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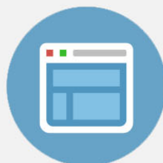

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Optical properties of InGaN quantum dots grown by SiN_x nanomasks

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InGaN quantum dots (QDs) deposited on SiN_x nanomasks have been investigated by atomic force microscopy, photoluminescence (PL), and photoluminescence excitation (PLE) measurements. It was found that the size of QDs can be well controlled by SiN_x nanomasks, enabling the manipulation of quantum confinement effect. The PL spectra of InGaN QDs contain several fine structures, and the main peaks can be attributed to families of QDs with different sizes. The emission arising from InGaN QDs and GaN buffer layer can be clearly distinguished based on PLE measurement, which can be used to improve the interpretation in the previous reports. Our study indicates that the quantum confined Stark effect due to piezoelectric field plays a very important role in the optical properties of InGaN QDs, which is very useful for the application of optoelectronic devices. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2717258]

I. INTRODUCTION

The group III-nitride wide-band-gap semiconductors have been recognized as very important materials for many optoelectronic devices, such as blue ultraviolet (UV) light emitting diodes (LEDs), laser diodes (LDs), and high-temperature/high-power electronic devices.[1–3] For example, InGaN/GaN quantum well (QW) structures have been successfully used as the active layers in LEDs and LDs.[1–3] However, the threshold current density is still high for LDs with InGaN QW structures. It is expected that using quantum dots (QDs) instead of QWs as the active layer can improve the performance of LDs.[4] Consequently, growth and characterization of InGaN QDs become very important for highly efficient blue and violet semiconductor lasers.[4] To ensure suitability for the optoelectronic applications, high density and uniform size of QDs must be achieved. Recently, several growth modes have been employed to grow InGaN QDs, including the Stranski-Krastanow mode and the deposition of antisurfactant such as Si or a SiN_x nanomasks facilitating the formation of auto-organized QDs.[4–6] However, a clear picture to describe luminescence from InGaN QDs is still not yet achieved. The detailed luminescence mechanism is quite complicated, which strongly depends on several different factors, including material composition, internal strain, QD size, excitation condition, as well as the method of QD formation. For example, the true indium content of small clusters is difficult to determine due to the shape and the small size of clusters. The large mismatch between InN and GaN lattices causes difficulties in their growth while film instability against spinodal decomposition often causes nonuniform distribution of indium aggregation.[7] Besides, due to large piezoelectric effect in this material, the quantum-confined Stark effect could be very important.[8] It has been controversial whether the dominant luminescence mechanism in InGaN/GaN originates from the presence of the In-rich cluster or from the internal electric field. In this paper, we investigate optical properties of InGaN QDs grown by selective growth on SiN_x-deposited GaN epilayer. By changing the duration of the SiN_x treatment, the wavelength covering visible region can be easily controlled. We demonstrate the fine structures in the photoluminescence (PL) spectra arising from families of QDs with different sizes. The emissions from InGaN QDs and from GaN buffer layer can be unambiguously distinguished based on the photoluminescence excitation (PLE) measurement. In addition, our measurements clearly indicate that the properties of InGaN QDs can be significantly influenced by the piezoelectric field. In view of the strong piezoelectric (PZ) field in InGaN QDs, the results shown here should be very important for their practical application.

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II. EXPERIMENT

The studied ultra-high-density InGaN QDs were grown by metal-organic chemical vapor deposition (MOCVD). A 30-nm-thick undoped GaN layer was first deposited on *c*-face sapphire substrates at 550 °C, followed by a 2-μm-thick Si-doped GaN underlying layer at 1000 °C. Then, NH_3 and the diluted Si_2H_6 were flowed through the sample to grow a rough SiN_x layer on the *n*-type GaN underlying layer with increasing the SiN_x treatment time from 120 to 195 s. The temperature was then reduced to 800 °C to grow the InGaN layers, and a 10 nm undoped GaN cap layer was grown on the InGaN layer to complete the sample structure. Atomic force microscopy (AFM) images of the uncapped samples are shown in Fig. 1, which indicate that the morphology of the surface changes from nanoislands to sharp QDs as the duration of the SiN_x treatment increases. The average widths, heights, and densities of the QDs in the samples are estimated to change from 33 to 35 nm, from 0.64 to 2.46 nm, and from 2.1×10^{11} to 2.9×10^{11} cm^{-2} with increasing thickness of the SiN_x layer. Therefore, the forma-

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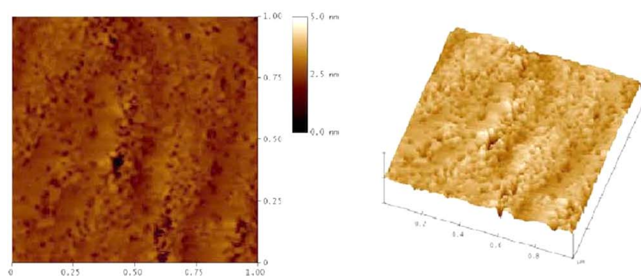
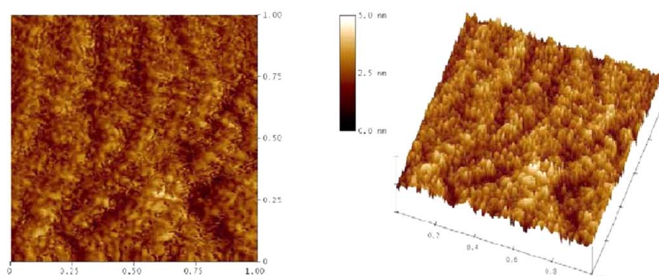
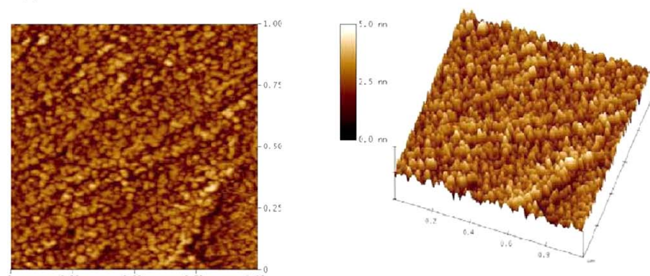
1(a) 120 s



1(b) 160 s



1(c) 178 s



1(d) 195 s

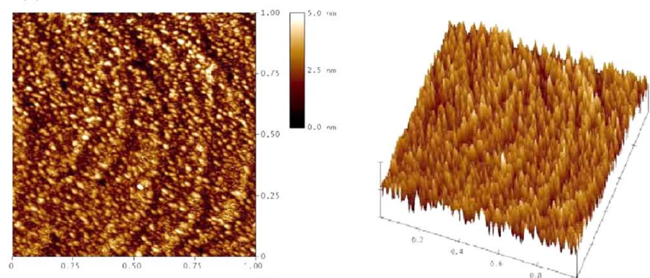


FIG. 1. (Color online) Atomic force microscopy images of InGaN layers with (a) 120 s, (b) 160 s, (c) 178 s, and (d) 195 s of SiN_x treatment on the underlying GaN layers.

tion of InGaN QDs could be controlled by just changing the duration of the SiN_x treatment. A detailed description of the growth process has been published elsewhere.[6] For the PL measurement, the sample was excited by a He–Cd laser working at 325 nm, dispersed by a Spex 0.85 m double-grating spectrometer, and the spectra were detected by a photomultiplier tube. Similar arrangements were used for the PLE measurement except the dispersed light from a Xe lamp was used as the continuous radiation.

III. RESULTS AND DISCUSSION

A. Dependence on SiN_x treatment time

Figure 2 shows the normalized PL spectra of InGaN QDs with the SiN_x treatment time of 120, 160, 178, and

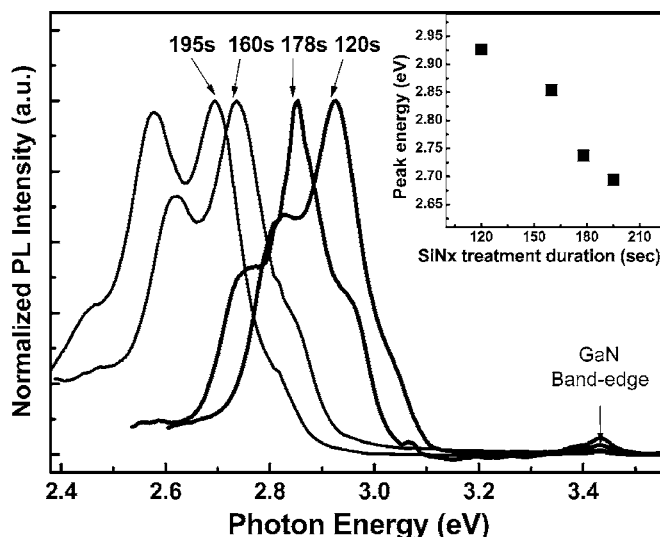


FIG. 2. Normalized photoluminescence spectra at room temperature of InGaN QDs with the SiN_x treatment time at 120 s, 160 s, 178 s, and 195 s. The inset shows the main peak energy of the PL spectra as a function of SiN_x treatment duration.

195 s. The InGaN QDs exhibit a very strong visible PL signal at room temperature, which even exceeds that of device quality of InGaN quantum wells. The strong PL signal results from the fact that the confinement of electron-hole pairs is very beneficial for enhancing radiative recombination, and the confined carriers have less probability to diffuse to nonradiative recombination centers. The PL spectra also clearly show that the signal arising from InGaN QDs is much stronger than that from GaN band gap emission. As shown in the inset of Fig. 2, the main peak has a redshift of about 232 meV when SiN_x treatment time increases from 120 to 195 s, which covers the entire visible region. According to the above AFM images, the redshift of the spectra could be simply attributed to the small quantum confinement effect with increasing the average height of InGaN QDs. It is worth noting that the PL spectrum of each InGaN QD sample contains two main peaks, which may arise from the transitions involving ground state and excited states of QDs, or families of QDs with different sizes.[9,10] As shown in Fig. 2, these two peaks have a comparable intensity and their energy separation is much larger than the thermal energy at room temperature; the possibility of involving ground and excited transitions can be ruled out because if the Boltzman distribution is taken into account, the carrier population in the excited state and the corresponding PL intensity should be very small. Apparently, this result clearly indicates that the double main peaks are due to different dot families.

B. Dependence on excitation intensity

To further investigate the optical properties of InGaN QDs, we have performed the PL measurements under different excitation densities, as shown in Fig. 3. It is found that the PL intensity is linearly proportional to excitation density, as shown in the inset of Fig. 3, indicating that the transition involves an excitonic recombination. It is also found that the main peak position of the PL spectra shows a blueshift as the

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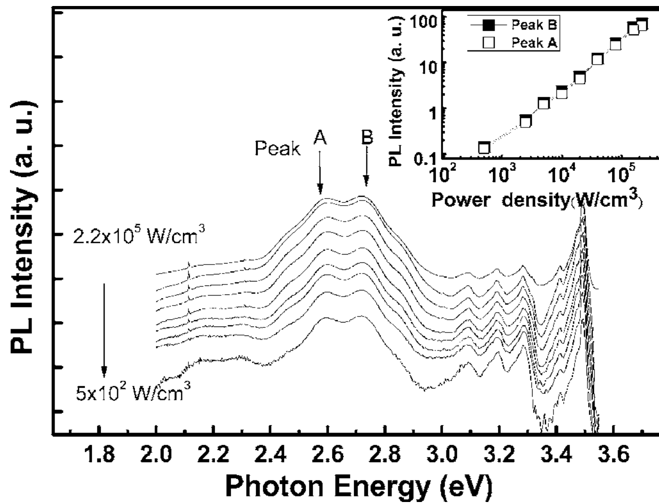

FIG. 3. Photoluminescence spectra of InGaN QDs with different excitation densities at room temperature. The inset shows the PL intensity of main peak as a function of excitation power density.

optical excitation density increases. In the typical case of the quantum confined Stark effect, such as in InGaN/GaN multiple quantum wells,[11,12] the blueshift was frequently attributed to the screening of built-in PZ field. The underlying physics is based on the spatial separation of photoexcited electrons and holes due to the built-in PZ field. Because the induced field is opposite to the internal PZ field, the resultant field is therefore reduced. This result demonstrates that the PZ field is important in determining the optical property of InGaN QDs.

C. Temperature dependence

Figure 4(a) shows temperature dependent PL spectra of InGaN QDs with the SiN_x treatment time of 195 s under high excitation density. When the temperature increases from 20 to 30 K, the peak shift of InGaN QDs is about 30 meV, while that of the InGaN bulk is about 90 meV. It is well known that the redshift of the PL peak position of a semiconductor with increasing temperature originates from electron-phonon interaction. Thus, the smaller redshift observed in InGaN QDs than that in bulk material can be explained by partial decoupling of the phonon-exciton interaction in QDs.[13] The structure covering from 3.0 to 3.3 eV at low temperature can be attributed to donor-acceptor pair (DAP) bands of GaN buffer layer. A clear evidence to support our assignment is that the phonon replica is in exact agreement with the LO phonon energy of GaN. Another feature is the yellow band at about 2.2 eV. The band is associated with deep levels due to point defects such as Ga or N vacancies.[14] It is worth noting that the integrated PL intensity from InGaN QDs is about 19 times higher than that from GaN at low temperature, and the difference increases with temperature. The improvement of optical recombination in QDs at high temperature can be related to the quantum confinement effect, in which the strong wave function overlap between electron and hole leads to an enhanced exciton binding energy and transition probability.

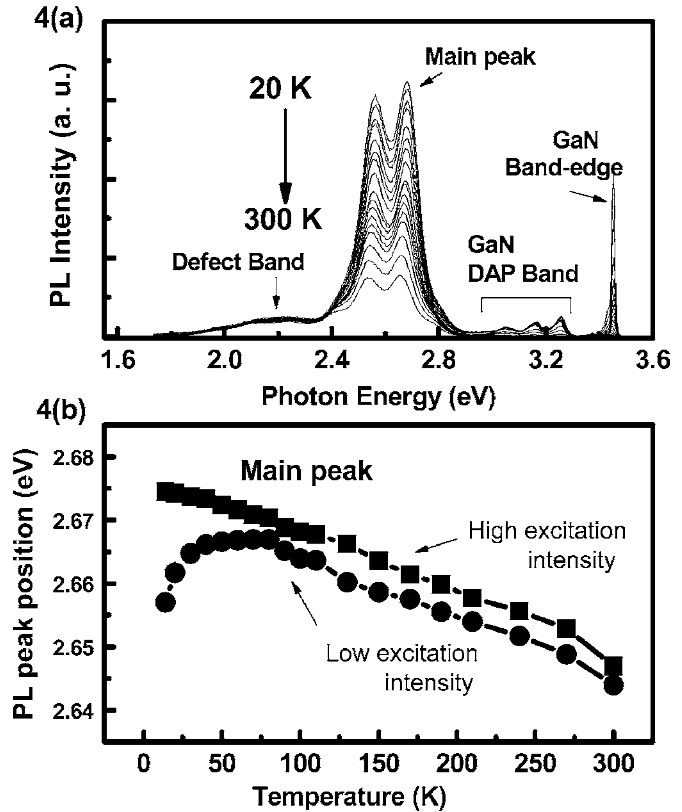

FIG. 4. (a) Temperature dependent photoluminescence spectra of InGaN QDs with the SiN_x treatment time at 195 s. (b) Temperature dependence of the photoluminescence peak position under high and low excitation intensities.

Figure 4(b) shows the PL peak position of InGaN QDs with the SiN_x treatment time of 195 s as a function of temperature under high and low excitation intensities. Under high excitation intensity, the piezoelectric field is severely screened and the PL temperature dependence is similar to that of bulk material, i.e., the band gap decreases with increasing temperature. On the other hand, under low excitation intensity, a blueshift in the PL peak is observed when temperature increases from 20 to 90 K. We can also understand this abnormal PL blueshift based on the screening of the piezoelectric field due to thermal excitation. At low temperature, carriers are frozen in nonradiative centers, and they will be thermally activated as temperature increases. This process will induce partial screening of the piezoelectric field and lead to the observed blueshift in the PL spectra. The temperature dependent PL spectra thus reflect a competition between the blueshift induced by the thermal screening of the piezoelectric field and the redshift induced by electron-phonon interaction.

In order to determine the activation energy of the electrons confined in InGaN QDs, we have performed integrated PL intensity as a function of the reciprocal temperature in InGaN QDs under different SiN_x duration times, as shown in Fig. 5. As the temperature increases, the variation of integrated PL intensity can be expressed as[15]

$$I(T) = I(0)/1 + C \exp(-E_A/k_B T), \quad (1)$$

where $I(0)$ is the integrated PL intensity at 0 K, E_A is the activation energy, C is a fitting parameter, and K_B is the

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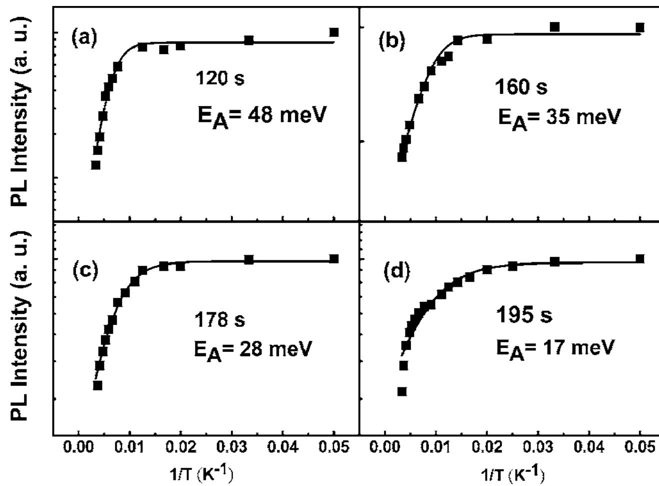

FIG. 5. Integrated photoluminescence intensity as a function of the reciprocal temperature of InGaN QDs with the SiN_x treatment time at (a) 120 s, (b) 160 s, (c) 178 s, and (d) 195 s. The continuous lines are the best fit for the samples according to the Arrhenius plot.

Boltzmann constant. Using Eq. (1), the activation energy E_A is 48, 35, 28, and 17 meV in InGaN QDs with SiN_x duration time of 120, 160, 178, and 195 s, respectively. Higher activation energy in InGaN QDs originates from a reduction in the dimensions of QDs, which is a consequence of the increased Columb interaction due to carrier confinement.

D. Photoluminescence excitation measurement

Figure 6 shows the PL and PLE measurements in InGaN QDs with the SiN_x treatment time of 195 s at room temperature. The PL and PLE spectra were measured by using the dispersed light from a xenon lamp as the excitation source. The PL spectrum is excited by a 3.35 eV radiation and the PLE spectra is monitored at different PL peaks, as shown in Fig. 6. Quite interestingly, the optical properties of PLE spectra with different monitoring wavelengths show very interesting behaviors. Especially, the emissions arising from InGaN QDs and GaN defects are quite different. The peak

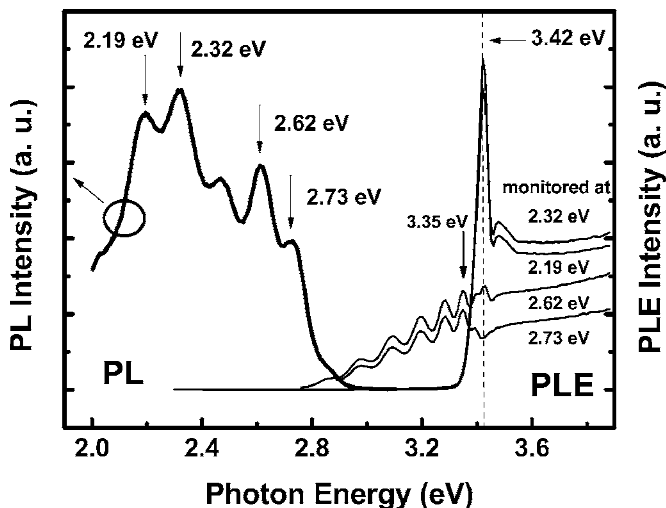

FIG. 6. Photoluminescence (PL) and photoluminescence excitation (PLE) spectra of InGaN QDs with the SiN_x treatment time at 195 s at room temperature.

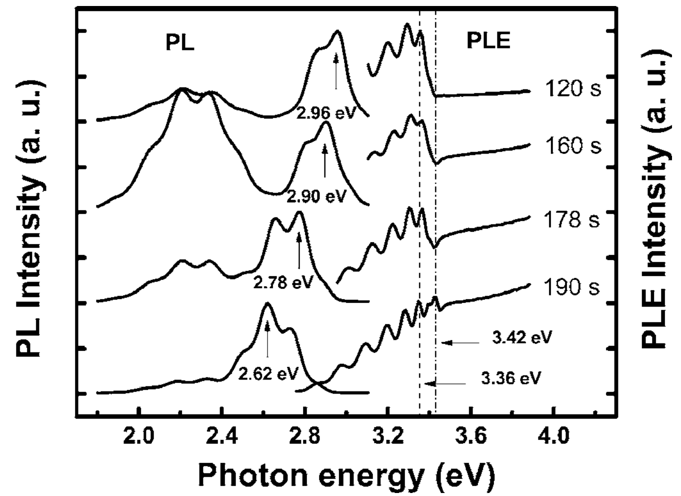

FIG. 7. Photoluminescence (PL) and photoluminescence excitation (PLE) spectra of InGaN QDs with the SiN_x treatment time from 120 to 195 s at room temperature.

energies at 2.19 and 2.32 eV can be attributed to the transitions in the GaN layer, because the PLE intensity can be greatly enhanced when the excitation energy is in resonance with the band gap transition of the GaN layer. However, PLE spectra monitored at 2.62 and 2.73 eV are quite different from those monitored at 2.19 and 2.32 eV. The PLE intensities monitored at 2.62 and 2.73 eV become larger even when the excitation energy does not belong to the transitions in the GaN layer. Therefore, they can be attributed to the excitonic transition in InGaN QDs. The spectral modulation below 3.36 eV in the PLE spectra is very interesting. Previously, its absorption has been identified as excitonic resonance in InGaN QDs, and therefore a giant Stokes shift was inferred.[16] As shown in Fig. 7, we summarize the PL and PLE spectra of InGaN QDs with the SiN_x treatment time from 120 to195 s at room temperature. All the PL spectra were excited by 3.36 eV using a xenon lamp as the excitation source, and the PLE spectra were monitored at the main peak positions, as shown in Fig. 7. Here, we only show the PLE spectra taken at the main PL emission peak in InGaN QDs with the SiN_x treatment time from 120 to 195 s. Similar behavior in PLE spectra was observed when the detection energies were varied in the region of QD radiation. All the PLE spectra in the region below 3.36 eV clearly show an oscillatory behavior, and the oscillatory structure does not shift with the change of QD size. Therefore, our result clearly indicates that the oscillatory behavior does not arise from the excitonic resonance in QDs, as reported previously.[16] Instead, it may originate from the absorption of the SiN_x or the wetting layer. We thus conclude that the PLE spectra monitored at different wavelengths are very useful to identify the origin of optical properties in multilayer structures.

IV. CONCLUSION

In conclusion, we have reported a highly efficient emission of ultra-high-density InGaN QDs with controllable optical properties by SiN_x nanomasks. Based on the temperature dependent PL spectra under high and low optical

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excitations, it is shown that the piezoelectric field plays a very important role in determining the emission from InGaN QDs. In addition, with the measurement of PLE spectra monitored at different photon energies, one can clearly distinguish the emission arising from QDs and buffer layer, which enables the clarification of the misunderstanding in previous reports. Our results also suggest that it is possible to fabricate full-color and high-brightness InGaN QDs LED based on the assistance of SiN_x nanomasks.

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