

Spatial Modulation of Linear and Quadratic Susceptibilities in Lithium Niobate Crystals by Using Femtosecond Laser Pulses

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15 *In this work we present the spatial control of the linear susceptibility (χ_1) in Lithium Niobate crystals by means of infrared (800 nm) femtosecond interaction. Diffraction gratings have been performed on the surface (relief) and inside (phase) of these samples by femtosecond laser writing. Also we have performed a spatial control of the quadratic susceptibility (χ_2) by direct writing of a pattern of ferroelectric domains on the surface of z cut substrates by using the second harmonic femtosecond pulses (400 nm). Finally, efficient photonic devices for second harmonic generation via quasi phase matching could be obtained following the experimental procedure presented in this work.*

Keywords

Q1

20 Introduction

During last years, femtosecond laser writing have represented an important tool to make photonic structures because of the simplicity, fast procedure and low cost to make integrated devices by using one step process [1–4].

25 The aim of these procedures deals on the interaction of femtosecond pulses with transparent materials giving rise to ionization processes. Focusing high intensity (10^{13} W/cm²) laser on the surface of transparent materials, for fluences above the ablation threshold, ionization process due to the multiphoton absorption takes place in the medium. For this energy regime, the material begins to be removed from the surface of the sample. On the other hand, focusing high intensity lasers inside transparent samples, and depending of the
30 energy range used in the experiments, the observed phenomenon can be identified as follow: 1) for laser intensities below damage threshold, it is possible to obtain a weak refractive index increment (about 10^{-3}) at the focusing region. This fact can be associated with the formation of colour centres and high density regions at the interaction zone; 2) for intensities above damage threshold, filaments due to Kerr effect can be observed whose length is

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large compared with the Raleigh zone from the focused beam. In this case, previous to the initial part of the filament, an important guiding region is also obtained [7].

Different 3D photonic devices have been recently reported by femtosecond laser writing in fused silica and other glass materials like fluoride, garnets and Ge-silica samples [8].

Addressing this type of photonic devices fabrication method for the use of electro-optic crystals like Lithium Niobate substrates it is possible boosting its application for optical communications such as splitters, Mach Zehnder interferometers, modulators, switches, erbium doped waveguides amplifiers (EDWAS), etc.

In this work we show the spatial control of the linear susceptibility (χ_1) trough of the fabrication of diffraction gratings on the surface (relief) and inside (phase) Lithium Niobate crystals by means of the direct femtosecond laser writing. Also it is presented the spatial control of quadratic susceptibility (χ_2) inducing the ferroelectric domains by using femtosecond writing at 400 nm on $-z$ face of LiNbO_3 substrates.

Experimental Procedure

The laser source to make the femtosecond laser writing was a Ti:Sapphire laser at 800 nm from Spectral Physics with 120 fs pulse width, 1 KHz repetition rate and up to 1 mJ energy delivered. The laser writing was made by using the experimental set-up shown in Fig. 1. The femtosecond infrared radiation was focused to the sample by means of the microscope objective 10 \times after passing through a 3 mm pinhole to get a cleaner beam. The sample was mounted on micro-positioning stages which are controlling by a PC. The minimal displacement from these stages is 0.5 μm . The diffraction gratings have an area of 4 mm^2 and a 10 μm pitch. To make the ablation groove, the writing laser was focused on the surface of the samples while for the phase gratings fabrication process, the laser was focused without pinholes, by using 20 \times and 40 \times microscope objectives at 200 μm below the surface of the z-cut LiNbO_3 substrates.

Finally, to get a spatial control of χ_2 , inducing the ferroelectric domains inversion by laser writing, the infrared femtosecond laser was doubled by using a Beta Barium Borate (BBO) non linear crystals cut to achieve 400 nm at the output to make this process. The optical poling [9] was performed by focusing the blue radiation on the $-z$ face of the sample by means the experimental set-up shown above. The z-cut LiNbO_3 crystal was impinged with blue radiation at energies below the ablation threshold. In order to reveal the ferroelectrics domains, after blue laser interaction, the sample was kept in HF solution during 1 hour.

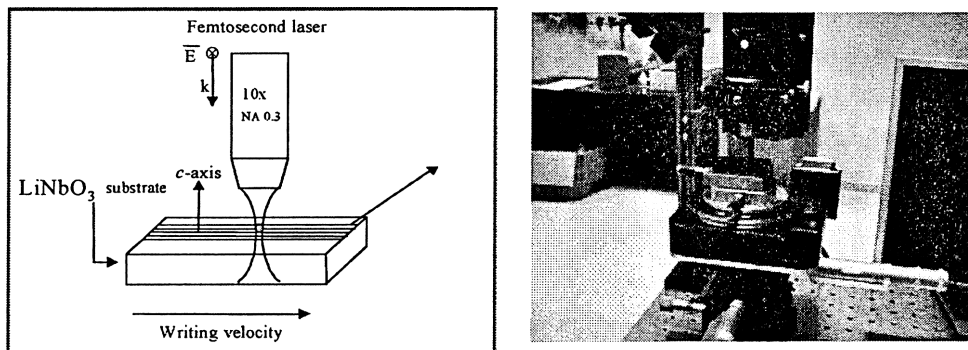


Figure 1. Experimental set up to perform the femtosecond laser writing on the z-cut Lithium Niobate substrates.

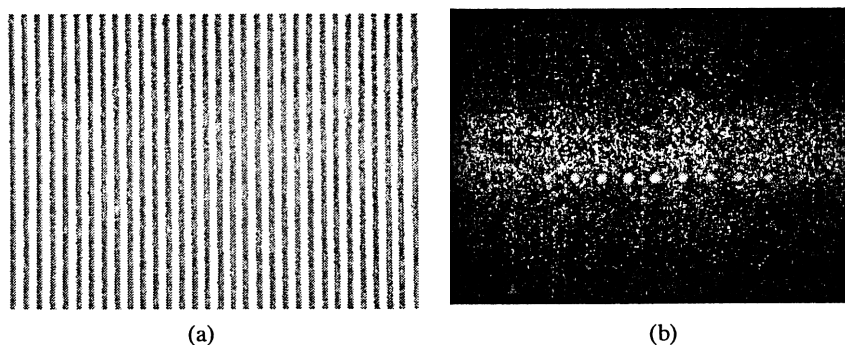


Figure 2. (a) Optical microscope photograph (working in reflection) of the relief diffraction grating fabricated by femtosecond laser writing. (b) He-Ne diffraction pattern from this grating.

Results and Discussion

70 The ablation gratings were made by impinging fluences of 1 J/cm^2 , slightly above the ablation threshold (0.7 J/cm^2). Figure 2a shows a photograph taken with microscope (working in reflection) of the relief grating performed at the surface of LiNbO_3 crystals by femtosecond laser radiation. As it can be seen, on the dark region have been carried out the ablation process and the material was removed over these regions. On the other hand, the white regions correspond to non-irradiated zones. The depth and width of the ablation channels are $5 \mu\text{m}$ and $6 \mu\text{m}$ respectively, while the pitch of the grating was set to be $10 \mu\text{m}$.
 75 Due to fabrication process, these gratings have a high refractive index contrast which is about 50% ($\Delta n = 1.2$). In order to characterize the diffraction behaviour of these photonic structures, a He-Ne laser at 632.8 nm was used at normal incidence for testing the device. A diffraction pattern with high orders were observed, as it is illustrated in Fig. 2b. The intensity of these diffraction orders show a very smooth decrease for consecutive orders. This fact can be related to the high refractive index contrast for the fabricated ablation gratings.

80 Figure 3a presents the optical contrast microscope image corresponding to phase grating recorded inside the Lithium Niobate crystal. As it can be seen the irradiated regions shows a thinner width ($2\text{--}3 \mu\text{m}$) than that observed for ablation gratings ($6 \mu\text{m}$).
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From these gratings a lower diffraction efficiency has been observed which can be associated to the weaker index increment for the experimental conditions performed. In

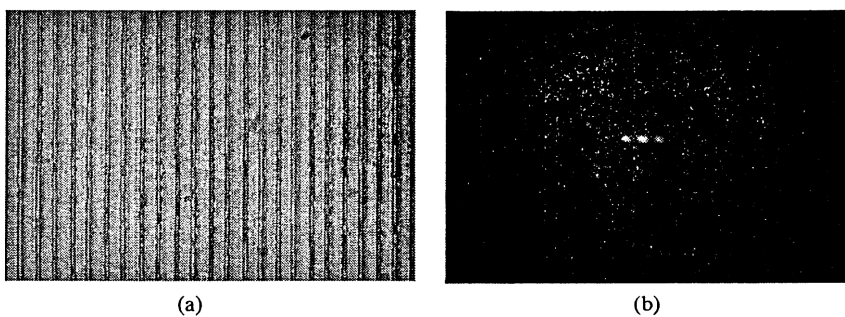


Figure 3. Optical microscope photograph (taken in transmission) of the phase grating written inside of Lithium Niobate crystals by femtosecond laser pulses.

Q3

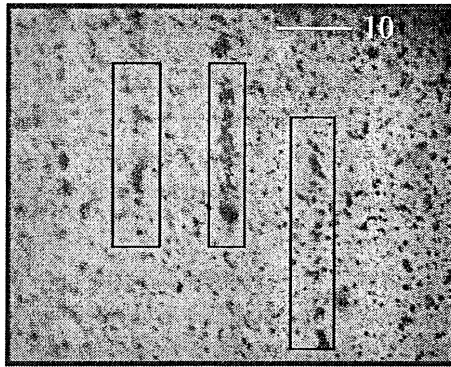


Figure 4. Ferroelectric domains induced by 400 nm femtosecond writing after revealing in HF solution.

our experiments, the induced index increment can be estimated to be $\Delta n = 10^{-3}$ which originates a refractive index spatial modulation up to 0.2%. In spite of the low refractive index increment, an important diffraction pattern from the 1 D photonic structures is obtained, as it is shown in Fig. 3b. It can be seen that high intensity orders reach up to ± 1 , and an appreciable decrease of intensity for the following orders is also observed. Moreover, for a similar refractive index increment induced in the Lithium Niobate crystal, efficient waveguides can be obtained by using several fabrication techniques [10].

In order to control the spatial distributions of the ferroelectric domains in a z cut lithium niobate substrate, the femtosecond laser writing process by using a wavelength of 400 nm for energies below to the ablation threshold was conducted. The blue irradiated regions, after keeping the sample on HF solution during 1 hour, show a clear intensified domain patterns on the surface of lithium niobate, as it is illustrated in Fig. 5. The surrounding regions to written tracks, also present some revealed ferroelectric domains which are due to the non monodomain initial sample.

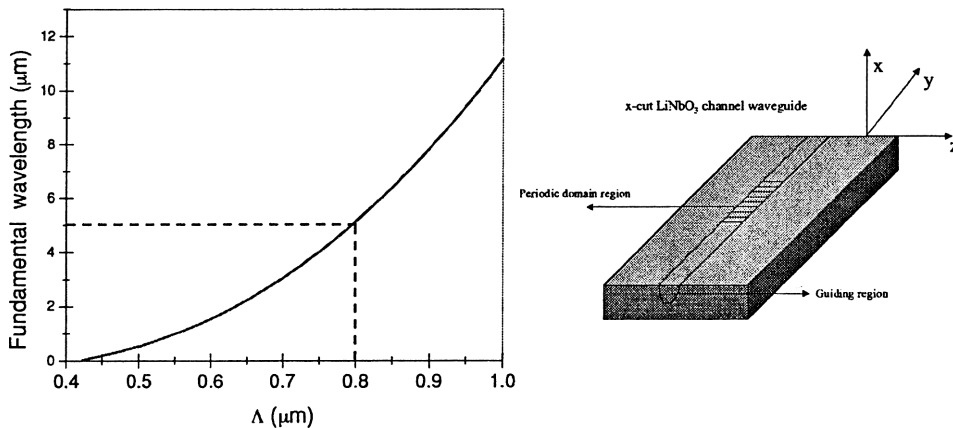


Figure 5. Dependence of the fundamental wavelength as a function of the length (Λ) of periodic domain structure to achieve Second Harmonic Generation (QPM) via Quasi Phase Matching for TE->TE conversion in x-cut LNB crystals.

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As it can be clearly observed from the figure, the region where the ferroelectric domains (denoted by hollow oblong) have been revealed correspond to the written tracks. From these preliminary results we can show that it is possible to obtain a spatial control of the ferroelectrics domains on the LiNbO₃ surface by writing the desired period structure with blue femtosecond laser pulses.

Further works will be addressed to describe the mechanisms involved in the phenomenology of ferroelectric domain orientation on the surface of lithium niobate crystals induced by using blue femtosecond radiation.

By combining the spatial control of quadratic susceptibility (χ_2) and writing a waveguiding region at the same region of the material, it is possible to make powerful integrated devices in waveguide configuration. In order to do this, a periodic ferroelectric structure on the surface or inside of x-cut or y-cut lithium niobate waveguides can be performed. For instance, to get second harmonic generation via quasi phase matching at 800 nm as fundamental wavelength it is necessary a ferroelectric periodic structure ranged between 5–10 μm , depending on the polarization state for both traveling waves inside the waveguides.

Figure 5 shows the dependence of the fundamental wavelength as a function of the of lattice pitch for quasi phase matching process. For efficient conversion, the second harmonic generation in lithium niobate crystals uses the highest second harmonic coefficient d_{33} , in this case x-cut or y-cut samples should be used [11]. A particular case, the fundamental and the second harmonic waves should be TE (ne, extraordinary index) polarized as it is sketched in the inset of Fig. 5.

Conclusions

In this work we present the spatial control of the linear susceptibility (χ_1) on the surface of Lithium Niobate substrates by means of femtosecond laser writing. In order to do that, efficient diffraction gratings either on the surface (relief) or inside (phase) of LNB crystals have been conducted.

Preliminary results of *optical poling* on the surface of Lithium Niobate crystals by writing at 400 nm also has been obtained. A spatial control of 1 μm of period of the quadratic susceptibility (χ_2) could be reached.

From the above experimental procedure we can fabricate integrated waveguides for second harmonic applications via the quasi phase matching in Lithium Niobate substrates by means of femtosecond laser writing.

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