Applied Physics Express 4 (2011) 035501
DOI: 10.1143/APEX.4.035501

Effects of Trimethylgallium Flow Rate on *a*-Plane GaN Growth on *r*-Plane Sapphire during One-Sidewall-Seeded Epitaxial Lateral Overgrowth

Hsiao-Chiu Hsu, Yan-Kuin Su[1]*, Shyh-Jer Huang, Ricky W. Chuang, Shin-Hao Cheng[1], and Chiao-Yang Cheng

Institute of Microelectronics, Department of Electrical Engineering, and Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 701, Taiwan, R.O.C.
[1]*Department of Electrical Engineering, Kun Shan University of Technology, Tainan 710, Taiwan, R.O.C.*

Received December 30, 2010; accepted January 25, 2011; published online February 15, 2011

The high crystalline quality of *a*-plane GaN growth on *r*-plane sapphire has been demonstrated successfully by using one-sidewall-seeded epitaxial lateral overgrowth (OSELOG). The dislocation density of OSELOG-grown GaN film is 3–4 orders of magnitude lower than that of the as-grown film and the coalescence thickness of OSELOG-grown GaN is less than 5 µm. Low temperature cathodoluminescence (CL) shows that the optimum trimethylgallium (TMGa) flow rate during OSELOG plays a significant role in enhancing the crystalline quality of *a*-plane GaN. The crystalline quality of *a*-plane GaN can be effectively improved using OSELOG compared with the other ELOG approaches.

Nonpolar *a*-plane GaN grown on r-sapphire by metal organic chemical vapor deposition (MOCVD) contains a high threading-dislocation density (TDD) of $\sim 10^{10}$ cm^{-2} and a basal-plane stacking-fault (BSF) density of $\sim 10^{5}$ cm^{-1}, due to the planar anisotropic nature of its growth and the large difference in lattice constant between the GaN film and sapphire substrate. Various research groups have devised different methods to overcome the problem, such as epitaxial lateral overgrowth (ELOG) or other modified ELOG techniques, which have been developed to minimize dislocation density.[1,2] Iida *et al.*[3] have also reported that the one-sidewall-seeded ELOG (OSELOG) could reduce the dislocation density effectively. The advantage of adopting the OSELOG approach can be derived from the high quality *a*-plane GaN films regrown via one-sidewall of an as-grown GaN seed by tuning the growth-rate ratio of the Ga-face to N-face of GaN films. As a result, the new dislocation is prohibited from propagating away from the coalescence area between the wing and window regions when compared with the conventional ELOG approach. This ELOG technique requires more than 10 µm to obtain the full coalescence film thickness with a better coalescence surface, which constitutes a difficulty in layer uniformity control. In this work, a modified OSELOG approach is introduced for improvement in *a*-plane GaN crystalline quality (TDD $\sim 10^{6}$ cm^{-2}) and reduction of full coalescence thickness (<5 µm). We have observed earlier that the growth rate of the full coalescence step is considered essential for OSELOG GaN film because it influences the strain relation and defect distribution in the regrown film. So far, there is little evidence to substantiate this finding.

Before the onset the of OESLOG process, the stripe pattern of a GaN seed on *r*-plane sapphire was fabricated. The OSELOG approach can be further divided into two steps involving the regrowth of GaN from one-sidewall and the lateral growth mode enhancement for coalescing the GaN film on a SiO_2 mask. In the first step, a seeded GaN layer was grown with a high V/III ratio of 1000. Then, the V/III ratio was subsequently changed to 220 during the GaN growth to obtain fully coalesced GaN on the SiO_2 mask. In fact, reducing the V/III ratio to 220 propelled an increase in the growth rate of the Ga-face sidewall, which resulted in the growth rates of the Ga-face sidewall and the N-face sidewall coming be nearly equal to one another.[4] At the second step, these samples were grown under the same V/III ratio but with TMGa flow rate varied during the coalesced step. The trimethylgallium (TMGa) flow of samples was varied with rates of 15, 30, and 45 sccm. The film surface was fully coalesced and the thickness of these samples was kept identical in order to evaluate the sample crystal quality as a result of varying the TMGa precursor flow rate. For comparison, an *a*-plane GaN film with a similar thickness was also grown and is henceforth denoted as the as-grown GaN. Figure 1 shows cross-sectional scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images of the regrown GaN film after completing the first step, where two regions are divided and shown as a wing region (2 µm) and a window region (1.6 µm), representing the as-grown GaN seed layer and the regrown GaN film, respectively. The cross-sectional SEM image of the grown GaN layer from one sidewall is shown in Fig. 1(a), where the regrowth of GaN film starts only from the N-face sidewall when the V/III rato is maintained at 1000. It also indicates that the growth rate of GaN film regrown from the Ga-face sidewall is extremely low compared to GaN film regrown from the N-face sidewall. This result seemingly agrees with Iida *et al.*'s study,[4] demonstrating that the growth rate of GaN film regrown from the Ga- and N-face sidewall could be controlled by changing the V/III ratio. Figure 1(b) shows the cross-sectional TEM imaging of the regrown GaN. It was observed

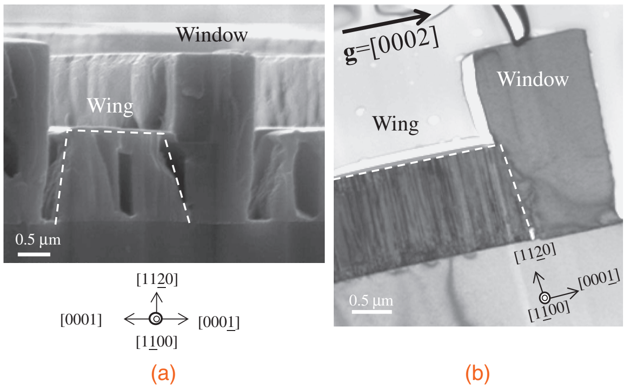

Fig. 1. Cross-sectional SEM image and TEM image of *a*-plane GaN after first step of GaN growth with V/III growth rate ratio of 1000.

*E-mail address: yksu@mail.ncku.edu.tw

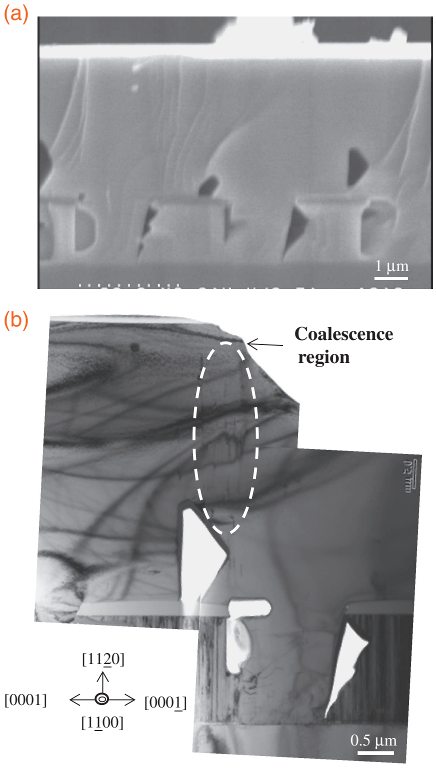

Fig. 2. Cross-sectional SEM image and TEM image of fully coalescence *a*-plane GaN after second step with V/III growth rate ratio of 220.

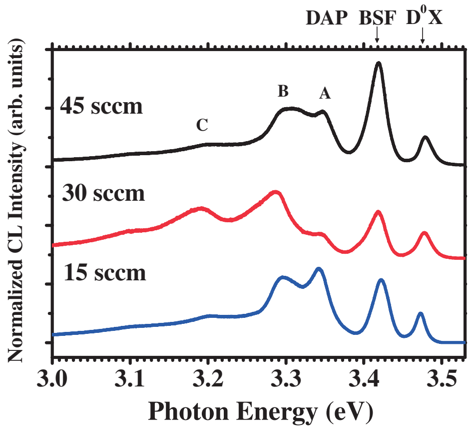

Fig. 3. Low-temperature CL spectra of OSLEOG samples with varied TMGa flow rates. A, B, and C are the multi peak emissions of DAP recombination.

that the dislocation density in the window region was 3–4 orders of magnitude lower than that in the wing region. It was also shown that with the OSELOG approach the high-quality GaN film regrown from one sidewall could be obtained with a significant reduction in the dislocation density compared to other approaches.

Figure 2 shows the cross-sectional SEM and cross-sectional TEM images of the regrowth GaN film after full coalescence by using the OSELOG technology with stripes patterned before the re-growth. Figure 2(a) shows the cross-sectional SEM image of the regrown GaN film. The coalescence thickness is 3.9 µm on the SiO_2 mask and the overall thickness is 5.5 µm if the as-grown GaN seed layer is also taken into account. The thickness was observed to be less than that reported earlier.[3)] The optimum growth conditions and narrower stripes contributed to the reduction in overall thickness. The result indicated that the modified OSELOG approach is able to reduce the process time and improve the yield when compared with the conventional ELOG technology. Figure 2(b) shows cross-sectional bright-field TEM image of the OSELOG GaN films after total coalescence, which can be used for measuring the dislocation distribution of the re-grown GaN films. It was found that there were dislocations in the regrown GaN film. It was proposed earlier that the SiO_2 mask could block the dislocation propagating from as-grown film to re-grown GaN film. New dislocations were formed at the interface when the Ga-face and N-face GaN were coalesced together. The air void adjacent to the SiO_2 mask was formed due to the difference in shape of the Ga- and N-faces of the GaN film. Generally, the Ga-face sidewall was formed in a arrowhead-like shape with inclined [11$\bar{2}$*n*] facets,[5)] while some voids formed in and near the wing region of GaN seed layer; the very phenomena observed was in fact due to various growth conditions that affect the outcome of the coalescence step, including high temperature, low V/III ratio, and high hydrogen gas feeding. It is known that GaN is capable of being decomposed to Ga and NH_3 when reacting with hydrogen gas at elevated temperatures, which results in etch of GaN film.[6)] The voids thus created afterward would have no impact on the crystalline quality, but rather their existence may prove beneficial for the strain relaxation in GaN film as the temperature is reduced.

To improve the crystalline quality of OSELOG GaN film, the growth rate was investigated by varying the TMGa flow rate during the growth process. Note that the NH_3 precursor flow rate was also varied in order to keep the same V/III ratio at this step. The full width at half maxima (FWHM) of different omega (ω) scans along the *c*-axis ([0001])/*m*-axis ([1$\bar{1}$00]) were measured by high-resolution X-ray diffraction (HR-XRD). The FWHMs measured along the *c*-axis of these OSELOG samples with different TMGa flow rates (15, 30, and 45 sccm) were 0.379, 0.141, and 0.387°, respectively. The FWHMs measured along the m-axis of these OSELOG samples with different TMGa flow rates (15, 30, and 45 sccm) were 0.301, 0.110, and 0.199, respectively. This indicated that the crystalline quality could be vastly improved by optimizing the coalescence rate when the TMGa flow was maintained at 30 sccm. Compared with the as-grown sample, a significant reduction of FWHM was observed as the scans were performed along the *m*-axis (0.527°) and the *c*-axis (0.398°).

Low-temperature (11 K) cathodoluminescence (CL) spectra of the OSELOG-grown sample surfaces with various TMGa flow rates are shown in Fig. 3. The CL spectra in Fig. 3 were taken over a relatively large area ($20 \times 20\,\mu m^2$). The main advantage in using CL to evaluate the defect distribution of a large area nonpolar GaN sample is an easy assessment of the defect distribution. Figure 3 shows the typically observed radiative emission bands and the related peak energies of GaN attributed to the donor-bound exciton (D^0X, 3.472 eV), BSF emissions (3.417 eV), and donor–acceptor pair (DAP) recombination (DAP, 3.345–2.8 eV).[7)] The BSF emission transition arises from the recombination of excitons bound (D^0X) to the so-called I_1 BSF.[8)] When the TMGa flow rates is decreased from 45 to 30 sccm and then down to 15 sccm, the corresponding D^0X peak energies of the OSELOG grown samples are 3.479, 3.477, and 3.473 eV, respectively. This result indicates that the strain in the GaN film seems to be reduced with a lower lateral growth rate when the D^0X peak is close to the strain-free value of 3.472 eV as the TMGa flow rate is decreased.[9)] Strain in the GaN film plays an essential role in the device behavior as it may affect the dislocation distribution in the epilayer, which in turn influences the optical and electrical performance in the end. This result shows that by tuning the TMGa flow rate, the strain in the GaN epilayer can be controlled during the growth. The high number of BSFs is the main cause for the suppression of the D^0X emission.[8)] Therefore, the ratio of BSF/D^0X emission intensity can be used to verify the crystalline quality of the OSELOG grown samples. As calculated, the BSF/D^0X ratios of these samples corresponding to different TMGa flow rates of 15, 30, and 45 sccm are 2.15, 1.74, and 3.54, respectively. The result shows that the intensity of BSFs is strongest when the TMGa flow rate is maintained at 45 sccm during the lateral growth, which renders the strong stress relaxation in epilayer and an enhancement in the formation of BSFs. The BSF/D^0X ratio of the OSELOG sample with a TMGa flow of 15 sccm is higher than that with a TMGa flow of 30 sccm. It is interesting to observe that the crystalline quality cannot be enhanced by reducing the lateral growth rate with the lowering of the TMGa flow rate. To investigate the possible mechanism dominating the crystalline quality during the growth, we focused on the DAP emission of these samples, as well as the D^0X and BSF peaks. The emission of DAP recombination in Fig. 3 shows a multipeaked curve, which are located at 3.345, 3.29, and 3.18 eV and depicted as “A”, “B”, and “C”, respectively. In the ELOG process, the impurity incorporation in the regrown GaN film is closely affected by the presence of the SiO_2 mask, which has a peculiar tendency of being decomposed to silicon or oxygen donor impurities at high temperature.[10)] In previous studies, the peak emission at 3.345 eV was reported represent the DAP transition from impurity incorporation on the N-terminated face before full coalescence.[11,12)] Generally speaking, the impurities are easily incorporated on the highly reactive N-terminated face as opposed to the more stable Ga-terminated face.[13)] Figure 3 shows that the impurity-related DAP intensity of the sample with a TMGa flow rate of 15 sccm is highest compared to other OSELOG grown conditions, indicating that there are more impurities incorporated in the OSELOG grown GaN film with a slower growth rate. For the defect observation of *a*-plane GaN, those imperfections act as nucleation sites for BSFs, which eventually terminate at a prismatic stacking fault (PSF).[8,13)] Furthermore, Häberlen *et al.* suggested that the peak at 3.29 eV in the DAP band, was contributed by the zero phonon line (ZPL) of the DAP band and PSF emission, which resulted in the line pulling effect on the individual peaks.[7)] This effect could generate the individual peaks associated with the DAP band which are not separated by a constant energy (91 meV) but rather by various energies.[10)]

The energy separations (Peaks A–B) of the DAP transition for the OSELOG grown samples are 51, 59, and 45 meV with the TMGa flow rates of 15, 30, and 45 sccm, respectively. It seems that the extent of the line pulling effect is related to the defect distribution in the OSELOG grown sample. The possible emission energy is contributed by different defect types such as impurity incorporation or strain relaxation. Furthermore, the observation of the peak at 3.18 eV in DAP emission is considered as the 1LO replica of the ZPL, which resembles the near band edge (NBE) emission but with less signal and more noise.[7)] It was also observed that the OSELOG grown sample with a TMGa flow rate of 30 sccm has a stronger DAP emission signal at 3.18 eV compared to other samples obtained with 15 and 45 sccm growth conditions. This provides strong evidence demonstrating the sample with a TMGa flow rate of 30 sccm has stronger NBE emission with excitonic recombination, vividly indicating that a better crystalline quality is obtained as a result of the well-controlled coalescence rate. Based on the observation from the NBE and DAP spectra, applying different growth rates evidently would make a pronounced effect on the stress and impurity incorporation in the GaN films during the OSELOG process, which in turn modify the crystalline quality and the dislocation distributions.

In conclusion, this study demonstrates an effective method to improve the crystalline quality of *a*-plane GaN film by using the OSELOG approach. The results conclude that the growth rate plays an important role during the coalescence step. All the above results demonstrate that OSELOG can serve as an effective and high throughput approach for the enhancement in the crystalline quality of *a*-plane GaN.

Acknowledgments The authors would like to thank the National Science Council, the Bureau of Energy, Ministry of Economic Affairs, Taiwan, for the financial support under Contract No. 98-D0204-6, TDPA program Nos. 97-EC-17-A-07-S1-105, NSC 97-2623-E-168-001-IT, and the assistance in film characterization provided by the LED Lighting and Research Center at National Cheng Kung University is also appreciated.

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