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UV laser-induced ordered surface nanostructures in congruent lithium niobate single crystals

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Abstract

Ultra violet illumination of the –z face of lithium niobate single crystals, under specific conditions, results in an organized arrangement of submicron etch resistant features that reflect the illuminating intensity distribution. Consequently, spatially resolved illumination can produce periodic structures with submicron periodicity. Furthermore, a size self-adjustment of the submicron etch resistant features was observed which is related to characteristic lengths (e.g. grating period) of the overall structure. The effect occurs for a narrow range of illuminating intensities and is attributed to a photo-induced electrostatic charge distribution, which modifies the electrochemical interaction of the acid with the surface. The size and periodicity of the structures, which can be achieved with this method, are suitable for the fabrication of 2D photonic crystal structures in this electro-optically tunable material.

Keywords: Submicron; Lithium niobate; Laser-induced; Nanostructure

1. Introduction

Lithium niobate is among the most important optical materials as it combines a variety of very useful properties. It exhibits significant optical non-linearity is electro-optic, photorefractive, pyroelectric, piezoelectric and has extended optical transparency over the infrared and visible spectrum [1], and therefore lithium niobate crystals are extensively used today in the photonics industry for the fabrication of electro-optic modulators and switches [2], acousto-optic devices [3] and non-linear frequency conversion via quasi-phase-matching in periodically poled material [4]. Furthermore, these properties also show the potential and further application of this material for the fabrication of tunable photonic devices.

However, a very important requirement for the realisation of such devices is the development of micro- and nano-fabrication methods. A very useful property commonly used in micro-fabrication is differential etching and lithium niobate exhibits significant differential etching behaviour in HF and $HF:HNO_3$ acid mixtures between ferroelectric domains of opposite orientation. The etch rate of

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the –z face of the crystal depends on the acid mixture and temperature [5], while the +z face remains unaffected by the etch process in our experience and this property has been used already in conjunction with ferroelectric domain engineering for the fabrication of a range of 2D and 3D microstructures [6,7].

Another method for the modification of the etching characteristics of the –z face of lithium niobate single crystals uses laser irradiation of the –z face of the crystal prior to acid etching. The interaction of the laser radiation with the crystal surface (under appropriate irradiation conditions) can reduce or even completely stop the etch process locally. This effect has been observed in Fe doped crystals using continuous visible laser irradiation during the etching process [8], and in undoped crystals using ultra violet (UV) laser radiation prior to acid etching [9,10].

In this paper, we show that spatially resolved pulsed UV laser irradiation of the –z face can act to organize the position and size of etch resistant features enabling the fabrication of submicron periodic structures. The ability for generation of such structures shows great potential for the fabrication of photonic structures with tuning capabilities, as the material is both nonlinear and electro-optic.

2. Experimental procedure

The experimental procedure followed in the present work involved exposure of the –z face of z-cut lithium niobate samples to ultra violet laser radiation and subsequent chemical etching in HF acid at room temperature. The time interval between exposure and etching varied from minutes to a few hours without any significant change in the results. The laser source which was used for the exposures was a KrF excimer laser emitting pulses of ~20 ns duration at a wavelength of 248 nm. The samples used were diced from commercial 500 μm thick z-cut congruent lithium niobate wafers, acquired from Crystal technology, USA, with both z faces optically polished.

Spatially resolved illumination of the surface was achieved by using either amplitude masks or phase masks depending on the desired resolution of the illuminating intensity pattern. Transmission electron microscopy (TEM) copper grids were used as amplitude masks. These grids were 11 μm thick, having circular shape of ~3 mm diameter and consisting of a two-dimensional metallic mesh with openings of either square or hexagonal shape. The widths of the grid openings were: 90, 50 and 35 μm.

Initial experiments suggested that the etch frustration results were sensitive to small variations of the illuminating intensity. For this reason the grids were attached to the crystal surface using narrow strips of adhesive tape instead of being pressed against the surface using a UV transparent optical flat, in order to avoid multiple reflections, which may well have confused the results. Consequently, the contact between the grid and the crystal surface was imperfect and near-field (Fresnel) diffraction effects were expected to play a significant role in the illumination of the area near the edges of the grid openings.

However, the overall feature size resulting from use of the contact amplitude mask method of illumination will of course be limited to the opening size of the grids and in order to increase the spatial resolution, phase mask illumination was used. The phase mask used in these experiments had a grating period, on the mask, of $\Lambda_m = 720$ nm, and hence was able to produce a near-field periodic intensity pattern with a 360 nm period. The phase mask was optimized for a wavelength of 246 nm ($\Lambda_m/2$). In order to ensure reproducible illumination conditions the spacing between the phase mask and the surface of the sample was kept constant at 125 μm using pieces of standard telecom fibre (with the polymer jacket removed) as spacers. The phase mask was then pressed against the sample in a specially designed holder. The beam profile of the excimer laser was rectangular with dimensions of ~30 mm × 15 mm. However, only the central 20 mm × 10 mm portion of the beam was used for spatially resolved exposures in order to obtain improved uniformity of illumination. The quality of excimer laser beams is inherently rather poor; therefore, there are always residual irregularities in the intensity profile even in the central portion of the beam. A number of exposures were performed using a portion of the beam with a significant intensity gradient. In this way it was possible to obtain a single spot mapping of the etch frustration behaviour as a function of the local intensity variation.

It is important to note that all the UV exposures were performed using laser fluences below the ablation threshold. The surface of the crystal was examined, after UV exposure, with an optical