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Periodic poling of optical waveguides produced by swift-heavy-ion irradiation in $LiNbO_3$

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Abstract The generation of periodically poled structures in waveguides prepared by swift-heavy-ion (SHI) irradiation, i.e. in the electronic stopping power regime, has been achieved following two different strategies. In one of them we have prepared bulk PPLN samples by an applied electrical field, followed by irradiation with F ions at 22 MeV. After the ion irradiation, a waveguide showing a high optical confinement is obtained, preserving the original PPLN structure. The second strategy consisted of electric periodic poling of previously fabricated swift-ion-irradiated waveguides. To our knowledge this method has not been, so far, successful for conventional implanted waveguides. The successful fabrication of PPLN structures on novel waveguides prepared by SHI irradiation offers a promising potential for nonlinear integrated optical devices.

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1 Introduction

Nowadays, a very active field of research is the generation of visible light of short wavelengths, for instance with the use of wavelength converters. These nonlinear optical devices have been widely investigated, the quasi-phase matching (QPM) in periodically poled $LiNbO_3$ (PPLN) being a widely used technique for efficient frequency conversion [1]. Since second-harmonic generation (SHG) is a nonlinear process, the waveguide configuration is often preferred because of the higher light intensities that can be obtained in this geometry. Moreover, shorter periods of the periodically poled structure can be achieved in waveguides than those obtained in bulk, due to the smaller thickness of the former configuration.

Nevertheless, the fabrication of PPLN on a waveguide does not always maintain the nonlinear and/or the guiding properties of the structure. Moreover, the result may depend on the kind of PPLN substrate, the type of waveguide, the period of the domain structure and even the fabrication sequence. For Ti-indiffused [2–5] and annealed or soft proton exchanged (APE or SPE) [6–8, 19] waveguides the periodic poling has been achieved either by fabricating the waveguide on a PPLN substrate or more recently by using the reverse procedure, i.e. waveguide fabrication followed by the poling. On the other hand, He^+ implantation at energies of several MeV and high fluences (10^{16}–10^{17} ions/cm^2) is also a known method to fabricate waveguides maintaining to a large extent the electro-optic and nonlinear optical properties of the bulk. However, to our knowledge, there are only a few reports dealing with the fabrication of PPLN structures on implanted waveguides [9, 10]. These works have shown that useful waveguides can be prepared by He implantation on PPLN substrates. However, the periodic poling of an implanted waveguide has not been, apparently, reported.

Recently, a novel type of $LiNbO_3$ waveguide has been fabricated [11, 12] by swift-ion irradiation using swift heavy ions (SHIs) and much lower fluences (10^{14}–10^{15} ions/cm^2) than in the conventional implantation procedure. A main difference in relation to the ion-implantation method is that the main mechanism responsible for the change in refractive index is electronic excitation instead of nuclear collisions. In fact, the features of the electronic damage are quite different from those associated with nuclear damage and implantation. These waveguides present very good properties for nonlinear frequency conversion [11, 12] that should compete favourably with other conventional waveguides. Specifically, they exhibit a sharp waveguide boundary and high light confinement, with index jumps, at $\lambda = 633$ nm, of around 0.1 for n_e and 0.2 for n_o (the highest index steps achieved in $LiNbO_3$ [13]). Moreover, the nonlinear coefficients are similar to those of the substrate and optical damage thresholds are of the order of or even higher than in the case of APE waveguides [14]. The potential of those SHI waveguides for other laser applications has been recently demonstrated [15].

The purpose of this work has been to complete the evaluation of the capabilities of the novel waveguides prepared by SHI irradiation for nonlinear optical applications. To this end, we have investigated the fabrication of periodically poled domain structures with periods in the micrometre range. Two different approaches have been followed: (i) waveguide fabrication on periodically poled lithium niobate (PPLN) substrates and (ii) electric field periodical poling of waveguides prepared by SHI. In all cases good periodically poled patterns have been achieved, and consequently the waveguides offer a good potential for quasi-phase matching in nonlinear optical applications. The results are discussed in relation to those obtained for proton-exchanged and for He (or H)-implanted waveguides.

2 Experimental details

2.1 Electric field poling

The conventional external electric field poling technique [16, 17] was applied for the periodic poling for both bulk samples (substrates) and SHI-irradiated waveguides. All the samples were z-cut congruent $LiNbO_3$ crystals of 0.5-mm thickness. The $+z$ face of the samples was periodically structured with photoresist electrodes of 15-µm period. Then, they were introduced into the poling chamber, where both the structured and non-structured faces were brought in contact with liquid electrodes (LiCl solution) and there a high electric field with a defined pulse shape was applied. The pulse shape was generated with a function generator (DS354, Stanford Research Systems) and amplified with a voltage amplifier (20/20C-L, Trek). The electric current produced during the poling process and the applied electric field pulse were registered with an oscilloscope.

2.2 Fabrication of planar waveguides by SHI irradiation

Z-cut congruent $LiNbO_3$ crystals purchased from PHOTOX were irradiated with F^{4+} ions at an energy of 20 MeV and fluences as low as 4×10^{14} ions/cm^2 in the 5-MV tandetron accelerator of the CMAM, as described in [11]. The incidence direction was along the z-axis (a small tilting of around 7° is introduced to avoid channelling through the crystal structure). The ion currents were kept low enough (<100 nA/cm^2) to prevent excessive heating and charging of the sample. The high-energy F ions penetrating the crystal lose their energy mostly by electronic excitation, as illustrated in Fig. 1a, which shows the electronic S_e and nuclear S_n stopping power curves as a function of depth. One notices that, except at the end of the ion range, $S_e \gg S_n$, i.e. the electronic losses are overwhelmingly dominant over those associated with nuclear collisions. Moreover, the experimental conditions have been chosen in such a way that there is a maximum of the electronic stopping power beneath the surface wherein the crystal is heavily damaged and becomes amorphous. This amorphized layer constitutes an optical barrier for light confinement and propagation. The layer structure induced by SHI is illustrated in the microphotograph of Fig. 1b, which was taken on a sample cut along the irradiation direction for a fluence of 4×10^{14} ions/cm^2. The depth scales for both Fig. 1a and Fig. 1b are the same. One clearly distinguishes the low-reflectivity buried amorphous layer caused by the electronic excitation mechanisms together with a fainter and deeper dark strip corresponding to the nuclear stopping region and ion implantation. On the other hand, the generated refractive-index profile derived from dark-mode characterization [18] together with the estimation of the amorphous layer depth obtained by Rutherford backscattering in channel configuration (RBS/C) ex-

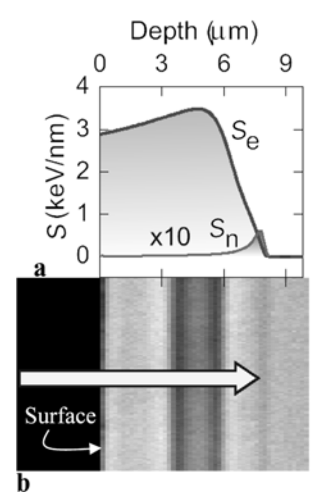

Fig. 1 (**a**) Stopping power curves of F ions at 22 MeV, where S_e represents the electronic stopping power and S_n the nuclear stopping power versus depth. (**b**) Microphotograph of a SHI-irradiated waveguide cut along the irradiation direction for a fluence of 4×10^{14} ions/cm^2. The buried amorphous layer caused by the electronic excitation mechanism and the nuclear stopping region can be seen

periments is shown in Fig. 2. It shows that the amorphous layer is thicker and the boundary separating the crystalline (where the light is guided) and amorphous layers is much sharper than in the case of He or H implantation. In summary, the SHI-irradiation method produces high step index jumps (0.2 for n_o, 0.1 for n_e at $\lambda = 632$ nm), allowing the propagation of highly confined modes, as can be seen in Fig. 2. The optical losses measured with the scattered light technique are around 1 dB/cm [11]. Moreover, the F^{4+} ions stop well inside the crystal, i.e. far from the waveguide and barrier, and so they do not affect the optical propagation.

2.3 Characterization of the domain pattern

In order to check the polarization reversal in the sample, a first hint is the displacement current which goes through the sample during the inversion process. Then, the most direct straightforward characterization technique is the observation of the sample under crossed polarizers, where the birefringence induced by the stress at domain walls makes possible a qualitative approximation to the actual area where polarization has been inverted. Finally, to characterize the quality of the domain structure induced in the samples, they were immersed in HF at room temperature. This procedure gives rise to a selective etching which distinguishes the $-z$ face from the $+z$ face, as well as the $+y$ and $-y$ faces, due to their different etch rates. In such a way we can observe with the microscope the domain pattern.

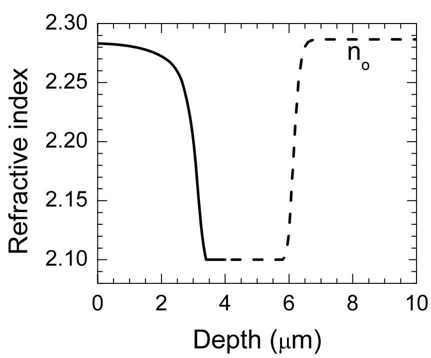

Fig. 2 Refractive-index profile for ordinary polarized light of a swift-ion-irradiated waveguide fabricated with F^{4+} ions with energy of 20 MeV and fluence of 4×10^{14} ions/cm^2, obtained from the dark-mode measurement technique

3 Results of the first strategy: periodic poling followed by irradiation

One starts with a previously generated PPLN z-cut substrate having a domain period of 15 μm. The profile of the voltage pulse used to pole the sample is similar to that illustrated in Fig. 3, but with slightly different applied voltages (the peak value used was 24.48 kV/mm). The sample was then cut into two equal pieces and one of them was then irradiated with fluorine ions at an energy of 20 MeV and a fluence of 4×10^{14} ions/cm^2. Finally, both surfaces of the twin samples (the irradiated and the non-irradiated) were chemically etched to reveal the periodic structure. As can be seen from the comparison between Figs. 4a and 4b, the domain structure is preserved and no disturbance of the domains or domain walls could be found.

The waveguides obtained by SHI of the poled substrates were characterized by the dark-mode technique. The refractive-index profiles were found to be the same as those fabricated on single-domain $LiNbO_3$ substrates, although the irradiation of heavy and energetic ions could have caused some depolarization. This result is similar to that found in the case of ion-implanted waveguides, i.e. He-implanted waveguides, in PPLN substrates, where there has been shown that the waveguide can be fabricated in both

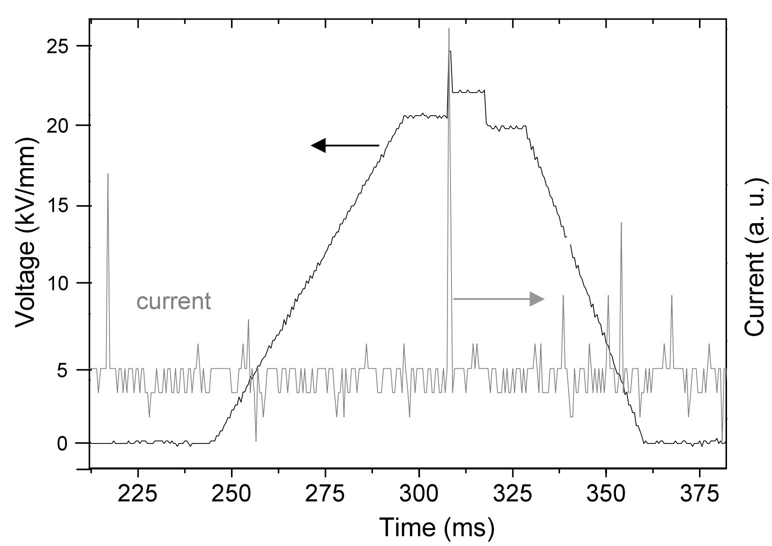

Fig. 3 Time profile of the voltage pulse and the current detected during the periodic poling of a SHI-irradiated waveguide

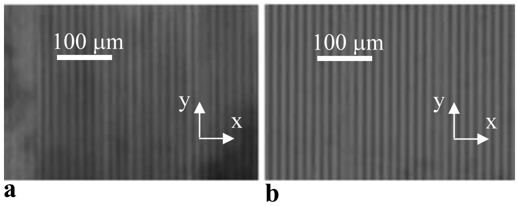

Fig. 4 (**a**) Etched surface of a PPLN sample fabricated by external electric field polarization. (**b**) Etched surface of the same PPLN sample after fabrication of a SHI-irradiated waveguide. It can be seen that the domain structure is preserved after the fabrication of the waveguide

substrates (PPLN and single domain) without affecting the PPLN structure [9].

4 Second strategy: electric field poling of waveguides

In this second approach, the sequence of the experiment was the following. Firstly, swift-ion-irradiated waveguides were fabricated on 0.5-mm-thick, z-cut congruent $LiNbO_3$ substrates with fluorine ions at 20 MeV and a fluence of 4×10^{14} ions/cm^2. Then, a photoresist pattern of 15-µm period and duty cycle of 0.5 was photolithographically made on the $+z$ face of the waveguides. Later, these samples were introduced in the poling chamber in order to proceed with the electric field poling process; we started with an applied field with a lower peak value than in the case of domain inversion in a bulk crystal. We tried a simple pulse with the same shape (shown in Fig. 3) as used in [19]. We increased successively the value of the applied field, maintaining the pulse shape, up to a value of 24.48 kV/mm where some current is detected. The peak current after three pulses is shown in Fig. 3.

To characterize the domain structure of the reversed area, one-half of the sample was chemically etched in HF at room temperature for some minutes and then it was observed with a microscope. As can be seen in Fig. 5a, the periodic domain pattern at the surface in some regions is quite good, with a duty cycle (which is defined as the proportion of reversed material with respect to the period length) of 0.5 in most regions. However, when the sample was observed under crossed polarizers, it was seen that the domain reversal had not taken place homogeneously throughout the whole sample, but only in some regions, the largest one being around 3-mm long. In order to gain additional information on the propagation of the domain reversal, some micrographs taken on a transversal cut of the sample along the irradiation direction have been taken after suitable etching, as can be seen in Fig. 5b. One first notices that both the waveguide and the amorphous barrier have been removed by the etching. Moreover, the domain pattern observed on the $+z$ face (Fig. 5a) appears to propagate somehow into the substrate. In fact, the periodic poling was not to be found on the $-z$ face where, instead, a single reversed domain appears.

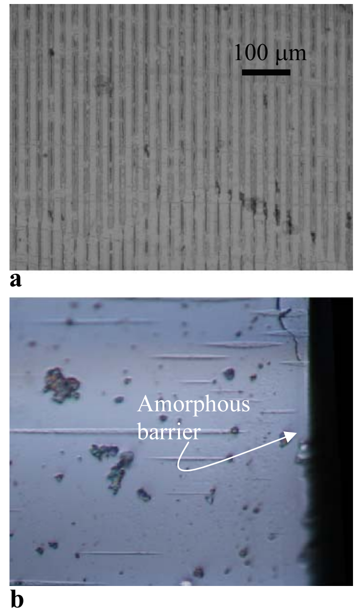

Fig. 5 (**a**) Domain structure of 15-µm period revealed by chemical etching in the $+z$ face of an external field periodically poled swift-ion-irradiated waveguide fabricated with F^{4+} ions at 20 MeV with 4×10^{14} ions/cm^2. (**b**) Transversal cut of the sample showing the irregular propagation of the domain pattern (note that the waveguide and barrier have been removed during chemical etching)

Finally, it was checked that, after the poling process, the waveguide structure and its functionality were maintained. A deeper study should be done in order to find the most appropriate pulse shape to obtain homogeneous PPLN throughout the whole sample on these SHI-irradiated waveguide structures.

5 Discussion

The processes of electrically induced poling (domain inversion) are quite complicated and not yet sufficiently understood. In particular, this is the case for the role played by lattice defects such as those introduced by doping, exchange, implantation or irradiation. On the other hand, it is not clear either how the defects associated with the various methods of waveguide fabrication influence a previously recorded poling structure. Therefore, a comparative analysis of the results achieved so far for periodic poling of different waveguides on $LiNbO_3$ is not an easy task. Here, we will put forward some physical arguments to discuss the available results obtained on the periodic poling of waveguides in $LiNbO_3$.

The first strategy used in this work has been successfully applied to proton-exchange waveguides and, very recently, to waveguides obtained by He implantation. This may be related to the low temperatures used in proton exchange in the first case and to the local character of the nuclear collision and implantation mechanisms (at the micrometre scale) in the second case. On the other hand, the method has not been successful for Ti-diffused waveguides due to the aggressive character of the diffusion process. In fact, the high temperature required for the incorporation of Ti in the crystal erases the previous poling pattern. It may be, here, surprising that the SHI-irradiation method has been successful even though a high local temperature rise (thermal spike) is produced at every single track along the ion trajectory. One may argue that this heating effect is very local (a few nanometres around the ion trajectory) and occurs in times of a few picoseconds and so it does not alter the poling pattern.

The second strategy followed in this paper has been mostly applied to Ti-diffused waveguides and very recently to proton-exchange waveguides [7, 19]. The attempts to apply it to He-implanted waveguides have failed so far [9]. In principle this appears surprising since the coercive field has been shown to decrease with implantation [20]. The reason for this failure has been attributed to the role of the amorphous barrier layer caused by the implantation, which may inhibit the domain-reversal propagation. This is not the case for our SHI-irradiated waveguides (see Fig. 5b), where a well-defined buried amorphous barrier is also formed. A possible reason lies in the possible different nature of the defects created by SHI irradiation and implantation and the different defect profiles. One should note that the domain inversion has crossed the amorphous layer although the progression is not uniform. For irradiated waveguides the defect concentration stays low up to the sharp amorphous boundary whereas for implanted samples the distribution is monotonic and reaches high levels even before amorphization [21]. One should also notice that even an implantation region developed in the irradiated samples occurs at larger depths (far from the waveguide region) and involves fluences much lower than those used for standard ion implantation.

In summary, a better knowledge of the basic physical processes involved in poling would be required in order to understand all the above-mentioned differences and optimize the processes for periodic poling on optical waveguides.

6 Summary and conclusions

We have demonstrated the possibility of producing PPLN structures on swift-ion-irradiated waveguides by applying a periodic electric field. Different from He-implanted waveguides, the two essayed strategies have been successful. In one of them, a low-fluence fluorine implantation (4×10^{14} ions/cm^2) at high energy (20 MeV) on a PPLN substrate produces a waveguide similar to that fabricated on a uniformly poled substrate. The domain structure is not significantly affected by the irradiation process. For the other strategy, the periodic polarization of a waveguide previously fabricated by SHI irradiation has also been achieved. This procedure has not been, so far, successful for He- or H-implanted waveguides. As a conclusion, SHI-irradiated waveguides are confirmed as a very promising option for nonlinear integrated optics devices.

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