High quality InAlN/GaN heterostructures grown on sapphire by pulsed metal organic chemical vapor deposition

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High quality InAlN/GaN heterostructures are successfully grown on the (0 0 0 1) sapphire substrate by pulsed metal organic chemical vapor deposition. The InAlN barrier layer with an indium composition of 17% is observed to be nearly lattice matched to GaN layer, and a smooth surface morphology can be obtained with root mean square roughness of 0.3 nm and without indium droplets and phase separation. The 50 nm InAlN/GaN heterostructure wafer exhibits a mobility of 1402 cm²/Vs with a sheet carrier density of 2.01 × 10¹⁳ cm⁻² and a low average sheet resistance of 234 Ω/cm² with a sheet resistance nonuniformity of 1.22%. Compared with the conventional continual growth method, PMOCVD reduces the growth temperature of the InAlN layer and the Al related prereaction in the gas phase, and consequently enhances the surface migration, and improves the crystallization quality. Furthermore, indium concentration of InAlN layer can be controlled by adjusting the pulse time ratio of TMIn to TMAl in a unit cycle, the growth temperature and pressure, as well as the InAlN layer thickness by the number of unit cycle repeats.

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1. Introduction

In the past decade, AlGaN/GaN-based high electron mobility transistors (HEMTs) have attracted considerable research attention and significant progresses have been made for high power and high-frequency electronics' applications [1,2]. The advantages of the nitride material system for such applications are attributed to their large band gap, high breakdown field, and strong spontaneous and piezoelectric polarization, which induce a two-dimensional-electron-gas (2DEG) with high electron concentration in the heterostructure interface. Usually, a high Al composition is used to increase the two-dimensional-electron-gas density, so as to improve device performance [3]. However, AlGaN barrier layer with a high Al composition is highly strained with respect to GaN, and the resultant large tensile strain could lead to plastic relaxation when the thickness of AlGaN layer is larger than its critical thickness [4,5]. Also, the increase in Al composition is accompanied with enhanced lattice mismatch effects, which rapidly degrade the crystalline quality of barrier layer, which in turn leads to a reduction of the carrier mobility. Moreover, the large strain between AlGaN and GaN would have further effects on the reliability of AlGaN/GaN HEMTs [6].

To circumvent this problem, InAlN can be adopted as an alternative barrier layer where the In composition can be adjusted to approximately 17.6%, in order to obtain an in-plane nearly lattice matched InAlN/GaN heterostructure. In this case, the polarization charge is completely determined by spontaneous polarization since the structure is free from strain so the piezoelectric polarization is nearly zero. Kuzmik has predicted a high 2DEG density of 2.7 × 10¹⁵ cm⁻² when InAlN is nearly lattice matched to GaN. It is due to the large spontaneous polarization (Psp) difference between InAlN and GaN (ΔPsp = 0.043 C/m²) and the large conduction band discontinuity (ΔEc = 1.1 eV), even though the piezoelectric component vanished due to the absence of strain [7]. However, from the epitaxial point of view, it is very difficult to grow high quality InAlN according to the state-of-the-art of epitaxial technology. The first problem is phase separation and composition inhomogeneity due to the large thermodynamic miscibility gap caused by the large difference of covalent bond length between AlN and InN [8–11]. Besides, InN is normally grown under the condition of low temperature (600 °C), high ammonia partial pressure, and relatively high chamber pressure, while it requires a temperature exceeding 1100 °C, low ammonia flow rates, and a low pressure for AlN deposition to avoid material losses that arises from gas-phase processes involving aluminum. The challenge in realizing high quality InAlN layer is to obtain the optimized growth conditions that work simultaneously for both AlN and InN. Recently, some research groups succeeded in the growth of InAlN/GaN heterostructures by molecular beam epitaxy
(MBE) and MOCVD [12–16]. Miyoshi et al. [17] have achieved a high 2DEG density of more than \( 2.6 \times 10^{13} \text{cm}^{-2} \) and high 2DEG mobility of 1170 cm\(^2\)V s for near lattice matched MOCVD-grown InAlN/GaN heterostructures with the barrier thickness of more than 15 nm. On the other hand, MBE grown samples for nearly lattice matched structures show the sheet carrier density of 2.68 \( \times 10^{13} \text{cm}^{-2} \) and mobility of 1080 cm\(^2\)V s at room temperature [13]. Especially, Lim et al. [18] presented nearly lattice matched In\(_{0.07}\)Al\(_{0.40}\)Ga\(_{0.53}\)N/GaN heterostructures grown by MBE exhibiting a sheet carrier density of 1.9 \( \times 10^{13} \text{cm}^{-2} \) and an electron mobility of 1460 cm\(^2\)V s recently. Particularly, the research group of the Ecole Polytechnique Federale De Lausanne lead by Marc Ilegems and then by Nicolas Grandjean has made good success in the growth of InAlN for distributed Bragg reflectors (DBRs) for use in vertical cavity surface emitting lasers and similar structures [19,20]. In addition to those traditional continual MOCVD method, we have successfully grown high quality InAlN/GaN heterostructures grown on the (0 0 0 1) sapphire substrate and detailed characterizations [19,20].

InAlN/GaN heterostructures were grown on 2 in. (0 0 0 1) sapphire substrate in a homemade vertical metal chemical vapor deposition (MOCVD) system with a 1 \( \times \) 2 in. close-coupled showerhead reactor. Fig. 1 shows schematic diagram of InAlN/GaN heterostructures in our experiments. After ultrasonic cleaning in organic solvents, chemical etching in a hot \( \text{H}_2\text{PO}_4: \text{H}_2\text{SO}_4 \) (1:3) solution, and rinsing in deionized water, sapphire substrate was placed on a silicon carbide coated graphite susceptor in the chamber and heated with rf induction. The growth temperature was the susceptor temperature and monitored using a thermocouple inserted under the susceptor. Trimethylgallium (TMGa), trimethylaluminum (TMAI), trimethylindium (TMIn), and ammonia (\( \text{NH}_3 \)) were used as Ga, Al, In, and N precursors, respectively. Prior to the epitaxial growth, the substrate was annealed at 1060 °C for 15 min in a nitrogen environment to remove the surface oxides. The growth was initiated with a 20 nm thick low temperature AlN nucleation layer at a temperature of 620 °C followed by a 60 nm thick high temperature AlN buffer layer grown at 1075 °C. Then, a 2 \( \mu \text{m} \) thick nominally undoped GaN layer was grown at 940 °C. Afterward, a 1 nm AlN interlayer was grown at a temperature of 940 °C to achieve a large electron wave function separation between InAlN barrier layer and the underlying GaN layer so as to diminish the impact of interface roughness and reduce alloy disorder scattering, resulting in an improved lateral transport property of carrier and better 2DEG confinement. GaN and AlN layers were grown using conventional MOCVD and the growth pressures were kept at 40 Torr. The fluxes of TMGa and ammonia for GaN growth were 56 \( \mu \text{mol/min} \) and 2500 sccm (sccm denotes cubic centimeter per minute at standard temperature and pressure), respectively, whereas the fluxes of TMAI and ammonia for InAlN growth were 3 \( \mu \text{mol/min} \) and 1500 sccm, respectively. Hydrogen was used as the carrier gas for the AlN and GaN layers growth with a flux of 2750 sccm, which was held constant.

The sample was then cooled down for the growth of a 14 nm thick InAlN barrier layer. Here, PMOCVD approach was used to deposit AlN/InN short period superlattices for InAlN barrier layer regions. The growth was carried out using the growth conditions previously optimized for nearly lattice matched InAlN/GaN heterostructures. InAlN epitaxial layer was grown using nitrogen as the carrier gas with a flux of 2750 sccm. Previously, we have optimized the growth conditions for InAlN, such as V/III ratio, precursor flux of TMAI and TMIn, and InAlN growth temperature and pressure. The optimized precursors’ fluxes of TMAI and TMIn were 2.3 and 5.8 \( \mu \text{mol/min} \), respectively. The ammonia flux was 1200 sccm and optimized V/III was \( \sim 6614 \). The best crystal quality was obtained under the growth pressure of 200 Torr at 760 °C. These efforts make it possible to grow single crystalline In\(_{0.07}\)Al\(_{0.40}\)Ga\(_{0.53}\)N (\( \sim 0.17 \)) film nearly lattice matched to GaN without phase separation. The use of PMOCVD allowed us to reduce the growth temperature and hence significantly increased indium incorporation, resulting in high quality InAlN layers. The growth sequence of the unit cells of AlN/InN is described in Fig. 2. As can be seen, there are two unit cells, 6 s pulses of TMAI, \( \text{NH}_3 \), and 24 s pulse of TMIn were introduced alternately into the MOCVD reactor. An ammonia pulse always followed the metal organic pulses. The precursor flux in an individual pulse was adjusted in such a way that the deposited thickness in each unit cell, determined from the total layer thickness divided by the number of unit cell repeats, was around 1.4 Å. The deposited thickness can be accurately controlled and the pulse cycle was repeated 100 times, resulting in a film thickness of \( \sim 14 \) nm. The whole layer thickness was estimated from the growth rate and confirmed by spectroscopy ellipsometry [21].

The crystalline quality and the indium composition of InAlN barrier layer were determined by high resolution X-ray diffraction (HRXRD). XRD was performed using a Bruker D8 high resolution diffractometer system, delivering CuK\(_a1\) (1.540 Å) radiation using a prodded mirror and a four-bounce Ge (2 2 0) symmetric monochromator. Surface morphology was observed by atomic force microscopy (AFM) using an Agilent 3500 and the surface roughness was deduced from root mean square (rms) roughness values of \( 5 \times 5 \) and \( 2 \times 2 \) \( \mu \text{m}^2 \) scans. The rms roughness of each sample was measured alone, i.e. one scan for
different scales. To understand temperature dependence of the 2DEG properties, the Hall effect measurement was carried out by the van der Pauw technique in the temperature range from 77 to 620 K using a Lake Shore Hall effect measurement system. Low frequency capacitance voltage (C-V) analysis was performed at 100 KHz using an Hg-probe contact. The sheet resistance and resistance nonuniformity of InAlN/GaN heterostructures were measured with Leighton contactless sheet resistivity mapping across the 50 mm wafer.

3. Results and discussion

Indium composition in the InAlN barrier layer was determined by XRD $\omega-2\theta$ scan. Owing to the thin InAlN barrier layer and the separate InAlN peak distinct from GaN peak, we could conduct (0 0 0 4) symmetric and (1 0 –1 5) antisymmetric reflections. Fig. 3 shows the (0 0 0 4) XRD $\omega-2\theta$ scan for a typical InAlN/GaN sample with InAlN thickness of 14 nm. The spectrum is dominated by the GaN peak at an angle of about 72.9° originating from the underlying GaN template. The additional peak seen at 76.8° is attributed to the AlN nucleation and interlayer. The InAlN barrier layer peak is observed at 74.6°. The lattice parameter $c$ can be directly calculated by the Bragg equation $c = \lambda/(2\sin \theta)$ [22], where $\lambda$ is the wavelength and $\theta$ is the Bragg angle. Along with parameter $c$, we can calculate lattice parameter $a$ from asymmetric ($1 0 1 5$) rocking curves; the dislocation density is unknown and is not given. Possible to measure XRD full width at half maximum (FWHM) of the biaxial strain in the layer, the indium composition can be determined by applying the relation [23]

$$x = \frac{\alpha(1 + v(x))}{\alpha(C_0 - \alpha_C(C_0 - \alpha_C + \alpha_C)}$$

where $\nu$ is Poisson’s ratio defined as $\nu = 2c_1/c_3$; $c_1$ and $c_3$ are the elastic constants of the hexagonal III-nitrides. The material constants used in this study are $a = 0.3111$ nm, $c = 0.498$ nm [24], $c_1 = 99$ GPa, and $c_3 = 389$ GPa for AlN [25]; $a = 0.35378$ nm, $c = 0.57033$ nm [26], $c_1 = 121$ GPa, and $c_3 = 182$ GPa for InN [27]. For In$_{x}$Al$_{1-x}$N ternary alloy, both lattice constants and Poisson’s ratio $\nu(x)$ are obtained by linear interpolation from the values of binaries. Finally, the indium composition of In$_{x}$Al$_{1-x}$N was calculated to be 0.17 ± 1%. Furthermore, it is noted that the wurtzite crystal structure of thin InAlN barrier layer has no indication of phase separation between InN and AlN from XRD analysis. Since the InAlN barrier layer is as thin as 14 nm, it is not possible to measure XRD full width at half maximum (FWHM) of rocking curves; the dislocation density is unknown and is not given.

We used an automated angle M-2000D rotating compensator from J.A. Woollam Co., Inc. in the wavelength range $\lambda = 400$–1000 nm at an angle of incidence $\theta = 65^\circ$ to determine the whole layer thickness. The thickness was determined by fitting the calculated dielectric functions to the measured wave forms using a Cauchy dispersion relationship model. Fig. 4 shows the excellent match between measured (dots) and calculated (lines) ellipsometric spectra. The thickness of the whole layer is 2076 ± 20 nm, which agrees well with the designed value (2094 nm). The values of refractive index $n$ are 2.08, 2.34, and 1.85 for AlN, GaN, and InAlN, respectively, at the incidence wavelength of 633 nm.

Fig. 5 shows the typical AFM surface images of InAlN/GaN heterostructures with a scan area of (a) $5 \times 5 \mu$m$^2$ and (b) $2 \times 2 \mu$m$^2$. A smooth surface with atomic steps and some small pits can be clearly seen. The pit density is approximately $4 \times 10^4$ cm$^{-2}$ in $2 \times 2 \mu$m$^2$ scan area. The flat surface accompanies spiral hillocks that can be related to the dislocations having screw components. In addition, the root mean square roughness was measured to be as low as approximately 0.3 nm for the $2 \times 2 \mu$m$^2$ scan area. This value is in good agreement with the previously reported data for MOCVD-grown InAlN/GaN heterostructures [17], and is less than the results grown by MBE [14], which suggests that PMOCVD is an alternative approach leading to a monoatomic smooth layer of thin InAlN film.

![Fig. 3. High resolution XRD triple-axis (0 0 0 4) $\omega-2\theta$ scan of InAlN/GaN heterostructures.](image)

![Fig. 4. Wave forms of (a) $\psi$ and (b) $\Delta$ for InAlN/AlN/GaN heterostructure on sapphire substrate obtained by spectroscopy ellipsometry at an incidence angle of 65° (dots) compared with calculated wave forms of $\psi$ and $\Delta$ (lines).](image)
The electrical properties of InAlN/GaN heterostructures were first measured by sheet resistance mapping of the 50 mm InAlN/GaN heterostructures wafer, as shown in Fig. 6. The wafer displayed a maximum resistance of 241 $\Omega$/cm² and a minimum value of 231 $\Omega$/cm². The average sheet resistance was 234 $\Omega$/cm² with resistance nonuniformity of 1.22%, showing the high quality of the GaN channel layer and the smooth interface. This low resistance nonuniformity has never been reported earlier in literature.

For additional electrical information, such as the sheet carrier concentration and the 2DEG confined position, $C-V$ profiling technique was applied, operating at room temperature with a test frequency of 100 kHz and a Hg probe with a contact area of 600 $\mu$m². Fig. 7 shows the Hall mobility ($\mu$), sheet carrier density ($n_s$), and sheet resistance ($R_s$) of PMOCVD-grown InAlN/GaN 2DEG as a function of temperature. It can be seen that the high 2DEG Hall mobility of 1402 $\pm$ 100 cm²/V s was achieved with a high sheet carrier density of (2.01 $\pm$ 0.2) × 10¹³ cm⁻² at room temperature. As temperature decreases, the Hall mobility gradually increases. When the temperature has been lowered to 77 K, the Hall mobility dramatically improves and exceeds 5348 cm²/V s due to the suppression of ionized impurity scattering, which suggests that conductivity is dominated exclusively by the carriers at the AlN/GaN heterointerface; those values are much higher than the data of Gonschorek et al. [14] (3170 cm²/V s at 77 K) and Hiroki et al. [28] (2600 cm²/V s at 77 K). Around 620 K, the Hall mobility decreases to 275 cm²/V s due to the significantly increased polar optical phonon scattering [29]. It can be observed that 2DEG density shows an almost constant value of approximately 2.0 × 10¹³ cm⁻² regardless of the different temperatures.

The structural and electrical characterization data presented above indicate that PMOCVD is a promising approach and may offer some advantages for producing InAlN based transistors in comparison with conventional MOCVD and MBE. We ascribe the
improvements to the following aspects. First, reactive atoms are separately supplied to the reactor at different times. This increases the surface mobility of the adatoms and enables them to find energetically favorable sites. Enhanced migration also lowers the InAlN growth temperature and increases indium incorporation into the InAlN alloy film. Reducing growth temperature could also avoid indium segregation, which improves material composition uniformity and reduces alloy disorder in the InAlN film. As a result, electrical mobility is improved significantly. In addition, we can control composition and thickness by adjusting the pulse time ratio of TMIn to TMAl in a unit cycle and the total number of unit cell repeats.

4. Conclusions

In summary, we have successfully grown nearly lattice matched InAlN/GaN heterostructures by PMOCVD. It can be confirmed that the InAlN barrier layer has a smooth surface morphology and no phase separation. High 2DEG mobilities of 1402 cm$^2$/V s and high 2DEG densities of $2 \times 10^{13}$ cm$^{-2}$ were obtained for InAlN/AlN/GaN heterostructures with the barrier thickness of 14 nm. The composition and thickness of InAlN barrier can be controlled by adjusting the ratio of pulse time of TMIn to TMAl in a unit cycle and the total number of unit cell repeats. PMOCVD can reduce the growth temperature of InAlN, increase the surface mobility of adatoms, consequently avoid indium separation, and improve the film quality. Our results indicate that PMOCVD is an effective way to grow the InAlN ternary alloy for high-frequency and high-power high electron mobility transistors (HEMTs) applications.

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