Letter

Photoinduced flipping of optical chirality during backward-wave parametric amplification in a chiral nonlinear medium

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We discuss the nonlinear process of backward-wave parametric amplification and oscillation, phase matched using a nonlinear grating inside a chiral medium. Our theory shows a remarkable feature manifesting as photoinduced flipping of optical chirality between the signal and the idler waves, without any change in the structural chirality of the medium. We show that circular birefringence, induced by chirality, leads to two different nonlinear processes with different phase-matching conditions that are associated with opposite states of circular polarizations. When the incident pump and signal beams are linearly polarized initially, their state of polarization evolves in an elliptical fashion because of the two competing nonlinear processes taking place in parallel. We also discuss the properties of such cavityless optical parametric oscillators based on distributed feedback provided by the nonlinear grating used for quasi phase matching.

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Chirality is a fundamental symmetry property of the constitution of matter, with fascinating and far-reaching implications in many areas of physics, chemistry, and biology [1–3]. The use of chirality for practical applications is also attracting attention. A chiral object or system is defined as the one whose structure and its mirror image (enantiomer) are not superposable by any symmetry operation (rotation or translation). Such a system also lacks spatial inversion symmetry. Although most of their physical properties are similar, a chiral object and its enantiomer may exhibit different responses and dynamics to external excitations. This feature is particularly striking for electromagnetic waves encountered in optics and photonics, both in the linear and nonlinear regimes [4–7]. As early as 1968, a chiral crystal was used for second-harmonic generation [8].

For a proper description of the optical response of a chiral medium, it is necessary to employ an appropriate multipole expansion of its interaction with the incident radiation [9]. The response of the chiral medium is expressed in terms of susceptibilities that govern the structural and dynamical aspects of the field-matter interaction. More specifically, the electric quadrupolar and magnetic dipolar terms have distinct impacts on the chiral features of the response. In particular, nonlocality through the field derivatives produces interesting properties such as optical spatial dispersion and other novel features such as gyrotropy [10]. It has been recognized that chirality plays a significant role in the domain of nonlinear optics with issues related to phase matching [4].

We have recently studied how the chirality of a nonlinear medium affects the process of optical parametric amplification (OPA) inside a chiral crystal [11,12]. We found that circular birefringence, induced by chirality, leads to two different nonlinear processes with different phase-matching conditions that are associated with the opposite states of circular polarizations. We also discussed the properties of conventional optical parametric oscillators (OPOs) made by placing a chiral nonlinear crystal inside an optical cavity.

In this Letter we apply our theory to a backward-wave optical parametric oscillator (BWOPO) and focus on the novel aspects introduced by the chiral nature of a nonlinear crystal, which sets up optical activity and manifests as circular birefringence and gyrotropy. The BWOPO makes use of three-wave mixing inside a nonlinear medium with finite second-order susceptibility $\chi^{(2)}$ but, in contrast to the conventional OPOs, the idler in a BWOPO travels in a direction opposite to that of the pump and signal waves [13–17].

Phase matching is realized through the technique of quasi phase matching (QPM) by alternating the sign of $\chi^{(2)}$ in a periodic fashion along the medium's length [18]. The nonlinear grating used for QPM also provides distributed feedback, which is useful for a cavityless BWOPO that does not require alignment of cavity mirrors, making the device simple and more reliable.

Our theory shows another remarkable feature of such OPAs and OPOs: photoinduced flipping of the chirality between the

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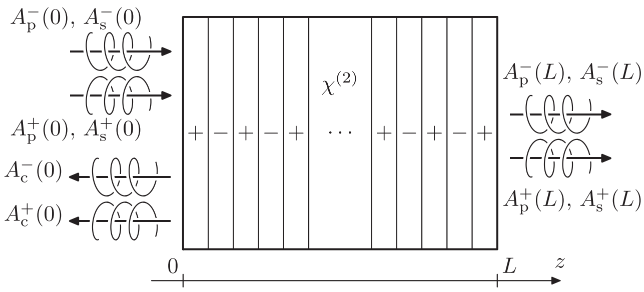

FIG. 1. Schematic showing parametric amplification inside a chiral structure, designed with a periodic modulation of the sign of the nonlinear coupling coefficient, required for quasi phase matching.

signal and the idler, without affecting structural chirality of the crystal. In a chiral crystal, structural chirality is set by the lattice structure during its growth, and it is challenging to manipulate the handedness of the crystal afterward. This is the reason why light-induced changes in chirality are likely to become important from a practical perspective. Recently, photoinduced chirality has been observed even in a nonchiral crystal [19].

Figure 1 shows the schematic of a chiral medium with a $\chi^{(2)}$ grating used for QPM. In the case of OPAs, the signal to be amplified is launched together with a pump beam in the forward direction. In the case of OPOs, only the pump beam is launched into the nonlinear medium. In both cases, the idler wave, generated during the three-wave mixing process, propagates in the backward direction and its frequency ω_c satisfies the energy-conservation condition $\omega_p = \omega_s + \omega_c$, where ω_p and ω_s are frequencies of the pump and signal beams.

The starting point of the analysis is the phenomenological constitutive relation for optically active media in the form $\mathbf{D} = \varepsilon_0(\varepsilon_r \mathbf{E} + \gamma \nabla \times \mathbf{E})$, where both ε_r and γ are tensors. The nonlocal character of this response is reflected by the presence of spatial derivatives of the electric field (rotation through the curl operation), which is a consequence of the electromagnetic induction and is compatible with the expressions derived using the interaction up to linear terms in the field derivatives.

For relatively wide optical beams propagating in the z direction, we can ignore diffraction and treat them as plane waves. However, the transverse components of the electric field are coupled by chirality of the medium such that the three plane waves are circularly polarized with different wave vectors for the left and right (LCP and RCP) components. Using the representation $\mathbf{E}(z,t) = \mathrm{Re}[(\mathbf{e}_+ E_+ + \mathbf{e}_- E_-)\exp(-i\omega_j t)]$, with $E_\pm = (E_x \mp iE_y)/\sqrt{2}$ in the complex-valued LCP/RCP basis $\mathbf{e}_\pm = (\mathbf{e}_x \pm i\mathbf{e}_y)/\sqrt{2}$, the components of the three plane waves can be written as

$$E_\pm(z,\omega_p) = A_p^\pm(z)\exp(ik_p^\pm z), \tag{1a}$$

$$E_\pm(z,\omega_s) = A_s^\pm(z)\exp(ik_s^\pm z), \tag{1b}$$

$$E_\pm(z,\omega_c) = A_c^\pm(z)\exp(-ik_c^\pm z), \tag{1c}$$

where the slowly varying amplitudes $A_j^\pm(z) = A^\pm(z,\omega_j)$, $j =$ p, s, c, evolve with z and with the wave numbers associated with the circularly polarized components of the beam at frequency ω_j given by

$$k_j^\pm = k_j \pm \alpha_j. \tag{2}$$

Here, $k_j = n_j\omega_j/c$ is the electric dipolar wave number with $n_j = n(\omega_j)$ being the refractive index, while $\alpha_j = f_c(\omega_j/2c)$ with f_c being a dimensionless quantity measuring chirality of the medium. Its presence in Eq. (2) is responsible for the circular birefringence of the chiral nonlinear medium.

In the plane-wave approximation, the LCP and RCP components of the electric field at the frequency ω_j satisfy the two coupled equations [11]

$$\left(\frac{\partial^2}{\partial z^2} + k_j^2 \pm 2i\alpha_j\frac{\partial}{\partial z}\right)E_\pm(z,\omega_j) = -\mu_0\omega_j^2 P_\pm^{(2)}(z,\omega_j). \tag{3}$$

The polarization term in this equation depends on the nonlinear crystal and the geometry used for a specific nonlinear process. We focus on a uniaxial anisotropic crystal of symmetry class 32 such that the z axis is aligned along its optic axis and the x axis lies along its twofold axis of symmetry. In the case of three-wave mixing, the circularly polarized components of the nonlinear polarization at the three frequencies ω_j can be written as [11]

$$P_\pm^{(2)}(z,\omega_s) = \frac{\epsilon_0}{2}\chi^{(2)}(z)A_c^{\mp *}(z)A_p^\mp(z)\exp[i(k_p^\mp + k_c^\mp)z], \tag{4a}$$

$$P_\mp^{(2)}(z,\omega_c) = \frac{\epsilon_0}{2}\chi^{(2)}(z)A_s^{\pm *}(z)A_p^\mp(z)\exp[i(k_p^\mp - k_s^\pm)z], \tag{4b}$$

$$P_\mp^{(2)}(z,\omega_p) = \frac{\epsilon_0}{2}\chi^{(2)}(z)A_s^\pm(z)A_c^\mp(z)\exp[i(k_s^\pm - k_c^\mp)z], \tag{4c}$$

where $\chi^{(2)}(z)$ denotes a specific component of the second-order nonlinear susceptibility involved in the three-wave mixing process. The z dependence of $\chi^{(2)}(z)$ takes into account of the nonlinear grating used for QPM (see Fig. 1).

We look for solutions of Eqs. (3) in the form of Eq. (1). As $\chi^{(2)}(z)$ is a periodic function, we follow a standard procedure and expand it in a Fourier series [18]. QPM occurs for a specific Fourier component with the wave number $K_q = 2\pi m/\Lambda$, where the integer m represents that component. Typically, the grating period Λ is chosen to ensure $m = 1$. We assume this to be the case and use $K_q = 2\pi/\Lambda$ in the following discussion.

In the paraxial-wave approximation, the use of Eqs. (3) and (4) provides the following three coupled equations describing evolution of the circularly polarized components of the three waves involved in the parametric amplification process,

$$\frac{dA_s^\pm(z)}{dz} = i\frac{\omega_s\chi_e^{(2)}}{4cn_s}A_c^{\mp *}(z)A_p^\mp(z)\exp(i\kappa_\mp z), \tag{5a}$$

$$-\frac{dA_c^\mp(z)}{dz} = i\frac{\omega_c\chi_e^{(2)}}{4cn_c}A_s^{\pm *}(z)A_p^\mp(z)\exp(i\kappa_\mp z), \tag{5b}$$

$$\frac{dA_p^\mp(z)}{dz} = i\frac{\omega_p\chi_e^{(2)}}{4cn_p}A_s^\pm(z)A_c^\mp(z)\exp(-i\kappa_\mp z), \tag{5c}$$

where $\chi_e^{(2)}$ is an effective nonlinear parameter and the residual phase mismatch is defined as

$$\kappa_\pm = (\Delta k - 2\pi/\Lambda)(1 \pm \beta), \quad \beta = \Delta\alpha/(\Delta k - 2\pi/\Lambda). \tag{6}$$

Here, Δk is the phase mismatch resulting from chromatic dispersion and $\Delta\alpha$ is the contribution resulting from the medium's chirality such that

$$\Delta k = k_p - k_s + k_c, \quad \Delta\alpha = \alpha_p + \alpha_s + \alpha_c. \tag{7}$$

The dimensionless parameter β in Eq. (6) indicates the relative contribution of chirality to the phase mismatch. Even though Δk is relatively large in a BWOPO because of backward propagation of the idler wave, the nonlinear grating is designed in practice such that its contribution to phase mismatch, $2\pi/\Lambda$, almost compensates for Δk. As a result, the difference $|\Delta k - 2\pi/\Lambda|$ is typically comparable to the chiral part $\Delta\alpha$ of the phase mismatch. For this reason, the value of β can vary over a wide range. Depending on the relative signs of Δk and $\Delta\alpha$, the chiral contribution can even be used for perfect phase matching by designing a BWOPO such that $\beta = \pm 1$. This will become clear later when we discuss numerical results for a specific configuration.

Equations (5) represent two sets of three coupled equations for the choice of $\pm$ signs corresponding to the LCP and RCP components of the signal and idler beams. Notice that the LCP component of the signal couples to the RCP component of the pump and idler beams; the opposite occurs for the RCP component. It is also important to note that the phase-matching conditions are different for these two nonlinear processes. In contrast, a single phase-matching condition needs to be satisfied when the nonlinear medium is not chiral. We note that QPM has been used with success for sum-frequency generation using chiral liquids [20–22].

Equations (5) show that the parametric amplification process in a chiral medium depends strongly on the initial state of polarization (SOP) of the pump and signal beams. The simplest situation occurs when the two beams are launched with opposite circular SOPs, say, RCP for the signal and LCP for the pump. In this case, the SOP of the idler wave matches that of the signal. All three waves remain circularly polarized inside the chiral medium.

The situation is quite different when the pump and signal beams are launched with identical linear SOPs (say, both polarized along the x axis). As two different nonlinear processes couple the circularly polarized components of these waves, requiring different phase-matching conditions, the SOPs of the signal and idler beams not only become elliptical but also change as the two beams are amplified by the pump beam. Moreover, the two beams exhibit opposite optical chirality in the sense that their circular birefringence switches signs. Such a photoinduced flipping of optical chirality is remarkable because it occurs without any change in the structural chirality of the crystal. Its origin can be traced to the phase-conjugate nature of the nonlinear interaction in Eqs. (5).

Equations (5) simplify considerably for a pump beam so intense that its depletion remains negligible inside the chiral medium and $A_p^{\mp}(z)$ can be treated as a constant, in which case the LCP and RCP components of the signal and idler beams (with $\pm$ sign) evolve with z as

$$\frac{dA_s^{\pm}(z)}{dz} = i\gamma_s A_p^{\mp} A_c^{\mp *}(z)\exp(i\kappa_{\mp} z), \tag{8a}$$

$$-\frac{dA_c^{\mp}(z)}{dz} = i\gamma_c A_p^{\mp} A_s^{\pm *}(z)\exp(i\kappa_{\mp} z), \tag{8b}$$

where the two nonlinear parameters are defined as

$$\gamma_s = \frac{\omega_s \chi_e^{(2)}}{4cn_s}, \quad \gamma_c = \frac{\omega_c \chi_e^{(2)}}{4cn_c}. \tag{9}$$

It is important to notice the minus sign in Eq. (8b), indicating that the idler travels in the backward direction. This feature requires that Eqs. (8a) and (8b) should be solved with boundary conditions imposed at the opposite ends of the nonlinear medium:

$$A_s^{\pm}(z=0) = A_{s0}^{\pm}, \quad A_c^{\mp}(z=L) = 0. \tag{10}$$

Consider first the OPA case in which a signal beam with the amplitude A_{s0}^{+} is launched with the pump beam. By differentiating Eq. (8a) and using Eq. (8b), $A_s^{+}(z)$ is found to satisfy

$$\frac{d^2A_s^{\pm}}{dz^2} - i\kappa_{\mp}\frac{dA_s^{+}}{dz} + (\gamma_s\gamma_c|A_p^{\mp}|^2)A_s^{\pm} = 0. \tag{11}$$

Its general solution can be written as

$$A_s^{\pm}(z) = [C_1^{\pm}\cos(b_{\pm}z) + C_2^{\pm}\sin(b_{\pm}z)]\exp(i\kappa_{\mp}z/2), \tag{12}$$

where

$$b_{\pm}^2 = \gamma_s\gamma_c|A_p^{\mp}|^2 + \kappa_{\mp}^2/4. \tag{13}$$

The constants $C_1^{\pm}$ and $C_2^{\pm}$ are found by imposing the boundary conditions given in Eq. (10). For a chiral medium of length L, we obtain [17]

$$A_s^{\pm}(z) = A_{s0}^{\pm}\exp(i\kappa_{\mp}z/2) \times \left[\frac{\cos[b_{\pm}(L-z)] + (i\kappa_{\mp}/2b_{\pm})\sin[b_{\pm}(L-z)]}{\cos(b_{\pm}L) + (i\kappa_{\mp}/2b)\sin(b_{\pm}L)}\right]. \tag{14}$$

The amplification factors for the LCP and RCP signals at $z = L$ become

$$G_s^{\pm} = \left|\frac{A_s^{\pm}(L)}{A_s^{\pm}(0)}\right|^2 = \left[\cos^2(b_{\pm}L) + (\kappa_{\mp}/2b_{\pm})^2\sin^2(b_{\pm}L)\right]^{-1}. \tag{15}$$

The amplitude of the idler wave is found following the same method. This wave propagates backward and its amplitude at a distance z is given by

$$A_c^{\mp}(z) = \frac{i\gamma_c A_p^{\mp}A_{s0}^{\pm *}\exp(i\kappa_{\mp}z/2)}{\cos(b_{\pm}L) - (i\kappa_{\mp}/2b_{\pm})\sin(b_{\pm}L)}\frac{\sin[b_{\pm}(z-L)]}{\sin(b_{\pm}L)}. \tag{16}$$

Flipping of the chirality for the idler is a consequence of the same phase factor $\exp(i\kappa_{\mp}z/2)$ appearing in Eqs. (14) and (16). As the idler propagates backward, the sign of $\kappa_{\mp}$ is reversed because of phase conjugation of the signal seen in Eq. (8b).

We next consider the case of BWOPOs, which create a tunable source at two wavelengths using a single pump beam. In a BWOPO, the signal and idler beams build up from noise photons (provided by parametric fluorescence). As the idler wave travels in a direction opposite to that of the the signal, distributed feedback provided by the nonlinear grating used for QPM is employed to make cavityless BWOPOs.

An important concept in the operation of any OPO is that of its threshold, defined as the minimum pump power needed before coherent radiation is emitted by the OPO at the signal

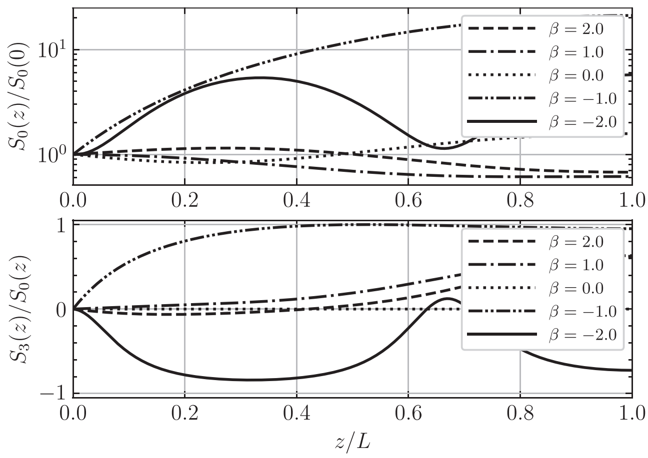

FIG. 2. Evolution of the signal intensity $S_0(z)/S_0(0)$ and ellipticity $S_3(z)/S_0(z)$ inside the chiral medium when the pump and signal are linearly polarized initially with $I_p/I_{th} = 2.0$. The relative chiral contribution varies from $\beta = -2$ to 2 for a fixed dipolar phase mismatch $(\Delta k - 2\pi/\Lambda)L/2 = 1.5$.

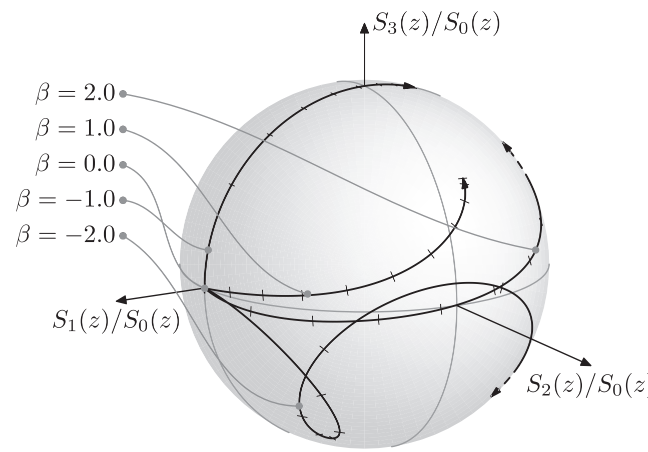

FIG. 3. Polarization state evolution of the signal mapped onto the Poincaré sphere for the five traces in Fig. 2.

and idler wavelengths. For conventional OPOs, this threshold is found by requiring that the optical field remains unchanged after each round trip inside the cavity. As a cavityless BWOPO is designed to make use of the distributed feedback provided by a built-in nonlinear grating, it should amplify few noise photons provided by parametric fluorescence by a large factor over its length L. In the case of perfect phase matching ($\kappa = 0$), the single-pass amplification factor is found from Eq. (15) to be $G_s^{\pm} = \sec^2(b_{\pm}L)$. This gain becomes infinitely large for $b_{\pm}L = \pi/2$. From Eq. (13), $b_{\pm}$ is related to the pump's intensity $I_p^{\mp} = |A_p^{\mp}|^2$, and the threshold is reached when

$$\gamma_s\gamma_c I_{th}^{\mp} = \pi^2/(2L)^2. \tag{17}$$

As a specific example, we consider OPA when both the pump and signal beams are linearly polarized initially such that $I_p/I_{th} = 2.0$. In Fig. 2, we show the evolution of the signal's intensity and ellipticity inside the chiral medium for five phase-mismatch situations using the Stokes parameters [23]

$$S_0 = |A_s^+|^2 + |A_s^-|^2, \quad S_1 = 2\mathrm{Re}[A_s^{+*}A_s^-],$$
$$S_3 = |A_s^+|^2 - |A_s^-|^2, \quad S_2 = 2\mathrm{Im}[A_s^{+*}A_s^-]. \tag{18}$$

As seen there, the signal's gain depends on the relative chiral contribution β to the phase mismatch [see Eq. (6)]. As discussed earlier, when $\beta = -1$, the residual phase mismatch can be fully compensated by the chiral contribution $\Delta\alpha$. Even more interesting is the introduction of polarization state selectivity during the OPA process. As shown by the ellipticity curves in Fig. 2, even a moderate change in the value of β, representing the ratio of chiral to electric dipolar phase mismatch, can switch the evolution from a circular polarization state to a periodically repetitive modulation of the ellipticity.

The remarkable features in the polarization state evolution induced by chirality of the nonlinear medium can clearly be seen in Fig. 3, where the Stokes parameters (S_1, S_2, S_3) are mapped onto the Poincaré sphere for the same parameter values used in Fig. 2. In the absence of a chiral contribution to the phase matching ($\Delta\alpha = 0$), the SOP remains fixed at its initial point $(1, 0, 0)$. When the relative chiral contribution exactly matches the dipolar mismatch ($\beta = -1$), the SOP remains linear but takes a meriodinal path around the Poincaré sphere, until it reaches the opposite hemisphere and becomes orthogonal to the initial polarization state. For other values of the chiral coefficient, the SOP either becomes LCP or RCP, depending on the sign of β, or enters a periodic oscillatory state (see the curve for $\beta = -2$).

Notice that the LCP component of the signal builds up from the RCP component of the pump; the opposite occurs for the RCP component. Both three-wave mixing processes occur simultaneously, but only one can be phase matched by the built-in nonlinear grating. Thus, irrespective of the SOP of the launched pump beam, the output of the BWOPO will be

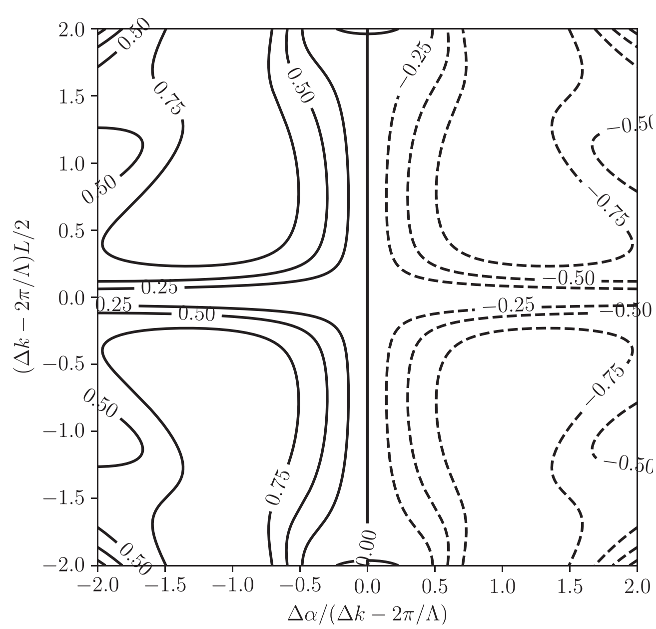

FIG. 4. Trajectories of constant ellipticity of the amplified signal, $S_3(L)/S_0(L)$, showing the interconnection between the dipolar phase mismatch and the chiral contribution to it governed by β. Both the pump and signal are linearly polarized initially with $I_p/I_{th} = 2.0$.

circularly polarized. Its nature (LCP or RCP) is governed by the phase-matching condition that is supported by the grating.

An intriguing question in this context is the symmetry aspects of interchangeability between the residual phase mismatch, $\Delta k - 2\pi/\Lambda$, and the chiral contribution $\Delta\alpha$. As shown in Fig. 4, where the ellipticity $S_3(L)/S_0(L)$ is plotted as functions of electric dipolar and chiral phase mismatch, although ellipticity exhibits perfect symmetry with respect to electric dipolar mismatch, it is perfectly antisymmetric with respect to the chiral coefficient $\Delta\alpha$. However, the relation between Δk and $\Delta\alpha$ in maintaining a constant ellipticity is nontrivial, with multivalued mutual solutions.

In conclusion, we have developed a theory behind the nonlinear process of backward-wave optical parametric amplification inside a chiral crystal. The process is phase matched by a nonlinear grating formed by the periodic structure in which neighboring sections have opposite signs of $\chi^{(2)}$. We show that circular birefringence, induced by chirality, leads to two different nonlinear processes with different phase-matching conditions that are associated with opposite states of circular polarizations. A single nonlinear process occurs only when the states of polarization of the incident pump and signal beams are both circular and orthogonal. When these beams are linearly polarized initially, the state of polarization of the signal being amplified evolves in an elliptical fashion because of the two competing nonlinear processes taking place in parallel. We have also found that the signal and idler beams exhibit opposite optical chirality in the sense that their circular birefringence switches signs. Such a photoinduced flipping of optical chirality is remarkable because it occurs without any change in the structural chirality of the crystal.

We also discuss the properties of a chiral BWOPO, which does not require any cavity and emits signal and idler beams in the opposite directions when pumped by a single pump beam. Its operation makes use of distributed feedback provided by the nonlinear grating used for QPM. We derive a simple expression for the threshold of such BWOPOs. Our main conclusion is that a BWOPO should emit circularly polarized light whose helicity depends on the specific nonlinear process that is nearly phase matched by the $\chi^{(2)}$ grating. Only when the nonlinear grating is designed such that both parametric processes exhibit some phase mismatch, the SOP of the emitted light may be linear or elliptical, depending on the SOP of the pump beam.

Experimental demonstration of a chiral BWOPO requires a crystal exhibiting molecular chirality that can also be poled in a periodic fashion for fabricating a $\chi^{(2)}$ grating. For example, a uniaxial anisotropic crystal of symmetry class 32, whose optic axis is aligned with the direction of propagating pump wave, would be a good candidate. An alternative is to employ a magnetoactive material, which becomes chiral and exhibits circular birefringence, when a static magnetic field is applied along a specific direction. Such materials are used commonly for making optical isolators. In the visible and near-infrared regions, these include terbium gallium garnet, yttrium iron garnet, and CeF_3 crystals [24]. Suitable materials in the mid-infrared region are $CeAlO_3$, EuF_2, and KTb_3F_{10}.

Data availability. The data that support the findings of this article are openly available [25].

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