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2012 J. Phys. D: Appl. Phys. 45 285103

(http://iopscience.iop.org/0022-3727/45/28/285103)

J. Phys. D: Appl. Phys. **45** (2012) 285103 (4pp) doi:10.1088/0022-3727/45/28/285103

Surfactant effects of gallium on quality of AlN epilayers grown via metal-organic chemical-vapour deposition on SiC substrates

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Received 24 February 2012, in final form 30 May 2012
Published 28 June 2012
Online at stacks.iop.org/JPhysD/45/285103

Abstract
Effects of gallium as a surfactant for the growth of AlN epilayers on SiC substrates by metal-organic chemical-vapour deposition have been studied. It was found that the use of gallium as a surfactant enables the growth of thick, crack-free AlN epilayers on SiC substrates. The photoluminescence and x-ray diffraction (XRD) analysis show that gallium surfactant can reduce some of the tensile strain in AlN epilayers and it improves the surface smoothness. XRD rocking curves yielded decreased full widths at half maximum for the (1 0 5) and (0 0 2) reflections, indicating a reduction in threading dislocation density in the AlN epilayers.

(Some figures may appear in colour only in the online journal)

Aluminium nitride (AlN) has emerged as a very important material due to recent advances in high-quality material growth [1–3]. Light-emitting diodes with emission wavelength of 210 nm have been demonstrated using pure AlN [4]. Furthermore, 200 nm metal–semiconductor–metal (MSM) deep ultraviolet (DUV) photodetectors based on pure AlN [5] were demonstrated. DUV photodetectors have a wide range of applications including secure space communication, missile threat detection, UV radiation monitoring in environment, and biological agent detection. As a consequence of its widest direct band gap (6.1 eV) among III-nitride semiconductors, combined with its outstanding electro-optical and physical properties, AlN also appears to be very promising for the development of vacuum UV and extreme UV detectors where Si-based photodetectors have reached their limits.

The growth of high-quality AlN epilayer directly on highly conductive SiC substrates naturally provides a hybrid Schottky barrier photodetector structure that does not require the growth of n-type ohmic contact layer. AlN layer has outstanding interfacial properties when deposited on SiC due to an excellent lattice match to SiC (1%) [6–10].

Studies of AlN (0 0 1) deposited on SiC (0 0 1) found lattice mismatch strains at the AlN/SiC interface to be compressive. The mismatch is believed to be partially relieved by the formation of misfit dislocations. However, there is tensile strain along the a-axis in the AlN layers grown on SiC, which tends to crack the surface of the nitride layers [11]. Cracking significantly reduces the performance of nitiride-based devices due to their current-scattering centres for light propagation as well as poor crystal quality [12]. Thus crack elimination from nitride layers is a crucial issue in realizing device application.

Different methods have been reported to remove cracks and to improve crystal quality of AlN epilayers. AlN growth is best performed at higher temperatures than GaN growth, because Al atoms are less mobile than Ga atoms. High structural quality crack-free AlN epilayers grown at temperatures as high as 1500 °C were reported [13, 14]. Because high-temperature metal-organic chemical-vapour deposition (MOCVD) systems require specialized heater and growth chamber designs, it is desirable to find methods for growing crack-free high-quality AlN at lower temperatures. High-quality AlN using a multilayer structure that alternates between two-dimensional and three-dimensional growth modes was reported [15, 16].

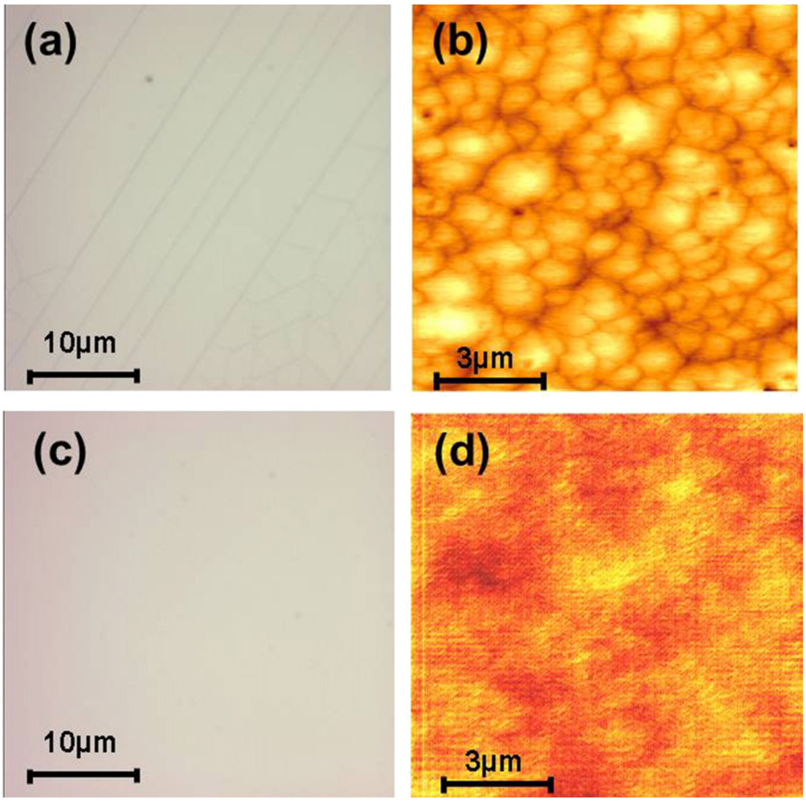

Figure 1. (*a*) A microscope image and (*b*) AFM image of AlN grown without Ga flow and (*c*) a microscope image and (*d*) AFM image of AlN grown with Ga flow.

In this work, we investigate the influence of gallium (Ga) as a surfactant on strain and cracking in AlN epilayers. AlN epilayers of 1.4 μm in thickness were grown on SiC substrate by MOCVD. A 12 nm low-temperature (920 °C) AlN nucleation layer was first grown on the *c*-plane (0 0 1) of SiC substrates followed by the growth of 1.4 μm high-temperature (1300 °C) undoped AlN epilayres. Trimethylaluminum, trimethylgallium (TMGa), and ammonia were used as sources for aluminum, gallium and nitrogen, respectively. During the growth of some AlN epilayers, TMGa was introduced into the gas stream, and various TMGa flow rates were used. The optimum flow rate of TMGa was 0.3 ml min^{-1} (which corresponds to 0.03 μmol min^{-1}).

It was found that the flow of Ga at the rate of 0.3 ml min^{-1} during the growth effectively prevents crack formation as shown by the microscopic images. Atomic force microscopy (AFM) images clearly reveal that Ga plays a role in improving the surface morphology of AlN epilayers. Energy dispersive x-ray (EDX) analysis on the AlN sample grown with Ga flow indicates the presence of a very low concentration of Ga (low enough to avoid alloy formation). X-ray diffraction (XRD) (0 0 2) ω–2θ scans show that Ga surfactant releases some of the tensile strain in the AlN and reduces the threading dislocation density through the reduction of the full-width at half-maximum (FWHM) of the (1 0 5) and (0 0 2) rocking curves. Low-temperature (10 K) photoluminescence (PL) measurements [17] indicate a less tensile strain in AlN epilayers grown using Ga surfactant compared with samples grown without Ga surfactant through a smaller red-shift for the band edge emission peak of AlN with Ga surfactant.

The surface cracking of AlN epilayer is examined using optical microscope and the results are shown in figure 1. The introduction of Ga during the growth of AlN prevents the formation of cracks completely (figure 1(*c*)). In comparison, the sample without Ga flow is observed to have a high density of cracks (figure 1(*a*)). Using Ga flow, the maximum crack-free layer thickness attempted is up to 1.8 μm.

To further identify the effect of Ga flow on growth mode, surface morphologies of the above-mentioned AlN samples were studied by AFM. Figure 1(*b*) shows the AFM image of AlN surface without Ga flow. Typical 3D growth mode results in a surface whose root-mean-square (RMS) roughness is 2.4 nm in a scanned area of $10 \times 10\ \mu m^2$. On the other hand, the surface of the AlN sample with Ga flow is very flat, with an RMS roughness as small as 0.4 nm, as shown in figure 1(*d*). This indicates that the Ga flow enhances surface mobility of Al and N adatoms which leads to improved 2D growth.

The presence of Ga in the AlN samples grown with Ga flow was verified by the EDX analysis. The EDX spectrum, shown in figure 2, indicates the presence of Ga in the AlN sample grown with Ga flow. The Ga concentration in the AlN sample is very low (0.08%), low enough to avoid alloy formation. This indicates that Ga behaves as a surfactant.

Figure 3 shows the XRD (0 0 2) ω–2θ scans of the 1.4 μm AlN films with and without Ga surfactant. The most intense peaks in the figure are the SiC substrate peaks. For the AlN peak of the film without Ga surfactant, it has a larger angular separation from the SiC peak than that of the film with Ga surfactant, which is an indicative of a slightly smaller *c* lattice

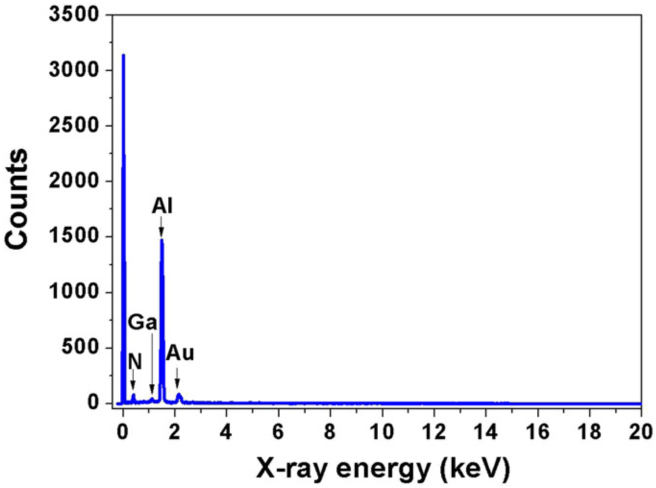

Figure 2. EDX spectrum of the AlN sample grown with Ga flow.

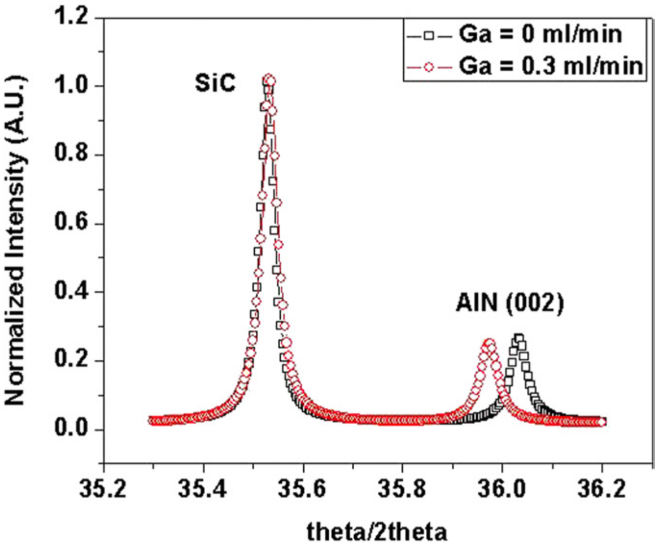

Figure 3. (0 0 2) ω–2θ scans of the two AlN samples grown with and without Ga surfactant.

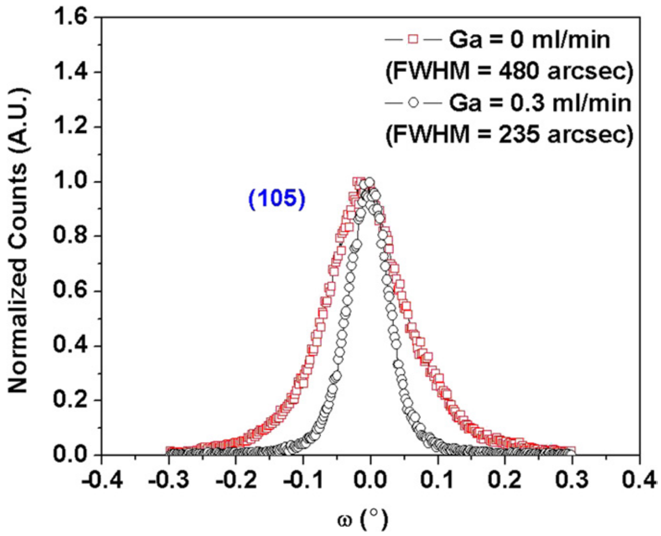

Figure 4. Rocking curves of the asymmetric (1 0 5) reflection peaks in AlN epilayers grown with and without Ga surfactant. FWHM is 235 arcsec for AlN sample with Ga surfactant and 475 arcsec for the AlN sample without Ga surfactant.

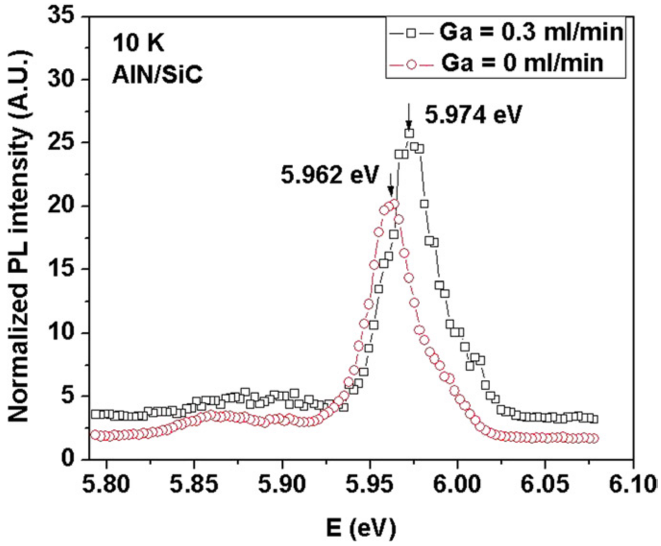

Figure 5. 10 K PL spectra of the two AlN samples grown with and without Ga surfactant, measured in a small spectral range from 5.8 to 6.1 eV.

constant. This shows that the film without Ga surfactant is under greater biaxial tension than the AlN with Ga surfactant, even though the strain in the AlN sample without Ga surfactant was partially released by crack formation. According to the lattice and thermal mismatch, AlN grown on SiC should experience about −1.0% biaxial compressive strain after the structure cooling down [16, 18, 19]. The main tensile strain that causes cracking results from grain coalescence [20]. The AlN sample with Ga surfactant has smaller tensile strain than that of the AlN sample without Ga surfactant, which already partially relaxed by forming cracks. This indicates that strain in the AlN sample with Ga surfactant is released through a relaxation process other than crack formation.

XRD rocking curves of the asymmetric (1 0 5) reflection peak of both AlN epilayers are shown in figure 4. A FWHM as low as 235 arcsec was obtained for the sample grown with Ga surfactant in comparison with 480 arcsec for the sample grown without Ga surfactant. Furthermore, the FWHM of the symmetric (0 0 2) XRD peak (not shown) of both samples with and without Ga surfactant was 118 arcsec and 180 arcsec, respectively. Obviously the threading dislocation propagation can be intercepted by the Ga surfactant especially for edge type dislocations. This leads us to attribute the reduced strain in the AlN sample with Ga surfactant compared with that without Ga surfactant to the reduction of the threading dislocation density [21].

To further study the effect of Ga surfactant during the growth on the residual stress in AlN films, PL measurements were conducted. Figure 5 shows the low-temperature (10 K) PL spectra of AlN epilayers with and without Ga surfactant measured in the energy range from 5.8 to 6.1 eV. Each PL spectrum exhibits a strong band edge emission line. The spectral peak position of the band edge emission line for the AlN sample grown using Ga surfactant is 5.974 eV and for the sample without Ga surfactant is 5.962 eV. Compared with the spectral peak position of the band edge emission line in the AlN grown on AlN bulk crystal (6.029 eV) [22], the band edge emission peak of both AlN epilayers grown with and without Ga surfactant is red-shifted due to tensile strain, where the AlN epilayer without Ga surfactant is under larger tensile strain which agrees with the XRD results and confirms that strain in the AlN sample with Ga surfactant is released through a relaxation process other than crack formation.

In summary, effects of using gallium as a surfactant during the MOCVD growth of AlN epilayers were investigated. It was found that gallium surfactant is an effective method to release tensile strain and to prevent crack formation in thick AlN epilayers; also, it improves the surface morphology of AlN epilayers. Moreover, gallium as a surfactant was also shown to narrow the XRD FWHM of (1 0 5) and (0 0 2) peaks, indicating a reduction in threading dislocation density.

Acknowledgment

TTU's work is partly supported by DOE (DE-FG02-09ER46552)

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