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Inhomogeneous incorporation of In and Al in molecular beam epitaxial AlInGaN films

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Plasma-induced molecular beam epitaxial AlInGaN heterostructures have been characterized by spatial resolved cathodoluminescence and x-ray energy dispersive microanalysis. Competitive incorporation of Al and In has been observed, with the formation of In-rich regions, showing enhanced luminescence around surface pinholes. These island-like In-rich regions are favored by growth at lower temperature due to the higher incorporation of indium into the alloy. The elastic strain relaxation associated to pinhole formation induces preferential local indium incorporation. The diffusion of carriers to these areas with reduced band gap enhances the luminescence emission of the quaternary film. The width and intensity of the luminescence appear to be sensitive to the mismatch between the quaternary film and the GaN layer below. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1407849]

Different GaN-based devices have been developed in the last several years. InGaN/GaN quantum well structures operate as efficient blue lasers[1] while transistors have been demonstrated on AlGaN/GaN heterostructures.[2] The lattice mismatch in the heterostructures introduces serious limitations for device design and/or operation. The emission of blue and green lasers with highly strained InGaN quantum wells as active layers shifts toward higher energies and also the indium content in the alloy is limited to a few percent.[3] On the other hand, the large lattice mismatch in Al-rich AlGaN/GaN heterostructures decreases the critical thickness of fully strained AlGaN barriers in GaN-based transistors. This effect introduces relaxation via misfit dislocations and cracks and degrades the electronic transport properties.[4]

In order to solve these problems, several authors have recently grown chemical vapor deposition (CVD)[3–7] and molecular beam epitaxy (MBE)[8] quaternary AlInGaN films. AlInGaN/InGaN or AlInGaN/AlGaN lattice-matched structures can be obtained by independently adjusting the band gap and the lattice constant through changes in the quaternary composition. The experimental conditions to obtain high quality films simultaneously satisfy the need of the high temperature required to grow Al-containing layers and the lower temperature required for indium incorporation.[8]

The AlInGaN samples described in previous works have been characterized by nonspatial resolved techniques like photoluminescence (PL), secondary ion mass spectroscopy (SIMS), or high resolved x-ray diffraction (HRXRD) that provide averaged values over large areas. In the work of Ref. 3, the authors found variations of the Al and In contents along the wafer indicated by their PL and x-ray energy dispersive spectroscopy (EDS) results. They suggest that these variations may be due to nonuniform fluxes of the reactant gases along the wafer. Transmission electron microscope images in Ref. 4 refer mainly to analysis of the atomically flat AlInGaN/GaN interface and no additional information on spatial distribution of defects is given.

Characterization with spatial resolution by means of cathodoluminescence (CL) and x-ray-microanalysis in the scanning electron microscope could provide information on the film structure and defect distribution related to the incorporation of Al and In in the alloy. In this work, a series of AlInGaN samples grown by MBE[8] was investigated by CL and x-ray microanalysis in order to study the spatial distribution of Al and In in the alloy as a function of the growth temperature.

The samples used in this work were grown by MBE, keeping the flux of Ga, In, and Al as well as the flux of nitrogen radicals constant, and varying the growth temperature in order to obtain different alloy compositions. Previous Rutherford backscattering spectroscopy (RBS) results[8] of the samples are summarized in Table I. The heterostructure consists of a thin AlN nucleation layer, about 5 nm, grown on a c-Al_2O_3 substrate, followed by 500 nm GaN and 130–230 nm of $Al_xIn_yGa_{1-x-y}N$. Further growth details are given in Ref. 8.

The CL measurements were carried out using a Hitachi S-2500 scanning electron microscope (SEM) at 77 K and at an accelerating voltage of 5 kV. In order to study the composition of the features observed in the SEM micrographs, mapping of Al, In, and Ga was carried out by EDS in a JEOL JXA-8900 M superprobe.

Secondary electron images show the existence of disk-shaped regions with typical size in the range of 10–50 μm on the film surface. These island-like structures appear to overlap or be dispersed as can be observed in Fig. 1, depend-

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TABLE I. Growth temperature, experimental compositions of the alloys obtained by RBS, and description of surface island-like features.

T (°C)	Al (%)	In (%)	Ga (%)	Island growth
650	12.0	14.5	73.5	Overlapped
715	17.0	6.0	77.0	Less overlapped
745	13.5	4.2	82.3	Dispersed, 10–50 μm in size
775	17.0	0.4	82.6	More dispersed ~10 μm in size

ing on the indium content of the sample (see Table I), and often a pinhole is associated to the bigger structures. CL images reveal that the island structures present more intense luminescence than the background. In Fig. 2 a CL image of a sample grown at 745 °C is shown for illustration. The dark spots on the bigger islands, marked by an arrow, correspond to pinholes. The CL spectra were recorded at a beam accelerating voltage of 5 kV in order to obtain luminescence only from the quaternary film (estimated penetration range of ~160 nm). All the samples show emission of a broad band peaked around 400 nm. No band gap emission edge is observed in the emission range estimated by Vegard's law even by including the bowing parameter (b) corrections from Ref. 7 (b_{AlGaN} equal to -1 eV, b_{InGaN} ranging from -1 to -4.8 eV, and $b_{AlInN}=-5$ eV). Therefore we attribute the detected emission to transitions between localized states. Deep level emission at around 400 nm was previously observed for similar AlInGaN films grown by CVD,[7] and was related to nonoptimum growth conditions for aluminum containing alloys. Figure 3 shows spectra of the sample grown at 745 °C. This sample shows higher luminescence intensity and a reduced emission band width compared to the rest of the samples. This observation correlates with the fact that this sample is lattice matched to the GaN film below, as the XRD reciprocal space maps of Ref. 8 show. There is no significant shift between the spectra recorded on the islands and on the background but the emission intensity of the 400 nm band increases when the spectra are recorded on the former. This

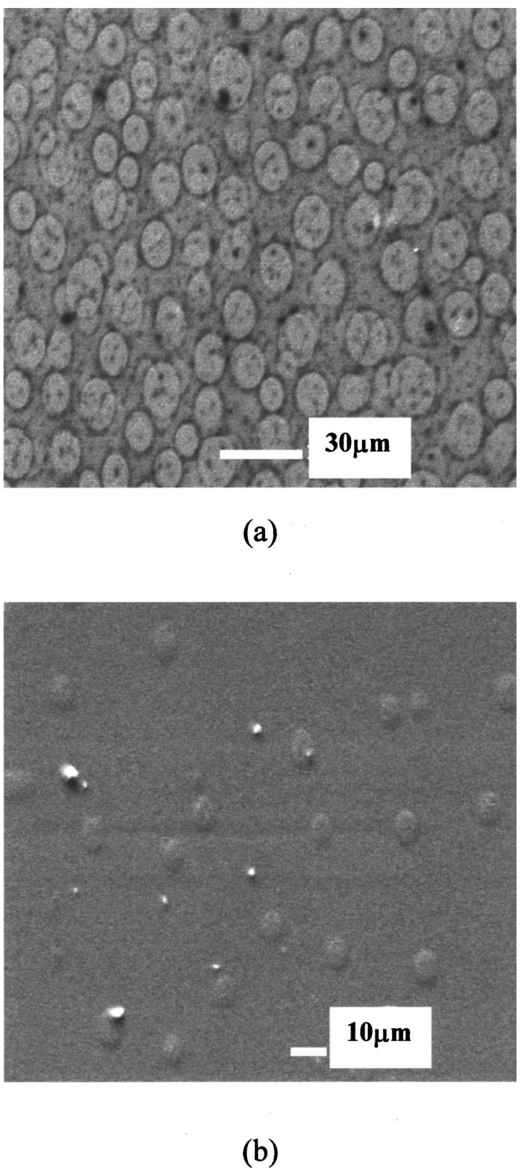

FIG. 1. Secondary electron images showing (a) overlapped disk-shaped regions of a sample grown at 650 °C (14.5% In) and (b) dispersed disk-shaped regions on the film surface of a sample grown at 775 °C (0.4% In). The samples grown at 715 and 745 °C show an intermediate topography between the ones shown here.

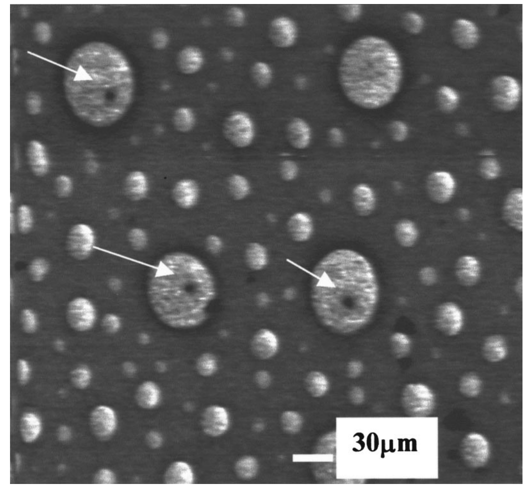

FIG. 2. Cathodoluminescence panchromatic image of a sample grown at 745 °C. Enhanced luminescence emission arising from the island-like regions is observed. Often pinholes, marked by arrows, are associated to the bigger islands.

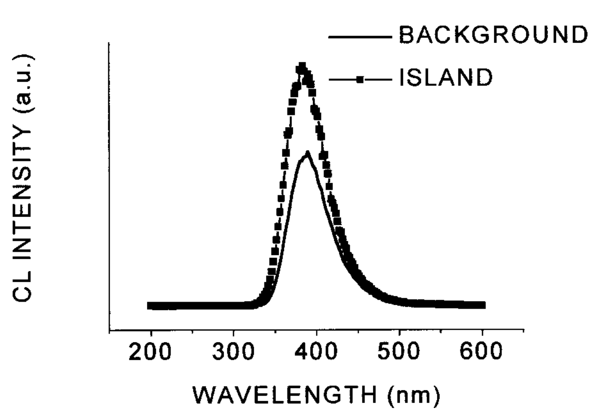

FIG. 3. CL spectrum of the lattice matched sample grown at 745 °C. The spectrum was recorded at 80 K with 5 kV beam accelerating voltage.

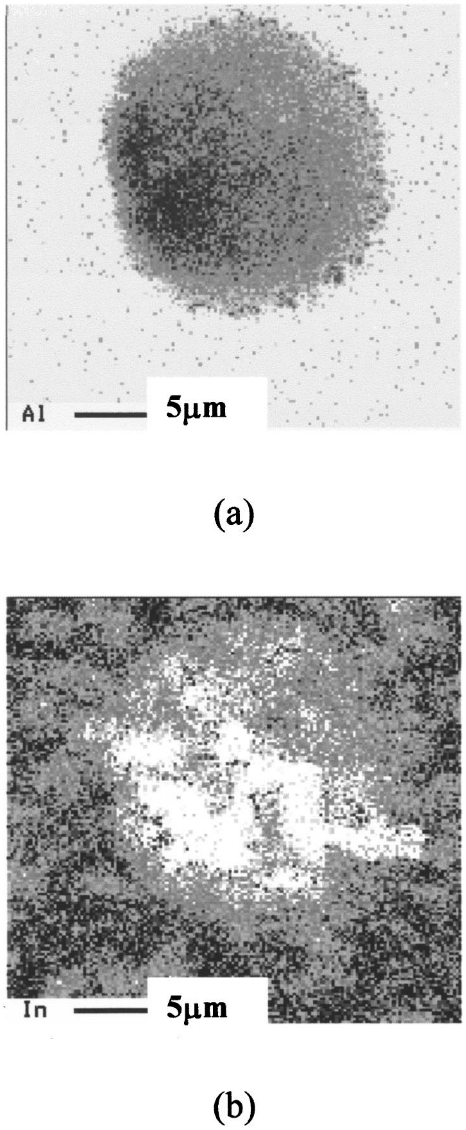

FIG. 4. EDS elemental mappings of (a) aluminum and (b) indium in a region showing an In-rich island of a sample grown at 745 °C. Complementary contrast is observed in both images, indicating competitive incorporation of indium and aluminum into the alloy. Dark regions in the image indicate a lower concentration whereas bright regions indicate a higher concentration of the mapped elements.

would be an indication of diffusion of part of the carriers generated to the islands prior to recombination.

X-ray mapping of Al, In, and Ga shows that competitive incorporation of aluminum and indium takes place during alloy growth. The observed islands have an enhanced In content while the background shows a higher concentration of aluminum. The Ga content remains almost constant along the wafer. In Fig. 4 mappings of Al and In in an island of the sample grown at 745 °C are shown. The x-ray data of the background composition of the samples are comparable to those obtained by RBS shown in Table I.

The SEM observations reveal that the islands are often associated with a hole in the center. This is particularly apparent in the bigger islands, probably due to the larger diameter of the pinhole. The density of pinholes, and hence the density of the pinhole related islands, has been found to increase with the indium content according to the results shown in Table I. This is due to the increase of misfit strain with In incorporation and elastic strain relaxation by the formation of pinholes. A similar observation was reported in previous work[9,10] on InGaN films and it was demonstrated that part of the strain in the layers is elastically reduced by the formation of pinholes. Strain can influence the luminescence by inducing preferential In incorporation at relaxed areas around pinholes. This incorporation will cause the diffusion of carriers to areas near the surface with reduced band gap due to a higher local In content in the alloy. The same mechanism appears to be responsible for the formation of In-rich islands around pinholes in the above mentioned InGaN films in previous work and in the quaternary compounds studied here. The CL and microanalysis results of this work support this idea.

In conclusion, the competitive incorporation of Al and In in AlInGaN alloys grown by MBE and its effect on the spatial distribution of the CL emission was investigated. Our data show that built-in strain in the layer determines the preferential incorporation of In atoms to the alloy in regions where the strain is relaxed via the formation of pinholes. The In-rich islands were observed to have more efficient luminescence compared to the background. This behavior is favored by growth at lower temperatures which causes a higher incorporation of indium into the alloy. Both the strain in the films and the temperature requirement are important factors in achieving high quality quaternary films.

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[1] S. Nakamura, Mater. Sci. Eng., B **50**, 277 (1997).
[2] L. Eastman, K. Chu, W. Schaff, M. Murphy, N. G. Weimann, and T. Eustis, MRS Internet J. Nitride Semicond. Res. **2**, 17 (1997).
[3] F. G. McIntosh, K. S. Boutros, J. C. Roberts, S. M. Bedair, E. L. Piner, and N. A. El-Masry, Appl. Phys. Lett. **68**, 40 (1996).
[4] M. A. Khan *et al.*, Appl. Phys. Lett. **76**, 1161 (2000).
[5] J. Zhang, J. Yang, G. Simin, M. Shatalov, M. A. Khan, M. S. Shur, and R. Gaska, Appl. Phys. Lett. **77**, 2668 (2000).
[6] G. Tamulaitis *et al.*, Appl. Phys. Lett. **77**, 2136 (2000).
[7] M. E. Aumer, S. F. LeBoeuf, F. G. McIntosh, and S. M. Bedair, Appl. Phys. Lett. **75**, 3315 (1999).
[8] A. P. Lima, C. R. Miskys, U. Karrer, O. Ambacher, A. Wenzel, B. Rauschenbach, and M. Stutzmann, J. Cryst. Growth **220**, 341 (2000).
[9] A. Cremades, M. Albrecht, J. M. Ulloa, J. Piqueras, H. P. Strunk, D. Hanser, and R. F. Davis, Mater. Res. Soc. Symp. Proc. **588**, 81 (2000).
[10] B. Jahnen, M. Albrecht, W. Dorsch, S. Christiansen, H. P. Strunk, D. Hanser, and R. F. Davis, MRS Internet J. Nitride Semicond. Res. **3**, 39 (1998).