GaN, a new material for integrated nonlinear optics

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Abstract

This article presents gallium nitride (GaN) as a new material for integrated nonlinear optics. The GaN optical properties such as refractive indices and nonlinear coefficents were measured. The technique developped to satisfy quasi phase matching with GaN is detailed. The propagation losses in GaN waveguides epitaxially grown on sapphire substrates were determined by prism coupling. The burying of the waveguides is shown to reduce these losses. Finaly, backward propagating nonlinear interactions can be envisaged since we demonstrated submicronic modulations of the sign of the GaN nonlinear coefficient.

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Introduction

Gallium nitride, GaN, is a wide bandgap semiconductor (3.4 eV) which possesses a large transparency zone covering the visible and the near and mid infrared domains. GaN and other III-nitrides have been widely studied since the early 90s for their unique optoelectronic properties in terms of light emitters and detectors. The applications such as the first blue laser diodes[1] became possible once the p-type doping problems were solved. Nowadays, the white light emitting diodes[1,2] (based on (In,Ga)N quantum wells) are getting very competitive in terms of power and efficiency and an intensive research is going on for the future general lightening. Many other works on III-nitrides deal with high-power and high-frequency electronics. Nevertheless, very few studies concerned GaN for optics or nonlinear optics. Most of them were dedicated to the measurement or calculation of the nonlinear coefficients $c^{(2)}$ of GaN[3-5]. Indeed, GaN crystallizes in the wurtzite structure which has hexagonal symmetry and therefore is non centro-symmetric. The $c^{(2)}$ published values present a significant discrepancy but they are of the same order as the lithium niobate ones. Therefore, optical frequency conversions can be envisaged over the whole transparency zone of GaN. Recently, the first second harmonic generation (SHG) performed in periodically poled GaN by quasi phase matching (QPM) was realized from 1660 nm to 830 nm by Chowdhury *et al*[6]. Yet, the conversion efficiency was extremely low.

In this paper, we present a different approach to achieve nonlinear interactions with GaN but still based on QPM[7]. This technique which compensates artificially the dephasing introduced along light propagation by the chromatic dispersion requires a periodic modulation of the sign of the nonlinear coefficient. The modulation period L can then be adapted in principle to any nonlinear process. For instance, in the simple case of SHG with a pump wavelength at l_w, the period is given by:

$$\Lambda = \frac{l_w}{2\left|n_{2w} - n_w\right|}$$

where n_w and n_{2w} are respectively the refractive indices at the fundamental and second harmonic wavelengths. This previous equation shows that QPM requires a "perfect" knowledge of the GaN chromatic dispersion. Indeed, the QPM condition is severe *i.e.* spectrally very narrow[8], which means that for the slightest index difference, the constructive interference effect is rapidly lost. In the literature, the GaN index measurements[3,9,10] present a too important discrepancy. Therefore, we have measured the ordinary n_o and extraordinary n_e refractive indices of GaN by prism coupling between 458 and 1570 nm using several laser sources.

First, the GaN chromatic dispersion from a thin film grown on sapphire will be presented. We will also give the nonlinear coefficients measured on the same sample. Then, we will describe the process used to obtain GaN structures dedicated to QPM. We will see that the main problem encountered concerns the propagation losses. A way to lower these losses will be discussed. Finaly, we will show that it is possible to get GaN structures with submicronic periods, thus opening the way to backward nonlinear optics interactions.

Chromatic dispersion, nonlinear coefficients, and propagation losses

The hetero-structures further presented were all grown by molecular beam epitaxy (MBE) using ammonia as nitrogen precursor. In order to measure the refractive indices of GaN, we grew a 1.9 µm thick GaN film on sapphire. A 25 nm thick low temperature (400°C) buffer layer is primarily grown on sapphire. Then, the GaN film is deposited at 800°C at a growth rate of ~ 1 µm/h under N-rich conditions. This film is an asymmetric planar waveguide which is strongly multimode at visible wavelengths as regards as its thickness and the index contrasts between guide and claddings. A classic prism coupling method is employed to determine the refractive indices by measuring the effective indices of the transverse electric (TE) and transverse magnetic (TM) propagation modes allowed in the waveguide. The experimental dispersion curves are shown in figure 1.

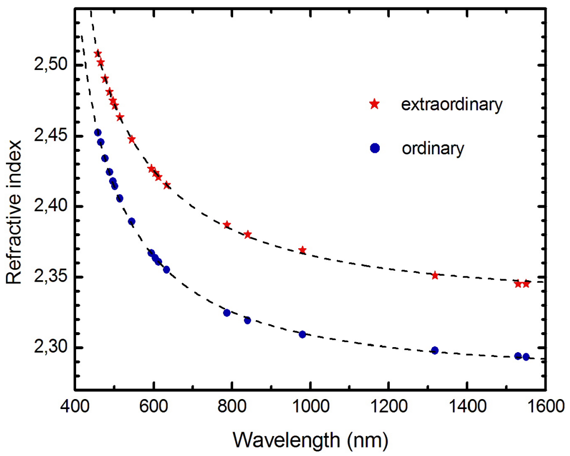

Fig. 1: Ordinary n_o and extraordinary n_e refractive indices of GaN as a function of optical wavelength. The dotted lines represent the experimental data adjustments.

These data are well adjusted with the 2-term Sellmeier-like equations given below:

$$n_o(l) = \sqrt{1 + \frac{0.298\, l^2}{l^2 - (342.0)^2} + \frac{3.909\, l^2}{l^2 - (144.6)^2}}$$

$$n_e(l) = \sqrt{1 + \frac{0.0117\, l^2}{l^2 - (422.5)^2} + \frac{4.437\, l^2}{l^2 - (177.0)^2}}$$

where l is expressed in nm. The refractive indices of several other GaN samples were also measured. Slight differences appeared between all these measurements. Nevertheless, we found out that there was a linear relation between the refractive indices and the stress state of the studied waveguides. Moreover, the ordinary index n_o appears to be more sensitive to stress variations than the extraordinary index n_e. These results, in terms of elasto-optic coefficients, are about to be published.

From the GaN/Sapphire film presented before, the nonlinear coefficients could be determined. They were measured by the rotational Maker fringes method. The pump laser was a pulsed Nd:YAG at 1064 nm. After adjustement of the angle resolved transmitted second harmonic signal generated in the GaN film, we found the following values for the non-zero elements of the $c^{(2)}$ tensor: $c_{33} = -10.6\, pm/V$ and $c_{15} = c_{31} = 5.3\, pm/V$. They are in good agreement with the latest published values by Sanford *et al*[5]. These are about one third of the lithium niobate coefficients.

Planar waveguide structures were grown directly on sapphire. The overall optical properties of these guides such as the effective indices or the propagation losses of the different TE and TM modes, were studied on Ga-polar GaN films. A strong increase of the losses from the fundamental mode (~ 1 dB/cm) to the highest order ones was evidenced. The same behaviour is observed for both the TE and TM modes. This is mainly due to light scattering at the guide interfaces. Indeed, the index contrasts are very high: 1.35 at the GaN/air interface and ~ 0.6 at the GaN/Sapphire interface at 632.8 nm. Figure 2 shows the intensity profiles of the four first TE modes of a GaN waveguide on sapphire. Here, these profiles are superimposed to the section image of such a guide as seen by transmission electron microscopy.

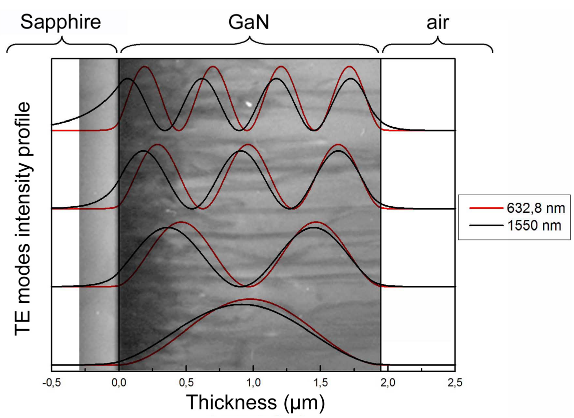

Fig. 2: Intensity profiles of the four first TE modes of a GaN planar waveguide on sapphire, at 632.8 and 1550 nm wavelengths. The guide thickness is 1.9 µm. The backward image is a transmission electron microscopy (cross section) picture of such a GaN film.

The dark lines are threading dislocations which originate from the strong lattice mismatch between GaN and sapphire (-15.9%). Their number decreases with the deposited film thickness. In general, the

GaN films grown by molecular epitaxy on sapphire substrates present a dislocation density of few 10^9 cm^{-2}. Yet, their potential influence on the propagation losses is still unclear. Note that the index variations that may occur along the guide thickness are negligible compared to the index contrasts responsible of the optical confinement.

GaN planar waveguides were also grown with differents inferior cladding layers consisting of AlN, $Al_{0.4}Ga_{0.6}N$, or $Al_{0.1}Ga_{0.9}N$ films. The index contrast between guide and cladding is all the more reduced as the Al composition decreases. In every case, the same behaviour of losses increase with the mode order is still observed. Therefore, its origin is essentially due to scattering at the GaN/air interface. Thus, the surface roughness plays a key role on the optical losses observed.

GaN structures dedicated to quasi phase matching

The QPM technique necessitates a periodic structuration of the sign of the nonlinear coefficient $c^{(2)}$ (with a typical period of few to tens of microns). Yet, the $c^{(2)}$ sign is linked to the crystal orientation. Therefore, the GaN films dedicated to QPM have to present an alternance of (0001) domains (said to be of Ga-polarity) and of $(000\bar{1})$ domains (N-polarity). The process used to get such periodically poled (PePo) structures involves two growth steps[11] and an intermediate etching step in order to define the periodic pattern. These different steps are summarized in figure 3.

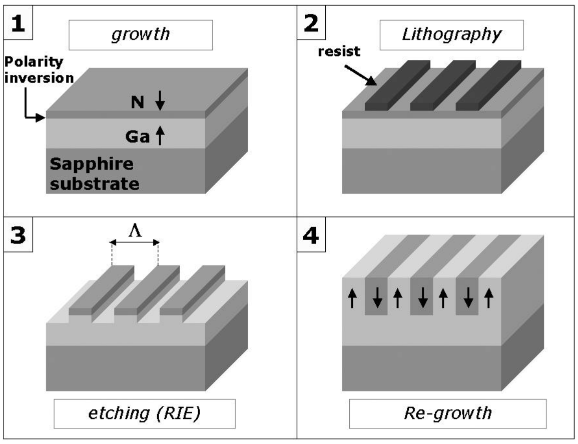

Fig. 3: Realization of periodically poled (PePo) GaN structures. (a) polarity inversion induced during GaN growth by a strong Mg doping. (b) resist deposition and UV lithography of the PePo pattern. (c) etching of the stripes down to the inferior Ga-polarity by reactive ion etching. (d) re-growth of the PePo film.

In our standard growth conditions, the layers naturally exhibit the Ga-polarity. The crystal orientation have first to be reversed. This is now well established by briefly and strongly doping the GaN film with Mg during growth[12]. The inversion process is checked in situ by reflexion high energy electron diffraction (RHEED). The diffraction diagrams are different whether the surface is the one of a Ga-polar or of a N-polar film. Then, the stripes pattern is fixed into some resist by UV lithography. An etching step is driven and the depth is controlled to be around 50 nm so that the inferior Ga-polar film is reached. The resist is further removed and the sample is re-introduced in the MBE chamber for a ~ 1h annealing at 800°C under ammonia flux. The re-growth is driven at a lower temperature, 770°C, which gives the lower roughness for the N-polar domains.

In the case of PePo planar waveguides (with Ga and N-polar domains), the propagation losses are very high (> 20 dB/cm) due to the superior roughness of the N-polar domains. Figure 4 shows a 3 µm thick regrowth of a PePo sample at 770°C. Figure 4a is a scanning electron microscopy (SEM) image of a sample of period 10 µm. The Ga and N-polar domains have almost the same thickness. Figure 4b is centered at the junction between two domains of a PePo pattern of period 40 µm. The surface roughnesses are well visible on this image. The Ga domain is smooth and presents a typical kinetic roughness[13]. The N domain is flat at a large scale but presents a high roughness at low scale. Although small pyramids can be seen, the surface is well smoother than for a regrowth at 800°C.

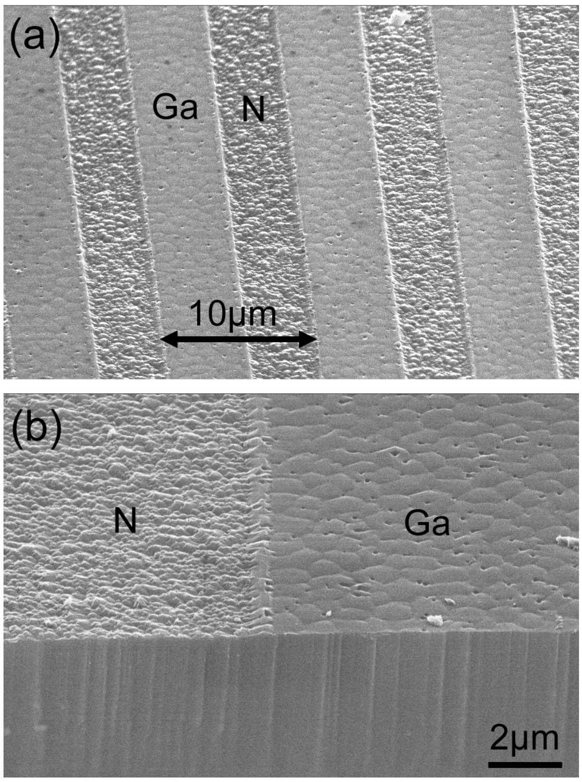

Fig. 4: Scanning electron microscopy images of a same sample (PePo GaN film having a thickness of 3 µm) for different periods. (a) period of 10 µm. (b) period of 40 µm.

The propagation losses should be reduced if the index contrast between guide and cladding (which in this case is air) is lowered. This can be realized by the growth of a superior (Al,Ga)N cladding layer with a low Al content (refractive index slightly inferior to the GaN one). With the deposition of such a film, a significant diminution of the losses has already been observed on Ga-polar waveguides. Works are currently driven towards the realization of PePo symmetric ridge waveguides.

Submicronic periods

Finaly, PePo structures with submicronic periods down to 170 nm have been grown[11]. In that case, the classic UV lithography is replaced by an etching by focused ion beam (FIB). The stripes are directly etched using Ga^+ ions accelerated at 35 kV with doses in the range 10^{16} to 10^{17} cm^{-2}. Before regrowth, no surface treatment is done, but the sample is annealed at 650°C under vacuum during 12h. Figure 5 is a SEM image of a 2 µm thick regrown submicronic PePo sample. It can be seen that the period is well respected. Yet, the Ga and N-polar domains do not have exactly the same thickness. Further work is needed to optimize the growth conditions (growth temperature, V/III ratio,…) in order to have equality of growth rates between the Ga and N-polar domains. Nevertheless, this result is very promising since it makes of GaN a candidate to backward propagating nonlinear interactions which have still not been achieved experimentally. Indeed, actual technological limitations prevent to reach, with the classic and widely used nonlinear materials such as lithium niobate[14], the submicronic periods that are required.

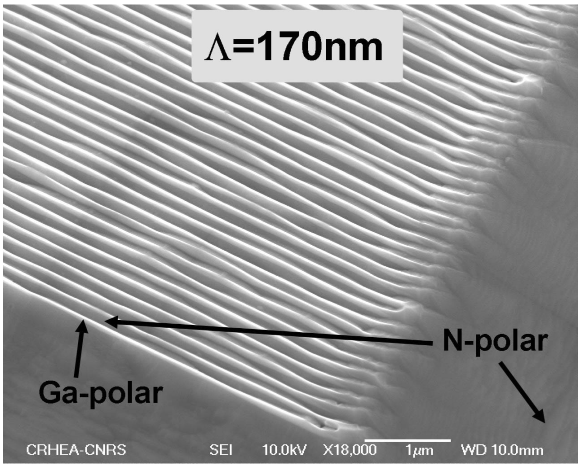

Fig. 5: Scanning electron microscopy images of a PePo film with a 170nm period. The re-grown thickness after focused ion beam process is 2 µm.

Summary

In conclusion, we have presented GaN as a new material for nonlinear optics. Periodically poled GaN structures dedicated to quasi phase matching are obtained by two growth steps with an intermediate etching step. The ordinary and extraordinary refractive indices are now well known on a wide spectral domain as well as their variations with stress. Yet, the propagation losses in periodically poled waveguides are high due to strong light diffusion at the rough N-polar domains surface. The burying of such waveguides with an (Al,Ga)N film should permit a significant reduction of the losses since it is already the case for Ga-polar buried waveguides. On the other hand, periodically poled GaN structures with submicronic periods were realized and backward propagating nonlinear interactions can then be envisaged.

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