Laser-Assisted Metal–Organic Chemical Vapor Deposition of Gallium Nitride

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Ammonia (NH$_3$) is commonly used as group-V precursor in gallium nitride (GaN) metal–organic chemical vapor deposition (MOCVD). The high background carbon (C) impurity in MOCVD GaN is related to the low decomposition efficiency of NH$_3$, which represents one of the fundamental challenges hindering the development of high-purity thick GaN for vertical high-power device applications. This work uses a laser-assisted MOCVD (LA-MOCVD) growth technique to address the high-C issue in MOCVD GaN. A carbon dioxide (CO$_2$) laser with a wavelength of 9.219 μm is utilized to facilitate NH$_3$ decomposition via resonant vibrational excitation. The LA-MOCVD GaN growth rate (GR, as high as 10 μm h$^{-1}$) shows a strong linear relationship with the trimethylgallium (TMGa) flow rate, indicating high effective V/III ratios and, hence, efficient NH$_3$ decomposition. [C] in LA-MOCVD GaN films decreases monotonically, as the laser power increases. A low [C] at 5.5 × 10$^{15}$ cm$^{-3}$ is achieved in the LA-MOCVD GaN film grown with GR of 4 μm h$^{-1}$. LA-MOCVD GaN films reveal high crystalline quality with room-temperature mobility of >1000 cm$^2$ V$^{-1}$ s$^{-1}$. LA-MOCVD provides an enabling route to achieve high-quality GaN epitaxy with low-C and fast GR simultaneously. This technique can be extended for epitaxy of other nitride-based semiconductors.

1. Introduction

Gallium nitride (GaN) has played an important role in optoelectronics, photonics, and radio-frequency (RF) electronic devices due to its wide direct bandgap (3.4 eV), high electron mobility (>1000 cm$^2$ V$^{-1}$ s$^{-1}$), high critical electric field (3.4 MV cm$^{-1}$), and high thermal stability. As a promising candidate for power electronic applications, the Baliga’s figure of merit (BFOM) of GaN is more than 500× and 3× higher than that of silicon (Si) and silicon carbide (SiC). With the availability of bulk GaN substrates, vertical GaN PN diodes with a breakdown voltage ($V_{BR}$) of 5 kV have been demonstrated.$^{[1]}$ To achieve vertical power devices with higher breakdown voltage, fundamental challenges in GaN epitaxy with low controllable doping, thick epilayer, and high mobility still need to be addressed. GaN thin films grown by metal–organic chemical vapor deposition (MOCVD) typically have a growth rate (GR) of 2–3 μm h$^{-1}$ with $N_d$–$N_a$ at low-10$^{16}$ to mid-10$^{15}$ cm$^{-3}$. Therefore, a high-quality GaN epifilm that can enable vertical GaN power devices with $V_{BR}$ > 10 kV is still lacking.

Trimethylgallium (TMGa) and ammonia (NH$_3$) are commonly used as the group-III and group-V precursors for MOCVD GaN growth. At the typical MOCVD growth temperature of GaN ($\approx$1000 °C), the decomposition efficiency of NH$_3$ is only at $\approx$1%.$^{[2,4-6]}$ High thermal stability of NH$_3$ leads to limited decomposition products for surface reactions. Therefore, the input V/III ratio during MOCVD GaN epitaxy is normally above 1000 to ensure sufficient N species reaching on the growth surface.

It has been suggested that carbon (C) impurity originating from metal–organic (MO) precursors is incorporated preferentially on nitrogen sites (C$_a$) in n-type GaN, giving rise to a deep acceptor level, which is responsible for compensating the n-type doping.$^{[7,8]}$ Carbon compensation in n-type GaN drift layer-based power devices can cause high leakage current and high on-resistance,$^{[9]}$ impeding device performance. Thus, suppression C incorporation in MOCVD n-GaN epitaxy is a key research goal for high power device applications.

Carbon incorporation in MOCVD growth of III nitrides is significantly affected by the stoichiometric balance, which relies on the effective V/III ratio on the growth surface. The input V/III ratio is determined by the molar flow of group-V and group-III precursors, whereas the effective V/III ratio refers to the active group-V and group-III species participating during the growth process. Theoretical investigations suggested that the high effective V/III ratio is an effective approach to reduce C incorporation.$^{[10-11]}$ Conventional approaches to suppress C incorporation in MOCVD GaN growth include: 1) increasing input V/III ratio via increasing NH$_3$ flow or decreasing TMGa flow$^{[2,7,9,12-15]}$; 2) increasing growth pressure$^{[2,9,12,13]}$; and...
increasing growth temperature.\cite{12,15} However, these approaches often lead to GaN growth with lower GRs, increased gas-phase reaction, and increased thermal decomposition. Fundamentally, the low decomposition efficiency of NH₃ in MOCVD GaN growth represents a key factor that leads to relatively high-C incorporation.

Laser CVD (LCVD) is a method that introduces laser as an additional energy source during the CVD process. There are two types of LCVD: 1) pyrolytic, in which a laser beam interacts with the substrate to create hot spot locally or globally for thermally assisted chemical reactions; and 2) photolytic, in which a laser beam interacts with the chemical reactants to break the chemical bonds and, thus, facilitate the growth.\cite{16} LCVD has been utilized to deposit various compound semiconductors, including selective-area deposition of III–V films\cite{17} and low-temperature MOCVD growth of GaN\cite{18} and AlN.\cite{19} Previously, carbon dioxide (CO₂) laser was also utilized in GaN MOCVD growth.\cite{20–22} At the resonant wavelength of 9.219 μm, strong emissions from NH₂ radicals were observed in the optical emission spectra (OES) from the resonantly excited NH₃ flows, together with other emission peaks from NH, N, N⁺, and H, which exhibited an effective dissociation of NH₃ molecules with CO₂ laser irradiation.\cite{22} It demonstrated GaN growth on (0001) sapphire substrate at a very low temperature of 250 °C in a custom-designed system. Fast GaN GRs of 12\cite{21} and 84 μm h⁻¹\cite{22} were demonstrated using CO₂ laser at the growth temperature of 600 and 750 °C, respectively. However, characterization of these materials indicates relatively low crystalline quality.\cite{21,22}

In this work, high-quality GaN thin films using a CO₂ laser-assisted MOCVD (LA-MOCVD) growth technique were demonstrated. The effects of the laser-assisted NH₃ decomposition on the GaN growth, impurity incorporation, and electron transport properties of LA-MOCVD GaN were systematically studied. Experimental results indicate that the effective V/III ratio at the growth surface can be significantly enhanced via CO₂ laser excitation in LA-MOCVD. C impurity incorporation can be reduced by more than 60% in LA-MOCVD GaN. GaN epifilm with low C (5.5 × 10¹⁵ cm⁻³) was achieved with a low input V/III ratio of 875 and a GR of 4 μm h⁻¹. LA-MOCVD GaN films show superior transport properties with the room-temperature mobilities over 1000 cm² V⁻¹ s⁻¹ for films grown on both GaN-on-sapphire templates and free-standing GaN substrates with the GR of 4.5 μm h⁻¹. The LA-MOCVD growth technique provides a new route to achieve high-quality GaN epifilms with low impurities and fast GRs promising for high power device applications.

2. Results and Discussion

The schematic of the LA-MOCVD system is shown in Figure 1. A PRC 1500 W tunable CO₂ laser was used as the laser source. A CO₂ laser beam is generated and introduced into the vertical rotating-disk MOCVD reactor (Agnitron Technology Inc., Agilis MOCVD R&D System). The laser beam is parallel to the wafer surface and susceptor. The width and the height of the laser beam are 14 and 4 mm, respectively. A beam dump is placed at the front viewport to absorb the residual laser power. The gas species were distributed via the high-speed rotation (1200 rpm). Note that the comparisons made between regular MOCVD and LA-MOCVD in this study were from the same areas on the wafers.

2.1. Interaction between CO₂ Laser and NH₃

CO₂ laser power absorption under different controlled chamber conditions was investigated. Figure 2a shows the laser power absorption as a function of the laser wavelength under a typical reactor condition for GaN growth (965 °C, 200 Torr, 4 standard liter per minute (slm) NH₃ flow, and 4 slm H₂ flow). The input laser power was fixed at 200 W. The most prominent absorption was observed at 9.219 μm (1084.71 cm⁻¹) where ~100% of the input laser power was absorbed. This wavelength matches with the rotational–vibrational transition of the N–H wagging mode (ν₃) of the NH₃ molecules at 1084.63 cm⁻¹.\cite{16,17,19} Therefore, 9.219 μm laser wavelength was selected as the operating wavelength in the following investigations. Figure 2b plots the laser power absorption (%) as a function of the NH₃ flow rate under a chamber pressure of 200 Torr, with a fixed laser wavelength at 9.219 μm and a laser power of 200 W. The laser power absorption increases, as the NH₃ flow rate increases with a quick saturation at an NH₃ flow rate of 0.75 slm. A further increase in NH₃ flow rate leads to a full absorption of the incident laser power. Our study also reveals a strong dependence of laser power absorption on the chamber pressure. As shown in Figure 2c, with a fixed NH₃ flow rate of 4 slm, the laser absorption was measured as a function of input laser power under different chamber pressures at 75, 100, 150, and 200 Torr. For the case of 75 Torr, the laser beam was fully absorbed when the input laser power was below 150 W. As the input laser power further increases beyond this threshold, the absorption % shows a gradual decrease. The similar trend was observed for all cases with different chamber pressures, but the input laser power threshold increases, as the chamber pressure increases. With a chamber pressure of 200 Torr, the input laser power threshold was measured at 350 W. From Figure 2d, the threshold input laser power shows a linear relationship with the chamber pressure. According to the ideal gas law: \( PV = nRT \), with fixed temperature (T), reactor volume (V), and gas composition,
the amount of NH₃ molecules (n) is proportional to the chamber pressure (P). Therefore, the threshold input power is proportional to the amount of NH₃ molecules along the beam pathway.

From the above-mentioned studies, CO₂ laser beam at a wavelength of 9.219 μm has a strong interaction with NH₃ molecules. The laser power absorption is strongly dependent on NH₃ flow rate and chamber pressure. The following sections discuss the GaN growth via the LA-MOCVD method and the comprehensive material characterization.

2.2. Growth of LA-MOCVD GaN

Typical MOCVD GaN growth is performed under N-rich conditions with the input V/III ratio up to several thousand. Thus, the GaN GR is limited by the mass transport of Ga species, i.e., the TMGa flow, and, therefore, has a linear relationship with the TMGa flow rate.¹⁴ As the TMGa flow rate increases, the effective V/III ratio decreases, which can lead to nonlinear relationship between the GR and the TMGa flow rate due to the limited N species. Figure 3 shows the GaN GR as a function of the TMGa flow rate under different input laser power: 0 (without laser), 150, and 250 W. The chamber pressure was fixed at 150 Torr, and NH₃ flow was kept at 4 slm. For the case without laser excitation (0 W), the GaN GR shows a trend of saturation, as the TMGa flow rate increases (>0.4 mmol min⁻¹), indicating the reduction of the effective V/III ratio. For the cases with laser excitation (150 and 250 W), the GaN GR increases linearly, as the TMGa flow rate increases, indicating that the GaN growths still fall under N-rich conditions. Thus, the introduction of laser beam during GaN MOCVD growth can effectively enhance NH₃ decomposition efficiency. Note that there is a slight reduction of the LA-MOCVD GaN GRs as compared with those of the conventional GaN growth (0 W). This could be due to the increased parasitic gas phase reactions facilitated by N radicals (NH₂), which form adducts and particles, and consume the
precursor supply to the growth surface. Nevertheless, the LA-MOCVD growth technique is a viable approach to achieve fast GR MOCVD GaN with relatively high effective V/III ratio.

Surface morphologies of two GaN samples grown on GaN on sapphires templates without and with laser excitation (250 W) were compared. The MOCVD growth condition including chamber pressure (150 Torr), input V/III ratio (415), and growth temperature (1025 °C) was kept the same for both samples. The corresponding GaN GRs were 9.3 and 8.5 μm h⁻¹ for the processes without and with laser, respectively. The total GaN film thickness was ≈2.8 μm for both samples. Figure 4a,b compares the macroscopic surface morphology of these two samples via optical imaging. Similar stripe morphologies were observed on both samples, indicating minimum impact of the laser excitation on the macroscopic surface morphology. Figure 4c,d compares the scanning electron microscopy (SEM) images taken from the same two samples. Hexagonal-shaped V-pits were observed on the GaN sample grown without the laser (Figure 4c). V-pits are commonly observed in N-deficient MOCVD GaN growth, indicating a low effective V/III ratio during the epitaxy. In contrast, the LA-MOCVD-grown GaN sample shows a smooth surface without observable V-pits, indicating a high-effective V/III ratio due to efficient NH₃ decomposition from the laser excitation. Figure 4e,f shows the comparison of the 10 × 10 μm² atomic force microscopy (AFM) imaging between the two GaN films. Both films were grown under step growth mode with the low root mean square (RMS) values of 0.606 nm (without laser) and 0.581 nm (with laser). From the investigation of the GaN surface morphologies, it is further confirmed that the LA-MOCVD provides more efficient NH₃ decomposition with the laser excitation, which facilitates GaN growth with higher V/III ratio.

2.3. Impurity Incorporation

Analytical models based on the Langmuir isotherm have been utilized to study the surface reconstructions and carbon coverages in GaN MOCVD under different growth conditions. It was found that the surface carbon coverage is positively related to the carbon incorporation in GaN. In this work, a theoretical
Langmuir adsorption model is utilized to investigate the effect of laser-assisted NH₃ decomposition on the GaN growth surface carbon coverage.

At equilibrium, the fractional coverage \( \theta \) of an adsorbate can be expressed as \[^2\]

\[
\theta = \frac{\exp \left( \frac{\mu_{\text{gas}} - \mu_{\text{gas}}^0}{kT} \right)}{1 + \exp \left( \frac{\mu_{\text{gas}} - \mu_{\text{gas}}^0}{kT} \right)} \tag{1}
\]

where \( \mu_{\text{gas}} \) is the chemical potential of gas species, and \( \epsilon \) is the absorption energy. For a multispecies absorption process, the fractional coverage of the \( i \)th adsorbate can be rewritten as

\[
\theta_i = \frac{\sum_j p_j \exp \left( \frac{\mu_{\text{gas}}^0 - \mu_{\text{gas}}^0_j}{kT} \right)}{1 + \sum_j p_j \exp \left( \frac{\mu_{\text{gas}}^0 - \mu_{\text{gas}}^0_j}{kT} \right)} \tag{2}
\]

where \( p_i \) represents the partial pressure of the \( i \)th gas species, and \( p^0 \) represents the reference (atmosphere) pressure. To qualitatively illustrate how the laser-assisted NH₃ decomposition influences the carbon incorporation, key absorption processes were considered in this model. There are two major reaction pathways in GaN MOCVD: the decomposition route and the adduct route. The decomposition route consists of a series of reactions starting from the decomposition of TMGa to (CH₃)₂Ga (DMGa) by removing a methyl group. Similarly, under high temperature, DMGa decomposes into CH₃Ga (MMGa) and then Ga, respectively. The formation of adducts between TMGa and NH₃ is another possible chemical pathway in which TMGa and NH₃ form TMGa:NH₃ adducts at low temperature and decompose by removing CH₄. It is suggested that, considering both reaction pathways, Ga, MMGa, and CH₃ are major reactive Ga and C species near the high-temperature growth surface.\[^2\] Therefore, the absorption processes of Ga, MMGa, and CH₃ are considered in this model. Three N-related processes (the absorption of NH₃, NH₂, and NH) are considered. Regarding C absorption, MMGa and CH₃ were suggested as the main sources.\[^2\] which were considered in this model. The absorption processes and the corresponding absorption energies cited from prior density-functional theory (DFT) reports are listed in Table 1. It is worth noting that the simplified model in this work intends to qualitatively study the impact of the NH₃ decomposition on C incorporation in GaN MOCVD.

Assuming that the reactive NH₃ decomposition products (NH₂ and NH) are from the direct decomposition of NH₃, the two-step decomposition processes are considered: 1) First-step decomposition: \( \text{NH}_3(g) \rightleftharpoons \text{NH}_2(g) + \text{H}(g) \); and 2) Second-step decomposition: \( \text{NH}_2(g) \rightleftharpoons \text{NH}(g) + \text{H}(g) \). Figure 5 plots the carbon coverage as a function of NH₃ decomposition percentage. Typical MOCVD GaN growth condition (growth temperature of 1050 °C, growth pressure of 200 Torr, and input V/III ratio of 1000) was used in this model. The chemical potentials of gas-phase species were taken from the previous study.\[^3\] Here, \( a_0 \) and \( a_1 \) represent the equilibrium conversion percentage of each reaction. For example, \( a_1 = 1\% \) and \( a_2 = 1\% \) mean that 1% of NH₃ decomposes into NH₂, and 1% of the produced NH₂ decomposes into NH. Figure 5 shows that surface C coverage can be greatly influenced by NH₃ decomposition. As \( a_1 \) and \( a_2 \) increase from 1% (typical for NH₃ decomposition at \( \approx 1000\) °C) to 100%, the C coverage reduces by more than three orders of magnitude. Note that NH₃ and NH have higher chemical potentials than NH₂, suggesting that they are more chemically active. Furthermore, the larger adsorption energies of NH₂ and NH as compared with NH₃ enable higher adsorption on the growth surface. Both theoretical\[^4\] and experimental\[^5\] studies show that C prefers to occupy N sites and form a deep acceptor state (C₃N). As C and N adatoms compete to occupy the same sites on the growth surface, more absorption of NH₂ and NH results in less C incorporation. Moreover, the H radicals generated from the NH₃ decomposition could also react with hydrocarbon radicals (CH₄) to form chemically stable CH₄, thus reducing the C incorporation. Although complex gas-phase and surface reactions can influence the absolute C incorporation in the GaN MOCVD growth process, this simplified analytical model indicates that enhancing NH₃ decomposition is a promising route to suppress C incorporation in GaN MOCVD.

A series of GaN sublayers were grown under various growth conditions for quantitative secondary ion mass spectroscopy (SIMS) characterization to probe C impurity concentration. The detailed growth conditions are listed in Table 2, which

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Table 1. N-, Ga-, and C-related absorption processes and their corresponding absorption energies calculated by DFT from the literature.

| Absorption process | Absorption energy (eV) | Reference |
|--------------------|------------------------|-----------|
| NH₃ (gas) \( \rightleftharpoons \) NH₃ (ad, T1) | 1.17 | [46] |
| NH₂ (gas) \( \rightleftharpoons \) NH₂ (ad, Br) | 3.97 | |
| NH (gas) \( \rightleftharpoons \) NH (ad, H3) | 6.43 | |
| Ga (gas) \( \rightleftharpoons \) Ga (ad, T1) | 3.77 | [47] |
| Ga (gas) \( \rightleftharpoons \) Ga (ad, T4) | 2.48 | |
| Ga (gas) \( \rightleftharpoons \) Ga (ad, H3) | 3.49 | |
| MMGa (gas) \( \rightleftharpoons \) MMGa (ad, T1) | 2.83 | [48] |
| CH₃ (gas) \( \rightleftharpoons \) CH₃ (ad) | 5.07 | [29] |

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Figure 5. Contour of surface carbon coverage as a function of the first and second steps of NH₃ decomposition obtained from theoretical modeling: 1050 °C temperature, 200 Torr pressure, and 1000 input V/III ratio were selected as the modeling condition.
are categorized into two groups: Group I (growth pressure: 150 Torr) and Group II (growth pressure: 200 Torr). Among each group, the input laser power was varied to investigate the impact of laser power on C incorporation and GaN GR.

Figure 6 plots the C concentration and GaN GR as a function of the laser power. Cases from Group I show that $[C]$ in GaN layers decreases as the laser power increases. $[C]$ was reduced from $2.5 \times 10^{16} \text{cm}^{-3}$ (0 W) to $1.2 \times 10^{16} \text{cm}^{-3}$ (250 W). The GaN GR among Group I reduced from 5.2 $\mu\text{m h}^{-1}$ (0 W) to 4.6 $\mu\text{m h}^{-1}$ (150 W) and 4.4 $\mu\text{m h}^{-1}$ (250 W). The slight reduction in GR is due to enhanced parasitic gas phase reaction as discussed earlier.

A more prominent $[C]$ reduction in LA-MOCVD GaN growth was observed from cases in Group II, in which the growth pressure was set as 200 Torr. The $[C]$ was reduced from $1.4 \times 10^{16} \text{cm}^{-3}$ (0 W) to $5.5 \times 10^{15} \text{cm}^{-3}$ (200 W), whereas the GR decreased from 5.2 $\mu\text{m h}^{-1}$ (0 W) to 4 $\mu\text{m h}^{-1}$ (200 W). This group of data directly demonstrates the effective reduction of $[C]$ in LA-MOCVD GaN growth due to enhanced NH$_3$ decomposition efficiency.

Figure 7a summarizes the $[C]$ in LA-MOCVD GaN from this work as compared with the representative $[C]$ in reported MOCVD GaN growth as a function of the input V/III ratio. The dashed line indicates the slope of $[C]$ versus input V/III ratio among the best reported data, considering that $[C]$ is reversely proportional to the input V/III ratio. One can observe that significantly high V/III ratios are required to achieve $[C] < 10^{16} \text{cm}^{-3}$. Using the LA-MOCVD growth technique, the achieved $[C]$ at $\approx 5.5 \times 10^{15} \text{cm}^{-3}$ with a V/III ratio at 875 represents the lowest value as compared with those grown at the similar V/III ratios.

In addition to the V/III ratio, the absolute flow rate of TMGa or NH$_3$ can significantly impact the background $[C]$ incorporation as well. This factor can be considered from the GaN film GR, considering that $[C]$ is positively related to the GaN GR.[14,32] Therefore, $[C]$ is plotted as a function of the input V/III ratio/GR from this work as compared with the data reported from the literature, as shown in Figure 7b. The dashed line indicates the slope of $[C]$ versus input V/III ratio/GR for the best reported data, considering that $[C]$ is reversely proportional to V/III ratio and 1/GR. It shows the trade-off between $[C]$ and GR as well as the V/III ratio. In conventional MOCVD GaN growth, to achieve low $[C]$ impurity, low GR and high V/III ratio are necessary. In contrast, LA-MOCVD GaN growth enables low $[C]$ with fast GR. With the increase in laser power, the LA-MOCVD showed significant $[C]$ reduction without a notable increase in input V/III ratio/GR. This is due to the high effective V/III ratio at the growth surface through laser-assisted NH$_3$ decomposition. As compared with previous reports, the LA-MOCVD GaN growth achieved low $[C]$ at mid-$10^{15} \text{cm}^{-3}$ with the lowest input V/III ratio/GR.

### Table 2. Growth conditions of GaN SIMS stacks with the corresponding GRs and carbon concentrations.

| Group  | Pressure [Torr] | V/III ratio | Laser power [W] | GR $[\mu\text{m h}^{-1}]$ | $[C] (\times 10^{16} \text{cm}^{-3})$ |
|--------|----------------|-------------|-----------------|-----------------|-----------------|
| Group I | 150            | 876         | 0               | 5.2             | 2.5             |
|         |                |             | 50              | 5.2             | 2.4             |
|         |                |             | 150             | 4.6             | 1.5             |
|         |                |             | 250             | 4.4             | 1.2             |
|         | 200            |             | 0               | 5.2             | 1.5             |
|         |                |             | 200             | 4               | 0.55            |

Figure 6. $[C]$ and GRs of LA-MOCVD GaN as a function of input laser power. The growth conditions of the samples are listed in Table 2.
2.4. Transport Properties

Figure 8a compares the room-temperature mobilities of MOCVD grown n-GaN films on both GaN/sapphire templates and semi-insulating free-standing GaN substrates, with and without laser excitation. The GRs of conventional MOCVD and LA-MOCVD GaN films were 5.2 and 4.5 μm h⁻¹, respectively. A high C layer ([C] ≈ 10¹⁸ cm⁻³) with 2 μm thickness was grown first to isolate the charges at the growth interface. Then, an n-GaN layer was grown on top of the high C layer for Hall measurement. The trend indicates that that GaN films grown via LA-MOCVD have superior room-temperature mobilities as compared with those grown without laser excitation. The electron mobilities of GaN films grown without laser excitation ranged from 600 to 750 cm²V⁻¹s⁻¹. LA-MOCVD GaN films show higher electron mobilities ranging between 750 and 1275 cm²V⁻¹s⁻¹. The enhanced electron mobilities from LA-MOCVD GaN samples are most likely due to the reduced impurity scattering effect from lower carbon concentration.[33] Figure 8b compares the room-temperature electron mobilities of GaN grown via LA-MOCVD from this work as compared with those grown via molecular beam epitaxy (MBE), halide vapor phase epitaxy (HVPE), and regular MOCVD. The LA-MOCVD GaN films are among the few reports that achieved room-temperature mobility over 1000 cm²V⁻¹s⁻¹. More importantly, LA-MOCVD GaN films contain a GR at 4.5 μm h⁻¹, much higher than those grown via MBE or conventional MOCVD conditions. It is worth noting that, in general, the electron mobilities of GaN grown on free-standing GaN substrates are higher than that grown on GaN templates. This could be ascribed to the higher dislocation scattering effect for GaN grown on GaN templates.[34]

3. Conclusion

LA-MOCVD GaN epitaxy was developed to facilitate NH₃ decomposition and, thus, to achieve high-quality GaN films with low-C and fast GR. It is confirmed that NH₃ molecules are most efficiently decomposed with CO₂ laser excitation at 9.219 μm. The strong linear relationship of the GaN GR (as high as 10 μm hr⁻¹) with TMGa flow rate and pits-free surface morphology at high GR (8.3 μm hr⁻¹) from LA-MOCVD GaN indicate high effective V/III ratios due to efficient NH₃ decomposition. The background [C] in LA-MOCVD GaN films decreased monotonically with the increase in the laser power. Low [C] at 5.5 × 10¹³ cm⁻³ was achieved in the LA-MOCVD GaN film grown with the GR of 4 μm h⁻¹. LA-MOCVD GaN exhibited lower [C] incorporation at low input V/III ratio and high GRs as compared with the conventional MOCVD GaN growth. High room-temperature mobilities over 1000 cm²V⁻¹s⁻¹ were achieved by LA-MOCVD with a GR of 4.5 μm h⁻¹ grown on both free-standing GaN substrate and GaN/sapphire template. The LA-MOCVD GaN growth technique provides a promising approach to obtain high-quality, low-C GaN with fast GR for power device applications. The similar method can also be extended to the epitaxy of other nitride semiconductors.

4. Experimental Section

GaN films were grown on 2 in. (0001) GaN-on-sapphire templates (GaN templates, threading dislocation densities (TDD) of 2 x 10⁶ cm⁻²) and ammonothermal Mn-doped free-standing GaN substrates (TDD < 10⁴ cm⁻²) in a commercial vertical rotating-disk MOCVD reactor (Agnitron Technology Inc., Agilis MOCVD R&D System). The temperature of growth surface was measured in situ via a pyrometer. TMGa and NH₃ were used as Ga and N precursors, respectively. H₂ was used as the carrier gas. Diluted silane (SiH₄) balanced in N₂ was used as Si dopant. Impurities of selected samples were characterized by quantitative SIMS, with the carbon detection limit (DL) of 1 x 10¹⁵ cm⁻³. Surface morphologies of GaN films were characterized by a Huvitz HRM-300 optical microscope, Thermo Scientific Apreo SEM, and Bruker AFM. Room-temperature electron transport properties were measured by van-der-Pauw Hall configuration using the Ecopia HMS-3000 Hall effect system.

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