Frustrated Optical Instability: Self-Induced Periodic and Chaotic Spatial Distribution of Polarization in Nonlinear Optical Media

Junji Yumoto and Kenju Otsuka

Musashino Electrical Communication Laboratory, Nippon Telegraph and Telephone Public Corporation, Musashino-shi, 180 Tokyo, Japan

(Received 21 January 1985)

The universal nature of self-induced polarization rotation in nonlinear optical media having a third-order nonlinearity is depicted in terms of phase-space trajectories characterized by separatrix orbits. The existence of nonlinear eigenpolarization, and of periodic synchronized spatial as well as chaotic spatial distributions of the polarization of light, are found in the mutual interaction of counterpropagating laser beams (collinear degenerate four-wave mixing geometry). Such spatial polarization turbulences cause a frustrated optical instability in input- versus output-intensity characteristics, when a fixed output polarization is selected.

PACS numbers: 42.65.Bp, 05.40.+j

Maker, Terhune, and Savage predicted that the axes of the vibrational ellipse for elliptically polarized light rotate as a function of distance in isotropic centrosymmetric media having an intensity-dependent refractive index.[1] Nonlinear eigenpolarization has been predicted to exist for interfering beams (collinear degenerate four-wave mixing geometry) in isotropic materials by Kaplan.[2] He briefly mentioned the possibility of spatial polarization instabilities without theoretical demonstration. In this Letter, we extend these works to include anisotropic crystals and provide new views into the nonlinear theory of wave propagation, in particular spatial polarization instability.

Nonlinear eigenpolarization, the self-induced polarization-state change for linearly and elliptically polarized monochromatic light, as well as symmetry-breaking instability, will be shown by use of the universal phase-space trajectory for a beam characterized as having a separatrix orbit in phase space. This system is explained quite well by nonlinear mechanical pendulum models (nonlinear optical pendulum equation).

Nonlinear eigenpolarization and periodic synchronized spatial distributions of polarization will be shown to exist when the mutual interaction of counterpropagating laser beams exists in anisotropic crystals. The chaotic spatial distribution of polarization is found when two beams are not identical in amplitudes. Time-domain self-oscillations and optical chaos have been predicted by Silberberg and Joseph for a similar interfering-beam scheme in a third-order nonlinear isotropic medium under the limitation of transit time being comparable to the medium response time.[3] Up to now, however, optical chaos has been mostly restricted to the time-domain turbulence in dissipative systems which are far from thermal equilibrium.

The conceptual model of the system is detailed in Fig. 1. Assume that there is a forward electric field $\mathbf{E}_1$ and a backward electric field $\mathbf{E}_2$ propagating along x with the same frequency ω, where $x,y,$ and z lie along the major axes of the anisotropic crystal. If we set

$$\mathbf{E}_i = E_{iy}\exp\{(-1)^i jk_y x - j\phi_{iy}\}\hat{\mathbf{y}} + E_{iz}\exp\{(-1)^i jk_z x - j\phi_{iz}\}\hat{\mathbf{z}}, \tag{1}$$

where $i=1,2$ and $j^2=-1$, the total field $\mathbf{E}$ (spatial part) in the anisotropic medium is expressed as

$$\mathbf{E} = E_y\hat{\mathbf{y}} + E_z\hat{\mathbf{z}} = \textstyle\sum_i E_{iy}\exp\{(-1)^i jk_y x - j\phi_{iy}\}\hat{\mathbf{y}} + \sum_i E_{iz}\exp\{(-1)^i jk_z x - j\phi_{iz}\}\hat{\mathbf{z}}. \tag{2}$$

Here, E_{iy} and E_{iz} are amplitudes of E_i along the y and z directions, k_y and k_z are linear propagation constants, and ϕ_{iy} and ϕ_{iz} are nonlinear phase terms. The nonlinear components of the polarization along the y and z directions are expressed as

$$P_y^{NL} = \epsilon_0\{\chi_{yyyy}E_yE_yE_y^* + \chi_{yyzz}E_yE_zE_z^* + \chi_{yzyz}E_zE_yE_z^* + \chi_{yzzy}E_zE_zE_y^*\}, \tag{3a}$$

$$P_z^{NL} = \epsilon_0\{\chi_{zzzz}E_zE_zE_z^* + \chi_{zzyy}E_zE_yE_y^* + \chi_{zyzy}E_yE_zE_y^* + \chi_{zyyz}E_yE_yE_z^*\}, \tag{3b}$$

with the assumption that the frequency arguments of $\chi^{(3)}$ are $(-\omega,\omega,\omega,-\omega)$. These expressions are valid for anisotropic crystals which belong to the cubic, tetragonal, hexagonal, and orthorhombic symmetry groups.[4] For anisotropic crystals belonging to other symmetry groups, other small but nonvanishing $\chi^{(3)}$ tensor components exist. Additionally, a much more complicated nonlinearity is expected.

In the following analysis, we employ a cubic-symmetry crystal, which does not have a linear birefringence, and each component of $\chi^{(3)}$ satisfies the following equations[4]:

$$k_y = k_z \equiv k, \quad \chi_{yyyy} = \chi_{zzzz} \equiv (k/k_0^2)\chi_1, \quad \chi_{yyzz} = \chi_{yzyz} = \chi_{zzyy} = \chi_{zyzy} \equiv (k/k_0^2)\chi_2, \quad \chi_{yzzy} = \chi_{zyyz} \equiv (k/k_0^2)\chi_3.$$

With the assumption that the crystal is lossless and the time response of $\chi^{(3)}$ is so fast that the time-varying terms of Debye's equation are neglected, the propagation characteristics of light are described by Maxwell's equations.

We also assume here that the medium response time τ is much shorter than the light transit time through the medium, i.e., $\tau << L/c$ (L is crystal length and c is velocity of light). In this regime, the light intensity threshold for the buildup of time-domain oscillations becomes extremely high.[3] Therefore, we restrict the analysis below to the steady state. With use of the slowly varying envelope approximation, the following coupled nonlinear differential equations are obtained for the steady state:

$$dE_{iy}/dx = \chi_2 E_{iz} E_{(3-i)y} E_{(3-i)z} \sin(\phi_1+\phi_2) + \tfrac{1}{2}\chi_3 E_{iy} E_{iz}^2 \sin 2\phi_i + (-1)^{i-1}\chi_3 E_{iz} E_{(3-i)y} E_{(3-i)z} \sin(\phi_1-\phi_2), \tag{4a}$$

$$dE_{iz}/dx = -\chi_2 E_{iy} E_{(3-i)y} E_{(3-i)z} \sin(\phi_1+\phi_2) - \tfrac{1}{2}\chi_3 E_{iy}^2 E_{iz} \sin 2\phi_i - (-1)^{i-1}\chi_3 E_{iy} E_{(3-i)y} E_{(3-i)z} \sin(\phi_1-\phi_2), \tag{4b}$$

$$d\phi_i/dx = \tfrac{1}{2}\chi_1\{E_i'^2 + 2E_{(3-i)}'^2\} - \chi_2\{E_i'^2 + E_{(3-i)}'^2 - (E_{iz}/E_{iy} - E_{iy}/E_{iz})E_{(3-i)y}E_{(3-i)z}\cos(\phi_1+\phi_2)\} - \tfrac{1}{2}\chi_3\{E_i'^2\cos 2\phi_i - 2(E_{iz}/E_{iy} - E_{iy}/E_{iz})E_{(3-i)y}E_{(3-i)z}\cos(\phi_1-\phi_2)\}, \tag{4c}$$

$$E_i' = E_{iy}^2 - E_{iz}^2, \quad \phi_i = \phi_{iy} - \phi_{iz}, \quad i = 1,2.$$

Here, we also consider the energy interaction between $\mathbf{E}_1$ and $\mathbf{E}_2$. From Eqs. (4a) and (4b), we obtain

$$\frac{d\mathbf{E}_i \cdot \mathbf{E}_i^*}{dx} = \frac{d}{dx}(E_{iy}^2 + E_{iz}^2) = 2E_{iy}\frac{d}{dx}E_{iy} + 2E_{iz}\frac{d}{dx}E_{iz} = 0, \quad i = 1,2.$$

Therefore, the energy exchange between $\mathbf{E}_1$ and $\mathbf{E}_2$ does not occur.

Let us first analyze a single beam only. In this case, from Eqs. (4a)–(4c), we obtain

$$d\theta_1/dx = -\tfrac{1}{4}\chi_3 E_1^2 \sin 2\theta_1 \sin 2\phi_1, \tag{5a}$$

$$d\phi_1/dx = \tfrac{1}{2}E_1^2 \cos 2\theta_1(\chi_1 - 2\chi_2 - \chi_3\cos 2\phi_1), \tag{5b}$$

$$\tan\theta_1 = E_{1z}/E_{1y}.$$

In this respect, let us examine the properties of the solutions of Eqs. (5a) and (5b) in the phase-space trajectory (θ_1, ϕ_1). In particular, Fig. 2(a) shows the phase-space trajectory when a centrosymmetric cubic crystal, $KTa_{0.65}Nb_{0.35}O_3$ (KTN), is assumed, where $\chi_2/\chi_1 = -0.179$ and $\chi_3/\chi_1 = 1.081$ ($\chi_1 = 0.452\times10^{-22}$ V^2/m^2 and $g_{11} = 0.136$ m^4/C^2).[5] It is clear from the figure that the present system has two kinds of singular points, centers (C) and saddles (S). The centers correspond to the circular polarization, which does not change its state during the propagation since phase velocity $v = 0$, where

$$v = [(d\theta_1/dx)^2 + (d\phi_1/dx)^2]^{1/2}.$$

Therefore, circular propagation is determined to be one of the nonlinear eigenpolarizations.

The other nonlinear eigenpolarizations are found to be linearly polarized light having $\theta = 0°$, $\pm45°$, and $\pm90°$. However, the linearly polarized light having $\theta = \pm45°$ locates at the unstable saddles (S), which are the cross points of the separatrices. This means that a slight fluctuation in time from the values of $\theta = \pm45°$ and $\phi = 0°$ (homoclinic point, potential maxima) results in a symmetry-breaking instability. Such polarization instabilities are also brought about by random noise in the stochastic layer[6] surrounding the separatrix orbit. These behaviors are very much analogous to the motion of a pendulum which is also expressed by a separatrix.[7] For an isotropic material,

$$\chi_1 - 2\chi_2 - \chi_3 = 0,$$

and then the separatrix disappears as shown in Fig. 2(b).

Let us next consider the case of two counterpropagating beams having the same frequency. Nonlinear eigenpolarizations are found to exist even in this case. These are obtained by setting

$$dE_{iy}/dx = dE_{iz}/dx = 0, \tag{6a}$$

$$d\phi_i/dx = 0. \tag{6b}$$

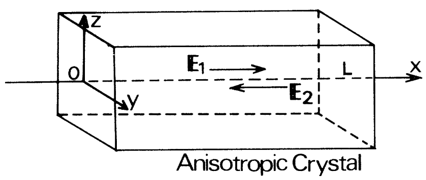

FIG. 1. Conceptual model of the system.

From Eq. (6a), there are at least three cases which satisfy the nonlinear eigenpolarization condition. These are (i) $\phi_1=\phi_2=0$; (ii) $\phi_1=\phi_2=\pm\pi/4$; (iii) $\phi_1=-\phi_2=\pm\pi/4$. In the case of (i), we obtain the following equations from Eqs. (6a) and (6b):

$$8k_1k_2B=\{-4(k_1^2+k_2^2)+(k_1+k_2)^2\pm[3(k_1+k_2)^2(3k_1-k_2)(k_1-3k_2)]^{1/2}\}A, \tag{7a}$$

$$\left(\frac{E_2}{E_1}\right)^2=\frac{-(k_1+k_2)a(1-a^2)(1+b^2)}{2\{k_1a(1-b^2)+k_2(1-a^2)b\}(1+a^2)}, \tag{7b}$$

where

$$A=\frac{\tan\theta_1}{1-\tan^2\theta_1},\quad B=\frac{\tan\theta_2}{1-\tan^2\theta_2},$$

$$k_1=\chi_1-\chi_2,\quad k_2=-\chi_2-\chi_3,$$

$$a=\tan\theta_1,\quad b=\tan\theta_2,\quad \theta_1,\theta_2\neq\pm\tfrac{1}{4}\pi.$$

From Eqs. (7a) and (7b), θ_1,θ_2, and E_2/E_1 are determined for the nonlinear eigenpolarizations. For example, with the assumption that θ_1 equals 40°, θ_2 and E_2/E_1 must satisfy −52.41° and 0.466, or −48.35° and 2.154, respectively. Linearly polarized beams having their arbitrary amplitude ratio (E_2/E_1) either parallel ($\theta_1=\theta_2$) or orthogonal to each other ($\theta_1=\theta_2\pm\pi/2$) have been reported to correspond to the nonlinear eigenpolarizations for the isotropic materials.[2] When θ_1 and θ_2 equal $\pm\pi/4$, Eqs. (6a) and (6b) are also satisfied. Then, in the case of (ii) and (iii), Eq. (6b) is satisfied only when $\theta_i=\pm45°$. That is, when $\mathbf{E}_1$ and $\mathbf{E}_2$ are corotating or counterrotating circularly polarized light, they satisfy the nonlinear eigenpolarization conditions. These polarization configurations have also been shown to be the nonlinear eigenpolarizations for isotropic crystals.[2]

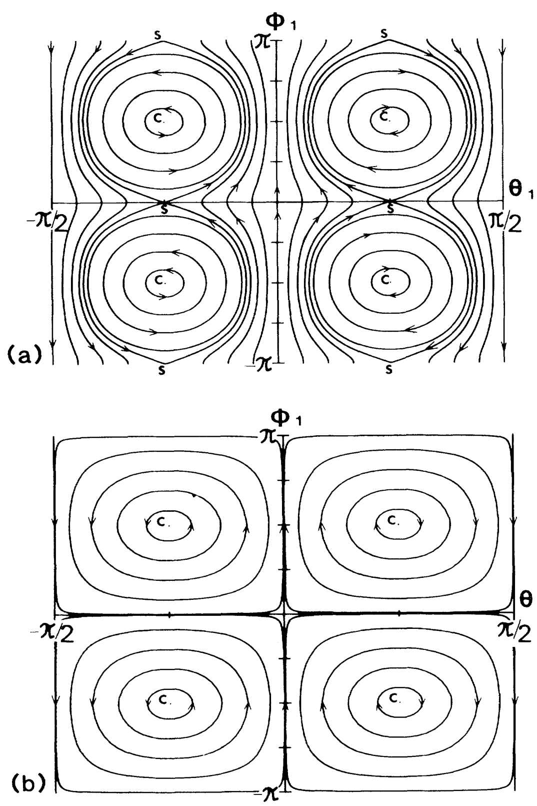

FIG. 2. Phase space trajectory. (a) anisotropic crystal (KTN); (b) isotropic material.

When two beams have the same polarization at any spatial point, the polarization states of both beams are synchronized during the propagation by following essentially the same trajectory as in Fig. 2(a).

When the polarization states or intensities of two incoming beams are not identical, the cylindrical phase trajectory[7] in Fig. 2(a) is destroyed and the chaotic spatial distribution of polarization is brought about as shown in Fig. 3, where $L=2$ and $E_1=E_2$. In order to assess the spatial optical chaotic nature of the solution with some confidence, we examined Poincaré maps as shown in Fig. 4, where $L=2000$. These maps were obtained by collecting (θ_1,ϕ_1) whenever the trajectory crossed the constant plane ($\phi_2=2n\pi$, $n=0,\pm1,\pm2,\ldots$). For $E_1=E_2$, the individual points are found to fall in a closed loop, suggesting that the long-term spatial regularity still remains, even in the chaotic regime. As θ_1 approaches θ_2, the closed loop shrinks. When $\theta_1=\theta_2$, the closed loop becomes a fixed point, which corresponds to the synchronized state. When $E_1\neq E_2$, the width of the loop widens, and the spatial chaotic nature in the polarization of light is pronounced. These phenomena may be interpreted by

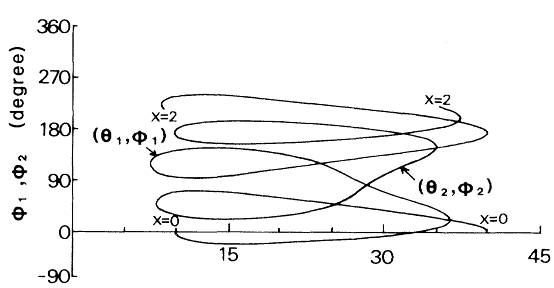

FIG. 3. Chaotic spatial distribution of the polarization in the phase space (θ_i,ϕ_i), where $E_1=E_2=5$, $\theta_1=10°$, $\theta_2=40°$ and $\phi_1=\phi_2=0°$ at $x=0$.

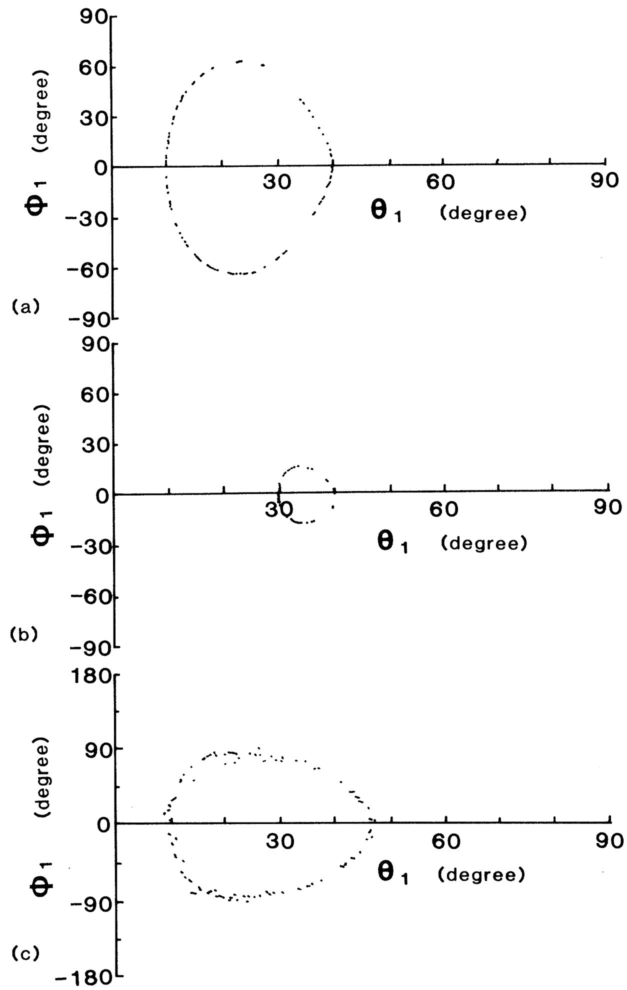

FIG. 4. Poincaré maps ($L=2000$). (a) $E_1=E_2=5$, $\theta_1=10°$, $\theta_2=40°$, $\phi_1=\phi_2=0°$ at $x=0$; (b) $E_1=E_2=5$, $\theta_1=30°$, $\theta_2=40°$, $\phi_1=\phi_2=0°$; (c) $E_1=3$, $E_2=5$, $\theta_1=10°$, $\theta_2=40°$, $\phi_1=\phi_2=0°$.

the motion of two pendulums coupled nonlinearly to each other.[7]

As a consequence, if we look at either of the two beams in the chaotic regime, the polarization state of the outgoing beam changes unpredictably for a slightly different initial condition. This means that the output intensity through a polarizer is frustrated by a change either in the polarization state or in the intensity of the incoming beam (frustrated optical instability). Such spatial optical turbulenece of light as predicted in this Letter is interesting in the sense that it represents a new example of deterministic instability in a conservative dynamic system.

Finally, let us briefly describe the experimental possibility of observing such spatial polarization turbulences. If we assume a KTN crystal, $E=1$ corresponds to the power density of 3.4 GW/cm^2, which can be readily attained with a Q-switched yttrium aluminum garnet laser, where $L=1$ stands for the crystal length of 1 cm.

The authors wish to gratefully acknowledge the discussion with Dr. Yoji Aizawa of Kyoto University. They are also indebted to Dr. Hiroshi Kanbe and Dr. Hitoshi Kawaguchi for their comments.

[1]P. D. Maker, R. W. Terhune, and C. M. Savage, Phys. Rev. Lett. **12**, 507 (1964).

[2]A. E. Kaplan, Opt. Lett. **8**, 560 (1983).

[3]Y. Silberberg and I. Bar Joseph, Phys. Rev. Lett. **48**, 1541 (1982).

[4]M. D. Levenson and J. J. Song, in *Coherent Nonlinear Optics,* edited by M. S. Field and V. S. Letokhov, Topics in Currents Physics Vol. 21 (Springer-Verlag, New York, 1980).

[5]J. E. Geusic, S. K. Kurtz, L. G. Van Uitert, and S. H. Wemple, Appl. Phys. Lett. **4**, 141 (1964).

[6]T. Y. Petrosky, Phys. Rev. A **29**, 2078 (1984).

[7]N. Minorsky, *Nonlinear Oscillations* (Van Nostrand, New York, 1962).