Surface domain engineering in congruent lithium niobate single crystals: a route to sub-micron periodic poling.

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Abstract

We describe a technique for surface domain engineering in congruent lithium niobate single crystals. The method is based on conventional electric field poling, but involves an intentional overpoling step that inverts all the material apart from a thin surface region directly below the patterned photoresist. The surface poled structures show good domain uniformity, and the technique has so far been applied to produce domain periods as small as ~ 1μm. The technique is fully compatibile with nonlinear optical integrated devices based on waveguide structures.

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Domain engineering in ferroelectric crystals such as $LiNbO_3$ and $LiTaO_3$ is an increasingly important and ever more versatile technique for applications in areas as diverse as harmonic generation and parametric processes [1], electro-optic Bragg gratings [2], and piezoelectric micro-actuated devices [3]. Over the past decade or so, highly efficient quasi-phase-matched nonlinear interactions have been achieved via precise periodic domain inversion in z-cut crystal samples, using periods for example of order a few μm for near infrared to blue or near UV harmonic generation [4,5]. Research on periodically poled lithium niobate, PPLN, (and to a lesser extent tantalate, PPLT,) continues to generate considerable interest from the fundamental viewpoint of materials research through to the fabrication of practical nonlinear optical and electro-optical devices. PPLN with periods for standard conversion wavelengths is now commercially available from several sources.

Fabrication of periodically poled materials with arbitrarily small values of period, particularly at sub-micron scales, remains an elusive goal however. The high coercive field, E_c, required for domain inversion in congruent $LiNbO_3$ (E_c ~220kV cm^{-1}), together with inherent non-uniformities and defects that are always present in commercially available materials, restricts the applicability of the standard electric field poling technique to periods of order >4-5μm in samples of thicknesses ~500μm. It is not an easy task to routinely fabricate high quality PPLN and in many cases the crystal must be polished down to thicknesses of order 100μm -150 μm to achieve finer periods than this [6,7].

Two approaches to overcome this apparent limit in domain period have recently met with some success however. The first technique, referred to as controlled spontaneous backswitching, has been applied to bulk samples of typical thickness 500 μm, to generate periods of 4μm [8], and more recently 2.6μm [9]. The second technique, applied to MgO:$LiNbO_3$ which has the benefit of improved resistance to photorefractive damage, utilized multiple short current pulses, generating a period of 2.2μm and depth of 1.5μm, which when used in conjunction with a waveguide geometry, has produced a high conversion efficiency [10].

This last result is significant in that, for waveguide geometries at least, it is not necessary to achieve domain inversion to depths exceeding the guide depth itself. Many of the earlier reports on domain inversion applied to typical commercial material supplied as either 300μm or 500μm thick wafers, and it is clearly harder to maintain high aspect ratio, short period, high quality domain patterning over these large and (for waveguide geometries) unnecessarily large depths. In this letter, we discuss a new technique for achieving superficial, or surface, domain inversion that has been used to achieve periods of 1μm, and that can be used we believe for achieving the periods of ~0.3μm required for waveguide implementation of backward wave parametric generation and tunable Bragg grating structures.

The technique relies on *overpoling* the sample which achieves the apparently undesirable effect of domain spreading and merging beneath the lithographically patterned

photoresist layer. For normal electric field poling, the established practice is to first calculate the charge, Q, corresponding to the patterned area intended for domain inversion. The formula used for this calculation is $Q=2\times A\times P_s$, where Q is the calculated charge, A is the area corresponding to the developed part of the photolithographic pattern (the area where the conductive liquid or gel is in contact with the crystal surface) and P_s is the spontaneous polarization of lithium niobate (0.72 $\mu C/mm^2$). An additional external empirical factor (EF) is also usually taken into account, to correct for variations in supplier dependent material stoichiometry, precise values of thickness across the sample and specific electrical characteristics of the poling supply itself. An EF value exceeding unity is often used to achieve the desired high quality periodic domain patterning, resulting in a calculated Q value of $2\times A \times P_s\times EF$.

This empirical factor thereby controls domain spreading within the crystal volume: values of EF<1 lead to *underpoling*, whereby domains are inverted preferentially in areas where nucleation is easier, for example at the edges of the photoresist patterns or areas of increased surface roughness. If EF is too large however, then the inverted domains, once nucleated, spread laterally, extending their volume more rapidly than required for an ideal 50/50 mark-to-space ratio grating. This case is referred to as *overpoling*, and is illustrated schematically in figure 1, where for large values of EF, small regions of material beneath the photoresist can remain in their original poled state. If overpoled, using values of EF exceeding the theoretical value of ~2, then the sample appears almost uniformly poled when observed between crossed polarizers. Once etched with HF/HNO_3 acids however, careful investigation reveals that some non-inverted domain regions survive beneath the

photoresist patterned surface, and that these can extend a few microns into the –z crystal face.

Using this technique, we have performed an initial parametric study of surface poling versus value of EF and imposed photoresist period. It should be noted that this technique will not work with other electrode materials such as directly deposited metals, as charge accumulation is thereby prohibited. We have used both conventional photolithographic patterning for periods between 2.5μm and 4.0μm and also laser exposure through a phase mask. For the latter technique, final domain widths of order 0.5 μm and periods of 1μm have been obtained. We have examined these surface domains for the former case, and find that they extend to depths of between 6μm and 11μm, entirely compatible with waveguide depths and good overlap of guided modes.

For the larger range of periods studied, the $LiNbO_3$ -z face was spin-coated with 1.2 μm thick photoresist and UV-exposed through a periodic amplitude mask. After photoresist development, gel electrodes were applied to both the unpatterned +z face and the patterned -z surface. The samples were poled using a computer-controlled supply that dynamically varied the applied field in order to maintain a constant current, and the poling process terminated when a pre-defined charge Q (= $2\times A \times P_s \times EF$) had passed through the crystal. A typical single pulse poling curve is shown in Figure 2, and illustrates the difference between this technique and that reported in [8,9]. No backswitching occurs in our overpoling process, and we feel this represents a

fundamentally simpler technique for achieving controlled small period surface domain inversion.

A variety of surface poling results can be obtained that depend on the value of EF used. Figure 3 shows a typical scanning electron microscope (SEM) picture of a surface poled sample, obtained with an EF value of 8. It is clearly seen following the HF/HNO_3 etching that the domains only exist in the near surface region (shown here to a depth of ~3 μm). Other EF values can and have been used, but to date we have not performed a full parametric study of depth or uniformity as a function of EF value. The surface domain depth however is clearly an inverse function of EF value.

In figure 4 we show the results of measured domain depth as a function of the period of the imposed photolithographic pattern, for an EF value of 8. Although the variation of measured domain depth (taken for between 30 and 100 periods) is rather large, two clear points emerge. Firstly, there is a minimum in the domain depth achieved, an obvious requirement for intended waveguide applications. Secondly, the mean depth is seen to scale approximately linearly with period. For applications that require sub-micron periodicity, this is again a useful observation as overlap between the guided modes and domain inverted regions is a pre-requisite for efficient nonlinear interactions. Figure 4 shows two fits: one (dashed line) includes the point (0,0) as a further implicit data point. The close agreement between these two gradients further confirms the approximate linearity stated above.

Finally, we show in figure 5 details of a ~1μm periodicity surface grating, fabricated using exposure of the photoresist via a phase mask. Following acid etching, sub-μm features are revealed that are of order 1μm in depth. We believe that such interferometric exposure (via phase mask or two beam interferometry) holds much promise, as domain patterning down to periods of order 0.3 μm required for backward wave interactions at a wavelength of 1.5 μm should be readily achievable using exposure with near UV laser irradiation.

In summary therefore, we have presented a novel single step approach for achieving surface domain inversion to depths that are consistent with single mode waveguides in $LiNbO_3$. The overpoling technique is simple to implement, and appears to work down to periodicities of at least 1μm. Further work is in progress to examine the optimum choice for the EF value used, and to fabricate first order gratings in waveguide materials, with the required periodicities of ~2μm.

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References.

1. M. Yamada, N. Nada, M. Saitoh, K.Watanabe, Appl. Phys. Letts., **62**, 435, (1993)

2. M. Yamada, M. Saitoh, H. Ooki, Appl. Phys. Letts., **69**, 3659 (1996)

3. C.L. Sones, S.Mailis, V.Apostolopoulos, I.E.Barry, C.B.E.Gawith, P.G.R.Smith, R.W.Eason, Journal of Micromechanics and Microengineering **12,** 53, (2002)

4. R. G. Batchko, V. Y. Shur, M. M. Fejer, R. L. Byer, Appl. Phys. Lett. **75**, 1673 (1999)

5. D. J. L. Birkin, E. U. Rafailov, G. S. Sokolovskii, W. Sibbett, G. W. Ross, P. G. R. Smith, D. C. Hanna, Appl. Phys. Lett. **78**, 3172 (2001)

6. K. Kintaka, M. Fujimura, T. Suhara and H. Nishihara, Electr. Lett. **32**, 2237 (1996)

7. M. Yamada, M. Saitoh, J. Appl. Phys. **84**, 2199 (1998)

8. R.G.Batchko, V.Y.Shur, M.M.Fejer, R.L.Byer, Appl. Phys. Letts., **75**, 1673, (1999).

9. V.Y.Shur, E.L.Rumyantsev, E.V.Nikolaeva, E.I.Shishkin, R.G.Batchko, G.D.Miller, M.M.Fejer, R.L.Byer, Ferroelectrics, **236**, 126, (2000)

10. T.Sugita, K. Mizuuchi, Y.Kitaoka, K.Yamamoto, Jap.J.Appl. Phys., Part 1. **40,** 1751, (2001)

Figure Captions.

Figure 1. Schematic of overpoling process for fabrication of surface domain structures. EF is an empirical factor.

Figure 2. Single pulse poling signatures for current and voltage. Note that no backswitching is observed in this process.

Figure 3. Scanning electron microscope picture of surface domains revealed by HF/HNO_3 acid etching.

Figure 4. Experimental measurements of domain depth versus domain period, as determined by optical microscopy, using an EF value of 8.

Figure 5. Scanning electron microscope picture of 1μm periodic surface domains written using a phase mask.

FIGURE 1 A. C. Busacca *et al*

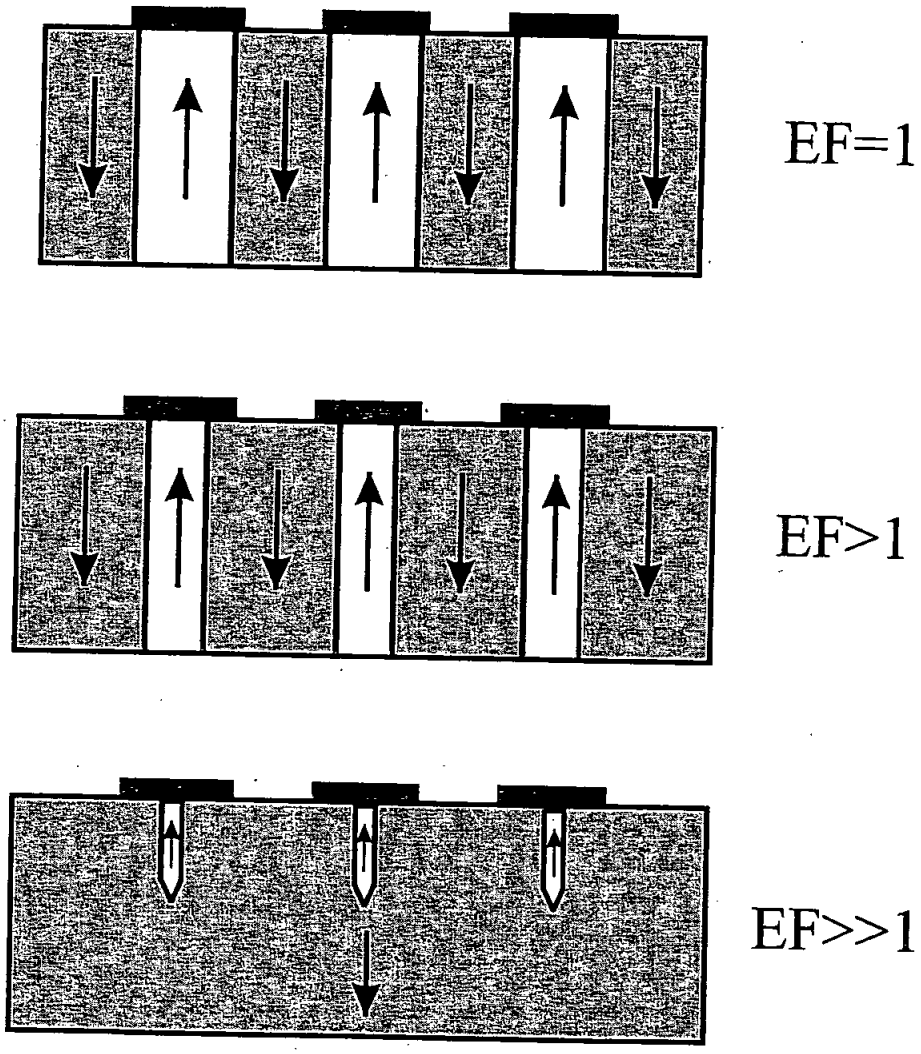

FIGURE 2 A. C. Busacca *et al*

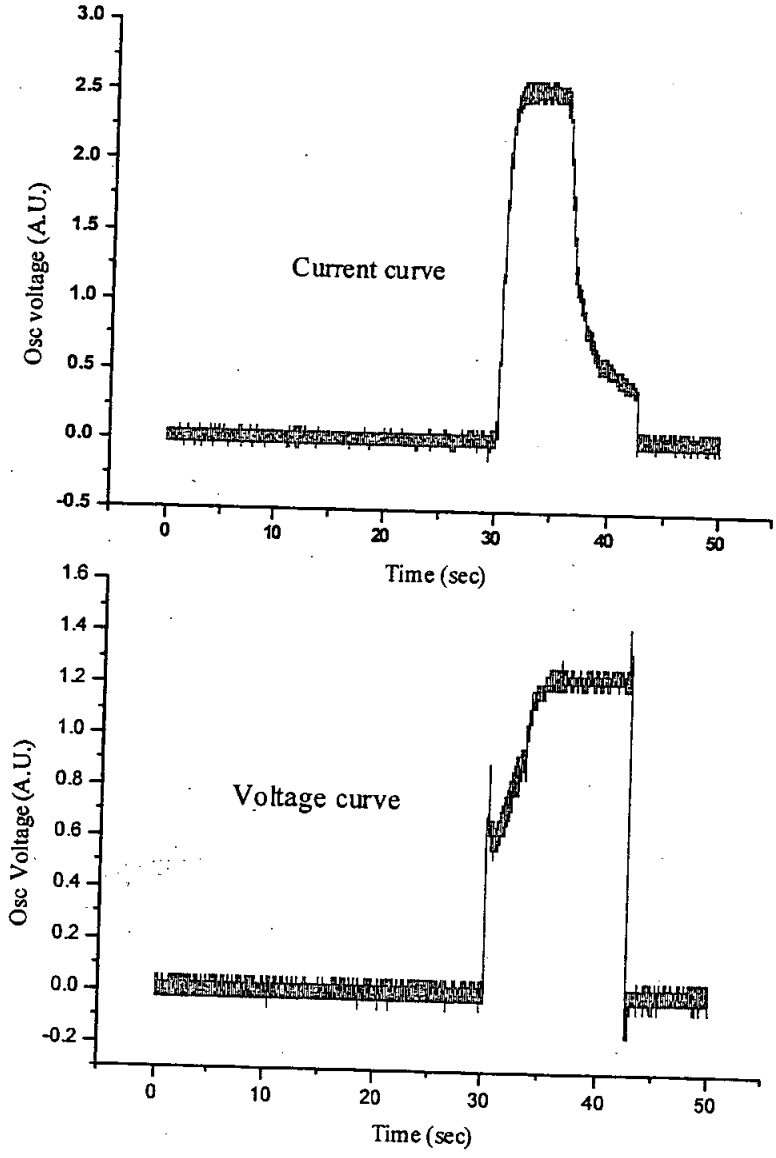

FIGURE 3 A. C. Busacca *et al*

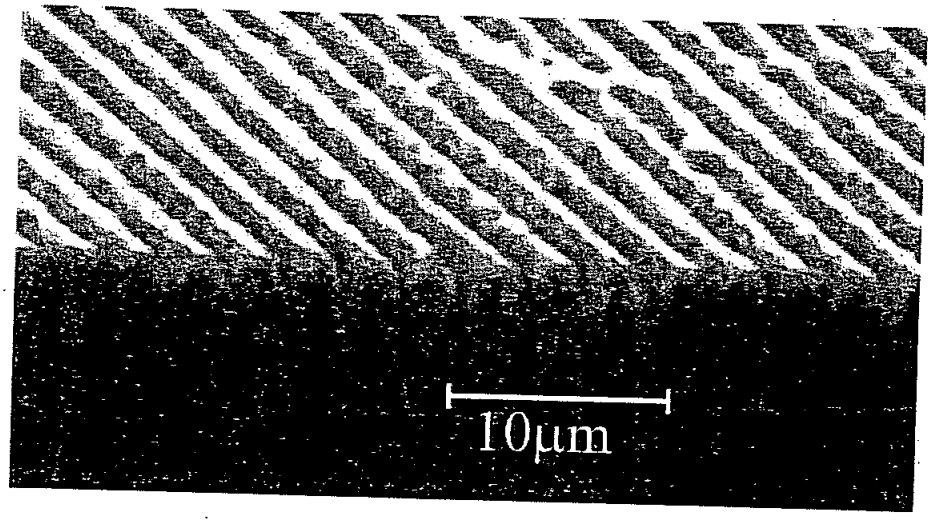

FIGURE 4 A. C. Busacca *et al*

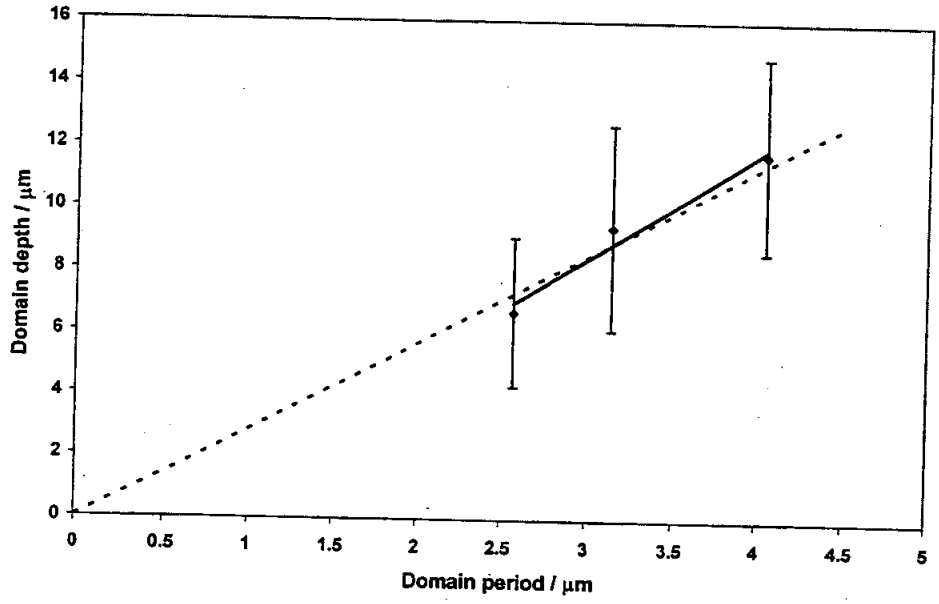

FIGURE 5 A. C. Busacca *et al*