Nonlinear optical transformation of the polarization state of circularly polarized light with holographic-cut cubic crystals

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ABSTRACT

We show theoretically and prove experimentally that with holographic-cut cubic crystals circularly polarized light can also be nonlinearly modified. Although the efficiency of the process is lower it is comparable with the efficiency of cross-polarized wave generation with linearly polarized input light.

Keywords: Femtosecond pulses; Optical nonlinearities; Circular polarization; Cross-polarized wave generation

1. INTRODUCTION

It is well known that circularly-polarized (CP) light cannot change its polarization state nonlinearly when propagating through any isotropic nonlinear media no matter how big the nonlinearity and the intensity are. The same is valid for *z*-cut cubic crystals.[1] Polarization change is an important issue to know when high-intensity laser projects are designed. Propagation of intense laser beams trough materials and the nonlinear optical transformation of the pulse parameters is an issue of great importance for many applications. While in some cases nonlinear change of the pulse properties is a drawback there are applications where such change is essential. In all cases the knowledge of the light-matter interaction at high intensities is valuable.

At sufficiently high light intensities it is possible to reach the threshold levels of various nonlinear effects, e.g. critical self-focusing, beam filamentation, etc. For most of these effects the polarization state of the incident light is crucial. Generally, the threshold is higher when the light is circularly polarized. Some researchers have systematically investigated the influence of the input polarization state on some of these effects. It was even shown that some nonlinear phenomena are impossible if CP light is used. For example, it was shown that CP input beams cannot undergo multiple filamentation provided that they are perfectly cylindrically symmetric.[2]

The threshold of multi-photon ionization and multi-photon excitation was found to be highest for CP and lowest for linearly polarized (LP) light.[3]

CP could be advantageous for pulse compression techniques in gases. By using CP the incident energy in the gas cell can be increased by more than 1.5 times due to the reduce in both the Kerr nonlinearity and the ionization rate of the gas at equivalent intensities. This allows for obtaining shorter compressed pulses through filamentation in gases or using gas-filled hollow fibers.[4,5]

Studies of white-light generation showed that linearly polarized light generates white light with higher intensity than circularly polarized light.[6] CP being the least efficient light polarization state for super-continuum generation may thus be used for suppression of such a generation in case it is an unwanted effect.

Another important nonlinear optical effect is the self-focusing (SF) of the beam. It is well known that for linearly isotropic media the nonlinear change in the index of refraction is minimum for circularly polarized incident light compared with any other polarization state (see e.g. Ref. 7). At a given intensity the SF is less pronounced for this kind of polarization. The critical power for self-trapping of the beam is also minimal for CP light. Since optical bulk damage of transparent materials by ultrashort optical pulses is primarily connected with self-focusing then optical damage

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threshold is higher for CP light. So femtosecond pulses with higher energy could be transmitted through optical elements by using CP.

As we see, circular polarization is an important polarization state for many applications. Therefore the knowledge of possible preservation or modification of this polarization state is of great importance.

In the recent years we investigate the process of cross-polarized wave (XPW) generation which is a nonlinear modification of input polarization (frequency degenerate polarization non-degenerate four-wave mixing process).[8] XPW generation became a key effect for temporal and spatial contrast improvement devices in the field of femtosecond laser science and technology. Systematic investigations of this process have been conducted mainly using cubic crystals with linearly and elliptically polarized input beam. In the most cases the crystals are oriented along their optical axis (z-cut). CP is an eigen polarization for *z*-cut cubic crystals and does not change neither linearly nor nonlinearly. Recently we proposed different orientation of the cubic crystals, holographic-cut ([110] or equivalent cut) orientation, which gives many advantages over *z*-cut.[8,9] The properties of holographic-cut orientation were investigated only for linearly polarized input. We, however, derived theoretically and prove experimentally that circular polarization state can be modified efficiently with holographic-cut cubic crystals. In the following sections we present the obtained results.

2. THEORETICAL PART

2.1. Numerical model

Our analysis is based on the full system of differential equations published in Ref. 8 that describe the process of cross-polarized wave generation in plane-wave approximation:

$$\frac{\mathrm{d}A(\zeta)}{\mathrm{d}\zeta} = i\gamma_1 AAA^* + i\gamma_2 AAB^* + 2i\gamma_2 ABA^* + 2i\gamma_3 ABB^* + i\gamma_3 BBA^* + i\gamma_4 BBB^*, \quad (1a)$$

$$\frac{\mathrm{d}B(\zeta)}{\mathrm{d}\zeta} = i\gamma_5 BBB^* + i\gamma_4 BBA^* + 2i\gamma_4 ABB^* + 2i\gamma_3 ABA^* + i\gamma_3 AAB^* + i\gamma_2 AAA^*. \quad (1b)$$

In the above equations A denotes the complex amplitude of the fundamental (or pump) wave and B – the complex amplitude of the generated orthogonally polarized wave. ζ is the longitudinal coordinate in the light propagation direction. Nonlinear coupling coefficients γ_1 and γ_5 are responsible for the self-phase modulation (SPM) while γ_2 and γ_4 govern the XPW conversion process from A to B and from B to A waves through the last terms in (1b) and (1a), respectively. γ_3 contributes to the cross-phase modulation (XPM). Nonlinear coupling coefficients depend on the orientation of the nonlinear medium and on the components of the $\chi^{(3)}$-tensor.[8] The Eqs. (1) apply for linearly non-birefringent media and hereafter we restrict our considerations to nonlinear crystals (NLC) that belong to *m*3*m* cubic symmetry group. Assuming that the photon energy is below the half band-gap so that the two-photon absorption could be neglected then the $\chi^{(3)}$-tensor is purely real tensor with only two independent components for crystals of *m*3*m* cubic symmetry:[10] $\chi^{(3)}_{xxxx}$ and $\chi^{(3)}_{xxyy}$. The relation between them is usually defined[10] as $\sigma = \left(\chi^{(3)}_{xxxx} - 3\chi^{(3)}_{xxyy}\right)/\chi^{(3)}_{xxxx}$ and is referred to as the anisotropy of the $\chi^{(3)}$-tensor. Nonlinear coupling coefficients for *z*- and holographic-cut *m*3*m* cubic crystals are summarized in Table 1 where the angle β is the angle between the linear input polarization and the *x*-axis of the crystal.

	z-cut	holographic-cut
γ_1	$\gamma_0[1-(\sigma/2)\sin^2(2\beta)]$	$\gamma_0[1-(\sigma/4)\cos(2\beta)+(3\sigma/16)\cos(4\beta)-7\sigma/16]$
γ_2	$-\gamma_0(\sigma/4)\sin(4\beta)$	$\gamma_0[(\sigma/8)\sin(2\beta)-(3\sigma/16)\sin(4\beta)]$
γ_3	$\gamma_0[1/3-(\sigma/4)\cos(4\beta)-\sigma/12]$	$\gamma_0[1/3-(3\sigma/16)\cos(4\beta)-7\sigma/48]$
γ_4	$\gamma_0(\sigma/4)\sin(4\beta)$	$\gamma_0[(\sigma/8)\sin(2\beta)+(3\sigma/16)\sin(4\beta)]$
γ_5	$\gamma_0[1-(\sigma/2)\sin^2(2\beta)]$	$\gamma_0[1+(\sigma/4)\cos(2\beta)+(3\sigma/16)\cos(4\beta)-7\sigma/16]$

Table 1. Nonlinear coupling coefficients for *z*-cut and holographic-cut *m*3*m* cubic crystals. $\gamma_0 = 6\pi\chi^{(3)}_{xxxx}/8n\lambda$

Notice that generally SPM and XPW generation are not the same for the A and B waves except in the special case of *z*-cut orientation. As a result of these non-equalities circular polarization will also be nonlinearly modified with holographic-cut crystals.

We solve numerically the system of equations (1) assuming an optical scheme shown in Fig. 1 and with initial

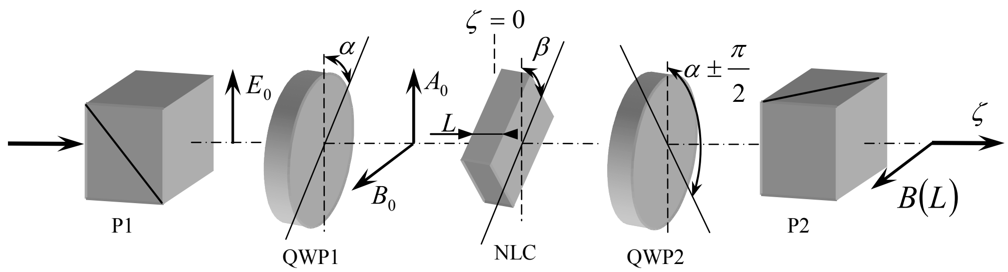

Figure 1. Optical scheme used in the model. P1, P2 – polarizers, QWP – quarter-wave plates, NLC – nonlinear crystal

conditions: $A_0 = |E_0|/\sqrt{2}$ and $B_0 = \pm i|E_0|/\sqrt{2}$ for circular polarization ($\alpha = 45°$); and $A_0 = E_0$ and $B_0 = 0$ for linear polarization ($\alpha = 0°$).

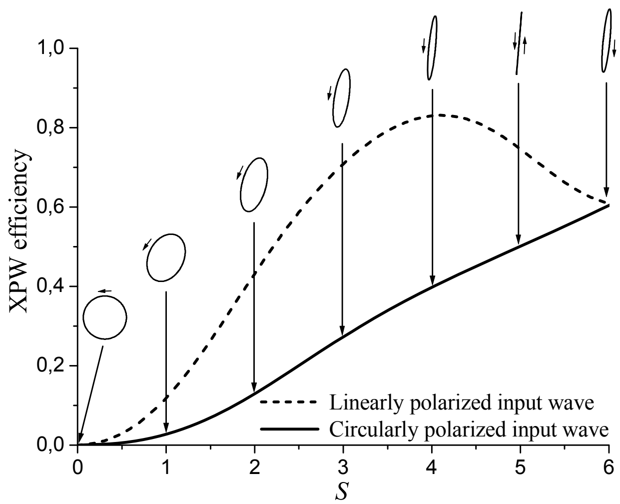

Figure 2. Simulation results for plane input wave. Solid curve – the efficiency obtained with circularly polarized input light. Intensity dependent polarization-state changes after the NLC are also depicted for the corresponding *S* values. Dashed curve – the efficiency obtained with linearly polarized input at optimal angle $\beta = 64.5°$

2.2. Numerical results

Simulation results for plane input wave are given in Fig. 2. XPW efficiency is defined as $|B(L)^2|/|E_0|^2$ (see Fig. 1). The dimensionless parameter S is proportional to the product: nonlinearity × input intensity × nonlinear crystal length. In practice S is limited to approximately 5 by the self-action effects.[11] In calculations $\sigma = -1.2$, a value that corresponds to the anisotropy of the BaF_2 is used.[12] Polarization state changes are also drawn in Fig. 2. It is seen that initially circular polarization transforms first into elliptical polarization. The orientation of the axes of the ellipse depends on the orientation of the crystalline axes. With further increasing S the polarization may become purely linear. After that the direction of rotation of the electric-field vector is changed and at sufficiently high S circular polarization with other handedness could be obtained (i.e. left-handed circular polarization can be nonlinearly transformed into right-handed one and vice-versa).

On the same Fig. 2 for comparison the efficiency of XPW generation with purely linear polarization is also given. Although the efficiency is lower for CP wave it is of the same order as for the linearly polarized input wave. Contrast-improvement devices based on holographic cut cubic crystals that use CP input beam can also be constructed.

3. EXPERIMENTS

3.1. Experimental setup

In our experiments we used a Ti:sapphire chirped-pulse amplifier laser system, based on a commercial 1 kHz, carrier-envelope phase-stabilized front-end amplifier from Femtolasers GmbH (Femtopower Compact Pro) followed by a homemade multipass amplifier. To obtain high-fidelity compressed pulses, a feedback loop (DazScope) is applied to an acousto-optic programmable dispersive filter (AOPDF) inside the laser front end for spectral phase measurement and compensation throughout the entire system. The typical pulse duration after optimization is approximately 25 fs.

The experimental setup is similar to that shown in Fig. 1. A part of the whole available laser power was steered to the experimental setup by using an uncoated wedge. This part of the beam was then focused onto the NLC sample by a 50-cm focal-length lens. For controllable attenuation of the incident pulse energy a reflective type filter wheel was placed before the lens. The output signal was collected with a short focal-length lens on a photodiode and was monitored on the screen of a digital oscilloscope. Another photodiode was used to monitor the energy of the input pulses. Absolute energy values were measured with an "Ophir" energy meter.

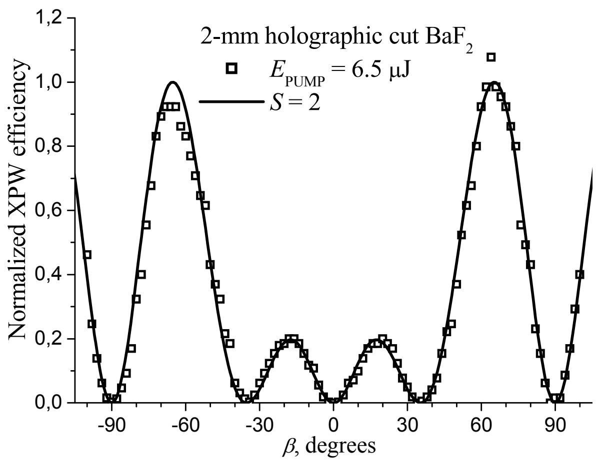

Figure 3. Experimental β-dependence of the XPW generation efficiency for linearly polarized input (symbols). The XPW efficiency is normalized to the average of the two highest maxima. Solid curve is the theoretical fit obtained assuming Gaussian spatial and temporal profiles of the input beam

During the experiments the laser pulse broadening in the air and in the other optical elements was compensated through setting proper parameters of the AOPDF. The criterion for good compensation was to find the maximum of the XPW signal.

We used holographic-cut BaF_2 crystals with different thicknesses: 1, 2 and 6 mm. The behavior of the crystals is similar but here we present results for 2-mm sample. For obtaining good efficiency of the nonlinear process the 1-mm crystal requires higher pump energy close to the surface damage threshold. On the contrary, the 6-mm sample requires less energy but in this case the critical self-focusing and the threshold of continuum generation are very easily reached.

Experimental β-dependence of the XPW generation with linearly polarized input pulses is shown in Fig. 3 for the 2-mm crystal. The experimental curve well corresponds to the theoretical curve for holographic-cut crystal. Similar curves were obtained with the other crystals (1 and 6-mm).

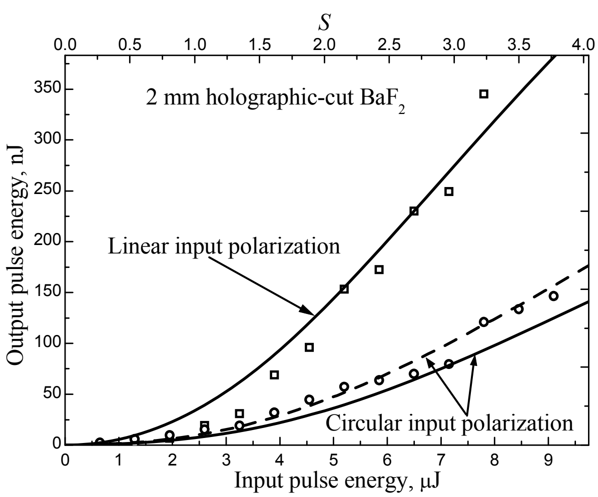

Figure 4. Experimental energy dependence: squares – with linearly polarized input beam at optimum angle $\beta = 64.5°$; circles with circularly polarized input beam. Curves are theoretical dependencies. The dashed theoretical curve is obtained for non-perfect circular polarization by using $\lambda/4 \pm \lambda/100$ phase plates. S-values for the theoretical dependencies are given on the top scale

3.2. Power dependence

Experimental dependences of the output pulse energy on the input pulse energy for both linear and circular input polarization are shown in Fig. 4. Experimental output pulse energies are not corrected for the linear losses that are due to reflection from the uncoated surfaces of the crystal and the output Glan. Theoretical curves in Fig. 4 are obtained assuming Gaussian spatial and temporal profiles of the input pulses by numerically solving the system (1) with initial conditions $E_0(r,t) \propto \exp(-r^2 - t^2)$ followed by numerical integration over r and t. We see a good qualitative agreement between theoretical curves and experimental data. The differences between them are due to propagation effects not included in the model. Such effects might be, for example, change in the beam size due to self-focusing, self-diffraction, residual and induced chirp, and multiphoton absorption. Contributions of higher-order nonlinearities are also possible.[13]

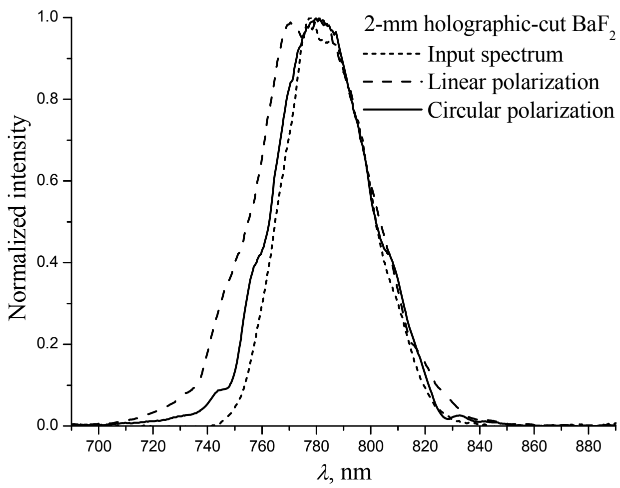

Figure 5. Spectrum of the output XPW signal for linearly and circularly polarized input beams. Dotted curve is the input beam spectrum

It can be seen from Fig. 4 that the XPW generation is more efficient with linearly polarized pump as predicted by the theory. Although the efficiency with CP is less it is of the same order of magnitude and we believe that it may find practical implementation.

Same experiments were conducted with other holographic-cut cubic crystals: CaF_2, SrF_2, as well as with new mixed crystals $Ca_xSr_{1-x}F_2$. They showed the same behavior only rescaled for the different values of the nonlinearity $\chi^{(3)}$ and anisotropy σ of the crystals.

3.3. Spectra

It is well known that the generated XPW pulses are shortened and spectrally broadened.[14] Measured spectra of the input pulses as well as of the output pulses with linear and circular input polarization are presented in Fig. 5. The spectra were taken with a spectrometer "Avaspec." All spectra were measured at equal input pulse energy 4 μJ, a value well bellow the threshold of critical self-focusing and continuum generation.

Both output spectra are broader than the spectrum of the input pulses which is an indication for pulse shortening. It is seen that spectral broadening is stronger with linearly polarized pump. This can be explained with the weaker SPM experienced by the circularly polarized light.[5]

4. CONCLUSION

In conclusion, we show theoretically and prove experimentally that with holographic-cut cubic crystals circularly polarized light can also be nonlinearly modified. As far as we know, it is the first demonstration of nonlinear modification of circularly polarized light in cubic crystals. The efficiency of the process is lower compared with the efficiency of cross-polarized wave generation with linearly polarized input light but is of the same order of magnitude. The demonstrated effect could be successfully used in practical devices in the field of femtosecond optics.

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REFERENCES

[1] Hutchings, D. C. and Werrett, B. S., "Theory of the anisotropy of ultrafast nonlinear refraction in zinc-blende semiconductors," Phys. Rev. B 52(11), 8150-8159 (1995).

[2] Fibich, G.. and Ilan, B., "Self-focusing of circularly polarized beams," Phys. Rev. E 67(3), 036622 (2003).

[3] Petit, S., Talebpour, A., Proulx, A., and Chin, S. L., "Polarization dependence of the propagation of intense laser pulses in air," Opt. Commun. 175, 323-327 (2000).

[4] Liu, J., Chen, X. W., Li, R. X., and Kobayashi, T., "Polarization-dependent pulse compression in an argon-filled cell through filamentation," Laser Phys. Lett. 5(1), 45-47 (2008).

[5] Chen, X., Jullien, A., Malvache, A., Canova, L., Borot, A., Trisorio, A., Durfee, C. G.., and Lopez-Martens, R., "Generation of 4.3 fs, 1 mJ laser pulses via compression of circularly polarized pulses in a gas-filled hollow-core fiber," Opt. Lett. 34(10), 1588-1590 (2009).

[6] Dharmadhikari, A. K., Rajgara, F. A., and Mathur, D., "Systematic study of highly efficient white light generation in transparent materials using intense femtosecond laser pulses," Appl. Phys. B 80, 61-66 (2005).

[7] Boyd, R. W., [Nonlinear Optics], Second ed., Academic Press (2003).

[8] Kourtev, S., Minkovski, N., Canova, L., Jullien, A., Albert, O., and Saltiel, S. M., "Improved nonlinear cross-polarized wave generation in cubic crystals by optimization of the crystal orientation," J. Opt. Soc. Am. B 26(7), 1269-1275 (2009).

[9] Canova, L., Kourtev, S., Minkovski, N., Jullien, A., Lopez-Martens, R., Albert, O., and Saltiel, S. M., "Efficient generation of cross-polarized femtosecond pulses in cubic crystals with holographic cut orientation," Appl. Phys. Lett. 92(23), 231102 (2008).

[10] Petrocelli, G.., Pichini, E., Scudieri, F., and Martellucci, S., "Anisotropic effects in the third-harmonic-generation process in cubic crystals," J. Opt. Soc. Am. B 10(5), 918-923 (1993).

[11] Jullien, A., Albert, O., Chériaux, G., Etchepare, J., Kourtev, S., Minkovski, N., and Saltiel, S. M., "A two crystal arrangement to fight efficiency saturation in cross-polarized wave generation," Opt. Express 14(7), 2760-2769 (2006).

[12] Dabbicco, M., Fox, A. M., von Plessen, G., and Ryan, J. F., "Role of $\chi^{(3)}$ anisotropy in the generation of squeezed light in semiconductors," Phys. Rev. B 53(8), 4479-4487 (1996).

[13] Gordienko, V. M., Mikheev, P. M., and Syrtsov, V. S., "Nonlinear rotation of the polarization of the intense femtosecond laser radiation in BaF_2," Bull. Russ. Acad. Sci.: Phys. 71(1), 122-125 (2007).

[14] Jullien, A., Canova, L., Albert, O., Boschetto, D., Antonucci, L., Cha, Y.-H., Rousseau, J., Chaudet, P., Chériaux, G., Etchepare, J., Kourtev, S., Minkovski, N., and Saltiel, S. M., "Spectral broadening and pulse duration reduction during cross-polarized wave generation: influence of the quadratic spectral phase," Appl. Phys. B 87, 595-601 (2007).