Fabrication and characterization of photonic structures in crystals of the KTiOPO$_4$ family

Fabricación y caracterización de estructuras fotónicas en cristales de la familia KTiOPO$_4$

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ABSTRACT:

Two different procedures have been used for the fabrication of photonic structures in crystals of the KTiOPO$_4$ family. We used ultrafast laser ablation to fabricate a 1D photonic crystal on the surface of an RbTiOPO$_4$ crystal, and we evaluated its properties as diffraction grating. We also present the recent advances we developed in a new procedure of fabrication of 2D and 3D photonic crystals of KTiOPO$_4$ grown on the surface of a KTP substrate by liquid phase epitaxial means within the pores of a silicon macroporous template, and their morphological, structural and optical characterization.

Key words: Diffraction Gratings, Photonic Crystals, Nonlinear Optical Materials, Nonlinear Optics, Laser Ablation, KTiOPO$_4$, Template Growth, Liquid Phase Epitaxy.

RESUMEN:

Se presentan dos procedimientos diferentes para fabricar estructuras fotónicas en cristales de la familia KTiOPO$_4$. En primer lugar hemos usado ablación mediante láser ultrarápido para fabricar un cristal fotónico 1D en la superficie de un cristal de RbTiOPO$_4$, y hemos evaluado sus propiedades como red de difracción. También presentamos recientes avances obtenidos mediante un nuevo procedimiento de fabricación de cristales fotónicos 2D y 3D de KTiOPO$_4$, crecidos epitaxialmente en la superficie de un substrato de KTP mediante fase líquida dentro de los macroporos de un molde de silicio. Se presentan resultados de su caracterización morfológica, estructural y óptica.

Palabras clave: Redes de Difarcción, Cristales Fotónicos, Materiales Ópticos No Lineales, Óptica No Lineal, Ablación Láser, KTiOPO$_4$, Crecimiento con Molde, Epitaxia de Fase Líquida.

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1. Introduction

Since 1987, when the first work about photonic crystals (PCs) was published [1,2], many interesting properties have been studied in one-, two- and three dimensions. Besides their peculiar linear optical properties, photonic crystals present many interesting features for controlling the nonlinear optical interactions. They provide the possibility to enhance [3] a second order nonlinear optical interaction and an alternative phase-matching mechanism [4]. The periodic modulation of both refractive index and second order nonlinear susceptibility might allow backward parametric oscillation [5], a nonlinear effect predicted many years ago but not yet observed experimentally.

To obtain a very efficient and durable nonlinear interaction in a PC it would be adequate to use a material with high second order nonlinear optical properties, such as KTiOPO$_4$ (KTP) and its isostructural RbTiOPO$_4$ (RTP). KTP has been recognized as one of the materials of choice for second harmonic generation of Nd:YAG light, due to its extremely low onset power threshold, high power conversion efficiency, and high threshold to laser-induced damage [6]. Among the isostructural materials to KTP, RTP presents a high surface damage threshold (9.0×10^6 MW m^{-2}) and large temperature matching bandwidth (50 K cm^{-1}), double than that of KTP [7].

In this paper we propose two different strategies to fabricate photonic crystals based on these non-linear optical materials. We fabricated diffraction gratings on the surface of an RTP crystal by ultrafast laser ablation, and evaluated its behaviour as a Bragg diffraction grating. One of the challenges for fabricating higher dimensional (2D and 3D) photonic crystals is the production of these structures with sufficient precision to prevent scattering losses blurring the crystal properties. We present here the advances we developed recently in a completely new procedure that combines top-down and bottom-up approaches to fabricate 2D and 3D PCs of non-linear dielectric materials [8]. These crystals are grown on the surface of a KTP substrate within the pores of a silicon macroporous template.

2. Fabrication of diffraction gratings

A 1D relief grating was recorded on the surface of a RTP sample by ultrafast laser ablation. This technique uses very short and intense laser pulses to

remove thin layers from the surface of a bulk target by means of physical mechanisms different from those taking place in conventional laser ablation. The collateral thermal and mechanical effects around the ablated area are diminished in such an extent, that precision and quality of the microstructures higher than those obtained with other techniques can be achieved. Non-linear absorption and ionization processes are on the basis of this technique. Focusing on dielectrics, within the duration of a pulse and for moderate intensities, a thin layer on the surface of the material is almost fully ionized by multiphoton and collisional mechanisms so that the number of free electrons rapidly amounts to the solid-state density. As a consequence of the poor electric transport properties of the material, surface charging takes place and the ultraintense "quasielectrostatic" field generated overcomes the binding energy of the ions and drags them out of the solid. This mechanism is known as Coulomb explosion and since thermal coupling with the lattice is negligible during this short period of time, the process is a purely non-thermal process.

For larger intensities, total ionization of the surface is achieved for the leading edge of the laser pulse. Therefore, free electrons can absorb energy from the laser pulse in the presence of the lattice atoms and ions by means of inverse bremsstrahlung mechanism. This absorbed energy contributes to raise the temperature of a deeper surface layer by electron heat diffusion to a value close to the thermodynamic critical temperature giving rise to a phase explosion process, resulting in violent expulsion of both vapor and equilibrium liquid droplets [9]. The depth of the ablated layer is determined by the electron heat diffusion length and the laser fluence. Although the process is still very fast, some thermal damage must be expected on the areas surrounding the microstructured region.

We have used a commercial Ti:Sapphire oscillator (Tsunami, Spectra Physics) and a regenerative amplifier system (Spitfire, Spectra Physics) based on chirped pulse amplification (CPA). The system delivers linearly polarized 120-fs pulses with central wavelength 795 nm with a repetition rate of 1 kHz. The maximum available pulse energy is 1 mJ but for the purpose of microstructuring the grating it was reduced to 0.78 µJ using a half-wave plate and a linear polarizer.

The transverse mode is gaussian and the beam width is 9 mm ($1/e^2$ criterion). The beam was then focused by a 50 mm achromatic lens resulting in a peak fluence of ~7 J cm^{-2} at focus.

The sample was placed on a motorized XYZ translation stage in order to achieve optimal focusing on the target surface. The focused beam moved in straight lines across the sample surface at a constant scanning speed of 130 µm s^{-1} avoiding iterative passes along the same line. The pitch between successive grooves was set to 15 µm. For this scanning speed, the number of pulses contributing to the ablation of a point within the sample surface was approximately 40. We have estimated the ablation threshold fluence to be 1.44±0.18 J cm^{-2} for 40 pulses. For multishot conditions (>100 pulses) the value for the threshold decreases to 1.18±0.15 J cm^{-2}.

Figure 1 shows a picture of the RTP sample taken with a Carl Zeiss Axio Imager A1 optical microscope after the ultrafast laser ablation process where it can be seen the diffraction grating generated on the surface of the sample. The lattice constant estimated from this microscope is approximately 15 µm.

We have recorded Bragg-diffraction spectra of this sample by using a FT-IR spectrometer (Bruker-Vertex 70) equipped with a special reflectivity attachment. The light source was a halogen tungsten lamp, and we collected the intensity of the diffracted light with a DLATGS detector in the spectral range from 7500 to 400 cm^{-1}. The incoming light was pointed perpendicular to the plane of the sample and the diffraction spectra were measured in a direction perpendicular to the grooves and at collection angles ranging from 24 to 60º in 2º steps. The measured data are represented in Fig. 2 with an intensity plot as a function of the wavelength and the diffraction angle.

To evaluate the lattice constant, the Bragg-diffraction spectra were fitted to the following 2-variable function:

$$I(\lambda, \sin\theta) = \sum_{n=1}^{3} \exp\left[\left(\frac{\sin\theta - \frac{1}{a}n\lambda}{w_n}\right)^2\right], \quad (1)$$

where w_n takes into account the width of the diffraction peaks, a is the lattice constant and n is an integer (the number of the diffraction order). The fitting of this function to the experimental data gives a robust estimation of the lattice constant

from the data, since all the measurements are taken into account simultaneously. The value of the lattice constant measured by this procedure was 14.92 μm, which was in good agreement with the value estimated by optical microscopy.

Fig. 1. Optical microscopy image of the diffraction grating generated on the surface of an RTP crystal by ultrafast laser ablation.
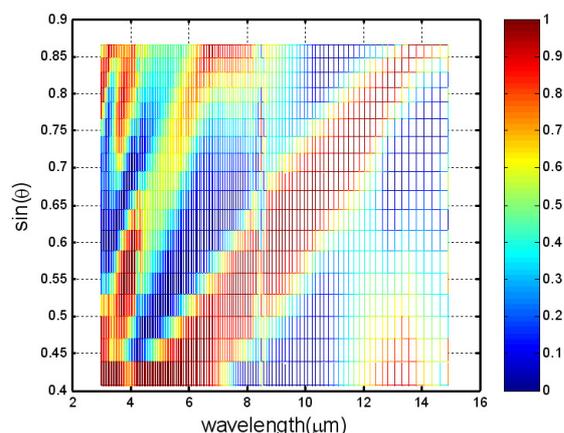

Fig. 2. 2D intensity plot as a function of the wavelength and the diffraction angle for the diffraction grating generated on the surface of an RTP crystal. Dark zones represent the diffraction orders.

3. Crystal growth of 2D and 3D photonic crystals by liquid phase epitaxy

The procedure for fabricating 2D and 3D photonic crystals we developed combines two well known techniques: the fabrication of 2D and 3D macroporous silicon membranes and the epitaxial growth of KTP within the pores of these membranes. This procedure, in addition to its simplicity, results in KTP and silicon integrated in a single structure that would eventually be used to generate or modulate light.

We fabricated these 2D and 3D PCs in four different steps which involved the preparation of high quality ordered macroporous silicon templates by light assisted electrochemical etching [10], the growth of the KTP epitaxial layer within the pores of the silicon template by liquid phase epitaxy, the polishing of the top or bottom surface of the KTP epitaxial layer, and finally, a selective etching of the silicon matrix.
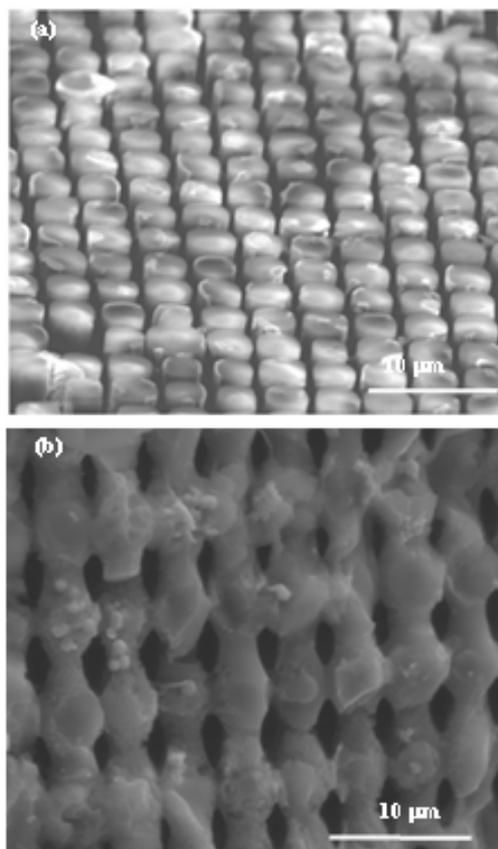

Fig. 3. SEM pictures of a (2D) and (b) 3D KTP photonic crystal.

The 2D or 3D silicon template was closely bound to a KTP single crystal substrate with typical dimensions 5 mm long, 3 mm wide and 1 mm thick. The crystal was oriented in such a way that the largest surface was perpendicular to the *c* crystallographic direction. The template/substrate set was immersed into a solution formed by mixing of K_2O, P_2O, TiO_2 and WO_3 with a molar % composition K_2O-P_2O-TiO_2-WO_3 = 42-14-14-30 for a period of time between 5 and 10 min at a supersaturation degree of about 2%. After growth, the top part of the KTP photonic structure was polished by using diamond powders ϕ = 0.1 μm. Once an optical-quality surface for the KTP

photonic structure was obtained, the last step in our approach was to remove the silicon template by selective chemical etching with TMAH diluted in distilled water (5 vol%) at 354 K. The effect of the selective etching is clearly visible in Figure 3, where a side-view image of the KTP photonic structure taken with the SEM is shown.

By using the aforesaid procedure we fabricated different KTP PCs with triangular and square lattices and lattice parameters ranging from 4.5 to 10 µm.

The crystallographic orientation of the substrate is transferred to the KTP photonic structure which we confirmed by X-ray texture analysis. The transference of the crystallographic orientation of the substrate to the KTP PC is advantageous for second order nonlinear applications since it may allow the use of the most appropriate nonlinear or electrooptic coefficient for a specific application in combination with a phase-matching condition, which would be provided by the photonic structure. This is not possible with bulk KTP where phase-matching relies on the birefringence and the coefficients with the largest nonlinearity cannot be used for efficient SH generation.

A more detailed optical characterization of a 2D KTP PC can be found in ref. [13]. Here we present the results of some basic light diffraction measurements performed on these structures. We placed the sample on a XYZ positioning stage mounted on a rotating stage to be able to change the angle of incidence to record the linear diffraction of the sample. Once mounted, the sample was illuminated with light at 527 nm. Some pictures of the observed diffraction patterns from samples with triangular and square lattices can be seen in Fig. 4 (a) and (b). In those pictures one can see the presence of an intense central spot corresponding to the specular reflection and some other spots that reflect the lattice of the sample.

The PC properties of a 2D KTP structure were demonstrated by performing a measurement of the specular reflection as a function of the wavelength of the incident field. We used a 2D KTP PC of KTP columns in air and periodicity of 4.5 µm. The sample was shined with p-polarized laser pulses, spectrally tuned in the range 940-1220 nm, at an angle of incidence of 25° with respect to the axis of the photonic structure. The specular reflectance spectrum, as shown in Fig. 4 (c), presents a dip at

1100 nm, which corresponds to the spectral position of the third-order Bragg reflection band, which was determined from a numerical calculation using the transfer matrix method.
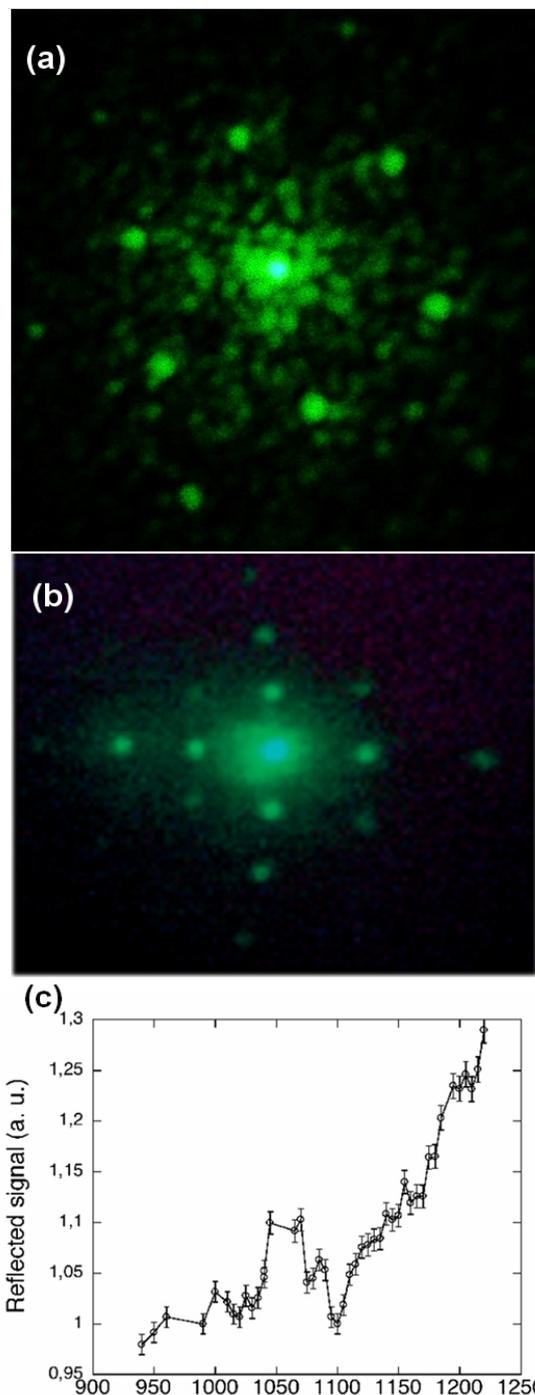

Fig. 4. Pictures of the linear diffraction pattern from a 2D KTP PC with (a) triangular lattice, and (b) square lattice. (c) Measurement of the specular reflection as a function of the wavelength of the incident light.

4. Conclusions

We have microstructured the surface of non-linear optical crystals, such as RbTiOPO$_4$, by ultrafast laser ablation for the fabrication of 1D photonic crystals.

We have also fabricated 2D and 3D KTP photonic crystals by the templated growth of KTP by liquid phase epitaxy within the pores of a Si template. In this way, the single-crystal substrate provided the desired crystallographic orientation, while the silicon template gave the desired final form to the thin layer.

In the data we collected from the growth procedure we followed there is no indication that KTP PCs with smaller periods could not be grown, as macroporous silicon templates or templates of other kind, with a smaller diameter pore become readily available. Additionally, this approach to fabricate 2D and 3D photonic structures can be extended to other nonlinear and optical materials that can be grown by liquid phase epitaxy.

With these photonic structures, in the future, we pretend to study different effects, such as the generation of light at different frequencies, parametric oscillation, and guiding and bending of light in previously designed line defects.