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2004 Jpn. J. Appl. Phys. 43 L264

(http://iopscience.iop.org/1347-4065/43/2B/L264)

Japanese Journal of Applied Physics
Vol. 43, No. 2B, 2004, pp. L264–L266

Ultra-High-Density InGaN Quantum Dots Grown by Metalorganic Chemical Vapor Deposition

Ru-Chin TU*, Chun-Ju TUN[1], Chang-Cheng CHUO, Bing-Chi LEE[2], Ching-En TSAI,
Te-Chung WANG, Jim CHI, Chien-Ping LEE[2] and Gou-Chung CHI[1]

Opto-Electronics and System Laboratories, Industrial Technology Research Institute, Hsinchu, Taiwan 310, Republic of China
[1]*Institute of Optical Science, National Central University, Chung-Li 32054, Taiwan, Republic of China*
[2]*Department of Electronic Engineering, National Chiao Tung University, Hsinchu, Taiwan 30050, Republic of China*

(Received August 28, 2003; accepted September 30, 2003; published February 6, 2004)

This study examined how the duration of SiN_x treatment on an underlying GaN layer affects the optical property, surface morphology and density of following InGaN quantum dots (QDs). InGaN QDs with extremely high density of near $3 \times 10^{11}\,cm^{-2}$ exhibited strong photoluminescence (PL) emission at room temperature (RT). Increasing the duration of the SiN_x treatment of the underlying GaN layer, the RT-PL peak of the following InGaN nano-islands and QDs was found to be red-shifted from the violet to the greenish region, and the spectrum was broadened. Additionally, the average height of InGaN nano-islands and QDs increased with the duration of SiN_x treatment, explaining the redshift of the RT-PL peak.
[DOI: 10.1143/JJAP.43.L264]

KEYWORDS: InGaN, quantum dot, density, SiN_x, photoluminescence, atomic force microscopy

GaN and related materials are currently the subjects of intense research due to their applications in laser diodes (LDs) and light-emitting diodes (LEDs) that operate between the ultraviolet and the visible regions.[1)] InGaN/GaN quantum wells (QWs) structures have successfully been used as the active layers in LEDs and LDs.[1)] However, the threshold current density is high for LDs with InGaN QWs structures. Having quantum dots (QDs) instead of QWs as the active layer is expected to improve the performance of LDs. LDs with QDs structures in the active layer have been theoretically predicted to have superior characteristics, including lower threshold currents and narrow spectra.[2)] Moreover, because of the localization of carriers trapped at dislocations, QDs structures have been expected to increase the efficiency of the luminescence of LDs.[3)] To ensure suitability for QDs laser applications, QDs layers with high spatial density and of uniform size must be grown.[4)] Several approaches have been investigated for fabricating InGaN QDs, including the Stranski-Krastanow growth mode[5,6)] and growth using anti-surfactant.[7,8)] The deposition of silicon anti-surfactant or a SiN_x nano-mask alters the morphology of the AlGaN films from that of step flow to that of a three-dimensional island, facilitating the formation of GaN[7)] QDs and InGaN QDs[8)] on the AlGaN.

This study investigates the optical property, the surface morphology and the density of InGaN QDs following different durations of SiN_x treatment on the underlying GaN layer before the InGaN layers were deposited. InGaN QDs with a very high density of near $3 \times 10^{11}\,cm^{-2}$ and strong photoluminescence (PL) intensity were obtained. Adjusting the duration of the SiN_x treatment of the underlying GaN layer was found to shift the room temperature (RT)-PL peak from the violet to the greenish region and to broaden the spectrum. The relationship between the average height and the PL-peak wavelength of InGaN nano-islands and QDs is also addressed.

Eight InGaN samples that had undergone different durations of SiN_x treatment of the underlying GaN layer before the InGaN layer was deposited, were grown on c-face sapphire substrates by metalorganic vapor phase epitaxy (MOVPE).[9–11)] A 300 Å-thick low-temperature GaN nucleation layer was first grown at 550°C. Then, the reactor temperature was increased to 1000°C to grow a 2 μm-thick underlying Si-doped GaN underlying layer, providing a step-flow grown surface as confirmed by atomic force microscopy (AFM). Then, a rough SiN_x layer with varying thickness (or different durations of SiN_x treatment) was grown on the n-type GaN underlying layer. During the treatment of the SiN_x layer, the flow rates of NH_3 and the diluted Si_2H_6 were 5 slm and 50 sccm, respectively. The temperature was then ramped down to 800°C to grow the $In_xGa_{1-x}N$ layers. As soon as the InGaN layers deposition was complete, the growth temperature was reduced to room temperature. During the growth of the InGaN layers, the vapor phase ratio TMIn/(TMIn+TEGa) was fixed at 0.35. Additional eight InGaN layers capped with a 10 nm un-doped GaN layer, were grown to investigate the optical property of the InGaN layers. The detailed growth conditions of InGaN samples that had undergone various durations of SiN_x treatment of the GaN were examined, as listed in Table I. The optical characteristics of these eight epitaxial samples were evaluated using RT-PL with a low 5 mW HeCd laser operated at 325 nm. The surface morphology of all samples grown was characterized by AFM. Scans were performed over a surface area of 500 nm, using a Digital Instruments Nanoscope with a sharpened Si_3N_4 tip.

Figures 1(a)–1(f) show AFM images of the surface morphology of the samples QW-A, QD-B, QD-C, QD-D, QD-E and QD-G, respectively. Figure 1(a) shows a surface with a step-like pattern, but a slightly rough 20 monolayers (MLs) InGaN layer that had not undergone any SiN_x treatment of the GaN surface. In contrast, Figs. 1(b)–1(f) show that the morphology of the surfaces changes from that of network-like nano-islands to sharp QDs as the duration of the SiN_x treatment increases. Therefore, three-dimensional growth was observed on the samples that had undergone SiN_x treatment (or to which Si was applied as an anti-surfactant) and the formation of InGaN QDs could be controlled by just increasing the duration of the SiN_x treatment. Notably, as the duration of the SiN_x treatment increased from 390 to 420 s, the average height and the dot density of InGaN QDs were estimated to increase approximately from 3.6 to 4.1 nm and from $2.1 \times 10^{11}\,cm^{-2}$ to $2.9 \times 10^{11}\,cm^{-2}$, respectively.

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*E-mail address: RuChinTU@itri.org.tw

Table I. Detailed growth conditions of InGaN samples investigated in this study.

Sample No.	SiN_x Treatment Time	InGaN QDs Deposition	PL Peak Wavelength (with 10 nm GaN Cap)	QDs Height	QDs Density
QW-A	0 s	20 MLs	NA	NA	NA
QD-B	240 s	20 MLs	435 nm	0.8 nm	NA
QD-C	320 s	20 MLs	445 nm	1.8 nm	NA
QD-D	360 s	20 MLs	465 nm	2.7 nm	NA
QD-E	390 s	20 MLs	480 nm	3.7 nm	2.1×10^{11} cm^{-2}
QD-F	420 s	10 MLs	458 nm	NA	NA
QD-G	420 s	20 MLs	495 nm	4.1 nm	2.9×10^{11} cm^{-2}
QD-H	420 s	30 MLs	510 nm	6.0 nm	1.9×10^{11} cm^{-2}

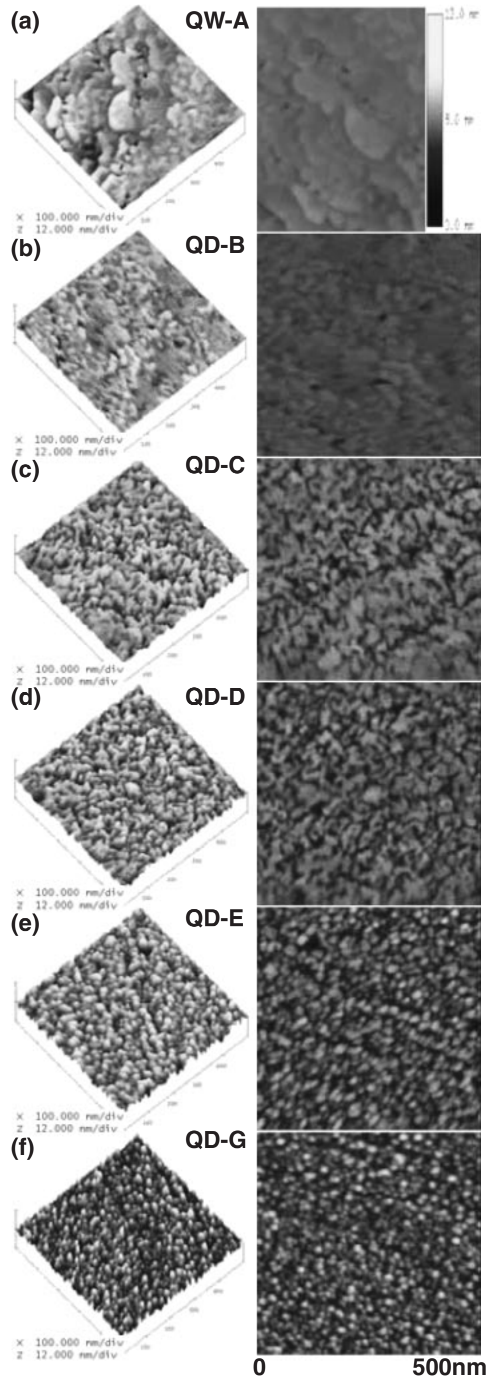

Fig. 1. 500 nm × 500 nm AFM images of InGaN layers with (a) 0 s, (b) 240 s, (c) 320 s, (d) 360 s, (e) 390 s, and (f) 420 s of SiN_x treatment on the underlying GaN layers.

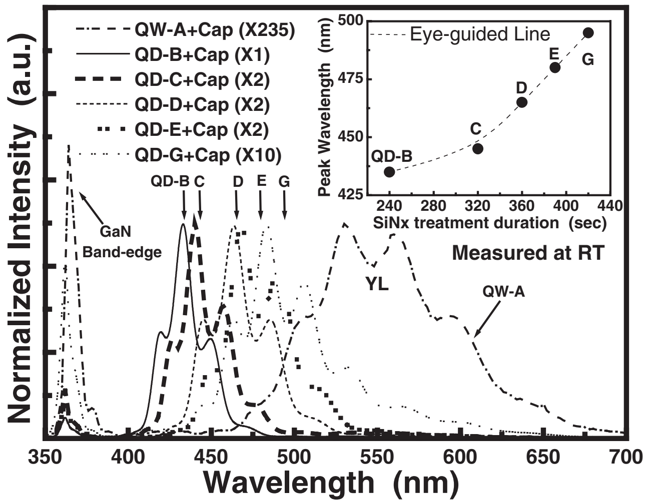

Fig. 2. Normalized RT PL spectra from the samples QW-A, QD-B, QD-C, QD-D, QD-E and QD-G. The inset plots the approximate RT-PL-peak wavelength with increasing duration of SiN_x treatment.

InGaN QDs, capped with a 10 nm GaN layer grown at the same temperature as the InGaN QDs, were grown to investigate the optical properties of InGaN QDs. Figure 2 shows the corresponding normalized RT PL spectra from the samples QW-A, QD-B, QD-C, QD-D, QD-E and QD-G. The inset in Fig. 2 presents the approximate RT-PL-peak wavelength of the InGaN QDs samples as the duration of SiN_x treatment increases. The RT-PL-peak wavelength of the InGaN QDs samples is red-shifted as the duration of SiN_x treatment increases. As listed in Table I, the average heights of the InGaN nano-islands and QDs were approximately estimated from the increase in AFM from 0.8 nm to 4.1 nm as the duration of the SiN_x treatment increased from 240 to 420 s. The redshift as the duration of the SiN_x treatment increased could be attributed to the increasing height of the QDs (or nano-islands) due to quantum effects. For comparison, the RT-PL emission of the InGaN QW-A sample with an estimated thickness of 5 nm (20 MLs), based on growth rate, was much weaker than those of the nano-islands and QDs, indicating that the efficiency of luminescence was increased by the additional carrier confinement in the nano-islands and QDs samples.

Next, the dependence of the InGaN QDs' properties on the amount of InGaN deposited was examined. Figures 3(a)–3(c) show AFM images of the surface morphology of the InGaN QDs samples QD-F, QD-G and QD-H, respectively.

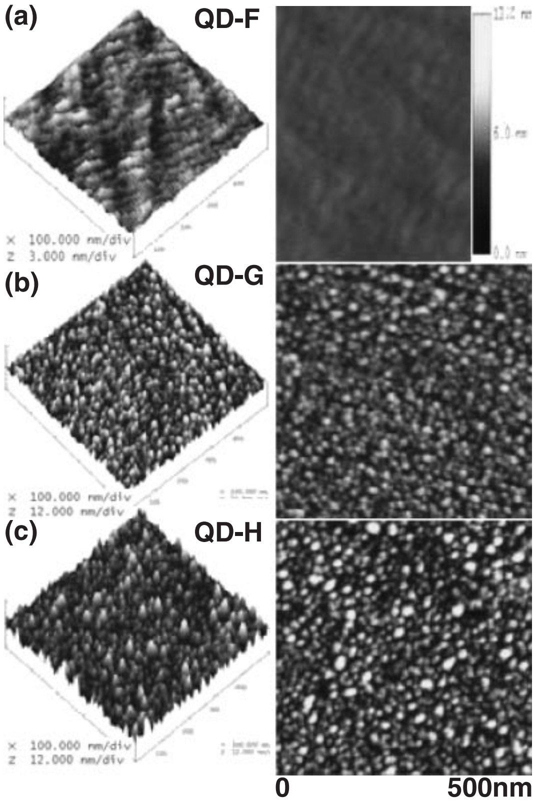

Fig. 3. 500 nm × 500 nm AFM images of InGaN layers for the deposition of (a) 10 MLs, (b) 20 MLs, and (c) 30 MLs.

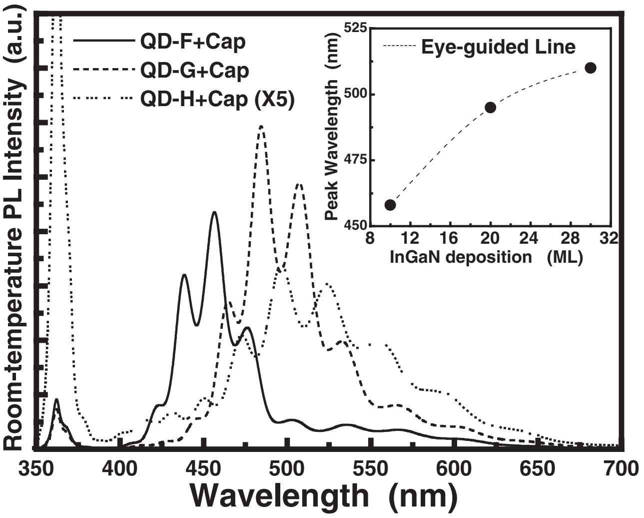

Fig. 4. RT PL spectra of the InGaN QDs samples F, G and H. The inset plots the approximate RT-PL-peak wavelength as the amount of InGaN deposited is increased.

The corresponding amounts of InGaN deposited in the samples QD-F, QD-G and QD-H, are 10, 20 and 30 MLs, respectively, at a fixed gas flow rate, growth temperature and SiN_x treatment duration of 420 s. At 10 MLs, only network-like nano-islands, but no dots were formed. InGaN QDs were formed when 20 MLs of InGaN were deposited, and the QDs density was then $2.9 \times 10^{11}\,cm^{-2}$. At 30 MLs, the InGaN QDs density decreased to $1.9 \times 10^{11}\,cm^{-2}$. The decline in the density of QDs with the increase in the amount of InGaN deposited from 20 to 30 MLs could be attributed to the merging of two or three small QDs into one bigger single one. Figure 4 presents the corresponding RT PL of the InGaN QDs samples QD-F, QD-G and QD-H. As shown in Table I, the average height of the InGaN nano-islands and QDs was estimated approximately from the increase in AFM from 4.1 to 6.0 nm as the amount of InGaN QDs increased from 20 to 30 MLs. The inset in Fig. 4 also presents the approximate RT-PL-peak wavelength as the amount of InGaN deposited increased. Therefore, the red-shift that occurred as the amount of InGaN deposited increased, could be attributed to heightened nano-islands and QDs, due to quantum effects.

In conclusion, this study has investigated the optical properties, surface morphology, and density of InGaN QDs that had undergone different durations of SiN_x treatment of the underlying GaN layer, before the InGaN layers were deposited. InGaN QDs with very high densities of near $3 \times 10^{11}\,cm^{-2}$ and high RT PL intensity were obtained. Increasing the duration of the SiN_x treatment of the GaN layer red-shifted the RT-PL peak of the InGaN nano-islands and QDs from the violet to the greenish region, and broadened the spectrum. Additionally, the average height of the InGaN nano-islands and QDs increased with the duration of the SiN_x treatment, explaining the red-shift of the RT-PL peak.

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