Abnormal polarization switching phenomenon in a-plane $Al_xGa_{1-x}N$

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Abstract: The optical polarization properties of *a*-plane $Al_xGa_{1-x}N$ films have been investigated by polarization-dependent photoluminescence (PL). The degree of polarization decreased with increasing the Al composition, and the main optical polarization direction switched from $\varepsilon \perp c$ to $\varepsilon // c$ at about $x = 0.07$ due to the valence band switching, representing that the optical transition energy of $\varepsilon // c$ is surpassing that of $\varepsilon \perp c$. However, with the Al composition larger than $x = 0.1$, the higher energy optical transitions of $\varepsilon // c$ exhibited the stronger PL intensity, opposite to the normal situations that higher energy states commonly have weaker PL intensity than the lower energy states. We utilized the 6 × 6 *k.p* model and the lambertian-like radiation pattern assumption to explain this abnormal optical polarization switching behavior in the *a*-plane $Al_xGa_{1-x}N$ layers and obtained good agreement with the experimental results.

OCIS codes: (260.5430) Polarization; (250.5230) Photoluminescence.

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1. Introduction

The conventional *c*-plane Gallium-nitride (GaN)-based multiple quantum well (MQW) structure suffers from the quantum confinement Stark effect (QCSE) as a result of the existence of the strong polarization, leading to the degradation of the carrier recombination transition rate, the red-shifted peak emission and the reduction of oscillator strength, which significantly limited the device performance [1–3]. The growth along non-polar orientations such as [11–20] *a*-plane [4–6] and [1-100] *m*-plane [7–9] has been proved to diminish or even eliminate the polarization-induced effects. In addition, the growth with non-polar orientations usually accompanies the strong polarized light emission. The top valence-band states are strongly modified by the lattice mismatch-induced in-plane anisotropic strain, so that the energy splitting and polarization selection of transitions have been obtained [10,11]. Therefore, under the influence of the strong in-plane strain, the valence-band states will separate large enough to raise the polarization ratio. Recently, the improvement and enhancement of the optical polarization for non-polar orientation materials has been widely discussed, due to the possible applications in liquid-crystal display (LCD) backlight modules utilizing the polarized light [12]. Several groups have paid efforts to investigate the effects of polarized light emission originated from the non-polar InGaN material system by theoretical simulation and experimental measurement [13–16]. Few studies reported the strain induced effect in the valence-band states and optical polarization properties of *a*-plane (Al, Ga)N material system [17,18]. In this study, we grew a series of *a*-plane $Al_xGa_{1-x}N$ films with different Al compositions to establish generalized understanding in non-polar III-group Nitrides suffering from in-plane anisotropic tensile strain. We observed the abnormal polarization switching phenomenon and utilized the 6×6 *k.p* model and lambertian-like radiation pattern to investigate the optical polarization mechanism.

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2. Experimental details

The non-polar *a*-plane $Al_xGa_{1-x}N$ films were deposited on *r*-plane sapphire substrates with different Al compositions. The films were grown at 1060 °C and 1.33×10^4 Pa by low-pressure metal-organic chemical vapor deposition (MOCVD) using the trimethylgallium (TMGa), trimethylaluminum (TMAl), and ammonia (NH_3) as sources of Ga, Al, and N in whole epitaxial process. The thickness of these films was smaller than 300 nm to avoid the formation of surface cracks. The $Al_xGa_{1-x}N$ films were varied in Al compositions x and characterized by high-resolution X-ray diffraction (XRD). For optical measurements, the polarization-dependent photoluminescence (PL) was carried out at room temperature. The excitation source was a frequency tripled Ti:sapphire laser at 266 nm, with the pulse width and repetition rate of 200 femtosecond and 76 MHz, respectively. The luminescence was dispersed by a 0.55 m monochromator with the 2400 grooves/mm grating and detected by a high sensitivity photomultiplier tube for ultraviolet-visible wavelengths. The light polarization is measured at the surface normal direction with a collection angle of 8 degrees. An ultraviolet linear polarizer was set in front of the light collection fiber to examine the polarization property. The numerical aperture (N.A.) of the optical measurement system is about 0.15. In the case of the *a*-plane sample, a polarized angle of 0 degrees is defined to be parallel to the *c*-axis ($\varepsilon // \mathrm{c}$) while the 90 degrees is along *m*-axis ($\varepsilon \perp \mathrm{c}$). The optical polarization emission was also simulated by *k.p* method for calculating the dispersion relation of the valence-band and the corresponding wave functions to explain the issue of valence-band states mixing.

3. Results and discussion

By the X-ray diffraction measurement results, Al composition x of *a*-plane $Al_xGa_{1-x}N$ films were obtained to range from 0 to 0.28. The results of asymmetric XRD reciprocal space mappings (RSM) indicated that $Al_xGa_{1-x}N$ layer was almost fully strained and had no relaxation in both *m*- and *c*-directions. The in-plane anisotropic tensile strains increased with Al composition x under the coherent growth [19]. Figure 1(a) shows the spontaneous emission spectrum of the *a*-plane $Al_xGa_{1-x}N$ films with different Al compositions ($0 \leqq x \leqq 0.28$) at room-temperature. The dominant emission peak is attributed to the near-band-edge transition. The emission peak energy shifts to higher energy with increasing the Al composition x for both polarization direction. The degree of polarization (DOP) is defined as $P=(I_\perp - I_{//})/(I_\perp + I_{//})$, where $I_\perp$ and $I_{//}$ are the PL intensity for two orthogonal polarization directions. The Al composition dependence of the DOP for *a*-plane $Al_xGa_{1-x}N$ films with polar angle φ ranging from 0 to 360 degrees is shown in Fig. 2. A distinct switch of dominant polarization direction from ($\varepsilon \perp \mathrm{c}$) to ($\varepsilon // \mathrm{c}$) can be observed with increasing the Al composition due to the valence band switching between x = 0.07 and x = 0.1, where $\mathcal{E}$ represents the electric field. The peak energy shift is defined as $\Delta \mathrm{E} = (\mathrm{E}_{\varepsilon//c} - \mathrm{E}_{\varepsilon \perp c})$, where $\mathrm{E}_{\varepsilon//c}$ and $\mathrm{E}_{\varepsilon \perp c}$ are the peak energy for the polarization directions of $\varepsilon // \mathrm{c}$ and $\varepsilon \perp \mathrm{c}$, respectively. The DOP and $\Delta \mathrm{E}$ of the investigated *a*-plane $Al_xGa_{1-x}N$ films with different x are listed in Table 1. Interestingly, not only the dominant polarization direction is switched as increasing the Al composition, but the main PL intensity is switched from the lower optical energy level transition ($\mathrm{E}_{\varepsilon \perp c}$) to higher optical energy level transition ($\mathrm{E}_{\varepsilon//c}$) as shown in Fig. 1(a). This abnormal polarization switching phenomenon is opposite to the normal situations that higher energy states commonly have weaker PL intensity than the lower energy states due to the carrier distribution and probability of recombination of electrons and holes.

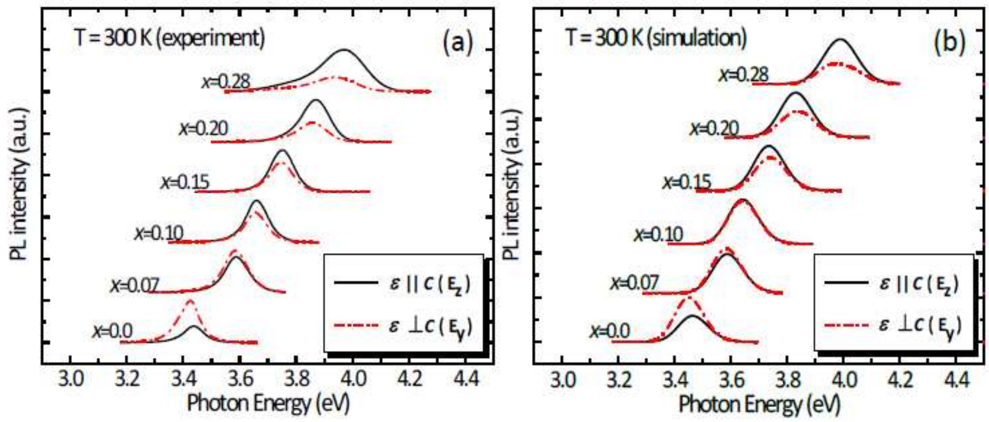

Fig. 1. (a) Experiment measurements and (b) simulation calculation of the PL intensity and spontaneous emission rate of *a*-plane $Al_xGa_{1-x}N$ films with the different aluminum compositions.

The effect of polarization switching in *c*-plane $Al_xGa_{1-x}N$ films has been investigated previously. K. B. Nam *et al.* presented that the emission intensity with polarization of $\varepsilon \perp c$ as well as the DOP decreases with increasing x [20]. Comparing with our results, *a*-plane $Al_xGa_{1-x}N$ films were also exhibited similar switching phenomenon, but the distinct difference of the emission peaks with the different polarization directions was only observed in *a*-plane $Al_xGa_{1-x}N$ films. Therefore, the unusual polarized light properties in *a*-plane $Al_xGa_{1-x}N$ films were discussed further.

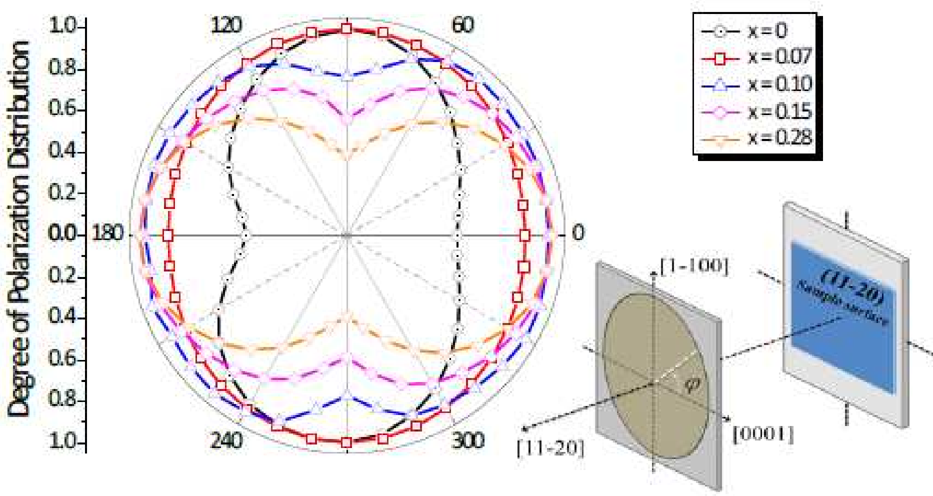

Fig. 2. The degree of polarization (DOP) of *a*-plane $Al_xGa_{1-x}N$ films with increasing Al composition x as a function of polarization angle starting at c-direction.

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Table 1. The degree of polarization and the difference of peak energy for *a*-plane $Al_xGa_{1-x}N$ films are shown

Aluminum Composition	0 (GaN)	0.07	0.10	0.15	0.20	0.28
DOP (Experiment)	33%	6%	−12%	−25%	−38%	−47%
DOP (Simulation)	27%	4%	−5%	−24%	−39%	−42%
ΔE(meV) (Experiment)	11	6	3	3	10	23
ΔE(meV) (Simulation)	12	3	0	−4	−8	9

To investigate the light emission polarization properties and to explain the abnormal polarization switching, we applied the 6 × 6 *k.p* model to calculate the *E*-*k* dispersion relation and the wave functions of the *a*-plane $Al_xGa_{1-x}N$ layer [21]. Then the spontaneous emission spectra were calculated to compare with the results of PL measurement. Figure 3(a) and 3(b) show the energy dispersion relation of the $Al_{0.07}Ga_{0.93}N$ and $Al_{0.28}Ga_{0.72}N$ films, respectively. As we know, the anisotropic tensile strain effect will lift up the $\left|X\right\rangle$ state. Also, the crystal field force will lift up the $\left|Z\right\rangle$ state, but the influence is weaker than the strain effect. Hence, the topmost state is the $\left|X\right\rangle$ state for most cases. The second state is usually mixed with $\left|Y\right\rangle$ and $\left|Z\right\rangle$-liked states and the $\left|Z\right\rangle$-liked state will become more dominated for the second band as the aluminum composition increases due to the larger effect of the crystal field energy. The transition involving the $\left|Z\right\rangle$-liked state occurs at higher energy and is expected to be *z*-polarized ($\varepsilon /\!/ \mathbf{c}, \mathrm{E}_Z$), and the transition involving the $\left|Y\right\rangle$-liked state is expected to be *y*-polarized ($\varepsilon \perp \mathbf{c}, \mathrm{E}_\mathrm{y}$).

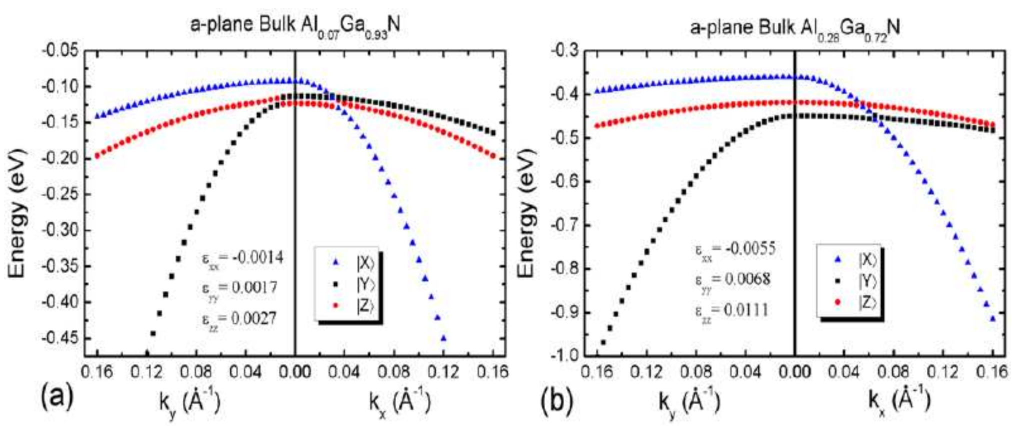

Fig. 3. The valence band energy dispersion relation under anisotropic tensile strained condition of the *a*-plane $Al_xGa_{1-x}N$ films (a) $x = 0.07$ and (b) $x = 0.28$.

Because the surface of the *a*-plane AlGaN film is rough, the light emitted from the $Al_xGa_{1-x}N$ film will reflect and scatter inside the sample structure. Hence, we assume that the emission pattern is the lambertian-like [22], having **cosθ** dependence. Therefore, the dominated *x*-polarized light corresponding to the surface normal direction of *a*-plane will contribute to the TE mode (*y*-polarized and *z*-polarized light) due to scattering projection. Since the light polarization is measured at the front of the growth direction with a collection angle of 8 degrees, the emission of each polarization component can be expressed as

$$P = \int_0^{90} \cos\theta \times 2\pi r \sin\theta r d\theta \tag{1}$$

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$$P_x = \int_{82}^{90} \cos\theta \times 2\pi r \sin(\frac{\pi}{2} - \theta)\cos(\frac{\pi}{2} - \theta) r d\theta / P \quad (2)$$

$$P_y = P_z = \int_{0}^{8} \cos\theta \times 2\pi r \sin\theta \cos\theta r d\theta / P \quad (3)$$

$$I_y^x = (I_x P_x)/2 + I_y P_y \quad (4)$$

$$I_z^x = (I_x P_x)/2 + I_z P_z \quad (5)$$

where the P_x, P_y, and P_z are the projections of *x*-polarized, *y*-polarized, and *z*-polarized light emitted along the growth direction, respectively. The I_y^x and I_z^x represent the intensity of the *y*-polarized and *z*-polarized light for the growth direction.

Figure 1(b) shows the calculated spontaneous emission spectra of the *a*-plane $Al_xGa_{1-x}N$ films. Note that the intensities of each case have been normalized because we emphasize the relative intensity of two polarized light and the peak energy. Since the topmost state of the *a*-plane $Al_xGa_{1-x}N$ is $|X\rangle$-like state, the measured light at the growth direction (*x*-direction) is mainly contributed by the recombination of the second and third bands. Moreover, the energy separation between the second and third bands increases as the Al composition increases, while the second band is getting dominated by $|Z\rangle$ state and the third band is getting dominated by $|Y\rangle$ state. Therefore, the intensity of *z*-polarized light (@003@004@005 $\varepsilon // c$ @003@005) increases as the Al composition increases, which have a good agreement with the experimental results. The DOP and ΔE of the simulated *a*-plane $Al_xGa_{1-x}N$ films with different *x* are also listed in Table 1 for comparison. It is worth noting that the emission spectra of *y*-polarized light (@003@004@005 $\varepsilon \perp c$ @003@005) and *z*-polarized light ($\varepsilon // c$)would be partly affected by the projection of the *x*-polarized light, originated from the optical transition from the conduction band to the $|X\rangle$ band as shown in Fig. 4. For the low Al composition case, the predominant optical transitions are contributed by the two topmost $|X\rangle$ and $|Y\rangle$ bands, comprising the main polarized emission along (@003@004@005 $\varepsilon \perp c$ @003@005) direction. On the other hand, for the higher Al composition case, the fractional contribution from the $|X\rangle$ band combining with that from the $|Y\rangle$ bands constitutes the weaker PL emission along (@003@004@005 $\varepsilon \perp c$ @003@005) with a smaller peak energy shown in Fig. 4(a). As a result, the spectrum shows the abnormal behavior that the stronger PL emission along ($\varepsilon // c$) belongs to a higher peak energy shown in Fig. 4(b).

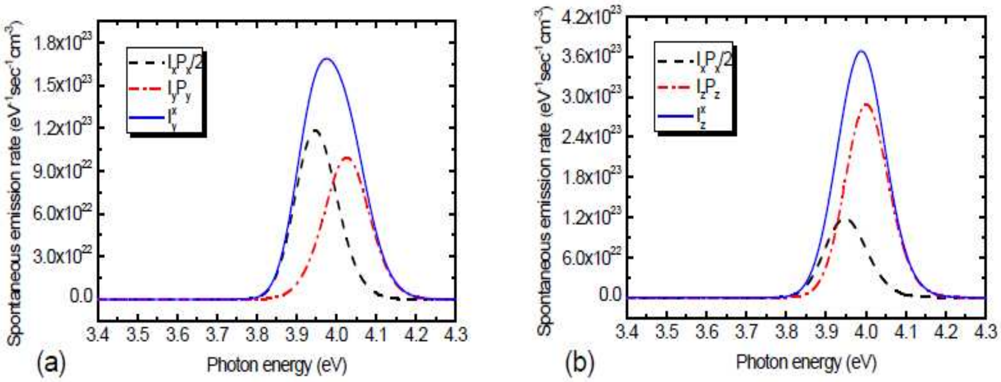

Fig. 4. (a) and (b) are the schematic illustrations of the influence of I_xP_x to the y-polarized light and z-polarized light, respectively.

4. Conclusion

We have investigated the polarization-dependent optical properties of a-plane $Al_xGa_{1-x}N$ films with different Al composition. The DOP decreases with increasing Al composition x. A distinct optical polarization switch is observed between $x = 0.07$ to $x = 0.1$ due to the valence band switching. In addition, we applied the $k.p$ method with the lambertian-like emission pattern to obtain the E-k dispersion relation and the wave functions of the a-plane $Al_xGa_{1-x}N$ layer. Base on this model, the calculation of the spontaneous emission spectra have a good agreement with the experimental results and the abnormal polarization switching phenomenon is explained.

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