Surface domain engineering in congruent lithium niobate single crystals: A route to submicron periodic poling

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() 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 photore sist. The surface poled structures show good domain uniformity, and the technique has so far been applied to produce domain periods as small as ~1 μm. The technique is fully compatible with nonlinear optical integrated devices based on waveguide structures.

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,$_1$ electrooptic Bragg gratings,$^2$ and piezoelectric microactuated 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 the order of 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 lithium tantalate), 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 submicron scales, remains an elusive goal however. The high coercive field, $E_c$, required for domain inversion in congruent LiNbO$_3$ ($E_c \sim 220$ kV 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 on the order of $>4–5$ μm in samples of thicknesses $\sim500$ μm. It is not an easy task to routinely fabricate high-quality PPLN and, in many cases, the crystal must be polished down to thicknesses on the order of 100–150 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 with a typical thickness of 500 μm, to generate periods of 4 μm,$^8$ and more recently 2.6 μm.$^9$ 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 2.2 μm and depth of 1.5 μm, 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 300 μm or 500 μm thick wafers. 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 method for achieving superficial, or surface, domain inversion that has been used to achieve periods of 1 μm, and that can be used, we believe, for achieving the periods of $\sim0.3 \mu$m required for waveguide implementation of backward wave parametric generation and tunable Bragg grating structures.

The technique for surface domain inversion is based on conventional electric-field ($E$-field) poling at room temperature. The procedure is as follows: One of the $z$ faces of the crystal is covered with a photolithographically patterned photoresist layer with a thickness on the order of 1 μm in order to achieve the appropriate $E$-field contrast which is necessary for a spatially selective domain inversion. Both $z$ faces are then covered with conductive gel electrodes, and a single high-voltage (HV) pulse is applied across the sample. The value of the HV varies with the thickness of the sample but the applied electric field must be on the order of 22 kV mm$^{-1}$.

For normal $E$-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 patterned 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 μC/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 peri-
Fig. 1. Schematic of the poling process as a function of the empirical factor $EF$. (a) underpoling ($EF<1$), (b) normal poling ($EF\sim1$), and (c) overpoling ($EF\gg1$) used for fabrication of surface domain structures.

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

Osc voltage ($\Delta V$)

0.5 1.0 1.5 2.0

Time (sec)

Current

Voltage

2 μm

0.5

5.0

4.5

4.0

3.5

3.0

2.5

2.0

1.5

1.0

0.5

0.0

Underpoling ($EF<1$)

Inverted area <50%

Ideal domain inversion has occurred over the whole area

Inverted area ~50%

Domain inversion has occurred over the majority of the material volume

Inverted area >50%

 overdic domain patterning, resulting in a calculated $Q$ value of $2A \times P_x \times EF$. The E-field is applied until the appropriate amount of charge according to the expression for $Q$ is detected. It is clear therefore, that the duration of the E-field application is a function of both the area to be poled and the $EF$ value.

This $EF$ 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 photosis patterns or areas of increased surface roughness. If $EF\sim1$, normal poling occurs which can be of good quality with 50/50 mark-to-space ratio and large scale uniformity for long period poled structures. Finally, 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. The three regimes, according to the domain spreading, are illustrated schematically in Fig. 1. Concentrating our attention on Fig. 1(c) which describes the state of the sample after poling using large values of $EF$, the schematic shows that small regions of material beneath the photosis can remain in their original poled state. If overpoling, using values of $EF$ exceeding the theoretical value of $\sim2$, 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 noninverted domain regions survive beneath the photosis patterned surface, and that these can extend a few microns into the $\bar{z}$ crystal face. The technique which is described here relies on overpoling the sample which achieves the apparently undesirable effect of domain spreading and merging beneath the lithographically patterned photosis layer. It is also able to create large scale uniform fine period surface inverted domain structures.

Using this technique, we have performed an initial parametric study of surface poling versus the value of $EF$ and imposed photosis 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 photolitho-
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. First, there is a minimum in the domain depth achieved, an obvious requirement for intended waveguide applications. Second, the mean depth is seen to scale approximately linearly with the period. For applications that require submicron periodicity, this is, again, a useful observation as the overlap between the guided modes and domain inverted regions is a prerequisite 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 just stated.

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

In summary, therefore, we have presented a single-step approach for achieving surface domain inversion to depths that are consistent with single-mode waveguides in \( \text{LiNbO}_3 \). The overpoling technique is simple to implement, and appears to work down to periodicities of at least 1 \( \mu \)m. 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 \( \sim 2 \) \( \mu \)m.

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1. M. Yamada, N. Nada, M. Saitoh, and K. Watanabe, Appl. Phys. Lett. 62, 435 (1993).
2. M. Yamada, M. Saitoh, and H. Ooki, Appl. Phys. Lett. 69, 3659 (1996).
3. C. L. Sones, S. Mailis, V. Apostolopoulos, I. E. Barry, C. B. E. Gawith, P. G. R. Smith, and R. W. Eason, J. Micromech. Microeng. 12, 53 (2002).
4. R. G. Batchko, V. Y. Shur, M. M. Fejer, and 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, and D. C. Hanna, Appl. Phys. Lett. 78, 3172 (2001).
6. K. Kintaka, M. Fujimura, T. Suhara, and H. Nishihara, Electron. Lett. 32, 2237 (1996).
7. M. Yamada and M. Saitoh, J. Appl. Phys. 84, 2199 (1998).
8. R. G. Batchko, V. Y. Shur, M. M. Fejer, and R. L. Byer, Appl. Phys. Lett. 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, and R. L. Byer, Ferroelectrics 236, 126 (2000).
10. T. Sugita, K. Mizuuchi, Y. Kitaoka, and K. Yamamoto, Jpn. J. Appl. Phys., Part 1 40, 1751 (2001).