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Influence of Al doping on lattice strain and electrical properties of epitaxial GaN films grown by metalorganic chemical vapor deposition on Al_2O_3 substrate

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The effects of the isoelectronic Al doping of epitaxial GaN films grown by metalorganic chemical vapor deposition on a (0001) Al_2O_3 single crystal substrate were investigated. It was found that the threading screw and edge dislocation densities of the GaN film decreased to less than half of that of the undoped GaN film up to Al doping concentration of 0.45%. The in-plane and out-of-plane strains were simultaneously reduced with the decrease in dislocation density as a result of the solution hardening effect. Accordingly, the electron mobility of the 0.45% Al-doped GaN film ($524\ cm^2/Vs$) was greatly improved compared to that of the undoped GaN film ($178\ cm^2/Vs$). However, the threading dislocation densities and strains were increased at a 0.64% Al concentration, and the electron mobility decreased accordingly. Therefore, the improvement in the electron mobility by Al doping up to 0.45% is the result of a decrease in the threading dislocation density and not a decrease in the number of point defects (Ga-site vacancy) as suggested earlier [Lee *et al.*, Appl. Phys. Lett. **83**, 917 (2003)]. © *2004 American Institute of Physics.*
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$Al_xGa_{1-x}N$/GaN heterostructure field effect transistors have recently attracted a great deal of interest for their applications in areas such as high power, high temperature, and microwave devices, due to their high electric breakdown field, large band gap, high thermal stability and high saturation-electron-drift velocity.[1,2] However, it is still difficult to grow high-quality epitaxial GaN films because a high density of treading dislocations and deep levels are inevitably generated as a result of the large lattice mismatch (16%) and the difference in the thermal expansion coefficients between the GaN thin film and the sapphire (Al_2O_3) substrate. Lee *et al.* reported that isoelectronic doping of a small concentration of Al (<1%) in GaN was effective in improving the device performance.[3] They attributed these improvements to the reduction in the number of point defects [Ga-site vacancies (V_{Ga}) and V_{Ga} complexes].[4] However, it is difficult to understand why Al doping reduced the V_{Ga} during the film growth because V_{Ga}, as a point defect, is not the V_{Ga} until the GaN crystal is formed and the incorporation of Al atoms does not have any specific reason to fill out the V_{Ga} during the film growth. Furthermore, how the strain state of the GaN films changes with increasing Al concentration has not been explored.

In this letter, it was shown that the improvement in the electron mobility is attributed to the reduction of the threading dislocation density of GaN with Al doping (<0.45%) and not by the decrease in point defect density. It was also found that Al doping relieves both the in-plane and out-of-plane strain of the GaN film up to 0.45% as a result of the solution hardening effect. However, at a high concentration of 0.64% Al, the electron mobility of the GaN seriously deteriorated rapidly presumably by the substitution of Ga ions with Al ions ($Al_xGa_{1-x}N$ alloy formation).

Undoped GaN films and a series of Al-doped GaN films were grown on a (0001) sapphire (Al_2O_3) substrate by a metalorganic chemical vapor deposition (MOCVD) at a growth temperature of 1020 °C. Trimethylgallium (TMGa), ammonia (NH_3), and trimethylaluminum (TMAl) were used as the source materials for Ga, N, and Al, respectively. Details of the MOCVD conditions are reported elsewhere.[3,4] Five samples were grown at different TMAl flow rates of 0 (undoped), 3, 6, 10, and 15 μmol/min to obtain differently doped epitaxial GaN films.

The Al concentrations in the GaN films were estimated using the measured photoluminescence (PL) and transmission data, assuming a bowing parameter of 0.69 eV.[4] The x-ray measurements were performed using a high-resolution x-ray diffractometer (Philips X'Pert MRD) with a Cu sealed anode and a four-crystal monochromator in a Ge (220) configuration. The rocking curve (ω scan) and radial 2θ-ω scans of the (002) and (302) Bragg spots were measured, the latter was in skew symmetric diffraction geometry using an Eulerian cradle. The lattice constants, c and a, were obtained using the symmetric (002) 2θ-ω scan and asymmetric (105)

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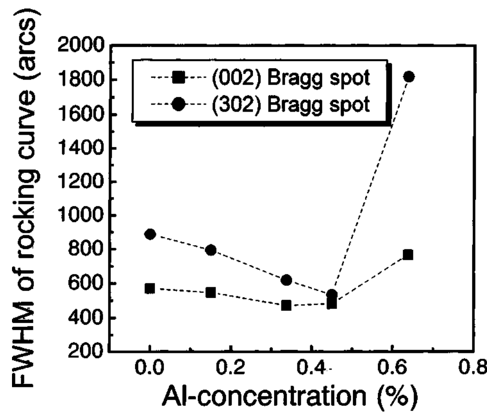

FIG. 1. FWHM values of the rocking curve of the GaN (002) and (302) Bragg reflections in TCD mode as a function of the Al-doping concentration.

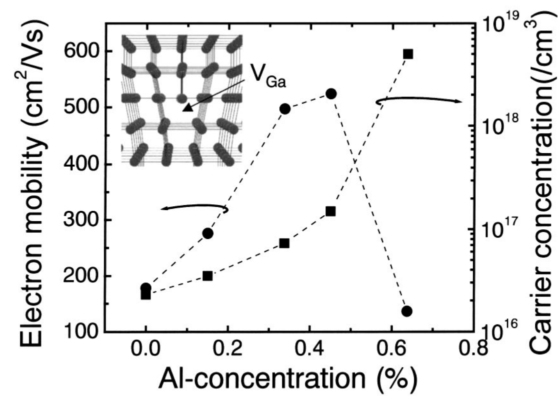

FIG. 3. Variations in the electron mobility and free carrier concentrations of the Al-doped GaN films as a function of the Al-doping concentration. The inset shows a schematic diagram of the V_{Ga} related to the dislocation cores.

reflections, respectively. The experiments were performed in triple crystal diffraction geometry taking the refraction correction into account.

Figure 1 shows the variation in the full width at half-maximum (FWHM) values of the rocking curve of the GaN (002) and (302) Bragg spots as a function of the Al-doping concentration. The FWHM values decrease with increasing Al concentration up to 0.45% and then increase at a 0.64% Al concentration. It is well known that screw threading dislocations with the Burgers vector $\mathbf{b}=[0001]$ causes a tilt of the $[0001]$ oriented GaN layers, while the edge dislocation with $b=1/3\langle 11\bar{2}0\rangle$ induces a twist by forming small angle boundaries in the GaN films.[5,6] Because the broadening of the symmetric (002) and asymmetric (302) reflections are caused mainly by the tilt and twist of the small domains, the data in Fig. 1 can be considered to represent the screw and edge dislocation densities, respectively.[5,6] Therefore, it can be understood that the threading dislocation densities decreased with increasing Al-doping concentration up to 0.45%, and increase rapidly at 0.64% Al concentration. The overall threading dislocation density of the GaN films with a 0.45% Al concentration ($\sim 1.8\times 10^9$ cm^{-2}) was reduced to less than half of that of the undoped GaN film ($\sim 5.0\times 10^9$ cm^{-2}). The reason for the decrease in the dislocation density can be found from the following results.

Figure 2 shows the variations in the strain along the out-of-plane $[\Delta c/c=(c-c_0)/c_0\times 100(\%)]$ and in-plane $[\Delta a/a=(a-a_0)/a_0\times 100(\%)]$ directions measured by the

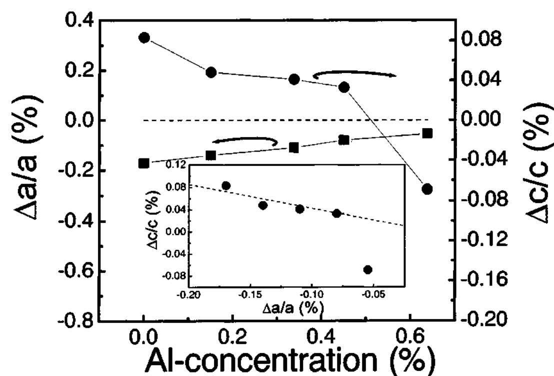

FIG. 2. Strains for the lattice constants c and a for the Al-doped GaN epitaxial films as a function of the Al-doping concentration. The inset shows the correlation between the strains for the lattice constants c and a.

position shifts of the GaN (002) and (105) Bragg spots. It can be understood that the in-plane strain decreases with increasing Al concentration up to 0.64% whereas the out-of-plane strain decreases up to 0.45% and then increases at the 0.64% of Al concentration. The negative and positive signs of the in- and out-of-plane strain, respectively, up to the 0.45% Al concentration suggests that the film is under in-plane compressive stress due to lattice mismatch. However, it can be understood that the film is under a different stress state at the 0.64% Al concentration from the negative values of both the in- and out-of-plane strains.

The inset in Fig. 2 shows the correlation between the in- and out-of-plane strains. The dashed line in the inset shows the theoretical correlation between the two strains, $\Delta c/c=-2(C_{13}/C_{33})\Delta a/a$, where C_{13} and C_{33} are the elastic stiffness coefficients of the bulk GaN. C_{13}/C_{33} of GaN is 0.23.[7] It can be understood that the plot of $\Delta c/c$ vs $\Delta a/a$ lies along the theoretical line for the Al concentration up to 0.45%. It is believed that the incorporated Al ions do not substitute for Ga ions but locate at the core of the threading dislocations, as schematically shown in the inset in Fig. 3, up to a 0.45% Al concentration due to their smaller radius (0.05 nm for Al^{3+} and 0.062 for Ga^{+3}). The filling out of the V_{Ga}'s at the dislocation cores by the isoelectronic Al ions eliminates the acceptor-like defects, and the electron density increases and the mobility is improved.

The deviation of the strain from the theoretical predictions at the 0.64% Al concentration suggests that some of the Al begins to substitute for Ga ions forming an $Al_xGa_{1-x}N$ mixed crystal at this Al concentration. The substituted Al ions in the Ga sites will cause local lattice strain and an increased threading dislocation density due to the difference in the atomic radii, as shown in Fig. 1(a).

It is important to note that Al doping into GaN results in a reduction of the threading dislocation density as well as the strain of GaN films at the same time up to a 0.45% Al concentration. Yamaguchi *et al.*[8] also reported a similar result in that the isoelectronic In-doped GaN became more strain-free and had a lower dislocation density than the undoped GaN. In the case of heteroepitaxy, the films generally grow coherently with respect to the substrate lattice up to a certain critical thickness, which depends on the lattice mismatch and the stiffness of the growing film, but is usually <10 nm without

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forming misfit dislocations. Above the critical thickness, misfit dislocations are generated in order to minimize the sum of the strain and dislocation energies of the film for a given lattice mismatch. Therefore, a larger dislocation density in the thick epitaxial film suggests less strain in the film. However, the results in this study are certainly in contrast to the general trend. Therefore, a new model will be required to explain these experimental results.

The fact that both the in- and out-of-plane strains decrease with increasing Al doping concentration up to 0.45% in Fig. 2 suggests that the GaN films become stiffer with increasing Al-doping concentration. In other words, the GaN crystal lattice become closer to what it should be even under the influence of the substrate lattice. This might be understood from the following reasons. When the doped Al atoms reside at the dislocation core, the impurity Al atoms will interact with the strain field induced by the dislocation, forming a Cottrell atmosphere.[9] Hence, a pinning the glide of the dislocation is expected, leading to the so-called ''solution hardening.'' Therefore, the Al-doped GaN becomes stiffer and recovers its lattice parameters. A stiffer material must have a lower dislocation density under a given stress as a result of the lattice and thermal expansion mismatches. In fact, the decrease in the threading dislocation density as a result of Al doping can also be understood from the solution hardening effect where the local strain introduced by the impurity suppresses the dislocation generation, motion, and multiplication.

Figure 3 show the variations in the electron mobility and free electron concentration of the Al-doped GaN epitaxial films as a function of the Al-doping concentration. Data points up to 0.45% were taken from Ref. 4 and those for the 0.64% were measured. The electron mobility of the GaN thin film significantly increased from 178 cm^2/Vs of the non-doped sample to 524 cm^2/Vs of the 0.45% Al-doped sample. However, further increases in the Al concentration results in a degradation of the electron mobility, and hence, it falls to 138 cm^2/Vs at a 0.64% Al concentration.

Wright *et al.*[10] suggested based on *ab initio* calculations that V_{Ga} or V_{Ga} complex at the threading edge dislocation in the n-type GaN should behave as electron traps and therefore be negatively charged. It should be noted that these V_{Ga}'s are not the usual point defects but are associated with the edge dislocation cores, as shown by the inset in Fig. 3. The charged edge dislocation will scatter the conduction electrons resulting in the mobility degradation.[11–13] Therefore, it can be concluded that the increasing electron mobility of the GaN films with increasing Al-doping concentration up to 0.45% Al is mainly due to a reduction of the threading edge dislocation density. It should be noted that the edge dislocation density of $\sim 5.0\times 10^9$ cm^{-2} and 2.4×10^9 for undoped and 0.34% Al-doped GaN corresponds to a dislocation core-associated V_{Ga} density of 1×10^{17} and 4.8×10^{16} cm^{-3}, respectively. Therefore, when the Al-doping concentration of GaN increases from 0% to 0.34%, the increase in the electron concentration by $\sim 5.2\times 10^{16}$ cm^{-3} should be obtained if this increase is due to a reduction in the edge dislocation density. Figure 3 shows that the net electron density increases by 5.0×10^{16} cm^{-3} when the doping concentration changes from 0% to 0.34% Al. This agreement confirms that the improvement in the electron mobility of the Al-doped GaN is mainly due to a reduction in the dislocation density.

However, the electron mobility for the 0.64% Al-doped GaN decreased, which was attributed to the increased ionized-impurity scattering as a result of rapidly increasing dislocation density above that concentration. It is well known that in GaN with a high dislocation density ($>10^8$ cm^{-2}), dislocation scattering dominates below a certain electron concentration ($\sim 10^{17}$ cm^{-2}), while ionized-impurity scattering is dominant above that electron concentration. Figure 3 shows that the free carrier concentration of the Al-doped GaN films monotonously increases with increasing Al concentration.

In summary, the structural and electrical properties of the isoelectronic Al-doped GaN epitaxial films grown by MOCVD were investigated. The incorporated Al ions, up to a 0.45% concentration, were found to reduce the overall threading dislocation of GaN films and relieve the in-plane and out-of-plane strains simultaneously. The improvement in the electron mobility of the Al-doped GaN was mainly due to a reduction in the threading edge dislocation density, and thus, the electron scattering centers.

However, at a 0.64% Al concentration, the threading screw and edge dislocation densities increased rapidly and the electron mobility became seriously degraded because some of the doped Al ions substituted for Ga ions at this Al concentration forming $Al_xGa_{1-x}N$ alloy.

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