Quasi-phase-matched blue light generation in bulk lithium niobate, electrically poled via periodic liquid electrodes

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Periodic poling of z-cut lithium niobate is reliably achieved in 0.2mm thick samples, patterned with photoresist on one face and subjected to pulsed electric fields via liquid electrodes. The acceptance bandwidth for third order second-harmonic generation reveals that the full 3.3mm length contributes to the conversion.

Introduction: Periodic poling was one of the first suggestions as to how to solve the phase-matching problem in nonlinear parametric interactions [1]. When the nonlinear coefficient alternates in sign with the correct spatial period, quasi-phase-matching (QPM) is achieved. Compared to birefringent phase-matching, QPM allows both access to new wavelengths and a higher conversion efficiency.

Several papers report frequency doubling (FD) in periodically poled crystals grown via a modified Czochralski process [2, 3]. It is, however, a major challenge to achieve highly uniform periodicity over centimetre lengths. A later variation on this technique is laser-heated pedestal growth [4]. Periodic poling has also been achieved by photolithographically defined diffusion and ion exchange. The resulting domain structure is created only in a shallow surface layer, is a faithful replica of the mask, and is useful for nonlinear interactions in waveguides but not in bulk samples.

Recently, electron-beam writing and application of high-voltage pulses via patterned aluminium electrodes have been demonstrated as techniques for room-temperature fabrication of bulk QPM crystals [5, 6]. In this Letter we present some further developments of high-voltage poling technology and bulk FD in the resulting crystals.

Fabrication: Liquid electrodes are well known for their benefits in the electric field poling of ferroelectrics [7]. We have found that 1 μm of standard photoresist acts as an effective barrier to domain inversion with liquid electrodes. It can thus be patterned to produce periodic poling.

Our setup was similar to that used by Camlibel [7]: Filter paper was soaked in a solution of LiCl in water and placed on both sides of a sample with a photoresist pattern on one side. This sandwich was then clamped between electrodes connected to ground and a high-voltage pulse generator. In the initial trials, it made little difference whether the photoresist pattern was applied to z^- or z^+. For the later fabrication of periodic structures, we patterned z^- with satisfactory results.

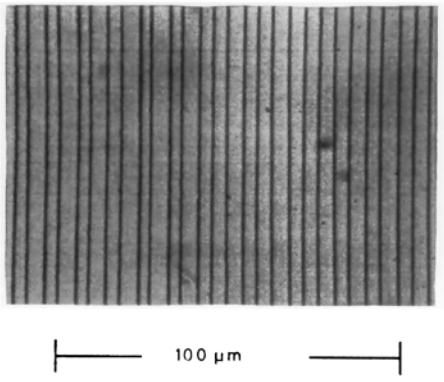

Fig. 1 *Domain structure of patterned side, revealed by etching*

This was initially the negative z-side

Typically we used 1–10 high-voltage pulses with durations in the range 0.1–10ms. The pulse amplitude was chosen to result in a slightly higher field strength than the coercive field of ~23kV/mm. The peak current through the electrodes during poling ranged from 0.5 to 50mA. The final domain structure does not depend strongly on the chosen parameter values. It is, for instance, possible to achieve complete domain inversion with one pulse. The single most important feature is that the transferred charge should be twice the spontaneous polarisation multiplied by the desired area of inversion. To keep the poling voltage below 6kV, we limited the processing to 0.1 and 0.2mm thick samples of lithium niobate. Figs. 1 and 2 show typical results for the domain walls on the patterned face and the side of the planar electrode, respectively. Evidently, there is a larger random variation in the position of the domain walls on the latter side.

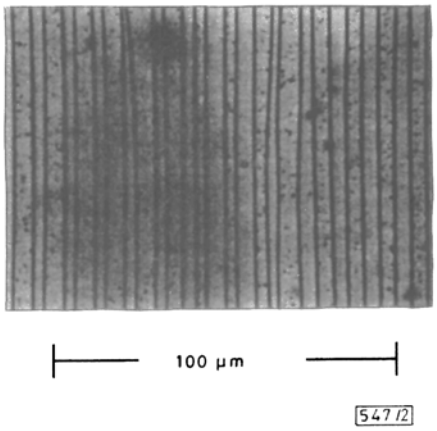

Fig. 2 *Domain structure on planar electrode side*

Optical experiments: Our best results to date were obtained in a 0.2mm thick sample with a period of 9μm (domain width 4.5μm), designed to give 3rd order quasi-phase-matched FD from 832nm to 416nm. The crystal length was 3.3mm. A tunable Ti:sapphire laser with a 0.04nm bandwidth was used in the experiments. The waist spot size inside the crystal was $w_0 = 18\mu m$ which gives a ratio of sample length to confocal length $L/b = 0.63$. Lenses with shorter focal length did not give any higher efficiency. The blue power level varied considerably with position across the sample, indicating that there are regions where the domains are less regular. The output power from a good position as a function of input power is shown in Fig. 3. It is proportional to the square of the input all the way up to an output of 20μW for an input of 300 mW, which gives an indication that photorefractive damage is not

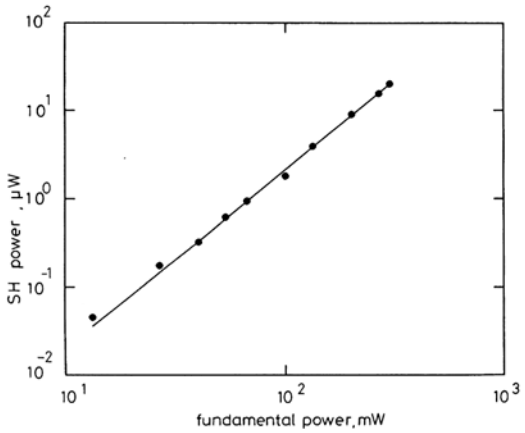

Fig. 3 *Second-harmonic power as function of input power*

a problem. This was also tested separately using the 488 nm line of an argon ion laser. At a power level of 10μW, the beam was focused to a waist spot size of 10 μm inside the sample and an aperture was inserted on the output side that blocked 50% of the power. This value of 50% remained constant up to the maximum power of 110mW.

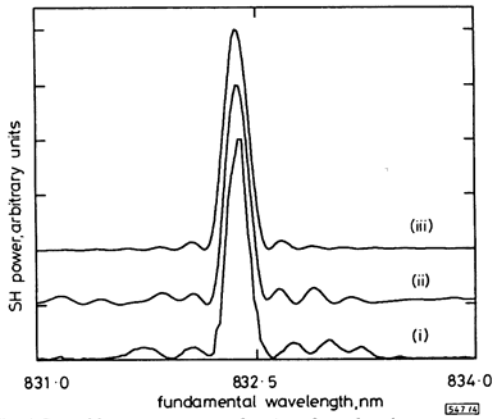

Fig. 4 *Second-harmonic power as function of wavelength*

The curves are shifted vertically for improved visibility
(i) experimental results
(ii) computation for crystal with domain-wall offset with standard deviation of 0.8 μm; scale of *y*-axis is same as for experimental curve
(iii) computation for perfectly uniform crystal

In Fig. 4 the second-harmonic output is shown as a function of fundamental wavelength. The phase-matching peak has the bandwidth predicted by theory, slightly less than 0.2nm, demonstrating that the full length of the crystal is involved in the interaction. The conversion efficiency of 0.07%/Wcm is ~10 times smaller than the theoretical value for a perfectly periodic crystal. Note that we have normalised against the physical length of the sample, not an 'effective' length derived from the phase-matching bandwidth. It has been shown that for an independent random offset from the ideal domain-wall position with Gaussian distribution, a standard deviation of roughly 0.5 coherence lengths gives a tenfold reduction of output power [8]. In our case, this means 0.8μm, which seems quite reasonable judging from Figs. 1 and 2. Fig. 4 curve (ii) is the trace for a simulated crystal with a standard deviation of $\sigma = 0.8$ μm. This displays both a shape and a reduction in peak efficiency that are similar to the experimental curve.

Conclusion: High voltage pulses, applied using liquid electrodes and a periodic photoresist pattern, produce effective periodic domain inversion in lithium niobate. The experimental third-order phase-matching bandwidth of a 3.3mm bulk crystal was equal to the theoretically expected value, showing that the whole crystal took part in the conversion. The conversion efficiency was reduced to 10% of the theoretical value by a random variation of the domain-wall positions. These values are expected to improve when we develop our processing to first-order gratings.

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