

Directional dependence of the second harmonic response in two-dimensional nonlinear photonic crystals

P. Molina,¹ M. O. Ramírez,^{1,a)} B. J. García,² and L. E. Bausá¹

¹Dept. Física de Materiales, Universidad Autónoma de Madrid, 28049 Madrid, Spain

²Dept. Física Aplicada, Universidad Autónoma de Madrid, 28049 Madrid, Spain

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A two-dimensional periodic arrangement of alternating ferroelectric domains in LiNbO₃, with asymmetric domain duty cycle has been used to demonstrate that counterpropagating beams along the polar axis can generate disparate second harmonic patterns, which are nonsymmetrical with respect to the source and the point of observation. These findings provide alternative routes to generate dissimilar light-matter interaction processes in two-dimensional structures assembled onto polar surfaces, including metals for plasmonics or biological compounds. © 2010 American Institute of Physics. [doi:10.1063/1.3459975]

Symmetry and anisotropy govern the physical and chemical properties of materials. Indeed, a great variety of physical systems are sensitive to the direction of applied external fields with its subsequent impact on their applications.^{1–3} This is particularly true when a loss of a point-symmetry operation leads to the existence of two equivalent states that differ only in their orientation and/or handedness. In ferroelectric crystals, symmetry breaking occurs at the phase transition resulting in a ferroelectric phase in which the crystals have two equivalent states which differ only in the orientation of the spontaneous electric polarization, \mathbf{P}_s . The presence of a defined \mathbf{P}_s is at the base of important applications of ferroelectrics such as capacitors, transducers, or nonvolatile memories, where the orientational states of \mathbf{P}_s can be used as the two states of a binary system.^{4–6} More recent examples of relevant phenomena related to unidirectionality in ferroelectrics are the demonstration of diode-like effect in BiFeO₃,⁷ or the use of polar surfaces to direct chemical interactions onto a surface to assembly nanostructures.⁸ Symmetry breaking in the ferroelectric phase leads to noncentrosymmetric crystal structures, therefore, showing a nonzero value of the nonlinear susceptibility, $\chi^{(2)}$, which makes them useful in nonlinear optical devices such as frequency mixers, optical parametric oscillators, or light modulators.^{9,10} Here we show that the sense of \mathbf{P}_s in trigonal ferroelectrics can lead to a pronounced spatial dependence of the second harmonic generated signal. By using a two-dimensional (2D) periodic arrangement of alternating ferroelectric domains in LiNbO₃, with asymmetric domain duty cycle (97%/3%), we demonstrate that counterpropagating beams along the polar axis can generate disparate SHG patterns which are nonsymmetrical with respect to the source and the point of observation. These findings provide alternative routes to generate dissimilar light-matter interaction processes in 2D structures assembled onto polar surfaces, including metals for plasmonics or biological compounds.

To reveal the effect that the sense of counterpropagating beams along the polar axis, z , produces on the quadratic nonlinear response of the system, nonsymmetric SHG spatial patterns are required. The simplest case corresponds to pat-

terns lacking axial symmetry with respect to a fundamental beam propagating along z . For this aim, we have fabricated a 2D nonlinear photonic crystal (2D-NLPC) in LiNbO₃ by creating a periodic arrangement of ferroelectric domains with alternating orientation of \mathbf{P}_s . We have employed direct electron beam writing on the $-z$ face of the crystal by means of a Philips XL30 Schottky field emission gun electron microscope driven by an Elphy Raith nanolithography software. Further details on the irradiation process can be found elsewhere.^{11,12} Figure 1(a) shows a picture of a portion of the 2D distribution of ferroelectric inverted domains. The fabricated NLPC consisted of a 2D hexagonal array of hexagonal column-shaped polarization inverted domains ($\mathbf{P}_s \downarrow$) embedded into a single domain LiNbO₃ crystal of opposite polarization ($\mathbf{P}_s \uparrow$). The inverted domain columns were directed along the polar axis of the crystal traversing the whole sample thickness (1 mm). The diameter of the inverted domains in the xy plane was 3 μm and the lattice parameter of

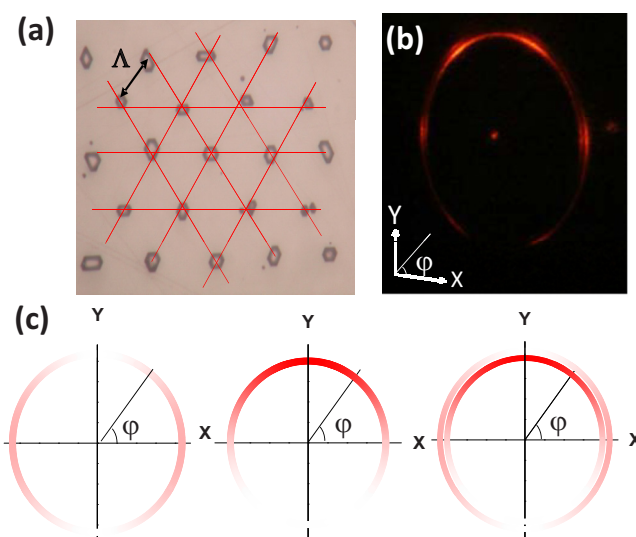


FIG. 1. (Color online) (a) Optical micrograph showing the 2D distribution of ferroelectric inverted domains. Lines on the picture are a guide to show the hexagonal symmetry of the domain pattern. (b) Far field SHG pattern ($\mathbf{k}_o \parallel z$). (c) Projection of the intensities of the ordinary (left), extraordinary (center), and combination of both (right) SHG beams, as follows from Eq. (1).

^{a)}Electronic mail: mariola.ramirez@uam.es.

the hexagonal array was $\Lambda = 20 \text{ } \mu\text{m}$. The spatial extension of the pattern was $2 \times 2 \text{ mm}^2$. The ratio, f , of the area of the inverted region to the entire area, was ~ 0.03 . Therefore, the orientation of \mathbf{P}_s of the original single domain sample is largely dominant. We use such a particular structure as a way to detect SHG to the naked eye when the fundamental beam is launched along the polar direction, which otherwise is not detectable.

2D-NLPC are being the object of several works.^{13–15} In these systems the structure of ferroelectric domains produces a modulation of the sign of the second order nonlinear susceptibility $\chi^{(2)}$ providing a set of reciprocal lattice vectors in the plane of modulation, xy . As a result, SHG occurs at multiple directions and nonlinear Bragg diffraction patterns can be obtained.^{16,17} Figure 1(b) shows the far field SHG pattern obtained when the fundamental beam propagates parallel to z . In this configuration the SHG waves propagate with a conical angle θ between the wave vectors \mathbf{k}_ω and $\mathbf{k}_{2\omega}$ defined by the momentum conservation law, which forces the particular angular dispersion $\cos \theta = 2k_\omega/k_{2\omega} = n^o(\omega)/n^{o,e}(2\omega)$, where $n^{o,e}(\omega)$ is the ordinary/extraordinary refractive index of the material at a frequency ω .¹³ Accordingly, our pattern corresponds to the far field of the beams generated inside the crystal which are distributed onto a circular ring for a particular color. The SHG pattern also shows a hexagonal distribution of more intense and better defined regions which are related to the nonlinear diffraction produced by the hexagonal symmetry of the ferroelectric domain walls.¹⁸

The generated pattern exhibits two following main features: (i) the coexistence of two SHG rings associated with two types of SHG processes $[e, oo]$ and $[o, oo]$ due to the birefringence of LiNbO_3 ¹⁸ and (ii) a marked azimuthal dependence of the SHG intensity which depends on the type of SHG process, $[e, oo]$ and $[o, oo]$. For the fundamental wavelength used in this work ($\lambda_\omega = 1200 \text{ nm}$), the external conical angles for the extraordinary and ordinary waves are $\theta_{\text{ext}} = 16.4^\circ$ and $\theta_{\text{ord}} = 17.6^\circ$, respectively. Considering the $3m$ point group symmetry of LiNbO_3 , the azimuthal intensity profiles can be written in terms of the nonlinear effective coefficients $d_{\text{eff}}^{\text{ord}}$ and $d_{\text{eff}}^{\text{ext}}$ as follows:¹⁷

$$I_{\text{SHG}}^{\text{ord}} \approx (d_{\text{eff}}^{\text{ord}})^2 = [d_{22} \cos(\varphi + 2\gamma)]^2,$$

$$I_{\text{SHG}}^{\text{ext}} \approx (d_{\text{eff}}^{\text{ext}})^2 = [d_{31} \sin \theta + d_{22} \sin(\varphi + 2\gamma) \cos \theta]^2, \quad (1)$$

where $d_{22} = d_{yxx} = -d_{yyy}$ and $d_{31} = d_{zxx}$ are the relevant nonlinear coefficients, φ the azimuthal angle measured counter-clockwise from the x axis and γ the polarization angle of the linearly polarized fundamental incident beam measured from the x axis ($\gamma = 0$ for polarization along the x axis). The results of the simulation for each process, as well as the composition of both rings, are shown in Fig. 1(c) for $\gamma = 0$. The far field pattern obtained for the ordinary process shows a twofold symmetry axis, while the extraordinary one lacks of axial symmetry. As observed, the intensity of the experimental SHG nonlinear pattern is modulated by the azimuthal dependence of d_{eff} . In LiNbO_3 the value of d_{31} is approximately twice larger than that of d_{22} ,¹⁹ thus, according to Eq. (1) the intensity of the extraordinary SHG beam dominates the pattern.

To analyze the effect that the sense of the fundamental beam along the polar direction produces on the SHG pattern, two independent symmetry operations, $C_2(x)$ and $C_2(y)$, have

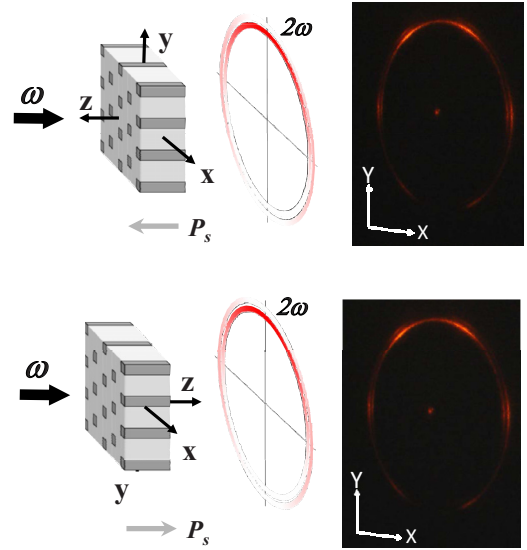


FIG. 2. (Color online) Effect of a $C_2(x)$ rotation of the nonlinear crystal on the far field SHG patterns. The result is consistent with the effect of the electrical poling along the z axis which lead the patterns unchanged. The dominant orientation of \mathbf{P}_s is marked.

been considered. In Fig. 2 we compare the SHG patterns produced by two different linearly polarized fundamental beams ($\gamma = 0$) propagating with opposite directions along the z axis as a result of a $C_2(x)$ operation on the crystal. Upon this rotation the SHG far field images are identical, regardless the direction $+z$ and $-z$ of the fundamental incident beam. The result is well reproduced by Eq. (1) taking into account that a $C_2(x)$ rotation on the nonlinear coefficient matrix of LiNbO_3 produces a change of sign on d_{22} and d_{31} . In fact, the effect of a $C_2(x)$ rotation is equivalent to the effect of the electrical poling along the z axis by applying an electric field. In LiNbO_3 reversing \mathbf{P}_s by applying an electric field along the z axis produces not only a reversion of the cationic chain along the polar axis but also a reversion in the chain of cations along the y axis.²⁰ Accordingly, the polarization inversion can be seen as a 180° rotation of the crystal structure around the x crystallographic axis, $C_2(x)$, which maintains the crystal structure unchanged. The results observed in Fig. 2 show that is not possible to distinguish the SHG patterns produced by two different counterpropagating fundamental beams when the nonlinear interaction experiences the physical changes on both z and y axis after the $C_2(x)$ rotation, which geometrically connect the two real polarization states $\mathbf{P}_s \uparrow$ and $\mathbf{P}_s \downarrow$ in the ferroelectric crystal.

However, by using a $C_2(y)$ rotation we demonstrate that SHG pattern is sensitive to the propagation sense of the fundamental beam along the z axis. Figure 3 shows the two far field SHG images obtained for two linearly polarized beams (again $\gamma = 0$) propagating with opposite sense after a $C_2(y)$ rotation of the crystal. In this case the SHG pattern differs intriguingly for light propagating in opposite directions: the orientation of the pattern changes from “up” to “down” when the fundamental beam propagation is changed from $-z$ to $+z$. The result is reproduced by Eq. (1) taking into account the twofold rotation around the y axis, $C_2(y)$, for which d_{31} changes its sign, while d_{22} remains unchanged. This operation changes the SHG pattern from “up” to “down” leading to a disparate nonlinear optical response for fundamental counterpropagating beams along the polar axis. We note that

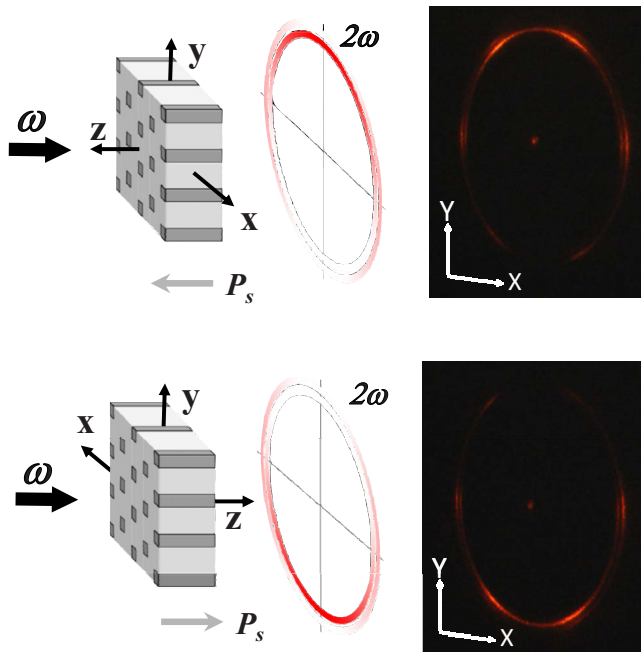


FIG. 3. (Color online) Effect of a $C_2(y)$ rotation of the nonlinear crystal on the SHG far field patterns. The SHG pattern is reversed from “up” to “down” when the fundamental propagation direction is changed from $-z$ to $+z$.

the 180° rotation around the y axis does not lead to the physical polarization inversion under electrical poling along the z axis, since it only changes the sign of the z axis, leaving unchanged the sign of y . (Let us recall that the sign of x is not physically relevant since the chain of cations along this axis is the same for $+x$ and $-x$). Therefore, the results are analog to what would happen in the crystal if solely the chain of cations along the z axis would be changed after inverting the polarization.

The effect of the polarization of the fundamental beam on the directionality of the SHG patterns is shown in Fig. 4. As derived from Eq. (1), the azimuthal intensity profile of the SHG waves depends on twice the polarization angle of input light, 2γ . Hence, for fundamental beams linearly polarized along x and y directions, ($\gamma=0$ and $\gamma=\pi/2$, respectively) it is possible to generate “complementary” SHG patterns [Figs. 4(a) and 4(b)]. Consequently, the interaction of a circularly polarized fundamental beam with the crystal leads to nonlinear diffraction patterns with two perpendicular symmetry planes [Fig. 4(c)]. Therefore, it is possible to switch from unequivalent to equivalent nonlinear optical responses by changing the polarization state of the fundamental beam.

In conclusion, quadratic SHG is revealed in this work as phenomenon with unidirectional character, as those many others inherent to the sense of \mathbf{P}_s in ferroelectric crystals on which are based the powerful applications of these systems. The work has been focused on LiNbO_3 but the results are applicable to other ferroelectric systems and may be used to expand the multifunctional character of 2D-NLPC (as nonlinear optical deflectors or self-frequency doubling lasers) when placed into optical cavities. Additionally, the disparate optical behavior is extensible to a large variety of quadratic three-wave mixing processes. Further, the results obtained on the directionality of the SHG response shows the potential of ferroelectric patterning as a valuable tool to generate original

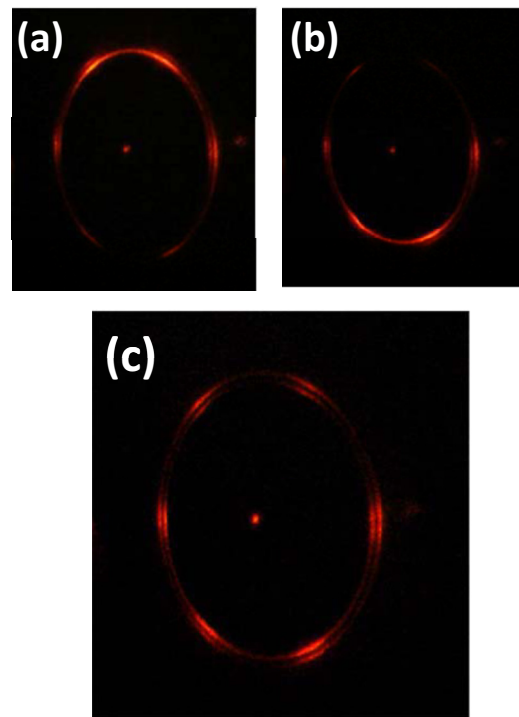


FIG. 4. (Color online) Far field patterns obtained for linearly polarized fundamental beams with polarization states (a) parallel to x axis, $\gamma=0$ and (b) parallel y axis, $\gamma=\pi/2$. (c) Far field pattern obtained for a circularly polarized fundamental beam.

spatially selective light-matter interaction processes.

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