Probing the relationship between structural and optical properties of Si-doped AlN

B. N. Pantha, A. Sedhain, J. Li, J. Y. Lin, and H. X. Jiang[a)]
Department of Electrical and Computer Engineering, Texas Tech University, Lubbock, Texas 79409, USA

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Much efforts have been devoted to achieve conductivity control in the ultrahigh band gap ($\sim$6.1 eV) AlN by Si doping. The effects of Si-doping on the structural and optical properties of AlN epilayers have been investigated. X-ray diffraction studies revealed that accumulation of tensile stress in Si-doped AlN is a reason for the formation of additional edge dislocations. Photoluminescence (PL) studies revealed that the linewidths of both band-edge and impurity related transitions are directly correlated with the density of screw dislocations, N_{screw}, which increases with the Si doping concentration (N_{Si}). Furthermore, it was formulated that the band-edge (impurity) PL emission linewidth increases linearly with increasing N_{screw} at a rate of $\sim 3.3 \pm 0.7$ meV/10^8 cm^{-2} (26.5 ± 4 meV/10^8 cm^{-2}), thereby establishing PL measurement as a simple and effective method to estimate screw dislocation density in AlN epilayers. © *2010 American Institute of Physics*. [doi:10.1063/1.3374444]

AlN has the widest direct band gap ($\sim$6.1 eV) among the III-nitride semiconductors and possesses outstanding properties such as high temperature stability, high thermal conductivity, and deep ultraviolet (DUV) transparency. These properties make AlN a good candidate for various device applications, such as templates[1] or active materials for DUV light emitters[2] and photodetectors,[3,4] and quantum cascade lasers.[5,6]

However, the insulating nature and high density of threading dislocations (TDs) hinder the commercial development of devices based upon AlN. The presence of triply/doubly negatively charged aluminum vacancies $V_{\mathrm{Al}}^{3-/2-}$ is believed to compensate the Si donors[7,8] while the lattice mismatch between the substrate and AlN is the main reason for high density of TDs.[9–12] While several groups have demonstrated n-type conductivity in AlN by Si doping,[1,13–15] doping causes crystal imperfections which can affect the structural and optical properties of the film. There are numerous reports on Si-doped AlN,[13–16] but the investigation on the impact of Si incorporation on structural and optical properties is limited. In this letter, we report on the correlation between structural/optical properties and Si doping concentration in AlN epilayers.

Undoped and Si-doped AlN epilayers of about 1 μm thickness were grown on c-plane sapphire by metal organic chemical vapor deposition. The sources of Al, N, and Si were trimethylaluminum, ammonia (NH_3), and silane (SiH_4), respectively. The Si doping concentration (N_{Si}) was varied from 5×10^{17} to 1×10^{19} cm^{-3}. We employed x-ray diffraction and DUV photoluminescence (PL) spectroscopy to investigate the structural and optical properties of these AlN epilayers.

The θ-2θ scans of (002) planes for undoped and Si-doped AlN epilayers are shown in Fig. 1(a). Regardless of N_{Si}, all curves have retained their symmetric shape. This is attributed to the fact that there is no gradient of stress normal to the surface.[17] As N_{Si} increases, the 2θ position shifts to a higher angle corresponding to a smaller c lattice constant. In contrast to this, at higher N_{Si} ($>2\times10^{18}$ cm^{-3}), the 2θ position shift is reversed. The shift in peak position is related to the change in stress in the film. In-plane lattice constant a was determined by the c lattice constant and d value of the (102) plane using the following equation,

$$\frac{1}{d_{hkl}^2} = \frac{4}{3}\left(\frac{h^2+hk+k^2}{a^2}\right) + \frac{l^2}{c^2}, \tag{1}$$

where h, k, l are miller indices, and c and d are lattice constants for the (002) and (hkl) planes, respectively. We have estimated the in-plane stress $\sigma_{\parallel}$ from the following equation:[18,19]

$$\sigma_{\parallel} = \frac{a-a_o}{a_o}\left(C_{11}+C_{12}-2\frac{C_{13}^2}{C_{33}}\right), \tag{2}$$

where C_{ij} are the elastic constants of AlN (C_{11}=410 GPa, C_{12}=140 GPa, C_{13}=100 GPa, and C_{33}=390 GPa) (Ref. 20) and a_o=3.112 Å is the lattice constant for stress-free AlN.[19] The variation in in-plane stress with N_{Si} is shown in Fig. 1(b). The graph shows that incorporation of Si in AlN has caused a relaxation in the compressive stress, which is in the order of 1 GPa in undoped AlN, and induced tensile stress

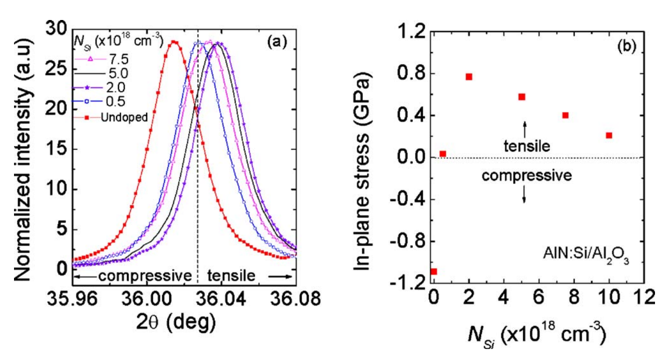

FIG. 1. (Color online) (a) Angular shift (2θ) of (002) reflection in θ-2θ scan mode in undoped and Si-doped AlN epilayers. (b) In-plane stress in Si-doped AlN epilayers as a function of the Si doping concentration (N_{Si}).

[a)]Electronic mail: hx.jiang@ttu.edu.

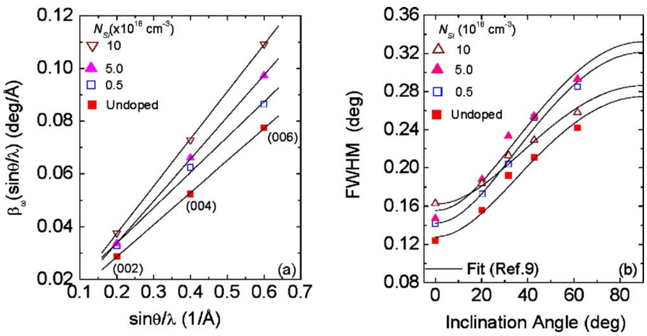

FIG. 2. (Color online) (a) The Williamson–Hall plot (W–H plot) using integral width of the (002), (004), and (006) rocking curves for undoped and Si-doped AlN epilayers, (b) FWHM of the rocking curves of the (*hkl*) planes as a function of lattice plane inclination angle with respect to sample surface for undoped and Si-doped AlN epilayers and fitted with an equation described in Ref. 9.

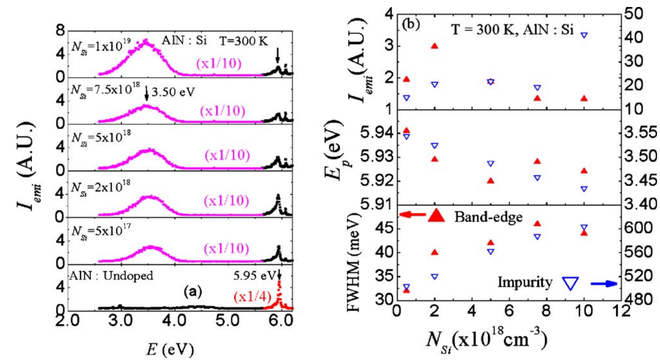

FIG. 4. (Color online) Room temperature (a) PL spectra of Si-doped AlN for different N_{Si}, (b) PL emission intensity (top), PL energy peak position (middle), and PL FWHMs (bottom) of Si-doped AlN as functions of N_{Si}. Closed (open) triangles on the left (right) scales describe the PL properties of band edge (impurity) emission.

when $N_{Si} > 5\times 10^{17}$ cm^{-3}. Tensile stress was a maximum ($\sim$0.8 GPa) at $N_{Si}=2\times 10^{18}$ cm^{-3}. Since the variations in in-plane stress and edge dislocation density N_{edge} (Fig. 3 will be discussed later) with N_{Si} were found to be very similar, we believe that relaxation of compressive stress and accumulation of tensile stress was the reason for the increase in edge dislocations in Si-doped AlN. Normally, dislocations are formed to reduce the stress.

Tilt and twist angles were obtained from the Williamson–Hall plots[21] as described by Lee *et al.*[9] The corresponding plots are shown in Figs. 2(a) and 2(b). Screw and edge TD densities (N_{screw} and N_{edge}) were estimated using the following classical formulae:[22]

$$N_{screw} = \beta_{tilt}^2/4.35 b_c^2, \tag{3}$$

$$N_{edge} = \beta_{twist}^2/4.35 b_a^2, \tag{4}$$

where b_c and b_a are the Burgers vector of c and a-type TDs, respectively, with $|b_c|=0.4982$ nm and $|b_a|=0.3112$ nm.

The variations in N_{screw} and N_{edge} with N_{Si} are shown in Fig. 3. We observed that N_{screw} increased monotonically with N_{Si}. N_{screw} was found to be about two times higher ($N_{screw} > 8\times 10^8$ cm^{-2}) in heavily doped samples ($N_{Si} > 7\times 10^{18}$ cm^{-3}) compared to undoped sample ($N_{screw}=4.2\times 10^8$ cm^{-2}). In contrast, N_{edge} was found to increase only in moderately doped samples ($N_{Si}=0.5$ to 5×10^{18} cm^{-3}) and

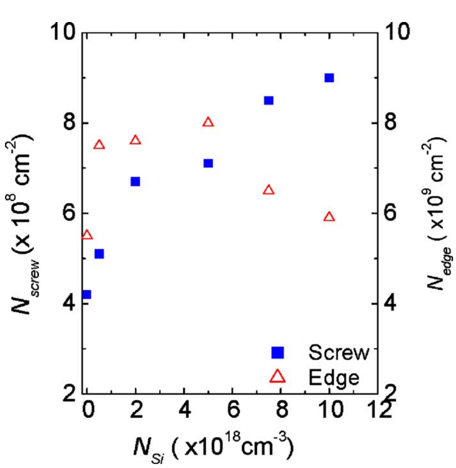

FIG. 3. (Color online) Screw (N_{screw}) and edge (N_{edge}) dislocation densities as functions of N_{Si}.

then decreased thereafter. N_{edge} was found to be about an order of magnitude higher than N_{screw}. Reduction in N_{edge} at $N_{Si} > 5\times 10^{18}$ cm^{-3} could be due to the bending and annihilation of these dislocations[16,23] to accommodate for the change in tensile stress [see Fig. 1(b)]. The results have established the fact that edge dislocations are correlated with the in-plane stress.

Fig. 4(a) shows the PL spectra of undoped and Si-doped AlN epilayers with N_{Si} varying from 5×10^{17} to 1×10^{19} cm^{-3}. All Si-doped samples exhibit weak emission lines due to donor bound exciton transition (I_2) at 5.93 eV. This emission line is about 20 meV below the free excitonic (FX) line in undoped AlN.[24] Spectra from all the Si-doped samples show a strong impurity band around 3.5 eV, about two orders of magnitude stronger than the I_2 emission, which is related to the aluminum vacancies, V_{Al}. This triply/doubly negatively charged aluminum vacancy ($V_{Al}^{3-/2-}$) (Refs. 7 and 8) is responsible for trapping electrons, resulting in poor room temperature electrical conductivity in Si doped AlN. The detailed PL features of both I_2 and impurity transitions as functions of N_{Si} are plotted in Fig. 4(b). Figure 4(b) (top) shows that the I_2 (impurity) emission intensity, I_{emi} decreases (increases) as N_{Si} increases, indicating that the incorporation of Si increases defect density and deteriorates material quality. This is further supported by the systematically increasing in full width at half maxima (FWHM) of both I_2 and impurity related transitions as seen in Fig. 4(b) (bottom).

The direct correlation between N_{screw} and the PL spectra FWHM is illustrated in Fig. 5. Figure 5 shows PL spectra FWHM of both I_2 and impurity transitions as functions of N_{screw} and fitted by the following equations:

$$\mathrm{FWHM}_{\text{band-edge}}(N_{screw}) = \mathrm{FWHM}_{\text{band-edge}}(\text{intrinsic}) + \alpha_{\text{band-edge}} N_{screw}, \tag{5}$$

$$\mathrm{FWHM}_{\text{impurity}}(N_{screw}) = \mathrm{FWHM}_{\text{impurity}}(\text{intrinsic}) + \alpha_{\text{impurity}} N_{screw}. \tag{6}$$

FWHMs were found to increase linearly with increasing N_{screw} at a rate of $\sim 3.3\pm 0.7$ meV/10^8 cm^{-2} for I_2 and $\sim 26.5\pm 4$ meV/10^8 cm^{-2} for impurity transitions. An intrinsic linewidth of $\sim$17 meV for I_2 emission was estimated when $N_{screw}=0$. This value is very close to the linewidth of the I_2 transition ($\sim$13 meV) measured experimentally in

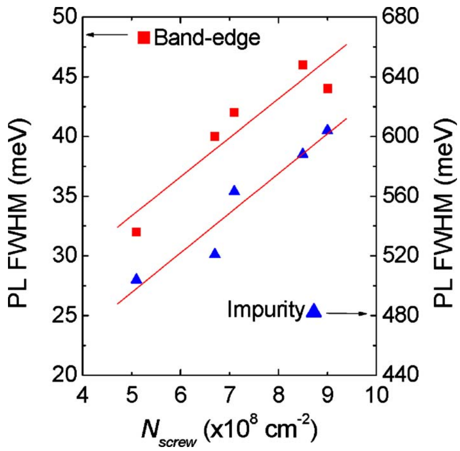

FIG. 5. (Color online) FWHMs of impurity and band edge PL emissions as function of N_{screw}. The solid lines are the linear fit of data using Eqs. (5) and (6).

c-plane AlN homoepilayers.[24] Homoepilayers can be considered as almost free of TDs. The results thus indicate that PL characteristics are very sensitive to N_{screw} in AlN epilayers and PL measurement is an effective method to assess relative N_{screw} in AlN epilayers.

In summary, we have investigated the effects of Si-doping on the structural and optical properties of AlN epilayers. The undoped AlN epilayer was found to be under compressive stress that was almost fully relaxed by doping with Si of about $N_{\text{Si}}=5\times10^{17}$ cm^{-3}. Further increase in N_{Si} between 2×10^{18} and 10^{19} cm^{-3} induced tensile stress in the epilayer. A direct correlation between the density of edge dislocations and in-plane stress in the films suggests that accumulation of tensile stress in Si-doped AlN is a reason for the increase in edge dislocations. Both screw and edge type TDs were found to increase with N_{Si}. However, N_{edge} decreased in highly doped ($N_{\text{Si}}>5\times10^{18}$ cm^{-3}) AlN samples. The most significant effect of Si-doping on structural properties was that N_{screw} increased monotonically with N_{Si}. A direct correlation between N_{screw} and the PL characteristics was observed. Specifically, the emission linewidth increased linearly with increasing N_{screw} at a rate of $\sim3.3\pm0.7$ meV/10^8 cm^{-2} for I_2 and $\sim26.5\pm4$ meV/10^8 cm^{-2} for impurity transitions. The results thus establish that PL is an effective method to estimate N_{screw} in AlN.

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