In situ growth monitoring of AlGaN/GaN distributed Bragg reflectors at 530 nm using a 633 nm laser*

Wen Feng(文锋), Huang Lirong(黄黎蓉)†, Jiang Bo(姜波), Tong Liangzhu(童梁柱), Xu Wei(徐巍), and Liu Deming(刘德明)

(Wuhan National Laboratory for Optoelectronics, College of Opto-Electronics Science & Engineering, Huazhong University of Science & Technology, Wuhan 430074, China)

Abstract: The metal-organic chemical vapor deposition (MOCVD) growth of AlGaN/GaN distributed Bragg reflectors (DBR) with a reflection peak at 530 nm was *in situ* monitored using 633 nm laser reflectometry. Evolutions of *in situ* reflected reflectivity for different kinds of AlGaN/GaN DBR were simulated by the classical transfer matrix method. Two DBR samples, which have the same parameters as the simulated structures, were grown by MOCVD. The simulated and experimental results show that it is possible to evaluate the DBR parameters from the envelope shape of the *in situ* reflectivity spectrum. With the help of the 633 nm laser reflectometry, a DBR light emitting diode (LED) was grown. The room temperature photoluminescence spectra show that the reflection peak of the DBR in the LED is within the design region.

Key words: light emitting diode; metal-organic chemical vapor deposition; GaN; distributed Bragg reflector
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1. Introduction

Low extraction efficiency in light emitting diodes (LEDs) is one of the important problems limiting the efficiency[1]. Distributed Bragg reflectors (DBRs) have been demonstrated to be effective in improving the light output of LEDs[2, 3]. Semiconductor based DBRs are always grown under the LED. In this way, they do not reduce the electrical injection efficiency compared with dielectric DBRs[4, 5]. In addition, semiconductor DBRs can be used in vertical cavity surface emitting lasers to improve the spectral characteristics.

GaN LEDs are promising for general lighting, and GaN based optical lasers are a crucial part for high-density digital versatile discs. Nitride based DBRs could improve the properties of GaN LEDs[6, 7] and GaN based lasers[8]. The AlGaN/GaN DBR is one of the general nitride DBRs demonstrated by metal-organic chemical vapor deposition (MOCVD)[9] or molecular beam epitaxy (MBE)[10, 11]. The key to obtaining a desired DBR is to control the thickness of every period and Al composition in the AlGaN layer exactly. Schenk[12] monitored an AlGaN/GaN DBR using laser reflectometry during the growth process. In his experiment, the wavelength of the reflectometry is the same as the peak wavelength of the designed AlGaN/GaN DBR. Thus the evolution of *in situ* reflected intensity could reflect the evolution of reflectivity of the AlGaN/GaN DBR at the design's peak wavelength in real time. Schenk shows that *in situ* reflectivity is a viable tool to control the growth for nitride-based DBRs. But in many situations, the wavelength of laser reflectometry equipped in MOCVD systems is different from the reflection peak wavelength of the designed DBR, which means that the *in situ* reflectivity is not the real reflectivity of the DBR at the design's peak wavelength. If we want to make best use of the laser reflectometry tool, we have to extract information from the evolution of *in situ* reflected intensity.

In this paper, the relation between the evolution of *in situ* reflected intensity and the *ex situ* reflectivity spectrum of an AlGaN/GaN DBR is first discussed in theory. Then two kinds of AlGaN/GaN DBRs are grown by MOCVD to compare with the simulation. Finally, a DBR LED is successfully grown by MOCVD with the help of 633 nm laser reflectometry.

2. Simulations

In this paper, the DBR structure was designed to have a peak reflectivity at a wavelength of 530 nm based on $Al_xGa_{1-x}N$/GaN quarter-wave (QW) materials. In other words, the thickness d of one period in the $Al_xGa_{1-x}N$/GaN DBR is equal to the sum of d_{GaN} and d_{AlGaN}, where d_{GaN} is the GaN QW thickness for λ = 530 nm, $d_{Al_xGa_{1-x}}N$ is the $Al_xGa_{1-x}N$ QW thickness for λ = 530 nm. The wavelength of laser reflectometry in the MOCVD system is 633 nm. The transfer matrix method was used to simulate the evolution of *in situ* reflectivity and *ex situ* reflectivity[13]. For incident light λ, the matrix at layer k is written as

$$\boldsymbol{M}(k) = \begin{pmatrix} \cos g_k & \mathrm{i}u_k^{-1} \sin g_k \\ \mathrm{i}u_k \sin g_k & \cos g_k \end{pmatrix}, \tag{1}$$

in which

$$\begin{aligned} g_k &= 2\pi\lambda^{-1}(n_k d_k \cos\theta_k), \\ u_k &= \begin{cases} n_k \cos\theta_k, & \text{s polarized,} \\ n_k \sec\theta_k, & \text{p polarized,} \end{cases} \end{aligned} \tag{2}$$

θ_k, d_k, and n_k are the incident angle, layer thickness and

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† Corresponding author. Email: hlr5649@163.com
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Table 1. Parameters of structures A, B and C.

Structure type	d_{GaN} (nm)	$d_{Al_xGa_{1-x}N}$ (nm)	d (nm)
Structure A	45 (< QW_{GaN})	57 (= $QW_{Al_{0.1}Ga_{0.9}N}$)	102
Structure B	55 (< QW_{GaN})	56 (< $QW_{Al_{0.1}Ga_{0.9}N}$	111
Structure C	67 (> QW_{GaN})	56 (< $QW_{Al_{0.1}Ga_{0.9}N}$)	123

refractive index at layer k, respectively. The matrix $\boldsymbol{M}$ for the whole DBR is expressed as

$$\boldsymbol{M} = \begin{pmatrix} m_{11} & \mathrm{i}m_{12} \\ \mathrm{i}m_{21} & m_{22} \end{pmatrix} = \prod_{k=1}^{2m} \boldsymbol{M}(k), \tag{3}$$

m is period number. The reflectivity R of the DBR is shown as

$$R = \left| \frac{(m_{11} + \mathrm{i}m_{12}u_{\mathrm{s}})u_0 - (\mathrm{i}m_{21} + m_{22}u_{\mathrm{s}})}{(m_{11} + \mathrm{i}m_{12}u_{\mathrm{s}})u_0 + (\mathrm{i}m_{21} + m_{22}u_{\mathrm{s}})} \right|^2. \tag{4}$$

The refractive index of $Al_xGa_{1-x}N$ used for the reflectivity spectra simulations is expressed as[14]

$$n(h\upsilon) = \left\{ a(x) \left(\frac{h\upsilon}{E_{\mathrm{g}}} \right)^{-2} \left[2 - \left(1 + \frac{h\upsilon}{E_{\mathrm{g}}} \right)^{0.5} - \left(1 - \frac{h\upsilon}{E_{\mathrm{g}}} \right)^{0.5} \right] + b(x) \right\}, \tag{5}$$

in which $h\upsilon$ is the photon energy, E_{g} is the energy band gap, x is the Al composition of $Al_xGa_{1-x}N$, $a(x) = 9.82661 - 8.21608x + 31.59x^2$, $b(x) = 2.73591 + 0.84249x - 6.29321x^2$.

Evolutions of *in situ* reflectivity and *ex situ* reflectivity spectra of three $Al_{0.1}Ga_{0.9}N$/GaN DBR structures, A, B and C, were simulated for comparison. QW_{GaN} and $QW_{Al_{0.1}Ga_{0.9}N}$ are equal to 56 nm and 57 nm, respectively. QW_{sum}, which is the sum of QW_{GaN} and $QW_{Al_xGa_{1-x}N}$, is equal to 113 nm. The thicknesses of GaN templates under the DBR are the same in the three structures, 23 QW_{GaN} to guarantee a large reflectivity. The other parameters are shown in detail in Table 1.

The thickness d of structure A is 102 nm, which is much smaller than the desired QW_{sum}. The thickness d of structure B is 111 nm, which is only 2 nm smaller than the desired QW_{sum}. The thickness d of structure C is 123 nm, which is larger than the desired QW_{sum}.

Figures 1(a) and 1(b) are the simulated evolutions of *in situ* reflectivity and *ex situ* reflectivity spectra for structures A, B and C. The smaller peaks in Fig. 1(a) are due to the variation of total film thickness. The envelope originates from the DBR. As shown in Fig. 1(b), the reflection peaks of reflectivity spectra for structures A, B and C are 479, 525 and 580 nm, respectively. This indicates that, when the thickness d is smaller than QW_{sum}, the period of the envelope and the modulation depth are smaller. The simulated *ex situ* reflection peak of structure A is blue-shifted compared with the desired 530 nm. When the thickness d is larger in structure C, the period of the envelope and the modulation depth are larger. The reflection peak of the DBR red shifts to 580 nm compared with the desired 530 nm. Although d in structure B is only 2 nm less than QW_{sum}, the reflection peak of structure B blue shifts about 5 nm compared with the desired 530 nm. According to the simulations, the number of smaller peaks in every period of the envelope is related to the thickness d of the 1-period AlGaN/GaN DBR. When the modulated depth is higher, Al composition is higher for the samples with the same thickness d. This shows that it is possible to evaluate the properties of the DBR by the shape of the envelope in Fig. 1(a).

3. Experiments

As shown in Fig. 2, DBR samples D and E were grown in this section according to the simulated evolution of *in situ* reflectivity of structures C and B, respectively. The two samples were grown on a c-plane sapphire substrate by MOCVD (Thomas Swan Close Coupled Showerhead system). Trimethylgallium (TMGa), trimethylindium (TMIn), and ammonia (NH_3) were used as precursors for Ga, In, and N elements, respectively. The carrier gas was hydrogen or nitrogen. Before growing, the substrates were thermally baked at 1100 ℃ in hydrogen to remove surface contamination for 0.5 h. The whole growth processes were *in situ* monitored by 633 nm Thomas Swan reflectometry. The evolution of measured *in situ* reflected intensity is shown in Fig. 3(a). For sample D, the growth process proceeded according to the following sequence. After the deposition of the 30 nm low growth temperature GaN nucleation layer at 530 ℃ (from time T_{A} to time T_{B} in Fig. 3(a)), 1.6 μm high temperature (HT) GaN was grown at 1050 ℃ (from time T_{B} to time T_{C} in Fig. 3(a)), followed by 30-period QW AlGaN/GaN DBR (from time T_{C} to time T_{D} in Fig. 3(a)). During the whole growth process of the DBR, the growth pressure and temperature were maintained at 200 Torr and 1050 ℃, respectively. For sample E, the growth temperature for GaN and AlGaN were 1030 and 1080 ℃, respectively; the growth pressures for GaN and AlGaN were 260 and 130 Torr, respectively. The DBR growth time for sample E ranges from time T_{C} to T_{E}. It takes more time to grow sample E due to the variation of growth temperature between the GaN layer and the AlGaN layer.

Figure 3(a) is the measured evolution of laser reflected intensity of samples D and E. The number of minor peaks in one envelope period for sample D is 13, which is the same as the simulated result of structure C in Fig. 1(a). The number of minor peaks in one envelope period for sample E is 5.5, which is the same as the simulated result of structure B in Fig. 1(a). These results mean that the thickness d of sample D is the same as structure C, and the thickness d of sample E is the same as structure B. As shown in Fig. 3(b), reflectivity spectra of samples D and E were measured *ex situ* at normal incidence, using a Perkin-Elmer Lambda 35 spectrometer. The reflection-peak wavelengths of samples D and E are 580 and 525 nm, respectively, which agree well with the simulated reflection-peak wavelengths of structures C and B in Fig. 1(b). The modulation depths of envelopes of samples D and E in Fig. 3(a) are larger than those of structures C and B, respectively. Also, the peak intensities of samples D and E are higher than those of structures C and B, respectively. The simulated and experimental results of modulation depth and peak intensity come down to one point: the Al composition x in sample D is higher than structure C; the Al composition x in sample E is higher than

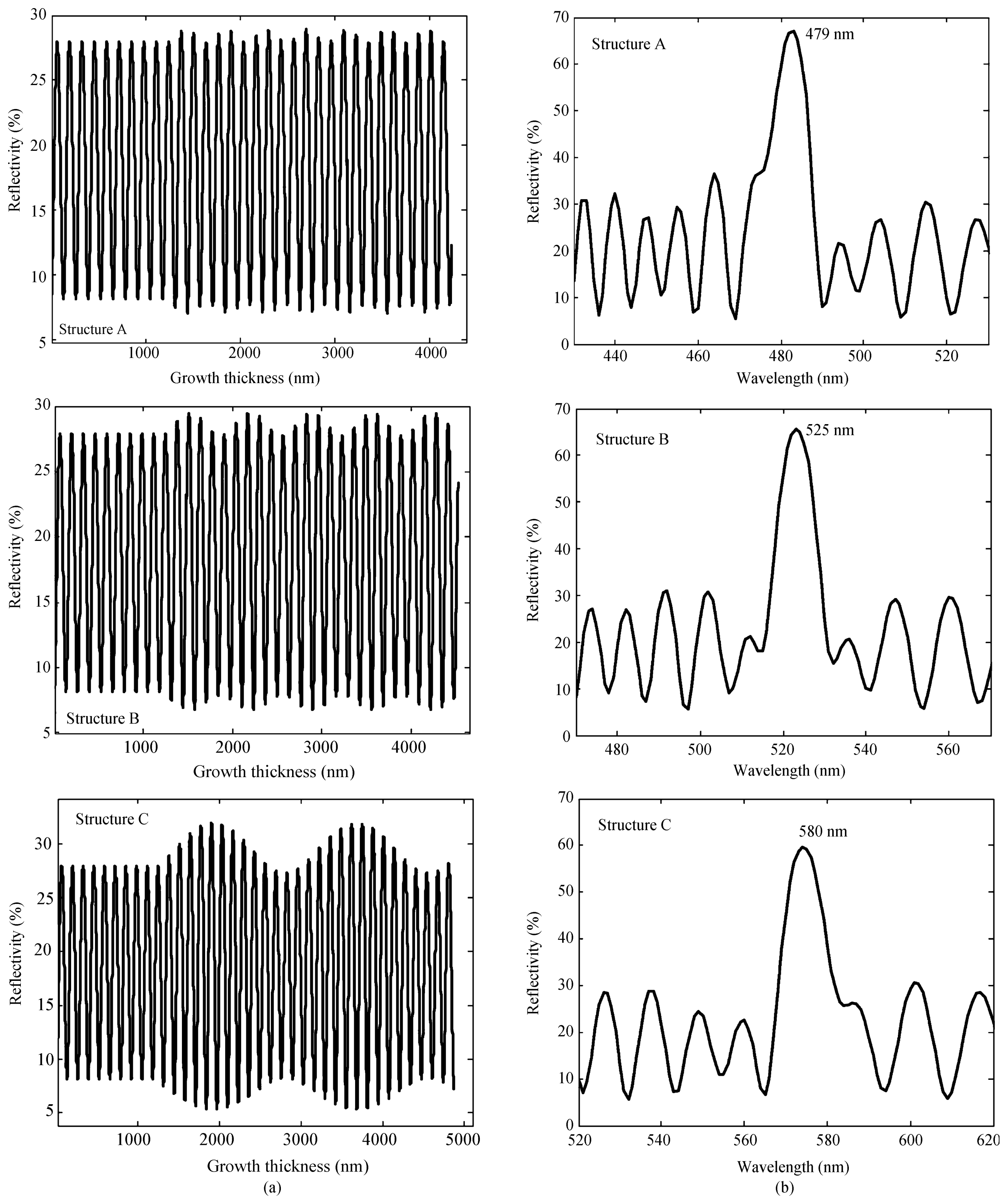

Fig. 1. (a) Simulated evolution of the reflected laser intensity during DBR growth. (b) Simulated reflectivity spectra of DBR.

structure B.

High resolution X-ray diffraction (HRXRD) measurements, performed by a Philips X-pert double crystal X-ray diffractometer with Ge (220) single crystal, were carried out to evaluate structure parameters. Figure 4 shows the $2\theta/\omega$ HRXRD scans of samples D and E for the (0002) reflection. The upper thicker curves are the experiment results. The highest peak originates from the substrate. Keen-edged satellite peaks on the left of the substrate peak are related to the GaN layer in the AlGaN/GaN DBR, and keen-edged satellite peaks on the right of the substrate peak are related to the AlGaN layer in the AlGaN/GaN DBR. The discernable satellite peaks in both samples show their good growth quality. The lower fine lines are simulated curves based on the Philips X'pert epitaxy and Smooth fit program. Al composition x, $Al_xGa_{1-x}N$ and GaN DBR thicknesses were estimated to be around 0.11 and

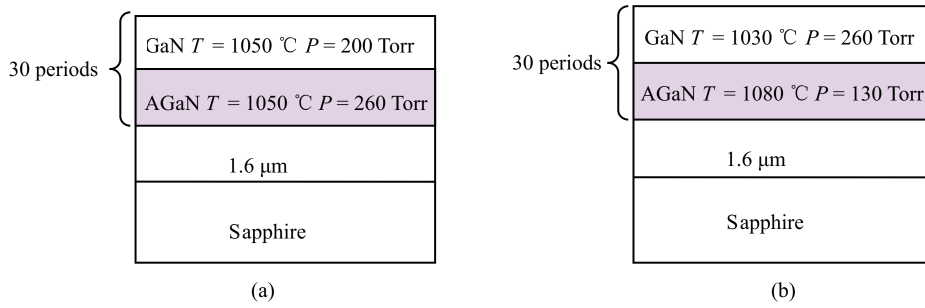

Fig. 2. (a) Sample D. (b) Sample E.

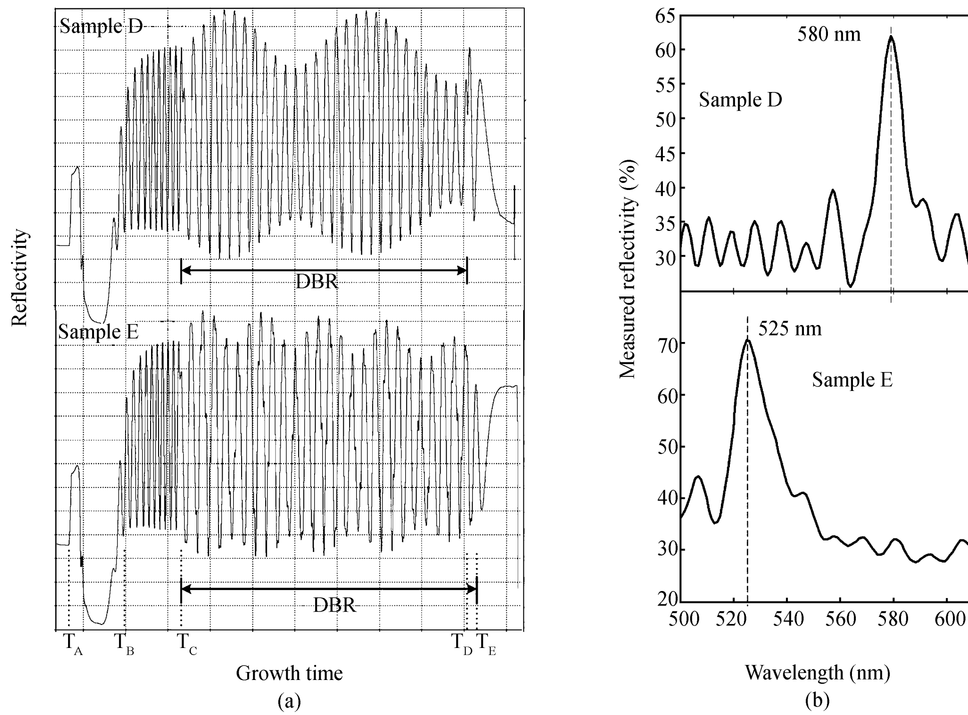

Fig. 3. Evolution of *in situ* reflectivity. (b) Measured *ex situ* reflectivity.

Table 2. HRXRD results for DBR in samples D and E.

Sample type	d_{GaN} (nm)	$d_{Al_xGa_{1-x}N}$ (nm)	x	d (nm)
Sample D	67	56	0.11	123
Sample E	55	56	0.20	111

56 nm and 67 nm for sample D, and 0.2 and 56 nm and 55 nm for sample E, which are listed in Table 2. According to the HRXRD simulated results, the thicknesses d for samples D and E are 123 and 111 nm, respectively, which are in agreement with the evaluations from evolution of *in situ* reflected intensity.

Then two dual wavelength multiple-quantum well LEDs were grown by MOCVD: one is sample F with a 20-period DBR, the other is sample G without a DBR. The schematic structures of the two LEDs are shown in Fig. 5. The growth processes were monitored by 633 nm laser reflectometry, and the measured evolution of the *in situ* reflected intensity of sample F is shown in Fig. 6(a). The growth process for the GaN template is from time t_A to time t_C, which is the same as the growth process from time T_A to time T_C in Fig. 3(a). The DBR in sample F was grown from time t_C to time t_D in Fig. 6(a). The growth parameters for the DBR in sample F are the same as those in sample D except that the growth time for the GaN layer was reduced from 170 to 142 s. After growing the DBR, n-type GaN was grown (from time t_D to time t_E in Fig. 6(a)), and then four green $In_xGa_{1-x}N$ quantum wells and barriers were deposited at 700 and 930 ℃, respectively. After the growth of the green quantum wells, two blue $In_yGa_{1-y}N$ quantum wells and barriers were deposited at 760 and 1070 ℃, respectively. The growth process for all the quantum wells ranges from time t_E to time t_F in Fig. 6(a). Finally, a 40-nm-thick p-$Al_{0.2}Ga_{0.8}N$ layer, followed by a 200-nm p-GaN layer (both at 930 ℃), were grown. The structure of sample G is the same as sample F except that there is no DBR between the high temperature GaN and the n-type GaN.

Figure 6(b) is the simulated evolution of reflected intensity of QW AlGaN/GaN DBR during the growth process, in which d_{GaN} = 56 nm, d_{AlGaN} = 57 nm. During DBR growth time ranging from t_C to t_D, there are 6 minor peaks in one envelope period in Fig. 6(a), which is the same as the peak number in

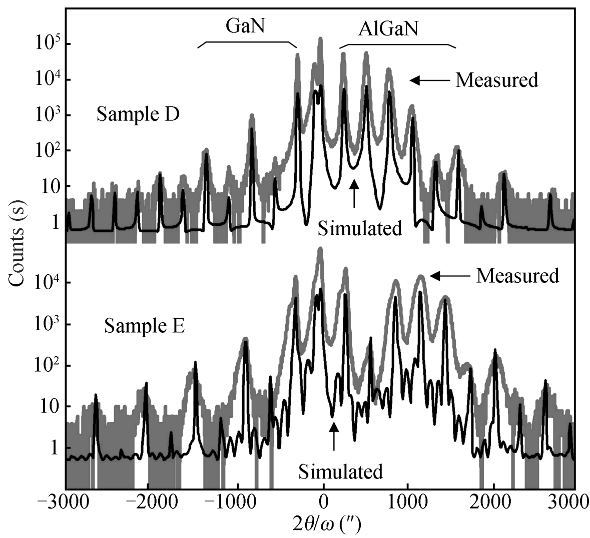

Fig. 4. HRXRD $2\theta/\omega$ scans for the (0002) reflection of samples D and E.

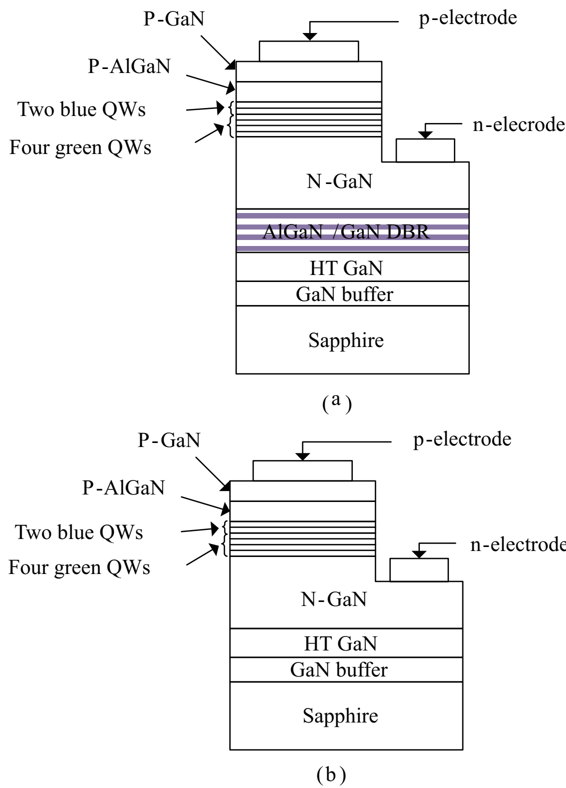

Fig. 5. (a) Schematic structure of sample F. (b) Schematic structure of sample G.

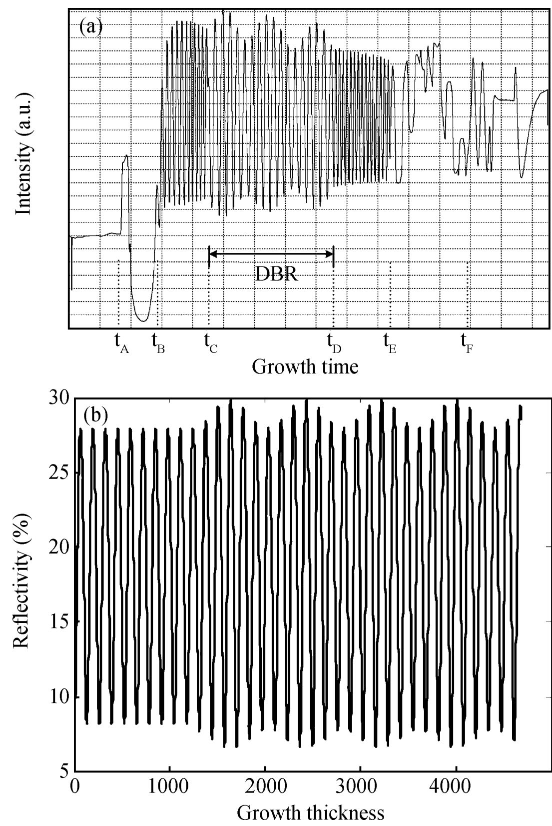

Fig. 6. (a) Measured evolution of *in situ* reflectivity of sample F. (b) Simulated evolution of *in situ* reflectivity of sample F.

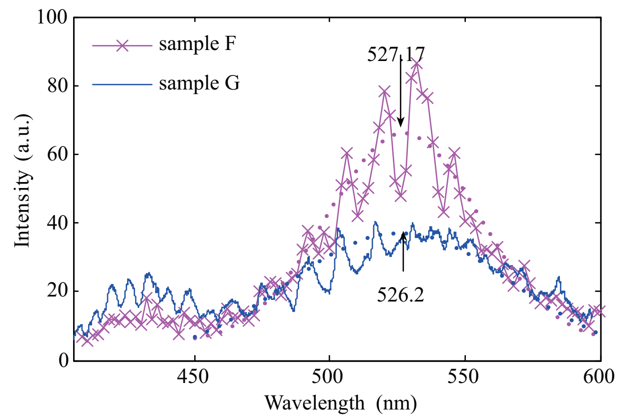

Fig. 7. Room temperature PL spectra of samples F and G (the dotted lines are Gaussian fitting).

one envelope period in Fig. 6(b). According to the simulation, the reflection peak wavelength of the DBR is estimated to be around 530 nm.

Room temperature photoluminescence (PL) was used to analyze the optical properties of samples F and G; a He–Cd laser operating at 325 nm was used for excitation source. The solid lines in Fig. 7 give the PL spectra of the two LED samples, whereas the dotted lines are Gaussian fitting curves. The small multiple peaks in the measured PL spectra are due to the Fabry–Perot effect (multi-wavelength interference between the GaN/air interface and GaN/sapphire interface or AlGaN/GaN DBR interface). After eliminating the multi-peak effect by Gaussian fitting, a major and a minor peak can be observed in both samples. The minor peak around 440 nm originates from the blue quantum wells. The major peak is due to the emission of the four green quantum wells at the bottom of the epitaxial structures. The major peak wavelength for sample F is 527.17 nm, while the major peak wavelength for sample G is 526.2 nm. The PL intensity in the green region of sample F is much higher than sample G, which is believed to be due to the enhancement by the DBR. This proves that the peak wavelength

of DBR reflection in sample F is close to our desired 530 nm.

4. Conclusions

We have studied *in situ* growth monitoring of AlGaN/GaN distributed Bragg reflectors with a reflection peak wavelength at 530 nm using 633 nm laser reflectometry in theory and experiments. The theoretical and experimental results show that although the laser wavelength is different from the peak wavelength of AlGaN/GaN DBR reflection, it is still possible to evaluate the DBR structure parameters from the period thickness of the DBR and the modulation depth of the envelope of *in situ* reflectivity spectra. An AlGaN/GaN DBR LED was successfully grown, and the peak reflected wavelength is in our design region. With the help of laser reflectometry, it is easy to obtain DBR information in real time.

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