Effects of swift argon-ion irradiation on the proton-exchanged $LiNbO_3$ crystal*

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A proton-exchanged $LiNbO_3$ crystal was subjected to 70-MeV argon-ion irradiation. The lattice damage was investigated by the Rutherford backscattering and channeling technique. It was found that the lattice disorder induced by the proton exchange process was partially recovered and the proton-exchanged layer was broadened. It indicated that the lithium ions underneath the initial proton-exchanged layer migrated to the surface during the swift argon-ion irradiation and supplemented the lack of lithium ions in the initial proton-exchanged layer. This effect was ascribed to the great electronic energy deposition and relaxation. The swift argon-ion irradiation induced an increase in extraordinary refractive index and formed another waveguide structure beneath the proton-exchanged waveguide.

Keywords: swift heavy ion, lithium niobate, optical waveguide

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1. Introduction

Lithium niobate ($LiNbO_3$) has long been used in a wide range of photonic devices due to its outstanding acousto-optic, electro-optic, and nonlinear properties. Devices such as electro-optical switches, power dividers, and frequency doublers have been used in optical information processing and optical communication.[1−3] The proton exchange is a conventional method used to fabricate optical waveguide structures in $LiNbO_3$ crystals. The replacement of lithium ions with protons takes place when a $LiNbO_3$ crystal is immersed in a benzoic acid melt at 200–250 °C,[4] which induces an increase of the refractive index at the surface and forms a waveguide. The lattice deformation and the nonlinear coefficient degradation induced by the proton exchange process can be partially recovered by thermal annealing.[5] The second-harmonic generation with high conversion efficiency has been achieved in periodically poled $LiNbO_3$ waveguides.[6,7]

The swift heavy-ion (SHI) irradiation has been widely used to modify various physical properties of materials. The passage of SHIs through a target material mainly leads to electronic excitation and ionization. This high electronic energy deposition has been confirmed to play an important role in many spectacular effects, such as amorphous track formation[8,9] and phase transformation.[10] Additionally, the SHI irradiation leads to the crystallization of amorphous alloys,[11] polymers,[12] and semiconductors.[13−16] It has been shown that the damage caused by 700-keV I ions implanted in SiC crystals can be almost completely washed out after the 827-MeV Pb-ion irradiation.[16]

In this study, we investigated the effects of SHI irradiation on a proton-exchanged $LiNbO_3$ crystal. Our main effort focused on the measurement of lattice disorder in the proton-exchanged layer and the waveguide properties before and after the SHI irradiation.

2. Experiments

An x-cut, optically polished $LiNbO_3$ crystal was immersed in a benzoic acid melt at 230 °C for 10 min. Then, it was cut into two pieces. One was subjected

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to 70-MeV Ar^{8+}-ion irradiation six months later. The other was kept at room temperature for comparison. The SHI irradiation was carried out at the Heavy Ion Research Facility in Lanzhou (HIRFL). The fluence was 1×10^{12} ions/cm^2. The initial beam had an energy of 792 MeV and was slowed down by passing through a 293-μm-thick Al foil.

Both samples were characterized by Rutherford backscattering and channeling (RBS/C) analyses using 2.1-MeV He^+ ions as the probing beam delivered by a 1.7-MV tandem accelerator at Shandong University.

The optical modes of the two samples were detected with a Metricon 2010 prism coupler. Transverse electric-polarized (TE-polarized) light from a 633-nm He–Ne laser struck the base of the rutile prism. The intensity of the light reflected from the base of the prism was detected and plotted as a function of the incident angle, a sharp drop in the intensity profile corresponded to a propagation mode.

Two end facets of the samples were carefully polished for end-face coupling experiments. The light from a 633-nm He–Ne laser was injected into and out of the waveguides through two 25× microscope objective lenses. The samples' output facets were imaged onto a charge-coupled device (CCD) camera recording the fields coupled out of the waveguides.

The sample without the SHI irradiation was post-baked at 230 °C for 20 min and then analyzed by all of the techniques mentioned above for comparison.

3. Results and discussion

3.1. Lattice disorder recovery

The channeling spectra of proton-exchanged $LiNbO_3$ (PELN) and swift-heavy-ion-irradiated PELN (SHII-PELN) are depicted in Fig. 1. The channeling and the random spectra of a pristine $LiNbO_3$ crystal are also depicted in Fig. 1 for damage calculation. The crystal lattice was deformed in the proton-exchanged layer where many lithium ions were replaced by protons.

The spectrum of the SHII-PELN sample shows two effects of swift argon-ion irradiation on PELN. i) The lattice disorder in the proton-exchanged layer was partially recovered. ii) The proton exchange process extended deeply into the crystal. Lithium ions underneath the proton-exchanged layer were replaced by protons during the swift argon-ion irradiation, migrated from the inside to the crystal's surface, and supplemented the lack of lithium ions in the initial proton-exchanged layer. Therefore, the pre-damaged crystal lattice was recovered.

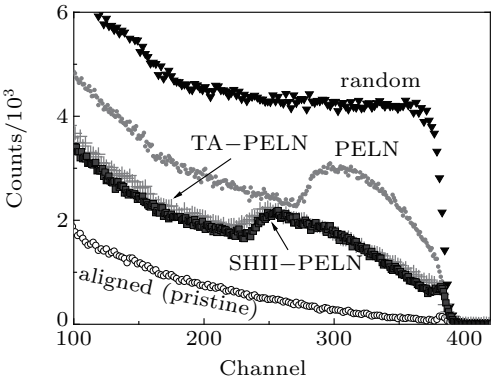


Fig. 1. RBS/C spectra of PELN, SHII-PELN, and TA-PELN samples.

The channeling spectrum of the PELN sample after the thermal annealing (TA-PELN) was recorded and is depicted in Fig. 1 by crosses. The thermal treatment reduced the lattice disorder in the $LiNbO_3$ crystal and broadened the proton-exchanged layer, just as the swift argon-ion irradiation was observed to do.

The depth profiles of the displaced Nb atoms were extracted from the RBS/C spectra. This was achieved by using a two-beam approximation[17] with the dechanneling estimated by a multiple scattering function. The calculations were implemented in an iterative procedure starting from the sample's surface. The obtained disorder profiles are shown in Fig. 2. The PELN sample had a damage peak with the maximum at 55% of the relative disorder. After the swift argon-ion irradiation or the thermal annealing, the disorder profiles appeared similar. The maximum relative disorder was reduced to about 30%, and the proton-exchanged edge was propelled from the depth of 0.65 μm to the depth of 0.9 μm.

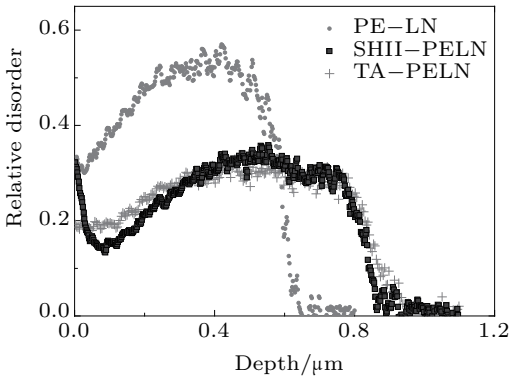


Fig. 2. Damage profiles calculated from RBS/C spectra of PELN, SHII-PELN, and TA-PELN samples.

The profiles of electronic and nuclear energy deposition of 70-MeV argon ions in a $LiNbO_3$ crystal were simulated by the SRIM 2006 program[18] and are shown in Fig. 3. The swift argon ions mainly caused electronic excitation in the $LiNbO_3$ crystal. Nuclear collisions did not dominate in the proton-exchanged layer, which was at the surface. It is reasonable to ascribe the recrystallization effect of the SHI irradiation to the massive electronic energy deposition.

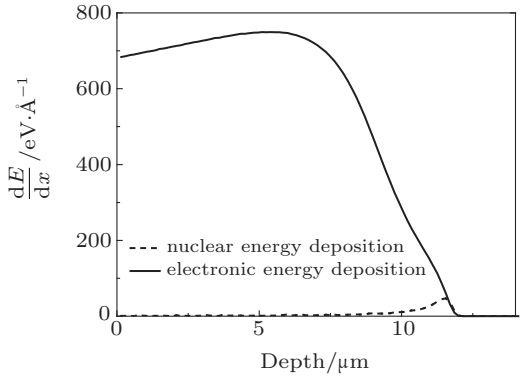


Fig. 3. Depth distributions of electronic and nuclear energy deposition of 70-MeV argon ions in $LiNbO_3$ crystal simulated by SRIM 2006 program.

The kinetic energy of the incoming ions was deposited almost exclusively via excitation and ionization of the electronic system of the solid. The relaxation of energy occurred in the electronic system through electron–electron collisions before the energy was transmitted to the lattice atoms by electron–phonon interactions. According to the thermal spike model, an equilibration of the electronic system takes place first (10^{-15}–10^{-14} s), and then the heating of the lattice takes place (10^{-14}–10^{-12} s) along the ion track.[15] It seems that for the pre-damaged lattice, the electronic energy deposition may act like the conventional thermal treatment, which excites the atoms in random directions. The thermal spike model is frequently invoked to explain many effects induced by swift heavy ions, although the temperature increase around the ion track is not easy to define because the energy deposition is a nonequilibrium process. In this study, the SHI irradiation did not wash out the lattice disorder completely as it did in Ref. [16]. The reason may be that the electronic energy deposition was not high enough or the fluence was too low. Moreover, there was a lack of lithium ions in the proton-exchanged layer.

3.2. Waveguide properties

There was a waveguide structure in the PELN sample with an extraordinary refractive index n_e. The TE-polarized light propagating along the y-direction in the x-cut $LiNbO_3$ had a refractive index of n_e. The prism coupling result of the PELN waveguide is depicted in Fig. 4(a). It shows two modes (TE_0 and TE_1) in the PELN waveguide with effective indices of 2.2818 and 2.2038. The prism coupling results of PELNs after the swift argon-ion irradiation and the thermal annealing are depicted in Figs. 4(b) and 4(c), respectively. There are also two modes in both cases. The effective indices of the modes are 2.2874 and 2.2277 in Fig. 4(b), and 2.2810 and 2.2242 in Fig. 4(c). Both the swift argon-ion irradiation and the thermal annealing raised the index of the TE_1 mode to similar values. However, the swift argon-ion irradiation raised the TE_0 mode, while the thermal annealing decreased it slightly.

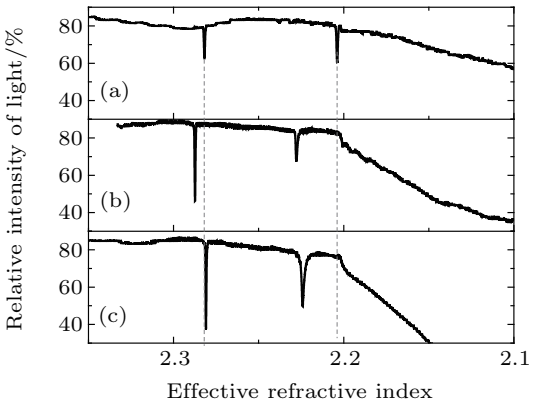


Fig. 4. Measured relative intensities of the light reflected from the prism versus the effective refractive index of the incident light for (a) PELN, (b) SHII-PELN, and (c) TA-PELN samples.

Near-field intensity distributions exiting the output facet of the SHII-PELN waveguide were recorded by a CCD camera in the end-facet coupling experiment and are shown in Figs. 5(a)–5(c). They were obtained by moving the sample up and down with the laser and the objective held fixed. It can be seen that there are two waveguide structures in the SHII-PELN sample. Figure 5(a) shows a narrow mode that is similar in size to the mode profile of the TA-PELN sample (not shown in this paper). Therefore, it is the TE_0 mode of the proton-exchanged layer. The mode in Fig. 5(c) is wider than the TE_0 mode but is compatible with the range of argon ions. This demonstrates that the swift argon-ion irradiation alone also formed

a waveguide structure, which means that the high electronic energy deposition induced an increase of n_e in the $LiNbO_3$ crystal. This may explain why the two modes had indices slightly higher in the SHII-PELN sample than the two modes in the TA-PELN sample. Figure 5(b) shows that the two waveguides can be excited together with the incident beam impinging upon an appropriate position of the input facet.

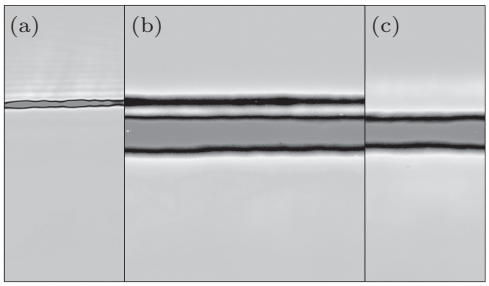


Fig. 5. Field intensity distributions of TE_0 mode of (a) the proton-exchanged waveguide and (c) the SHI-irradiated waveguide exiting the output facet of the SHII-PELN sample. Two modes are excited at the same time in panel (b).

4. Conclusion

In summary, we analyzed the effects of the swift argon-ion irradiation on the proton-exchanged $LiNbO_3$ crystal. The lattice disorder was measured by the RBS/C technique. The results show that the swift argon-ion irradiation affected the deformed lattice in a manner similar to that of thermal annealing. The lattice disorder was partially recovered, and the proton-exchanged layer was broadened, which indicates that the lithium atoms migrated along the direction opposite to the incoming ions during the swift argon-ion irradiation. This phenomenon was ascribed to the massive electronic energy deposition and relaxation within the electron system before the energy was transmitted to the lattice. Unlike the thermal annealing, the recrystallization induced by the SHI irradiation took place at room temperature and was achieved within several minutes. Additionally, the high electronic energy deposition induced an increase in n_e and formed a waveguide structure underneath the proton-exchanged layer.

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