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Christos Flytzanis ; Fredrik Jonsson ; Govind P. Agrawal

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Christos Flytzanis,[1,a)] Fredrik Jonsson,[2,b)] and Govind P. Agrawal[3]

AFFILIATIONS

[1] Laboratoire de Physique de l'École Normale Supérieure, ENS, Université PSL, CNRS, Sorbonne Université, Université Paris Cité, F-75005 Paris, France
[2] Division of Electricity, Department of Electrical Engineering, Uppsala University, SE-75103 Uppsala, Sweden
[3] The Institute of Optics, University of Rochester, Rochester, New York 14627, USA

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[a)] **Author to whom correspondence should be addressed:** christos.flytzanis@phys.ens.fr
[b)] **Also at:** Mycronic AB, SE-18303, Sweden.

ABSTRACT

We discuss the nonlinear process of optical parametric amplification inside a chiral crystal. We show 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. 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, 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 singly and doubly resonant optical parametric oscillators made by placing a chiral nonlinear crystal inside an optical cavity.

I. INTRODUCTION

Chirality, an intriguing property of matter, has been attracting considerable attention in recent years.[1–3] Chiral media have been used for extensive linear and nonlinear spectroscopic studies to probe their structural and dynamic features. In particular, the interaction of electromagnetic waves with a chiral medium produces interesting properties such as optical activity, spatial dispersion, and the breakdown of inversion symmetry. Even though such phenomena do not affect much the magnitude of linear refractive index, they impact considerably the state of polarization (SOP) of light and introduce novel features such as gyrotropy.

In this context, the nonlinear effects, particularly those sensitive to the breakdown of spatial inversion symmetry, are of growing interest for devices with specific functionalities involving gyrotropy.[4–8] In this work, we address the issue of optical parametric amplification (OPA) and its use for making an optical parametric oscillator (OPO). Non-chiral nonlinear crystals are routinely used for making OPAs and OPOs, and the discussion of such devices can be found in any book on nonlinear optics.[9–12] Here, we focus on the novel aspects introduced by the chiral nature of a nonlinear crystal and, specifically, the impact of the chirality through the optical activity on the phase-matching issue, which determines the efficiency of the nonlinear optical processes in general and in fact their very functionality. Our results show that new features emerge from the nonlocality and the breakdown of spatial inversion symmetry, owing to the contributions of electric quadrupolar and magnetic dipolar interactions, in addition to the electric dipolar ones, and strongly affect the phase matching procedure through the involvement of the optical activity.

For a proper description for a chiral medium, it is necessary to employ an appropriate multipole expansion of the interaction of that matter with incident radiation.[13,14] In this approach, nonlocality enters through the field derivatives, and the response of the chiral medium is expressed in terms of susceptibilities that govern the structural and dynamical aspects of the field–matter interaction. The quadrupolar and magnetic dipolar terms in the multipole expansion have distinct meaning, and their impact on the dielectric response can be studied using Maxwell's equations.

In this work, our objective is to discuss OPA in a nonlinear chiral dielectric with a finite second-order susceptibility. To simplify the following discussion, we ignore the nonlinear effects in Sec. II and focus on the chiral aspects of a linear dielectric medium. The nonlinear effects are included in Sec. III, where we discuss the nonlinear process of three-wave mixing inside a chiral crystal exhibiting a finite second-order susceptibility. A set of three coupled nonlinear equations is derived in this section for both the left- and right-circularly polarized (RCP) components associated with the pump, signal, and idler waves. This set of equations is solved approximately in Sec. IV under the assumption that the pump beam remains nearly undepleted inside the nonlinear crystal. The results show that, owing to the chirality, the parametric gain and the phase-matching condition are generally different for the left- and right-circularly polarized components of the signal and idler waves. The use of chiral nonlinear crystals for making an OPO is discussed in Sec. V, and the main conclusions are summarized in Sec. VI.

II. LINEAR CHIRAL MEDIA

The starting point of the analysis is the phenomenological constitutive relation for optically active media in the form $\mathbf{D} = \varepsilon_0 (\varepsilon\mathbf{E} + \gamma\nabla\times\mathbf{E})$, where both ε 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) being a consequence of the electromagnetic induction and is compatible with the expressions derived using the interaction up to linear terms in the field derivatives. A specific component of the electric flux density $\mathbf{D}$ can be written as

$$D_i = \varepsilon_0\varepsilon_{ij}E_j, \tag{1}$$

with the dielectric permeability tensor including first-order spatial derivatives involving rotation, with i, j, and k taking three values x, y, and z (or 1, 2, 3). Here, ε_{ij} is taken as a linear combination,

$$\varepsilon_{ij} = \varepsilon_{ij}^{(0)} + \gamma_{ijl}\frac{\partial}{\partial x_l}, \tag{2}$$

of a term $\varepsilon_{ij}^{(0)}$ without rotatory dispersion and a term γ_{ijl} being a third-rank tensor obeying the [anti-]symmetry condition $\gamma_{ijl} = -\gamma_{jil}$ for a non-absorbing dispersive medium. We consider propagation along the optic axis of a cubic or doubly refracting crystal so that γ_{ijl} reduces to the form

$$\gamma_{ijl} = \frac{cf}{\omega}e_{ijl}, \tag{3}$$

where c is the vacuum speed of light, ω is the angular frequency of radiation, and f is a pseudoscalar measuring the magnitude of specific rotation at this frequency. The unit permutation tensor of third rank e_{ijl} is also known as the Levi–Cività tensor.

The use of Eq. (2) in Maxwell's equations for a nonmagnetic dielectric medium leads to the frequency-domain wave equation,

$$\left[(\nabla^2 + k^2)\delta_{ij} + \gamma_{ijl}\frac{\partial}{\partial x_l}\right]E_j(\omega) = 0, \tag{4}$$

where δ_{ij} is the Kronecker delta function and k is defined as

$$k^2 = (\omega/c)^2\varepsilon^{(0)}. \tag{5}$$

For a plane wave propagating along the z direction, the transverse electric field components, $E_x(\omega,z)$ and $E_y(\omega,z)$, vary with z but do not depend on x or y. In this case, the solutions of Eq. (4) are left-circularly polarized (LCP) and right-circularly polarized (RCP) plane waves with different wave vectors. If we denote the electric field of these two waves as $\mathbf{E}^{\pm} = E_x^{\pm}\hat{x} + E_y^{\pm}\hat{y}$, the four field components satisfying Eq. (4) can be written as

$$E_x^{\pm}(\omega,z) = \pm iE_y^{\pm}(\omega,z) = E_0\exp(ik^{\pm}z), \tag{6}$$

where E_0 is the common amplitudes and $k^{\pm}$ are the wave numbers associated with the LCP and RCP waves. Their magnitudes differ by a small amount because of chirality such that

$$k^{\pm} \approx k \pm \alpha, \tag{7}$$

where the chirality parameter α is defined as

$$\alpha = (k^+ - k^-)/2 = \omega f/2c. \tag{8}$$

This result shows that the chirality of a medium introduces circular birefringence in that medium.

A linearly polarized wave can be decomposed into its LCP and RCP parts that acquire different phase shifts when $\alpha \neq 0$. After propagating some distance into the chiral medium, its electric field can be written as

$$E_j(\omega,z) = \frac{1}{2}[E_j^+(\omega,z) + E_j^-(\omega,z)] \tag{9}$$

for $j = x, y$. Using Eq. (6) in this expression, we obtain

$$E_x(\omega,z) = E_0\exp(ikz)\cos(\alpha z), \tag{10a}$$

$$E_y(\omega,z) = E_0\exp(ikz)\sin(\alpha z). \tag{10b}$$

These relations show that the initially linear state of polarization (SOP) of a plane wave remains linear but rotates at the rate $\alpha = \omega f/2c$ as the plane wave traverses the chiral medium.

From the quantum-mechanical expressions[15,16] of the response functions in Eqs. (1) and (2), it is clear that γ measures the photoinduced modification of the orbital (angular) momentum through electromagnetic induction; this sets up a time varying magnetic field, which induces the additional electric moment related to the optical activity in Eq. (1). This is analogous to the electronic spin–orbit coupling energy in semiconducting compounds, being also a (nonlocal) manifestation of the electromagnetic induction and provide photospin–orbit coupling in photonic structures.

III. OPA IN NONLINEAR CHIRAL MEDIA

We now extend the preceding treatment to the case of a nonlinear chiral medium following the approach of Bey and Rabin.[16] The second-order nonlinear process of OPA involves the interaction of three waves, known as the pump (p), the signal (s) and the idler (c), at the frequencies ω_p, ω_s, and ω_c, respectively. The energy conservation requirement for the OPA process leads to the following relation among these frequencies:

$$\omega_p = \omega_s + \omega_c. \tag{11}$$

In the nonlinear case, the frequency-domain wave equation in Eq. (4) should be modified to include the second-order nonlinear polarization setup by the effective fields generated inside a dielectric medium by the incident ones. In the present case, the effective fields have an additional component arising from chirality, together with the components in Eq. (10). Moreover, the second-order polarization has a small term containing third-order susceptibility that we neglect in the following analysis. Owing to the chiral nature of the nonlinear medium, we need to consider the two circularly polarized components at each frequency separately.

Assuming a time dependence of the form $\exp(-i\omega_i t)$ for a wave oscillating at the frequency ω_i and making the plane wave approximation, the x and y components of the plane wave at the frequency ω_i are found to satisfy the following two coupled equations:

$$\left(\frac{\partial^2}{\partial z^2}+k_i^2\right)E_x^{\pm}(\omega_i,z)+\left(\frac{\omega_i f_i}{c}\right)\frac{\partial E_y^{\pm}(\omega_i,z)}{\partial z} = \frac{1}{\varepsilon_0}\left(\frac{\omega_i}{c}\right)^2 P_x^{(2)\pm}(\omega_i,z), \tag{12a}$$

$$\left(\frac{\partial^2}{\partial z^2}+k_i^2\right)E_y^{\pm}(\omega_i,z)-\left(\frac{\omega_i f_i}{c}\right)\frac{\partial E_x^{\pm}(\omega_i,z)}{\partial z} = \frac{1}{\varepsilon_0}\left(\frac{\omega_i}{c}\right)^2 P_y^{(2)\pm}(\omega_i,z), \tag{12b}$$

where the last term accounts for the induced nonlinear polarization in the chiral medium, $f_i = f(\omega_i)$ accounts for the frequency dependence of chirality, and k_i is defined as

$$k_i^2 = \varepsilon^{(0)}\omega_i/c \quad (i = p,\ s,\ c). \tag{13}$$

As in the case of a linear chiral medium, the wave number $k_i^{\pm}$ can be written in the form

$$k_i^{\pm} \approx k_i \pm \alpha_i, \quad \alpha_i = \omega_i f_i/2c. \tag{14}$$

Similar to the linear case, we look for solutions of the two inhomogeneous wave equations in Eq. (12) in the form

$$E_x^{\pm}(\omega_i,z) = \pm i E_y^{\pm}(\omega_i,z) = E^{\pm}(\omega_i,z)\exp(ik_i^{\pm}z). \tag{15}$$

Using $P_x^{(2)\pm}(\omega_i,z) = \pm i P_y^{(2)\pm}(\omega_i,z)$ in Eq. (12) and making the paraxial-wave approximation, the slowly varying amplitudes $E^{\pm}(\omega_i,z)$ of the electric field satisfy (for $i = \mathrm{p,s,c}$)

$$\frac{dE^{\pm}(\omega_i,z)}{dz} = \frac{i}{2k_i\varepsilon_0}\left(\frac{\omega_i}{c}\right)^2 P_x^{(2)\pm}(\omega_i,z)\exp(-ik_i^{\pm}z). \tag{16}$$

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. The incoming pump and signal waves are polarized linearly along the x axis, and their wave vectors are directed along the z axis. In this situation,

$$E^{+}(\omega_{\mathrm{p}},0) = E^{-}(\omega_{\mathrm{p}},0), \tag{17a}$$

$$E^{+}(\omega_{\mathrm{s}},0) = E^{-}(\omega_{\mathrm{s}},0), \tag{17b}$$

$$E^{+}(\omega_{\mathrm{c}},0) = E^{-}(\omega_{\mathrm{c}},0) = 0. \tag{17c}$$

The nonlinear polarizations at the frequencies of the three waves involved in the OPA process are obtained following the approach of Bey and Rabin.[16] The result is

$$P_x^{(2)\pm}(\omega_{\mathrm{s}},z) = \pm i P_y^{(2)\pm}(\omega_{\mathrm{s}},z) = \frac{\varepsilon_0}{2}d_{11}E^{\pm *}(\omega_{\mathrm{c}},z)E^{\mp}(\omega_{\mathrm{p}},z)\exp\left(i(k_{\mathrm{p}}^{\mp}-k_{\mathrm{c}}^{\pm})z\right), \tag{18a}$$

$$P_x^{(2)\pm}(\omega_{\mathrm{c}},z) = \pm i P_y^{(2)\pm}(\omega_{\mathrm{c}},z) = \frac{\varepsilon_0}{2}d_{11}E^{\pm *}(\omega_{\mathrm{s}},z)E^{\mp}(\omega_{\mathrm{p}},z)\exp\left(i(k_{\mathrm{p}}^{\mp}-k_{\mathrm{s}}^{\pm})z\right), \tag{18b}$$

$$P_x^{(2)\pm}(\omega_{\mathrm{p}},z) = \pm i P_y^{(2)\pm}(\omega_{\mathrm{p}},z) = \frac{\varepsilon_0}{2}d_{11}E^{\mp}(\omega_{\mathrm{s}},z)E^{\mp}(\omega_{\mathrm{c}},z)\exp\left(i(k_{\mathrm{s}}^{\mp}+k_{\mathrm{c}}^{\mp})z\right), \tag{18c}$$

where d_{11} depends on a specific component of the second-order nonlinear susceptibility $\chi^{(2)}$. The complete treatment with a depleted pump for the OPA and OPO with associated Manley–Rowe relations will be presented in Jonsson *et al.*[17]

Upon substituting Eq. (18) into Eq. (16), we obtain the coupled amplitude equations describing the evolution of the circularly polarized components of the three waves involved in the OPA process as follows:

$$\frac{dE^{\pm}(\omega_{\mathrm{s}},z)}{dz} = \frac{id_{11}}{4k_{\mathrm{s}}}\left(\frac{\omega_{\mathrm{s}}}{c}\right)^2 E^{\pm *}(\omega_{\mathrm{c}},z)E^{\mp}(\omega_{\mathrm{p}},z) \times \exp\left(i(\Delta k \mp \Delta\alpha)z\right), \tag{19a}$$

$$\frac{dE^{\pm}(\omega_{\mathrm{c}},z)}{dz} = \frac{id_{11}}{4k_{\mathrm{c}}}\left(\frac{\omega_{\mathrm{c}}}{c}\right)^2 E^{\pm *}(\omega_{\mathrm{s}},z)E^{\mp}(\omega_{\mathrm{p}},z) \times \exp\left(i(\Delta k \mp \Delta\alpha)z\right), \tag{19b}$$

$$\frac{dE^{\pm}(\omega_{\mathrm{p}},z)}{dz} = \frac{id_{11}}{4k_{\mathrm{p}}}\left(\frac{\omega_{\mathrm{p}}}{c}\right)^2 E^{\mp}(\omega_{\mathrm{s}},z)E^{\mp}(\omega_{\mathrm{c}},z) \times \exp\left(-i(\Delta k \pm \Delta\alpha)z\right), \tag{19c}$$

where we used the relation $\omega_{\mathrm{p}} = \omega_{\mathrm{s}} + \omega_{\mathrm{c}}$ together with the following definitions:

$$\Delta k = k_{\mathrm{p}} - k_{\mathrm{s}} - k_{\mathrm{c}}, \quad \Delta\alpha = \alpha_{\mathrm{p}} - \alpha_{\mathrm{s}} - \alpha_{\mathrm{c}}. \tag{20}$$

Equation (19) represents two sets of three coupled equations. These sets describe the OPA of LCP and RCP components of the signal inside a chiral nonlinear medium, corresponding to the choice of + and − signs. Each nonlinear process also creates an idler (or conjugate) wave with the same state of polarization. It is important to note that the phase-matching conditions, $\Delta k = \pm\Delta\alpha$, are different for these two nonlinear processes. In contrast, a single phase-matching condition, $\Delta k = 0$, needs to be satisfied when the nonlinear medium is not chiral.

Equation (19) shows that the OPA in a chiral medium depends strongly on the initial SOP of the pump and signal. 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 phase matching requires $\Delta k = \Delta\alpha$, and 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 both the pump and signal beams are launched into the chiral medium with the identical linear SOPs, e.g., both polarized along the x axis. As is evident from Eq. (19), two different OPA processes couple circularly polarized components of these waves, requiring different phase-matching conditions. As it may be difficult to satisfy both of these conditions simultaneously, efficiencies of the two OPA processes would be different in practice, implying that all three waves would have different elliptical SOPs that would also evolve inside the nonlinear chiral medium. The technique of quasi-phase matching has been used with success in chiral liquids for sum-frequency generation and other applications,[18–20] and it may be useful for OPAs as well.

IV. SIMPLIFIED THEORY OF OPAS

Equation (19) can be simplified considerably when the pump beam is so intense initially that its depletion remains negligible along the entire length of the nonlinear crystal. Negligible pump depletion implies that E_p can be treated as a constant in Eq. (19), which reduces to two sets of two linearly coupled equations between $E_s^\pm$ and $E_c^\pm$. Using the notation $E_j(z) = E(\omega_j, z)$ for $j = p, s, c$, one of these sets can be written as

$$\frac{dE_s^+}{dz} = iK_s E_p^- E_c^{+*} \exp\left(i(\Delta k - \Delta\alpha)z\right), \tag{21a}$$

$$\frac{dE_c^{+*}}{dz} = -iK_c^* E_p^{-*} E_s^+ \exp\left(-i(\Delta k - \Delta\alpha)z\right), \tag{21b}$$

where

$$K_s = \frac{d_{11}}{4k_s}\left(\frac{\omega_s}{c}\right)^2, \quad K_c = \frac{d_{11}}{4k_c}\left(\frac{\omega_c}{c}\right)^2. \tag{22}$$

The second set is obtained by interchanging the + and − superscripts in Eq. (21).

Both sets of equations can be solved by writing the solution in the form

$$E_s^\pm = \mathscr{E}_s^\pm \exp\left(i\gamma_s^\pm z\right), \quad E_c^\pm = \mathscr{E}_c^\pm \exp\left(i\gamma_c^\pm z\right), \tag{23}$$

where $\gamma_s^\pm$ and $\gamma_c^\pm$ are the effective propagation constants of the signal and idler waves. Substituting this form into Eq. (21), we obtain a set of four coupled linear equations, which we write in a matrix form as

$$\begin{pmatrix} \gamma_s^+ & -K_s E_p^- & 0 & 0 \\ -K_c E_p^{-*} & \gamma_c^{+*} & 0 & 0 \\ 0 & 0 & \gamma_s^- & -K_s E_p^+ \\ 0 & 0 & -K_c E_p^{+*} & \gamma_c^{-*} \end{pmatrix} \begin{pmatrix} \mathscr{E}_s^+ \\ \mathscr{E}_c^{+*} \\ \mathscr{E}_s^- \\ \mathscr{E}_c^{-*} \end{pmatrix} = \mathbf{0}. \tag{24}$$

In addition, $\gamma_s^\pm$ and $\gamma_c^\pm$ satisfy the relations

$$\gamma_s^\pm = -\gamma_c^{\pm *} + \Delta\aleph_\mp, \tag{25}$$

where $\Delta\aleph_\pm = \Delta k \pm \Delta\alpha$.

Equation (24) shows that a nontrivial solution exists when the determinant of the matrix vanishes. This requirement leads to two quadratic equations as follows:

$$\gamma_s^{+2} - \gamma_s^+ \Delta\aleph_- + K_s K_c |E_p^-|^2 = 0, \tag{26a}$$

$$\gamma_s^{-2} - \gamma_s^- \Delta\aleph_+ + K_s K_c |E_p^+|^2 = 0. \tag{26b}$$

The solutions of the two quadratic equations are

$$\gamma_{s\pm}^+ = \frac{1}{2}\left(\Delta\aleph_- \pm ig_+\right), \tag{27a}$$

$$\gamma_{s\pm}^- = \frac{1}{2}\left(\Delta\aleph_+ \pm ig_-\right), \tag{27b}$$

where the parametric gain for the two OPA processes is defined as

$$g_\pm^2 = \left(4K_s K_c |E_p^\mp|^2\right)^2 - \Delta\aleph_\mp^2. \tag{28}$$

The preceding solution can be used to determine $\mathscr{E}_s^\pm$ and $\mathscr{E}_c^\pm$ using the boundary conditions at the entrance of the crystal at $z = 0$.[9] In the general case where both the signal and idler are launched at $z = 0$ with the electric fields $E_s^\pm(0)$ and $E_c^{\pm *}(0)$, the solution at a distance z is found to be

$$E_s^\pm = \left[\mathscr{E}_{s+}^\pm \exp\left(-g_+ z\right) + \mathscr{E}_{s-}^\pm \exp\left(g_+ z\right)\right] \exp\left(i\Delta\aleph_\mp z\right), \tag{29a}$$

$$E_c^{\pm *} = \left[\mathscr{E}_{c+}^\pm \exp\left(-g_- z\right) + \mathscr{E}_{c-}^\pm \exp\left(g_- z\right)\right] \exp\left(-i\Delta\aleph_\mp z\right), \tag{29b}$$

where

$$\mathscr{E}_{s\pm}^\pm = \pm\frac{1}{2g_+}\left[\left(i\Delta\aleph \mp g_+\right)E_s^\pm(0) - \frac{\omega_c}{\omega_s}\left(\frac{k_c}{k_s}\right)^{1/2} g_0^\mp E_c^{\pm *}(0)\right], \tag{30a}$$

$$\mathscr{E}_{c\pm}^\pm = \mp\frac{1}{2g_-}\left[\left(i\Delta\aleph \pm g_-\right)E_c^{\pm *}(0) - \frac{\omega_c}{\omega_s}\left(\frac{k_s}{k_c}\right)^{1/2} g_0^\mp E_s^\pm(0)\right]. \tag{30b}$$

These expressions simplify considerably when only the pump and signal beams are incident on the nonlinear crystal, and the idler wave is generated inside that crystal. In this situation, we can set $E_c^\pm(0) = 0$ in Eq. (30).

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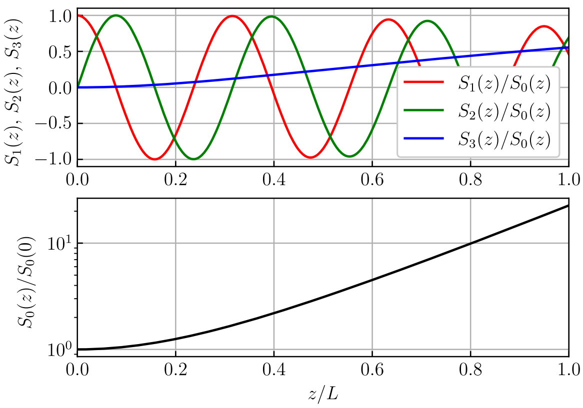

FIG. 1. Evolution of the polarization state and amplification of the signal in the case $|\Delta\alpha| < |\Delta k|$ for the parameter values $4K_sK_c|E^{\mp}(0)|^2L = 2.5$, $\Delta kL = 3$, and $\Delta\alpha L = -1$.

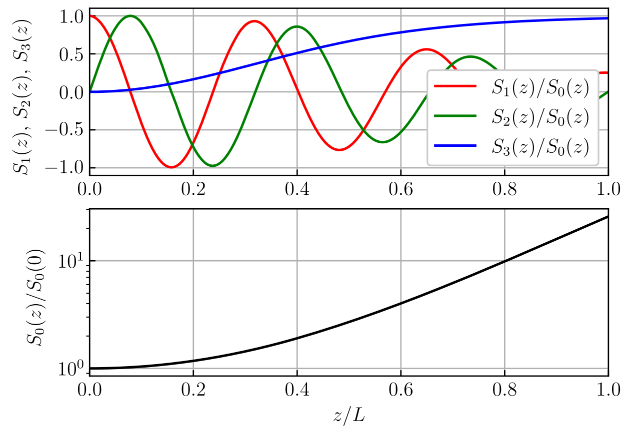

FIG. 3. Evolution of the polarization state and amplification when the chiral contribution to the phase matching matches in magnitude the electric-dipolar phase mismatch: $\Delta kL = -\Delta\alpha L = 3$; other parameters are equal to those used in Fig. 1.

The evolution of the polarization state and the amplification of the signal are conveniently expressed in terms of its Stokes parameters as follows:[21]

$$\begin{aligned} S_0 &= |E_s^+|^2 + |E_s^-|^2, \quad S_3 = |E_s^+|^2 - |E_s^-|^2, \\ S_1 &= 2\mathrm{Re}[E_s^{+*}E_s^-], \quad S_2 = 2\mathrm{Im}[E_s^{+*}E_s^-], \end{aligned} \tag{31}$$

where the elevation S_3/S_0 provides the normalized ellipticity and the azimuth $(S_1, S_2)/S_0$ indicates the axes' orientation of the polarization ellipse.

Figure 1 shows the evolution of the polarization state and the signal's amplification along the OPA's length when the chiral contribution to the phase-matching condition is less than the electric-dipolar term, $|\Delta\alpha| < |\Delta k|$, with the parameter values given there. The corresponding Stokes trajectory is shown in Fig. 2 on the Poincaré sphere. As seen there, the initially linear polarization state $[S_3(0) = 0]$ evolves gradually toward an elliptic one, following a spiral path on the Poincaré sphere. In cases where the sign of the chirality coefficient α is opposite, going from dextrorotation to levorotation, the Stokes trajectory is effectively mirrored in the plane $S_3 = 0$.

A similar behavior occurs when the chiral contribution to phase matching is larger but does not exceed the dipolar contribution. The specific case in which the magnitude of chiral contribution matches the dipolar one ($|\Delta\alpha| = |\Delta k|$) is shown in Figs. 3 and 4. The signal is amplified in nearly the same fashion, but evolution of the polarization state is different. Not only the pitch of the spiral increases on the Poincaré sphere, but the final state also corresponds to a circularly polarized eigenmode.

The evolution of the signal becomes quite different when the chiral contribution significantly exceeds the dipolar contribution to the phase mismatch. An example is shown in Figs. 5 and 6 using $\Delta kL = 3$ and $\Delta\alpha L = -10$. The signal's power no longer increases monotonically and exhibits an oscillatory pattern. In fact, it never exceeds the input level, indicating that parametric amplification

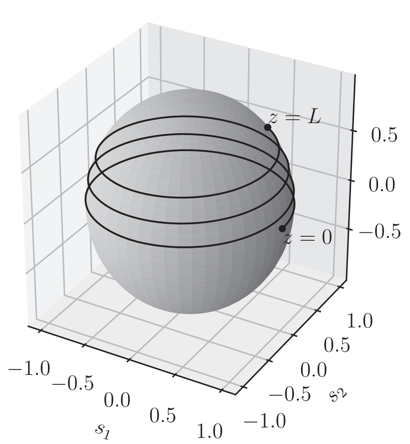

FIG. 2. Evolution of the Stokes parameters shown in Fig. 1 plotted as a trajectory on the Poincaré sphere.

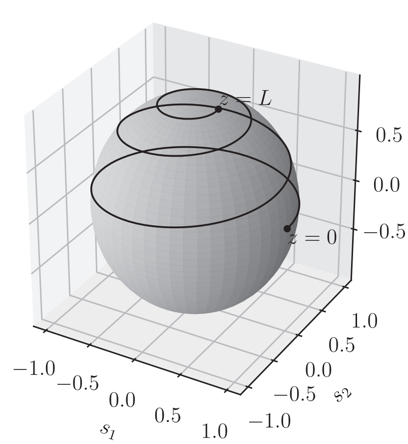

FIG. 4. Evolution of the Stokes parameters shown in Fig. 3 plotted as a trajectory on the Poincaré sphere.

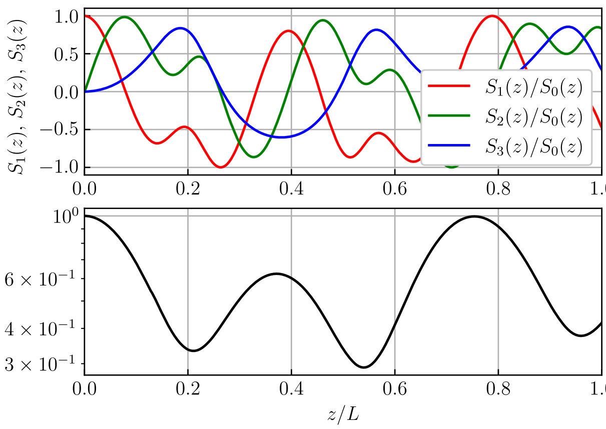

FIG. 5. Evolution of the polarization state and amplification when the chiral contribution to the phase matching exceeds the electric-dipolar contribution: $\Delta kL = 3$ and $\Delta\alpha L = -10$; other parameters are equal to those used in Fig. 1.

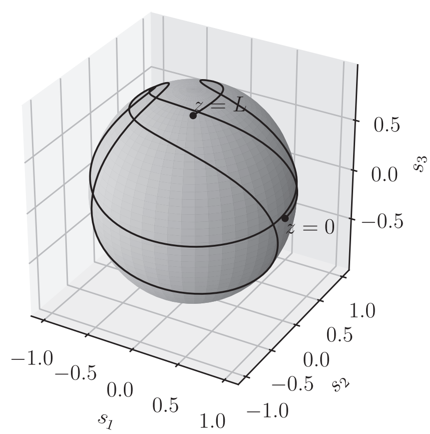

FIG. 6. Evolution of the Stokes parameters shown in Fig. 5 plotted as a trajectory on the Poincaré sphere.

ceases to occur. The Stokes parameters also vary in a zigzag fashion, resulting in a complicated pattern of the polarization state on the Poincaré sphere. These features are due to a periodic transfer of energy back and forth between the signal and idler waves.

V. OPTICAL PARAMETRIC OSCILLATORS

OPOs make use of the optical gain provided by an OPA to create a tunable source at one or two wavelengths that are different from the laser used to pump the OPO.[9,11,12] This is achieved in practice by placing an OPA between two properly designed mirrors forming an optical cavity. In a doubly resonant OPO, mirrors are made transparent at the pump wavelength but are highly reflective at both the signal and idler wavelengths. In contrast, the mirrors reflect only one wavelength (signal or idler) in a singly resonant OPO and are transparent at the other two wavelengths. Since only the pump is incident on the nonlinear crystal, the signal and idler beams build up inside this crystal from noise photons (provided by parametric fluorescence) over multiple round trips inside the cavity.

An important concept in the operation of an OPO is that of its threshold, defined as the minimum pump power needed before which coherent radiation is emitted by the OPO at the signal and/or idler wavelengths. A simple way to estimate this threshold consists of requiring that the optical field remains unchanged after each round trip inside the cavity. In the case of a non-chiral medium and a singly resonant OPO, the signal field after one round trip can be written as

$$E_s(z + 2L) = R \exp\left[2(g - l_i)L + i\phi_r\right] E_s(z), \tag{32}$$

where R is the reflectivity of two mirrors (assumed to be the same), g is the parametric gain inside a nonlinear crystal of length L, l_i is the internal loss of the crystal, and ϕ_r is the phase shift acquired over one round trip. Frequencies of the cavity modes are found by setting $\phi_r = 2m\pi$, where m is an integer. Using the condition $E_s(z + 2L) = E_s(z)$, the threshold is reached when the gain is high enough to satisfy $R \exp[2(g - l_i)L] = 1$ or

$$g = l_i + l_m, \qquad l_m = \frac{1}{2L} \ln(1/R), \tag{33}$$

where l_m is the loss at two mirrors, distributed over the cavity's length. This condition can be used to find the threshold value of the pump power after noting that g scales linearly with the pump power.

The preceding threshold condition can be used for a chiral nonlinear medium as well with one difference. Because of different parametric gains for the LCP and RCP states in a chiral crystal, as indicated in Eq. (28), the threshold conditions are different for these two polarization states and can be written as

$$g_+ = l_{i+} + l_m, \qquad g_- = l_{i-} + l_m, \tag{34}$$

where the internal losses can be different for the two polarization states. Using $g_\pm$ from Eq. (28) with the same internal loss, pump powers needed to reach the threshold are given for the LCP and RCP modes as

$$P_\pm = |E_p^\pm|^2 = \frac{(l_{\text{tot}}^2 + \Delta\aleph_\pm^2)^{1/2}}{4K_s K_c}, \tag{35}$$

where $l_{\text{tot}} = l_i + l_m$ is the total loss of the cavity. It follows that the threshold pump power depends on the phase mismatch and is smaller for the polarization state for which the phase mismatch is smaller. In practice, a chiral OPO will operate in only that circular polarization state (RCP or LCP) for which the phase-matching condition is nearly satisfied. The reason is that once the lower threshold is reached, that particular polarization state builds up exponentially over multiple round trips inside the cavity. As a result, even a small difference in the values of g_+ and g_- suffices to force the circular polarization state with the larger gain to dominate the OPO output.

As is discussed in several textbooks,[9,11,12] the case of a doubly resonant OPO is more complicated. The reason is that such lasers provide output simultaneously at two wavelengths corresponding to the signal and idler waves. It is thus necessary to consider how these two waves build up after each round trip and impose the condition that the electric field does not change after each round trip for both of them. Following the discussion in Ref. 11, the threshold condition for a non-chiral doubly resonant OPO is

$$\cosh(gL) = 1 + \frac{(1 - R_s e^{-l_i L})(1 - R_c e^{-l_i L})}{R_s e^{-l_i L} + R_c e^{-l_i L}}, \qquad (36)$$

where the mirror reflectivity is allowed to be different for the signal and idler waves that are being amplified inside the nonlinear crystal.

We can use Eq. (36) for a chiral OPO as well provided that we replace g with $g_\pm$ for the two circular polarization states. Once again, the polarization state with the largest gain has a lower pump power and will reach the threshold first and will dominate at the OPO output. In practice, the threshold pump power is decreased by a large factor for doubly resonant OPOs. In spite of this advantage, such OPOs are rarely used because singly resonant OPOs are much more stable compared to them.

VI. CONCLUDING REMARKS

In this paper, we have developed the theory behind the nonlinear process of optical parametric amplification inside a chiral crystal. As is well known, chirality makes the nonlinear medium circularly birefringent. We show that circular birefringence, induced by chirality, leads to two different OPA processes with different phase-matching conditions that occur simultaneously and 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, their states of polarization evolve in an elliptical fashion because of the two competing OPA processes taking place in parallel. The idler wave generated during the OPA also becomes elliptically polarized.

We also discuss how the OPA process can be used to make an OPO by placing a chiral nonlinear crystal between two properly designed mirrors forming an optical cavity. We consider both the singly and doubly resonant OPOs and show that they both have different threshold pump powers for the LCP and RCP modes. Our main conclusion is that such OPOs will emit circularly polarized light, no matter how they are pumped. The helicity of the emitted radiation depends on which of the two nonlinear processes, involving the RCP and LCP components of the pump, signal, and idler waves, is nearly phase-matched and thus reaches the threshold first.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Christos Flytzanis: Investigation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Fredrik Jonsson**: Investigation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Govind P. Agrawal**: Investigation (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

REFERENCES

[1] L. M. Haupert and G. J. Simpson, "Chirality in nonlinear optics," Annu. Rev. Phys. Chem. **60**, 345–365 (2009).

[2] P. Lodahl, S. Mahmoodian, S. Stobbe, A. Rauschenbeutel, P. Schneeweiss, J. Volz, H. Pichler, and P. Zoller, "Chiral quantum optics," Nature **541**, 473–480 (2017).

[3] S. Yu, X. Piao, and N. Park, "Chirality in non-Hermitian photonics," Curr. Opt. Photonics **3**, 275–284 (2019).

[4] P. Fischer, A. D. Buckingham, and A. C. Albrecht, "Isotropic second-order nonlinear optical susceptibilities," Phys. Rev. A **64**, 053816 (2001).

[5] M. A. Belkin, S. H. Han, X. Wei, and Y. R. Shen, "Sum-frequency generation in chiral liquids near electronic resonance," Phys. Rev. Lett. **87**, 113001 (2001).

[6] M. A. Belkin, Y. R. Shen, and C. Flytzanis, "Coupled-oscillator model for nonlinear optical activity," Chem. Phys. Lett. **363**, 479–485 (2002).

[7] R. W. Boyd, J. E. Sipe, and P. W. Milonni, "Chirality and polarization effects in nonlinear optics," J. Opt. A: Pure Appl. Opt. **6**, S14–S17 (2004).

[8] J. Vogwell, L. Rego, O. Smirnova, and D. Ayuso, "Ultrafast control over chiral sum-frequency generation," Sci. Adv. **9**, eadj1429 (2023).

[9] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, 1984).

[10] *Quantum Electronics: A Treatise*, edited by H. Rabin and C. L. Tang (Academic Press, New York, 1975), Vol. I–II.

[11] R. W. Boyd, *Nonlinear Optics*, 4th ed. (Wiley, 2020).

[12] J.-M. Liu, *Nonlinear Photonics* (Cambridge University Press, 2022).

[13] J. Fiutak, "The multipole expansion in quantum theory," Can. J. Phys. **41**, 12 (1963).

[14] C. Flytzanis, "Theory of nonlinear optical susceptibilities," in *Quantum Electronics: A Treatise*, edited by H. Rabin and C. L. Tang (Academic Press, New York, 1975), Vol. I, Part A, pp. 9–207.

[15] F. Jonsson and C. Flytzanis, "Photospin-orbit coupling in photonic structures," Phys. Rev. Lett. **97**, 193903 (2006).

[16] P. P. Bey and H. Rabin, "Coupled-wave solution of harmonic generation in an optically active medium," Phys. Rev. **162**, 794 (1967).

[17] F. Jonsson, G. P. Agrawal, and C. Flytzanis, "Optical parametric processes in chiral nonlinear media" (unpublished) (2024).

[18] B. Busson, M. Kauranen, C. Nuckolls, T. J. Katz, and A. Persoons, "Quasi-phase-matching in chiral materials," Phys. Rev. Lett. **84**, 79–82 (2000).

[19] L. Z. Liu, K. O'Keeffe, and S. M. Hooker, "Optical rotation quasi-phase-matching for circularly polarized high harmonic generation," Opt. Lett. **37**, 2415–2417 (2012).

[20] I. C. Khoo, C.-W. Chen, T.-M. Feng, and T.-H. Lin, "Nonlinear liquid crystalline chiral photonic crystals for visible to mid-infrared optical switching and modulation," in *Optica Nonlinear Optics Topical Meeting 2023* (Optica Publishing Group, 2023), p. Tu1B.3.

[21] M. Born and E. Wolf, *Principles of Optics*, 7th ed. (Cambridge University Press, 1999).