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# 1 In-situ holograms and two-wave mixing 2 amplification of conical diffraction vector waves

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10 **Abstract:** Holographic recording and selective reconstruction and amplification of conical  
11 diffraction vector waves is demonstrated using a nonlinear photorefractive  $\text{Sn}_2\text{P}_2\text{S}_6$  crystal  
12 acting simultaneously as medium producing the conical diffraction effect and as holographic  
13 storage material. It is shown that upon propagation of the object wave along one of the optical  
14 axes of the biaxial crystal, the azimuthal spreading of the local linear polarizations across the  
15 conical diffraction ring allows its holographic recording with any combinations of the object  
16 and reference wave's input polarizations, including mutually orthogonal ones. We discuss the  
17 dependence of the recording and read-out polarizations on the recovered and amplified patterns  
18 and provide a simplified model qualitatively accounting for all the main observed features. The  
19 approach permits a tailoring of the reconstructed or amplified vector waves in terms of intensity  
20 and polarization distributions and opens interesting perspectives for their static or dynamic  
21 manipulation.

## 22 1. Introduction

23 Internal conical diffraction (CD) is a natural optical effect that produces specific types of  
24 vector beams having a spatially-dependent polarization. This phenomenon is observable upon  
25 propagation of a focused beam along one of the optic axes of an optically biaxial crystal (BC)  
26 and gives rise to a slanted cone with circular base [1–3]. The latter represents the locus of the  
27 Poynting vectors associated to the wavevector propagation along the singular direction of the  
28 optic axis. Even for a homogeneously polarized input wave, different linear polarizations are  
29 found to spread azimuthally across the ring section of the cone. Despite a nearly 200 years  
30 history since Hamilton's prediction [4], this phenomenon has seen a renewed interest in the last  
31 two decades. This has been enabled by a deeper theoretical understanding owing to a paraxial  
32 diffraction theory describing the CD vector waves [1, 5] and is motivated by both fundamental  
33 and applied aspects. In this context we may cite the interesting orbital angular momentum  
34 characteristics [6–10] and the new features allowed by novel configurations with a series of BC in  
35 cascade [11–15] that open new venues for the complex shaping of these vector beams, up to the  
36 case where the CD beams no longer possess the usual circular symmetry [16, 17]. Potential and  
37 recently demonstrated applications of CD include optical trapping of particles or Bose-Einstein  
38 condensates [18–20], free-space multiplexing for communication [21], beam shaping [15, 16, 22],  
39 polarimetry [9, 23, 24] or super-resolution imaging [25, 26].

40 Conical diffraction is a manifestation of the linear optical properties of the crystal and the  
41 above mentioned investigations have all been performed in the linear regime with the BC acting as  
42 passive elements. Nevertheless, few studies have combined CD with nonlinear optics, where CD  
43 at the second-harmonic wavelength [27–29] or at the sum-frequency generation wavelength [30]  
44 have been studied. Properly doped BC of  $\text{KGd}(\text{WO}_4)_2$  or  $\text{KY}(\text{WO}_4)_2$  have also been employed as

45 active elements within a cavity for realizing conical diffraction lasers with specific polarization  
46 properties [31–35].

47 Some of the biaxial crystals giving rise to CD also show another nonlinear optical effect,  
48 the photorefractive effect [36]. This is an indirect effect leading to refractive index changes  
49 already upon low power light illumination. In this work we combine conical diffraction with  
50 photorefraction within the same medium to demonstrate conical diffraction holograms as well as  
51 the selective amplification of the CD wave via dynamic two-wave mixing. Section 2 describes  
52 briefly the experimental approach and the  $\text{Sn}_2\text{P}_2\text{S}_6$  (SPS) crystal used both as medium giving  
53 rise to CD and as nonlinear holographic recording material. Section 3 illustrates the major  
54 experimental results showing that propagation of the object wave along one of the optical axes of  
55 the BC is crucial and critical. Indeed, provided that the object wave propagates along the optical  
56 axis and gives rise to CD, holographic recording and two-wave mixing amplification can be  
57 efficiently achieved with any combination of the polarizations of the object and reference waves,  
58 including mutually orthogonal ones. The underlying physical phenomenon is highly complex and  
59 involves the physics of the photorefractive effect in strongly anisotropic configurations as well as  
60 the constraints associated to the propagation of light in singular directions within the BC. While  
61 a complete theory would require the knowledge of several still unknown material parameters, it  
62 is possible to qualitatively catch the main experimental features with a simplified model which is  
63 presented and discussed in Sect. 4. Finally, Sect. 5 gives brief remarks and conclusions.

## 64 2. Crystal sample and experimental approach

65 The BC used for the investigations is a nominally undoped ferroelectric SPS, which belongs to the  
66 monoclinic point group  $m$  at room temperature. SPS is an attractive nonlinear optical material  
67 owing to its transparency range extending from 530 to 8000 nm wavelength, its interesting  
68 electro-optical [37, 38] and nonlinear optical properties [39] and the high photorefractive  
69 sensitivity [40, 41]. The anisotropic optical properties and their dispersion were studied in detail  
70 in [42] and optical activity near the ferroelectric-paraelectric phase transition was evidenced  
71 in [43]. A summary of some of the most relevant material parameters is given in Ref. [41]. A  
72 11.2 mm-long SPS sample was cut from a crystal grown by the vapor transport method [44] and  
73 electrically polarized to insure a ferroelectric monodomain state. The sample was oriented with  
74 the parallel beam entrance and exit faces being nearly perpendicular to one of the two optic axes.

75 A simplified scheme of the experimental arrangement for holographic recording and two-wave  
76 mixing is shown in Fig. 1. Figure 1(a) is for the holographic recording and read-out of the CD  
77 beam created in-situ by the object wave. A He-Ne laser beam at the wavelength of 632.8 nm  
78 is split into a reference and an object wave by taking care that the path lengths difference is  
79 within the coherence length of the laser. Their polarizations are initially left and right circular,  
80 respectively. Before each wave reaches the SPS crystal, the polarizations can be individually  
81 modified by polarization controllers (Pol-contr) in each path being composed of linear polarizers,  
82 wave-plates or adjustable liquid crystal retarders. The Gaussian input waves have initially an  
83 equal diameter and are focused by the spherical lenses  $L_1$  (focal length  $f = 200$  mm) and  $L_2$   
84 ( $f = 75$  mm) towards the SPS crystal, where they interfere. The positions of the lenses are chosen  
85 in such a way that the reference wave has a slightly larger diameter than the object wave inside  
86 the crystal ( $\approx 2.1$  mm vs. 1.5 mm). The focal plane of lens  $L_2$  corresponds to the focal image  
87 plane where the CD ring pattern formed by the object wave is the sharpest. The spherical lens  
88  $L_3$  images this plane onto a CCD camera to observe the transmitted, amplified or reconstructed  
89 CD object wave with the corresponding power being detected in parallel by a photodiode. For  
90 holographic recording (but not for two-wave mixing) the input powers of the two beams are  
91 adjusted in such a way that the reference and object waves have nearly the same intensity inside  
92 the SPS crystal ( $I \approx 30$  mW/cm<sup>2</sup>). For holographic recording (Fig. 1(a)) shutter  $S_1$  on the object  
93 wave is kept open during 90 seconds. This allows to record a photorefractive grating composed

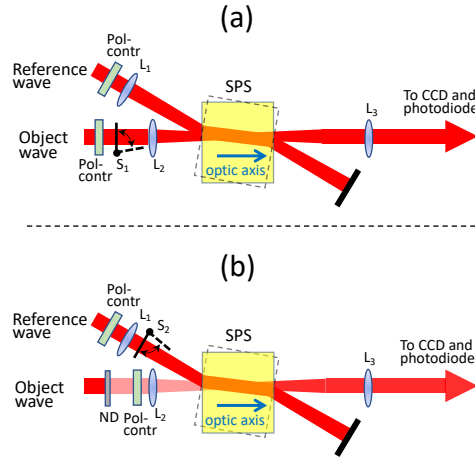


Fig. 1. Scheme of experimental arrangement for the holographic recording and read-out of the CD object wave (a) and for its amplification by two-wave mixing (b). SPS:  $\text{Sn}_2\text{P}_2\text{S}_6$  biaxial crystal;  $S_1$ ,  $S_2$ : shutters;  $L_1$ ,  $L_2$ ,  $L_3$ : spherical lenses; Pol-contr.: polarization controller; ND: neutral density filter. The optic axis of the SPS crystal is aligned with the object wave (yellow shape). The dashed shape give an alternative (misaligned) crystal orientation.

94 of a fast primary grating nearly fully compensated by a slow secondary grating, this behavior is  
 95 typical for type I nominally undoped SPS crystals [41]. When the shutter  $S_1$  is being closed, the  
 96 primary grating disappears quickly revealing the secondary grating that results in the diffraction  
 97 of the reference wave into the reconstructed object wave. For the examples presented in the next  
 98 section this reconstructed wave is observed 5 seconds after closing shutter  $S_1$ .

99 The situation for two-wave mixing (Fig. 1(b)) is similar, however with different boundary  
 100 conditions. Here the object wave is first attenuated by a neutral density filter (ND) to an intensity  
 101 less than  $1/100$  (inside the BC) with respect to the one of the reference. In this case, injection of  
 102 the reference wave by opening shutter  $S_2$  leads to an amplification (or eventual depletion) of the  
 103 CD object wave. Importantly, we recall that the object wave produces the CD vector beams only  
 104 if it propagates along the optic axis (yellow orientation of SPS in Fig. 1). By misaligning the  
 105 crystal (dashed orientation) CD no longer occurs and the behavior differs strongly, as discussed  
 106 in the next section. In our experiments the horizontal laboratory plane is spanned by the optic  
 107 axis of the crystal and by the  $x$  crystallo-physical axis of the orthogonal Cartesian system used to  
 108 describe the tensorial properties of SPS. The convention taken is the same one as in Ref. [42]  
 109 (see Fig. 1 therein), the  $x$ -axis is close to the crystallographic  $a$ -axis of the monoclinic SPS and  
 110 to the direction of the spontaneous polarization. The examples given below are obtained for  
 111 an external crossing angle of  $31^\circ$  between the two waves, what corresponds to a spatial grating  
 112 period of  $\Lambda \approx 1.2 \mu\text{m}$ , however other crossing angles give qualitatively similar results.

### 113 3. Experiments

114 As mentioned above, propagation along the optical axis leads to spreading of different linear  
 115 polarizations across the CD ring pattern. If the input wave is circularly polarized, all the linear  
 116 polarizations are equally important and the circular ring pattern is homogeneous in intensity.  
 117 The polarization spreading can be easily visualized by filtering the output CD ring with a linear  
 118 polarization analyzer placed before the observation CCD camera. This is shown in Fig. 2 which  
 119 gives the transmitted CD object wave (with no reference wave) for circularly polarized input after

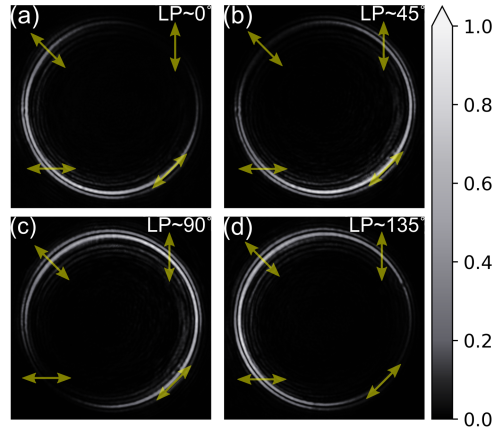


Fig. 2. Polarization-filtered object wave with a linear polarizer (LP) placed before the CCD camera and oriented in four different directions, i.e. (a) 0°, (b) 45°, (c) 90° and (d) 135°. The input object wave is circularly polarized. The nodes in the intensity distribution correspond to the positions where the local polarization in the output CD ring is perpendicular to the LP transmission direction. The arrows give the corresponding local linear polarization directions. The grayscale bar gives the relative intensity scale.

120 filtering with a polarization analyzer (LP) oriented at 0°, 45°, 90° or 135° in the laboratory frame.  
 121 Clearly, upon filtering the local intensity depends on the azimuthal position on the ring because  
 122 the locally transmitted intensity depends quadratically on the projection of the local polarization  
 123 on the analyzer transmission axis. The positions of the nodes in Fig. 2(a)-(d) correspond thus  
 124 to the azimuthal positions where the local polarization is perpendicular to the analyzer axis.  
 125 It can be easily recognized that axially opposite points on the ring are associated to mutually  
 126 orthogonal local polarizations, as generally expected in the case of CD. It is worth noticing that  
 127 using a linearly rather than circularly polarized input, that is placing the linear polarizer before  
 128 rather than after the BC, leads to the same type of crescent-like pattern as in Fig. 2, the full ring  
 129 being obtained only if the input wave is polarized circularly. The above-discussed azimuthal  
 130 polarization spreading is a remarkable property of the CD wave in view of the ability to record  
 131 photorefractive holograms with any combinations of the input beam polarizations. It is indeed  
 132 always possible to find ring sections of the object wave where the local polarization is such as to  
 133 produce strong interference with a uniformly polarized reference wave.

134 We illustrate first this mutual polarization versatility in the case of dynamic two-wave mixing  
 135 (TWM). This is shown in Fig. 3 which represents the temporal build-up of the spatially integrated  
 136 object wave amplification  $\bar{\gamma}_0$  for various polarizations of the reference wave acting as a pump  
 137 and a unique polarization of the object wave. The quantity  $\bar{\gamma}_0$  is defined as the object wave total  
 138 power in presence of the reference pump wave divided by the transmitted object wave power  
 139 without the reference wave. For Fig. 3 the incident object wave is aligned to the optic axis  
 140 of the BC and is kept right circularly polarized (RCP) while the reference wave polarization  
 141 is either left circular (LCP) or linear at +45° or -45°. Clearly, upon injection of the reference  
 142 wave at  $t = 0$  [opening of shutter  $S_2$  in Fig. 1(b)], a significant amplification is observed for  
 143 all polarization combinations, including the case where the two incident waves are mutually  
 144 orthogonal (RCP and LCP). The amount of amplification differs slightly for the three cases as the  
 145 strength of the underlying photorefractive gratings and the electro-optic coupling is a function of  
 146 the wave polarizations. The dynamics seen in Fig. 3 shows the rise of the primary photorefractive  
 147 grating in a time of the order of one second. Here the maximum of  $\bar{\gamma}_0$  does not correspond to a

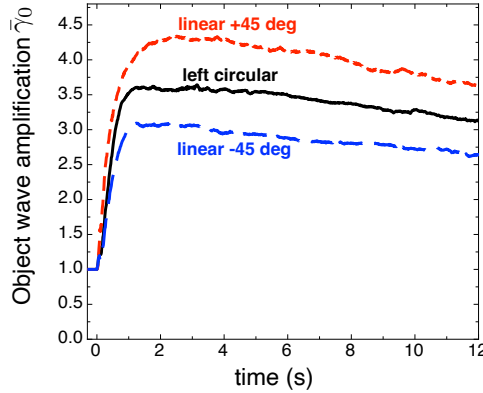


Fig. 3. Two-wave mixing amplification of the object wave as a function of time. The input object wave is aligned to the crystal optic axis and is right circularly polarized. Results for three different polarizations of the incident pumping reference wave are shown: left circular polarization (solid black curve), linear polarization (LP) at  $+45^\circ$  (red dashed curve) and LP at  $-45^\circ$  (blue dashed curve).

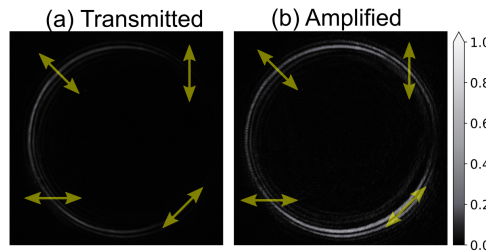


Fig. 4. Two-wave mixing amplification for interfering linearly polarized orthogonal eigenwaves. (a) Transmitted CD object wave in absence of the reference pump wave. (b) Amplified CD object wave. The arrows give the local linear polarization directions at the output and the grayscale bar gives the relative intensity scale.

148 steady-state because, as mentioned above, for type I SPS crystals a secondary grating starts to  
 149 compensate the primary one, resulting in the slight decrease of the amplification seen after the  
 150 maximum. This compensation would be nearly complete after a time of  $\approx 100$  s. It is worth  
 151 noting that the amplification  $\bar{\gamma}_0$  shown in Fig. 3 is an average amplification over the CD ring  
 152 as obtained by integrating all the power in the object wave using a photodiode. In reality the  
 153 amplification differs for different sections of the ring, that correspond to different local linear  
 154 polarizations of the CD wave. This point will be discussed below in connection with Fig. 4.  
 155 Finally, we would like to point out that larger amplifications than those shown in Fig. 3 are  
 156 possible for situations where the two input waves share a same polarization. This is the usual  
 157 condition under which photorefractive two-wave mixing experiments are normally performed. By  
 158 choosing a reference wave polarization corresponding to one of the two eigenpolarizations in SPS  
 159 and the corresponding nearly parallel linear polarization for the input object wave, we have observed  
 160 in separated experiments a maximum amplification  $\bar{\gamma}_0 \approx 10.2$ , as compared to a maximum of  
 161  $\bar{\gamma}_0 \approx 4.3$  in Fig. 3.

162 As mentioned above, the TWM amplification differs locally, what can be easily seen in Fig. 4.  
 163 Here the reference wave polarization is linear and corresponds to one of the eigenpolarizations in  
 164 the SPS crystal. The polarization of the object wave is linear too but is associated to the other  
 165 (orthogonal) eigenpolarization when the crystal is slightly misaligned. Figure 4(a) shows the

166 transmitted object wave for the aligned configuration in absence of the reference beam (shutter  
167  $S_2$  in Fig. 1(b) closed). As expected, the observed CD ring pattern is inhomogeneous in intensity  
168 as a result of the linear input polarization. After amplification (Fig. 4(b), three seconds after  
169 opening shutter  $S_2$ ) one observes in this specific case a much stronger increase of intensity in the  
170 regions on the right side of the CD ring, that were initially weaker. The amplification for the left  
171 side of the ring is significantly less pronounced, what give rise to a kind of power equalization  
172 within the CD ring. Importantly, we have verified that the amplified CD conserves the same  
173 polarization distribution than the linearly transmitted one (see Fig. 4) , meaning that the light  
174 amplified by the nonlinear process respects the constraints given by the linear properties of the  
175 medium. Note also that in the present example the spatially averaged amplification is  $\bar{\gamma}_0 \approx 4.5$ . It  
176 is interesting to compare this value with the one obtained under the same conditions except for the  
177 crystal being turned (misaligned) so that the the object wave no longer propagates along an optic  
178 axis. In this case the orthogonal eigenwaves prevent the creation of an efficient photorefractive  
179 grating. We have checked this case for an internal misalignment of  $\approx 6^\circ$ , finding that the object  
180 wave is essentially unaffected by the pumping with the reference beam, the amplification being  
181 less than 12% ( $\bar{\gamma}_0 \approx 1.12$ ).

182 The same kind of behavior can be observed for grating diffraction at a photorefractive hologram  
183 recorded with orthogonal eigenwaves, as for the case of Fig. 4. Figure 5(a) shows the linear  
184 transmission of the object wave under CD conditions, while Fig. 5(b) shows the reconstructed  
185 CD wave after closing shutter  $S_1$  in Fig. 1(a) at the end of the recording. Clearly, also here a  
186 reconstructed CD vector wave (with same local polarization as the transmitted object wave) is  
187 being observed despite for the orthogonal eigenwaves associated to the object and reference  
188 beams. As for TWM, also here one observes a locally stronger reconstruction on the right side of  
189 the ring, corresponding to a region where the transmitted object wave is weaker. Figure 5(c) and  
190 (d) are for the case where the crystal is misaligned and the object wave makes an internal angle  
191 of about  $6^\circ$  with the optic axis. In this case the object wave experiences double refraction. The  
192 latter can be recognized in Fig. 5(c) that shows its transmission through the SPS crystal. Here the  
193 saturated left spot corresponds to the transmission of the dominant object eigenwave while the  
194 weak spot on the right corresponds to a spurious intensity on the other (orthogonally polarized)  
195 eigenwave . If a hologram is recorded with the reference wave on the orthogonal eigenwave,  
196 upon read-out one observes merely a weak diffraction at the position of the right spot [Fig. 5(d)].  
197 Under the same conditions used for the aligned configuration, the corresponding diffracted power  
198 in direction of the object wave is about 30 times weaker, which confirms the specific importance  
199 of propagating the object wave along the optic axis.

200 The section of the ring which is most strongly reconstructed depends on the input polarization  
201 of the reference wave. This is illustrated with the help of Fig. 6, that shows the results of hologram  
202 recording and read-out with linear horizontal or vertical polarization (in the laboratory frame) for  
203 the reference wave, together with a circular polarized object input wave at recording. Clearly  
204 the horizontal (H-pol) and vertical polarized (V-pol) reference lead to predominant revealing  
205 of different sections of the CD ring at read-out, as seen in Fig. 6(b) and (c), respectively. The  
206 corresponding angular-dependent powers can be best visualized by integrating the intensity across  
207 a narrow radial section across the CD ring for different azimuthal angles along the ring, what gives  
208 the solid red curve (H-pol) and the dotted blue curve (V-pol) in Fig. 6(d). This behavior can be  
209 put in relation with the expectations of a simplified theory, as discussed in the next section.

#### 210 **4. Background theory**

211 The detailed explanation of our above observations in connection with CD holography and  
212 TWM is a tedious task. It involves on one hand the physics related to the anisotropic optical  
213 properties leading to the CD effect and, on the other hand, the complex physics related to the  
214 photorefractive effect [36] in such a low symmetry monoclinic crystal. The latter is concerned by

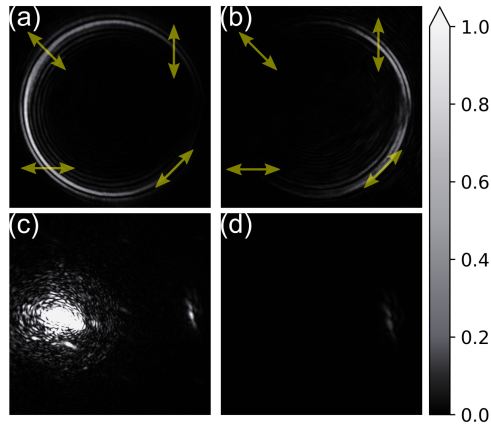


Fig. 5. Holographic recording with orthogonal eigenwaves. Left column: transmitted object wave prior to hologram recording for the aligned (a) and misaligned (c) configurations. Right column: object wave reconstruction at read-out after hologram recording, again for the aligned (b) and misaligned (d) configurations. The arrows in (a) and (b) give the local linear polarization directions for the transmitted and reconstructed object wave, respectively.

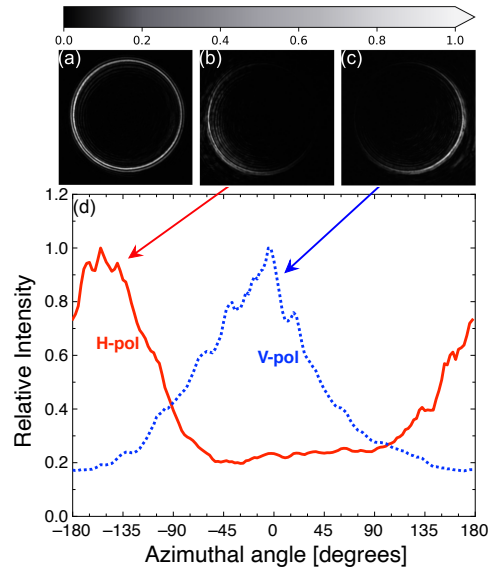


Fig. 6. CD holography with circularly polarized input object wave and linearly polarized (horizontal (H) or vertical (V) in laboratory frame) reference wave. (a) CD object wave transmission prior to hologram recording; (b) Object wave reconstruction for H-polarized reference; (c) Object wave reconstruction for V-polarized reference; (d) Normalized azimuthal dependence of local power on the reconstructed ring for cases (b) and (c).

215 the photoinduced charge transport leading to the formation of a space-charge field distribution  
 216 and by its conversion into a refractive index modulation (phase hologram) by means of the  
 217 material's electro-optic response. However, the space-charge electric field in the case of a  
 218 photorefractive grating is periodically modulated in space and the electro-optic response gets



219 an additional level of complexity as compared to the case where the field would be spatially  
 220 homogeneous. A thorough quantitative description requires the knowledge of all clamped  
 221 electro-optic coefficients, as well as of all the elastic, elasto-optic and piezo-electric properties of  
 222 the crystal, a significant part of which is still unknown for SPS. More details on these aspects can  
 223 be found in [45, 46] and references therein. For the above reasons the purpose of this section is  
 224 not to find a quantitative agreement between experiments and theory, but rather to confirm the  
 225 plausibility of the observation in the framework of a highly simplified theoretical approach.

226 As discussed above in connection with Fig. 2, the local polarization direction on the CD varies  
 227 along the ring circumference and spans an angle  $\theta$  of  $180^\circ$  over a full revolution of the polar  
 228 angle  $\varphi$  by  $360^\circ$ , the variation in  $\theta$  being always half the variation in  $\varphi$ . Considering different  
 229 spatial positions on the ring is therefore equivalent to considering a given linear polarization  
 230 component for the object wave among all the possible ones in the plane perpendicular to the  
 231 object wave  $\vec{k}$ -vector. We recall that for a circularly polarized input object beam the different  
 232 linear polarizations are equally probable, giving thus a complete ring. The easiest situation is  
 233 the one where the reference wave is linearly polarized (as for instance in Fig. 6). In this case the  
 234 problem of estimating the azimuthal dependence of the hologram diffraction efficiency and TWM  
 235 gain reduces essentially to an evaluation of the coupling strengths between a plane reference  
 236 wave with a fixed polarization and components of the object wave with all possible polarization  
 237 directions taken as a variable parameter. This coupling strength follows from coupled wave theory  
 238 for anisotropic gratings [47] and is a function of several material and experimental parameters.  
 239 On one hand, in the case of a grating created by means of the photorefractive effect the coupling  
 240 strength depends on the modulation depth amplitude of photorefractive space-charge field created  
 241 by the two considered wave components. On the other hand it depends on an effective scalar  
 242 electro-optic coefficient  $r_{eff}$  specific to the pair of waves being considered. This coefficient is a  
 243 function of the unit vector  $\hat{K}$  in the direction of the grating vector, of the anisotropic electro-optic  
 244 properties expressed by a tensor  $r_{ijk}^{eff}$  (see Ref. [45]), and of the two unit polarization vectors  $\hat{d}^{ref}$   
 245 and  $\hat{d}^{obj}$  for the reference and object waves, respectively. Importantly, here  $\hat{d}^{obj} \equiv \hat{d}^{obj}(\theta)$  is  
 246 the local object wave polarization corresponding to a given azimuthal position on the CD ring.  
 247 The expression for the scalar electro-optic coefficient determining the coupling is [45]

$$r_{eff} = \hat{d}_i^{obj} \cdot (r_{ijk}^{eff} \hat{K}_k) \cdot \hat{d}_j^{ref}, \quad (1)$$

248 where the Einstein summation convention over equal indices is utilized. Importantly, all the  
 249 vectors in the above equation have to be expressed in the orthogonal crystallo-physical reference  
 250 frame  $(xyz)$  in which the electro-optic tensor and other tensorial physical properties of the crystal  
 251 are usually expressed. This crystallo-physical system is oblique with respect to the laboratory  
 252 frame. As an example, the unit vector along the optic axis of propagation of the object wave is  
 253 expressed in the  $xyz$ -system as  $(0.4678, 0.7660, 0.4408)$ . In the laboratory frame the same vector  
 254 would possess only a unique unity component along the longitudinal coordinate perpendicular to  
 255 the entrance surface plane of the crystal. As discussed in [45], the third-rank tensor  $r_{ijk}^{eff}$  to be  
 256 used in (1) is a function of the clamped (high-frequency) electro-optic tensor elements and  
 257 depends also on the elastic, piezoelectric and elasto-optic properties of the material. Unfortunately  
 258 there is a lack of sufficient data on these mechanical properties for SPS, so that we do not have  
 259 access to the correct values to be used for the coefficients of the tensor  $r_{ijk}^{eff}$ . Instead we take a  
 260 rather rude approximation and we utilize the unclamped (free) electro-optic tensor elements  $r_{ijk}^T$   
 261 that were measured in [37] and [38] for what matters the first and third column of the tensor,  
 262 respectively. The employed values (in units of pm/V) are:  $r_{111}^T=174$ ,  $r_{221}^T=92$ ,  $r_{331}^T=140$ ,  $r_{131}^T=-25$ ,  
 263  $r_{113}^T=-67$ ,  $r_{223}^T=-22$ ,  $r_{333}^T=-25$  and  $r_{133}^T=14$ . The two still unknown coefficients  $r_{322}^T$  and  $r_{122}^T$   
 264 are expected to be rather small as they are associated to electric field directions perpendicular to  
 265 the crystallographic mirror plane containing the crystal's spontaneous polarization. Both these

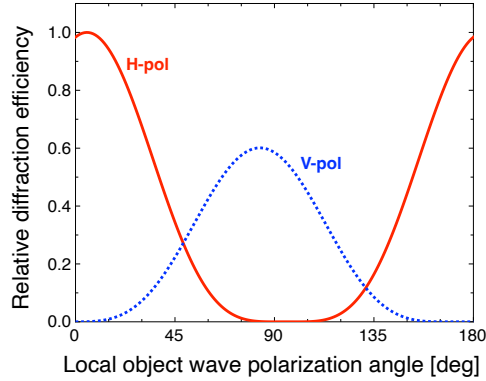


Fig. 7. Expected relative diffraction efficiency based on the simplified model described in Sect. 4. The object input wave is assumed circularly polarized and contain all linear polarization angles between  $0^\circ$  and  $180^\circ$ . The reference wave is assumed horizontally (red solid curve) or vertically polarized in the laboratory frame (blue dotted curve).

266 coefficients are set equal 0 for the following evaluation. Note also that the third column (index  
 267  $k = 3$ ) diagonal coefficients  $r_{113}$ ,  $r_{223}$  and  $r_{333}$  are taken here with the opposite sign as in the  
 268 original publication [38]. The reason is the different convention for the positive direction of the  
 269  $z$ -crystallo-physical axis. Here we stick to the the convention in Ref. [42] which is consistent  
 270 with the IEEE Standard on Piezoelectricity [48], with positive  $x$ - and  $z$ -direction such that both  
 271 piezoelectric coefficients  $d_{111}$  and  $d_{333}$  are positive, while in [38] a convention with  $d_{333}$  negative  
 272 was taken. It is also worth mentioning that for our experimental configuration the photorefractive  
 273 space-charge electric field has the largest projection on the  $x$ -axis and the elements of the first  
 274 column ( $k = 1$ ) of the electro-optic tensor largely determine the resulting scalar electro-optic  
 275 coefficient of Eq. (1).

276 The expected local hologram diffraction efficiency  $\eta$  is proportional to the square of the product  
 277 of  $r_{eff}$  with the amplitude  $E_{sc}$  of the photorefractive space-charge grating responsible for the  
 278 coupling of the two waves, i.e. [47]

$$\eta \propto [r_{eff}(\theta)E_{sc}]^2 . \quad (2)$$

279 This relation is valid for moderate diffraction efficiencies of the phase hologram, what is the case  
 280 for our gratings where the spatially averaged diffraction efficiency is typically of the order of  
 281 2-3 %. For what matters the space-charge field amplitude, we assume here that it is proportional  
 282 to the light modulation index induced by the two local polarizations, i.e. to the scalar product  
 283  $E_{sc} \propto (\hat{d}^{obj} \cdot \hat{d}^{ref})$ . This is a rather rude approximation for various reasons, the principal one  
 284 being associated to the fact that the space-charge field saturates at large modulation (for nearly  
 285 parallel  $\hat{d}^{obj}$  and  $\hat{d}^{ref}$ ), what leads to a sublinear dependence of  $E_{sc}$  on the modulation depth in  
 286 this regime. Therefore, the use of this approximation may lead to an excessive contrast in the  
 287 simulated curves for  $\eta$  as a function of the object wave polarization angle. Such a simulation is  
 288 shown in Fig. 7 for the case of a circularly polarized input object wave and a linearly polarized  
 289 reference (horizontal or vertical), as for the case of Fig. 6. It can be easily recognized in Fig. 7  
 290 that the diffraction for the horizontal and vertical polarization of the reference beam are nearly  
 291 (but not exactly) in anti-phase, meaning that their maxima are separated by approximately  $90^\circ$   
 292 in the object wave polarization, what corresponds to  $180^\circ$  on the CD ring. Despite for the many  
 293 simplifying assumptions, such a behavior agrees qualitatively with the experimental results  
 294 shown in Fig. 6.

295 In the case of two-wave mixing with weak signal wave, the local amplification  $\gamma(\theta)$  is expected

296 to depend exponentially on the exponential gain  $\Gamma(\theta)$  as [36]

$$\gamma(\theta) = e^{\Gamma(\theta)d} , \quad (3)$$

297 where  $d$  is the propagation distance in the crystal. In absence of an anisotropy of the photoexcitation  
298 process [46] the exponential gain can be expressed as

$$\Gamma \cong B(\hat{e}^{obj} \cdot \hat{e}^{ref})r_{eff}(\theta)\tilde{E}_{sc,im} , \quad (4)$$

299 where  $B$  is a constant that depends on the refractive indices associated to the two waves, on  
300 the light wavelength, and on the weak walk-off angle for the reference wave. Here  $\tilde{E}_{sc,im}$  is  
301 the imaginary part of the space-charge field amplitude. This is the nonlocal component of the  
302 space-charge field ( $90^\circ$  phase shifted with respect to the intensity modulation) and is responsible  
303 for the TWM gain. In materials with diffusion dominated charge transport, as in SPS [41], this  
304 is the dominant component of the space-charge field. Importantly, this quantity depends on  
305 experimental and materials parameters such as the beams crossing angles, the trap density in the  
306 crystal or its static dielectric properties. However,  $\tilde{E}_{sc,im}$  does not depend on the relative intensity  
307 of the two-waves and the corresponding light modulation depth, so that it can be considered as a  
308 constant for our purposes. In (4) the unit vectors  $\hat{e}^{obj}$  and  $\hat{e}^{ref}$  are along the electric fields of the  
309 object and reference waves, respectively. Due to the weak walk-off their scalar product can be  
310 safely approximated by the corresponding scalar product of the electric displacement unit vectors  
311  $\hat{d}^{obj}$  and  $\hat{d}^{ref}$ , so that

$$\Gamma \cong C(\hat{d}^{obj} \cdot \hat{d}^{ref})r_{eff}(\theta) , \quad (5)$$

312 where  $C$  is again a constant that depends on the specific experimental arrangement. In this  
313 equation the dependence on the local object wave polarization angle  $\theta$  appears twice, in the  
314 effective scalar electro-optic coefficient  $r_{eff}(\theta)$  and in the scalar product between the two  $\hat{d}$   
315 vectors. Figure 8 shows the expected amplification of the object wave for the case where the  
316 input polarizations of the object and reference wave are linear and mutually orthogonal and the  
317 reference wave polarization is aligned to one of the eigenpolarizations. This situation corresponds  
318 to the one of Fig. 4. The evaluation follows from the simplifying approach of Eqs. (3), (5) and (1)  
319 where the constant  $C$  was adjusted to give an average amplification  $\bar{\gamma}_0 \approx 4.5$ . In Fig. 8 the angle  $\theta$   
320 is given relative to the input polarization angle  $\theta_{0,obj}$  of the object wave ( $\theta_{0,obj} = -63.2^\circ$ ), so that  
321 the input wave intensity distribution (assumed proportional to  $\cos^2(\theta - \theta_{0,obj})$ ) is centered on the  
322 graph (dashed red curve). It can be recognized that the central region where the object wave has  
323 a maximum in absence of TWM interaction (corresponding to the left region in Fig. 4(a)) is not  
324 being amplified. This is due to the vanishing scalar product in (5) at this point. The maximum  
325 power in the amplified wave is predicted for two regions with a deviation of roughly  $\pm 60^\circ$  in  $\theta$   
326 ( $\pm 120^\circ$  along the ring) from this position. These maxima have different heights and are mainly a  
327 result of a combination of the effects of  $r_{eff}(\theta)$  together with the initial  $\theta$ -dependence of the  
328 unaffected CD wave. Even though the gain  $\Gamma$  tends to increase when the polarization direction  $\theta$   
329 gets closer to the one of the reference (at the two borders of the graph in Fig. 8), from a certain  
330 point on the lack of input light possessing such polarization components becomes dominant,  
331 what gives the minimum at  $\pm 90^\circ$  in Fig. 8. Experimentally this minimum corresponds to the dark  
332 region on the right-hand side of the circle on Fig. 4(b). Again, despite the highly simplifying  
333 assumptions, one can conclude that our modeling gives a satisfactory qualitative agreement with  
334 the observations. It is also worth mentioning that, even though here the average amplification is  
335 only slightly more than a factor of 4, locally the light can be amplified much stronger with peaks  
336 exceeding a factor of 40.

## 337 5. Discussion and conclusion

338 We have considered conical diffraction vector waves in a nonlinear photorefractive and optically  
339 biaxial material and we have shown, for the first time to our knowledge, that it is possible

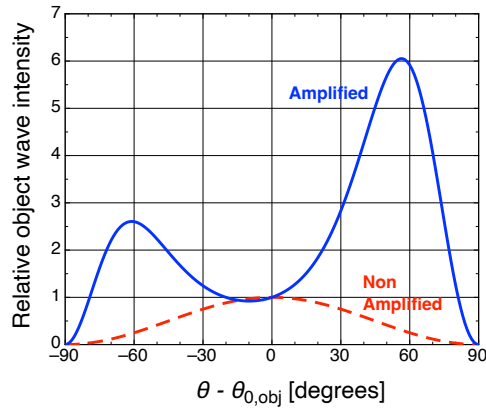


Fig. 8. Expected TWM amplification of the CD object wave as a function of the local angle of polarization  $\theta$  for the case of orthogonal linearly polarized input waves with the reference wave as one of the eigenpolarizations. The angle  $\theta_{0,obj}$  is the polarization angle of the input object wave, for which the non amplified CD wave is locally maximum. Red dashed curve: non amplified wave; Blue solid curve: amplified object wave with  $\tilde{\gamma}_0 \approx 4.5$ .

340 to amplify locally or record in-situ holograms of such waves. The biaxial crystal acts both  
 341 as generator of the CD vector wave and as a recording material. The holographic recording  
 342 process possesses a kind of angular resonance for the object wave direction parallel to one of the  
 343 optical axes. It was shown that under these aligned CD conditions of the crystal the recording  
 344 is possible even for orthogonal polarizations of the two interacting input beams thanks to the  
 345 polarization spreading resulting from the CD process. This property is remarkable since the  
 346 hologram recording is driven by the intensity distribution, as in conventional holography, and not  
 347 by the polarization states of the interacting waves, as in the framework of polarization holography  
 348 in polarization-sensitive media [49]. It was found that the pattern reconstruction depends on the  
 349 combination of recording polarizations and on the read-out polarization. For two-wave mixing  
 350 we obtain a varying local amplification across the CD ring, which can partially lead to a more  
 351 homogeneous power around the circle. The observed effects could be used as an additional  
 352 way to dynamically control the structure of the CD ring beam, for instance in connection to  
 353 optical particle trapping. We have analyzed the situation with the help of a highly simplified  
 354 theoretical approach based on the model of the photorefractive effect and general features of  
 355 conical diffraction. Despite its simplicity and the number of assumptions, the model allows a  
 356 surprisingly good qualitative description of the main experimental features. A more thorough  
 357 modeling would require the determination of a number of still unknown anisotropic material  
 358 parameters, as well as the consideration of details regarding CD under the conditions of optical  
 359 activity [50,51], which is little investigated to date and was neglected for the purpose of this work.  
 360 The present investigations have dealt with CD in a single crystal without cascades. However,  
 361 more complex vector patterns can be obtained with CD cascades, up to the cases where their  
 362 circularly symmetry is being broken [16, 17]. Since in such cases the output patterns are obtained  
 363 in the last focal image plane (near the last crystal in the cascade), we expect that recording and  
 364 local amplifications of such more complex patterns shall be possible as well if this last crystal is  
 365 able to store a hologram.

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369 **Data Availability.** Data underlying the results presented in this paper are not publicly available at this  
370 time but may be obtained from the authors upon reasonable request.

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