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Tailoring of refractive index profiles in LiNbO₃ optical waveguides by low-fluence swift-ion irradiation

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Abstract

Proton-exchange LiNbO₃ planar optical waveguides have been irradiated with swift ions (Cl 30 MeV) at very low fluences in the range $5 \times 10^{10} - 5 \times 10^{12} \text{ cm}^{-2}$. Large modifications in the refractive index profiles, and therefore in the optical performance, have been obtained due to the generation of amorphous nano-tracks by the individual ion impacts. Moreover, a fine tuning of the refractive index can be achieved by a suitable control of the fluence ($\delta n / \delta \phi \sim 10^{-14} \text{ cm}^2$ or $\delta n \approx 10^{-5}$ for $\delta \phi = 10^9 \text{ cm}^{-2}$). An effective medium approach has been used to account for those changes and determine the amorphous fraction of material. The results have been compared with information extracted from complementary RBS channelling experiments. From the calculated amorphous fractions a radius of $\sim 2 \text{ nm}$ for the amorphous tracks have been estimated.

1. Introduction

It has been recently shown that irradiation with high-energy medium-mass ions having specific energies $>0.1 \text{ MeV/amu}$ (swift ions) causes structural damage due to massive electronic excitation [1]. It presents very different features, not yet well understood, to those induced by nuclear collision (and implantation) damage [2]. In particular, bombarding ions whose electronic stopping power is above a certain threshold create amorphous tracks [3–5] of nanometre diameter in LiNbO₃ and many dielectric materials. Moreover, the track diameter increases with the stopping power. Since the refractive index of amorphous LiNbO₃ is significantly lower than the two principal indices of the crystal [6], those tracks modify in a significant and controllable way the average refractive index profile at the irradiated region [7]. This may be used to fabricate a number of photonic devices, such as optical waveguides [8]. Above a critical fluence value, assuring full coverage of the sample surface, homogeneous amorphous

layers with low and isotropic refractive index ($n = 2.10$ at $\lambda = 633 \text{ nm}$) are obtained [9].

There are several well-documented methods to fabricate optical waveguides in LiNbO₃ with interesting potential for integrated optic devices. In particular, the β_1 -phase obtained by suitable proton-exchange treatments [10, 11] shows high optical confinement and undergoes very reduced photorefractive damage [12], making it adequate for high power linear applications in the visible spectral region. Alternatively, α -phase proton-exchanged waveguides present lower confinement but their SHG coefficients are similar to those of the bulk crystal. The changes in optical response induced in those, either α - or β_1 - waveguides, by swift-ion irradiation open a number of technological possibilities to tailor the refractive index profile of a given integrated optics device. For example, they could be used to introduce predetermined refractive index patterns in a planar waveguide by suitable masks and to directly record certain device architectures (e.g. channelled waveguide structures). Here, one may take advantage of the straight ion trajectories in the electronic stopping power regime and the sharp boundaries

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that may be achieved between irradiated and non-irradiated regions. Also the electronic irradiation effects may serve to achieve a fine tuning of the phase-matching conditions in nonlinear applications. In a future perspective, one may also profit from the nanostructuring caused by the non-overlapped tracks in a low-fluence irradiation. For example, one may consider, as a long-term application, the fabrication of optical sensors by etching the nano-tracks and use the molecular trapping and filtering capabilities of the formed nano holes. Finally, these kinds of electronic excitation effects, induced by swift ions that have been scarcely investigated so far, may be relevant when using optoelectronic waveguide devices in radiation environments (fission and fusion reactors, space missions etc).

The specific purpose of this work has been to explore the use of swift ion-beam irradiation to generate non-overlapped amorphous nano tracks in optical waveguides and determine the induced changes in the refractive index profiles. Proton-exchange waveguides have been selected because of the simplicity for fabrication and good optical performance [10, 11]. It will be shown that the shape of the waveguide refractive index profile is essentially preserved by the irradiation but the refractive index jump and effective indices can be finely tuned in units of 10^{-5} or even smaller by playing with the fluence. The results obtained have been discussed in terms of the formation of individual amorphous tracks, using a simple averaging (effective-medium) approach to describe the optical permittivity.

2. Experimental

2.1. Sample preparation

β_1 -phase waveguides have been prepared from *X*-cut congruent LiNbO₃ wafers (integrated optics grade, 1 mm thick), purchased from Photox Optical Systems (Oxford). They have been obtained by immersion in a benzoic acid melt buffered with 1% lithium benzoate at 300 °C for 0.5 h within a sealed ampoule. This treatment led to an exchange degree of 50–70% and a step-like refractive index profile, with $\Delta n_e = 0.11$ and a thickness of 1.1 μm , supporting 3 modes at $\lambda = 488 \text{ nm}$. The structure of the phase [11] is different from the one corresponding to congruent bulk LiNbO₃. For the purpose of comparison some α -phase (annealed or soft proton-exchanged, SPE) guides have been directly produced by immersion in a benzoic acid melt buffered with 3% lithium benzoate at 300 °C for 18 h within a sealed ampoule. This procedure gave rise to a guide supporting 2 modes, with an estimated surface refractive index change of $\Delta n_e = 0.012$ and a thickness of about 3 μm , at $\lambda = 488 \text{ nm}$. The SPE method is known to produce smooth, Gaussian-like refractive index profiles. The composition of the phase is known to be $\text{H}_x\text{Li}_{1-x}\text{NbO}_3$ with x in the range 0.10–0.15, and the structure is the same as that for the LiNbO₃ substrate.

2.2. Swift-ion irradiation

The ion irradiations using Cl at 30 MeV were performed in the 5 MV accelerator installed at CMAM (www.uam.es/cmam). The stopping power curve as a function of depth is shown in figure 1 which assures that it lies above the amorphization

Figure 1. Electronic stopping power S_e as a function of depth in LiNbO₃ for irradiations with Cl ions having 30 MeV. The value S_{th} for the amorphization threshold is represented by a dashed line.

Figure 2. Schematic view of the waveguide and of the nano-tracks created by the Cl 30 MeV ions.

threshold value [9] ($S_{th} \sim 5 \text{ KeV nm}^{-1}$) from the surface down to around 3–4 μm depth. In principle, this is expected to be the length of the amorphous tracks (see a simple scheme in figure 2). In this regime the dielectric constant of the irradiated layer should be the average of the value for the non-irradiated crystal (or waveguide) and that for the amorphous region. Fluence was kept below 10^{13} cm^{-2} so that we are in the regime of non-overlapped tracks. Currents were kept below a few nanoamps to avoid excessive heating of the samples.

2.3. Waveguide characterization

The optical characterization of the irradiated samples was made by means of the standard dark-mode technique, coupling light through a rutile prism. The emission from an Ar laser at 488 nm with polarization control (TE mode) was used to determine the extraordinary effective refractive indices.

3. Results

3.1. β_1 -phase waveguides

These are relevant waveguides for linear optical applications. For all used fluences the three TE modes observed in the unirradiated waveguides are preserved. The dependence of the effective refractive indices at 488 nm, N_m , versus $(m+1)^2$, m being the mode order, is plotted in figure 3 for different fluences. An essentially linear dependence is obtained, as expected for a step-like profile, although a slight deviation

Figure 3. Square of the effective refractive index versus $(m+1)^2$, m being the mode order, for different irradiation fluences on a β_1 -phase proton-exchanged waveguide.

appears for the third mode at the two higher ion fluences. In addition, the exact waveguide wave equation with a testing profile (Fermi-type) has been numerically solved using a multilayer method to obtain its effective indices i.e. the resultant eigenvalues. These indices are compared with the experimental ones and the testing profile is varied using an iterative method until an error below 0.5% is reached. The conclusion is that a step-like profile is a good approximation and it has been assumed to calculate the effect of ion irradiation. The flat profile at the β_1 -waveguide region is consistent with the approximate constancy of the electronic stopping power (and so track diameter). As expected, the waveguide thickness essentially coincides (except for a small broadening) with that of the initial β_1 -phase.

The resultant profiles are plotted in figure 4 where it can be seen that the flat refractive indices of both, the waveguide and the substrate at 488 nm, steadily and significantly decrease with the fluence although the change is greater at the surface. This information is better appreciated in figure 5, where the effective refractive indices N_m (solid symbols) are represented versus fluence ϕ . In the same plot the indices at the sample surface (open triangles) and the substrate (open rhombus) are included for comparison. It can be seen that N_m decreases appreciably and monotonously with fluence and a fine control of it up to $(\Delta N / \Delta \phi) \approx 2.5 \times 10^{-14} \text{ cm}^{-2}$ can be achieved, i.e. for a feasible low irradiation fluence of 10^9 cm^{-2} one would have $\Delta n \approx 2.5 \times 10^{-5}$.

3.2. α -phase waveguides

For these waveguides the study is less reliable due to the much smaller refractive index jump (and so the lower number of observable modes). In figure 6 the effective refractive index of the fundamental mode is plotted as a function of fluence. In fact, the first excited mode of the irradiated sample disappears after irradiation at a fluence of $4 \times 10^{11} \text{ cm}^{-2}$. As in the case of the β_1 -phase the effective refractive index decreases monotonously with fluence but it is worth noting that for any fixed ionic fluence, ΔN is smaller for the α -phase. For instance, at $2 \times 10^{12} \text{ cm}^{-2}$ and for the fundamental mode ΔN are 0.012 and 0.06 for α - and β_1 -phases, respectively. It is worth noting that the change in the refractive index should be accompanied by a reduction in the effective second order nonlinear optical coefficient, as previously reported for bulk

Figure 4. Calculated refractive index profiles for the β_1 -phase waveguide modified by irradiation with Cl 30 MeV ions at different fluences.

Figure 5. Effective refractive index of the three modes of the β_1 -phase waveguide modified by irradiation with Cl 30 MeV at different ion fluences ($\blacksquare$, $\bullet$, $\blacktriangle$), $\lambda = 488 \text{ nm}$. The refractive index of the waveguide (∇) and substrate ($\diamond$) obtained from the calculations assuming step-like profiles are also included (see figure 4).

Figure 6. Effective refractive index of the fundamental mode of the α -phase waveguide modified by irradiation with Cl 30 MeV ions at different ion fluences. Different symbols indicate different samples.

samples irradiated under similar conditions [13]. Anyhow, the work shows that a substantial fraction of the coefficient is preserved for the fluences used in the present work.

With regard to losses in our nanostructured α and β_1 -phase irradiated waveguides one should expect that they are enhanced in comparison with those of unirradiated proton-

exchange waveguides (0.2–0.4 dB cm⁻¹). An estimate of the track contribution to losses can also be inferred from measurements performed on waveguides that are directly formed by irradiation with Cl at 46 MeV in LiNbO₃ (see [13]). For relatively high fluences, above 10¹² cm⁻², losses of around 10 dB cm⁻¹ have been measured, which should be mostly associated with the presence of the nano-tracks.

4. Discussion of optical results

The main trends obtained for the modification of the refractive index of the waveguides can be qualitatively understood in line with the formation of the nanometre-size amorphous regions generated by the ion impacts [3–5, 7]. Since the electronic stopping power of the ion is above the amorphization threshold at the surface (see figure 1), amorphous tracks ($n_a = 2.14$ for 488 nm) are generated under irradiation leading to a decrease in the average refractive index with regard to that of the substrate. This effect reaches both the waveguide and the substrate because the tracks are longer than 3 μm, whereas the waveguides have 1.1 μm (β_1 -phase) and 3 μm (α -phase) depths, respectively.

The decrease in the effective refractive index, both in the waveguide and in the substrate, can be described with a simple effective-medium model for the dielectric constant of the irradiated material. With the fluences used in our experiments ($\phi \geq 10^{10}$ cm⁻²) the average distance between tracks (100 nm for $\phi = 10^{11}$ cm⁻²) is well below light wavelength and so the concept of an effective refractive index makes sense. Due to the large length of the tracks in comparison with the waveguide thickness and the approximate constancy of the stopping power in that depth one may reasonably assume cylindrical tracks (diameter independent on depth). For every irradiation fluence the square of the refractive index $\bar{n}_e(\phi)^2$ (i.e. the dielectric permittivity) should be an average between the values for the amorphized material, n_a^2 , and for the non irradiated waveguide n_e^2 ,

$$\bar{n}_e(\phi)^2 = n_e^2 - f_a(\phi)(n_e^2 - n_a^2), \quad (1)$$

where n_e and n_a are the refractive indices of the waveguide at the surface and of the amorphous LiNbO₃ respectively, and $f_a(\phi)$ the fraction of the material that has been amorphized. In fact, this has approximate validity since it has recently been shown that one should also take into account the contribution to the refractive index of the disordered region around the track core [7]. However, the effect is relatively small, (around 10–20%) and so it can be neglected in a first approximation. Therefore, the change in the refractive index of the irradiated with regard to the initial material depends on two factors, the difference ($n_e^2 - n_a^2$) and the amorphous fraction $f_a(\phi)$. The first one is a known quantity that obviously does not depend on fluence. Its value is 0.54 for the α - and 1.04 for the β_1 -phase. Then, it is greater for β_1 waveguides contributing to the observed higher modification under irradiation. In turn, the second factor f_a does depend on irradiation fluence and it is not known *a priori*. Nevertheless, it can be obtained from (1) as

$$f_a(\phi) = \frac{\bar{n}_e(\phi)^2 - n_e^2}{n_e^2 - n_a^2}. \quad (2)$$

Figure 7. Amorphous fraction generated in β_1 -phase waveguide and LiNbO₃ substrate by irradiation with Cl (30 MeV) as a function of fluence obtained from optical measurements (○ and □, left axis). The disorder fraction obtained from RBS/C measurements versus the fluence (△, right axis) for the β_1 -phase has been also included (see section 5).

Then, one can calculate the amorphous fraction for the β_1 waveguide and the substrate using the measured profiles plotted in figure 4 that provide $\bar{n}_e(\phi)$. Results for $f_a(\phi)$, displayed in figure 7, are essentially concordant for the two regions without any systematic significant deviations. This is consistent with the generation of core tracks having the same diameter in, both, the waveguide and the substrate. Moreover, a roughly linear dependence of f_a with fluence can be observed consistent with non-overlapping track regime. From the slope of the linear fit and taking into account that $f_a = \phi \pi r^2$, the radius of amorphous track can be estimated as ~ 2 nm. This value is similar to that reported for bulk LiNbO₃ in a previous work [7].

5. RBS channelling data and analysis

In order to gather additional information on the disorder induced by ion-beam irradiation and compare it with that measured optically, RBS channelling (RBS/C) spectra, using 3.8 MeV protons, have been taken on the proton-exchange waveguides. Results are summarized in figure 8, where the Nb spectra corresponding to channelling and random incidence are included for a β_1 -phase waveguide, figure 8(a) and for an α -phase waveguide, figure 8(b). The spectrum for a LiNbO₃ substrate irradiated with a fluence of 10¹² cm⁻² is also plotted for comparison. Several features are inferred from the figure. First, one observes that the α waveguides behave under irradiation in a similar way to the bulk crystal. This is the expected behaviour taking into account that their structure is very similar to the bulk crystal [11]. On the other hand, the channelled spectrum for the unirradiated β_1 waveguide shows a clear peak in the energy yield at the sample surface. The width of the peak is consistent with the thickness of the β_1 waveguide (~ 1 μm) as measured by the dark-mode method. The occurrence of the peak and the decrease in yield at the waveguide-substrate boundary should be attributed to a slight misorientation of the channels in the two structures. Taking into account the typical width of the channelling dips in LiNbO₃ the misfit angle should be around 0.1°, so that channelling is still quite good in the β_1 -phase. The lattice

Figure 8. RBS/channelling spectra (3.8 MeV protons) of irradiated β_1 -phase (a) and α -phase (b) waveguides with three ion fluences: (1) $4 \times 10^{11} \text{ cm}^{-2}$, (2) $1 \times 10^{12} \text{ cm}^{-2}$ and (3) $2 \times 10^{12} \text{ cm}^{-2}$. The spectra of the non irradiated waveguides (0) and of a full amorphous sample (random) are also included for comparison. Also for comparison, the spectrum for a LiNbO_3 substrate irradiated with a fluence of 10^{12} cm^{-2} (----) is plotted.

misfit is not surprising if one considers the relative changes in lattice parameters and strains in the waveguide (in the range 10^{-3} – 10^{-2}) [11].

Additionally, the fraction of disorder induced by irradiation in the β_1 -phase can be estimated from the RBS/C yields from figure 8(a) and it is included (as triangles) in figure 7 for comparison. In the waveguide region, close to the surface, intrinsic dechannelling effects are not expected to be significant. It appears that the fraction measured from RBS/C spectra is much larger (a factor 2–3) than inferred from the optical data for the β_1 -phase. In fact, this result is not surprising if one takes into account that the amorphous core track is surrounded by a more extensive halo constituted by isolated defects [7]. Although these defects may have a significant effect on the RBS/C channelling yield due to heavy distortion and blocking of the crystallographic channels, they do not substantially modify the refractive index of the crystal. In fact, the halo is expected to have a radius around 2–3 times that of the core [7], in agreement with our present data (see figure 7). This result emphasizes that the type of disorder to which this technique is sensitive differs from that affecting optical measurements. This point may be quite relevant when comparing amorphization thresholds reported in the literature.

6. Summary and conclusions

Ultra-fine tuning of the refractive index of alpha and beta phase waveguides by means of swift-ion irradiation at low fluences (5×10^{10} – $5 \times 10^{12} \text{ cm}^{-2}$) has been demonstrated. The variations of the waveguide refractive index have been characterized by the dark mode technique by determining the effective mode refractive indices. For the β_1 -phase the approximate modified refractive index profiles have also been calculated and satisfactorily compared with the predictions of a simple effective-medium model for the dielectric response. From this model, a radius of 2 nm is estimated for the amorphous tracks. Moreover, the amount of disorder introduced by irradiation, obtained from the RBS/C results, may appear consistent with the optical data if one takes properly into account the occurrence of the halo surrounding the amorphous track core. In summary, swift-ion irradiation provides a reliable and reproducible method to directly pattern the refractive index of a dielectric crystal.

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