Quasiregular quantum-dot-like structure formation with postgrowth thermal annealing of InGaN/GaN quantum wells

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Postgrowth thermal annealing of an InGaN/GaN quantum-well sample with a medium level of nominal indium content (19%) was conducted. From the analyses of high-resolution transmission electron microscopy and energy filter transmission electron microscopy, it was found that thermal annealing at 900 °C led to a quasiregular quantum-dot-like structure. However, such a structure was destroyed when the annealing temperature was raised to 950 °C. Temperature-dependent photoluminescence (PL) measurements showed quite consistent results. Blueshift of the PL peak position and narrowing of the PL spectral width after thermal annealing were observed. [DOI: 10.1063/1.1467983]

Quantum-dot-like structures have been observed in InGaN/GaN quantum wells (QWs). Such structures are formed because of the large lattice mismatch between InN and GaN.[1–3] With high-resolution transmission electron microscopy (HRTEM), randomly distributed clusters of indium aggregation and phase-separated InN were widely observed. The cluster structures form potential minimums (called localized states) for trapping carriers for efficient photon emission.[4–7] Such indium-aggregated distributions are basically located in the quantum-well layers; however, they usually diffuse into barriers, extensively under certain conditions. Typically, such out-diffusion becomes more extensive in a sample with a higher nominal indium content.[3]

It has been reported that postgrowth thermal annealing could alter the sizes and distributions of self-organized InAs quantum dots.[8,9] In InGaN compounds, postgrowth thermal annealing processes for changing cluster structures and their photon emission properties were also reported.[10–12] In one of the previous results, a better-confined quantum-well structure, i.e., weaker indium out-diffusion, after thermal annealing was reported.[12] The photoluminescence (PL) intensity was increased with the stronger quantum-well confinement effect. Postgrowth thermal annealing can provide means for device manufacturers to tune the photon emission wavelength or to tailor the gain spectrum by changing the size/composition of the quantum-dot-like structures. The information of thermal annealing is also helpful for crystal growers to design growth procedures. In this letter, we report the formation of quasiregular quantum-dot-like structures from randomly distributed indium-aggregated clusters after postgrowth thermal annealing of an InGaN/GaN QW sample. It was found that with thermal annealing temperature at 900 °C, indium distribution was better confined in the quantum-well layers. Although the better confinement in a high-indium sample (>45%) has been reported,[12] the observation of quasiregular quantum-dot-like structures has never been reported. In particular, the indium content of our sample is much lower (19%). Also, we found that such a trend of regular structure disappeared when the thermal annealing temperature was 950 °C. The measurements of optical properties showed quite a consistent trend.

The sample used in this study was grown with a low-pressure metal–organic chemical-vapor deposition reactor. The InGaN/GaN QW sample consisted of ten periods of InGaN wells with 3.5 nm thickness. The barrier was 10 nm GaN. In the sample, the QW layers were sandwiched between a 1.5 μm GaN buffer layer on a (0001) sapphire substrate and a 50 nm GaN cap layer. The growth temperatures were 1010 and 720 °C for GaN and InGaN, respectively. The designated indium content of the sample was about 19%. As-grown samples were thermally annealed in a quartz tube furnace at different temperatures ranging from 800 to 950 °C in nitrogen ambient for 30 min. HRTEM and energy filter transmission electron microscopy (EFTEM) were used to characterize material structures.

For HRTEM observation, cross-sectional samples were prepared in the conventional manner by grinding, dimpling,

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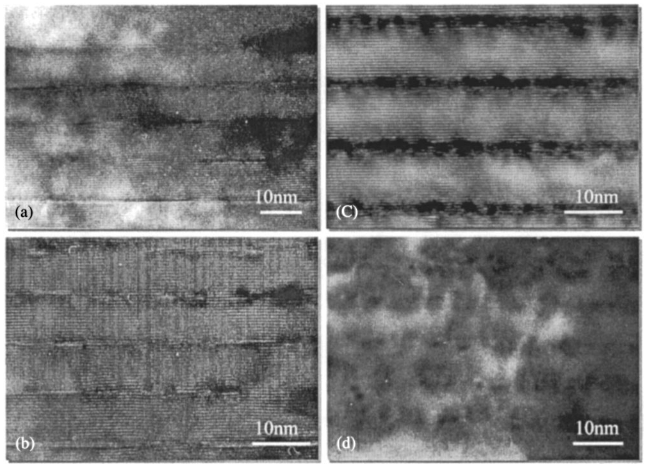

FIG. 1. HRTEM bright-field images of the as-grown (a), 800 °C-annealed (b), 900 °C-annealed (c), and 950 °C-annealed (d) samples.

and Ar^+-ion milling with 6 kV, 1 mA, and an incident angle of 4°. Based on previous experiences, for samples containing indium, the ion-milling step needs to be carried out with the specimen holder at liquid-nitrogen temperature in order to minimize ion-beam damage. The HRTEM investigations were conducted with a 200 keV Philips CM 200 and a 300 keV JEM 3010 microscope. All the high-resolution micrographs were taken at Scherzer defocus and the sample was viewed along a [11–20] zone axis. The 300 keV JEM 3010 microscope was equipped with a 2 k×2 k slow-scan charge-coupled-device camera and a Gatan imaging filter (GIF). Regarding the EFTEM images, the drift between the two preedge and the postedge images was recorded by using the cross-correlation algorithm available in the Digital Micrograph software of the GIF. In order to remove the background contribution underneath the ionization edges and to obtain elemental maps, the jump-ratio method was used to avoid noise and artifacts. However, it cannot be quantified.

Figures 1(a)–1(d) show the HRTEM bright-field images of the as-grown (a), 800 °C-annealed (b), 900 °C-annealed (c), and 950 °C-annealed (d) samples. The contrast variations in the pictures represent the fluctuations of indium composition. The diffusive InGaN/GaN QW interfaces can be clearly seen in the as-grown sample [Fig. 1(a)]. The average size of indium-rich clusters is larger than 10 nm. The clusters are irregularly dispersed and extended into the GaN barriers. Increasing the annealing temperature leads to a better confinement of indium-rich clusters near InGaN QWs, as shown in Figs. 1(b) and 1(c). The average size of the indium-rich clusters becomes smaller after the annealing treatment. The size homogeneity of quantum-dot-like structures was also improved. Interestingly, one can observe that very-fine indium-rich quantum-dot-like structures (2–5 nm) were regularly dispersed inside the InGaN QWs after the annealing treatment at 900 °C [Fig. 1(c)]. However, further increase of annealing temperature to 950 °C leads to a highly irregular structure, as shown in Fig. 1(d). EFTEM scanning was performed for the samples. Indium composition profiles across and along the QWs were obtained. Line-scan results of indium composition (%) across QWs in the as-grown sample (a), and samples annealed at 900 °C (b) and 950 °C (c), are shown in Fig. 2. Compared to the as-grown sample, the

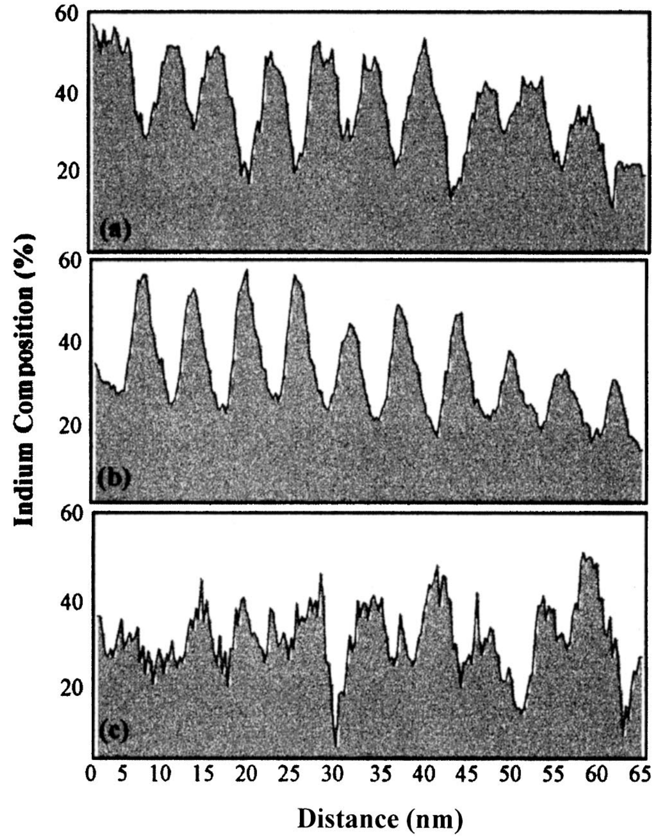

FIG. 2. EFTEM line-scan results of indium composition (%) across QWs in the as-grown sample (a), and samples annealed at 900 °C (b) and 950 °C (c).

sample annealed at 900 °C manifests better confinement of indium. However, after annealing at 950 °C indium becomes dispersive again. Figure 3 shows EFTEM line-scan results of indium composition (%) along a QW in the as-grown sample (a), and samples annealed at 850 °C (b), 900 °C (c), and 950 °C (d). Annealing did result in more regular structures of clusters. Quasiregularly arrayed quantum-dot-like structures with nearly the same indium concentration at the cores of the quantum-dot-like clusters can be observed in the sample with 900 °C annealing. The irregular distribution of compositional fluctuation can again be observed after postgrowth annealing at 950 °C. In the InGaN/GaN QW sample, thermal energy at 900 °C is sufficient to drive the system to a state of lower potential energy, i.e., a quasiregular structure. The relationship between the period of the indium-rich clusters and the width and nominal indium content of QWs is worth investigating further. When the annealing temperature reached 950 °C, the higher temperature caused coarsening of indium-rich clusters with the sizes exceeding the QW width.[8] Such a process destroyed the QW structure, leading to the results shown in Figs. 1(d), 2(c), and 3(d).

Figures 4 and 5 show the PL characteristics of these samples. Because of the oscillatory variations in PL spectra due to the Fabry–Pérot effect, normal PL peak position and full width at half maximum could not be accurately calibrated. PL peak positions were then obtained through Gaussian fitting, and the root-mean-square (rms) spectral widths were calculated. Figure 4 shows the blueshifts of PL peak positions after thermal annealing. The usually observed S-shape variation of PL peak position is unclear in Fig. 4.

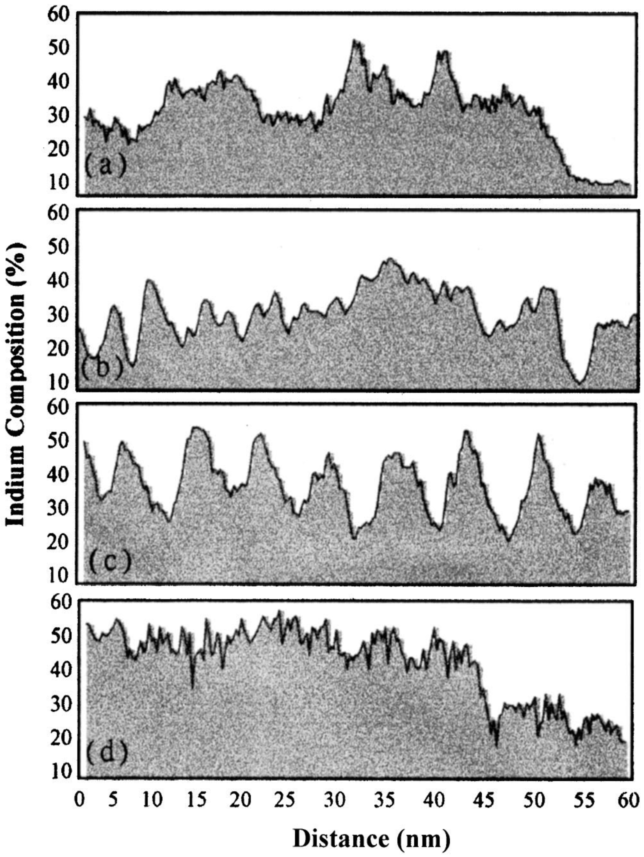

FIG. 3. EFTEM line-scan results of indium composition (%) along a QW in the as-grown sample (a), and samples annealed at 850 °C (b), 900 °C (c), and 950 °C (d).

Typically, annealing at higher temperature results in a larger blueshift. However, the result of 950 °C annealing seems to have different behaviors between the low- and high-temperature ranges. Figure 5 shows the PL rms width variations of various samples. The as-grown sample shows an irregular variation in the high-temperature range. Typically, thermal annealing leads to more regular indium cluster structures. Therefore, the PL spectral width is expected to be

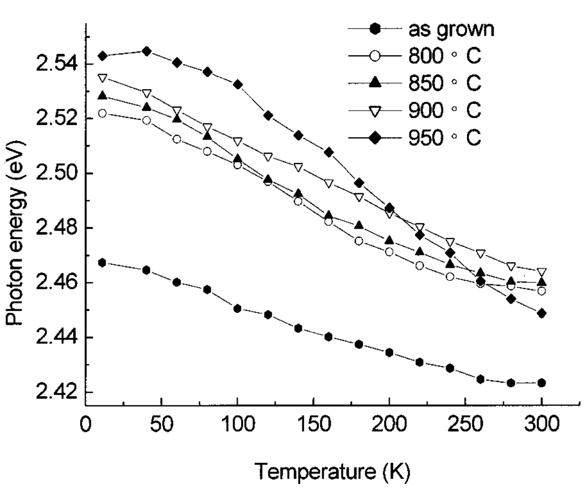

FIG. 4. Fitted PL peak positions as functions of temperature of various samples.

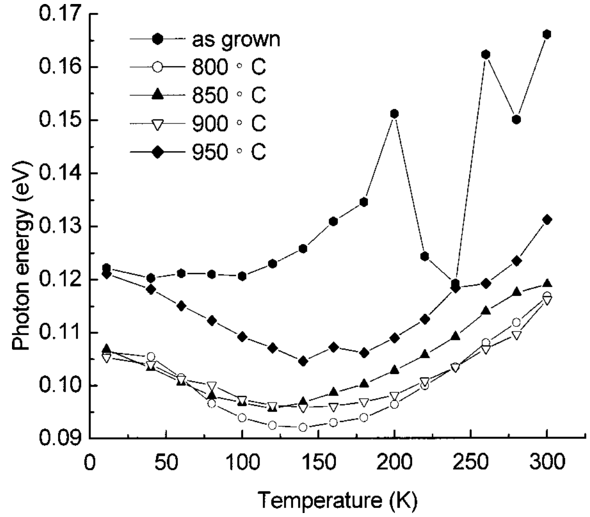

FIG. 5. PL root-mean-square spectral widths as functions of temperature of various samples.

smaller, as confirmed with the behaviors of the annealed samples in Fig. 5. The larger rms spectral width of the 950 °C annealing sample, compared with the other three annealed samples, is consistent with the irregular structure shown in Figs. 1(d), 2(c), and 3(d).

In summary, we have conducted postgrowth thermal annealing of an InGaN/GaN QW sample with a medium level of nominal indium content. From material analyses, we found that thermal annealing at 900 °C led to quasiregular quantum-dot-like structure. However, such a structure was destroyed when the annealing temperature was further raised to 950 °C.

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