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Surface Domain Inversion in Ferroelectric Lithium Niobate

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Periodic inversion is reported for ferroelectric domains near the surface of z-cut lithium niobate crystals by a modified electric field poling technique. The depth of the inverted domain region extends to values of order a few μm (5–10 μm) below the surface of the crystal, thereby removing the high aspect ratio instability problems associated with bulk poling, and therefore allowing the fabrication of fine period ferroelectric domain structures. Using this method periods as short as 1 μm have been achieved. Such periodic domain distributions have been fabricated in Ti-indiffused and proton exchanged lithium niobate waveguides and our first quasi-phase-matching results are shown.

Keywords: Lithium niobate waveguides; ferroelectric domain engineering; frequency conversion

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As the area of integrated optics continues to expand there is an increasing demand for compact and efficient integrated nonlinear optical devices. Lithium niobate and lithium niobate waveguides are already offering a significant contribution in this direction. Due to the important and useful properties of congruent lithium niobate as well as the ability to reproducibly grow high quality single crystals this material has become an industry standard for the production of high speed optical modulators.

In order to make use of the highest nonlinear coefficient, d_{33}, which is not accessible through the technique of birefringent phase matching, periodic inversion of the ferroelectric domains has been proposed [1] and has been successfully implemented for blue light generation [2, 3].

The fabrication of high quality periodically inverted domain distributions in bulk crystals however, is restricted to longer periods (> few μm) because of high aspect ratio instabilities. For commercially available lithium niobate

wafers with thickness of typically 300 μm or 500 μm the successful fabrication of large scale uniform inverted domain gratings is typically restricted to periods of ~6–10 μm. In order to fabricate finer period domain distributions further thinning of the substrate must be applied [4, 5] but this additional fabrication step is best avoided if possible.

There is a class of applications however, that does not require domain inversion to extend throughout the thickness of the crystal. For the case of guided wave devices the major requirement, apart from the high and uniformity of the domain distributions, is good overlap of the domain inverted volume with the guided optical fields which therefore requires domain depths typically of order a few μm only. Such a method for the fabrication of periodically inverted fine period ferroelectric domain distributions, which extend only a few μm below the surface of lithium niobate crystals, is presented here.

The method is based on conventional electric field (E-field) poling with an intentional "overpoling" step. As in conventional E-field poling the crystal is covered with photolithographically patterned photoresist on one of the two z faces (−z, or +z). Conductive gel electrodes are applied on both faces and finally a high voltage is applied. The patterned photoresist provides enough electric field contrast so that only the areas where the electric field exceeds the value of the coercive field (~22 kV/mm), the developed parts of the photoresist, will invert their polarity. The whole process is computer controlled in such a fashion that the applied field stops when the exact predetermined amount of charge has passed through the crystal. An additional important feature of our poling equipment is that the voltage is controlled in order to keep the current constant.

The amount of charge which is required to domain invert an area A is: $Q = 2 \times A \times P_s$, where Q is the calculated charge, and P_s is the spontaneous polarization of lithium niobate (0.72 μC/mm^2). An additional external empirical factor (EF) is also usually taken into account in conventional poling, to correct for variations in supplier dependent material stoichiometry, precise values of thickness across the sample and specific electrical characteristics of the power supply itself. An *EF* value exceeding unity is often used to achieve the desired high quality periodic domain patterning, resulting in a modified calculated Q value of $2 \times A \times P_s \times EF$.

Furthermore, the *EF* factor determines the state of the sample after poling. Specifically for values lower than 1 the sample becomes *underpoled*, where only a portion of the patterned area is successfully domain inverted, while for values far greater than 1 it becomes *overpoled* where the sample appears uniformly poled regardless of any initial photoresist patterning.

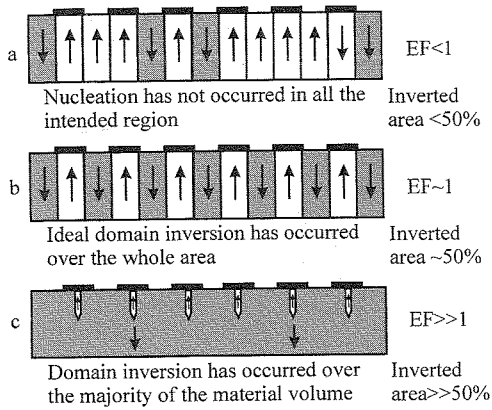

Figure 1. Schematic of the resultant inverted domain structures as a function of the empirical factor *EF*.

The evolution of domain inversion as a function of *EF* is outlined in Fig. 1. As shown in Fig. 1a for $EF < 1$ only part of the intended area has been inverted. For values of $EF \sim 1$ good quality domain inversion can extend all the way through the crystal, while for $EF > 2$ essentially complete domain inversion has occurred apart from a small region directly under the photoresist which maintains its original polarization state. This is possibly due to the presence of compensating charges which are trapped between the insulating photoresist and lithium niobate surface and which will create a local electric field opposing the externally applied one.

Poled samples are typically examined by brief etching in HF acid which utilizes the differential etching between + and −z faces and provides a visual indication of the poled structure. The morphology of the samples after poling depends on the poling regime as specified by the value of *EF*. In the underpoling regime patches of poled areas are observed unevenly distributed across the surface, due to domain nucleation beginning from randomly distributed surface defects.

In the *overpoling* regime however (for $EF \geq 2$), although the sample appears to be uniformly poled, more careful examination following etching shows a surface relief pattern corresponding to the initial photoresist period that shows that a portion of the areas under the photoresist that have maintained their original polarization state. Figure 2 shows an SEM picture of an *overpoled* sample patterned with a period of ~2.5 μm where the inverted ferroelectric domains have been made visible after brief etching in HF acid.

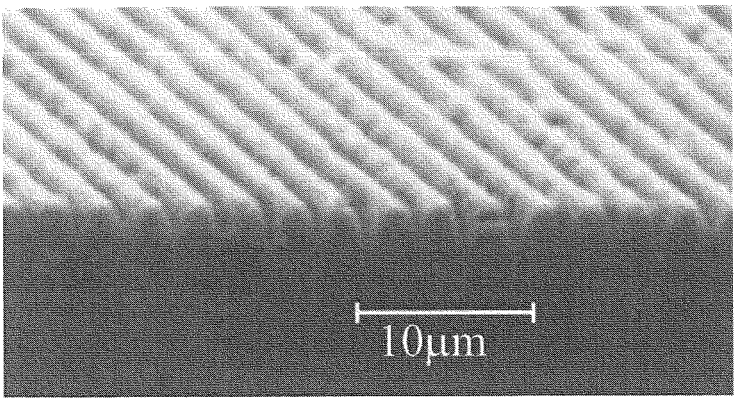

Figure 2. Scanning electron microscope picture of surface domains revealed by HF/HNO_3 acid etching. The period of the domain inverted structure is 2.5 μm.

Side etching of the y-face showed that the surface inverted domains extend to depths of a few microns and that their depth is a function of the period. Measurements of the depth which were performed by side etching of overpoled samples with different periods showed that the depth of the surface domains decreases as the period decreases. The results are summarized in Fig. 3 where the average domain depth, as determined by Scanning electron Microscopy measurements of the side etched overpoled samples, is plotted against the inverted domain period for samples that have been poled with an $EF = 8$. The plot of Fig. 3 shows that there is an approximately

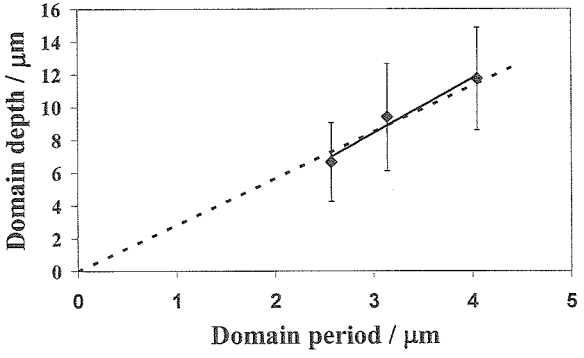

Figure 3. Experimental measurements of domain depth versus domain period, as determined by optical microscopy, using an EF value of 8.

Figure 4. Scanning electron microscope picture of 1 μm periodic surface domains written using a phase mask.

linear decrease of the average domain depth with decreasing domain period and also that there is a constraint in the overlap of the domain inverted region with waveguide modes for submicron periods. The dash line fitting line shown in Fig. 3 takes into account the point (0, 0) as a valid data point while the solid line doesn't. However there is close agreement between the two gradients which confirm the linearity statement.

Domain periods down to 1 μm have so far been achieved using this method as shown in the scanning electron microscopy picture of a briefly etched surface poled sample which is presented in Fig. 4. In order to achieve such fine periodic patterning the photoresist was exposed through a phase mask.

In order to prove the utility of the method for optical waveguides applications both proton exchanged and Ti indiffused lithium niobate channel waveguide samples were fabricated and overpoled. HF etching of the poled waveguide samples showed that good quality uniform inverted domain distributions can be achieved as is shown in the optical microscope pictures of Fig 5. Figure 5a shows a poled proton exchanged waveguide while Fig. 5b shows a poled Ti:indiffused waveguide.

A continuous wave Ti:sapphire laser was used in order to perform quasi phase matched second harmonic generation experiments in poled waveguides for blue light generation. Preliminary second harmonic generation

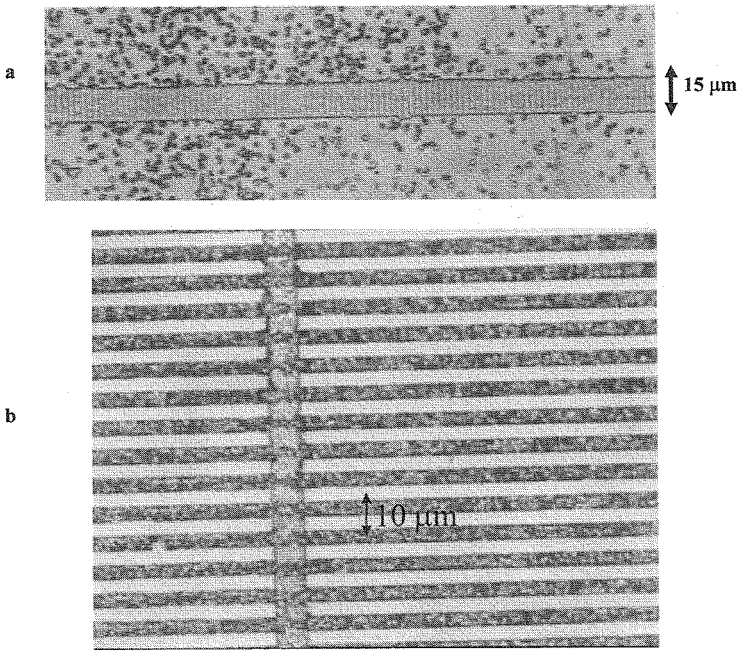

Figure 5. Sections of surface periodically poled lithium niobate waveguides with revealed inverted periodic domains after etching: (a) proton exchanged (b) Ti indiffussed.

results have been obtained from a periodically poled soft proton exchanged lithium niobate channel waveguide and a typical temperature tuning curve of the quasi phase matched (QPM) interaction is presented in Fig. 6. The phase matched wavelength is 855.6 nm but as we used a multimode waveguide structure at the wavelengths of the fundamental optical wave the net conversion efficiency was lower than optimal.

However, single mode waveguide structures have been fabricated using Ti indiffusion in lithium niobate and the QPM second harmonic generation results showed increased efficiency. For a single mode (at the fundamental wavelength) optical waveguide with surface periodically poling we observed 3.46 mW of blue light (after correcting for the filter transmission) at 412.6 nm for a pump intensity of 70 mW at a temperature of 205°C. The corresponding value of the internal conversion efficiency is therefore, 126 %/W cm^2. To further improve the performance of such nonlinear devices several aspects of the poling as well as the waveguide fabrication must be considered. Firstly

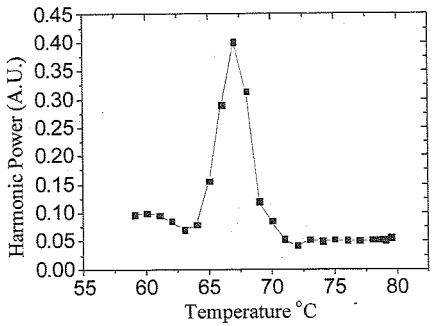

Figure 6. Phase matching curve from a surface periodically poled soft proton exchanged lithium niobate waveguide obtained using a continuous wave tunable Ti:sapphire laser. The phase matching wavelength is 855.6 nm.

the mark to space ratio of the inverted domains, which in our case is not the ideal 50/50 ratio, will need to be corrected and secondly, fabrication of tolerant optical waveguide structures has also to be addressed in order to compensate for fabrication errors which results in reduced conversion efficiency.

In conclusion, a method for the fabrication of surface periodically inverted ferroelectric domain structures in lithium niobate single crystals based on E-field poling has been presented. The inverted domain depth is of order a few microns depending on the period of the distribution, and is sufficient to support QPM interactions in lithium niobate waveguides. Finally efficient first order QPM second harmonic generation in both soft proton exchanged and Ti indiffussed waveguides has been demonstrated.

REFERENCES

[1] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, *Phys. Rev.* **127**, 1918 (1962).
[2] G. Batchko, V. Y. Shur, M. M. Fejer, and R. L. Byer, *Appl. Phys. Lett.* **75**, 1673 (1999).
[3] D. J. L. Birkin, E. U. Rafailov, G. S. Sokolovskii, W. Sibbett, G. W. Ross, P. G. R. Smith, and D. C. Hanna, *Appl. Phys. Lett.* **78**, 3172 (2001).
[4] K. Kintaka, M. Fujimura, T. Suhara, and H. Nishihara, *Electr. Lett.* **32**, 2237 (1996).
[5] M. Yamada and M. Saitoh, *J. Appl. Phys.* **84**, 2199 (1998).