broader bandwidth of the excited states, which shifts T_f of the higher levels to the lower temperature.

The FWHM of the PL peak from the ground state of InAs QDs is 30 meV at room temperature, and this is smaller than the 40–60 meV usually reported at room temperature (with the exceptional report of 21 meV at room temperature).²⁴ Since this value is usually associated with inhomogeneous broadening of QDs, the InAs QDs embedded in the $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL will be more uniform.

In order to identify the mechanism of PL quenching, the temperature dependence of the integrated PL intensities of two-period stacked InAs QDs embedded in a $\text{GaN}_{0.007}\text{As}_{0.993}$ SCL was investigated. The temperature dependence of the integrated PL intensity of each excited level evaluated from the fit is shown in Fig. 8, where the dependence of the PL intensity on the temperature can be written by the following equation:

$$I(T) = A/[1 + B \exp(-Ea/kT)],$$

where I is the integrated PL intensity, A and B are two constants, Ea is the thermal activation energy and k is the Boltzmann constant. The estimated thermal activation energy Ea values are shown in Table I for the *s*, *p*, *d* and *f* states. From the fit in the higher temperature range, it is found that the activation energy also shows an excited-state dependence.

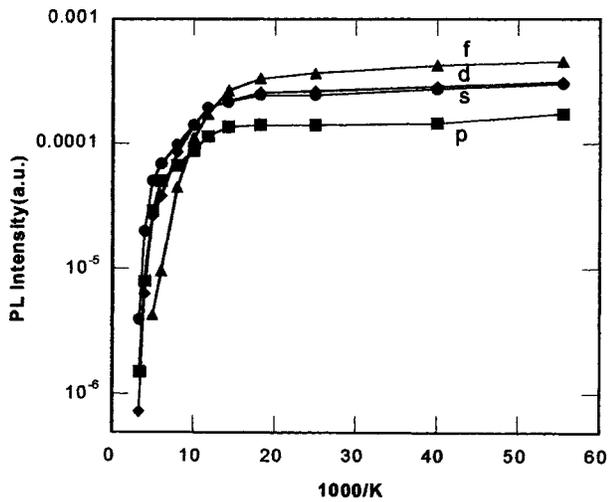


FIG. 8. Temperature dependence of integrated PL intensities of two-period stacked InAs QDs embedded in a GaN_{0.007}As_{0.993} SCL. The integrated intensities of the ground state and excited state are denoted by *s*, *p*, *d* and *f* curves, respectively.

The differences in energy between the PL emission of InAs QDs and the estimated band gap of the GaAsN layer are also given in Table I for the *s*, *p*, *d* and *f* states. These values are very close to the estimated thermal activation energies of the *s*, *p*, *d* and *f* states. Therefore the reduction of the PL intensities with an increase in temperature will be mainly due to the thermal activation of carriers out of the InAs quantum dots into the GaNAs barrier layers, followed by the nonradiative recombination during diffusion in the barrier layers.

Table II shows the barrier band gap energy E_g , thermal activation energy E_a , PL peak energy E_{PLs} of the *s* state, and PL peak energy E_{WL} of the additional peak of InAs QDs embedded in GaNAs and of the wetting layer of those embedded in GaAs spacer layers previously reported. We can see in Table II, the E_a value for the ground state obtained from the InAs QDs embedded in the GaN_{0.007}As_{0.993} SCL is about two times larger than the corresponding values reported for those in GaAs (105 meV).²² This result indicates that the InAs QDs embedded in the GaNAs SCL have a better quantum confinement effect. The thermal activation energy will be determined by the difference in energy ΔE between the PL emission of InAs QDs and the band gap energy of the spacer layers or wetting layer for high quality materials. We can see in Table II, the difference in energy ΔE for InAs quantum dots embedded in GaAs and GaNAs spacer layers is 175 and 236 meV, respectively. This indicates the thermal activation energy for high quality InAs QDs embedded in the GaNAs SCL is much larger than that

TABLE I. Thermal activation energies E_a of the *s*, *p*, *d*, and *f* states of InAs QDs and the differences in energy between the band gap of GaAsN and the PL emission of the *s*, *p*, *d* and *f* states of InAs QDs.

	<i>f</i> (meV)	<i>d</i> (meV)	<i>p</i> (meV)	<i>s</i> (meV)
E_a	84	112	157	222
$\Delta E = E_g - E_i$ <i>i</i> = <i>s, p, d, f</i>	77	119	181	236

TABLE II. Band gap energy E_g , and thermal activation energy E_a . PL peak energy E_{PLs} of the *s* state and PL peak energy E_{WL} of the wetting layer of InAs QDs embedded in both GaNAs and GaAs spacer layers. ΔE denotes the difference in energy between the PL emission of InAs QDs and the band gap energy of the lower barrier, either the spacer layer or wetting layer.

Spacer layer	E_{PLs} (eV)	E_{WL} (eV)	E_g (eV)	ΔE (eV)	E_a (eV)
GaN _{0.007} As _{0.993}	1.094	(1.42)	1.33	0.236	0.222
GaAs	1.255 ^a	1.43 ^b	1.51 ^a	0.175 ^a	0.105 ^a

^aReference 22.

^bReferences 13–16.

embedded in GaAs spacer layers. In the InAs QDs embedded in GaAs spacer layers, however, the thermal activation energy is much less than the difference in energy ΔE . This indicates that InAs quantum dots embedded in GaAs spacer layers have a higher density of defects and dislocations. Any defects and dislocations in the material will provide nonradiative channels to quench the luminescence, which will reduce the thermal activation energy.

The PL peak energy that appeared at 1.42 eV is very close to that of a wetting layer previously reported with GaAs capping layers.^{13–15} However the observed PL peak energy is larger than the band gap energy of the GaN_{0.007}As_{0.993} spacer layer. This difference in energy cannot be explained by the normal quantum confinement effect in the InAs WL. PL peaks at similar energy position have been reported after thermal annealing, and their origin was attributed to intermixed states near the heterointerface.²⁵ Details of the origin of this additional PL peak is under study.

IV. CONCLUSIONS

Self-assembled InAs quantum dots embedded in a GaN_{0.007}As_{0.993} SCL were grown by MOMBE on a GaAs (001) substrate with a high density of $1 \times 10^{11} \text{ cm}^{-2}$. The PL properties were studied for two-period stacked InAs quantum dots embedded in the GaN_{0.007}As_{0.993} SCL. Four well-resolved excited-state peaks were observed in PL spectra for InAs QDs embedded in the GaN_{0.007}As_{0.993} SCL in spite of the high QD density. This was attributed to improved uniformity by the relief of compressive strain in InAs QDs with the tensile GaNAs SCL. The state filling effect was also observed. It was found there was a slight redshift of the PL peaks with an increase in excitation intensity. This was explained by band gap renormalization due to a carrier–carrier exchange interaction. Strong photoluminescence was observed, and even a PL spectrum for the low excitation intensity of 0.019 W/cm^2 was obtained, which indicates that the InAs QDs embedded in the GaN_{0.007}As_{0.993} SCL have excellent quality. It was found that the linewidth of PL spectra decreased for the ground state, and the linewidth first decreased and then increased for the excited state with an increase in temperature. This was interpreted by the fact that relaxation of the photogenerated carriers can involve two different processes, the repopulation process and electron–phonon scattering. The thermal activation energy of each state was obtained, and we attribute the photoluminescence quenching mechanism as being due to the escape of carriers

from InAs QDs to nonradiative recombination centers through the GaNAs SCL. The FWHM of the PL spectrum from the ground state InAs QDs is about 30 meV at room temperature.

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