Influence mechanism of growth temperature and pressure on surface morphology and defects of InGaN materials

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Abstract
We studied the influence of temperature and pressure on the surface morphology and V-defects of the InGaN films. It was found that an appropriate increase in the growth temperature enhanced the mobility of Ga and In atoms, smoothened the surface of the InGaN thin film samples, and improved the growth quality. Simultaneously, increasing the temperature appropriately reduced the surface roughness of the sample and the defect density of the V-defects. It is also found that under the same temperature conditions, a lower pressure weakens the incorporation barrier of atoms, enhances the incorporation efficiency of In atoms, and improves the growth quality of InGaN.

1. Introduction

GaN-based materials have become a hot topic in the current research field of laser diodes (LD) and light-emitting diodes (LED). In the field of laser processing, blue laser welding is more prominent for precision processing. In the field of visible light communication, the LD-related modulation rate is higher, and the data throughput far exceeds that of the traditional data transmission methods [1, 2]. In terms of laser displays, owing to their wide color gamut, uniform brightness, and strong directionality, laser displays based on three primary colors of visible LDs are expected to become the core of next-generation displays [3]. In laser diodes, InGaN can be used as the upper and lower waveguide layers and cladding layers in GaN-based laser LDs, and can also be used as the good layers of the quantum well light-emitting region [4]. From the device-level analysis, the quality of epitaxial growth is the basis of device quality [5]. For the quality analysis of InGaN, we previously studied the influence of the growth rate of InGaN in the active region and the thickness of the cap layer on the carrier injection and luminous efficiency [6]. Studies have shown that there are many factors in the quality analysis of InGaN, such as how to incorporate more In under limited conditions, and how to solve the problem of In-rich clusters formed by different contents of incorporated In also requires more research [7, 8]. At the same time, during the growth of InGaN, due to the lattice mismatch between the sapphire substrate and the GaN template layer, a large number of dislocations are generated at the interface between the substrate and GaN and extend to the InGaN layer, resulting in a high defect density of InGaN [9]. In addition, studies have also shown that the growth of InGaN causes the rate of the (10–11) plane to be different from that of the (0001) plane, resulting in the formation of V-defects [10, 11]. Because the content of In affects the analysis of the material quality, we ensured that the content of In was approximately the same in the experiment. Based on this, we studied the influence of different pressure and temperature conditions on the surface morphology of the material, and simultaneously, the V-defects on the surface, from the perspective of improving the growth quality of InGaN. This study aims to analyze the mechanism of material morphology change from the perspective of growth and improve the growth quality of InGaN.
2. Materials and methods

We grew InGaN epitaxial films on GaN in a tightly coupled spray reactor using a metal-organic chemical vapor deposition (MOCVD) system. Firstly, a layer of GaN was grown on a c-plane sapphire substrate, then a 25 nm GaN buffer layer was grown at 540 °C, and finally, a 1 μm of unintentionally doped GaN was grown at 1100 °C. During the growth of the InGaN layer, trimethylgallium (TEGa), trimethylindium (TMIn), and ammonia gas (NH3) were used as precursors, and N2 was used as the carrier gas at 785, 805 and 810 °C. We studied the effect of different temperature changes on the quality of InGaN materials. The In composition may differ owing to the different indium incorporation efficiencies at different temperatures and pressures. Therefore, in order to make a comparable analysis of the epitaxial growth process, we adjusted the TMIn flow rate to ensure the same indium composition of approximately 3.2% for the studied InGaN samples grown under different temperatures or pressures. The samples were set as A, B, and C, which were grown under three different temperature conditions, and their growth pressures were all 200 Torr. At the same time, sample D was grown at the same growth temperature as sample B, but with a different pressure of 400 Torr.

We performed ω−2θ scans of the samples using an x-ray diffractometer (Rigaku Smart Lab) to determine their structural parameters. Simultaneously, defects on the surface of the samples were observed using scanning electron microscopy (SEM). The surface morphologies of the InGaN films were measured using a Bruker atomic force microscope (AFM). Using a 325 nm He-Cd laser as the excitation light source, the room-temperature photoluminescence (PL) spectra of the samples were measured using a grating spectrometer.

3. Results and discussion

We performed x-ray diffraction (XRD) tests on samples A, B, C, and D. Figure 1 shows the results of the ω−2θ scan of the XRD on the (0002) plane. The sharp peak corresponds to GaN and the peak on the left side corresponds to InGaN. The growth temperature, pressure, flow rate, and In content of the InGaN films under these experimental conditions are listed in table 1. We analyzed the FWHM of each sample by means of Gaussian fitting. The FWHMs of A, B, C, and D are 0.0099, 0.0115, 0.0127, and 0.0120 arcsec, respectively. The indium
content of all samples was between 3.2% ± 0.1%, and their difference in In content was very small, which has only little effect on the XRD measured results.

The AFM scan images of samples A, B and C are shown in figures 2(a)–(c), respectively. The roughness values of the surfaces of samples A, B and C are 0.548, 0.686 and 0.823 nm, respectively. This shows that the mobility of Ga atoms is low because of the lower temperature, so Ga atoms tend to aggregate at the nucleation point, bond with themselves, and incorporate into the lattice. When the growth temperature was increased to 810 °C (sample A), the mobility of Ga atoms was further enhanced, which was more favorable for incorporation into the edge of the step and flattening the surface. The main reason for the smoother surface of our analyzed samples may be the

Table 1. Growth conditions of four InGaN samples A, B, C and D and the composition of In obtained by XRD.

| Samples | Temperature(°C) | Pressure (torr) | In(%) | FWHM (arcsec) |
|---------|----------------|----------------|-------|--------------|
| A       | 810            | 200            | 3.29  | 0.0099       |
| B       | 805            | 200            | 3.25  | 0.0115       |
| C       | 785            | 200            | 3.22  | 0.0127       |
| D       | 805            | 400            | 3.21  | 0.0120       |

Figure 3. SEM scan results (a), (b) and (c) of the local area of the three samples A, B and C near the central area, and the larger magnification of sample C (d). In (d), the morphology of the V-defects can be observed more clearly.

Figure 4. Roughness (solid circle) and defect density (solid square) of samples A, B and C as a function of temperature.
improved surface migration ability of the external atoms at higher temperatures. In addition, the growth temperature must be appropriately reduced, which reduces the mobility of Ga atoms.

Furthermore, we performed scanning electron microscopy (SEM) on samples A, B and C. The microimage results are shown in figures 3(a)–(c). Figure 3(d) shows that the main defects observed on the surface of the sample were V-defects with hexagonal openings.

After counting the calculation, the results of three samples A, B and C under magnifications of 3.0 kv, 7.8 mm and 50 k are more obvious. The densities of samples A, B and C are $3.40 \times 10^8$, $3.52 \times 10^8$ and $5.09 \times 10^8$ cm$^{-2}$. The defect density of sample C was the highest, whereas that of sample A was the smallest. At the same time, the FWHM of sample A is the smallest, further indicating that an increase in the growth temperature is beneficial for reducing the defect density. The curves of roughness and defect density of the samples as a function of temperature are shown in figure 4. We found that an increase in the growth temperature to a certain extent effectively reduces the defect density on the surface of the sample, and the roughness of the sample also decreases to a certain extent.

According to Shiojiri [11], during the growth stage of InGaN, In atoms are trapped and isolated in the strain field of the threading dislocation (TD) core, which hinders the migration of Ga atoms to the TD core and results in a higher growth rate than indium-rich In’s TD core. Small pits were then formed on the TD core. When the growth temperature of InGaN is lowered, the growth rate of the (10–11) plane is lower than that of the (0001) plane, causing the formation of V-defects along the six sloping (10–11) sidewalls. On one hand, increasing the temperature enhances the surface Ga and In atomic mobility, reducing the number of pit cores. However, the (10–11) plane and (0001) plane decrease at higher temperatures, which can suppress craters with (10–11) sidewalls [12, 13]. Sharma [14] proposed that each V-defect has a penetrating edge dislocation at its center, and the defect starts at the core of the edge dislocation in the presence of In. The lower temperature of InGaN/GaN quantum well growth facilitates the formation of V-defects. Therefore, by adjusting the growth conditions, the formation of V-defects could be suppressed by appropriately increasing the temperature.

We further tested the surface topography of the three samples using AFM over a larger area, and the results are shown in figure 5. It indicates that surface morphology deteriorates and the defect densities increases when the growth temperature decreases from 810 °C to 785 °C, which is attributed to the different growth rate between (10–11) and (0001). It is reported that the different growth rate on (10–11) will decrease when growth temperature decreases, and the surface migration of Ga and In atoms will also decrease Therefore, increasing the temperature can effectively reduce the defect density of V-defects to a certain extent.

In the experiment, although the flow rate of TMIn was slightly increased in order to ensure a similar In content of different samples, comparing samples B and D, it was found that the In content of sample D was slightly lower than that of sample B. On the one hand, In will desorb with the increase in temperature [15]; on the other hand, the increase in the pressure of 400 Torr in group D compared to the pressure of 200 Torr in group B will weaken the boundary layer of TMIn passing through the growing film transmission capacity. At lower pressures, the atomic incorporation barrier will be lowered [16], and the sample will shift from 3D to 2D step flow growth [17, 18]. Therefore, the group B samples are grown at a lower growth pressure (200 Torr), which will

Figure 5. Surface morphologies of three samples A, B, C and D measured by AFM (a), (b), (c) and (d).
lead to a higher In incorporation efficiency of this group of samples than that of the D group, while also having better growth quality.

Figure 6 shows the room temperature PL spectra of three InGaN samples, which is measured with 325 nm-laser excitation source. The results show that there are three obvious luminescence peaks in the figure, which are the GaN peak at 370 nm, the InGaN peak at 500–520 nm, and the yellow light peak at 650 nm. The absorption coefficient is $1.2 \times 10^4$ cm$^{-1}$, and the intensity decays to $\frac{1}{e}$ at 120 nm after light entering the material. This band was generated by the transition from the bottom of the conduction band to the bottom of the valence band. There is broad emission around 500 nm–520 nm, whose intensity is the strongest. The corresponding range of this output is approximately 500 nm–520 nm. At the same time, there was a relatively obvious but weak yellow light peak (YL) at 650 nm for all three samples. There are a lot of literatures showing that the yellow light peak at this place is directly related to the complex formed by carbon impurities [18]. When the temperature increases, the carbon impurities decrease, and the peak value of the corresponding peak also decreases to a certain extent.

We performed a Gaussian fitting of the PL spectra at room temperature and calculated that the full width at half maximum (FWHM) of the three samples was 32.529, 33.480 and 34.279 nm, which further indicated that the screw dislocation of sample A was lower, and an appropriate increase in temperature would improve the growth quality of the sample. Compared with A, the PL peak of C exhibits a larger decrease, indicating that the number of nonradiative recombination centers has increased [19]. We believe that this may be due to the following factors: There is a large stress in InGaN owing to the lattice mismatch between InN and GaN, which is beneficial...
for introducing many defects, such as point defects and plane defects. Under the combined action of the thermal and stress fields, new defects were generated, which affected the peak intensity of InGaN.

We further normalized and fitted the GaN peak of the measured PL spectra, as shown in figure 7. With a change in wavelength, the PL luminescence intensity of the GaN peak at room temperature firstly decreased and then increased. When the In content is low (<6%), the valence band order between InGaN and the surface GaN is lower (<3KT), and a part of the photogenerated minority carriers are thermally excited to the surface for recombination [20]. When the temperature was appropriately increased, the In content increased, and the carrier confinement effect was strengthened. This increased the peak value of InGaN. When the In content is high, the luminous intensity decreases with an increase in the composition owing to the decrease in the material quality. Although the InGaN film is thin, localized states still occur in the In-rich region, which will affect the process of radiative recombination.

4. Conclusions

We grew several groups of InGaN thin films by studying the different growth conditions of MOCVD. We performed XRD, AFM, and PL tests at room temperature on these samples to study the effects of growth temperature and pressure on the surface morphology and V-defects of InGaN. The results show that when the growth temperature is low, the surface migration ability of atoms is weak, and the difference in growth rate between the (0001) and (10−11) plane is large, which is not conducive to inhibiting the formation of V-defects. Under the same temperature conditions, when the pressure is lower, it is beneficial to accelerate the desorption of impurity atoms, reduce the incorporation barrier of atoms, promote the growth of samples toward the two-dimensional step direction, and improve the growth quality of the InGaN thin films. We believe that this research will be beneficial for improving the growth quality of InGaN.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Disclosures

The authors declare no conflicts of interest

Ethics statement

Informed consent from patients was not required, and the institutional review board (IRB) review was waived based on institutional policy.

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