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## Second-harmonic generation in Zn-diffused periodically poled LiNbO<sub>3</sub> channel waveguides

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Received: 16 May 2001/Revised version: 7 September 2001  
Published online: 30 October 2001 • © Springer-Verlag 2001

**ABSTRACT** In this work second-harmonic generation by quasi-phase matching (QPM) in Zn-diffused periodically poled lithium niobate channel waveguides is presented. A stable TM→TE conversion by QPM has been found. The results are in good accordance with theoretical estimations obtained by the phase-matching condition, either for the polarisation character of the second-harmonic wave as well as for the spectral range, taking into account the periodicity of the domains.

PACS 42.82.-m; 42.65.Ky; 77.84.-s

### 1 Introduction

Optical frequency conversion by quasi-phase matching (QPM) has been demonstrated using different ferroelectric crystals [1–3]. Particularly interesting is lithium niobate (LiNbO<sub>3</sub>) due to the large effective non-linear coefficients of periodically poled lithium niobate (PPLN) [1]. Several non-linear devices based on this material, in which the periodic modulation is combined with the high power densities available in waveguiding structures, have been reported [4–7].

The potential advantages of Zn diffusion have been previously reported [8, 9] as a fabrication technique to obtain LiNbO<sub>3</sub> waveguides. It has also been recently reported that Zn diffusion from the vapour phase at low temperatures produces low-loss waveguides in this material, and preserves the initial domain pattern of the substrate [10, 11]. Therefore, with this technique it is possible to use PPLN substrates in which the periodic modulation is induced during the crystal-growth process.

In this work the non-linear character of Zn-diffused periodically poled LiNbO<sub>3</sub> channel waveguides is explored. A CW Ti:sapphire laser has been used to investigate the optical frequency conversion by QPM. Second-harmonic generation (SHG), at room temperature, in the blue spectral region has been detected for fundamental waves in the near-infrared range. A good accordance between experimental data and theoretical estimations has been found.

### 2 Experimental procedure

Periodically poled lithium niobate crystals have been grown along the *x*-axis in an air atmosphere with automatic diameter control by a crucible weighing system [12]. The initial melts, containing the congruent LiNbO<sub>3</sub> ([Li]/[Nb] = 0.945), have been doped with erbium and ytterbium oxides with a purity grade of 99.99%. The concentration of both dopants in the initial melts was 0.5 mol%. This technique allows us to induce the formation of periodic domain structures during the crystal growth by using the appropriate growth conditions [13–15].

After cutting and polishing to optical grade, the ferroelectric domain pattern of a *Y*-cut substrate from one of the PPLN crystals has been revealed by chemical etching in a diluted solution of HF:HNO<sub>3</sub> (1:2 by volume) at room temperature for 1 h. Scanning electron microscope (SEM) measurements have been done to characterise the ferroelectric domains.

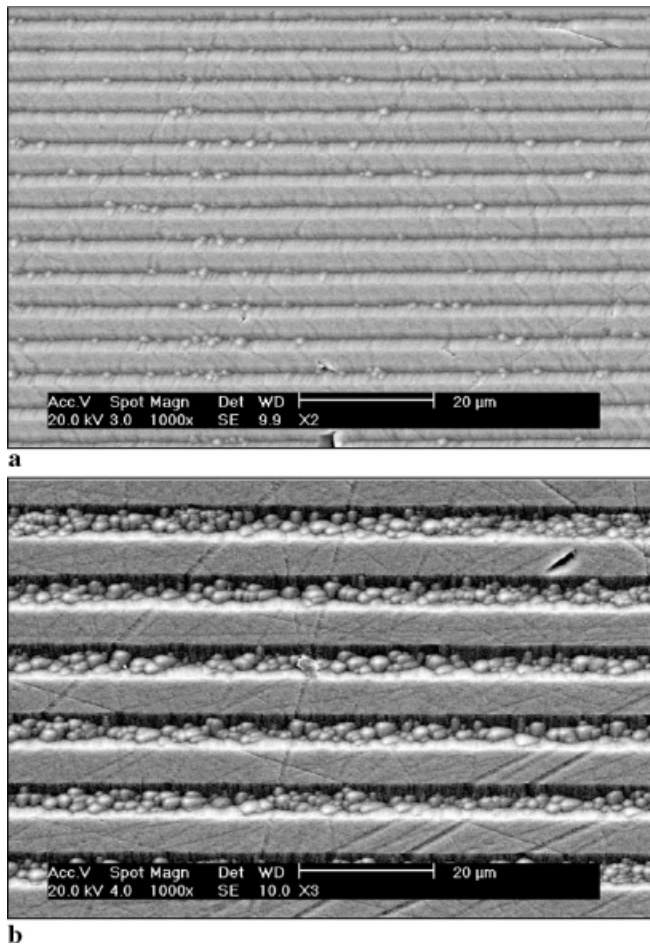
The wafer was again polished to a flat surface with optical quality in order to perform the channel-waveguide fabrication. Ultraviolet photolithographic techniques, adapted from microelectronic technology, have been used to define the channels in which the metal diffusion takes place [16]. The Zn diffusion was performed following the two-step procedure reported in [10]. In the first step the sample was placed in a Zn atmosphere at 550 °C for 2 h, and a second step of diffusion at 850 °C for 4 h was selected in order to produce low-loss waveguides.

A CW Ti:sapphire laser, with a tunability range from 720 nm to 870 nm, was used to perform the characterisation of the SHG in the Zn-diffused PPLN channel waveguides. The intensity of the second-harmonic wave was detected by using an EMI-9558QB photomultiplier tube.

### 3 Results and discussion

The ferroelectric domain pattern of the substrate was revealed by chemical etching and then analysed by using a scanning electron microscope. The SEM measurements confirm that different periodically poled (PP) regions, about 1 mm in length and of different periods, were generated during the crystal-growth process. Figure 1a and b show the micrographs corresponding to two regions with periods of 5 μm and 10 μm respectively. Although there are different

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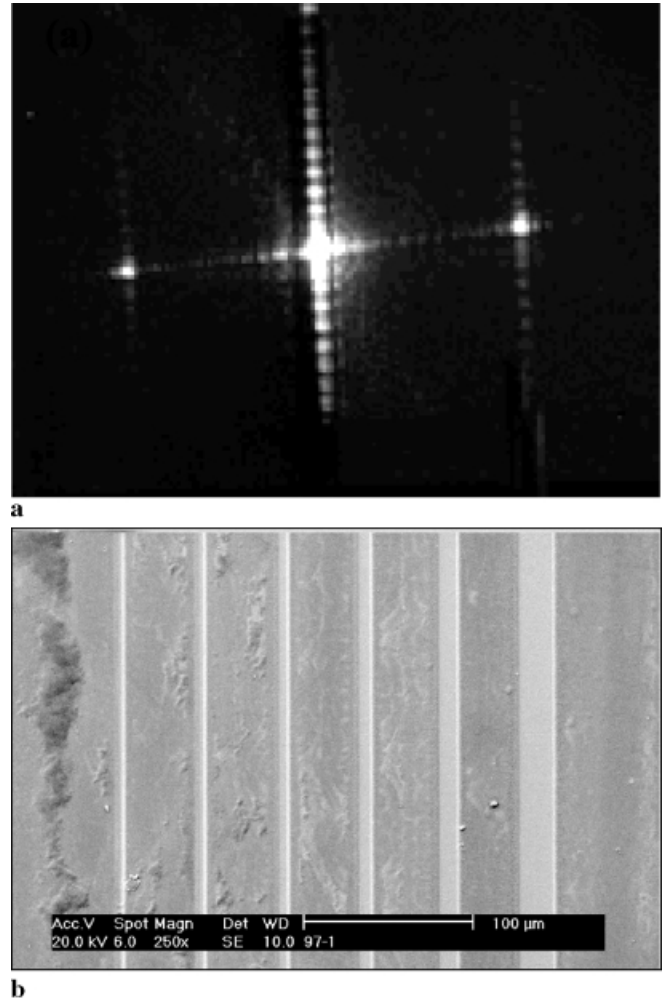
**FIGURE 1** SEM micrographs of PP regions with **a** 5- $\mu\text{m}$  and **b** 10- $\mu\text{m}$  periods

regions with periods ranging from 5  $\mu\text{m}$  to 16  $\mu\text{m}$ , most of these periodically poled regions have a period value around 6–8  $\mu\text{m}$ .

The initial stage in the fabrication of the channel waveguides is the definition of the mask that stops the Zn diffusion. The motifs of a  $\text{Cr}_2\text{O}_3$  commercial mask have been transferred to a  $\text{SiO}_2$  film, previously deposited over the substrate, by UV photolithographic techniques [16].

The perpendicularity between the channels in the  $\text{SiO}_2$  stopping mask and the PP domains in the  $\text{LiNbO}_3$  substrate has been checked, prior to the Zn diffusion, by diffraction techniques. A helium–neon laser (632.8 nm) was used to illuminate the sample surface that contains the channels; Fig. 2a shows one of the diffraction patterns obtained. The diffraction in the vertical line corresponds to the periodic series of channels in the  $\text{SiO}_2$  film, while the pattern in the horizontal line is induced by the presence of a periodic modulation in the ferroelectric domains, showing that a good perpendicularity between channels and PP domains has been achieved.

In Fig. 2b a SEM micrograph of the Zn-diffused channels (with dimensions 1-cm long and 1-, 2-, 4-, 6-, 10- and 20- $\mu\text{m}$  wide), before removing the  $\text{SiO}_2$  stopping mask, is presented. The channel waveguides are parallel to the  $x$ -axis (the  $c$ -axis perpendicular to the channels).

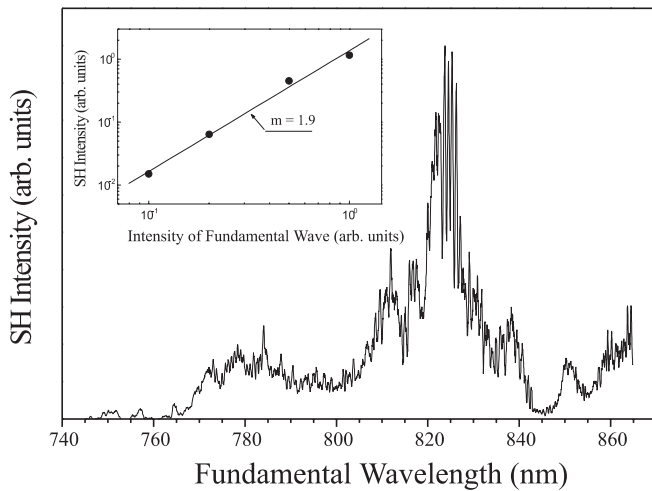


**FIGURE 2** **a** Diffraction pattern at 632.8 nm showing the perpendicularity achieved between the channels in the  $\text{SiO}_2$  stopping mask and the PP domains; *vertical* and *horizontal patterns* respectively. **b** SEM micrograph of Zn-diffused waveguides (1-, 2-, 4-, 6-, 10- and 20- $\mu\text{m}$  widths)

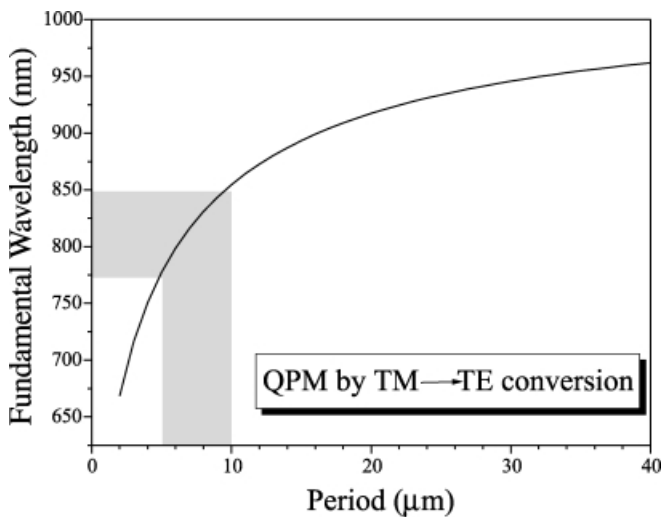
The non-linear character of the Zn-diffused channel waveguides has been explored in continuous-wave mode at room temperature by using a CW Ti:sapphire laser. The fundamental wave ( $\omega$ ), polarised along the ordinary direction (in order to produce an interaction with the  $d_{32}$  coefficient), was coupled into the waveguides by using a microscope objective. A second objective, at the other end, was used to collect the second-harmonic wave ( $2\omega$ ). A strong polarised character (higher than 90%) has been found along the extraordinary direction for the  $2\omega$  wave, indicating that the QPM involves a  $\text{TM} \rightarrow \text{TE}$  conversion.

Figure 3 shows the phase-matching curve for the  $\text{TM} \rightarrow \text{TE}$  conversion measured in the 2- $\mu\text{m}$  channel waveguide with 1-cm length (containing non-uniform PP regions, as has been mentioned previously). As can be seen, the phase-matching condition is satisfied for fundamental wavelengths higher than 770 nm, the  $2\omega$  wave having its maximum intensity for excitation at around 822 nm. The inset in this figure shows the expected quadratic dependence of the SH intensity on the intensity of the fundamental wave.

Figure 4 shows the calculated relationship between the ferroelectric domain period ( $\Lambda$ ) and the fundamental wavelength



**FIGURE 3** Phase-matching curve at room temperature measured in the 2- $\mu\text{m}$  channel waveguide. The *inset* shows the dependence of the SH wave on the intensity of the fundamental wave



**FIGURE 4** Calculated relationship, in the case of QPM by TM $\rightarrow$ TE conversion, between the ferroelectric domain period and the fundamental wavelength

( $\lambda_\omega$ ) at first order for QPM by TM $\rightarrow$ TE conversion, which is given by [3]:

$$\Lambda = \frac{\lambda_\omega}{2[n_e(2\omega) - n_o(\omega)]} \quad (1)$$

where  $n_o(\omega)$  and  $n_e(2\omega)$  are the ordinary and extraordinary refractive indices at the corresponding wavelengths.

This theoretical calculation proves that by taking into account the different periods in the ferroelectric pattern of the substrate, which has been indicated in Fig. 4 by a shadowed region, it should be possible to obtain SHG for a wide range of fundamental wavelengths. The QPM relationship is satisfied from 770 nm to 850 nm, being in perfect agreement with the experimental phase-matching curve. The maximum conversion in the generation of blue light is reached for fundamental

wavelengths close to 820 nm. From (1), it can be deduced that the periods needed for this conversion should be close to 7  $\mu\text{m}$ . This value is also in good accordance with the initial SEM analysis of the substrate (mentioned above), which reveals the dominant period value (around 6–8  $\mu\text{m}$ ) in the periodically poled regions.

Preliminary measurements related to the photorefractive damage indicate that, at the incident power level used in these experiments (around 0.1 W), the fundamental mode pattern and the SH intensity remain stable, although a quantitative estimation of the optical damage threshold has not been obtained. Detailed evaluation of this figure is currently in progress.

#### 4 Conclusions

The feasibility of Zn diffusion from the vapour phase to produce non-linear waveguides in lithium niobate has been demonstrated. Optical frequency conversion by quasi-phase matching has been obtained in Zn-diffused LiNbO<sub>3</sub> channel waveguides via TM $\rightarrow$ TE conversion. The SH wave exhibits a broad phase-matching curve in consequence of variations in the period associated with the PP ferroelectric domains.

Further investigations in order to precisely control the domain period during crystal growth are needed in order to obtain a higher efficiency in blue-light generation.

**ACKNOWLEDGEMENTS** This work was partially supported by DGES (PB97-0019) and Comunidad de Madrid (07T/0026/1998). R.E. Di Paolo is supported by a grant from Ministerio de Educación y Cultura (Spain).

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