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Photoreflectance and photoluminescence of thick GaN layers grown by HVPE

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Very thick (up to 100 μm) GaN layers grown by HVPE are investigated by photoreflectance (PR) and photoluminescence (PL) spectroscopies. The layers were deposited on a GaN buffer layer which was grown on a c-plane sapphire substrate by MOVPE. Both, N- and Ga-polar layers were selected to these investigations. We have observed a strong dependence of the optical properties on the polarity of GaN surface. We have obtained that the bandgap-related emission for Ga-polar layers is stronger and narrower than the emission for N-polar layers. Also, significant differences have been found in PR spectra of the two type layers. In the case of Ga-polar layer a broad PR resonance with Franz-Keldysh oscillation (FKO) related to the surface electric field (215 kV/cm) has been observed, while in the case of N-polar layer narrow resonances have been found as being predominant. No-FKO for N-polar layer indicates that the surface electric field for this layer is weak. It means that the surface barrier for N-polar GaN is much smaller than for Ga-polar GaN layer.

Keywords: GaN, HVPE, Photoreflectance, N-face, Ga-face.

1. Introduction

GaN and related III–V nitride materials have been investigated for various applications in electronic and optoelectronic devices, such as blue and green light-emitting diodes, laser-diodes, and high-power and high-temperature electronic devices [1,2]. Although the growth of group III-nitride materials has progressed rapidly, it is still difficult to obtain high-quality layers due to the large dislocations densities and deep levels inherited from the lattice mismatch between the GaN layer and the sapphire substrate. Much improved device performance could be facilitated by the development of high quality freestanding GaN substrates with tailored electrical properties. So far, the homoepitaxy of GaN layers on GaN substrate grown at high temperatures and at high hydrostatic pressures has been successfully applied to obtain high-quality GaN layers [3,4]. However, other faster and cheaper methods of the grown are still sought-after. One of the rapidly advancing sources of thick, freestanding GaN wafers is hydride vapour phase epitaxy (HVPE) [5]. This method makes possible a fast growth of GaN layers with the thickness of a few hundreds of micrometers. Such layers can be separated from the sapphire substrate by laser-induced lift-off [6,7].

Very important issue for GaN layers is the polarization of GaN surface. The c-plane sapphire on which GaN layers are grown and GaN do not share the same atomic stacking order. Consequently, the crystal direction [0001] of a GaN layer can either be parallel or antiparallel to the growth direction, leading to epilayers with two different polarities, Ga- and N-polar layers. Previous investigations showed that the two polar layers have very different structural and electrical properties [8–16]. A Ga-polar layer has a relatively smoother and more stable surface [9–10], lower impurity contamination [11], and different height of Schottky barrier [12–15]. The photoluminescence properties of Ga-polar layers also seem better than N-polar layers [16, 17]. Therefore, it is often believed that the overall quality of the Ga-polar layer is better than N-polar layers. Optical properties of both Ga- and N-polar thick GaN layers grown by HVPE are investigated by using photoreflectance (PR) and photoluminescence (PL) spectroscopies in this paper. On the basis of the analysis of PR spectra we are able to estimate the surface electric field [18,19] in GaN layers. The advantages of PR technique are its contactless character and high sensitivity even at room temperature [19,20].

2. Experiment

The thick layers were deposited by HVPE on a GaN buffer layer which was grown on a c-plane sapphire substrate in a

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vertical flow MOVPE system at atmospheric pressure [21,22]. The MOVPE growth was carried in home made MOVPE system with horizontal quartz reactor (atmospheric pressure) with RF heating. The thickness of GaN buffer layers was about 2 µm. The thick GaN layers were grown in conventional HVPE system: three-temperature zone furnace and horizontal quartz reactor. Nitrogen (6N) was used as the carried gas. GaCl was formed by the reaction of gaseous HCl (6N) and liquid Ga (6N) at 920°C. HCl (6N) was diluted by nitrogen. NH_3 (7N) was used as the source gas. Total gas flow was about 2500 ml/min. The temperature in a grown zone was kept at 1060°C. Two-step deposition method was performed. First step was the growth of GaN with a small amount of HCl (approx. 4 ml/min.) The second step growth was faster – flow of HCl was 10 ml/min. Both, Ga- and N-polar layers with the thickness of ~100 µm were selected to these investigations. The polarity of these layers was determined on the basis of the chemical etching.

The PR measurement was performed in the so-called bright configuration where the sample was illuminated by white light from a halogen lamp (150 W) serving, as a probe, beam source at near normal incidence. The reflected light was dispersed through a 0.55-m focal length single grating monochromator and detected by R647P Hamamatsu photomultiplier. For photomodulation of 300 nm line an Ar^+ laser was used as a pump beam, which was mechanically chopped at the frequency of 284 Hz. The signal was recorded by a model SR830 DSP lock-in amplifier. More details about PR set-up can be found elsewhere [20]. The PL measurement was performed using the same apparatus. The spectral resolution was kept on the level of several tenths of meV.

3. Results and discussion

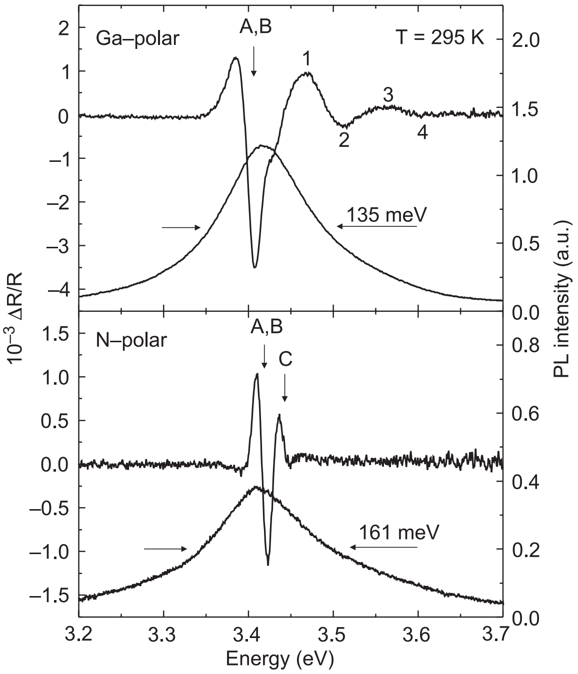

Fig. 1. Photoluminescence and photoreflectance spectra of Ga-and N-polar thick GaN layers grown by HVPE.

Figure 1 shows a comparison of the room temperature PR and PL spectra obtained for Ga-polar and N-polar GaN layers. The intensity and the full half width maximum (FHWM) of PL band related to bandgap emission change with the change in the sample polarity. The integrated intensity of emission peak for Ga-polar layer is about three times higher than the intensity for N-polar layer, and the full half width maximum of PL band increases form 135 to 161 meV with the change in the GaN surface polarity from Ga- to N-polar one. We have also measured other Ga-polar and N-polar GaN layers grown at similar conditions, and we have obtained the results very similar to these presented in Fig. 1. It means, that the quality of Ga-polar GaN layers is better than the quality of N-polar layers. This conclusion is also supported by an analysis of the sample surface preformed by using a common optical microscope as we presented in Fig. 2. We have observed less structural defects for Ga-polar layers than for N-polar ones.

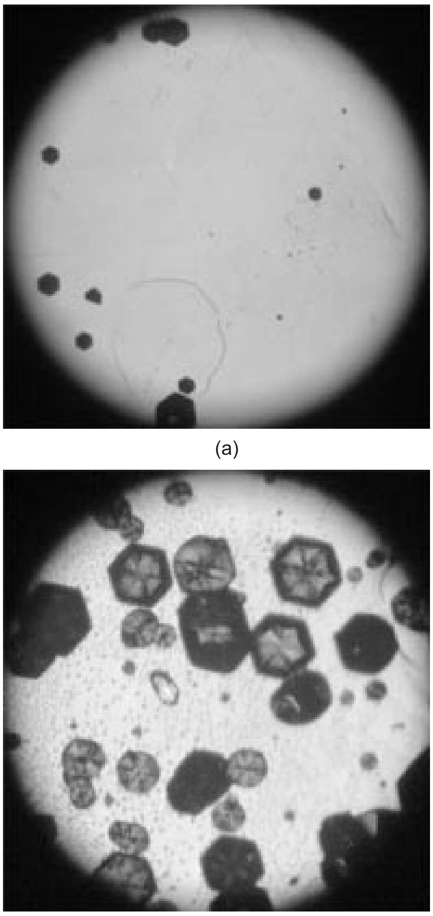

Fig. 2. A representative photo for Ga-polar (a) and N-polar (b) surfaces of GaN layers prepared by camera with a common optical microscope.

Very interesting results for the two types of GaN layers have been obtained by measuring PR spectra. As it can be seen in Fig. 1, PR spectra recorded for Ga- and N-polar

GaN layers look very different. In the case of Ga-polar GaN layer, a PR resonance with characteristic Franz-Keldysh oscillations (FKOs) is observed while in the case of N-polar GaN layer quite narrow PR lines without any oscillations are observed. The presence of FKO signal is associated with an internal electric field existing in the sample, i.e., a surface electric field or a field at interfaces existing in the structure. In the case of such a structure as our one, i.e., 100-µm thick GaN layer, it is the surface field. An electric field on GaN(HVPE)/GaN(MOVPE) interface cannot be probed because the thickness of HVPE layer is to big, about 250 times bigger than the wavelength of a probing beam. The FKO was analysed using the asymptotic expression for electroreflectance [18,19]

$$\frac{\Delta R}{R} \propto \exp\left[\frac{-2\Gamma\sqrt{E-E_g}}{(\hbar\theta)^{3/2}}\right] \times\cos\left[\frac{4}{3}\left(\frac{E-E_g}{\hbar\theta}\right)^{3/2}+\phi\right]\frac{1}{E^2(E-E_g)}, \tag{1}$$

$$F=\frac{(\hbar\theta)^{3/2}\sqrt{2\mu}}{e\hbar}, \tag{2}$$

where $\hbar\theta$ is the electro-optic energy, Γ is the linewidth, ϕ is the phase factor, F is the electric field, and μ is the electron hole reduced mass (μ = 0.2 m_e). The field estimated from the period of FKOs is 215 kV/cm (see Fig. 3). Such a huge value of the built-in electric field is typical for GaN-based structures [13,23]. We have observed PR signal similar to this one shown in Fig. 1(a) also for other Ga-polar samples. In addition, we have observed, that the intensity of the high energy part of this spectrum, i.e., the part with FKO, changes from sample to sample. Such behaviour is understood, because the intensity of FKO is very sensitive to sample quality. In the case of N-polar layer, no-FKOs indicate that the surface electric field is very weak. PR features observed for this layer could be attributed to both excitonic and band-to-band transitions. The character of the transition depends on the sample quality. In the case of high quality layers at room temperature an excitonic absorption is expected while in the case of poor quality only the band-to-band absorption is expected. Also, the built-in electric field influences the character of optical absorption. In the regime of strong built-in electric fields the band-to-band absorption is expected while for low built-in electric fields both the band-to-band (at high temperatures) and the excitonic (at low temperatures) absorption are expected. The presence of FKOs proves about the significant built-in electric field, therefore the PR feature with FKOs is associated with the band-to-band absorption and cannot be attributed to the excitonic absorption. However, it is not excluded that a feature associated with the excitonic absorption can interfere with the FKO-like PR line. Such situation is understood within the band diagram shown in Fig. 4. In the case of situation as in Fig. 4(a), i.e., a strong surface electric field, an excitonic absorption is possible far from the surface where the built-in electric field is weak. Therefore, we do not exclude such possibility for our Ga-polar layers, especially that at the energy of 3.41 eV an individual PR feature is visible. This feature could be attributed to absorption in the region of a weak built-in electric field. In the case of N-polar GaN layer, we expect a weak surface electric field as in Fig. 4(b) therefore PR resonances can be attributed to the excitonic and/or the band-to-band absorptions.

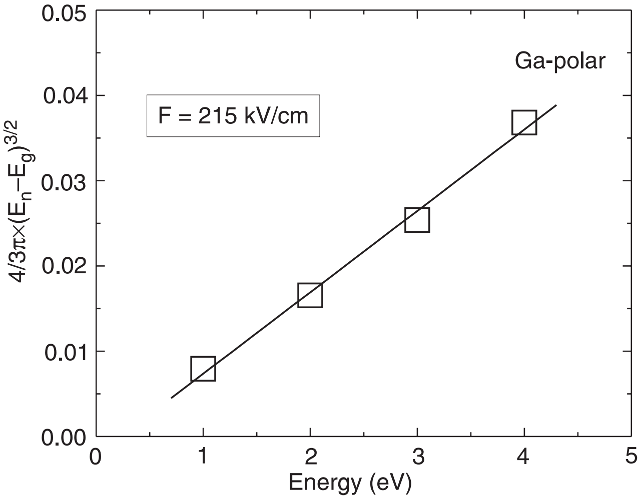

Fig. 3. Analysis of FKO observed for Ga-polar GaN layer.

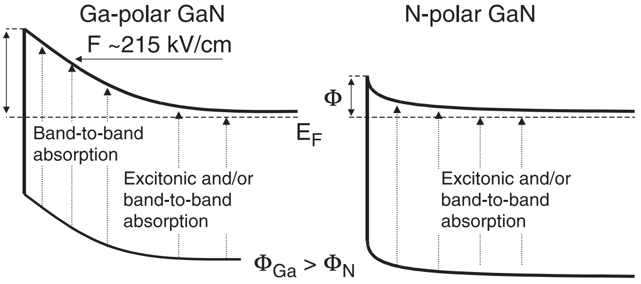

Fig. 4. Schematic band diagram for the Ga-polar (a) and N-polar (b) GaN layers.

Moreover, we observe that at the same conditions of photomodulation, the intensity of PR signal for the N-polar layer decreases of about 50% in comparison to the intensity of PR signal measured for Ga-polar layer. The intensity of PR signal is proportional to the changes in the built-in electric field. Therefore smaller intensity of PR signal, at the same conditions of photomodulation indicates different surface properties, i.e., different surface barrier ***Φ***. This conclusion is consistent with the presented in Fig. 4 bandgap diagram which assume that the surface barrier for Ga-polar GaN layer is bigger than the barrier for N-polar layer.

On the basis of PL and PR results we have concluded that the optical properties of the thick GaN layer are poor in comparison to thin GaN layers grown by MOVPE or MBE. But, we expect that the quality of such thick GaN layers will be worst than the quality of thin layers. The origin of poor quality for thick layers grown on a sapphire substrate is the 15% lattice mismatch between the substrate and the lattice constant of GaN crystal. However, the quality of the layers is sufficient for using them as a substrate for GaN homoepitaxy.

4. Conclusions

Thick Ga- and N-polar GaN layers were grown by HVPE on GaN buffer layer, which was grown on a c-plane sapphire substrate by MOVPE. The optical properties of these layers have been discussed in this paper. We have found that the bandgap-related emission for Ga-polar layer is stronger and narrower than the emission for N-polar layer. It proves about better quality of the Ga-polar layer. Moreover, we have found that PR spectra for the two types of layers are completely different. This phenomenon is attributed to different surface electric field, i.e., different surface barrier of the potential. In the case of Ga-polar layer, the surface electric field is strong (215 kV/cm), i.e., a surface barrier is high, and hence the resonance with FKO is observed in PR spectrum. For N-polar layer no-FKO is observed because the surface electric field is weak. It means that the Φ_N is significantly smaller than the Φ_{Ga}.

Acknowledgments

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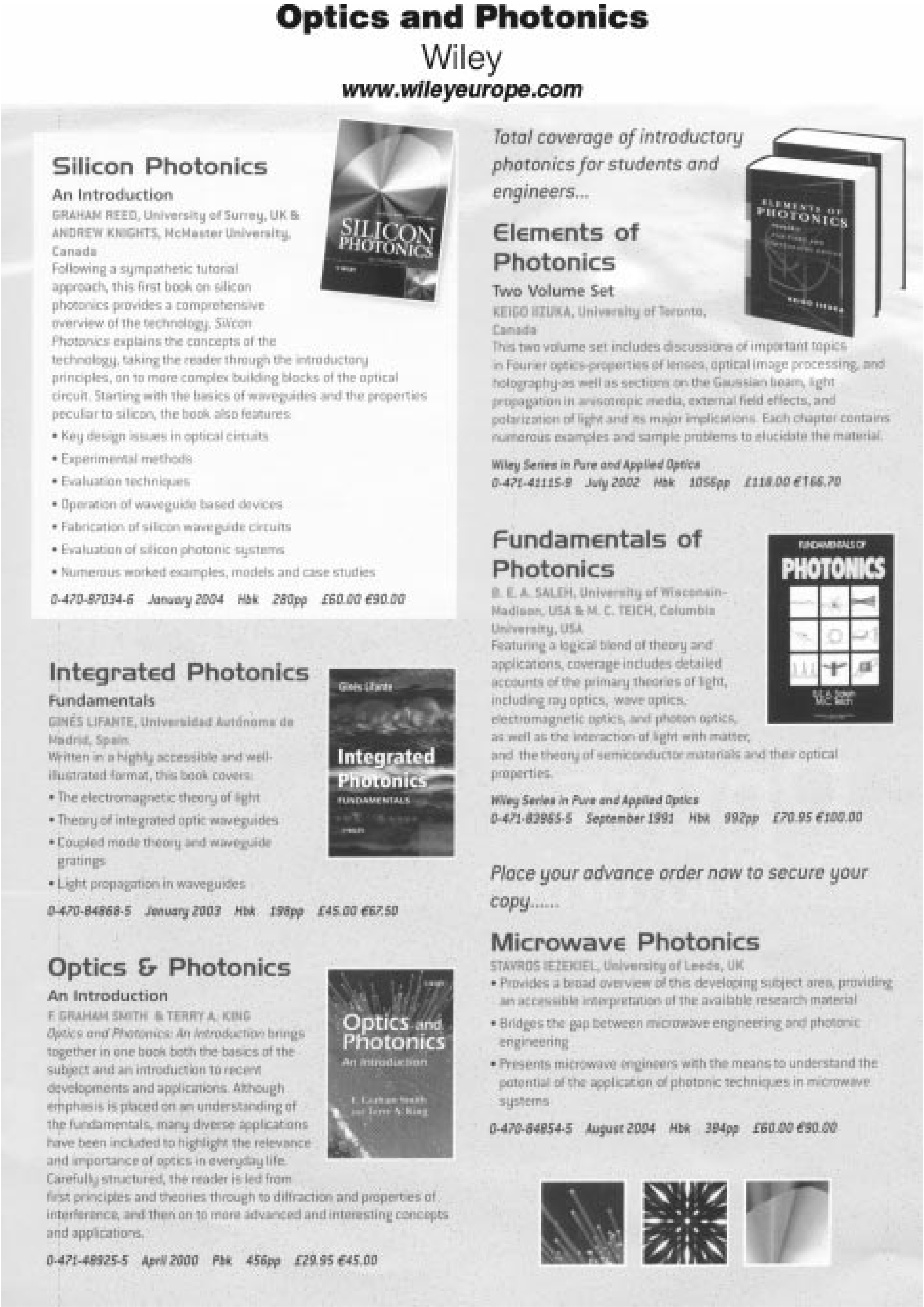

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