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Micro structuring of $LiNbO_3$ by using nanosecond pulsed laser ablation

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Abstract

In this work, we report on the fabrication and characterization of surface microstructures in lithium niobate thin films and single crystal by using KrF excimer laser ablation technique at 248 nm with 6 ns pulse width. Ablation is carried out through a mask projection set-up. Various experimental conditions are used in order to evaluate the potential of laser direct writing for photonic waveguides fabrication in lithium niobate. The surface morphology of the processed structures was studied by optical and atomic force microscopy. Laser processing mechanism is investigated by using micro-Raman spectroscopy.

Keywords: Laser micromachining; Laser ablation; Lithium niobate; Photonic structure

1. Introduction

Lithium niobate ($LiNbO_3$) is a ferroelectric material of great interest in the field of optics and telecommunications, due to its large electro-optic and non-linear optical coefficients [1]. It has been widely used as electro-optic modulator, wavelength converter and optical waveguide. Moreover, it is expected that this material might be of great interest as a photonic crystals thanks to its linear and non-linear optical properties.

Recently, several techniques have been reported for lithium niobate microstructuring such as reactive ion etching (RIE) [2], electron beam lithography (EB) [3], focused ion beam (FIB) [4] and light induced frustration of etching (LIFE) [5]. Pulsed laser ablation is one of the most commonly used methods for materials machining. It offers many benefits such as wide design flexibility, relatively high speed and low cost. For instance, laser machining has been proposed as an alternative technique to create photonic elements such as gratings and microholes [6]. Deshpande et al. [7] performed a study on femtosecond laser processing of lithium niobate surface and subsurface structures by using a Ti:sapphire femtosecond laser (800 nm, 300 fs). The investigation of the modification induced in z-cut congruent $LiNbO_3$ bulk crystal by femtosecond laser irradiations has been reported in ref. [8]. It was found that the utilization of an amplified Ti:sapphire laser system with a repetition rate of 1 kHz and energy per pulse of 1 mJ allows a precise control of the microstructure size. Generally speaking, the utilization of amplified femtosecond lasers allows athermic drilling processes which are needed for sub-micron structuring without relief modification at the periphery of the holes. However, cheaper and less complex lasers in the nanosecond regime can also be used. For instance, in ref. [9] the authors reported the fabrication of bulk-$LiNbO_3$ phase gratings with a 2 μm period and 110 nm depth by using a nanosecond excimer laser ablation (248 nm, 30 ns) through a phase mask. To improve the quality of the ablated patterns realized with a nanosecond KrF excimer laser on x-cut bulk lithium niobate surface, Chong et al. [10] deposited a 1 μm thick silicon dioxide overlayer on the sample prior to ablation.

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In this work, we report on the study of UV-laser-induced surface microstructures in $LiNbO_3$ bulk crystal and thin films. We aim to perform a direct writing of those structures by using a KrF excimer laser ablation process with 6-nanosecond pulse widths. We study the influence of several experimental conditions in order to find out the optimal set of parameters to obtain efficient photonic waveguides structures. Micro-Raman spectroscopy is used to identify and analyse the changes in the material structure induced by laser ablation. Nomarski optical microscopy as well as atomic force microscopy were used to characterize the surface morphology.

2. Experimental procedures

For our experiments, we use $LiNbO_3$ single crystal (from Crystal Tech) and $LiNbO_3$ thin film deposited at 490 °C onto (0 0 0 1) sapphire substrates using radio-frequency magnetron sputtering from a target constituted of a cold pressed pellet of a commercial $LiNbO_3$ powder. The sputtering unit was equipped with a radiofrequency generator operating at 13.56 MHz. In the experiments, the substrates were kept at a distance of 80 mm from the target. The power and the total pressure were maintained at 180 W and 4 N/m^2, respectively. Depositions were performed in a Ar:O_2 (60:40) gas mixture. Further details on $LiNbO_3$ thin films fabrication process are reported elsewhere [11].

Laser micromachining of $LiNbO_3$ thin films and bulk crystal is performed using a nanosecond KrF excimer laser system (ATLEX 300 SI Model) operating at 248 nm with a pulse repetition rate of 50 Hz and 6 ns pulse-width. The samples are mounted on a motorized *x*–*y* translation stage. All the process is performed in air under normal conditions of temperature and pressure. The sample ablation is carried out in a mask projection set-up. An aluminium-on-quartz mask pattern is directly imaged onto the samples to create 10 μm wide holes. The laser fluence is varied from 0.3 to 2.3 J/cm^2 for each pulse, while the number of pulses varies from 1 to 100 in order to control the holes depth in $LiNbO_3$ thin film and single crystal. After laser ablation, the surface morphology and the depth of the patterned structure are examined by a Nomarski optical microscope and atomic force microscope (AFM). Finally, the induced structural changes of the ablated area are characterized by using μ-Raman spectroscopy.

3. Results and discussions

Prior to investigate the ablation process, we determine the crystalline structure and the quality of $LiNbO_3$ thin film deposited on sapphire by using X-ray diffraction (XRD). $LiNbO_3$ film is found to be monocrystalline and exhibit a single orientation (0 0 0 6). The study of the waveguiding properties of the unprocessed thin film was performed using *m*-lines spectroscopy [12]. The obtained results demonstrated that our thin film is monomode both TE and TM with an ordinary and extraordinary refractive index of $n_o = 2.262$ and $n_e = 2.203$, respectively. The optical losses are measured using a charge-coupled device (CCD) technique, and are found to be around 1 dB cm^{-1}.

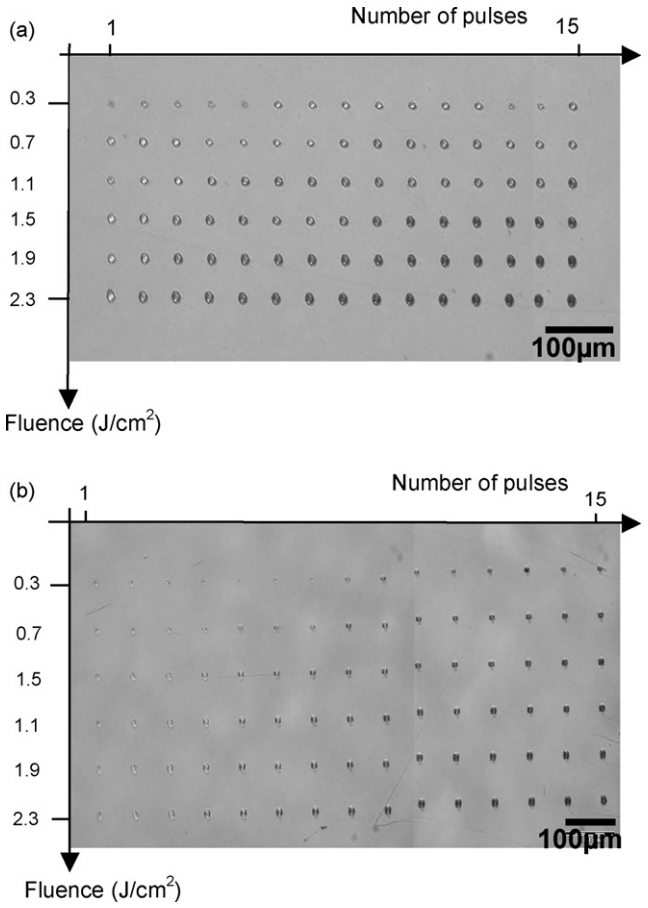

Fig. 1. Optical microscopy images of an array of holes patterned in (a) $LiNbO_3$ thin film and (b) crystal, with 1–15 pulses and various fluences.

Subsequently, we firstly investigate the effects of pulse repetition rate (from 10 to 100 Hz) on the ablation process. We find that the holes properties were not affected by this parameter: in other words, the time delay between two consecutive pulses does not influence the drilling process.

Secondly, we investigate the effect of beam fluence and of the number of pulses on the ablation process. A two-dimensional array of circular holes was written on $LiNbO_3$ thin film and crystal surfaces by varying the number of pulses (from 1 to 15 pulses) and the beam fluence (from 0.3 to 2.3 J/cm^2) as displayed on Fig. 1. The repetition rate used here is 50 Hz. As expected, the observations demonstrate that the holes depths as well as their shape strongly depend on the number of pulses and the fluence used. For $LiNbO_3$ thin film the ablation process takes place even with a single pulse at 0.3 J/cm^2. The surface contrast darkens with increasing number of pulses and fluence which means that the holes depth increases. The better shape (circular) for the holes is obtained with a high number of pulses and high fluences (Fig. 2a shows the influence of the fluence).

In the case of $LiNbO_3$ crystal, the evolution of the hole-shape is also a function of increasing number of pulses and fluence. In Fig. 2b, we can notice that the ablated holes have an oval shape whereas the mask pattern used for ablation was circular. The general trends already observed for the thin film samples are the same here.

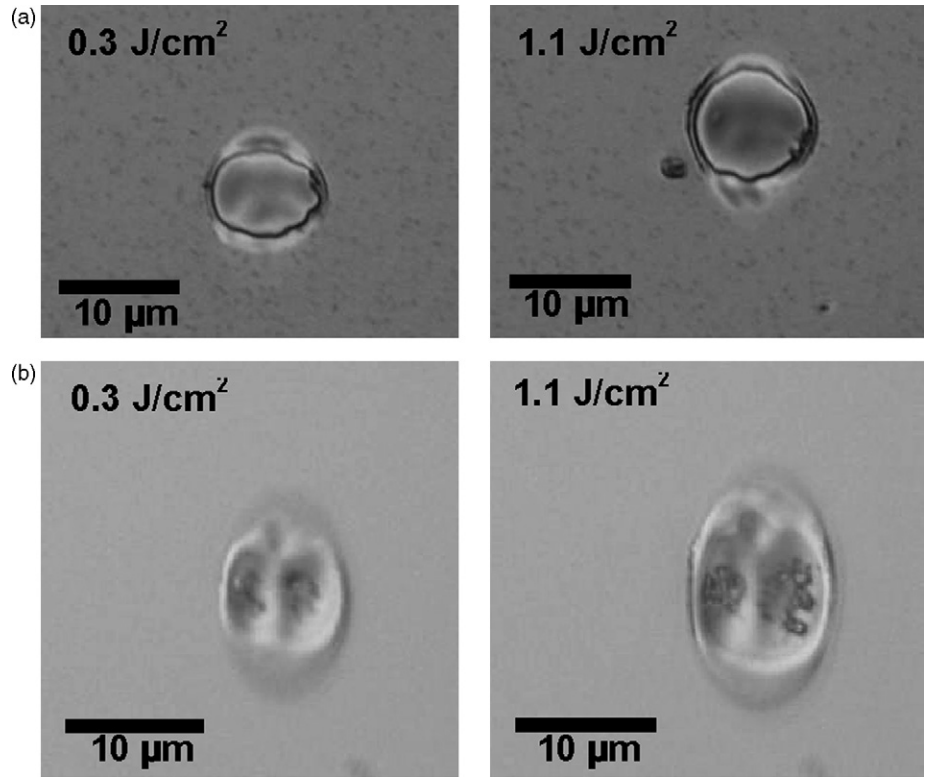

Fig. 2. Optical microscopy images of holes ablated in (a) $LiNbO_3$ film and (b) crystal, with 10 pulses and various beam fluences.

In both samples, a rim around the holes is observed: it is caused by resolidification of molten material (lithium niobate) on the edges. This is clear on Fig. 3 which displays AFM images of two holes ablated with five pulses on $LiNbO_3$ thin film. This ring of matter is more pronounced in the case of $LiNbO_3$ crystal. Its height also increases with increasing number of pulses and with fluence as well. The surface of the ablated $LiNbO_3$ thin film for large number of pulses and high fluence becomes rougher and blacken from 10 pulses and fluence of 1.5 J/cm^2. This can be explained by the amorphization of $LiNbO_3$ thin film due to thermal effect generally expected in the ablation in the nanosecond regime [13]. Another possible explanation is that the sapphire substrate might be reached, as it will be discussed later in the μ-Raman results.

In addition to ring formation at the edge of the holes, we observe the formation of a collateral damaged zone. This region known as heat affected zone is prevalent in the ablation with nanosecond lasers if a high fluence and/or large number of pulses are used due to the dominance of thermal diffusion [6,14,15]. From this study, we can emphasize that the ablation of $LiNbO_3$ crystal is mostly dominated by melting and resolidification process which prevents sufficient mass expulsion, and results in surface and volume damage.

In order to study the structural modifications caused by nanosecond laser ablation in and around the holes patterned on $LiNbO_3$ thin film and crystal, we performed micro-Raman experiments in back-scattering geometry [16,17]. In this configuration, the expected modes of y-cut $LiNbO_3$ crystal

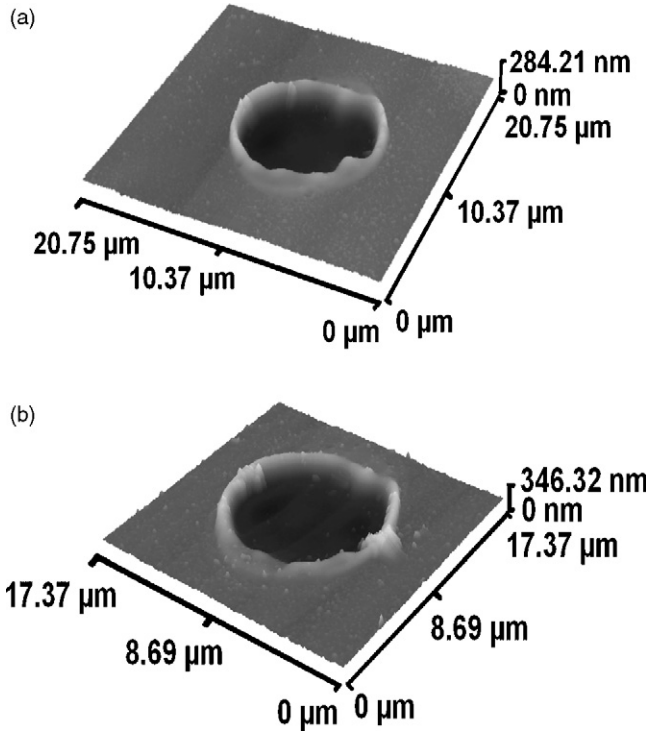

Fig. 3. Surface morphology of two holes ablated on $LiNbO_3$ thin film surface with five pulses and fluence of (a) 0.3 J/cm^2 and (b) 1.5 J/cm^2, recorded by AFM.

substrate are E(TO). For instance, Fig. 4a displays micro-Raman spectra in $y(xz)\bar{y}$ configuration, obtained for a hole ablated with 20 pulses and a fluence of 2.3 J/cm^2 on $LiNbO_3$ crystal surface. From a first inspection of these data, we can see that all the Raman peaks corresponding to the unprocessed area, which are typical for scattering geometry $y(xz)\bar{y}$, are also observed in the processed area. This demonstrates that no amorphization of $LiNbO_3$ crystal surface has been induced by nanosecond ablation under our experimental conditions. However, the relative intensity of all peaks is affected. This could be due to the introduction of defects, disorder and changes in the crystal structure of $LiNbO_3$. In case of ablation with 100 pulses the signal is totally flattened and most of the Raman modes present in the non-modified $LiNbO_3$ disappear, suggesting a laser-induced amorphization of the material.

For holes ablated in the same conditions in $LiNbO_3$ thin film the obtained Raman spectra are shown in Fig. 4b. Data of the unprocessed area correspond to Raman spectrum of bulk-$LiNbO_3$ measured in $z(xz)\bar{z}$ configuration. We observe the activation of A_1(LO) and E(TO) modes as predicted by the group theory. We also obtain typical Raman peaks of sapphire substrate as indicated on this figure. However, the recorded spectrum on the processed area shows a total vanishing of all peaks that were apparent in the Raman spectrum measured for the unprocessed area. The signal is totally flattened, only the peaks which correspond to sapphire substrate remain. This indicates that $LiNbO_3$ film had undergone melting and lost of its crystalline structure and the remaining very thin film became completely amorphous, or that the holes are deep enough to reach the sapphire substrate as previously mentioned.

Finally, one dimensional (1D) laser-induced structures forming ridge optical waveguides were performed. The laser writing is carried out by translating the sample ($LiNbO_3$ thin film or bulk crystal) perpendicular to the beam direction. For $LiNbO_3$ thin films we scan the surface at a speed of 71 μm/s and using a fluence of 1.1 J/cm^2. The grooves are made by a single

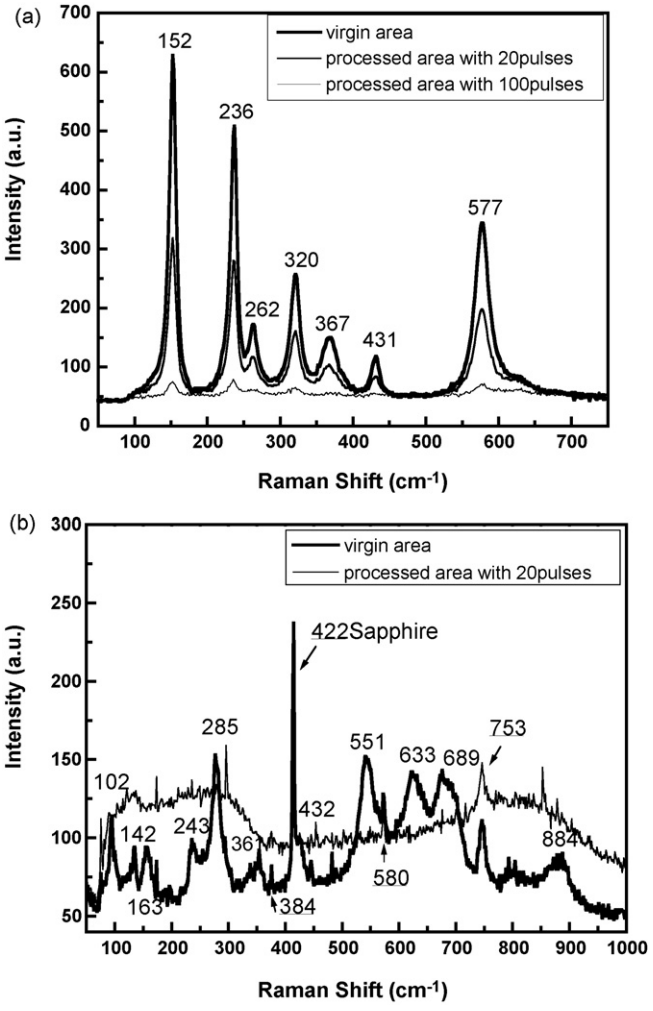

Fig. 4. Comparative plots for Raman shift for the unprocessed and processed areas (with 20–100 pulses and 2.3 J/cm^2), on $LiNbO_3$ (a) crystal and (b) thin film.

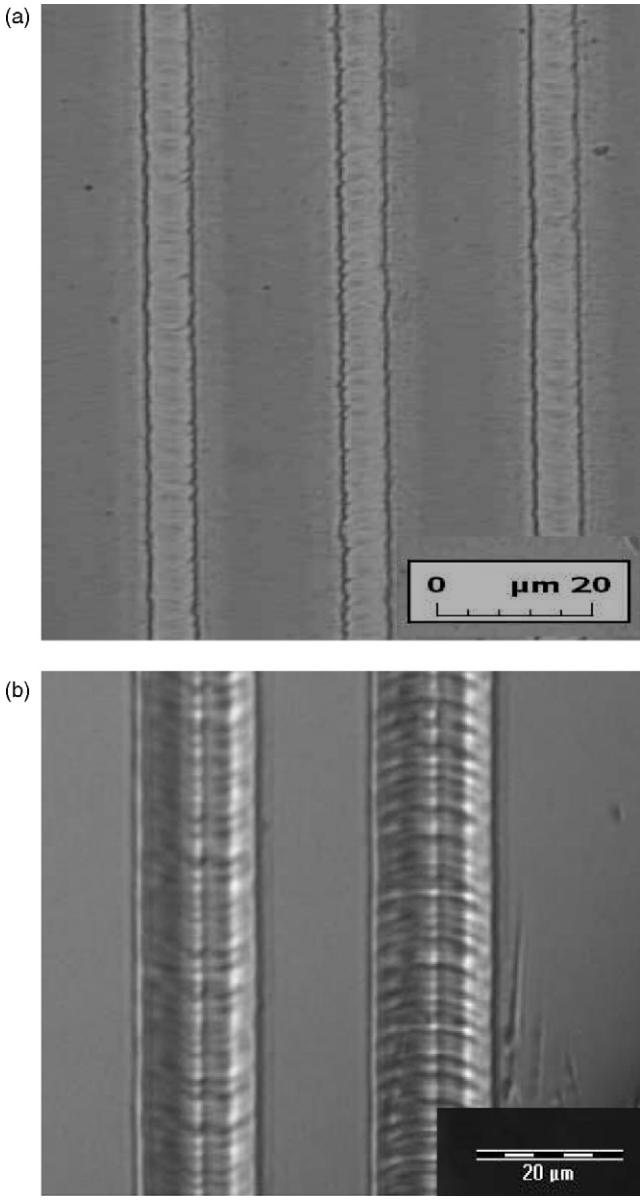

Fig. 5. Optical microscopy images of grooves patterned in (a) $LiNbO_3$ thin films and (b) crystal.

pass of the laser beam. Fig. 5a shows an optical microscopy image of the ridge defined by two adjacent grooves. The ridges are uniform without extensive damage of the patterned area. Periodic marking are also observed at the bottom of the grooves. Fig. 5b shows grooves achieved on $LiNbO_3$ crystal with a translation speed of 50 μm/s and a fluence of 2.3 J/cm^2. These preliminary results indicate that nano-second laser ablation could be used to obtain ridge optical waveguides. However, further studies are needed to totally characterize the obtained structures. For instance, end-fire coupling technique should be used to demonstrate waveguiding properties of those structures.

4. Conclusion

In this work, we report the investigation of KrF excimer laser-ablation of lithium niobate thin film and single crystal. Ablations are carried out in a mask projection set-up. We studied various conditions of ablation in order to assess the applicability of laser direct writing of photonic waveguides in $LiNbO_3$. Our results indicate that the optimal circular shape for the holes is obtained with a large number of pulses and high fluences. The pulse repetition rate does not affect the ablation process. Moreover, nanosecond laser ablation seems to induce an amorphization of $LiNbO_3$ thin film due to thermal effects. We also emphasize that the ablation of $LiNbO_3$ crystal is dominated by melting and resolidification process which prevents sufficient mass expulsion, and results in surface and volume damage rather than a purely etched structure. These results have been confirmed by using μ-Raman spectroscopy. Finally, our work shows that nanosecond laser ablation could be used in order to create ridge optical waveguide.

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