Lateral phase separation in AlGaN grown on GaN with a high-temperature AlN interlayer

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The influences of a high-temperature (HT) AlN interlayer (IL) on the phase separation in crack-free AlGaN grown on GaN have been studied. The depth-dependent cathodoluminescence (CL) spectra indicate a relatively uniform Al distribution in the growth direction, but the monochromatic CL images and the CL spectra obtained by line scan measurements reveal a lateral phase separation in AlGaN grown on relatively thick HT-AlN ILs. Moreover, when increasing the thickness of HT-AlN IL, the domain-like distribution of the AlN mole fraction in AlGaN layers is significantly enhanced through a great reduction of the domain size. The morphology of mesa-like small islands separated by V trenches in the HT-AlN IL, and the grain template formed by the coalescence of these islands during the subsequent AlGaN lateral overgrowth, are attributed to be responsible for the formation of domain-like structures in the AlGaN layer. © *2005 American Institute of Physics*.
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AlGaN wide band gap semiconductors have tremendous potential applications for electronic and optoelectronic devices spanning the visible to the ultraviolet spectral range.[1] The coherent growth of AlGaN on GaN results in tensile stress in the epilayer due to the large lattice mismatch, which is up to −2.4% for AlN/GaN at room temperature. This stress may lead to crack formation during the deposition process,[2] and block the growth of crack-free thick AlGaN layer with a high mole fraction of AlN. Considerable efforts have been made to overcome the crack problem by inserting a low-temperature (LT) thin AlN interlayer (IL)[3,4] between GaN and AlGaN layers, and to study the influence of LT-AlN IL thickness on the twist and tilt mosaic in AlGaN,[5] as well as the origin of stress reduction by LT-AlN ILs.[6] Meanwhile, a high-temperature (HT) AlN IL is also able to suppress crack formation in AlGaN,[7,8] and multiple HT-AlN ILs can even sustain the crack-free growth of 60 pair $Al_{0.2}Ga_{0.8}N/GaN$ distributed Bragg reflectors.[9] However, very few efforts have addressed the effects of HT-AlN ILs on the subsequent growth of AlGaN layers. In this letter, we present a detailed study on the influences of the HT-AlN IL thickness on the phase separation in the AlGaN layer.

Six samples were prepared using low pressure metalorganic chemical vapor deposition in a showerhead reactor. Trimethylgallium, trimethylaluminum and ammonia were used as precursors for Ga, Al and N, respectively. H_2 served as the carrier gas. First, a 2.5 μm GaN layer was grown on (0001) sapphire substrate via a LT GaN nucleation layer. Then a HT-AlN IL was deposited on GaN at 1040 °C for samples A, B, C, and D, with a nominal thickness of 9, 18, 26, and 40 nm, respectively. Finally, a 1.1-μm-thick $Al_{0.25}Ga_{0.75}N$ layer was deposited on AlN ILs with nominally identical growth parameters for all the samples. For comparison, samples E and F were prepared using a LT-AlN IL with a nominal thickness of 8 and 40 nm, respectively.

A triple-axis x-ray diffractometer (JPN Rigaku SLX-1A) was used to measure the diffraction patterns of these samples. Low-temperature cathodoluminescence (CL) investigations were performed in a scanning electron microscope equipped with an Oxford mono-CL2 and He-cooling stage operating at 9 K. A grating monochromator and a cooled charge-coupled device array were used to disperse and detect the CL signal, respectively. High-resolution transmission electron microscopy (TEM) observations were carried out using a Philips CM200 field emission gun microscope operating at 200 kV.

Figure 1 shows the (0004) triple-axis $\omega/2\theta$ x-ray diffraction (XRD) patterns of samples A, B, C, and D. For sample A with the thinnest HT-AlN IL, only one sharp XRD peak was observed for the AlGaN film. When increasing the HT-AlN IL thickness, the AlGaN XRD peak shifts to a lower angle due to the enhanced relaxation of the AlGaN layers. More interesting is that for sample B, a shoulder appears on the left side of the AlGaN XRD peak, and it turns into a separate peak for samples C and D. These XRD results indicate that there are two distinct AlN mole fractions in the AlGaN layer grown on a relatively thick HT-AlN IL, and the proportion of AlGaN alloys with the relatively low AlN mole fraction increases with increasing HT-AlN IL thickness. In contrast, it is found that only a single XRD peak appears in the (0004) $\omega/2\theta$ scan of the AlGaN layer (not shown here) for samples E and F grown with a LT-AlN IL.

The double XRD peaks might come from a lateral phase separation in the AlGaN layer or two AlGaN layers with

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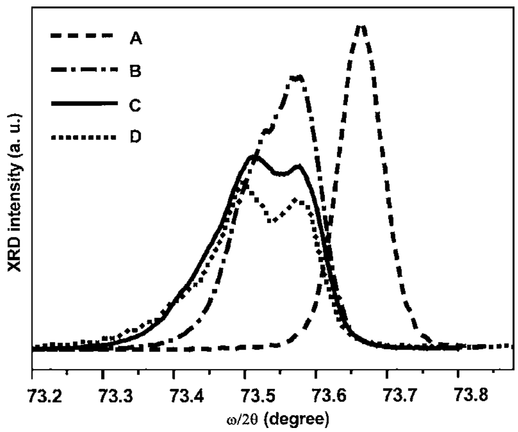

FIG. 1. The (0004) triple-axis $\omega/2\theta$ x-ray diffraction patterns of samples A, B, C, and D.

different AlN mole fractions in the growth direction. In order to distinguish these two possibilities, we performed detailed CL investigations on these samples. It is found that there is no remarkable shift of the AlGaN CL spectra from varying excitation depth of the AlGaN layer, when the accelerating voltage of the incident electron beam increases from 5 to 30 kV (not shown), indicating a negligible phase separation in the growth direction. However, the monochromatic CL images taken at fixed wavelengths with a 10 keV electron beam reveal a lateral inhomogeneous distribution of the Al composition. The CL images [Figs. 2(a)–2(d)] from samples A, B, C, and D show clearly domain-like structures with a strong contrast, while those from samples E and F

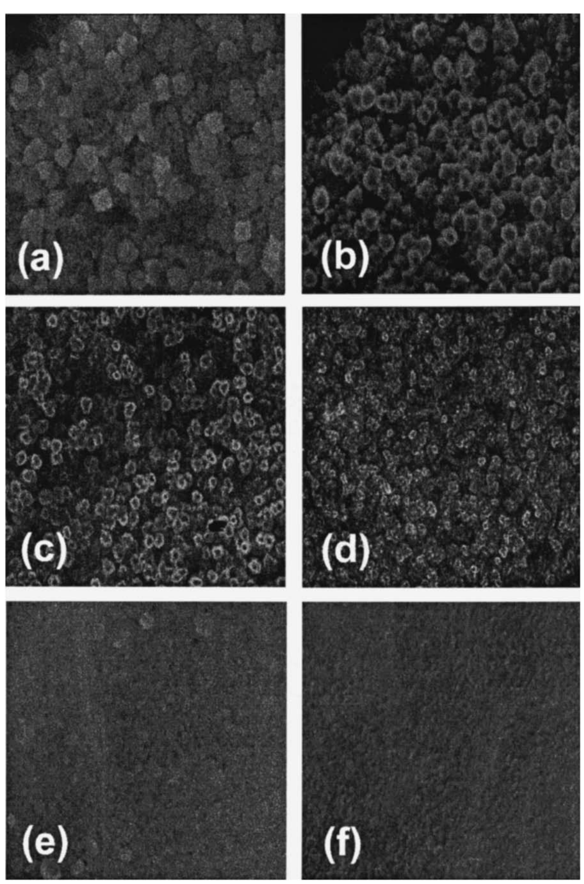

FIG. 2. The 233 μm × 173 μm monochromatic CL images of samples A (a), B (b), C (c), D (d), E (e), and F(f) taken at 4.008, 4.008, 3.995, 3.981, 4.016, and 4.016 eV, respectively.

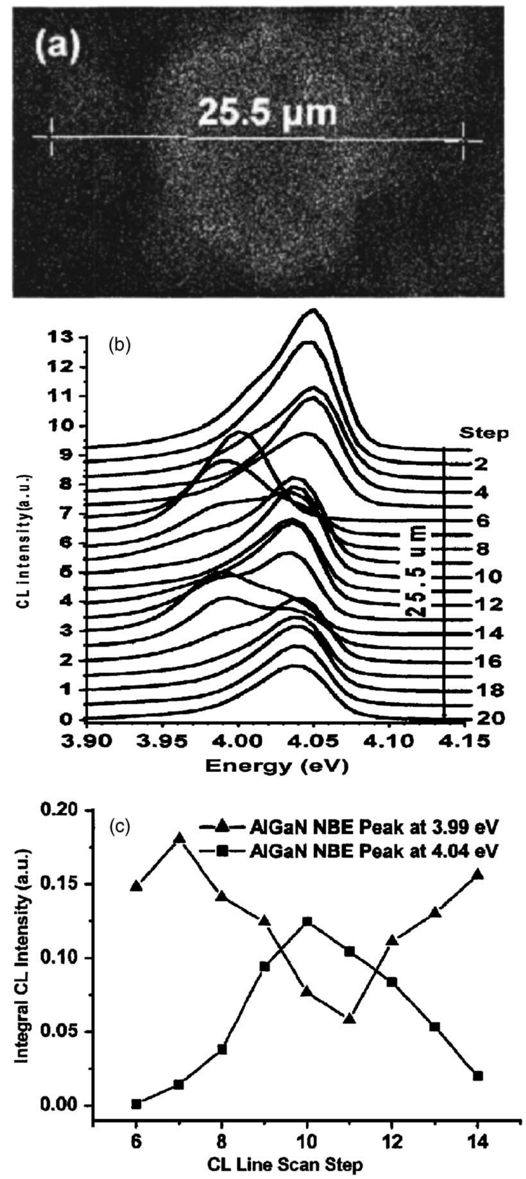

FIG. 3. Monochromatic CL image of a domain in sample B taken at 4.008 eV (a). The lateral line scan CL spectra across the domain (b). The CL spectra have been vertically shifted for clarity. The spatial distribution of the integral CL intensity across the domain (steps 6–14) of the 4.04 eV peak (square) and the 3.99 eV peak (triangle) is shown. Each step of the line scan covers a lateral distance of 1.34 μm.

using a LT-AlN IL [Figs. 2(e) and 2(f)] are much more homogeneous. It is noted that the domain-like contrast is weakest in the CL image [Fig. 2(a)] of sample A with the thinnest HT-AlN IL, which is consistent with the single XRD peak in the $\omega/2\theta$ scan of the AlGaN (0004) reflection (Fig. 1) and indicates a relatively uniform Al distribution in sample A. However, for the AlGaN layers with a thicker HT-AlN IL, the contrast of domain-like structures becomes much clearer and the domain size decreases significantly, which demonstrates a severer lateral phase separation in accord with the XRD results (Fig. 1).

Furthermore, a series of CL spectra were measured by a lateral line scan of the electron beam across a domain to obtain more detailed information about the domain-like distribution of the AlN mole fraction. A set of AlGaN CL spectra [Fig. 3(b)] was acquired by the line scan crossing the domain in sample B, as shown in Fig. 3(a). The line scan measurements were performed from the left side towards the

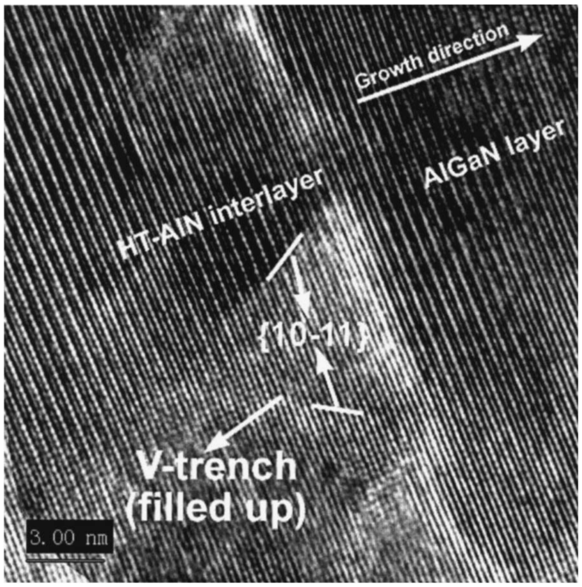

FIG. 4. Cross-section high-resolution TEM image showing V trenches formed in the HT-AlN IL of sample B.

right side along the line shown in Fig. 3(a), which corresponds to the steps 1–20 marked in Fig. 3(b). Each step of the line scan covers a lateral distance of 1.34 μm. It is clearly shown that the two distinct CL peaks at 4.04 and 3.99 eV mainly come from the central region (e.g., steps 10–12) and periphery (e.g., steps 6 and 7) of the domain, respectively. It is noted that they are the near-band-edge emissions of $Al_{0.25}Ga_{0.75}N$ alloy film. The integral CL intensity of the AlGaN peak at 3.99 eV decreases gradually from the periphery towards the center of the domain, while that of the AlGaN peak at 4.04 eV alters in the opposite phase, as shown in Fig. 3(c). It means that the AlN mole fraction is a bit lower in the periphery than in the central region of the domains. The CL line scan measurements of samples C and D present a similar result.

To shed light on the nature of the influence of HT-AlN ILs on the subsequent AlGaN growth, we performed detailed x-ray (10.5) asymmetric reciprocal space mapping (RSM) measurements and cross-section TEM analyses for the samples. Although there is only one weak diffraction spot from the LT-AlN IL in the RSMs (not shown) of samples E and F, two dispersed and separate reciprocal spots from the HT-AlN IL were simultaneously observed, indicating two different Al contents in the HT IL. It implies that the nominal HT-AlN IL has turned into an Al-rich AlGaN IL consisting of two parts with different Al compositions, due to the intermixing of the high-temperature grown AlN IL with the GaN underlayer and/or the AlGaN overlayer.[4] Meanwhile, the cross-section high-resolution TEM image of sample B (Fig. 4) reveals the existence of V trenches in the HT IL, which were filled up during the subsequent AlGaN layer growth. This feature of mesa-like islands separated by V trenches in the HT-AlN layer grown on GaN[10,11] may lead to an inhomogeneous distribution of strain and Al composition in the HT IL. The Al content is a little smaller in the filled V trench regions than in the mesa-like islands. During the initial growth stage of the AlGaN layer, adjacent small islands are coalesced into large grains due to the lateral overgrowth, and the periphery of these grains mainly comes from the regions over V trenches. These grains serve as a template for the further AlGaN growth.[12,13] The AlGaN growth on the central area of these grains is under a higher compressive strain than that on the outer region of the grains, leading to a higher Al incorporation in the former. The inhomogeneous distribution of AlN mole fraction seeded during the initial growth can extend through the epitaxial film.[14] Thus, the domain-like structures are formed in the AlGaN film due to the lateral phase separation. When a thicker HT-AlN IL is grown on GaN, the relaxation between them is enhanced.[10] As mentioned before, our x-ray RSM results have revealed that the original HT-AlN IL turns into an Al-rich AlGaN IL after the subsequent growth of the $Al_{0.25}Ga_{0.75}N$ layer, and the Al contents in the HT IL increase with increasing its thickness. Due to the larger lattice mismatch between $Al_{0.25}Ga_{0.75}N$ and the HT IL, the subsequent AlGaN growth on the thicker HT IL is under a higher compressive strain, which may diminish the size of the template grains grown on the thicker HT IL. These smaller template grains will cause an enhanced phase separation in the AlGaN layer, accompanied by a reduction of the domain size.

In conclusion, a careful study of lateral phase separation in crack-free AlGaN grown on GaN using a HT-AlN IL is reported. It is found that the phase separation occurs mainly in the lateral direction, but not significantly in the growth direction. In the central region of the domain-like structures, the AlN mole fraction of AlGaN is a bit higher than in the periphery of the domains. In order to suppress phase separation during the AlGaN growth, the HT-AlN IL must be as thin as possible, because the phase separation may be greatly enhanced when increasing the thickness of the HT-AlN IL. Based on the cross-section TEM image and the x-ray RSM results, a simple model is proposed to illuminate the origin of the lateral phase separation in the AlGaN layer grown on HT-AlN ILs.

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