Improvement on epitaxial grown of InN by migration enhanced epitaxy

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(Received 5 June 2000; accepted for publication 23 August 2000)

Epitaxial growth of InN on (0001) sapphire with an AlN buffer layer was studied by migration-enhanced epitaxy, which is composed of an alternative supply of pure In atoms and N_2 plasma. A series of samples were prepared with different substrate temperatures ranging from 360 to 590 °C. As-grown films were characterized by x-ray diffraction (XRD), reflective high-energy electron diffraction, atomic-force microscopy (AFM), and Hall measurements. Both XRD θ–2θ and ω scans show that the full width at half maximum of the (0002) peak nearly continuously decrease with increasing growth temperature, while InN grown at 590 °C shows the poorest surface morphology from AFM. It is suggested that three-dimensional characterization is necessary for an accurate evaluation of the quality of the InN epilayer. Hall mobility as high as 542 $cm^2/V\,s$ was achieved on film grown at ~500 °C with an electron concentration of 3×10^{18} cm^{-3} at room temperature. These results argue against the common view that nitrogen vacancies are responsible for the high background *n*-type conductivity of InN. To illuminate the relationship between Hall mobility and carrier concentration, the electrical properties of all InN films grown recently were summarized. © *2000 American Institute of Physics.* [S0003-6951(00)00942-6]

Indium nitride is an important III–V compound semiconductor with various potential microelectronic and optoelectronic applications. The use of InN and its alloys with GaN and AlN makes it possible to extend the emission of nitride-based light-emitting diodes from ultraviolet to red.[1] Although the band gap of InN is 1.89 eV at room temperature, which is smaller than that of GaN which is 3.39 eV, it is larger than that of GaAs, 1.42 eV, and is certainly large enough for many high-power and high-energy applications. More importantly, the inherent material characteristics of InN make it superior to GaN in some very important aspects. A much higher peak drift velocity compared with GaN was predicted by theoretical calculations over a wide range of temperatures from 150 to 500 K and a doping concentration up to 10^{19} cm^{-3}.[2] The transient electron transport, which is expected to be the dominant transport mechanism in submicron-scale devices, was also studied on InN.[3] It is found that InN exhibits the highest peak overshoot velocity and that this velocity overshoot lasts over the longest distance when compared with GaN and AlN.

However, unlike the intensively studied GaN, InGaN, and other nitride compounds, InN remains as one of the least studied nitride materials. This is believed to be mainly due to the difficulty in preparation of high-quality InN epilayers. Because of the low InN dissociation temperature, and the high equilibrium N_2 vapor pressure over the InN film, the preparation of InN requires a low growth temperature.[4] Meanwhile, for common III–V nitride epitaxy techniques, such as metalorganic vapor-phase epitaxy and molecular-beam epitaxy (MBE), low growth temperature means a short migration distance of group-III atoms. That is, many group-III atoms will have less energy to travel long enough on the surface to locate their energy minimum sites before they react with N to form small less-mobile nitride islands. As a result, nitride film with high defect density will form. Therefore, in order to achieve a high-quality InN epilayer, the above problem must be solved.

In this study, we report epitaxial growth of InN on (0001) sapphire with an AlN buffer layer by migration-enhanced epitaxy (MEE), which is composed of an alternative supply of pure In atoms and nitrogen plasma. The advantage of MEE has been well studied.[5] It is exactly a good way to enhance the migration distance of group-III atoms during low-temperature epitaxial growth. A series of samples were prepared with different substrate temperatures ranging from 360 to 590 °C. We investigated the dependence of crystallization, surface morphology, carrier concentration, and mobility for the InN films over this range of substrate temperatures. Hall mobility as high as 542 $cm^2/V\,s$ was achieved on film grown at ~500 °C with an electron concentration of 3×10^{18} cm^{-3} at room temperature. This is among the best results ever reported on InN in recent years.

The growth of InN was performed with a turbopumped Varian GEN-II gas-source MBE chamber with a background pressure of 1×10^{-10} Torr, conventional effusion cells, and an EPI unibulk rf plasma source for generation of nitrogen radicals. The nitrogen flux through the plasma source is fixed at 0.7 sccm, causing a nitrogen partial pressure in the MBE chamber of 1.8×10^{-5} Torr during growth. The whole growth can be separated into three steps. In the first step, (0001) sapphire wafers with backside sputter-coated Ti/W were nitridated by nitrogen plasma at the high temperature of 1100 °C for 3 min; then, a 2 nm AlN nucleation layer was deposited at 950 °C by the conventional MBE technique. Finally, the substrate was cooled down slowly to the desired InN growth temperature. The MEE growth was carried out with an alternative supply and consisted of a four-step sequence: In supply (2 s), interrupt (2 s), nitrogen plasma (2 s),

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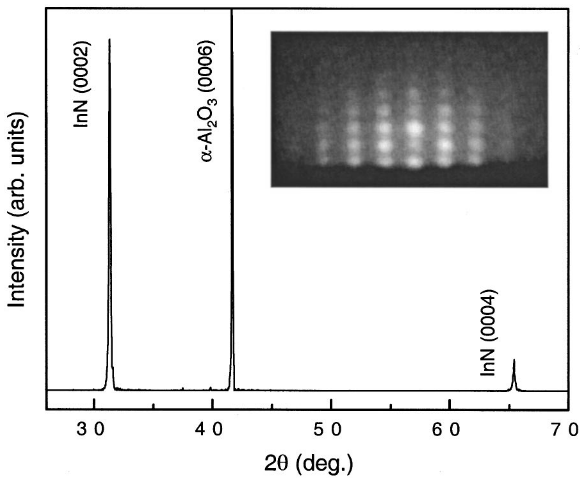

FIG. 1. Typical θ–2θ XRD profile of the grown InN by MEE. The inset is a typical spotty RHEED pattern observed along the $[10\bar{1}0]$ azimuth during growth.

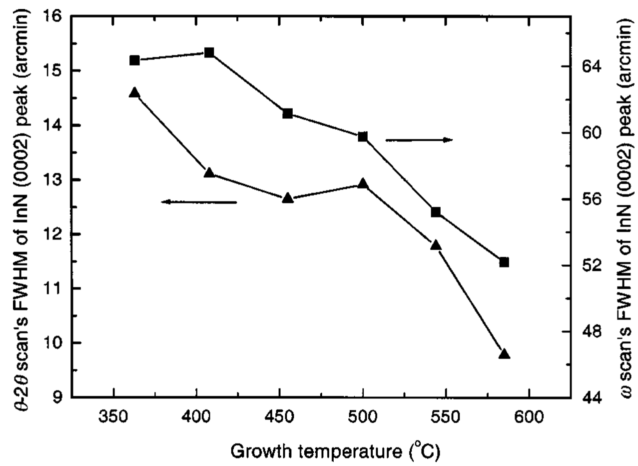

FIG. 2. Substrate temperature dependence of the FWHM of the InN (0002) peak in the XRD θ–2θ scan and ω scan.

and interrupt (2 s). A series of InN films were prepared with different substrate temperatures to optimize the best growth condition. All of them were grown on 1/4 2-in.-size wafers with a final film thickness around 0.1 μm. Thicker InN films grown on larger size wafers were also tried in this study. The growth rate was found to be $\sim$0.1 μm/h for all the grown films, which is nearly independent of growth temperature in the range studied.

X-ray diffraction (XRD) measurements and reflective high-energy electron diffraction (RHEED) were used to characterize the crystalline structure of the grown films. Figure 1 shows a typical θ–2θ XRD profile of the grown InN film. As shown in Fig. 1, wurtzite-type InN (0002) and (0004) peaks were clearly observed, indicating that a highly *c*-axis oriented InN film was formed. All the growth processes were monitored by RHEED. Usually, a spotty RHEED pattern will appear to replace the streaky AlN RHEED pattern soon after the MEE begins and will keep until the end of growth. The spotty RHEED pattern of InN suggests a nonplanar growth front and three-dimensional growth mode. The inset of Fig. 1 is a typical spotty RHEED pattern of the grown InN, which again verifies its epitaxial character. From the evolution of the RHEED patterns throughout the growth, the relationship of the crystallographic orientations between InN, the AlN buffer, and the sapphire substrate is found to be $[2\bar{1}\bar{1}0]_{\mathrm{InN}} \| [2\bar{1}\bar{1}0]_{\mathrm{AlN}} \| [10\bar{1}0]_{\alpha\text{-}\mathrm{Al_2O_3}}$.

Figure 2 shows the temperature dependence of the full width at half maximum (FWHM) of the InN (0002) peak in the XRD θ–2θ scan and ω scan. In Fig. 2, the FWHM in both scan modes nearly continuously decreases with increasing growth temperatures. Since it is commonly believed that in XRD measurements, the ω and θ–2θ scans represent the crystalline mosaicity and degree of distribution of the lattice constant, respectively, this observation implies that the best crystalline quality can be obtained at the highest growth temperature of 590 °C.

However, in atomic-force microscopy (AFM) measurements, while most of the samples grown below 550 °C exhibit acceptable surface morphology with root-mean-square (rms) roughness around 10 nm, and InN film grown at around 590 °C shows a very rough surface with rms roughness near 30 nm, even though this sample has the smallest FWHM of the (0002) peak in the XRD measurement. Therefore, the above observations indicate that only *c*-axis quality alone cannot reflect all the crystalline aspects of the grown InN. In-axis XRD combined with AFM or off-axis XRD is necessary for an accurate characterization. The rough surface should result from the dissociation effect of InN at high temperature, while the improved crystal quality may be due to the annealing process[6] occurring at the same time. Thus, those high-temperature-grown films should consist of small InN islands of good crystalline quality. In this study, InN films grown at temperature higher than 600 °C were also tried. All of them exhibit a rougher surface with tiny In droplets visible under a microscope.

The electrical properties were determined by Hall measurements with Van der Pauw geometry. Figure 3 shows the room-temperature Hall mobility and carrier concentration of the InN films as a function of the substrate temperature. All the films show *n*-type conductivity with carrier concentrations in the order of 10^{18}–10^{19} cm^{-3}. The Hall mobility first increases with raising the substrate temperature from 350 to 500 °C, and then decreases as the temperature is further

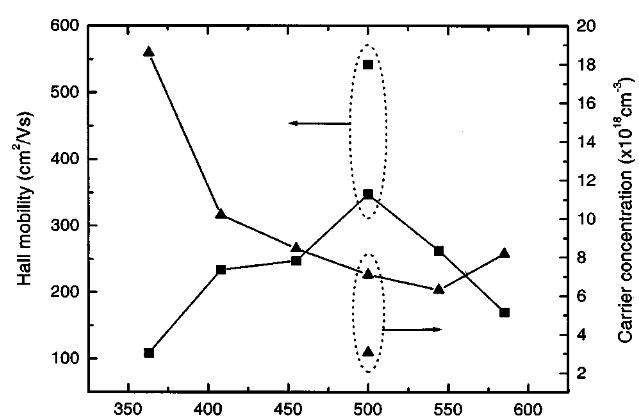

FIG. 3. Room-temperature Hall mobility and carrier concentration of the InN films as a function of substrate temperature. The separate points on the figure correspond to the electrical properties of a special sample grown on 2 in. sapphire with 0.2 μm thickness.

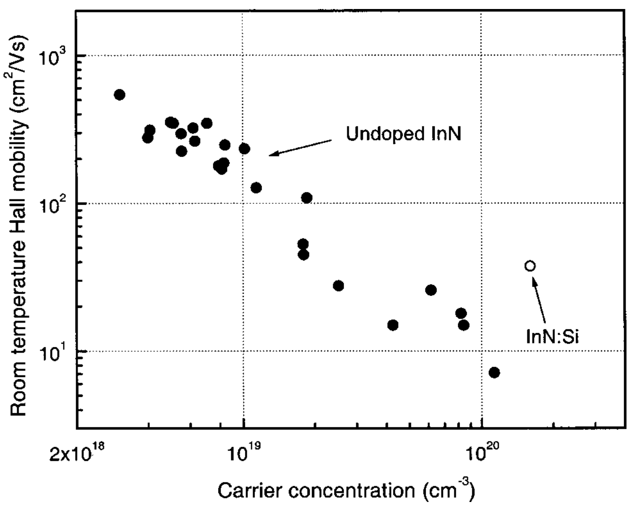

FIG. 4. Summary of Hall data of all InN films grown recently.

raised. Its peak value at substrate temperature 500 °C is 348 cm^2/V s with a carrier concentration 7.1×10^{18} cm^{-3}. The evolution of carrier concentration with substrate temperature is nearly of the opposite trend compared with Hall mobility. It is expected that the improvement of electrical properties of InN films with increasing substrate temperature in the range of 360–500 °C is associate with better crystalline quality at higher growth temperature, which has been reflected in the XRD measurement. Meanwhile, the degradation of electrical properties in the temperature range of 500–600 °C should be a result of the dissociation effect of InN at high temperature, which has also been revealed form the poor surface morphology of the AFM measurements. The electrical properties of corresponding InN films grown by the conventional MBE technique are not shown in Fig. 3 for comparison. It is because those films grown by MBE at the same system usually have a very poor surface. Some of them are even powder-like. It is difficult to make metal contacts on them for Hall measurements and even when made, their data will be unreliable. Therefore, the advantage of MEE over MBE for growing InN is apparent in this study.

The separate points in Fig. 3 correspond to the Hall data of an InN sample grown on 2 in. wafer with a film thickness of 0.2 μm at 500 °C. Its mobility is as high as 542 cm^2/V s at room temperature with a low carrier concentration of 3×10^{18} cm^{-3}. Compared with its counterpart, which was grown on a 1/4 2 in. wafer with a film thickness of 0.1 μm, its electrical properties were further improved. The reason for this improvement is under investigation. It is expected that a more uniform temperature field can be achieved on a larger substrate during growth and better epitaxial character may be obtained for thicker films. Actually, it has been observed that the spotty RHEED pattern of grown InN can become a little streaky if the growth time is prolonged.

It is worth noting that the results of Fig. 3 argue against the common view that nitrogen vacancies are responsible for the high background *n*-type conductivity of InN. With increasing substrate temperature, more nitrogen atoms should evaporate and the concentration of the nitrogen vacancies should increase. If the vacancies were the major donors, the carrier concentration should monotonically increase with increasing substrate temperature. This picture is clearly different from our observation. Therefore, other defects are probably responsible for the high background *n*-type doping.

To further explore the relationship between the Hall mobility and carrier concentration of InN films, all the Hall data of recent InN growths are summarized in Fig. 4, which include not only the data in the above discussion, but also the data of InN films grown with different buffer-layer treatments and InN films grown at varied substrate temperatures. From Fig. 4, it is clear that in order to achieve InN films with higher mobility, the carrier concentration should be further reduced. Thus, more attention should be focused on the origin of background doping of InN. In addition, these data are of the same range as the calculations by Transley's group.[7]

In summary, epitaxial growth of InN by migration-enhanced epitaxy was studied with various substrate temperatures. The optimum substrate temperature was found to be around 500 °C, at which a high Hall mobility of 542 cm^2/V s was achieved with a carrier concentration of 3×10^{18} cm^{-3} at room temperature. The advantage of MEE growth for InN over the conventional MBE technique was demonstrated. The results of this study do not agree with the common view that nitrogen vacancies are responsible for the high background *n*-type doping of InN.

The authors would like to thank Dr. Michael J. Murphy for his kind tutoring on the MBE machine and Dr. Roman Dimitrov for the fruitful discussions with him. This work is supported by ONR Contract No. N000149910936 monitored by Dr. Colin Wood.

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