Stress, structural and electrical properties of Si-doped GaN film grown by MOCVD*

Xu Zhihao(许志豪)†, Zhang Jincheng(张进成), Duan Huantao(段焕涛), Zhang Zhongfen(张忠芬), Zhu Qingwei(朱庆玮), Xu Hao(徐浩) and Hao Yue(郝跃)

(Key Laboratory of Wide Band-Gap Semiconductor Materials and Devices, School of Microelectronics, Xidian University, Xi'an 710071, China)

Abstract: The stresses, structural and electrical properties of n-type Si-doped GaN films grown by metalorganic chemical vapor deposition (MOCVD) are systemically studied. It is suggested that the main stress relaxation is induced by bending dislocations in low doping samples. But for higher doping samples, as the Si doping concentration increases, the in-plane stresses in the grown films are quickly relaxed due to the rapid increase of the edge dislocation densities. Hall effect measurements reveal that the carrier mobility first increases rapidly and then decreases with increasing Si doping concentration. This phenomenon is attributed to the interaction between various scattering process. It is suggested that the dominant scattering process is defect scattering for low doping samples and ionized impurity scattering for high doping samples.

Key words: Si-doped GaN; stress relaxation; defect; electrical properties

DOI: 10.1088/1674-4926/30/12/123003 **PACC:** 7280E; 6110; 7360L

1. Introduction

Research interest in GaN has been driven by the significant technological importance of this material in recent years. Indeed, GaN is used in the fabrication of a range of electronic and photonic devices. Due to its important role in these applications, Si-doped GaN has been extensively studied in these years. It is well known that GaN films are usually heteroepitaxially grown on sapphire substrates. However, the lattice constants and thermal expansion coefficients of them are not well matched. This mismatch leads to a large compressive stress in the as-grown film when the temperature falls from the growth temperature (about 1000 °C) down to room temperature, resulting in a high dislocation density in the grown film[1]. Dislocations are electrically activity, have an influence on the device properties, and act as scattering centers for the carriers[2–4]. It was also observed that dislocations were involved in the yellow luminescence transition in GaN film[5–9]. Therefore, the influence of Si doping on the dislocations and the optical properties has been of great interest. So far, extensive research has been done to investigate the properties of n-type GaN. Kucheyev *et al.*[10] reported that the yellow band emission in n-type GaN originated from point defects such as Ga vacancies by investigating the optical properties of a series of Si-doped samples. Lee *et al.*[11] reported that the residual stresses in Si-doped GaN films were gradually relaxed by incorporating Si dopant in grown films. Furthermore, the electrical properties of n-type GaN film have been investigated by many research groups[12–14]. However, most of the previous research only focused on one aspect of the properties of the n-type GaN film, and systematical investigations of n-type GaN film are rarely found. In fact, up to now, not only are many underlying mechanisms in n-type GaN, such as the origin of the yellow luminescence still under debate, but also many problems, such as the connections of the stress, structural quality and electrical properties, are not well understood. Actually, these properties are very important among the parameters characterizing n-type GaN films. Therefore, it is of importance to know the relationships between these properties.

In this paper, the relationships between the stress, defects and carrier mobility of n-type GaN are presented. By combining an extensive set of characterization techniques, the stress relaxation mechanism and the carrier transport property of n-type GaN are analyzed in detail. This may be helpful to the growth of n-type GaN film.

2. Experiments

The six samples studied in this paper were grown on *c*-plane (001) sapphire substrates by a cold-wall low-pressure MOCVD system. Triethylgallium (TEGa), ammonia (NH_3) and hydrogen diluted silane (SiH_4) were used as the Ga, N and Si precursors, respectively, and highly purified hydrogen (H_2) was used as the carrier gas. Before the growth of the GaN buffer layer, a GaN nucleation layer with a thickness of 50 nm was initiated grown at 500 °C, then an unintentionally doped GaN layer with a thickness of 450 nm and a Si-doped GaN

* Project supported by the Major Program and Key Project of National Natural Science Foundation of China (Nos. 60890191, 60736033) and the National Key S&T Special Project (No. 2008ZX01002).

† Corresponding author. Email: forman1115@163.com

Received 16 May 2009, revised manuscript received 12 July 2009

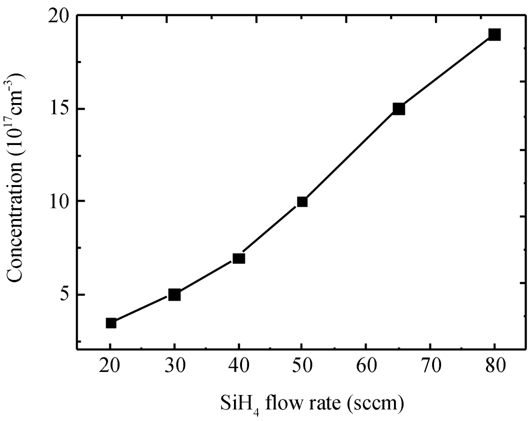

Fig. 1. SiH_4 flow rates versus carrier concentrations for the samples.

layer with a thickness of 500 nm were grown on the nucleation layer at about ∼1000 °C. The chamber pressure was held at 40 Torr during the whole growth process. The total thickness of each sample was measured at about 1 μm by a transmission spectrum analyzer measurement[15]. By controlling the diluted SiH_4 flow rate, the carrier concentrations in the six samples were 3.5×10^{17}, 5.0×10^{17}, 7.0×10^{17}, 1.0×10^{18}, 1.5×10^{18} and 1.9×10^{18} cm^{-3}, respectively, labeled as sample 1 to sample 6. Figure 1 shows the relationship between the diluted SiH_4 flow rates and the carrier concentrations for the samples. It is obvious that the carrier concentration is proportional to the SiH_4 flow rate. For characterizations of the samples, the structural qualities of the films are determined by high resolution X-ray diffraction (HRXRD) measurements, and van der Pauw Hall measurements are used to characterize the electrical properties.

3. Results and discussion

3.1 . Relationship between stress and defects

To determine the crystal qualities of the samples, X-ray rocking curve measurements of the (002) symmetric and the (102) skew symmetric diffraction were first performed using a Bruker D8 discover diffractometer, shown in Figs. 2 and 3. As seen in Fig. 3, as the Si doping concentration increases, the full width at half maximum (FWHM) of the (102) diffractions of the GaN samples decreases when the carrier concentration is low, while it quickly increases in the samples where the carrier concentrations are higher than 7.0×10^{17} cm^{-3}. It was also found that the FWHM of the (002) diffractions shows no obvious variations in the whole investigated doping range. It is well known that the FWHM for the (002) plane and the (102) plane indirectly represent the screw dislocation density and the edge dislocation density of a film, respectively. Thus, the above-mentioned results imply that the screw dislocation densities of the as-grown GaN layers are not particularly influenced by the Si doping concentrations in the investigated doping range. On the other hand, it is found that there is a well-regulated relationship between the Si doping concentration and the edge dislocation density, i.e., the edge dislocation densities of the layers first decrease and then increase as the Si doping

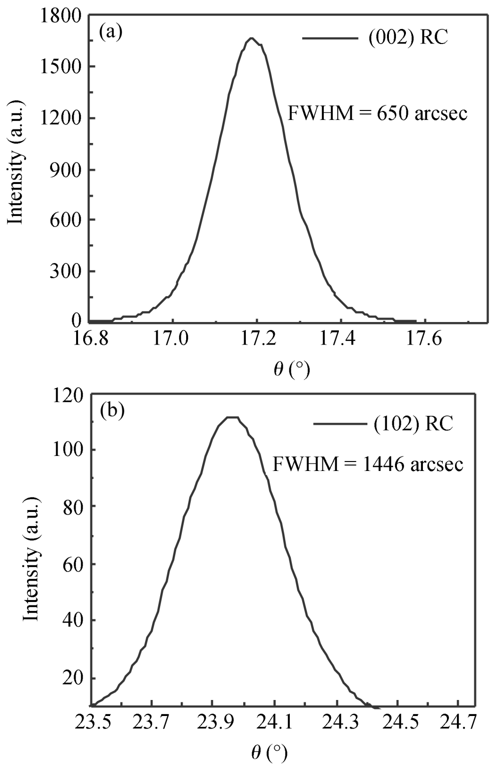

Fig. 2. XRD rocking curves of sample 3: (a) (002) plane; (b) (102) plane.

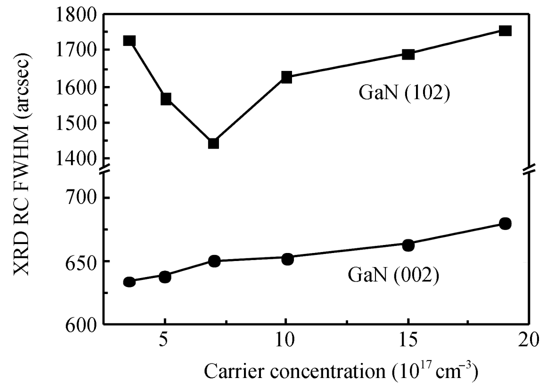

Fig. 3. XRD FWHM of the samples versus carrier concentrations.

concentration increases.

In order to determine the in-plane stresses in the samples, 2θ–ω scans of the (002) diffractions were performed using HRXRD. Figure 4(a) shows the measured 2θ–ω curves of the samples. The sapphire (006) peak (which is located at 41.62 °C) is used as the reference, thus the c lattice constant of the GaN film can be exactly calculated. The in-plane stresses of the samples can be roughly estimated by the following equation:

$$\sigma_{xx} = \varepsilon_{zz} B/\upsilon,$$

where B is the bulk modulus (200 GPa)[16], υ is the Poisson ratio (0.23)[17] and ε_{zz} is the relative strain of the c axis in a GaN film. The ε_{zz} in the above equation can be calculated by the following equation:

$$\varepsilon_{zz} = (c - c_0)/c_0,$$

where c represents the c lattice constant measured by XRD and

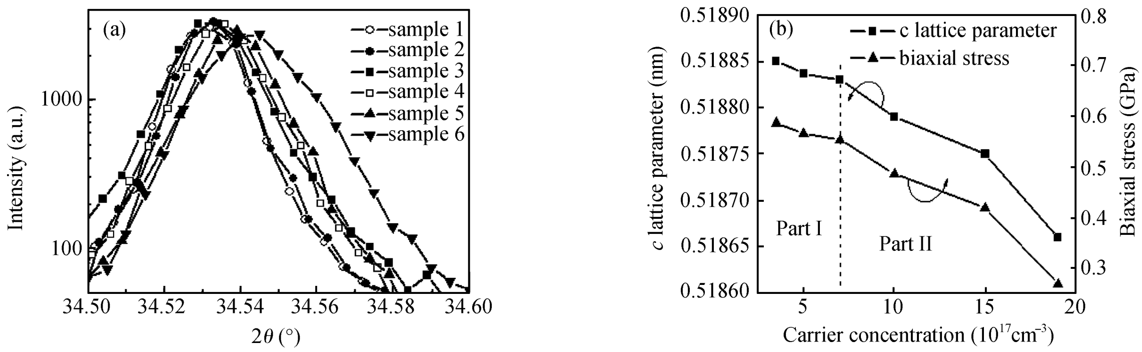

Fig. 4. (a) 2θ–ω curves of the samples measured by HRXRD; (b) Dependence of the c lattice constant and the in-plane stress on the carrier concentration.

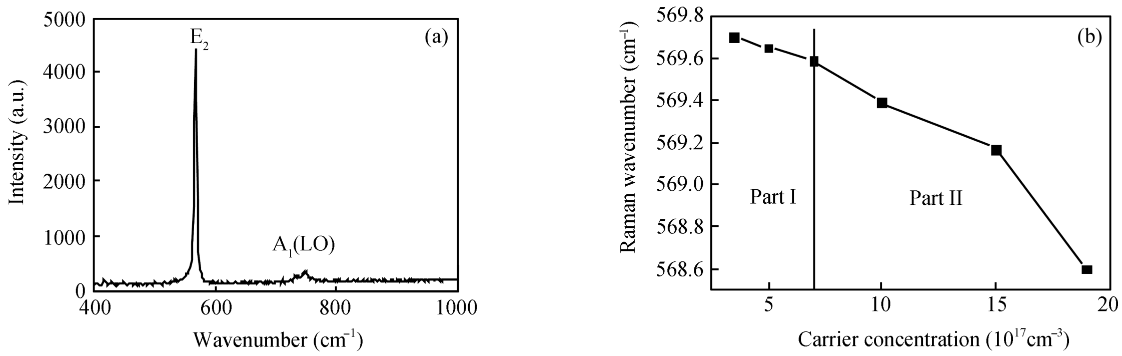

Fig. 5. (a) Raman spectrum of sample 6; (b) Raman wave numbers for the samples.

c_0 is the c lattice constant for strain-free bulk GaN (c_0 = 5.185 Å)[18]. Figure 4(b) shows the c lattice constant and the in-plane stress as functions of the carrier concentration in the samples. From Fig. 4(b), it is easily found that the in-plane stresses in the GaN films are relaxed with increasing the doping concentration. Studying Fig. 4(b) carefully, it is found that the curves can be divided into two parts: part I and part II. As the doping concentration increases, it is obvious that the biaxial stresses in the films decrease slowly in part I while they decrease quickly in part II, i.e., the in-plane stresses in the films are gradually relaxed in part I while the stresses are quickly relaxed in part II. This result implies that though the incorporation of Si dopant in a film can lead to relaxation of the in-plane stress, the stress relaxation in the grown film is correlated with the Si doping concentration. The relaxation of the stress is gradual and unobvious when the doping concentration is low. However, in the sample where the carrier concentration exceeds the concentration of 7.0×10^{17} cm^{-3}, the in-plane stress is quickly relaxed and can be obviously observed in the curves.

To further confirm this phenomenon, micro-Raman measurements are used to determine the in-plane stresses in the grown films. Figure 5(a) shows the measured result of sample 6. The measurement was taken under a backscattering configuration with the incident ray parallel to the c-axis of the measured film. Two peaks are observed in Fig. 5(a). It is clarified that only two allowed modes can be observed under the above-mentioned configuration according to the Raman selection rules[19]. Thereby, the strong peak centered at about 568 cm^{-1} and the weak peak centered at about 742 cm^{-1} correspond to the E_2 mode and the A_1(LO) mode, respectively. The peak corresponding to the E_2 mode is fitted with a Lorenz lineshape (not shown here). Therefore, the in-plane stress in the measured film can be evaluated with the wavenumber of the E_2 mode. As seen in Fig. 5(b), as the Si doping concentration increases, the in-plane stress in the GaN film is gradually relaxed at first. However, the stress is then quickly relaxed when the carrier concentration exceeds the threshold value, which is 7.0×10^{17} cm^{-3} in our samples. This result is in good agreement with the result measured by HRXRD shown above.

Combining the HRXRD ω scan measurements, the above-mentioned phenomenon is analyzed in detail. It is found that many bending dislocations are formed when a little Si dopant is incorporated into the GaN epilayer[20, 21]. Thus, a fraction of the in-plane stress in the film will be relaxed due to the lateral misfit components of the bending dislocations, i.e., it is suggested that in a sample with a low Si doping concentration, a part of the stress will be relaxed due to the existence of the bending dislocations. It is also confirmed that an annihilation of the threading dislocations occurs due to the reaction of the bending dislocations and the threading dislocations[21]. It is considered that the annihilation will in turn prevent the relaxation of the in-plane stress. Thus, the total effect of the complicating reactions is that the in-plane stress is gradually relaxed in the sample with a low doping concentration. However, as seen in Fig. 3, lots of edge dislocations will be formed by introducing more Si dopant, leading to a quick relaxation of the stress. In fact, it is inferred from the above-mentioned analysis that the in-plane stresses are mainly relaxed by bending dislocations in light doping samples; while the in-plane stresses in heavy doping samples are mainly relaxed by edge

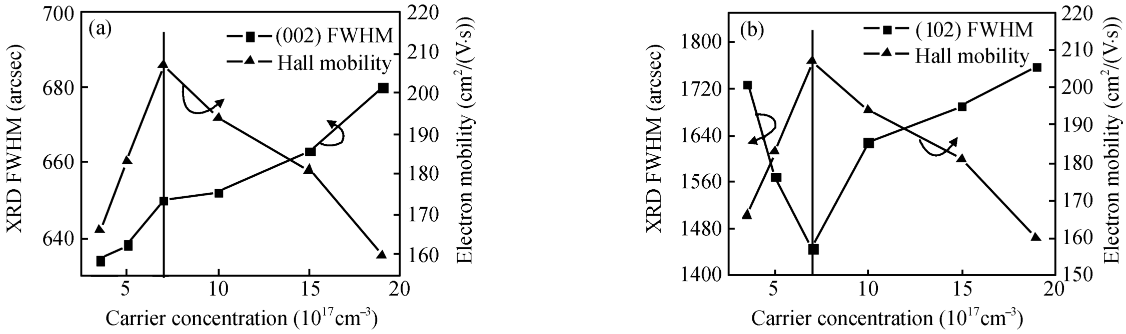

Fig. 6. (a) (002) and (b) (102) FWHM and electron mobility as functions of the carrier concentration.

dislocations.

3.2. Relationship between the defects and the electrical properties

To investigate the electrical properties of the GaN layers, van der Pauw Hall effect measurements are employed to determine the carrier mobilities of the layers. Figure 6 shows the carrier mobility and the XRD FWHM as functions of the carrier concentration in the samples. For convenience in the following discussion, the curve in Fig. 6 is divided into two regions according to the carrier concentration. The region in which the carrier concentration is lower than 7.0×10^{17} cm^{-3} is called the light doping region while the other region is called the heavy doping region. As seen from the curve, the carrier mobilities of the samples in the light doping region increase with increasing doping concentration. However, it is found that the carrier mobilities decrease with increasing doping concentration for the samples in the heavy doping region. This result disagrees with the conclusion obtained by Wang *et al.*[22]. It is thought that this discrepancy is mainly due to the Si doping concentrations being too heavy in the samples investigated by Wang *et al.*, and thus the phenomenon in the light doping region cannot be observed in their samples.

In order to deeply analyze the relationship between the defects and the carrier mobility, the connection between the dislocations and the carrier mobility is first studied. Focusing on the dependences of FWHM and carrier mobility on carrier concentration in the samples, it is easily found from the curves that the carrier mobility is not correlated with the screw dislocations but seems to depend strongly on the edge dislocations. In the whole investigated doping range, carrier mobility decreases with increasing edge dislocations, both in the light doping region and in the heavy doping region. This is mainly because the edge dislocation density in the grown epilayer is about two orders of magnitude higher than that of the screw dislocation density in the epilayer, and thus the carriers are scattered much more by edge dislocations. Studying Fig. 6(b) carefully, it is found that both the doping concentration and the edge dislocation density of sample 4 are higher than those of sample 2, but the carrier mobility of sample 4 is slightly higher than that of sample 2, i.e., though the carrier mobility is well correlated with the edge dislocation density in both regions, this does not hold true in the whole investigated doping range. It is suggested that this phenomenon can be attributed to the interaction between various scattering processes.

In order to study the above-mentioned phenomenon, a detailed analysis of the influence of various kinds of scattering processes on the carrier transport property of n-type GaN is given in the following discussion. It is well known that the carrier mobility of an n-type GaN film is mainly limited by the defect scattering process (including the vacancy scattering process and the dislocation scattering process), the ionized impurity scattering process and the lattice vibration scattering process at room temperature. In the following discussion, the lattice vibration scattering process is not considered because it is only dependent on temperature. In an undoped GaN film, the carriers are strongly scattered by the vacancies because of the enormous vacancies existing in the film, resulting in a very low carrier mobility of the undoped GaN film. By investigating the optical properties of n-type GaN, it is revealed that a fraction of the vacancies will be filled by Si atoms by incorporating Si dopant in a film[23, 24], resulting in a great reduction of the vacancy scattering. It is also reported by Ng *et al.*[25] that the introduction of Si dopant into a GaN film will screen the dislocation scattering by decreasing the Debye screening length. Therefore, in samples with low Si doping concentrations, the carrier mobility increases rapidly with the increase of the doping concentration due to the significant reduction of the defect scattering and the weak ionized impurity scattering caused by the low Si concentrations in the samples. Furthermore, as the doping concentration increases, the edge dislocation density decreases in these samples, weakening the dislocation scattering. In other words, the carrier mobilities of samples are mainly limited by the defect scattering in the light doping region. However, the ionized impurity scattering becomes stronger and dominates the carrier scattering process when the doping concentration exceeds the threshold concentration. In this case, as mentioned above, the edge dislocation density quickly increases when more Si atoms are introduced into the film, leading to a further reduction of the carrier mobility. However, because of the very low defect scattering, the carrier mobility decreases slowly when the doping concentration is slightly heavier than the threshold concentration, i.e., it is inferred that the carrier mobility decreases slowly when the doping concentration is not much heavier than the threshold concentration. Thus, it is possible that the carrier mobility of a sample with a higher doping concentration and a higher dislocation density is slightly higher than that of a sample with a lower doping concentration and a lower dislocation density. Actually, the above-mentioned analysis is in good agreement with the experimental results. As seen from the above analysis, the carrier mobility of n-type GaN is greatly affected by vacancy scattering, dislocation scattering and ionized impurity scattering at room temperature. As Si doping concentration increases, some of the scattering processes are strengthened while some of them are weakened, implying the influence of various scattering processes on the carrier mobility is not a simple one.

4. Conclusion

The stresses, structural and electrical properties of Si-doped GaN films grown by metalorganic chemical vapor deposition (MOCVD) are systematically studied in this paper. It is found that stress relaxation in the as-grown GaN films is correlated with Si doping concentration. This phenomenon is attributed to the following causes. For the sample with a light Si doping concentration, the layer is mainly relaxed by bending dislocations. Furthermore, as the Si doping concentration increases, a reduction of the edge dislocation density in the film occurs, slowing the stress relaxation in the film, while in the sample with a heavy Si doping concentration, the layer is quickly relaxed due to the rapid increase of edge dislocations. The van der Pauw Hall measurements reveal that the room temperature electron mobility in a Si-doped GaN film is mainly restricted by the defect scattering process and the ionized impurity scattering process. It is revealed that the dominant scattering process in light doping samples is the defect scattering process. In this case, the room temperature carrier mobility increases with decreasing dislocation scattering and vacancy scattering. As the Si doping concentration increases, the carrier mobility decreases due to the ionized impurity scattering process dominating the scattering process.

References

[1] Iwaya M, Terao S, Hayashi N, et al. High-efficiency GaN/Al$_x$Ga$_{1-x}$N multi-quantum-well light emitter grown on low-dislocation density Al$_x$Ga$_{1-x}$N. Surf Sci, 2000, 159: 405

[2] Cho H K, Kim C S, Hong C H. Electron capture behaviors of deep level traps in unintentionally doped and intentionally doped n-type GaN. J Appl Phys, 2003, 94: 1485

[3] Muret P, Philippe A, Monroy E, et al. Luminescence properties of defects in GaN. J Appl Phys, 2002, 91: 2998

[4] Hansen P J, Strausser Y E, Erickson A N, et al. Scanning capacitance microscopy imaging of threading dislocations in GaN films grown on (001) sapphire by metalorganic chemical vapor deposition. Appl Phys Lett, 1998, 72: 2247

[5] Soh C B, Chua S J, Lim H F, et al. Assignment of deep levels causing yellow luminescence in GaN. J Appl Phys, 2004, 96: 1341

[6] Reshchikov M A, Morkoc H. Luminescence properties of defects in GaN. Appl Phys Rev, 2005, 97: 061301

[7] Zhao D G, Jiang D S, Yang H, et al. Role of edge dislocations in enhancing the yellow luminescence of n-type GaN. Appl Phys Lett, 2006, 88: 241917

[8] Ponce F A, Bour D P, Gotz W, et al. Spatial distribution of the luminescence in GaN thin films. Appl Phys Lett, 1996, 68: 57

[9] Kwon Y H, Shee S K, Gainer G H, et al. Time-resolved study of yellow and blue luminescence in Si- and Mg-doped GaN. Appl Phys Lett, 1999, 76: 840

[10] Kucheyev S O, Toth M, Phillips M R, et al. Chemical origin of the yellow luminescence in GaN. J Appl Phys, 2002, 91: 5867

[11] Lee I H, Choi I H, Lee C R, et al. Stress relaxation in Si-doped GaN studied by Raman spectroscopy. J Appl Phys, 1998, 83: 5787

[12] Shiojima K, Suemitsu T, Ogura M. Correlation between current-voltage characteristics and dislocations for n-GaN Schottky contacts. Appl Phys Lett, 2001, 78: 3636

[13] Krtschil A, Dadgar A, Krost A. Decoration effects as origin of dislocation-related charges in gallium nitride layers investigated by scanning surface potential microscopy. Appl Phys Lett, 2003, 82: 2263

[14] Laukkanen P, Lehkonen S, Uusimaa P, et al. Structural, electrical, and optical properties of defects in Si-doped GaN grown by molecular-beam epitaxy on hydride vapor phase epitaxy GaN on sapphire. J Appl Phys, 2002, 92: 786

[15] Zhang J C, Hao Y, Li P X. Thickness measurement of GaN film based on transmission spectra. Acta Phys Sin, 2004, 53: 1243

[16] Xu Y N, Ching W Y. Electronic, optical, and structural properties of some wurzite crystals. Phys Rev B, 1993, 48: 4335

[17] Kisielowski C, Kruger J, Ruvimov S, et al. Edge-on micro-Raman assessment of trigonal modes in partially ordered GaInP$_2$. Phys Rev B, 1996, 54: 17754

[18] Detchprohm T, Hiramatsu K, Itoh K, et al. Relaxation process of the thermal strain in the GaN/α-Al$_2$O$_3$ heterostructure and determination of the intrinsic lattice constants of GaN free from the strain. Jpn J Appl Phys, 1992, Part 2, 31: 1454

[19] Arguello C A, Rousseau D L, Porto S P S. First-order Raman effect in wurtzite-type crystals. Phys Rev B, 1969, 181: 1351

[20] Cantu P, Wu F, Waltereit P, et al. Si doping effect on strain reduction in compressively strained Al$_{0.49}$Ga$_{0.51}$N thin films. Appl Phys Lett, 2003, 83: 674

[21] Ruvimov S, Liliental-Weber Z, Suski T, et al. Effects of Si doping on the dislocation structure of GaN grown on the A-face of sapphire. Appl Phys Lett, 1996, 69: 990

[22] Wang L, Zhang X P, Xi G Y, et al. Study on electrical properties of n-GaN grown at low temperature by metal-organic vapor phase epitaxy. Acta Phys Sin, 2008, 57: 5923

[23] Yang H C, Lin T Y, Huang M Y, et al. Optical properties of Si-doped GaN films. J Appl Phys, 1999, 86: 6124

[24] Suski T, Perlin P, Teisseyre H, et al. Mechanism of yellow luminescence in GaN. Appl Phys Lett, 1995, 67: 2188

[25] Ng H M, Doppalapudi D, Moustakas T D, et al. The role of dislocation scattering in n-type GaN films. Appl Phys Lett, 1998, 73: 821