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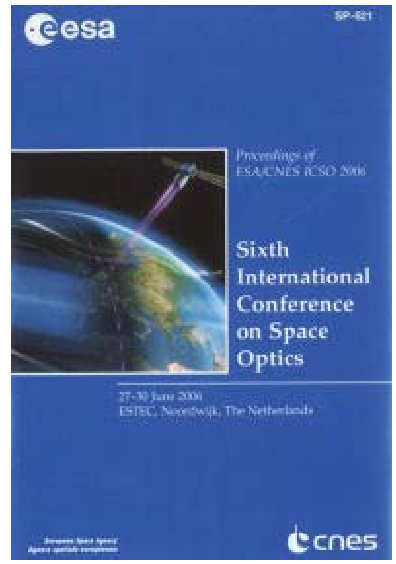

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NANOTECHNOLOGY IN LITHIUM NIOBATE FOR INTEGRATED OPTIC FREQUENCY CONVERSION IN THE UV

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ABSTRACT

In the domain of Earth Explorer satellites nanoengineered nonlinear crystals can optimize UV tunable solid-state laser converters. Lightweight sources can be based on Lithium Niobate (LN) domain engineering by electric field poling and guided wave interactions.
In this Communication we report the preliminary experimental results and the very first demonstration of UltraViolet second-harmonic generation by first-order quasi-phase-matching in a surface-periodically-poled proton-exchanged LN waveguide. The pump source was a Ti-Sapphire laser with a tunability range of 700-980 nm and a 40 GHz linewidth. We have measured UV continuous-wave light at 390 nm by means of a lock-in amplifier and of a photodiode with enhanced response in the UV. Measured conversion efficiency was about $1\%W^{-1}cm^{-2}$. QPM experiments show good agreement with theory and pave the way for a future implementation of the technique in materials less prone to photorefractive damage and wider transparency in the UV, such as Lithium Tantalate.

1. INTRODUCTION

In the domain of Earth Explorer satellites nano-engineered nonlinear crystals can optimize UV tunable solid-state laser converters [1]. Lightweight sources can be based on Lithium Niobate (LN) domain engineering by electric field poling and guided wave interactions. Ferroelectric periods below the micro scale are needed to fabricate integrated optical devices based on quasi-phase-matching (QPM) techniques. Conventional poling techniques are limited to periods greater than 6 microns. On the contrary, very promising results have been obtained with the technology of "Surface Periodic Poling" (SPP) on LN [2-3]. SPP with respect to bulk poling has a minimum period pitch shorter by two orders of magnitude as short as 200 nm [4]. Also, SPP -unlike bulk periodic poling- do not invert domains in the all sample thickness (0.5 mm), but just under the -z face. This means that only configurations based on interactions near the surface layer can be devised. Therefore, it seems quite promising to demonstrate the compatibility of SPP with channel waveguides. Such a short nonlinear grating is required for Quasi-Phase-Matched (QPM) Backward Second Harmonic Generation (BSHG) and Counterpropagating Optical Parametric Amplification (COPA) [5].
The proton exchange (PE) process is a consolidated technique for the fabrication of low losses waveguides and devices with excellent characteristics such as good electro-optical interactions, high nonlinear coefficient and low susceptibility to the photorefractive damage [6]. Therefore PE process control on LN and domain engineering in the nanoscale (by means of the SPP technique) will render it possible to optimize parametric conversion and also all-optical functions. This requires a better understanding of the nature and reproducibility of the SPP-PE combination, with a careful investigation of the poling patterns and of the channel waveguides [7]. Such an experimentation will make more evident physical mechanisms, phenomenology and potentiality of SPP-PE technology.
In this Communication we will give experimental results regarding all the technological issues related to LN SPP-PE compatibility and nonlinear measurements. In particular we report on QPM parametric conversion with a long period (16.8 μm) SPP pattern, useful to test the technology, and on the very first demonstration of UV SH generation by first-order quasi-phase-matching (2 μm period) in a channel waveguide configuration.

2. EXPERIMENTAL

We have fabricated, on z-cut LN crystals, periodic structures with pitch down to 750 nm. Firstly a 1.3 μm thick photoresist film (S1813 Shipley) has been spin coated on the –z crystal face of a 500 μm thick LN sample and, subsequently, the periodic pattern has been defined with standard photolithography. The developed pattern serves as the insulating layer to give electric field periodic contrast during high voltage poling.
The sample has been placed between two electrodes covered with a thin gel electrolyte layer to ensure

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proper electrical contact. The LN faces were in this way connected to the high voltage supplied by a 662 Trek amplifier of the waveform generated by an Agilent model 3220A; a digital oscilloscope (54641A Agilent) has allowed to monitor voltage and current during poling.

In order to exceed the LN coercive field and achieve the surface periodic inversion we have applied for an appropriate time interval a 1.3 kV pulse over a 10 kV bias so to satisfy the overpoling condition:

$$Q > 2 \cdot A \cdot P_s \tag{1}$$

where Q is the flowing charge, A the poled area and Ps the spontaneous polarization of the ferroelectric crystal. In this way, without an exact control of flowing charge, it is possible to let the domain walls merge completely on the +z face of the crystal but not underneath the insulated photoresist coated –z face regions, where still domains periodically keep their original orientation.

After the surface poling technology was satisfactorily set-up, the research efforts were devoted to the realization of suitable waveguides by proton exchange. In order to obtain a good conversion efficiency it is required an high optical power density in the all path of nonlinear interaction between input radiation and the generated radiation. For this reason the SPPLN structure is fabricated on channel optical waveguides.

In a first step we have experimented the higher index beta phase waveguides supporting a single mode at 1.55 μm. LN was initially coated with a SiO_2 layer, and then channel SiO_2 openings ranging between 1 to 7 μm have been fabricated with standard photolithography and HF etching.

The PE process has been carried out by dipping the LN crystal in a fused mixture of benzoic acid and 1% lithium benzoate, contained in a sealed ampoule, at about 300 °C. Within the LN crystal lattice it occurs a process of H^+ ions diffusion and substitution of lithium ions. As a result, a large increment of the extraordinary index of refraction and a reduction of the ordinary one is obtained in the surface layer where diffusion occurs. Nevertheless, as already known for bulk poling [6], crystal stoichiometry in the waveguide region was so different from the bulk values that both surface poling after PE was prevented and also domain erasure by PE was determined (Fig. 1).

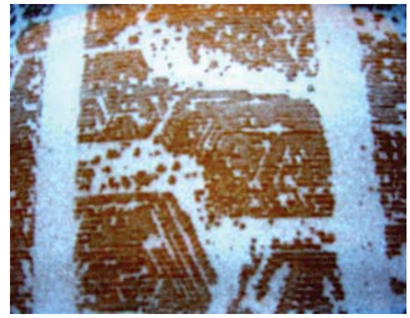

Fig. 1. Poling on a β-phase PE waveguide.

For this reason, we moved to the realization of alpha phase waveguides (Fig. 2) using a 3% lithium benzoate mixture at about 300 °C for 4 days [8]. In this case the index increment is smaller but LN stoichiometry is almost preserved, and actually first experiments show both that it is possible to superimpose the SPP over PE waveguides (Fig. 2), and that it is also possible to carry out PE on substrates which were previously surface poled.

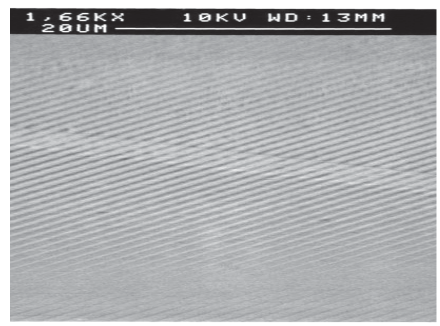

Fig. 2. Poling on a α-phase PE waveguide.

Morphology and distribution of ferroelectric domains after poling and PE have been revealed performing selective chemical etching, and inspected through electron microscopy and surface analysis techniques. In order to establish the ferroelectric nonlinear nature corresponding to surface relieves made evident by HF etching, nonlinear measurements have been carried out on a 16.8 μm SPP 4 μm wide single mode waveguide about 1540 nm.

A ps-pulsed laser beam was focused onto the waveguide input to study the nonlinear response of the sample by tuning both fundamental wavelength and power. The temperature was kept constant at room-value during all measurements. We measured the output second harmonic (SH) and, for each channel, we obtained SH tuning curves (SH conversion efficiency versus fundamental wavelength) as the one shown below (Fig. 3).

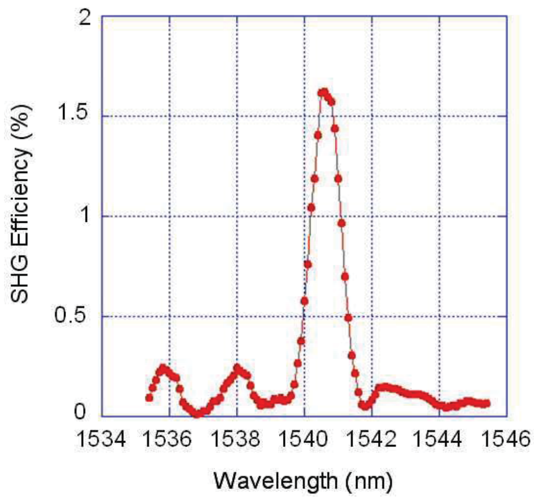

Fig. 3. SHG Efficiency at low FF power.

As we can observe, the full width at half maximum (FWHM) is about 1 nm. This is what is expected for a uniform domain structure over the whole 10 mm long sample (see Fig. 4 in the next page), as given by the expression of FWHM $\delta\lambda$ in a periodically poled sample:

$$\delta\lambda = \frac{0.4429\lambda}{L}\left|\frac{n_2-n_1}{\lambda}+\frac{\partial n_1}{\partial\lambda}-\frac{1}{2}\frac{\partial n_2}{\partial\lambda}\right|^{-1} \qquad (2)$$

being n_1 = 2.1396 and n_2 = 2.1848 the measured effective indices of the fundamental modes at FF and SH, respectively, λ is the FF wavelength (in nm) and L the sample length (in nm) and the derivatives evaluated at the pertinent wavelengths.

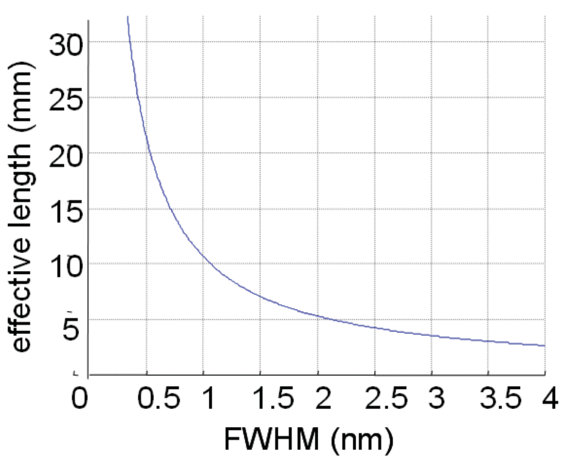

Fig. 4. Effective grating length vs FWHM.

We also observed that the optimum QPM wavelength (FF) varies with input (peak) power, as visible in the following Fig. 5.

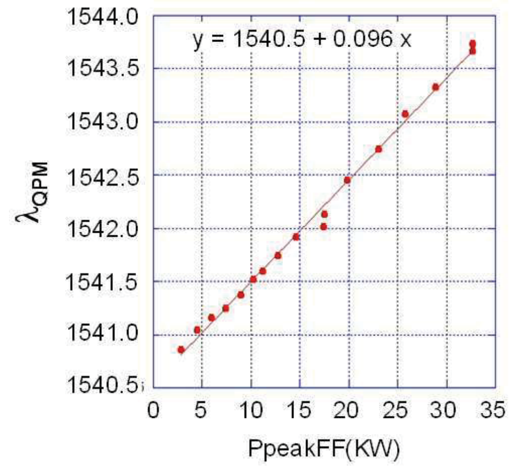

Fig. 5. Measured SH peak drift vs FF power.

Hence, by adjusting both FF wavelength and power simultaneously, higher SH conversion efficiencies can be achieved, as large as >10%.

Finally, with the 2 μm poling period sample, we carried out UltraViolet second-harmonic generation measurement. First-order quasi-phase-matching was obtained in this SPP-PE LN channel waveguide. The pump source corresponds to a 40 GHz linewidth of a tunable Ti:Sapphire laser. The LN sample was kept at a temperature of 250°C to reduce the strong photorefractive damage. We have measured UV continuous-wave light at approximately 390 nm by means of a lock-in amplifier at 1.5 KHz to reduce noise level. Measured spectral efficiency of generated UV, Fig. 6, has a peek of about $1\%W^{-1}cm^{-2}$.

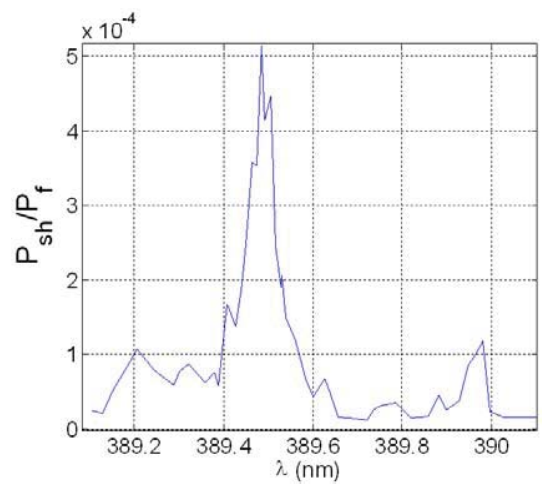

Fig. 6. UV SHG spectral efficiency.

A factor of 0.1 reduction in the measured efficiency was attributed to no optimum mark-to-space ratio (10:90) in the region of the guide. A quadratic response of the generated light with pump power is evident from the photodiode current plot shown in fig. 7.

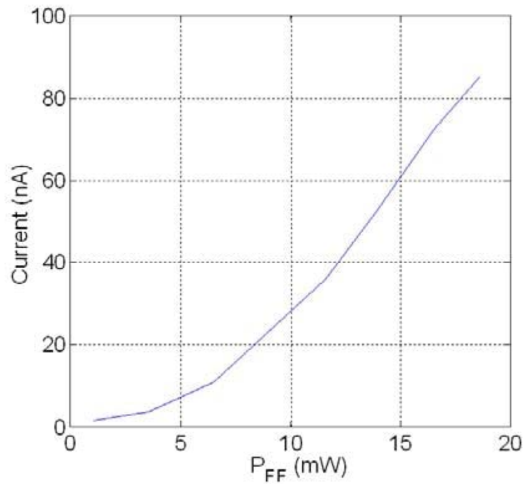

Fig. 7. Quadratic behaviour of generated UV light (photodiode current) versus pump power.

3. CONCLUSIONS

We have demonstrated surface periodic poling compatibility with alpha phase proton exchange waveguides. Nonlinear measurements have been carried on about 1540 nm pump wavelength to confirm SPP effectiveness and to evaluate periodic domain uniformity. With a proper poling period (2 µm) UV generation was subsequently demonstrated, reaching a 1%$W^{-1}cm^{-2}$ efficiency. Further experiments will be devoted to the optimization of domain thickness and mark-to-space ratio. New materials showing wider transparency in the UV and lower photorefractive damage, such as Lithium Tantalate, will also be investigated to improve device efficiency. Further research will focus on BSHG configuration measurements.

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