Analysis of the anisotropy in an *m*-plane GaN film via HVPE on a γ-$LiAlO_2$ substrate*

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Abstract: A 0.09 mm *m*-plane GaN film is deposited via hydride vapor phase epitaxy (HVPE) on a γ-$LiAlO_2$ substrate. To research the anisotropy between directions with different angles with the *c*-axis in the *m* plane, photoluminescence (PL) measurements were carried out. The results show that the electronic transition was influenced by the electric field along the *c*-axis, which results in an obvious anisotropy, but the influence was weakened by the hexagonal symmetry along the *c*-axis.

Key words: γ-$LiAlO_2$; GaN; HVPE; anisotropy; PL spectra
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1. Introduction

In recent years, because of its wide band gap and electron transitions in the form of direct transition, wurtzite GaN has been considered very suitable for the manufacture of semiconductor optical devices, such as blue-light diodes and UV detectors. Usually, the substrate for epitaxial growth of GaN is Al_2O_3 (0001), SiC (0001) or other material with heteroepitaxy along the [0001] direction of GaN[1]. The *c*-axis, as a polarity axis, of wurtzite GaN is along the [0001] direction, and thus researchers began to study γ-$LiAlO_2$ (100) as a substrate with heteroepitaxy normal to the $(1\bar{1}00)$ plane of GaN, which is referred to as the *m*-plane. Along this direction, nonpolarity is exhibited, so we can avoid the generation of spontaneous polarization and piezoelectric polarization[2–4]. Because of the absence of electric field along the $[1\bar{1}00]$ direction, the material demonstrates excellent optical properties and has great advantages when used in optoelectronic devices. In this paper, GaN film grown on $LiAlO_2$ substrates via HVPE has been carried out with a photoluminescence spectra (PL) test to analyze the anisotropy in the *m*-plane of the GaN film.

2. Experiment

We grew 0.09 mm *m*-plane GaN on the (100) plane of γ-$LiAlO_2$ with the HVPE system designed by our lab. After growth, the substrate cracked into small pieces and the *m*-GaN film was peeled off automatically. We tested the free-standing film with X-ray diffraction (XRD) to confirm its uniform orientation. In order to observe the optical properties, we tested the film with polarized Raman spectra and room temperature polarized photoluminescence spectra. We carried out the Raman test at room temperature with Jobin Yvon U1000 Raman spectroscopy, and used an Ar^+ laser device with a wavelength of 514.5 nm. In the PL test, we used a He–Cd gas laser device with output wavelength of 325 nm at room temperature; the polarization was varied in the *m*-plane by 15° per test from 0°, parallel to the *c*-axis, to 90°, perpendicular to the *c*-axis.

3. Results and discussion

We carried out the XRD test with two configurations: with the scattering plane parallel (ϕ = 0°) and perpendicular (ϕ = 90°) to the *c*-axis, respectively. The ω–2θ scan result that is plotted in Fig. 1 revealed only $(10\bar{1}0)$ reflection of GaN, from which we could tell the film was uniformly oriented. The full width at half maximum (FWHM) of the $(1\bar{1}00)$ reflection was 2.0174° when ϕ = 90° while it was 0.3802° when ϕ = 0° as shown in Fig. 2. This asymmetry in rocking curve width is a consequence of the in-plane anisotropy of the crystalline mosaic.

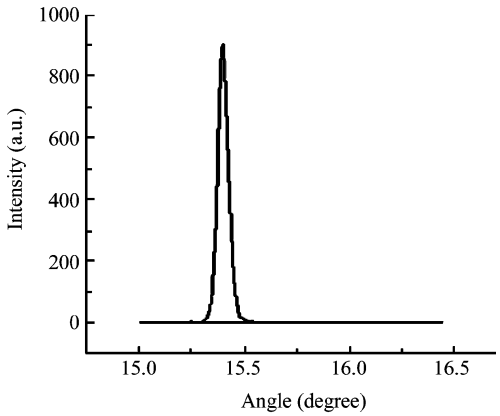

Fig. 1. ω–2θ scan of the $(10\bar{1}0)$ plane.

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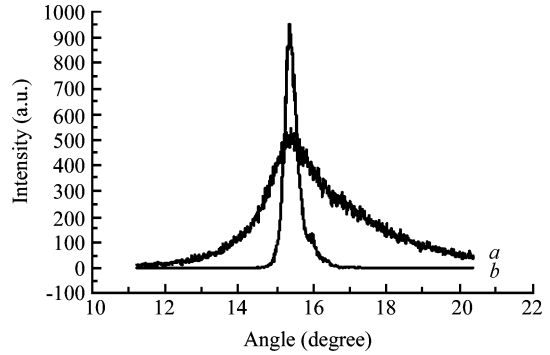

Fig. 2. ω scan of the $(10\bar{1}0)$ plane. Curve *a*: $\phi = 90°$; Curve *b*: $\phi = 0°$.

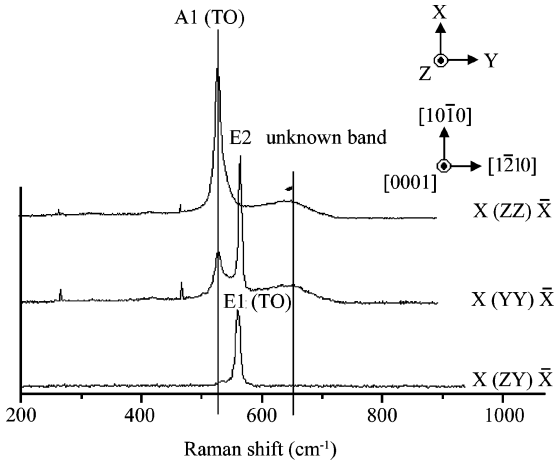

Fig. 3. Results of different polarization configurations $(X(YY)\overline{X}$, $X(ZZ)\overline{X}$, and $X(ZY)\overline{X})$ of the film′s Raman test. The orientations of *X*, *Y*, *Z* are parallel to $[10\bar{1}0]$, $[1\bar{2}10]$, and [0001] respectively. The result is in accord with the selection rule, and an unknown wide band appears in the $X(YY)\overline{X}$ and $X(ZZ)\overline{X}$ configurations.

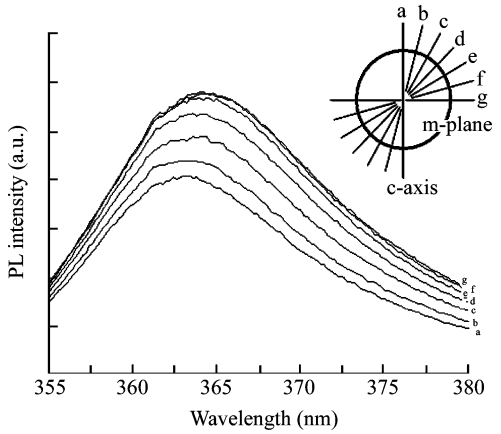

Fig. 4. PL spectra with different angles between the polarizations and the *c* axis. *a*: 0°; *b*: 15°; *c*: 30°; *d*: 45°; *e*: 60°; *f*: 75°; *g*: 90°.

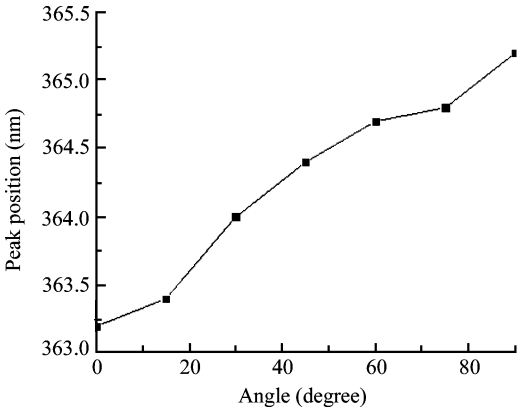

Fig. 5. Peak positions of PL spectra with different angles of *m*-GaN.

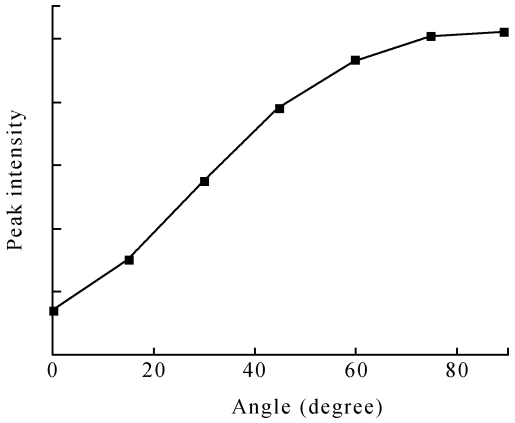

Fig. 6. Peak intensities of PL spectra with different angles of *m*-GaN.

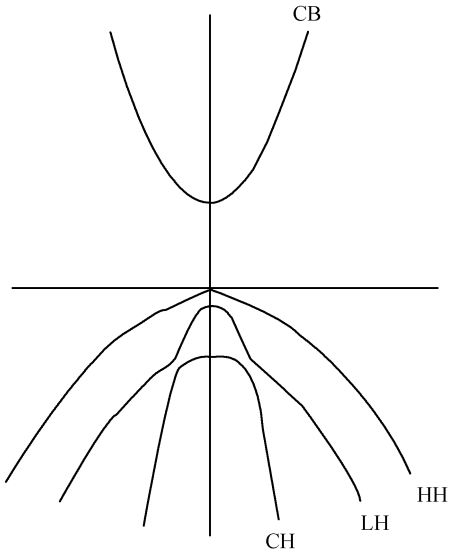

Fig. 7. Dispersion curve of GaN crystalloid.

Figure 3 is the Raman spectra of the *m*-plane sample with three different scattering configurations. The results are consistent with the selection rule, and there is an absence of the E2 phonon in the $X\ (ZZ)\overline{X}$ geometry which indicates that the sample is a single-domain *m*-plane film. Furthermore, we can see the in-plane orientation of the *c*-axis from this result: the orientations of *X*, *Y*, *Z* are parallel to $[10\bar{1}0]$, $[1\bar{2}10]$, and [0001] respectively.

Figure 4 shows the result of the PL spectra with different angles between the *c*-axis and the polarization. We can see that the bigger the angle changes, the longer the wavelength and higher intensity of the spectra peaks become. Figure 5 shows the relationship between the angle and the wavelength, while Figure 6 shows that between the angle and the intensity.

With the valence bands calculated by the $\boldsymbol{k \cdot p}$ method and the parabolic conduction band, we can get the dispersion curve as shown in Fig. 7. The spontaneous emission spectra can be simulated with the spontaneous emission rate expressed

as follows:

$$R_{\rm sp}(n,\,E) = \frac{\tilde{n}e^2E}{\pi m_0^2\varepsilon_0\hbar^2c^3} \times \int_{k_x=-\infty}^{\infty}\int_{k_y=-\infty}^{\infty}\int_{k_z=-\infty}^{\infty} |M|^2F(k)f_{\rm c}(k) \times\left[1-f_{\rm v}(k)\right]\frac{{\rm d}k_x{\rm d}k_y{\rm d}k_z}{4\pi^3}, \quad (1)$$

$$|M|^2 = \sum_{i=x,\,y,\,z}|nc_ia_i|^2|M_{\rm b}|^2, \quad (2)$$

$$|M_{\rm b}|^2 = \left(\frac{m_0^2}{m_{\rm e}}-1\right)\frac{E_{\rm g}(E_{\rm g}+\Delta_1+3\Delta_2)+2\Delta_1\Delta_2}{2(E_{\rm g}+2\Delta_2)}. \quad (3)$$

In these expressions, a_x , a_y , and a_z are unit vectors for each dipole of $|x\rangle$, $|y\rangle$, and $|z\rangle$; c_x , c_y, and c_z are mixing ratios of p functions along each axis, and x, y, and z represent the directions $[1\bar{2}10]$, $[1\bar{1}00]$, and [0001]. Δ_1 and Δ_2 are the band splittings by the crystal field and spin-orbit interaction, respectively. These parameters will definitely have been influenced by the variation of the film's anisotropy, therefore changing the output result of the PL test.

In the dispersion curve of the symmetrical hexagonal wurtzite crystal, the heavy hole (HH) band can be described as its eigenstate function: $|x + {\rm i}y\rangle$ like a p-orbital function, in which the dipole is perpendicular to the wave vector $\boldsymbol{k}$[5], and thus the wave vector $\boldsymbol{k}$ fixes the z-axis, which is the c-axis direction[6]. In wurtzite GaN, because of the difference in negative electricity between Ga atoms and N atoms, positive and negative charge centers are separated in the Ga–N covalent bond, resulting in the presence of an electric field along the c-axis in the crystal. Therefore the $\boldsymbol{k}$ vector is principally bound in that direction, and after overlaying of all the p function, there is a polarity[7, 8]. The m-plane which contains the c-axis is a polar plane, and the polarized PL spectra clearly show their anisotropy.

When the influence of the electric field along the c-axis is considered, the spectra with the angle of 0° demonstrate the transition between the crystal field split-off band (SCH) and the conduction band. The SCH band consists of $|z\rangle$ states like an atomic s-orbital function, and the spectra with angle 90° demonstrate the transition between the $|x\rangle$ part of the HH band as well as the light hole (LH) band and the conduction band. The two bands consist of $|x + {\rm i}y\rangle$ states[8, 9]. Because the state amount in the SCH band is smaller than that the HH and LH, the 0° intensity is lower, and because the energy of SCH is lower, the PL peak wavelength is shorter.

At the same time, we can see that in all cases of the different angles, changes between wavelengths and intensities of the PL peak are not particularly evident, and especially in the large angles, the PL spectra curves almost coincide. This shows that performance of in-plane anisotropy of the crystal is largely inhibited due to the hexagonal symmetry of the wurtzite crystal, resulting in a weakened anisotropy in the m-plane. In the crystal momentum with hexagonal symmetry, the dipole of HH and LH is always normal to the wave vector $\boldsymbol{k}$. With the wave vector $\boldsymbol{k}$ transmitting in all directions in the crystal, the total polarizations offset each other, and the result tends to be isotropic[10]. The distribution rate of the components of the dipole on each coordinate axis undergoes a certain amount of change because of the symmetry of the crystal, and the polarity along the c-axis is weakened. The fact that the wavelength changes very little with the angle shows the distribution changes: when the polarization is 0° or 90°, the states of the dipole do not totally stay in $|z\rangle$ or $|x+{\rm i}y\rangle$; meanwhile the transmission is not totally from SCH or HH and LH to the conduction band, but instead is mixed. This shows that in our GaN sample, the hexagonal symmetry was a more dominant influence on the optical anisotropy than the electric field along the c-axis.

4. Conclusion

An m-plane GaN film sample grown on a γ-$LiAlO_2$ substrate with the HVPE system is tested with polarized PL spectra to analyze its anisotropy. The results with different angles between the c-axis and the polarization direction reveal that the electron transition is influenced by the built-in electric field potential along the c-axis in the sample, resulting in an obvious anisotropy. However, the anisotropy is weakened by the hexagonal symmetry on the pivot of the c-axis of the GaN crystal.

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