doi:10.1017/S1431927613003711
Microsc. Microanal. 19 (Suppl 2), 2013
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Microscopic Characterization of MBE-grown Dilute-nitride Alloys of $GaAsN_x$ (0.01<x<0.04) on GaAs (001) for Photovoltaic Solar Cells

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Nitrogen is insoluble in many III-V semiconductors such as GaAs [1]. However, metastable nitrogen can still be incorporated by epitaxial growth at low temperatures. A small amount of nitrogen could significantly decrease the band gap of $GaAsN_x$ by increasing the N content of the dilute-nitride alloy [2]. This change would further extend the $GaAsN_x$ alloys into longer wavelengths of the solar spectrum, making them very attractive for a wide range of optoelectronic applications, such as infrared detectors and absorbers for high-efficiency tandem solar cells [2]. The dilute-nitride alloy films studied here were grown using an RF plasma-assisted molecular/chemical beam exitaxy system (M/CBE) in a Riber 32^{TM} reactor. All samples were fabricated on (001) nominal orientations of GaAs substrates. A 0.5μm undoped GaAs buffer layer was processed at 520 ˚C before growth of the dilute-nitride alloy films. The 100nm $GaAsN_x$ epilayers were grown on the buffer layer at 450˚C at a rate of 1μm/h [3]. Cross-sectional TEM samples of the dilute-nitride films were prepared using mechanical polishing and dimpling followed by argon ion milling. Most images were recorded using high-angle annular-dark-field (HAADF) imaging in a JEOL-2010F field-emission scanning transmission electron microscope (STEM) operated at 200 keV, which provides a nominal probe size of ~2 Å. High-resolution TEM images were taken using a JEM-4000EX operated at 400 keV, with a structural resolution of 1.7 Å.

Four dilute-nitride alloy samples were investigated in this work, with increasing N concentrations of 1.44%, 1.82%, 3.05% and 3.67%, respectively, as determined using X-ray diffraction (XRD). The band gaps of the four samples were determined by photoluminescence (PL) analysis to be 1.21 eV, 1.15eV, 1.02 eV and 0.92 eV, respectively. All samples were firstly characterized using HAADF-STEM (Z-contrast) imaging. All of the $GaAsN_x$ epilayers had higher contrast than the GaAs buffer layers, as shown in Fig. 1, which is a HAADF image of the $GaAsN_{0.0144}$ sample. This contrast implies that the alloy films contain relatively higher atomic weight after growth, according to the Z sensitivity of HAADF imaging. It further implies that the N atoms incorporated into the $GaAsN_x$ films must occupy interstitial sites of the GaAs lattice, which would cause the increased atomic weight. Additionally, it was interesting to notice that internal stress due to the nitrogen content increased as more interstitial nitrogen was incorporated into the sample. It appeared that stress relief in the dilute-nitride alloy films occurred frequently by cracking, as shown for the $GaAsN_{0.0367}$ sample in Fig. 2(a). Figure 2(b) is a HAADF lattice image showing the interface region between the $GaAsN_{0.0367}$ film and the GaAs buffer. It appears that the interface is not completely abrupt but no defects are visible. In addition, contrast variations are present in the $GaAsN_{0.0305}$ sample, as shown in TEM image 3(a) and HAADF image 3(b), which suggest that parasitic N-rich layers are present. These N-rich layers could severely affect the carrier extraction and transport properties of the alloys, but changes to growth procedures are in progress to alleviate these problems [3]. Further microanalysis and structure-property correlations as a function of annealing treatment are being investigated and will be described.

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[4] This work was supported by DOE/NSF Cooperative Agreement No. EEC-1041895. The authors acknowledge the use of facilities in the John M. Cowley Center for HREM at ASU.

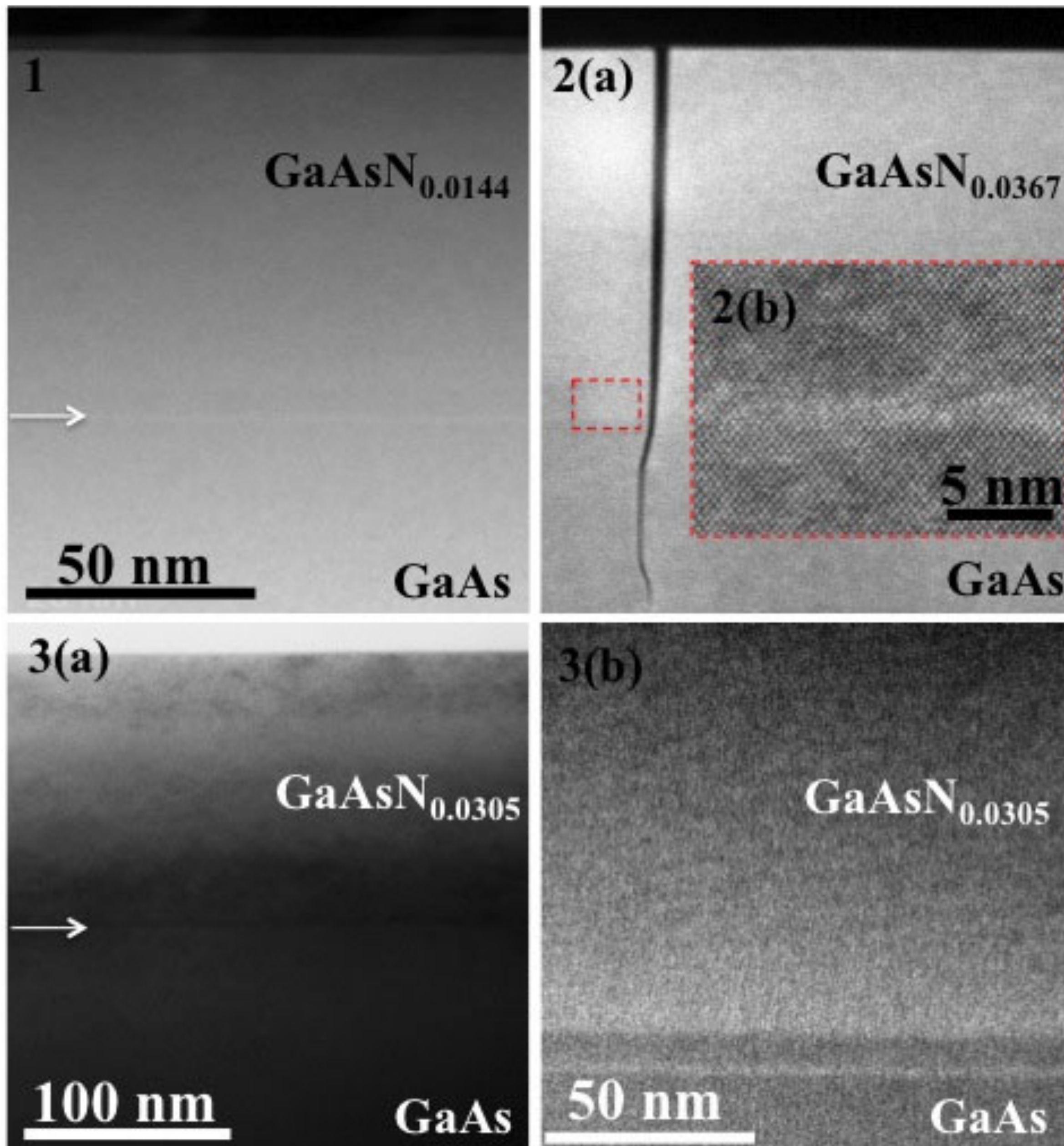

Figure 1. HAADF-STEM image showing $GaAsN_{0.0144}$ alloy grown on GaAs buffer.
Figure 2. (a) HAADF-STEM image showing $GaAsN_{0.0367}$ alloy grown on GaAs buffer; (b) Enlarged HAADF-STEM lattice image showing interface region between $GaAsN_{0.0367}$ alloy and GaAs buffer.
Figure 3. (a) TEM and (b) HAADF-STEM images showing $GaAsN_{0.0305}$ alloy grown on GaAs buffer.

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