New Exact Solutions and Bifurcations in the Spatial Distribution of Polarization in Third-Order Nonlinear Optical Interactions

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By means of the Poincaré-sphere representation, new exact solutions for the evolution of the polarization in anisotropic crystals exhibiting a third-order nonlinearity and subjected to a dc electric field are found. This allows us to discover the existence of bifurcations of the nonlinear eigenpolarizations. A stability analysis of the eigenpolarizations shows how the intrinsic instabilities evolve in the interaction between self-induced and dc-field–induced birefringence.

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An intense electromagnetic wave propagating in a nonlinear medium experiences the effects of a self-induced birefringence.[1] In isotropic media this effect has long been known as "ellipse rotation,"[2] but only recently has there been recognized the existence of unstable nonlinear eigenpolarizations (i.e., waves which propagate unchanged in the nonlinear medium[3]) in the case of anisotropic media.[4] The problem of the interaction of an optical-field–induced birefringence with a dc-field–induced birefringence has also been analytically solved by Sala for an isotropic medium.[5] This is formally equivalent to the consideration of the presence of linear and nonlinear directional coupling between two integrated dielectric waveguides,[6,7] or the interaction of nonlinear and intrinsic birefringence in a single-mode optical fiber.[8,9]

By introducing a formalism based on the Poincaré-sphere representation,[10] we describe the evolution of the polarization in isotropic or anisotropic nonlinear media. New analytical solutions are obtained, and a new type of bifurcations of the nonlinear eigenpolarizations leading to spatial polarization instability is found as a consequence of the competition between linear and nonlinear birefringences.

We consider a monochromatic electric field $\mathbf{E}$ at frequency ω propagating in the z direction, where x,y,z lie along the major axes of the crystal (Fig. 1). We further choose the z axis so that the section with the (x,y) plane of the index ellipsoid is circular. Therefore the crystal has no birefringence, but we allow for a possible anisotropy in the third-order nonlinear susceptibility tensor $\chi^{(3)}_{ijkl}$. The electric field within the transparent crystal can be expressed as

$$\mathbf{E}=\hat{\mathbf{x}}E_1+\hat{\mathbf{y}}E_2=\hat{\mathbf{x}}\mathscr{E}_1\exp(jkz-j\omega t)+\hat{\mathbf{y}}\mathscr{E}_2\exp(jkz-j\omega t), \tag{1}$$

where k is the linear propagation constant. The nonlinear wave equation for third-order effects reduces, in the slowly varying approximation, to

$$-j\,dE_i/dz=kE_i+(2\pi\omega^2/kc^2)P_i^{(3)},\quad i=1,2. \tag{2}$$

For crystals belonging to the symmetry classes 422, $4mm$, $4/mmm$, $42m$ (tetragonal), 622, $6mm$, $6/mmm$, $6m2$ (hexagonal), 432, $43m$, and $m3m$ (cubic),[11] Eq. (2) can be written as

$$-j\,dE_i/dz=kE_i+E_i(\chi_1|E_i|^2+2\chi_2|E_{(3-i)}|^2)+\chi_3E_i^*E_{(3-i)}E_{(3-i)},\quad i=1,2, \tag{3}$$

where

$$\chi_1=(2\pi k_0^2/k)\chi^{(3)}_{1111}(\omega;\omega,\omega,-\omega),$$

$$\chi_2=(2\pi k_0^2/k)\chi^{(3)}_{1212}(\omega;\omega,\omega,-\omega),$$

and

$$\chi_3=(2\pi k_0^2/k)\chi^{(3)}_{1221}(\omega;\omega,\omega,-\omega);\quad k_0^2=\omega^2/c^2,$$

and $\chi^{(3)}_{ijkl}(\omega;\omega,\omega,-\omega)$ is strictly real. For isotropic media,[2] $\chi_3+2\chi_2-\chi_1=0$.

Under the nonlinear transformation of variables

$$S_0=|E_1|^2+|E_2|^2,\quad S_1=|E_1|^2-|E_2|^2,$$
$$S_2=E_1E_2^*+E_1^*E_2,\quad S_3=j(E_1^*E_2-E_1E_2^*), \tag{4}$$

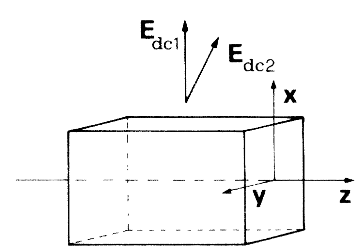

FIG. 1. Anisotropic crystal referred to its principal directions (x,y,z). E_{dc1} and E_{dc2} are oriented along the $\langle 100\rangle$ and $\langle 110\rangle$ directions, respectively.

system (3) can be written in the compact form

$$\dot{S}_0 = 0, \quad \dot{\mathbf{S}} = \boldsymbol{\Omega}_{\mathrm{NL}}(\mathbf{S}) \times \mathbf{S}, \tag{5}$$

where a dot denotes d/dz. The second equation is known as Euler's equation for the angular momentum of a rigid body in free rotation about a fixed point. The motion of the state of polarization as represented by the reduced Stokes vector $\mathbf{S} \equiv (S_1, S_2, S_3)$ is a nonrigid rotation of the Poincaré sphere $S_1^2 + S_2^2 + S_3^2 = S_0^2$ with angular velocity

$$\boldsymbol{\Omega}_{\mathrm{NL}}(\mathbf{S}) \equiv \chi(0, (1-\lambda)S_2, -\lambda S_3,),$$

where $\chi \equiv 2\chi_3$ and $\lambda = (\chi_1 - 2\chi_2 + \chi_3)/2\chi_3$. In the isotropic case we have $\lambda = 1$ and $\boldsymbol{\Omega}_{\mathrm{NL}}(\mathbf{S}) = (0, 0, -\chi S_3)$ (ellipse rotation).

System (5) admits the invariants

$$S_1^2 + S_2^2/\lambda = r_1,$$
$$S_1^2 + S_2^2 + S_3^2 = R^2. \tag{6}$$

The intersections of the above surfaces are the closed trajectories along which the vector $\mathbf{S}$ moves. The nonlinear dynamical system (5) is separable; the motion of S_1 is that of an anharmonic undamped Duffing oscillator,

$$\ddot{S}_1 + \alpha S_1 + \beta S_1^3 = 0, \tag{7}$$

where $\alpha = \chi^2\lambda[R^2 + (1-2\lambda)r_1]$, $\beta = 2\chi^2\lambda(\lambda - 1)$. With no loss of generality, we can choose $\lambda > 1$ (for example for $KTa_xNb_{1-x}O_3$, $KTaO_3$, and $BaTiO_3$,[12] $\lambda = 1.2$–1.3) and Eq. (7) is explicitly solved in terms of Jacobian elliptic functions as

$$S_1(z) = \begin{cases} S_{10}\,\mathrm{cn}(z(\alpha + \beta S_{10}^2)^{1/2}, m), & m^2 = \beta S_{10}^2/2(\alpha + \beta S_{10}^2), \text{ for } E \geq 0, \\ S_{10}\,\mathrm{dn}(z(\beta/2)^{1/2} S_{10}, n), & n^2 = 2(1 + \alpha/\beta S_{10}^2), \text{ for } E < 0, \end{cases} \tag{8}$$

where $E = \frac{1}{2}\chi^2\lambda\Gamma_1(R^2 - \lambda\Gamma_1)$ is the energy of the oscillator and for simplicity we chose the origin on the z axis in such a way that $S_{20} = 0$ results. The parameters S_2 and S_3 are obtained from Eqs. (8) through the invariants (6). The resulting trajectories on the Poincaré sphere are depicted in Fig. 2. System (5) has four stable singular points (centers) located at $D_{1,2} = (\pm R, 0, 0)$ and $D_{3,4} = (0, 0, \pm R)$, and two unstable singular points (saddles) at $D_{5,6} = (0, \pm R, 0)$.[4] Whenever two light beams counterpropagate in the crystal, (5) modifies into the new conservative system

$$\begin{aligned} &\dot{S}_0 = 0, \quad \dot{\mathbf{S}} = [\boldsymbol{\Omega}_{\mathrm{NL}}(\mathbf{S}) + \boldsymbol{\Omega}'_{\mathrm{NL}}(\mathbf{W})] \times \mathbf{S}, \\ &\dot{W}_0 = 0, \quad -\dot{\mathbf{W}} = [\boldsymbol{\Omega}_{\mathrm{NL}}(\mathbf{W}) + \boldsymbol{\Omega}'_{\mathrm{NL}}(\mathbf{S})] \times \mathbf{W}, \end{aligned} \tag{9}$$

where $W_0, \mathbf{W}$ are associated with the field propagating in the $-z$ direction through (4) and $\boldsymbol{\Omega}'_{\mathrm{NL}}(\mathbf{W}) \equiv ([\chi' + \chi(2\lambda - 1)]W_1,\ (\chi' + \chi)W_2,\ (\chi' - \chi)W_3)$, and $\chi' \equiv 2\chi_2$. In the isotropic case, (9) admits four independent invariants[13] and no chaotic motion is possible;[14] however, symmetry-breaking bifurcations and unstable polarization eigenarrangements exist.[14] In the anisotropic case, chaotic behavior in the solutions of (9) has been reported.[4]

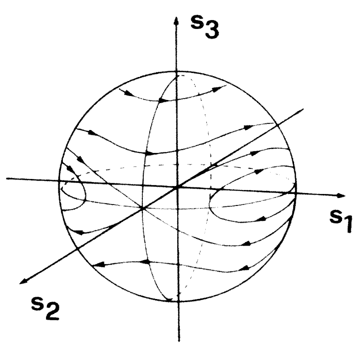

FIG. 2. Trajectories of the polarization state on the Poincaré sphere in an anisotropic nonlinear crystal.

If we consider centrosymmetric crystals (symmetry classes $4/mmm$, $6/mmm$, or $m3m$), in the presence of a static field the quadratic electro-optic effect modifies system (5) to

$$\dot{S}_0 = 0, \quad \dot{\mathbf{S}} = \boldsymbol{\Omega}(\mathbf{S}) \times \mathbf{S} = [\boldsymbol{\Omega}_L + \boldsymbol{\Omega}_{\mathrm{NL}}(\mathbf{S})] \times \mathbf{S}. \tag{10}$$

If, for example, a dc electric field of amplitudes $E_{\mathrm{dc}1}$ is directed along the $\langle 100 \rangle$ direction, then $\boldsymbol{\Omega}_L = (\eta_1, 0, 0)$ and $\eta_1 = E_{\mathrm{dc}1}^2[\chi_{1111}^{(3)}(\omega;\omega,0,0) - \chi_{1122}^{(3)}(\omega;\omega,0,0)]$. System (10) is integrable as follows. The invariants of the motion are

$$S_1^2 + S_2^2/\lambda + 2\eta_1 S_1/\chi\lambda = \Gamma_2,$$
$$S_1^2 + S_2^2 + S_3^2 = R^2, \tag{11}$$

and S_1 satisfies the equation

$$\ddot{S}_1 + \tau + (\alpha' + \delta)S_1 + \sigma S_1^2 + \beta S_1^3 = 0, \tag{12}$$

where

$$\alpha' = \chi^2\lambda[R^2 + (1-2\lambda)\Gamma_2], \quad \tau = \chi\eta_1(R^2 - 2\Gamma_2\lambda),$$
$$\delta = 4\eta_1^2, \quad \sigma = 3\eta_1\chi(2\lambda - 1).$$

The solution of Eq. (12) can be expressed through the elliptic integral

$$z = (1/\sqrt{2})\int_{S_{10}}^{S_1} dx/[Q(x)]^{1/2}, \tag{13}$$

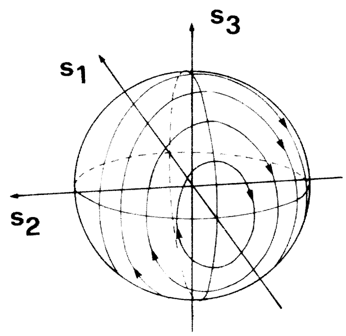

FIG. 3. Trajectories of the polarization in an anisotropic nonlinear crystal subjected to an external dc field $E_{\rm dc1}$ when $t_1 > R$.

where

$$Q(x) = E' - \tau x - \tfrac{1}{2}(\alpha^2+\delta)x^2 - \sigma x^3/3 - \beta x^4/4,$$

$$E' = \tfrac{1}{2}\chi^2\lambda\Gamma_2(R^2-\lambda\Gamma_2).$$

A stability analysis of the singular points of system (10) reveals how the interaction between the dc- and optical-field–induced birefringence affects the topological properties of the polarization trajectories. The eigenpolarizations $\hat{\mathbf{S}}$ are defined by $\mathbf{\Omega}(\hat{\mathbf{S}})\times\hat{\mathbf{S}}=0$, or explicitly

$$\hat{S}_2\hat{S}_3=0, \quad \hat{S}_1\hat{S}_3+t_1\hat{S}_3=0, \qquad (14)$$
$$\hat{S}_1\hat{S}_2+t_2\hat{S}_2=0.$$

We introduced the two bifurcation parameters $t_1 = \eta_1/(\chi\lambda)$ and $t_2=\eta_1/\chi(\lambda-1)$, $t_2>t_1$. Whenever $t_1>R$, system (10) admits the two stable singular points (centers) $P_{1,2}=(\pm R,0,0)$ (see Fig. 3). In this situation the dc-field–induced birefringence dominates, in the sense that the trajectories on the Poincaré sphere are topologically equivalent to those of a rigid rotation around S_1. If the intensity of the optical field (or the nonlinearity χ) is increased up to the point where $t_1 \leq R$, the point P_2 bifurcates into an unstable saddle point at $(-R,0,0)$ and two centers at $P_{3,4}=(-t_1, 0, \pm(R^2-t_2^2)^{1/2})$ (see Fig. 4). A trajectory of unperiodic motion (separatrix) determines a partition of the Poincaré sphere into three domains of periodic motion. Suppose that the input optical field polarization is alternately switched between two trajectories arbitrarily close to the separatrix, but situated in two different domains. Then after some finite propagation length the field will emerge in two nearly orthogonal polarization states. If the polarization state of an intense pump beam (located near the separatrix) is perturbed through coherent superposition with a weak signal beam, by placing at the crystal output an analyzer for circular polarizations, we obtain in principle (choosing the proper interaction length) a complete intensity modulation of the strong wave. This effect leads to conception of a new class of all-optical devices, such as coherent small-signal amplifiers and phase-sensitive switches or discriminators.[9]

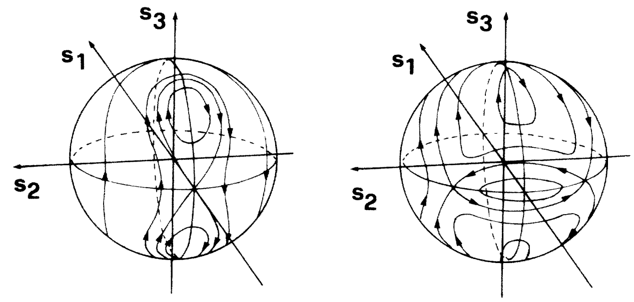

FIG. 4. Same as in Fig. 3, with $t_1 < R$ (left) and $t_2 < R$ (right).

If also $t_2 \leq R$, point P_2 bifurcates again into the stable center at $(-R,0,0)$ and two saddles at $P_{5,6} = (-t_2, \pm(R^2-t_2^2)^{1/2}, 0)$ (see Fig. 4). With further increase in the wave intensity or χ, the self-induced effects dominate and the trajectories in Fig. 4 approach those of Fig. 2 (with no applied dc field).

The observable features of the bifurcated solutions are conveniently described by considering the evolution of the components of $\mathbf{S}$. As shown in (12), S_1 moves in a quartic potential well: Each trajectory on the sphere fixes the energy E' and the shape of the potential. The bifurcated trajectories contained in a certain domain on the sphere correspond to oscillations in the same dimple of the well for S_1.[7] The information on the polarization variations along z provided by the orbits in Figs. 2–4 is usefully complemented by the knowledge of the orbital periods. Figure 5 shows the distribution of periods for the cases $t_1=2R$ (see Fig. 3) and $2t_2=R$ (see Fig. 4): The trajectories are determined by the choice of Γ_2 or, equivalently, the initial conditions $\mathbf{S}(0)=(S_{10}, 0, \pm(R^2-S_{10}^2)^{1/2})$. Note

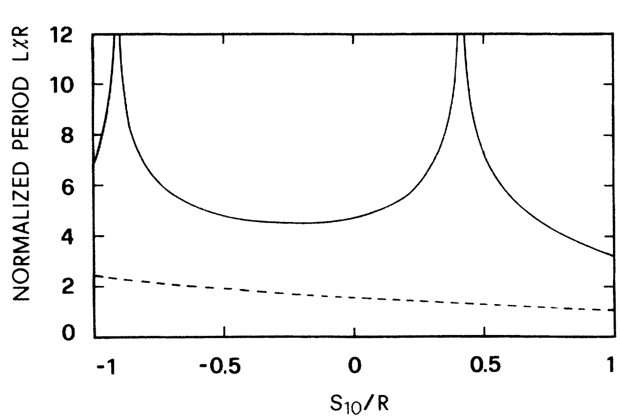

FIG. 5. Normalized period $L\chi R$ of the orbits on the Poincaré sphere vs the initial condition S_{10}/R ($S_{20}=0$) for $2R=t_1$ (dashed line) and $2t_2=R$ (solid line).

how the period approaches infinity in proximity of the separatrices.

The case of an electric field E_{dc2} applied along the crystal direction $\langle 110 \rangle$ can be examined by a similar procedure. Equation (10) holds with $\mathbf{\Omega}_L = (0, \eta_2, 0)$, where $\eta_2 = 2E_{dc2}^2 \chi_{1221}^{(3)}(\omega;\omega,0,0)$.

We limit ourselves here to a discussion of the stability of the eigenpolarizations. The singular points of the system (10) are defined by

$$\hat{S}_2\hat{S}_3 + u_1\hat{S}_3 = 0, \quad \hat{S}_1\hat{S}_3 = 0, \qquad \hat{S}_1\hat{S}_2 - u_2\hat{S}_1 = 0. \tag{15}$$

The bifurcation parameters are $u_1 = \eta_2/\chi$ and $u_2 = \eta_2/\chi(\lambda - 1)$ (for $\lambda < 2$, $u_1 < u_2$). If $u_1 > R$, the singular points are given by the two centers $Q_{1,2} = (0, \pm R, 0)$. When $u_1 \leq R$, Q_2 bifurcates into a saddle at $(0, -R, 0)$ and two centers at $(0, -u_1, \pm(R^2 - u_1^2)^{1/2})$. Finally, if $u_2 \leq R$, then also Q_1 bifurcates into a saddle at $(0, R, 0)$ and two centers at $(\pm(R^2 - u_2^2)^{1/2}, u_2, 0)$.

The approach based on the Poincare sphere to analyze the self-induced nonlinearities is powerful since it reduces the solution of the nonlinear wave equation to solving a one-dimensional anharmonic oscillator [Eq. (7) or Eq. (11)] which can be easily integrated. Furthermore the display of the trajectories provided by the sphere conveys all the amplitude and phase information on the evolution and stability of the optical field in the nonlinear medium in a very immediate way.

The prediction of bifurcations and instability phenomena of the kind described in this Letter can be extended to include nonlinear media with a higher degree of anisotropy,[15] such as birefringent or optically active crystals. The effects of material inhomogeneities and of a finite source bandwidth are the subject of further investigation.

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