Surface periodic poling of lithium niobate for efficient non-linear optical waveguide applications.

C.L. Sones, A.C. Busacca, V. Apostolopoulos, R.W. Eason, S. Mailis
Optoelectronics Research Centre, University of Southampton, Highfield, Southampton
SO171BJ, UK
E-mail: cls@soton.ac.uk

INTRODUCTION

Domain engineering of LiNbO₃ has been researched for a range of applications in areas as diverse as second harmonic generation and parametric oscillation, electro-optic Bragg gratings and piezoelectric micro-actuated devices. Non-linear frequency conversion achieved by quasi-phase-matched interaction remains an attractive route for realising efficient coherent blue-green light sources. Such quasi-phase-matching requires precise control of periodic domain inversion, with periods that can be as small as ~2 μm for first order conversion via second harmonic generation (SHG) from the near I.R. into the blue spectral region. Application of an electric field greater than the crystal's coercive field is the most widely employed route for fabrication of periodically inverted domain structures for non-linear quasi-phase matched interactions. It is experimentally very difficult to achieve such high aspect ratios in bulk poled material of typical thickness ~ 500μm. The high coercive fields required for domain inversion, together with the inherent non-uniformities and defects present in commercially available materials, restrict the routine applicability of electric field poling to periods of the order of ~4-5 μm in samples of this thickness. To circumvent this problem wafers can be thinned to ~100-150 μm to achieve such small domain periods, but this pre-treatment is both undesirable, expensive and for waveguide applications, unnecessary. Several other techniques such as controlled spontaneous backswitching, and the use of multiple short current pulses, have also been successfully used to generate periods of the order of 2.2-3.0 μm in bulk and waveguide geometries respectively. However fabrication of such poled crystals with very small periods particularly of sub-micron scales remains an elusive goal. Achieving large uniform periodically poled areas is difficult: we however have achieved periods of 1 μm by our technique which relies on over-poling the sample, thereby achieving the apparently undesirable effect of domain spreading and merging beneath the lithographically patterned photoresist layer. This technique results in superficial or surface domain inversion, which can be used in conjunction with a waveguide geometry for higher conversion efficiencies in non-linear interactions. Second harmonic generation experiments yielding blue light have been performed with surface poled annealed proton exchanged and Ti-indiffused waveguides confirming the utility of our technique.

TEXT

The technique we employed for surface poling, achieves the desired aim by simply over-poling the samples. The necessary modification of the conventional poling mechanism is achieved by increasing the value of the empirical factor, EF, which in the conventional poling case is used to correct for the variations in the supplier dependent sample stoichiometry and also accounts for thickness variations across the sample. Hence the empirical factor is often set to a value slightly greater than unity during conventional poling.

The samples to be poled were photolithographically patterned on the –z face with the desired periodic structure and the superficial domain inversion was then achieved with an application of an electric field. A single pulse of 11.1 kV, greater than the coercive field of the crystal was applied across the z-faces of the sample with the aid of a conductive gel. The applied voltage was computer-controlled and was terminated as soon as the predetermined charge, corresponding to the patterned domain inversion area had been transferred across the samples. The charge required for the domain inversion was calculated using the following equation:

\[ Q = 2P_z \times A \times EF \]

Where \( P_z \) is the spontaneous polarisation of the ferroelectric, A is the area within which the domains are to be inverted and EF is the empirical factor.

The empirical factor thereby effectively controls the domain spreading within the crystal after the initial stages of domain nucleation and coalescence. As the domain wall movement continues until the
applied voltage has been terminated, a much larger value of the empirical factor and hence a much larger value of the charge would effectively keep the computer-controlled voltage on for a correspondingly longer period of time. Hence a larger value of the EF would have the undesirable effect of lateral spreading of the domains, leading to periodic structuring with a non-ideal mark-to-space ratio. However the domain extensions into the un-patterned area can be affected by a careful selection of the EF. This effect, referred to as over-poling, results in domain inversion in all regions apart from just below the photoresist patterned sample face.

If over-poled with values of EF greater than two, the sample appears almost uniformly poled all across the patterned area when observed between crossed polarizers. However when etched in a HF acid, investigation reveals the survival of the patterned structure extending to a depth of a few microns beneath the surface. A scanning electron microscope image of the etched -z face of a sample surface poled with an EF of eight is seen in figure 1.

Initial parametric studies of this effect, for different values of the EF and the periods of the pattern imposed on the photoresist have been carried out. A conventional photolithographic technique using an amplitude mask was employed to pattern the photoresist with periods ranging from 2.47 µm to 4 µm. Domain widths of the order of 0.5 µm and periods of around 1 µm were obtained when laser exposure through a phase mask was used to pattern the resist. Etching studies of the crystallographic y-faces, of the samples patterned using the normal lithographic technique reveal the fact that the domains extend to depths of around 6 to 11 µm. The depth of the domains within the crystal is observed to change with the periods patterned on the photoresist and also with the value of EF used for poling, with the domain depths being larger for larger periods and smaller values of EF. The attained depths of a few microns for the near surface domains indicate that the technique has full compatibility with waveguide geometries formed via Ti-indiffusion and proton-exchange. A scanning electron micrograph of the etched -z face with domain inversion to a depth of ~3 µm only, is shown in figure 1.

![Fig 1. A SEM image of the etched domain structure.](image)

We have verified the applicability of this poling technique by poling annealed proton-exchanged and Ti-indiffused waveguides. Ti-indiffused channel waveguides spaced 100 µm apart and oriented parallel to the crystallographic x-direction, with widths varying from 1.5 µm to 8 µm were fabricated by a standard lift-off technique on the -z face of a 500 µm thick 30 mm long z-cut samples. A 100 nm thick titanium film was thermally evaporated on to the -z face of the sample and indiffused in an oxygen atmosphere at a temperature of 1050°C over a period of 11 hours. The -z face of the sample with the indiffused channel waveguides was then photolithographically patterned with a 0.7 mm wide pattern with a period of 2.47 µm. The periodic pattern had a mark to space ratio of 60:40. Domain inversion was then achieved by the superficial poling technique employing an empirical factor of magnitude 6.
With superficial poling however we did not find the need to take off the shallow LiClO out-diffusion induced domain-inverted layer, usually formed on the + z face during the high temperature metal indiffusion. This unavoidable polishing step, which ensures domains extending unidirectionally through the crystal, however can have an undesirable effect on the poling, as it would mean an undesired variation in the crystal thickness.

A wavelength tunable Ti:sapphire laser was then used to study how effective the superficial poling technique would be in achieving the domain uniformity required for quasi-phase matched second harmonic generation. For interaction with the optimal non-linear susceptibility tensor coefficient $d_{31}$ the incident beam was TM-polarised. The sample was maintained at an elevated temperature of $205^\circ$C to avoid any photorefractive damage that could be induced due to the generation of the second harmonic. We studied quasi-phase matched blue light generation from a surface-poled waveguide of width $\sim 2.5\mu$m. The phase-matching curve shown in figure 2 indicates a conversion from the TM$_{00}$ mode of the fundamental to the TM$_{00}$ and TM$_{01}$ modes of the second harmonic, with more efficient conversion from the TM$_{00}$ of the fundamental to the TM$_{00}$ of the second harmonic being achieved at 412.66 nm. The slight variation in the symmetry of the tuning curve could be as a result of structural imperfections along the length of the waveguide and also because of possible irregularity in the domain structure.

A maximum measured external second harmonic power of 3.5 mW was measured for a pump power of 70 mW, which corresponds to an effective length-normalised efficiency of 126 %/Wcm$^{-1}$ with an effective length of 0.75 cm deduced from the tuning curve. The lower value of the efficiency results from the non-ideal domain mark to space ratio of 80/20. The efficiency would however scale to four times the obtained value with a mark to space ratio of unity.

Fig 2. Phase-matching curve for surface poled Ti indiffused waveguides.
Similar experimental results were obtained with surface poled annealed proton exchanged waveguides. The tuning curve for the same is shown in figure 3 below.

![Tuning curve for SHG in surface poled annealed proton exchanged waveguide.](image)

**Figure 3.** Tuning curve for SHG in surface poled annealed proton exchanged waveguide.

**CONCLUSION**

In conclusion we have demonstrated a surface sensitive domain inversion technique. We are currently attempting to optimise the superficial poling technique and aim to achieve both uniform sub-micron periodically poled structures and samples of periodicity ~ 2.5µm with the ideal 50/50 mark to space ratio.

**References**