

# Novel and nanostructured optical waveguides by means of using electronic damage of swift heavy ions.

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**Abstract:** *We present a review and the new results regarding a new and very efficient method for the fabrication of novel optical waveguides in crystalline materials by means of using the processing capability of the high electronic excitations provided by high-energy ( $\sim 2\text{-}50$  MeV) heavy ( $A > 8$ ) ions. The control of the electronic damage both in intensity (i.e. keV/nm) and in spatial distribution (depth and lateral in single nano-track regime) allows the fabrication of new optical structures at moderate ( $\sim 10^{14}$  cm $^{-2}$ ) or even ultra-low fluences ( $\sim 10^{12}$  cm $^{-2}$ ). It is a clear alternative to the standard procedure (based on using the nuclear damage caused by light ions) which requires excessively high fluences ( $10^{16} - 10^{17}$  at/cm $^2$ ). Moreover new nanostructured waveguides are feasible with potential for size-controlled nanopores integrated in waveguides for novel optical devices.*

## Introduction

Optical waveguides are fundamental elements of many photonic devices. Various crystalline materials have been intensively studied for such broad field depending on the particular application. Ion-beam irradiation offers a general and flexible method to modify the structure and properties of materials with several advantages over other physical or chemical methods. So far, the technological applications of ion irradiation mostly rely on the effects caused by nuclear collisions and ion implantation. In particular, ion implantation of transparent materials with light ions (H and He) of a few MeV's energy has allowed for the fabrication of optical waveguides and a variety of integrated optics devices [1]. Particular attention has been given to lithium niobate (LiNbO $_3$ ) due to its excellent electrooptic and nonlinear optical properties and that large good quality crystals can be routinely grown at a reasonable cost. Essentially, the mechanism proposed to account for the generation of the waveguiding layer is the amorphization of the region at the end of the ion range as a consequence of nuclear collisions and ion implantation. It presents a markedly reduced refractive index in comparison to the unirradiated material and behaves as an optical barrier. However, this is at the expense of using quite high fluences ( $10^{16}\text{-}10^{17}$  cm $^{-2}$ ) that strongly hinders the practical usefulness of the method.

Recently, the use of heavier ions and higher energies to cause structural damage and modify the physical

response is starting to be explored. A variety of ions at energies ranging from 3 MeV to 50 MeV have already been investigated in LiNbO $_3$  [2-3]. Under these irradiation conditions the electronic stopping power becomes dominant over the nuclear stopping power. As a consequence of these studies it has been ascertained that the induced damage is mostly related to electronic excitation mechanisms. The features associated to these processes markedly differ from those applying to nuclear collisions. In particular, full amorphization of the lattice can only be achieved when the electronic stopping power is above a certain threshold (*thresholding behavior*) [4].

So far, most studies have dealt with the generation of latent (amorphous) tracks by single swift-ion (energies  $\geq 0.1\text{-}1$  MeV / amu) impacts in insulating materials which is well-documented [5-8]. Amorphous tracks with nanometric radius are generated along their trajectory when the electronic stopping power  $S_e$  is above a certain threshold  $S_{th}$  ( $\sim 5$  keV/nm for LiNbO $_3$ ). The radius of the track is a growing function of  $S_e$  and so it can be varied through the irradiation conditions.

Here we present a review and some new results regarding the electronic excitation damage by swift ions and its potential application to Photonics [4,7,8,10-15]. In particular, irradiation with high-energy ( $\sim 2\text{-}50$  MeV) medium-mass ( $A > 8$ ) ions has been shown to be a very efficient method for the modification of crystalline materials and fabricating novel optical waveguides. This results from the processing capabilities of the high electronic excitations provided by swift ions through the control of beam energy and intensity or better the electronic stopping profile inside the crystal. At variance with the classical implantation method the new optical structures can be produced at moderate ( $\sim 10^{14}$  cm $^{-2}$ ) [10] or even ultra-low fluences ( $\sim 10^{12}$  cm $^{-2}$ ) [13]. Most results (experimental and detailed analysis) have been obtained with LiNbO $_3$  and the potential of universal applicability to other crystalline materials has been explored with KGd(WO $_4$ ) $_2$  (KGW) [11].

## Experimental

Ion irradiations were performed with the 5 MV tandem accelerator recently installed in the Centro de Microanálisis de Materiales (CMAM) at University Autónoma of Madrid [9]. Maximum ion beam cur-

rents were in the range 10-50 nanoamps to prevent excessive charging and heating. Samples were tilted  $9^\circ$  with respect to beam direction to avoid ion channeling. The ion beam was overfocused and expanded so that, by means of a mask, only the central homogeneous part of the beam is used. The dimensions of the irradiated area were typically  $6 \times 6 \text{ mm}^2$ . The beam homogeneity, better than 10 %, was checked by means of measuring the ion induced luminescence in a  $\text{SiO}_2$  glass sample with a sensitive CCD camera. The main material studied has been congruent  $\text{LiNbO}_3$  having integrated optics quality purchased from PHOTOX and CASIX. KGW slides were purchased from Passat Ltd., Ontario, Canada.

The prism coupling dark m-lines method has been used to determine the refractive index profiles induced by the irradiations. By proper selection of TE and TM mode coupling and sample orientation under the prism, all the main refractive indices can be measured separately (two for the birefringent  $\text{LiNbO}_3$  and three for the biaxial KGW).

### Buried amorphous layers: High index jump optical waveguides

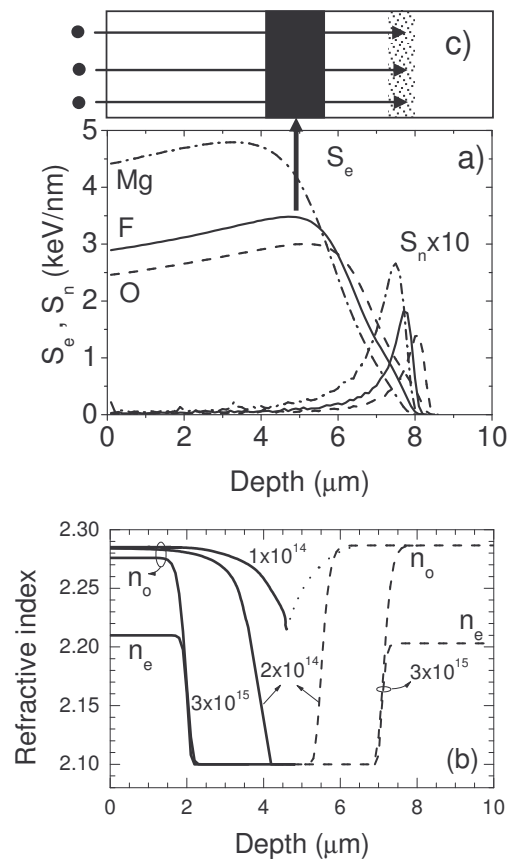
The novel strategy followed in these experiments is illustrated in Fig. 1. One looks for irradiation conditions where the maximum of the electronic stopping power  $S_e(z)$  curve, lies at a certain depth inside the crystal (well separated from the peak value of the nuclear stopping power  $S_n(z)$ ) and reaches a value close to the intrinsic threshold ( $S_{th} \approx 5 \text{ keV/nm}$ ). Our irradiation conditions F at 22 MeV, O at 20 MeV and Mg at 28 MeV fulfill this requirement as illustrated in Fig. 1a), derived from SRIM 2003 calculations. Then, it is expected that, after some *incubation fluence*, amorphization at every single ion trajectory starts at the location of the maximum of  $S_e(z)$  so that the process would proceed as illustrated in Fig. 1b): When the fluence guarantees full overlapping of the individual latent tracks ( $\sim 10^{13} \text{ cm}^{-2}$ ) a low refractive index homogeneous layer is formed inside the crystal and consequently a surface planar optical waveguide is generated at the surface.

For sufficiently high fluences the profile approaches a step-like shape, whose bottom level reaches a limit value,  $n = 2.10$ , approximately independent of the investigated ion as well as of light polarizations and coincides with the refractive index of amorphous  $\text{LiNbO}_3$ .

Another interesting feature of those waveguides is the low level of propagation losses ( $\sim 1 \text{ dB/cm}$ ), quite comparable to that measured for conventional implanted waveguides [10].

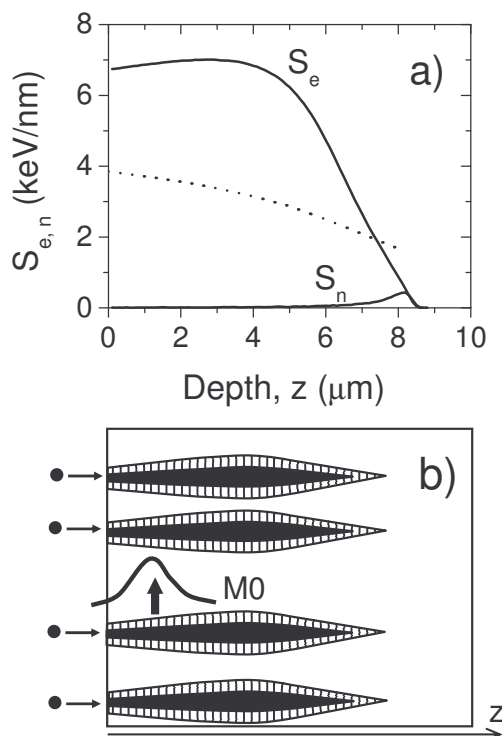
### Nanostructured optical waveguides at ultralow fluences

Next, we address the processing of  $\text{LiNbO}_3$  by means of swift-heavy ions in the very low fluence regime where single-track are not overlapping. In this case



**Fig. 1:** (a) Plot of the electronic and nuclear stopping power for F at 22 MeV (continuous line), O at 20 MeV (dashed line) and Mg at 28 MeV (dashed-dotted line); (b) Ordinary ( $n_o$ ) and extraordinary ( $n_e$ ) refractive index profiles obtained from the dark-mode data (solid lines) for some representative fluences. The estimated refractive index profile (using the low index optical resonances and RBS/channeling data) corresponding to the back amorphous-crystalline boundary is also shown with dashed lines (c) scheme, considering the F 22 MeV case for the illustration, of the generated buried amorphous layer (black strip) near the maximum of the electronic stopping curves. The dotted strip indicates the region of nuclear damage and implantation.

uniform layers are not formed but instead a nanostructured heterogeneous medium is achieved. By using optical methods (waveguide dark-mode analysis) we have learned about the depth morphology of the tracks and about the potential for tailoring the optical response of the irradiated region and lead to optical waveguide fabrication. The strategy of our method is illustrated in Fig. 2 where the stopping power  $S_e(z)$  for Cl (46 MeV) calculated with SRIM 2003 is plotted as a function of  $z$ , Fig. 2a, together with a qualitative illustration of the (average) expected track profile, Fig. 2b. It includes a low-index amorphous core ( $n_a = 2.10$  at  $\lambda = 633 \text{ nm}$ ) and a defective surrounding halo. Following the  $S_e(z)$  curve the track radius should increase with depth up to a maximum and then decrease with increasing  $z$ . In Fig



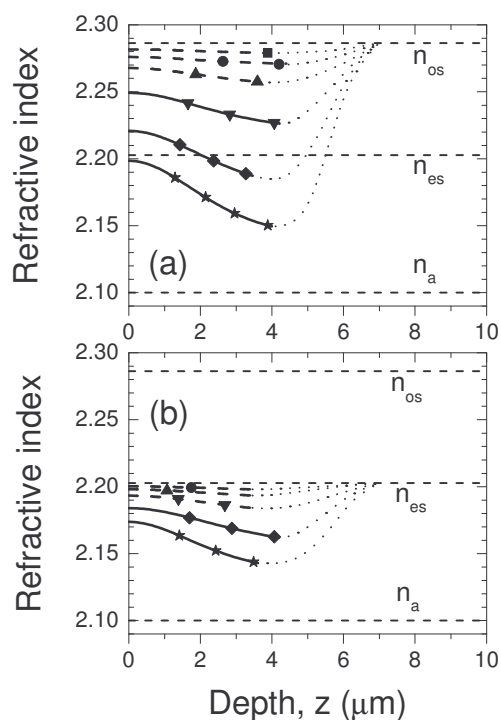
**Fig. 2:** a) Electronic,  $S_e$ , and nuclear,  $S_n$ , stopping power curves for Cl 46 MeV in LiNbO<sub>3</sub> calculated with SRIM2003. The dotted line shows the corresponding “amorphization” threshold curve. b) Schematic depth morphology of the tracks, showing the core (black) and the surrounding halo (dashed). An schematic light profile (M0) illustrates waveguiding behavior.

2a) the dotted line shows the predicted (“amorphization”) threshold according to recent data, that include the effect of ion velocity. The measured ordinary and extraordinary refractive index profiles,  $n_o(z)$  and  $n_e(z)$  induced by the Cl (46 MeV) irradiations are shown in Fig. 3a and Fig. 3b, respectively. It has been found that tracks are composed of an amorphous core surrounded by a *damaged (pre-amorphous) halo*. The measured radius of the track core at the surface shows a reasonable accordance with previously reported data.

The functionality of this novel waveguides made by partial amorphization of the LiNbO<sub>3</sub> crystal is a matter of concern. The measured SH coefficient ( $d_{33}$ ) vs. fluence is shown in Fig. 4. It is normalized to a virgin substrate. A remarkable high activity (> 60%) is still preserve at the fluences where optical waveguides are obtained. The gradual decrease of the  $d_{33}$  coefficient is logical following the increase in the amorphous fraction at the surface.

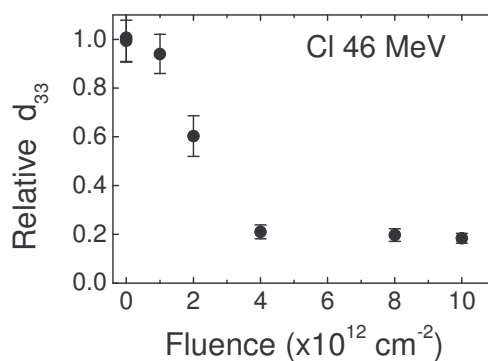
### Conclusions

By using the electronic excitation provided by high-energy medium-mass (i.e.  $A > 8$ ) ions it is possible to produce various novel optical waveguides, by playing with the shape of the electronic stopping curve. By using curves with a maximum stopping value



**Fig. 3:** Ordinary, a), and extraordinary, b), refractive index profiles (solid lines) at  $\lambda = 633$  nm for X-cut LiNbO<sub>3</sub> samples irradiated with Cl 46 MeV ions at the fluences (at/cm<sup>2</sup>) of:  $5 \times 10^{11}$  (squares),  $1 \times 10^{12}$  (circles),  $2 \times 10^{12}$  (up triangles),  $4 \times 10^{12}$  (inverted triangles),  $8 \times 10^{12}$  (rhombi), and  $1 \times 10^{13}$  (stars). Horizontal dashed lines show the refractive index values for the crystal ( $n_{os}$  and  $n_{es}$ ) and amorphous ( $n_a$ ) regions. A guess of the expected refractive index profiles behind the minimum is also plotted with a dotted line.

inside the crystal, buried amorphous layers are generated which produce good quality high index-jump step-like optical waveguides. This method requires the use of moderate fluences ( $\sim 10^{14}$  cm<sup>-2</sup>) to generate overlapping individual amorphous tracks. Using the same strategy but ultra-low fluences of  $\sim 10^{12}$  cm<sup>-2</sup>, i.e. in the isolated tracks regime, a nanostructured optical medium with an effective gradually decreasing refractive index profile is produced. This profile



**Fig. 4:** Fluence evolution of the second order non-linear optical coefficient  $d_{33}$  relative to the one of virgin LiNbO<sub>3</sub> for the LiNbO<sub>3</sub> samples irradiated with Cl 46 MeV ions.

supports waveguide modes and shows a high second order optical nonlinearity. Note that in all cases the fluences required are several orders of magnitude below those ones needed in standard implantation with light ions. The physical mechanisms and theoretical basis for the observed behavior have been summarily described.

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