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Influence of iron doping on spatial soliton formation and fixing in lithium niobate crystals

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ABSTRACT

We analyse the feasibility of using iron-doping in lithium niobate in order to stabilize and permanently fix light-induced integrated structures. General 3D optical interconnections were realized in bulk lithium niobate crystals by means of soliton waveguides exploiting the enhanced photorefractive properties obtainable using specific iron doping. We report an enhancement of the photorefractive properties in doped crystals that can be considered for permanently fixing the integrated circuits. This work opens new directions for realizing permanent self-assembled and self-aligned integrated electro-optic devices and photonic circuits.

1. Introduction

The continuously growing demand for data storage and processing stimulated the development of high-data-rate fibre-optic communication systems which in turn has created a need for high-capacity repeaters and terminal systems for processing optical signals and, therefore, a need for high-speed photonic switches. Similarly, the potential for optical computing can only be realized if large arrays of fast photonic gates, switches, and memory elements are developed. On the other hand several important technological sectors such as space industry, environment, biology and military applications would demand for the integration of devices able to store and process information at very high bit-rates in compact and stable integrated devices.

Photonic interconnections may be realized by use of optical waveguides with integrated-optic couplers or fibre-optic couplers and micro-lenses: the optical connections have also the option (with no analogy in electronic devices) to connect a large number of elements by using the propagation of light beams themselves. Conventional optical components may be used to create interconnection maps with simple patterns, such as shift, fan-in, fan-out, magnification, reduction, reversal, and shuffle. However, the full potential of the "optical" approach to information technology can be exploited only if it is available a way to create arbitrary interconnection networks. A way to arbitrarily pilot light signals is constituted by holographic control of the propagation of a beam [1–4]. An elementary volume phase hologram is, in general, an optical element where a periodic modulation of its refractive index is present. This modulation can be achieved by exploiting the photorefractive properties of a medium, i.e. by inducing the refractive index variation by proper inhomogeneous illumination. In this context, lithium niobate is known to be photorefractive in the visible spectral range: its photorefractive sensitivity (the maximum achievable refractive index change for a given light intensity) can be increased of many times by doping the material with metals possessing two states of valence, such as Fe^{2+}/Fe^{3+} since iron ions act as donors and traps of electrons [5,6], key parameters in the photo-excitation processes that drive photo-refractivity [7–9]. Lithium niobate technology has therefore the potential to include holographic stages into integrated optical circuits as already reported in bulk doped Fe:$LiNbO_3$ crystals by Ti in-diffusion [10]. Some data were reported also on the diffusion of Fe [11,12] and co-diffusion with Ti in $LiNbO_3$ crystals [13] with the purpose of realizing an Infrared integrated filter by means of a holographic grating. Most recent results concerns the realization of photorefractive optical waveguides in Fe:LN by ion implantation [14]. Using these configurations, the first demonstrators of optical interconnections integrated optical circuits and sensors with a holographic stage have been already reported [15–20]. The main limitation of these optical processing schemes is that their full potential is nowadays difficult to miniaturize and integrate in a

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device of reduced size and insensitive to environmental perturbations. Optical processors are inherently two dimensional, so that optical interconnections have to be performed in parallel with a three dimensional geometry [21]. Moreover when holographic elements are used, alignment problems are critical and their design has to be realized with a very tight tolerance. In this scenario the realization of complex optical circuits with included holographic elements for data storage and processing on a single monolithic chip of doped lithium niobate will be strongly recommended. The possibility of realizing optical circuits directly in the volume of the crystals by using the photorefractive effect represents a valid alternative: the optical circuit can be obtained as the result of a self-writing process, where all the elements of the system are simultaneously written by interaction of a suitable light beam arrangement with the material. By this way, all the optical elements will result self-aligned, with no need for any accurate design or optimization. In order to reach this goal, the exploitation of photorefractive solitons can be an answer [22]. This approach consists in letting a light beam propagate inside a lithium niobate crystal while the photorefractive effect creates a positive variation of the refractive index along the propagation path. The light beam experiences then a self-focusing effect which become more and more effective, until a stable situation is reached: the beam propagates without diffracting, confined into a self-written waveguide. These solitonic waveguides are stable even after the beam is turned off, exhibit very low losses and have the advantage that they can be created anywhere in the bulk of the substrate [21,23–38].

In this work we present some results of a feasibility of fixing soliton waveguides using iron doping. The fixing problem is a very hot topics for soliton waveguide. In fact, if an efficient fixing of soliton waveguides would be indeed possible, such technology will become an important and efficient technique for realizing complex integrated photonic circuits able to perform all-optical as well as electro-optical signal processing using the enormous amount of nonlinear properties of lithium niobate crystals.

2. Sample preparation

Several iron doped lithium niobate (Fe:LN) crystals and a co-doped Zr,Fe:LN one were grown by the Czochralski technique and poled at high temperature to get mono-domain structure at the University of Padova. All the boules have a congruent composition: the iron doped crystals have a nominal dopant content varying from 0.05 mol.% up to 0.10 mol.%, while in the co-doped boule the Zr and Fe concentrations are 3 mol.% and 0.055 mol.% respectively. The growth direction was along the *z*-axis in order to achieve a single domain state through the whole sample volume. By exploiting the X-ray diffraction technique, they were oriented and subsequently cut in slices with the major surface perpendicular to the *y*-axis, with a tolerance better than 0.4°. The oriented Fe:LN slices were polished to achieve optical quality surfaces, by standard procedures using a Logitech PM5 lapping machine. Since after the growing process the iron present in the samples is mainly in the 3+ valence state, some crystals were reduced by a suitable and controlled treatment in a gas mixture of Ar (98%) + H_2 (2%) for different annealing times at 500 °C, thus varying the amount of Fe^{2+}. Since Fe:LN has an absorption band in the visible range which is proportional to the amount of only Fe^{2+} [39], optical absorption measurements were performed for each sample in order to determine their reduction degree Fe^{2+}/Fe^{3+}. The measurements were realized by using a Jasco V-670 spectrometer in transmission mode in the spectral range 330–850 nm and with light polarized perpendicular to the lithium niobate ferroelectric axis. By exploiting the absorption cross section at 532 nm [39,40], the amount of Fe^{2+} and Fe^{3+} of each sample was derived, as reported in Table 1.

Table 1
Fe^{2+} and Fe^{3+} concentrations of each samples.

Sample	Nominal [Fe] ($\times 10^{18}$ at/cm^3)	[Fe^{2+}] ($\times 10^{17}$ at/cm^3)	[Fe^{3+}] ($\times 10^{18}$ at/cm^3)	[Fe^{2+}]/[Fe^{3+}] (%)
142.3.10	18.8 (0.1 mol.%)	0.46 ± 0.15	18.8 ± 6.1	0.2 ± 0.1
142.3.1	18.8 (0.1 mol.%)	1.27 ± 0.16	18.7 ± 2.4	0.7 ± 0.1
142.3.5	18.8 (0.1 mol.%)	5.80 ± 0.46	18.2 ± 1.5	3.2 ± 0.6
150.3	9.4 (0.05 mol.%)	6.47 ± 0.46	8.75 ± 0.96	7.4 ± 0.9
112.5.1	1.8 (0.01 mol.%)	0.50 ± 0.01	1.73 ± 0.32	2.8 ± 0.7
117.8.1	1.2 (0.007 mol.%l)	0.30 ± 0.01	1.16 ± 0.31	2.6 ± 0.7
117.12.1	1.2 (0.007 mol.%)	8.60 ± 0.02	0.39 ± 0.01	253 ± 9
117.12.2	1.2 (0.007 mol.%)	8.72 ± 0.02	0.39 ± 0.01	266 ± 9
122.4.1	0.9 (0.005 mol.%)	0.40 ± 0.01	0.86 ± 0.21	4 ± 1

3. Photorefractive properties

The photorefractive characterization of the material was performed by exploiting a depth-resolved experimental set-up as described in [41], i.e. combining a two-beam coupling technique with focused writing beams. In this way holograms with micrometer sizes were written in different areas of the sample, allowing the photorefractive response of the material to be spatially related to the iron concentration and the reduction degree. The scheme of the setup is shown in Fig. 1.

The light of a diode-pumped solid-state (DPSS) laser ($\lambda = 532$ nm) is expanded and split into two beams by using two lenses and a pseudo-Mach–Zehnder arrangement. After the second beam splitter, the two beams are slightly divergent and, after passing through a horizontal polarizer, they are parallelized by using a cylindrical plano-convex lens, which directs them towards a microscope objective. The cylindrical lens has the property to compress the beams only in the horizontal direction, keeping them unaltered in the vertical one. In this way, at the focal plane of the objective, the two beams are tightly focused in the vertical direction, presenting in this direction a waist of a few micrometers. The sample is mounted on a three-axis translator system with its *z*-axis parallel to the beam propagation direction, thus the grating vector K (of spacing $\Lambda = (5.2 \pm 0.3)\mu m$) and the polarization of the beams are parallel to the ordinary axes of the crystal. Behind the crystal an objective lens is used to collect the two beams and, with the help of a specially designed mechanical chopper and a lock-in amplifier, it is possible to monitor the time evolution of the diffracted part of one beam by periodically stopping for a short time the other one. The vertical and lateral sizes of the interference area are respectively (9.3 ± 0.8) μm and (57.6 ± 1.5) μm (at $1/e^2$ for both directions), therefore the maximum intensity used to record the holograms is $(16 \pm 2) \times 10^4$ W/m^2 (for a total of 270 μW of power on the sample) thus allowing us to use the theory of the one center band-transport model to describe the photorefractive behavior of our samples [42,1]. The hologram efficiency η, defined as the ratio between the intensities of incident beam (I_i) and the diffracted one (I_d), can be used to evaluate the value of the refractive index change at each time, by exploiting the Kogelnik formula [43]

$$\eta(t) = \frac{I_d(t)}{I_i} = \sin^2\left(\frac{\pi \Delta n(t) L}{\lambda \cos\theta}\right) \quad (1)$$

where L is the hologram thickness and θ is the half-angle between the two writing beams. In this way the time evolution of the refractive index change is derived. Finally, by combining the well-known formula of the electro-optic effect and the simplified solutions of the Kukhtarev equations, it is possible to fit the experimental data with the following equation