

Contents lists available at ScienceDirect

# Journal of Crystal Growth

journal homepage: www.elsevier.com/locate/jcrysgro



## Photoluminescence study of Si-doped a-plane GaN grown by MOVPE

Dabing Li <sup>a,b</sup>, Bei Ma <sup>a</sup>, Reina Miyagawa <sup>a</sup>, Weiguo Hu <sup>a</sup>, Mitsuhisa Narukawa <sup>a</sup>, Hideto Miyake <sup>a,\*</sup>, Kazumasa Hiramatsu <sup>a</sup>

#### ARTICLE INFO

Available online 19 January 2009

PACS: 78.55.Cr 81.15.Gh 68.55.Ln

Keywords:
A1. Photoluminescence
A1. Si doping
A3. MOVPE
B1. a-Plane GaN

#### ABSTRACT

Si-doped *a*-plane GaN films with different doping concentrations were grown by metal-organic vapor phase epitaxy. A mirrorlike surface without pits or anisotropic stripes was observed by optical microscopy. Detailed optical properties of the samples were characterized by temperature- and excitation-intensity-dependent PL measurements. A series of emission peaks at 3.487, 3.440, 3.375–3.350, 3.290 and 3.197 eV were observed in the low-temperature PL spectra of all samples. The origin of these emissions is discussed in detail.

© 2009 Elsevier B.V. All rights reserved.

### 1. Introduction

Nonpolar GaN and its alloys have attracted increasing attention for use in optoelectronic device applications because conventional c-plane III-nitride quantum well (QW) has a large polarization electric field, which has a significant effect on the efficiency of optoelectronic devices [1]. a-Plane  $(11\bar{2}0)$ GaN as one of the promising nonpolar nitrides has been extensively studied [2]; however, there are few detailed reports on the optical properties of Si-doped a-plane GaN [3,4]. Yu et al. have studied the emission properties of Si-doped a-plane GaN by temperature-dependent photoluminescence (PL) and they assigned the peak at 3.42 eV to donor-acceptor pair (DAP) emission based on the results of temperature dependence of its emission [3]. However, in undoped a-plane GaN, emission at the 3.42 eV has also been observed, which was attributed to the emission from excitons bound to basal plane stacking faults (BSFs) [2]. Lee et al. have reported the room-temperature (RT) PL spectra of Si-doped a-plane GaN and found that broad near-band-edge (NBE) emission ranged from 3.29 to 3.41 eV, which was assigned to stacking faults. However, detailed investigation of the origin of the emission was not carried out by the authors [4]. In this study, optical properties of 2-µmthick Si-doped a-plane GaN films grown on an 8-µm-thickundoped a-plane GaN/r-plane sapphire with different carrier concentrations were investigated in detail by temperature- and excitation-intensity-dependent PL measurements.

## 2. Experimental procedure

Approximately  $2-\mu m$ -thick Si-doped a-plane GaN films with different  $CH_3SiH_3$  flow rates (0, 2, 5 and 10 sccm) were grown by metal-organic vapor phase epitaxy (MOVPE). Prior to the growth of Si-doped a-plane GaN, unintentionally doped a-plane GaN with a thickness of about  $8 \mu m$  was deposited on nitrided r-plane sapphire, and no buffer layers were inserted between the undoped layer and the r-plane sapphire substrate. The growth temperature and pressure were  $1060\,^{\circ}C$  and  $500\,\text{Torr}$ , respectively. Trimethylgallium (TMG) and ammonia (NH<sub>3</sub>) were, respectively, used as the Ga and N precursors.  $CH_3SiH_3$  was used as the dopant source. The optical properties of Si-doped  $(11\bar{2}\,0)$  GaN films were investigated in detail by temperature- and excitation-intensity-dependent PL measurements using a  $325\,\text{nm}$  He–Cd laser. Optical microscopy and high-resolution X-ray diffraction were employed to characterize the surface morphology and crystalline quality.

## 3. Results and discussion

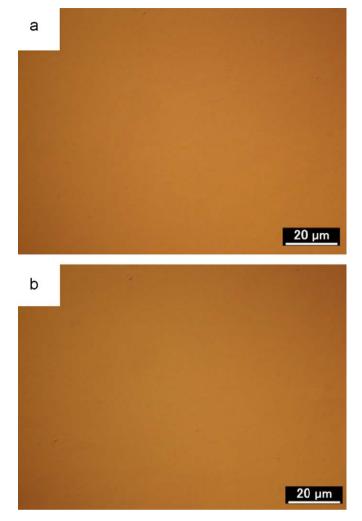
Fig. 1 shows differential optical microscopy images of Si-doped *a*-plane GaN obtained with CH<sub>3</sub>SiH<sub>3</sub> flow rates of 0 (undoped) and 10 sccm. According to Fig. 1, the surfaces of undoped and Si-doped *a*-plane GaN are mirrorlike and no pits or anisotropic morphology are observed, which means that the surface quality of our sample is good and that Si doping has no obvious effect on the surface morphology. The effect of Si doping on the surface morphology and structural properties will be reported in detail elsewhere of this issue[5].

<sup>&</sup>lt;sup>a</sup> Department of Electrical and Electronics Engineering, Mie University, 1577 Kurimamachiya, Tsu 514-8507, Japan

b Key Laboratory of Excited State Processes, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, PR China

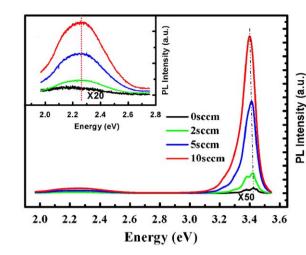
<sup>\*</sup>Corresponding author. Tel./fax: +81592319399. E-mail address: miyake@elec.mie-u.ac.jp (H. Miyake).

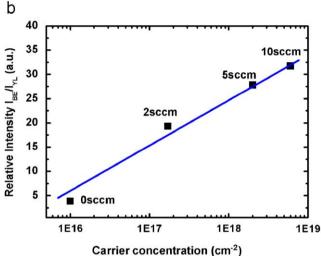
a



**Fig. 1.** Differential optical microscopy images of Si-doped *a*-plane GaN with CH<sub>3</sub>SiH<sub>3</sub> flow rates of (a) 0 sccm and (b) 10 sccm.

The RT-PL spectra of the Si-doped a-plane GaN are shown in Fig. 2. All the spectra exhibit NBE emission, which increase in intensity with increasing dopant flow rates. A yellow line (YL) emission peaked at 2.2-2.3 eV (Fig. 2(a)). Furthermore, the NBE emission shifts to the lower energy and the full-width at halfmaximum (FWHM) of the PL spectra becomes broader with increasing CH<sub>3</sub>SiH<sub>3</sub> flow rate, i.e., carrier concentration. This phenomenon has also been observed in Si-doped c-plane GaN, which was attributed to band-gap renormalization [6]. For aplane GaN, the above-mentioned phenomenon can also be assigned to the band-gap renormalization. The inset of Fig. 2(a) depicts an enlargement of the YL-emission spectra, whose intensity increases as the CH<sub>3</sub>SiH<sub>3</sub> flow rate increases from 0 to 10 sccm. Yu et al. attributed the origin of the YL emission of Sidoped a-plane GaN to the formation of a  $V_{Ga}$ - $O_N$  (Ga vacancy and O on N sites) complex [3]. As known from theoretical calculation and experimental measurement,  $V_{Ga}$  is the main defect responsible for YL emission in GaN [7,8]. The calculation demonstrated that the formation energy of  $V_{Ga}$  decreases with increasing Fermi level, i.e., increasing n-type carrier concentration. Therefore, Si doping can easily lead to the form of Ga vacancies, and thus the intensity of YL emission increases. In our case, the YL peak becomes stronger with increasing Si-dopant flow rates; thus, it is proposed that the Ga vacancies are the main origin of the YL emission in our Si-doped a-plane GaN. However, note that the ratio of the NBE-emission intensity ( $I_{BE}$ ) and

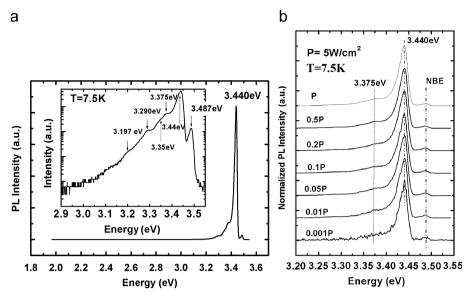




**Fig. 2.** Room-temperature PL spectra of Si-doped a-plane GaN with different CH<sub>3</sub>SiH<sub>3</sub> flow rates (a) and the dependence of the ratio of  $I_{\rm BE}$  to  $I_{\rm YL}$  on the carrier concentration (b). The straight line is a guide to the eyes. The inset shows the yellow emission of Si-doped a-plane GaN at different Si-dopant flow rates. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

YL-emission intensity ( $I_{\rm YL}$ ) does not decrease, but increases with increasing dopant concentration (Fig. 2(b)), which proves that the crystalline quality does not deteriorate with Si doping.

Fig. 3(a) shows the low-temperature (LT) PL spectra of the Sidoped a-plane GaN with a CH<sub>3</sub>SiH<sub>3</sub> flow rate of 10 sccm. The inset of the figure shows part of the LT-PL spectra with a logarithmic scale. The spectra reveal a series of emission peaks at 3.487, 3.440, 3.375-3.350, 3.290 and 3.197 eV. The 3.487 eV emission line is regarded as the NBE transition, which comprises free and bound exciton lines [3]. The 3.440 eV emission line is 20 meV higher than the reported 3.42 eV emission line in  $\alpha$ -plane GaN [2]. The 3.42 eV emission line is usually assigned to a DAP [3] and BSFs recombination [2]. In our case, for the 3.440 eV emission line, the origin of DAP recombination can be excluded according to the excitation-intensity dependence of the PL spectra (Fig. 3(b)) and the fact that no blue shift was observed upon increasing the excitation intensity by over three orders of magnitude. Further PL measurement demonstrates that the temperature dependence of the peak energy (for the 3.440 eV line) (Fig. 4) has a similar trend to that of the 3.42 eV emission line reported in Ref. [2], i.e., the peak position of the 3.440 eV emission undergoes a slight redshift and then a blueshift in the temperature range 7.5-90 K, and above



**Fig. 3.** Low-temperature (7.5 K) PL spectra of Si-doped *a*-plane GaN with a CH<sub>3</sub>SiH<sub>3</sub> flow rate of 10 sccm (a) and excitation-intensity dependence of PL spectra of Si-doped GaN with a CH<sub>3</sub>SiH<sub>3</sub> flow rate of 10 sccm (b). The inset of (a) shows part of the PL spectra with a logarithmic scale.

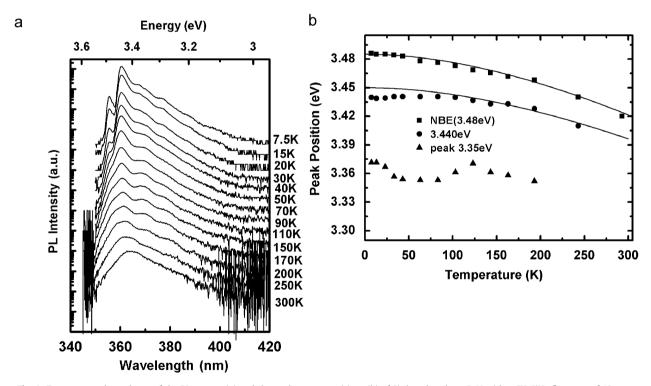


Fig. 4. Temperature dependence of the PL spectra (a) and the peak energy positions (b) of Si-doped a-plane GaN with a  $CH_3SiH_3$  flow rate of 10 sccm.

90 K the peak indicates the thermally induced band-gap shrinkage. Thus, in this study, the 3.440 eV line also originated from BSFs, and not from DAP recombination. The NBE-emission spectra can be well-fitted by Varshni's empirical equation  $E_{\rm NBE}=E_0-\alpha T^2/(T+\beta)$  with parameters of  $\alpha=8\times10^{-4}\,{\rm eV/K}$  and  $\beta=834\,{\rm K}$ , which are similar to the values reported for c-plane GaN [9,10]. The peak at 3.29 eV probably results from defect-related emission as previously reported [2], and the peak at 3.197 eV is its LO phonon replica. In Fig. 4, there is an emission peaked at 3.35–3.75 eV with an S-shaped temperature dependence of the peak position. Moreover, compared with the LT-PL (7.5 K) spectra of the undoped a-plane GaN, the intensity of this peak in Si-doped a-plane GaN is stronger than that in

undoped *a*-plane GaN. Liu et al. assigned this peak possible origin from prismatic stacking faults (PSFs) [11]; therefore, our results may demonstrate that this peak is related to Si doping inducing PSF defects, although further investigation is required since the emission mechanism of this peak is still not clear.

## 4. Conclusion

Si-doped *a*-plane GaN films with different doping concentrations were studied by temperature- and excitation-density-dependent PL measurements. At RT, all samples exhibited UV transitions peaking from 3.3991 to 3.4245 eV and deep-level YL

emission at 2.2–2.3 eV. The YL emission was attributed to a  $V_{\rm Gar}$  related transition. At a low temperature, a series of emission peaks at 3.487, 3.440, 3.375–3.350, 3.290 and 3.197 eV were observed in the PL spectra of all samples. The temperature- and excitation density-dependent PL measurements demonstrated that the nearband-edge emission (3.487 eV) closely fitted Varshni's formula. The 3.440 eV emission was attributed to recombination related to BSFs, and the peak range from 3.350 to 3.375 eV was thought to be related to Si-doping inducing PSF defects.

## Acknowledgments

This work was partially supported by the Akasaki Research Center at Nagoya University, and the Ministry of Education, Culture, Sports, Science and Technology by Grants-in-aid for Scientific Research (Nos. 18360008 and 18560010) and for the Scientific Research on Priority Areas (No. 18069006).

### References

- P. Waltereit, O. Brandt, A. Trampert, H.T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche, K.H. Ploog, Nature 406 (2000) 865.
- [2] P.P. Paskov, R. Schifano, B. Monemar, T. Paskova, S. Figge, D. Hommel, J. Appl. Lett. 98 (2005) 093519.
- [3] H.B. Yu, H. Chen, D. Li, Y.J. Han, X.H. Zheng, Q. Huang, J.M. Zhou, J. Cryst. Growth 263 (2004) 94.
- [4] S.N. Lee, H.S. Paek, J.K. Son, T. Sakong, O.H. Nam, Y. Park, J. Cryst. Growth 307 (2007) 358.
- [5] B. Ma, N. Miyagawa, W.G. Hu, D.B. Li, H. Miyake, K. Hiramatsu, J. Cryst. Growth, this issue, doi:10.1016/j.jcrysgro.2009.01.040.
- [6] M. Yoshikawa, M. Kunzer, J. Wagner, H. Obloh, P. Schlotter, R. Schmidt, N. Herres, U. Kaufmann, J. Appl. Phys. 86 (1999) 4400.
- [7] T. Mattila, R.M. Nieminen, Phys. Rev. B 55 (1997) 9572.
- [8] K. Saarinen, et al., Phys. Rev. Lett. 79 (1997) 3030.
- [9] M. Leroux, N. Grandjean, B. Beaumont, G. Nataf, F. Semond, J. Massies, P. Gibart, J. Appl. Lett. 86 (1999) 3721.
- [10] W. Shan, T.J. Schmidt, X.H. Yang, S.J. Hwang, J.J. Song, B. Goldenberg, Appl. Phys. Lett. 66 (1995) 985.
- [11] R. Liu, A. Bell, F.A. Ponce, C.Q. Chen, J.W. Yang, M.A. Khan, Appl. Phys. Lett. 86 (2005) 021908.