Polarization instability and multistability in nonlinear optics

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Temporal polarization instability is seen as an oscillatory or random variation in time of the polarization parameters of light interacting with a nonlinear system. Spatial polarization instability is the formation of "frozen" complex quasiperiodic or pseudochaotic distributions of the polarization parameters of a wave along the direction of propagation. The problem of polarization instability or multistability is intimately related to the polarization of eigenwaves in the nonlinear problem and their transformation as a result of "hard" and "soft" spontaneous polarization symmetry breaking. This paper presents a review of publications on polarization instabilities in passive nonlinear optical systems, including Fabry-Perot resonators, gyrotropic media, systems with strong two-photon absorption, birefringent crystals, fiber lightguides, and isotropic media.

1. INTRODUCTION. SPATIAL AND TEMPORAL POLARIZATION INSTABILITY, NONLINEAR EIGENPOLARIZATIONS, "HARD" AND "SOFT" SPONTANEOUS POLARIZATION SYMMETRY BREAKING

In low-intensity optics (linear optics), the polarization of an electromagnetic wave in a medium is independent of the intensity of light and is uniquely related to the polarization of the radiation incident on the separation boundary between vacuum and the medium under investigation. Waves of a particular polarization type that corresponds to the type of symmetry of the medium (eigenwaves) retain their polarization parameters. For example, in an isotropic optically active liquid (unracemized solution of chiral molecules), the eigenwaves are circularly polarized. In crystals, eigenwaves are, in general, elliptically polarized, although in the simple special case of nongyrotropic uniaxial crystals they are orthogonally polarized (the so-called O and E waves). In dissipative media exhibiting linear or circular dichroism, arbitrarily polarized waves tend to one of the eigenwaves of the medium as they propagate through it. The very important point is that the "output" polarization of light has no discontinuities. The derivatives of polarization parameters of the emerging wave with respect to the polarization parameters of the incident radiation do not exhibit discontinuities either.

In nonlinear optics, the situation is radically different and both the refractive index and the absorption coefficient of the medium are functions of the radiation intensity.

The following specifically nonlinear polarization effects have attracted increasing attention in recent years:

(a) polarization multistability and temporal polarization chaos. When the "input" intensity or polarization of light undergoes an adiabatic change, the "output" polarization is found to be a multivalued function containing both stable and unstable branches, i.e., the physical state of polarization depends on the "prehistory" of the system (this is the so-called polarization hysteresis). For a particular combination of the parameters of radiation and of the nonlinear medium, and steady-state "input" radiation, the "output" polarization exhibits a self-oscillatory or pseudochaotic variation of polarization with time, which has a continuous frequency spectrum (Fig. 1a).

(b) Polarization multistability and spatial polarization chaos. Here, we are concerned with the quasisteady-state case, i.e., the "freezing" in time of the distribution of polarization parameters along the light beam (light rays). In complete analogy with the temporal case, there are different possible stationary stable distributions in space (the analog of the temporal multistability), an oscillatory variation in the polarization parameters along the ray (analog of self-oscillatory solutions), and a chaotic polarization distribution (with a continuous spectrum of spatial frequencies). In the last case, the polarization parameters of the output radiation leaving the nonlinear medium are found to depend strongly and "unpredictably" on the initial conditions. This is described by the phrase *frustrated polarization instability* (Fig. 1b).

The formal subdivision into multistability and temporal and spatial instability is very arbitrary in the case of real systems. Nevertheless, in many cases, particular polarization-unstable systems can be justifiably assigned to one or the other class of objects. For example, when the temporal polarization instability has a local character, due to the interaction of light with an individual molecular oscillator, we have pure temporal instability.

Analyses of temporal and spatial polarization instabilities often resort to the concept of *spontaneous polarization symmetry breaking*. We shall distinguish two cases, namely, "hard" and "soft" spontaneous symmetry breaking. When the parameters of a nonlinear optical system or the intensity of the light wave reaches a certain definite (threshold) value, the linear polarization of the light wave *switches* to right- and left-handed elliptic polarization as a result of the development of fluctuations. Under ideal conditions, and for isotropic directions, this polarization switching occurs with equal probability to left- and right-handed states when the experiment is repeated a large number of times. This reflects the property of inversion symmetry of space when electromagnetic interactions are taken into account (P-invariance).[1)] We shall refer to this case as *hard*. Examples of hard spontaneous symmetry breaking are well known in mechanics. This property is exhibited by the simple pendulum in the gravitational field, when its point of suspension executes high-frequency horizontal oscillations,[1] or by the strain of a rod with a rectangular cross section, compressed in the longitudinal direction[2] (Fig. 2). In an optical system with *soft*

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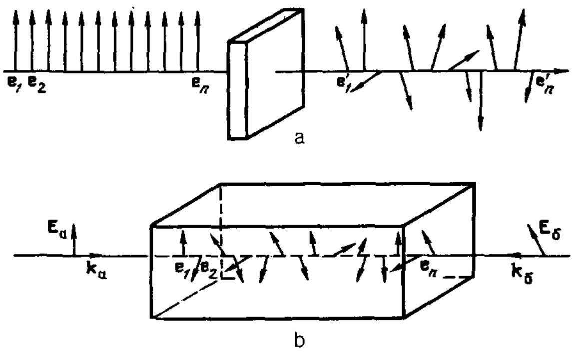

FIG. 1. Temporal polarization chaos (a). Interaction with the nonlinear system ensures that there is an unpredictable change in the polarization of light. e_m and e'_m are unit vectors in the direction of the electric field of the wave before and after the interaction, respectively. Subscripts indicate different instants of time. A specific example is discussed in Section 2. Spatial polarization chaos during the counter-porpagation of light beams in an extended isotropic medium with a Kerr-type nonlinearity (b). Subscripts on the unit vectors e_m, which define the direction of the electric field in one of the waves (E_a), label the spatial coordinate along the beam (see Section 6 for further details).

spontaneous polarization symmetry breaking, certain particular linear polarizations of light are unstable: the exponential development of fluctuations ensures that the radiation leaving the system can, with equal probability, be turned clockwise or anticlockwise, or can acquire left- or right-handed elliptic polarization. The soft effect does not have an intensity threshold (a special case is examined in Sec. 3). Systems with "soft" spontaneous symmetry breaking are also known in mechanics. An example is afforded by the gravitational pendulum held in an inverted vertical position by a spring with zero extension in the strictly vertical position of the pendulum. In this case, the parameter that experiences spontaneous symmetry breaking is the angle of deflection of the pendulum from its vertical position, and the force of gravity is the analog of the light intensity.

Other physically important concepts encountered in nonlinear polarization optics include the limiting direction of polarization[3] (Dykman and Tarasov, 1977) and nonlinear eigenpolarization[4] (Kaplan, 1983). These were introduced by analogy with the idea of the polarization of eigenwaves in linear optics. Here, we are concerned with polarization states that do not vary during the propagation of powerful radiation in nonlinear media. The concept of nonlinear eigenpolarization is much more complicated and extensive than its equivalent in linear optics. When several light beams (for example, counter propagating beams) interact nonlinearly, we speak of matched combinations of polarization parameters and intensities of all rays (in the case of two waves, matched pairs of polarization states and intensities), when their polarization does not vary during the propagation process. In a recent paper, Gaeta *et al.*[5] (1987) established a new property of nonlinear optical systems: allowance for the finite rate at which the nonlinear response is established (in practice, this means that the time for the nonlinearity to be established is commensurate with the time taken by light to cross the nonlinear medium) produces a departure from stability at least for some sets of nonlinear eigenpolarizations. This departure from stability can be classified as *hard* spontaneous polarization symmetry breaking (see Sec. 6 for further details).

As a rule, each particular case of polarization instability can be related genealogically to some known nonlinear polarization effect. The simplest class of such phenomena consists of polarization self-interaction effects, which have been analyzed in particular detail as phenomena stimulating polarization instability, multistability, or chaos. Here we have in mind the self-rotation of the polarization ellipse of a powerful electromagnetic wave[7] (Maker *et al.*, 1964), and the phenomenon of nonlinear gyrotropy, i.e., the variation in the optical activity of a medium in the field of a high-intensity linearly polarized light wave[8] (Akhmanov and Zharikov, 1967), and the self-induced rotation of polarization in cubic crystals, due to the anisotropy of nonlinear absorption[3] (Dykman and Tarasov, 1967). We note that some kind of polarization effect is significant for the initial instability development. The nonlinear transformation of polarization is then small and the contribution of different mechanisms can be analyzed additively. As a rule, nonadditive nonlinear mixing of the contributions of different nonlinear-optics polarization effects occurs for parameter values for which polarization multistability is observed.[8]

Studies of polarization instabilities in optics (regarded

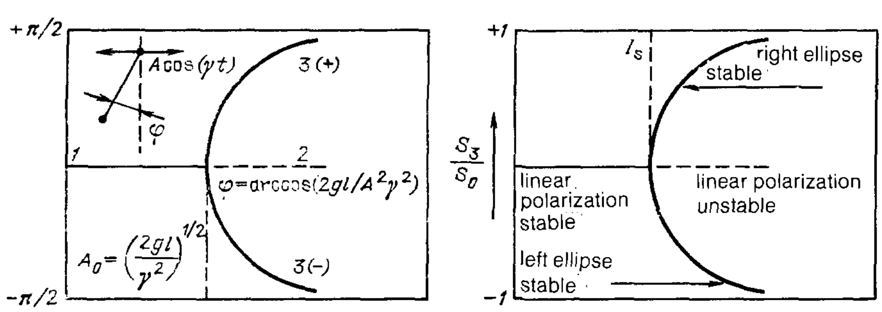

FIG. 2. "Hard" spontaneous symmetry breaking in mechanical and optical systems: a—gravitational pendulum, whose point of suspension oscillates as $A\cos\gamma t$ ($\gamma \gg (g/l)^{1/2}$) (g is the gravitational acceleration); when the oscillation amplitude A reaches the value $(2gl/\gamma^2)^{1/2}$ two equivalent stable symmetric states (left and right) become possible and the deflection φ from the vertical is given by $\cos\varphi = 2gl/A^2\gamma^2$; small fluctuations in the parameters produce a "jump" of the pendulum from the unstable vertical state (segment 2) to branch 3 (+) or 3 (−); b—spontaneous polarization symmetry breaking in an optical system, e.g., in a Fabry-Perot resonator filled with a nongyrotropic nonlinear isotropic medium in the case of linearly polarized exciting radiation; the light intensity at entry to the resonator is plotted along the horizontal axis and the ellipticity at exit from the optical system is shown along the vertical axis (this is a simplified picture; see Sec. 2 for further details) which experiences a jump from the linear to the elliptic state when the pump intensity exceeds the threshold value I_S.

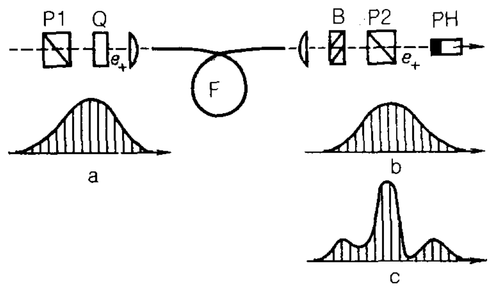

FIG. 3. Experimental demonstration of polarization instability in a weakly birefringent fiber lightguide. A bell-shaped train of picosecond light pulses (a) with circular polarization e_+ is launched into the fiber lightguide. For low power levels, the right-polarized component of the emerging radiation (b), detected by the photodector PH, repeats the shape of the pulse at entry to the system. When the critical intensity is reached, and the value of this can be estimated from the condition that the induced change in the refractive index must be equal to the initial birefringence in the fiber, the circular polarization ceases to be stable and a sharp circularly polarized pulse appears in the photodetector channel (c). P1 and P2 are polarizing prisms, Q and B are, respectively, the quarter-wave plate and the Babinet compensator, and F is the fiber lightguide.[118] Details of the experiment are discussed in Sec. 7.

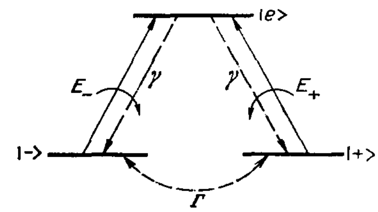

FIG. 4. Model of a two-level atom used to analyze polarization instability in atomic gases.[13] The main transition occurs without a change in the total angular momentum ($J = 1/2 \to J = 1/2$) and involves a degenerate Zeeman sublevel of the ground state (Λ-system).

as studies on phase instabilities) did not evolve out of nothing: radiophysics has long been concerned with phase multistability in nonlinear parametric systems for which bistable and multistable phase states were demonstrated as far back as the 1960's [see Refs. 9 (1963), 10 (1962), and 11 (1962)].

At present, research into polarization instability involves systems with external optical feedback, ring resonators, and Fabry-Perot resonators filled with nonlinear media with different types of symmetry, as well as systems without resonators but with intrinsic or hybrid (electrooptical) nonlinearity in single-ray or multiray configurations. Publications concerned with polarization instabilities in fiber lightguides constitute a numerous group.

Figure 3 shows a schematic diagram of one of the first optical fiber experiments on polarization instability. The necessary experimental techniques had been available for many years, but had to await the more recent theoretical developments in nonlinear polarization optics, optical bistability, and stochastic dynamics of simple systems before they could be systematically exploited in this very simple but striking experiment.

The aim of this paper is to provide a review of current studies of polarization instability which form a subset of an important topic in modern optics, namely, the physics of optical bistability.[12]

2. POLARIZATION INSTABILITY IN ATOMIC GASES. SPONTANEOUS SYMMETRY BREAKING, HYSTERESIS, AND SELF-OSCILLATIONS

The first experimental and theoretical publications on polarization multistability in atomic gases were the result of studies of the model proposed by Kitano, Yabuzaki, and Ogawa[13] (1981), which was subsequently called the Λ-system. This involves an ensemble of two-level atoms that have a degenerate Zeeman ground-state level and relax rapidly to the upper level (Fig. 4). Kitano, Yabuzaki, and Ogawa predicted that, if a gas of these hypothetical two-level atoms were to be placed in a Fabry-Perot resonator pumped by a linearly polarized electromagnetic wave of frequency close to the single-photon absorption resonance, the polarization of radiation leaving the resonator could assume the following three values after the intensity of light had reached a certain definite threshold value (hard regime): (1) linear polarization (as in the incident radiation); (2) elliptic polarization (right-handed ellipse), (3) elliptic polarization (left-handed ellipse). When the intensity reaches its threshold value, the linear polarization becomes unstable, i.e., the system tends toward spontaneous polarization symmetry breaking during fluctuations in the polarization of the incident radiation or fluctuational departures from the uniform distribution of atomic spin directions relative to the light-beam axis. An adiabatic reduction in light intensity then produces a switching of polarization to the linear state for a lower pump intensity (polarization-intensity hysteresis). In western literature, this effect is referred to as *optical tristability* (OT) or *polarization switching* (PS).

The existence of two stable elliptic polarizations above the threshold is due to the circular birefringence that arises as a result of the optical pumping of the corresponding Zeeman transition. It influences the change in the eigenfrequencies of the resonator and, hence, its transmission coefficient which is different for right and left circularly polarized waves. The stability of elliptic polarization, i.e., its tristability, is assured by a suitable choice of the relative position of the absorption line and the resonator mode in the spectrum.[2)]

The Λ-system simulates with sufficient precision the transition observed in sodium vapor in a buffer gas used to suppress hyperfine structure and hole burning. The required effect can be observed by suitably tuning the cavity resonator and choosing its Q-factor. When the light intensity is of the order necessary to saturate the transition, the polarization of the output radiation returns to the linear state.

The first experiments were performed by Cecchi *et al.*[16] (1982), who used a continuously-operating dye laser exploiting the ${}^2S_{1/2} \to {}^2P_{1/2}$ transition (589.6 nm) in sodium vapor. Polarization hysteresis was observed as the laser intensity was varied in the range 5–30 MW, and a small departure from linear polarization at entry to the cavity resonator could be used to select a right- or left-handed hysteresis cycle at exit from the system. An original resonator design with a windowless cell was proposed by Hamilton *et al.*[17] (1983) for the investigation of polarization switching states. It obviated the problem of residual birefringence in windows, and enabled them to perform a detailed study of polarization hysteresis and of polarization switching states for a frequency-scanned laser.

Kitano *et al.*[18] (1981) developed the idea of optical tri-

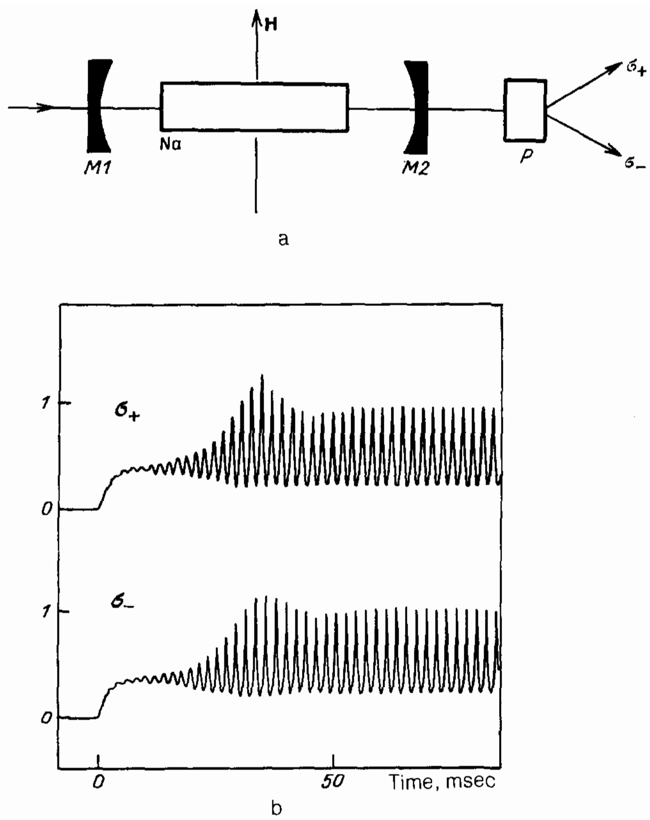

FIG. 5. The Λ-atom in the Fabry-Perot resonator in a transverse magnetic field[23] (a) The oscillograms show the establishment of the self-oscillatory regime for the intensities of the circular components σ^+ and σ^- at exit from the resonator containing a sodium cell in an external magnetic field H exceeding the critical value. Note that the intensity oscillations are in antiphase. The zero along the time axis corresponds to the instant at which the laser is turned on; P is a polarizing prism and $M\,1$-$M\,2$ are resonator mirrors. The radiation entering the resonator is linearly polarized (b).

stability and examined the behavior of the Λ-system in a resonator to which a static magnetic field was applied at right angles to the beam. They showed that, for certain particular ratios of the parameters of the system, the polarization at exit from the resonator exhibited self-oscillations with frequency of the order of the Larmor frequency. These self-oscillations were observed experimentally by Mitschke *et al.*[19] (1983) for a magnetic field of a few tens of microtesla. The critical field was found to depend on the Q-factor of the resonator and on the detuning from resonance (Fig. 5) [see also Refs. 20–22 (1984) and 23 (1986)].

A subsequent analysis of the Λ-system in a resonator, performed by Savage *et al.*[14] (1982), showed that, in the dispersion approximation (absoroption neglected), and when the saturation of transitions was taken into account, the asymmetric stable states could become unstable, and this would lead to the self-oscillatory polarization state in the absence of the external magnetic field, to the doubling of the period of the oscillations, or to chaotic motion, i.e., optical polarization turbulence. In a later paper (1983), Carmichael *et al.*[24] found a complicated chaotic structure with "windows" of periodic motion, typical for the Lorentz attractor or the Duffing oscillator [see, for example, Ref. 25 (1984)].

Arecchi *et al.*[16] (1983) found multistable (bistable and quadristable) solutions when they took into account the coherence of the ground states of the system. Giusfredi *et al.*[27] (1985) investigated a Fabry-Perot resonator containing sodium vapor without the buffer gas, but with competing transitions between hyperfine and Zeeman levels, which ensured the presence of multistable polarization states. This system is significantly different from the simple Λ-atom. Complicated hysteresis curves were recorded by frequency scanning and varying the temperature and the magnetic field. The influence of saturation of transitions, vapor density, and resonator tuning on the hysteresis cycles were investigated experimentally by Giacobino[28] (1985). Spontaneous polarization symmetry breaking in atomic vapor was investigated by Adonts *et al.*[29] (1984) for $J=1/2 \to J=3/2$ transitions. The papers of Hamilton *et al.*[30] (1982) and Areshev *et al.*[31,32] (1982, 1983) are devoted to studies of polarization instability in the ring resonator.

Kitano *et al.*[33] (1984) found that optical polarization instabilities could occur in the Λ-system even in the absence of the Fabry-Perot resonator. Delayed feedback, which is necessary for chaos to occur, can be produced with a $\lambda/8$ plate and a mirror that returns a substantial proportion of the radiation back to the gas-filled cell (Fig. 6). This example provides a simple and clear demonstration of the reasons for the optical polarization turbulence (chaos). The magnetization M_z in the direction of a ray is proportional to the population difference between the ground-state sublevels, and can be determined from the balance equation. If the normalized intensity I_+ of right-polarized light is high in comparison with I_- and the damping Γ/σ, the light field ensures that the spins of all the atoms point in the direction

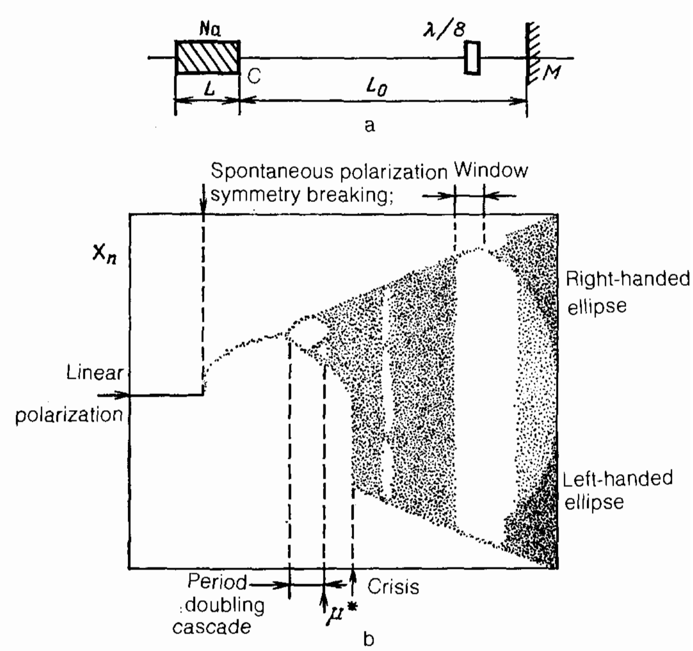

FIG. 6. Polarization chaos in an optical system without a resonator: a—optical system for observing polarization chaos;[32] the cell C contains a gas of two-level atoms (Λ-system), e.g., sodium vapor; mirror M returns part of the radiation to the cell after the $\lambda/8$ plate; b—when L_0 is large, the description of the magnetization of the gas reduces to the discrete map $X_{n+1} = \mu \sin X_n$, where μ is a measure of the intensity of light; on the birfurcation diagram, the chaotic variation in X for $\mu > \mu^*$ corresponds to the chaotic behavior of the gas magnetization, Faraday rotation, and, consequently, chaotization of polarization after the exit mirror M.

of the beam. The cell rotates the polarization of light by an angle proportional to the magnetization and to the thickness L of the cell, and the change in polarization produces a change in the reflection conditions in the mirror + wave plate system. When the delay $t_R = 2L_0/c$ in the optical delay line between the cell and the mirror and back again is taken into account, a self-consistent equation can be obtained for the normalized magnetization $m_z = M_z/M_0$, which gives the polarization at exit from the system (through the mirror):

$$\frac{dM_z(t)}{dt} = -(\Gamma + 2\sigma I_0)\, m_z(t) + R\sigma I_0 \sin[2klm_z(t - t_R)].$$

When the delay t_R is large enough, a linear change of variables in this equation will reduce it to the discrete map

$$X_{n+1} = \mu \sin X_n,$$

where

$$X = 2klm_z \quad и \quad \mu = 2klRI_0(\Gamma + 2I_0)^{-1}$$

and k is a function of the detuning from resonance.

Even a pocket calculator will then suffice to verify that this map demonstrates chaotic behavior (Fig. 6b). Which of the two branches of opposite symmetry will be taken up by the system at the point $\mu = 1$ when the normalized intensity μ is altered adiabatically will be determined by small fluctuations in the system (spontaneous symmetry breaking), e.g., fluctuations in the input polarization (the system is symmetric under the replacement of X with $-X$, i.e., under the replacement of right-handed with left-handed polarization ellipse). A change in light intensity is accompanied by period-doubling cascades, after which the polarization becomes a random function of time (polarization turbulence). More detailed analysis shows that chaotic motion in the system is demonstrated by the so-called *crises of chaos*,[34] in which the dimension of a chaotic attractor (amplitude of polarization fluctuations) can increase rapidly for a small change in intensity.

Yabuzaki *et al.*[35] (1984) investigated experimentally the Λ-system without a cavity resonator (sodium vapor with buffer gas) in the optical system of Fig. 6a, but without the optical delay line. Chaotic motion was not observed in this case, but the system did demonstrate spontaneous symmetry breaking. As in the Fabry-Perot resonator, a transverse magnetic field produces an oscillatory motion of polarization at a frequency close to the Larmor value. Transitions (hops) between the two stable branches (elliptic polarizations of opposite sign) do not occur as the parameters are varied, but they can be produced by an additional pulse of circularly polarized light. After careful adjustment of the system, i.e., accurate setting of the relative orientation of the polarizing prism and the axis of the wave plates, the symmetry breaking (exit to one of the symmetric elliptic solutions) at the threshold intensity is accompanied by "choice noise" which is observed for about a minute. The system demonstrates polarization hysteresis when the $\lambda/8$ plate in the feedback loop is replaced with a combination of a quarter-wave plate and a linear polarizer (Glan prism).

McCord and Ballagh[36] (1985) have pointed out an interesting possibility whereby the intensity of a circularly polarized beam can be efficiently controlled by a second beam of opposite circular polarization, using the selective pumping of Λ-atoms.

Optical transitions in Λ-atoms occur, as already noted, without a change in the total angular momentum ($J = 1/2 \to J = 1/2$). Ballagh and Jain[37] (1984) and Parigger *et al.*[38] (1985) have noted the interesting polarization phenomena that can be observed in Fabry-Perot resonators filled with atomic vapor when transitions with a change in the total angular momentum ($J = 1 \to J = 0$) are excited. This atomic system is fundamentally different from the Λ-system. Polarization asymmetry is not spontaneously nucleated in this case because of Zeeman coherence at the $J = 1$ level. However, when a longitudinal magnetic field is applied and destroys the Zeeman coherence of the ground state, the intensity-polarization hysteresis is observed. Samarium vapor was chosen for the investigation (${}^7F_1 \to {}^7F_0$ transition, $\lambda = 570.68$ nm). The principle of the experiment is analogous to that shown in Fig. 5, except that the magnetic field is applied longitudinally rather than transversely to the beam, using the Faraday scheme. The results of a detailed experimental investigation have been reported by Parigger *et al.*[39] (1986), who also investigated the polarization switching states. The right-polarized radiation component is a non-single-valued function of the intensity of the right-polarized radiation. The situation is mirror-inverted when the direction of the magnetic field is reversed. In other words, we can speak of polarization bistability controlled by the magnetic field.

At present, publications on polarization instability in Fabry-Perot resonators filled with atomic gases have almost completely ceased to appear. However, the basic ideas on polarization chaos and polarization multistability, put forward in the course of studies of Λ-atoms, have been found to be fruitful in the study of other systems.

3. LIMITING POLARIZATION DIRECTIONS, POLARIZATION OSCILLATIONS, POLARIZATION MULTISTABILITY, AND CHAOS IN CUBIC CRYSTALS

In linear optics, the polarization of a wave propagating in an arbitrary direction in a nongyrotropic cubic crystal ($m3m$, $m3$, $\bar{4}3m$) remains constant.[3)] Dykman and Tarasov [Ref. 3 (1977) and Ref. 4 (1978)] have investigated the dissipative mechanisms of nonlinear resonance interaction between light and impurity levels in cubic crystals with a triply degenerate excited state, which transforms according to the vector representation of the cubic group (Fig. 7), and noted the existence of a new polarization effect in which a linearly polarized ray propagating along the four-fold symmetry axis of the crystal (e.g., ⟨001⟩) retains its polarization state if, and only if, the electric vector points along ⟨100⟩, ⟨010⟩, ⟨110⟩, or ⟨$\bar{1}$10⟩. The saturation of impurity levels in a wave whose polarization is different from any of those just noted is accompanied by rotation of the polarization vector toward the nearest of the directions ⟨100⟩ or ⟨010⟩: the weaker field component (E_x or E_y) is absorbed more strongly. The ⟨110⟩ or ⟨$\bar{1}$10⟩ polarizations are unstable: a minute fluctuation grows exponentially with sample thickness, tending to the nearest four-fold symmetry axis. The system thus tends to soft spontaneous polarization symmetry breaking without a threshold (the first experimental observation of this effect is described in Ref. 42).

Zhadanov *et al.*[43] (1980) related in a more general

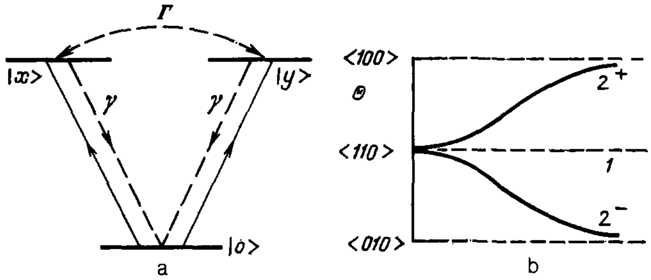

FIG. 7. "Soft" spontaneous polarization symmetry breaking without a threshold in a cubic crystal: a—model of the impurity in the local field of the cubic crystal;[3] the ∇-system with a threefold degenerate excited level (the state $|z\rangle$ not shown); the $x\rangle$ and $|y\rangle$ levels are populated by radiation polarized along the x and y axes, respectively; b—"soft" spontaneous polarization symmetry breaking in a dissipative cubic crystal (nonzero imaginary part of the anisotropic component of the tensor $\chi^{(3)}$); the intensity I incident on the crystal is plotted along the horizontal axis and the direction of the polarization of the light wave, θ, is plotted as a function of the position coordinate z in the crystal along the vertical axis; branch 1—unstable, branches 2^+ and 2^-—stable nonlinear eigenpolarizations; $\theta \sim \theta_0 \exp(\eta I z)$ on the initial segment.

form, the nonlinear transformation of polarization in cubic crystals to the sign of the anisotropic combination of the components of the cubic nonlinear susceptibility tensor $\Delta\chi^{(3)} = \chi^{(3)}_{1111} - 3\chi^{(3)}_{1122}$ and, having experimentally investigated the orientational dependence of the nonlinear self-rotation of the plane of polarization in gallium arsenide ($\bar{4}3m$), have discovered stable and unstable (repulsive) polarization eigenstates for linearly polarized light. This question was subjected to a detailed theoretical analysis by Zhdanov *et al.*[44] (1981), Zheludev[45] (1981), and Zheludev and Petrenko[46,47] (1984) for crystals of higher and intermediate categories.

The anisotropy of the third-order nonlinear susceptibility tensor is typical for all cubic crystals in which, in contrast to the isotropic medium, the combination of components $\Delta\chi^{(3)}$ is, in general, nonzero. Contributions to the evolution of the anisotropic part are provided not only by impurity transitions, but also by interband transitions as well as transitions involving the participation of excitons.[4)]

If the polarization is different from the eigenpolarization, its structure transforms as light propagates through the medium: the polarization rotates toward the nearest stable direction, and the rate of rotation depends on the imaginary part of the anisotropic component of the cubic nonlinear susceptibility, Im $\Delta\chi^{(3)}$, and on the intensity of light. Moreover, self-induced ellipticity is produced and depends on the intensity, the thickness of the crystal, and the real part of the anisotropic component of the tensor $\chi^{(3)}$. The sign of Re $\Delta\chi^{(3)}$ determines the sense in which the end point of the electric field vector traces out the ellipse.[5)] The sign of the anisotropic part of the tensor $\chi^{(3)}$ depends not only on the crystal type, but also on the wavelength of light. In principle, this dependence gives rise to a completely new type of polarization effect, i.e., spectral-polarization instability. A small change in the wavelength of light in the region $\Delta\chi^{(3)} = 0$ produces the *switching* of stable and unstable axes and, consequently, polarization switching at exit from the crystal by amounts that, in principle, can reach up to 45°. The scale of the effect is determined by the derivative $\partial\,\mathrm{Im}\{\Delta\chi^{(3)}\}/\partial\lambda$. This quantity has been measured for gallium arsenide ($T = 300$ K, $\partial\,\mathrm{Im}\Delta\chi^{(3)}/\partial\lambda \simeq 10^{-9}$ cgs/μm; Ref. 48). The heating of the crystal can also lead to a change in the sign of anisotropy; for example, in gallium arsenide, the point of nonlinear isotropy $[\Delta\chi^{(3)}(\nu_0) = 0]$ shifts toward the red as the temperature increases at the rate $\partial\nu_0/\partial T \simeq 3$ cm^{-1}/deg^{-1}. The effect is significant for lasers with relatively long pulses that succeed in producing appreciable heating of the crystal: as the intensity increases, radiant heating should give rise to observable interchange of stable and unstable polarization states and polarization jumps during the laser pulse (thermal polarization instability). The subdivision of these effects into self-induced rotation and self-induced ellipticity is valid only for small nonlinear transformations. Nonadditive mixing of the contributions due to different polarization self-interaction effects [see Ref. 51 (1982) and Ref. 8 (1984)] sets in under the conditions of strong transformation of polarization. The main features of this are examined in Ref. 51 for zinc blende crystals, taking into account the spatial dispersion of nonlinearity (Sec. 4). It is shown that a linear polarization at entry to the medium tends to become circular, and the sign of the latter depends on the orientation of the initial polarization relative to the symmetry axes of the crystal and on the relationships between the components of the nonlinear susceptibility tensor. A small change in the direction of the polarization vector in the region of unstable nonlinear eigenpolarization produces a large change in polarization at exit, i.e., from right- to left-handed circularly polarized waves. Nonlinear eigenpolarizations can be found in a general form in the case of self-interaction, without specifying the nature of the nonlinearity (it is sufficient to assume that $|D^{\rm nl}| \ll |D^{\rm l}|$ where $D^{\rm l}$ and $D^{\rm nl}$ are, respectively, the linear and nonlinear parts of the electric polarization of the medium. If we confine our attention to the analysis of the propagation of transverse waves in cubic crystals and in birefringent media along the optic axis, the condition that must be satisfied by the nonlinear eigenpolarization takes the form of the following expression:[44–46]

$$\frac{D_+^{\rm nl}}{E_+} = \frac{D_-^{\rm nl}}{E_-}$$

where $D_\pm^{\rm nl} = D_x^{\rm nl} \pm iD_y^{\rm nl}$ and $E_\pm = E_x \pm iE_y$. The nonlinear induction $D^{\rm nl}$ is calculated from the field E in the unperturbed (linear) problem, i.e., with linear absorption and gyrotropy. The solutions for the electric field components that satisfy the above relation (we note that complex numbers are being equated) correspond to polarization states that do not experience intensity-dependent changes against the background of natural gyrotropy. Of course, the medium must be nongyrotropic if we seek linearly polarized eigenwaves.

Dykman and Tarasov[53] (1982), Balashenkov and Kozlov[54] (1984), and Yumoto and Otsuka[55] (1985) used an analysis of phase trajectories to show that, for certain relationships between the components of the nonlinear susceptibility tensor, the polarization ellipse oscillates in space around the symmetry axes of the nondissipative cubic crystal. Sala[58] (1984) has given a clear formulation of the nonlinear-optics polarization problem for the Stokes vector,[6)] having reduced the analysis to the solution of a set of coupled nonlinear equations. Gregori and Wabnitz[59] (1986) and, independently, Tratnik and Sipe [see Ref. 60 (1986) and Refs. 61 and 62 (1987)] then found a profound analogy between the equations for the Stokes vector in the polarization problem and the Euler equation for the angular momentum of a rigid body rotating around a fixed point, and the propeller-airplane problem.[1(b)] It would appear that the set of coupled Euler equations for the Stokes vector is the most elegant and universal form of the relations of nonlinear polarization optics of nondissipative systems. The nonlinearity consists of the fact that the "precession frequency," i.e., the spatial period of polarization oscillations, is a function of the magnitude of the "precessing vector," i.e., of the intensity of light. It may be expected that, when this description is generalized to systems with absorption, it will lead to equations analogous to the equations of motion of a rigid body with one fixed point and friction. Even a small amount of friction can then result in loss of stability and the appearance of low-frequency components in the spectrum (the so-called secular instability; see, for example, Ref. 64).

We now turn to the consideration of cubic crystals. Following the paper by Gregori and Wabnitz,[59] we shall write the equation for the Stokes vector of a wave propagating in the ⟨001⟩ direction in a cubic crystal in the form

$$\frac{d\mathbf{S}}{dz} = [\boldsymbol{\Omega}_{nl}(\mathbf{S})\,\mathbf{S}],$$

where

$$\boldsymbol{\Omega}_{nl} = 4\pi k_0 \chi^{(3)}_{1212}\left\{0,\ \left(1 - \frac{\Delta\chi^{(3)}}{2\chi^{(3)}_{1212}}\right) S_2,\ -\frac{\Delta\chi^{(3)}}{2\chi^{(3)}_{1212}} S_3\right\}.$$

The variables can be separated in the nondissipative case, and the evolution of $S_1 = |E_x|^2 + |E_y|^2$ can be described by the Duffing equation without damping and the right-hand side given by

$$\frac{d^2 S_1}{dz^2} + \alpha S_1 + \beta S_1^3 = 0.$$

We thus have an interesting space-time analogy, namely, the variation of polarization parameters with sample thickness is analogous to the oscillations of a load suspended from a nonlinear (non-Hookeian) spring. The nondissipative Duffing equation can be solved exactly in terms of elliptic Jacobi functions, i.e., whatever the parameters of the problem, the polarization does not exhibit a chaotic variation with increasing thickness. Tratnik and Sipe [see Ref. 60 (1986) and 61 (1987)] have examined the general form of the equations for the Stokes vector in media with and without a center of inversion along the directions of twofold, threefold, fourfold, and fivefold (or higher) rotation symmetry axes. The case of the cubic crystal corresponds to a fourfold rotation axis. It is found that all nondissipative media have at least two integrals of motion, namely, the intensity of light and a more complicated integral that introduces the intensity into the free energy of the system. Actually, this means that, even in the most general case, the Euler equations for the three components of the Stokes vector are integrable, and there are no chaotic solutions of the polarization self-interaction problem.

A distribution of polarization parameters along the beam with a continuous spectrum of spatial frequencies (polarization chaos) arises in anisotropic cubic crystals when a more complicated model is analyzed. Yumoto and Otsuka[55] (1985) and Gregori and Wabnitz[59] (1986) have examined the counter propagation of two beams of equal frequency in a cubic crystal (two nonlinear coupled Euler equations for the Stokes parameters) and have determined the nonlinear eigenpolarizations of the steady-state problem. Depending on the initial polarization parameters of the waves entering the crystal from opposite directions, and depending on the ratios of the components of the cubic nonlinear susceptibility tensor of the medium, there are two solutions for the intensity ratio of these waves that determine the nonlinear eigenpolarizations. However, for an arbitrary intensity ratio and positive anisotropy of nonlinear refraction, waves polarized along the bisectors between the fourfold symmetry axes are always nonlinear eigenwaves. Two counter-propagating circularly polarized waves of the same or opposite chirality are also found to retain their polarization for any ratio of the components of the nonlinear susceptibility tensor. In all other cases, in which the polarizations or beam intensities are different, the attainment of threshold excitation in the medium is accompanied by the onset of polarization chaos in space (analogous to the temporal development of the oscillations of two coupled nonlinear oscillators). Tratnik and Sipe [see Ref. 60 (1986), Ref. 62 (1987), and Ref. 63 (1988)] have shown, however, that it is only in the case of propagation (direct or counter) along the symmetry axes different from C_n ($n = 1,2,4$) that there is the necessary number of integrals of motion sufficient for the complete integration of the equations for the Stokes vector. In other words, like any other dynamic chaos, polarization chaos is a consequence of an "excess of freedom" during the counter-propagation of rays in cubic crystals. A symmetric analysis of polarization bistability in its topological context has also been performed by Dykman[65] (1986). The random distribution of polarization within a cubic crystal during the interaction of two counter-propagating waves is a new form of deterministic instability in conservative systems, referred to by the authors of Ref. 55 as "frustrated": a small change in the intensity of polarization of one of the rays at entry to the crystal is accompanied by an unpredictable and very rapid change in polarization at exit from the crystal.

Counter propagation can be produced by reflection from an external mirror or a mirror deposited on the face of the crystal. It is precisely this case that is particularly important in practice (when it is combined with a Fabry-Perot resonator containing the nonlinear crystal). Otsuka and Yumoto[66,67] (1986) have examined this configuration and indicated the conditions for the onset of spatial polarization turbulence in the optical region (Fig. 8).

The first experimental studies of frustrated polarization instability were performed by Zheludev *et al.*[68,69] (1983), Dovchenko[70] (1984), and Dovchenko *et al.*[71] (1984), who investigated the polarization self-interaction of picosecond pulses in a Fabry-Perot resonator. The time to traverse the resonator was 20 ps and the pulse length was 40 ps, so that this case was closer to that of two counter-propagating beams, i.e., a beam incident on a crystal and a beam reflected from the back mirror of the resonator (Fig. 9).

A detailed analysis of quasistationary polarization states in an optical ring resonator containing a cubic crystal with nonzero nonlinear anisotropy has been carried out by Dykman and Tarasov[72,73] (1984) for nonlinear mechanisms relying on the saturation of absorption by impurity centers and on two-photon absorption. Nonlinear eigenpolarizations, their stability, type of bifurcation curves, and conditions of spontaneous polarization symmetry breaking were

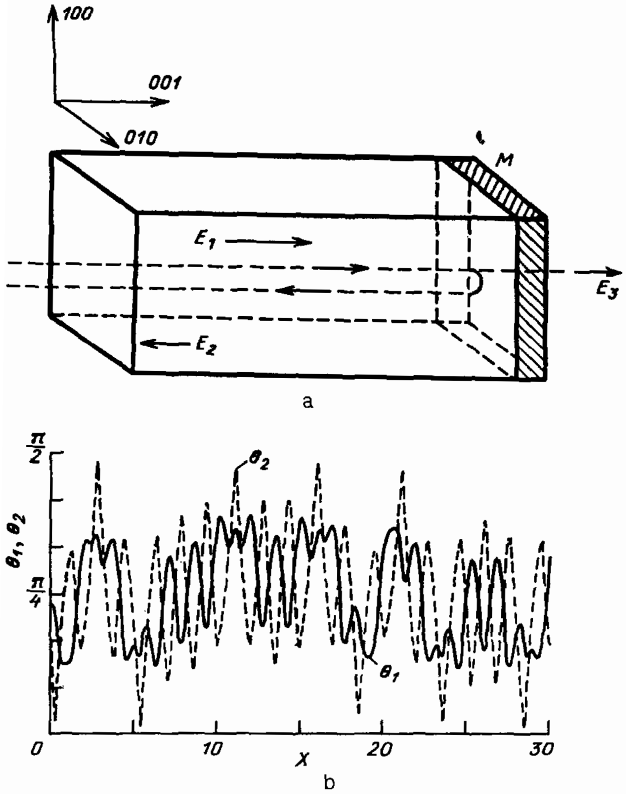

FIG. 8. Spatial polarization instability in a cubic crystal for two counter-propagating waves:[66,67] a—arrangement for observing frustrated polarization instability, using reflection from the mirror M; b—distribution of polarization along the x axis in the crystal; θ_1 and θ_2 are angles of rotation of the polarization of forward and backward waves, respectively, under the conditions of spatial polarization chaos (stationary problem).

investigated for the dissipative ($\mathrm{Im}\,\Delta\chi^{(3)}\neq 0$) and reactive ($\mathrm{Re}\,\Delta\chi^{(3)}\neq 0$) mechanisms of optical nonlinearity. The medium chosen for the observation of optical polarization bistability was a KCl crystal containing color centers F_A (Li),

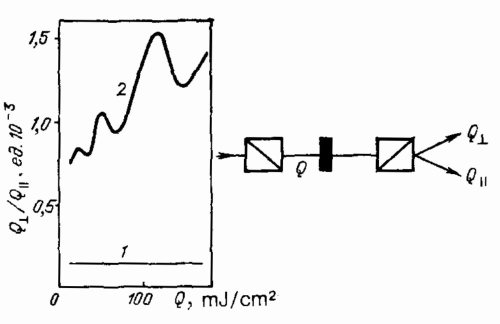

FIG. 9. Experimental data on frustrated polarization instability in cubic crystals.[49,70,71] The diagram shows the integrated degree of polarization $Q_\perp/Q_\parallel$ (ratio of the energies of orthogonal and parallel polarization components in the pulse) as a function of the energy density Q in the pump pulse after the interaction between the picosecond light pulse and the Fabry-Perot resonator consisting of a gallium arsenide crystal with mirrors deposited on the ⟨001⟩ faces ($r = 80\%$), for different input polarizations (resonator baseline $d = 900\,\mu m$). Curve 1 corresponds to the initial orientation of polarization along the $\theta = \pi/4$ direction, which is the nonlinear eigenpolarization. The extinction coefficient in this case is about 3×10^{-4} and is independent of intensity. It is determined by the quality of the polarizing prisms. Curve 2 corresponds to $\theta = \pi/8$. We note the nonmonotonic increase in depolarization as a function of the pump intensity.

for which optical bistability due to the "slow" nonlinearity of the color centers could be observed for very low levels of optical interaction (~ 1 mW), and the corresponding time constants were of the order of tens of seconds. Yumoto and Otsuka have suggested that a possible candidate for the observation of frustrated polarization instability is the KTN crystal ($KTa_xNb_{1-x}O_3$), which has a nonlinear anistropy amounting to $\Delta\chi^{(3)}\sim 10^{-13}$ cgs (Ref. 74). Polarization turbulence can probably be observed in this case for intensities of the order of 4 GW/cm^2. The nonlinear anisotropy has now been measured for a number of cubic crystals. The anisotropic part of the tensor $\chi^{(3)}$ has its maximum value near the one-photon absorption resonance of gallium arsenide ($\Delta\chi^{(3)}\sim 10^{-6}$–$10^{-7}$ cgs), for which a giant self-induced change in polarization has been recorded with a rate constant exceeding 10 000 deg/cm (Refs. 75 and 76), and the frustrated polarization instability can probably be observed for intensities of the order of 10 MW/cm^2.

Earlier in this Section, we discussed the spatial distribution of light-wave polarization in an anisotropic nonlinear medium, i.e., we were interested in *spatial* polarization instability. Akhmanov *et al.*[77] (1982) have predicted that it should be possible to observe temporal polarization instability in cubic crystals. This question has been analyzed by Zheludev *et al.*[8] (1984) and by Akhmanov and Zheludev[49] (1986). The conditions for the observation of spatial and temporal polarization instabilities are different. While the first can arise in a system for a sufficiently high degree of nonlinear transformation of polarization of light (high radiation intensity or large crystal length) in the presence of feedback (resonator) or counter-propagating waves, the instability considered in Ref. 77 can be observed locally without feedback or cavity resonator (intrinsic polarization instability) for each individual molecule of the medium. However, the important point here is that the cubic nonlinearity of molecular oscillators must be large enough. For example, the origin of polarization instability becomes clear if we consider the model in which orthogonal noninteracting nonlinear oscillators are excited by different orthogonal polarization components (model of a cubic crystal).[7)] This shows that hysteresis in the level of excitation of each of the orthogonal oscillators generates a hysteresis in the absorption of orthogonal components of the light field and, consequently, a polarization hysteresis. Numerical simulation of the behavior of a nonlinear oscillator and of a set of coupled oscillators in an external force field is known to lead to chaotic solutions, i.e., temporal polarization chaos should arise in a cubic crystal. The condition for the observation of this phenomenon is $|\chi^{(3)}|\ |E^2|\sim 1$. When the density of nonlinear centers in the crystal host is low, the polarization instability can have a small amplitude because, in this case, it is due to a local mechanism, and the resultant effect is proportional to the density of nonlinear centers.

In our opinion, polarization instability in cubic crystals is particularly important from the practical point of view because it provides a basis for the development of devices for the control of light by light, with polarization data encoding. The simplicity of the system and the availability of the technology for highly nonlinear semiconducting cubic crystals such as silicon, germanium, gallium arsenide, and the triple solution mercury-cadmium-tellurium opens up new avenues in different spectral ranges.

4. POLARIZATION INSTABILITY IN MEDIA WITH NONLINEAR GYROTROPY

There is undoubted interest, especially in nonlinear optics of biological objects and the spectroscopy of excitons in solids, in investigations into the polarization instability associated with nonlinear gyrotropy, i.e., the spatial dispersion of nonlinearity. The simplest manifestation of nonlinear gyrotropy is the dependence of the optical activity of an isotropic medium on intensity[8] (1967). Kovrighin *et al.*[43,44] (1981) have shown that the spatial dispersion of nonlinearity, introduced into the phenomenological description via the tensor $P_i = \gamma^{(3)}_{ijklm} E_j E_k \nabla_m E_l$, can lead to the self-induced rotation of polarization even in nongyrotropic crystals, for example, crystals with $\bar{4}3m$ symmetry. A more detailed analysis of nonlinear gyrotropy has been carried out by Zheludev[45] (1981) and by Zheludev and Petrenko[46,47] (1984). When the polarization interaction is weak, changes in polarization due to the nonlinear anisotropy ($\Delta\chi^{(3)}$) and the nonlinear gyrotropy ($\gamma^{(3)}$) are additive.

We note the important difference between the nonlinear rotation of the plane of polarization of light due to nonlinear anisotropy and that due to nonlinear gyrotropy. Rotation of the plane of polarization due to the nonzero anisotropic components of the imaginary part of the tensor $\chi^{(3)}$ is *P*-even, i.e., its symmetry is analogous to that of Faraday rotation: the small nonlinear angle of rotation is doubled in each forward and return transit. The effect due to the spatial dispersion of nonlinearity is *P*-even and is analogous to the rotation observed in sugar solutions, i.e., it is cancelled on the return path. Spatially dispersive rotation is nondissipative (reactive), i.e., *T*-invariant, and occurs under thermodynamically reversible conditions. The nonlinear rotation of polarization due to the anisotropy of the cubic crystal is dissipative and irreversible, i.e., *T*-noninvariant.[8)]

The inclusion of spatial dispersion of nonlinearity can lead (e.g., in crystals with zinc blende symmetry) to a change in the nonlinear eigenpolarizations when the direction of propagation of light is reversed.

Nonlinear gyrotropy is well defined in media with strongly nonlocal nonlinear response. The parameter representing the spatial dispersion, which determines the scale of nonlocality, can be taken to be the ratio of the characteristic microscopic dimension in the medium (δ), e.g., the crystal lattice parameter in the nonresonant case, to the wavelength. The corresponding susceptibility has been estimated[48] as being $\gamma^{(3)} \sim \delta\chi^{(3)}$. This is why effects due to the spatial dispersion of nonlinearity, and the polarization instability associated with them, may be significant in the first instance in cholesteric liquid crystals near the point of phase transition to the isotropic state ($\delta/\lambda \sim 10^{-1}$), chiral biological macromolecules ($\delta/\lambda \sim 10^{-1}$), and in the neighborhood of exciton and biexciton absorption resonances in semiconductors ($\delta/\lambda \sim 10^{-2}$–$2\times10^{-2}$).

There is only a small number of publications on the effects of nonlinear gyrotropy on polarization instability. Makarov *et al.*[83] (1986) and Makarov and Matveeva[84] (1988) have examined an isotropic nonlinear gyrotropic medium (in which rotatory power is a function of intensity) in an optical ring resonator. Allowance for spatial dispersion of the nonlinearity produces a considerable change in the polarization instability as compared with the case of nongyrotropic nonlinear media,[31,32] which is particularly well defined for a linearly polarized exciting radiation: hard spontaneous polarization symmetry breaking, which is typical for nongyrotropic right-left symmetric nonlinear media in the resonator, is now removed and "softened" by the chiral nonlinear optical interaction, i.e., the emerging radiation is elliptically polarized even for low intensities. For high intensities (above the threshold for spontaneous symmetry breaking in nongryrotropic systems), the dependence of the intensity and polarization parameters of the emerging radiation on the pump level is very well defined, i.e., nonlinear gyrotropy gives rise to new closed branches of the polarization-intensity curve, the stability region is shifted, and self-oscillations appear.

Akhmanov *et al.*[77] (1982) have calculated the nonlinear susceptibility of an ensemble of randomly oriented mirror-nonsymmetric molecules with the same chirality sign on the assumption that each of the molecules can be described by the nonlinear Kuhn model, i.e., a set of orthogonal interacting oscillators in the field of the electromagnetic wave:

$$\ddot{x} + \omega_0^2 \dot{x} + \xi x + \frac{a}{3!} x^3 + \frac{b}{3!}(y^3 + 3x^2 y) + cxy^2 = \frac{eE_x}{m} e^{-ik_z D/2} + \text{c.c.},$$

$$\ddot{y} + \omega_0^2 \dot{y} + \xi y + \frac{a}{3!} y^3 + \frac{b}{3!}(x^3 + 3y^2 x) + cyx^2 = \frac{eE_y}{m} e^{ik_z D/2} + \text{c.c.}$$

The method of slowly varying amplitudes can then be used to derive an implicit constitutive equation for the isotropic gyrotropic medium, which takes into account the cubic nonlinearity of the molecular oscillators:

$$\mathbf{P} = \chi^{(1)}\mathbf{E} - i\Gamma\,[\mathbf{kP}] + R\,(2\,(\mathbf{PP}^*)\,\mathbf{P} + (\mathbf{PP})\,\mathbf{P}^*) - i\Gamma_2\,(\mathbf{P}\,(k\,[\mathbf{PP}^*]) + [\mathbf{kP}]\,(\mathbf{PP}^*)),$$

and this allows many-valued solutions for the response of the medium. It has been shown that the specific gyrotropy and circular dichroism of this system are non-single-valued functions of the light intensity, i.e., for low intensities ($|E|^2\chi^{(3)} \ll 1$), there is the usual nonlinear optical activity, but polarization multistability arises for intensities $|E|^2\chi^{(3)} \sim 1$ and, for linearly polarized incident light, the sign of the resulting gyrotropy can be the same as or opposite to the sign of natural optical activity, depending on the prehistory and the intensity of light (Fig. 10).

It has been suggested that optical polarization bistability could be observed in gyrotropic crystals against the background of amplitude bistability due to increasing absorption,[85] and this looks very realistic. In a medium in which the absorption coefficient is a rapidly varying function of temperature, e.g., in semiconducting materials near the fundamental or exciton absorption edge, an increase in the intensity of light propagating through the crystal leads to an increase in its temperature because of the dissipation of part of the light energy. The rise in temperature is accompanied by an increase in absorption which, in turn, produces further dissipation of light energy. The positive feedback loop is thus closed, and hysteresis becomes possible on the pump-transmitted intensity plane (see, for example, Ref. 86 for further details). When the bistability due to increasing absorption in gyrotropic media is observed, amplitude bistability must, of course, be accompanied by polarization instability because of the temperature dependence of gyrotropy.

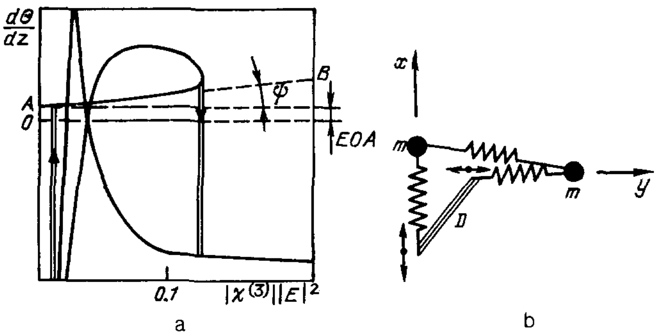

FIG. 10. Local polarization instability in a nonlinear gyrotropic liquid:[77] a—specific gyrotropy $d\theta/dz$ of an ensemble of randomly oriented chiral molecules; EOA—natural gyrotropy; the slope of the line AB that touches the graph at the point $|E|^2 = 0$ is proportional to the nonlinear optical activity constant; arrows show transitions between stable branches; multivalued solutions constitute the polarization multistability; b—nonlinear Kuhn model of an optically active molecule; it is assumed that the oscillators representing the molecules and the elastic coupling between them are nonlinear.

At this point, we note an important property of thermal effects: when continuously operating lasers are used, the threshold for the observation of optical bistability does not depend on the power density in the light beam, and is a function of only its total power because the rate of change of temperature in the hot-spot increases with decreasing diameter of the beam at approximately the same rate as the increase in the density of liberated power.[85] This actually means that the amplitude-polarization thermal bistability can be observed at arbitrarily low power densities (as long as the light-spot diameter is much smaller than the diameter of the crystal). In some materials, for example, ZnP_2 (Ref. 85), the nonlinear gyrotropy due to the thermal mechanism is found to reach the gigantic value of 2000 $\deg\cdot cm^{-1}\cdot W^{-1}$. Optical polarization chaos can probably be observed in a two-beam scheme, using a time-modulated pump and a nonlinear gyrotropic medium (Ref. 87).

It is now appropriate to mention that, in recent years, considerable research effort has been devoted in stereochemistry to spontaneous symmetry breaking in weak right-left asymmetric interactions (e.g., optical interactions). For some ranges of the external interction parameters (e.g., incident intensity), the stable state of the isotropic medium (a mixture of chiral molecules) is asymmetric: there is an excess of one of the mirror isomers. Outside these regions, the symmetric state, i.e., the so-called racemate (a mixture of equal amounts of right and left elements) is the only stable one.

The sensitivity of the racemic state to chiral interactions is exceedingly high. Kondepudi and Nelson[88] (1986) have discussed the possibility of using spontaneous symmetry breaking for the detection of parity violation in the weak interaction between the electron and the nucleus, which lifts the energy degeneracy of molecular levels of right-left isomers and produces an energy level splitting ΔE. The chiral selectivity is predicted to be $\Delta E/kT \sim 10^{-17}$–$10^{-15}$.

The interaction between a mixture of chiral molecules and light can be described by a three-level energy scheme, in which the uppermost level is the excited state that is identical for all the molecules and the two other levels correspond to the two mirror isomers[89,90] (Avetisov and Anikin, 1985; compare this with the Λ-system[13]). Since the detection of stable proportions of the right and left molecules in the mixture, and the investigation of the kinetics of the break-up of the racemic state, can also be performed optically, e.g., by measuring the natural gyrotropy or the circular dichroism of the medium, the above stereochemical effect in spontaneous chiral symmetry breaking is directly relevant to the question of polarization instability that we are discussing, and can be described in terms of nonlinear susceptibilities.

The stability of the racemic state under the influence of incident radiation is a question that involves the very basis of organic life, and is related to the problem of the chiral purity of the biological world.

The first publication has appeared[91] on the preferential photodestruction of the right-handed component of the racemate of an aromatic amino acid under the influence of linearly polarized light.

5. DEPOLARIZATION OF RADIATION IN TWO-PHOTON ABSORPTION

The instability of linearly polarized light in an isotropic medium with two-photon absorption was probably first discussed by Ritze and Bandilla[92] (1980), who analyzed the steady-state interaction between light and an ensemble of atoms in the case of transitions between levels with the same total angular momentum $J = 0$ (the Φ-system; see Fig. 11). The single-mode picture cannot account for two-photon absorption. Analysis shows that linearly polarized radiation interacting with the atomic system acquires an orthogonal component due to the nonclassical interference between orthogonally polarized modes with the same frequency and direction, which are absorbed in pairs between the same atomic levels. The quantum effects discussed by Ritze and Bandilla play a significant role in media with strong two-photon absorption. It is well known that "giant" two-photon absorption is observed in cuprous chloride crystals during the photoproduction of the bound states of two free excitons (biexcitons; see, for example, the review by Haug and Kling-

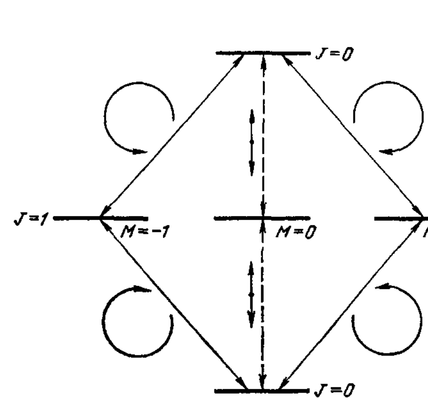

FIG. 11. The Φ-system. The selection rules for two-photon transitions between levels with the same total angular momentum $J = 0$, used in the analysis of problems on the insufficiency of the single-mode picture of two-photon absorption[92] and spontaneous polarization symmetry breaking in two-photon absorption. If the quantization axis lies along the light beam, two-photon excitation of the upper level can occur along two channels via the degenerate levels with $J = 1$, and opposite values of the quantum number M, corresponding to the operator for the z component of the magnetic moment. Interaction between light and the Φ-system produces the depolarized radiation component. If the quantization axis is parallel to the initial polarization, the two-photon transitions occur via the $M = 0$ levels, and there is no depolarized component.

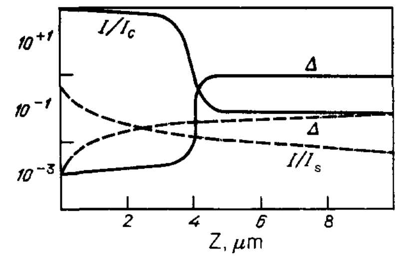

FIG. 12. Polarization instability in CuCl in the case of two-photon absorption. The figure shows the results of the simulation experiment[97] on the degree of depolarization $\Delta(z) = (I_+ - I_-)/(I_+ + I_-)$ and the normalized light intensity $I(z)/I_S$ as functions of the crystal thickness z for intensity $I(0) = 100I_S$ (solid lines) and $I(0) = 0.9I_S$ (dashed lines). The radiation wavelength lies near the biexciton absorption resonance, and the threshold intensity for spontaneous polarization symmetry breaking is $I_S = 0.25$ MW/cm^2. When the crystal thickness is 4 μm, the radiation corresponding to the intensity above the threshold transforms from linearly polarized to circularly polarized. The initial ellipticity of light is $\Delta(0) \sim 10^{-3}$.

shirn[93] (1981). The symmetry properties observed in two-photon transitions in cuprous chloride are analogous to those shown in Fig. 11 (Ref. 94, 1975). It is found that, when the dependence on the exciton and biexciton energy levels and the selection rules for two-photon transitions is taken into account for biexcitons excited in crystals similar to cuprous chloride during the propagation of linearly polarized light under the conditions of strong nonlinear absorption, this leads to hard spontaneous polarization symmetrybreaking [see Cho and Itoh[95] (1984), Inoue[96] (1986), and Kranz and Haug[97] (1986)]. As a consequence of the renormalization (shift) and population of exciton and biexciton states when a certain intensity threshold I_s is reached, two types of solutions of the dispersion relation for the wave vector **k** are found to arise, namely, $\mathbf{k}_{\rm left} = \mathbf{k}_{\rm right}$ (I) (unstable) and $\mathbf{k}_{\rm left} \neq \mathbf{k}_{\rm right}$ (II) stable, i.e., small fluctuations in the linear polarization of incident light produce elliptically polarized radiation at exit from the crystal. Kranz and Haug[97] (1986) estimate that the threshold pump power density is $I_s = 0.25$ MW/cm^2 ($\hbar\omega \sim 2.81$ eV). When the sample thickness is a few microns, and the initial ellipticity amounts to a fraction of a percent, the threshold intensity I_s must be exceeded by a factor of 10–100 to ensure that a practically complete transformation of the polarization of emerging radiation into one of the circular components can be observed (Fig. 12).

A number of the nonlinear-optics polarization experiments with cuprous chloride can be explained, at least partially, in terms of the phenomenon of spontaneous symmetry breaking in two-photon biexciton absorption. Zhdanov *et al.* (1980) have investigated the nonlinear optical activity (polarization self-interaction of linearly polarized light) in single-crystal films of cuprous chloride produced by evaporation in vacuum of recrystallized material of extreme purity onto the ⟨001⟩ cleavage plane of rock salt. The high quality of these single-crystal films, essential for experiments on NOA, was confirmed by electron diffraction studies. The measurements were performed in the intensity range 3–10 MW/cm^2 at the wavelength of 386 mm, using films 3–35 μm thick (Fig. 13). The giant two-photon absorption was observed, but NOA was not measured because the polarization of the radiation leaving the crystal within this intensity range was not at all linear, i.e., the transmission coefficient of the polarizer GP2 was practically independent of its orientation, whereas the extinction coefficient of the polarizer/sample/analyzer system at 0.532 μm wavelength (well away from the two-photon resonance) was 10^{-3}. We are inclined to look upon these data as a manifestation of spontaneous polarization symmetry breaking. Analogous results at helium temperatures were obtained by M. Kuwata (private communication, 1988). Intensity-dependent depolarization in cuprous chloride was also investigated by Itoh and Kahotno[99,100] (1985 and 1982, respectively) and by Kuwata and Nagasawa[101] (1987) (see also the discussion given in Ref. 98 of Ref. 100).

6. POLARIZATION INSTABILITY IN ISOTROPIC MEDIA: ROTATION OF THE POLARIZATION ELLIPSE AND INTERACTION BETWEEN TWO BEAMS

The rotation of the polarization ellipse of a powerful light wave (Refs. 7 and 102, 1964 and 1965, respectively) is the most common and widely investigated polarization phenomenon in nonlinear optics. The effect is possible in media of any symmetry, e.g., in isotropic media. By analyzing a model nonlinear medium consisting of chaotically oriented nonchiral nonlinear molecules[9] Akhmanov *et al.*[77] (1982) showed that, when the light-wave intensity reached $|E|^2 \sim (\chi^{(3)})^{-1}$ the self-rotation of the polarization ellipse ceased to be stable, and a local (for each individual molecule) hysteresis of the polarization response became possible. The effect occurs when the incident radiation is elliptically polarized. Actually, the polarization is unstable and may be randomized for intensities for which the nonlinear increment to the refractive index becomes comparable with the linear component. This condition is often difficult to satisfy because the necessary beam power density lies above the optical breakdown threshold.

Kaplan[4] (1983) and Kaplan and Law[104] (1985) have examined the spatial instability of polarization in the static problem of two counter-propagating waves in an isotropic medium with Kerr and strictional nonlinearity. When the counter-propagating waves are linearly polarized, and the two polarizations are orthogonal or parallel to one another,

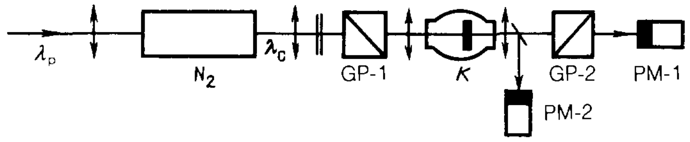

FIG. 13. Block diagram illustrating the experiment on the polarization self-interaction in cuprous chloride. The source of radiation is a liquid-nitrogen cooled SRS oscillator with $\lambda_c = 386.6$ nm, pumped by the third harmonic of the Nd^{3+}:YAG laser; $\lambda_p = 355$ nm; GP-1 and GP-2 are Glan prisms. Polarization measurements are performed with the photomultipliers PM-1 and PM-2, and the epitaxial single-crystal film of cuprous chloride on sodium chloride substrate was cooled in the nitrogen crystal K.

or the waves are circularly polarized with the same or opposite circular polarization, the polarization parameters of the waves do not change during the propagation process, i.e., we have matched pairs of nonlinear eigenpolarizations (in the isotropic nonlinear medium, the nonlinear eigenpolarizations of a progressive wave are circular and linear). Sala[58] (1984) has shown that, in addition, there are two initial elliptic polarization states for which the propagation of the wave in the presence of another strong elliptically polarized wave is not accompanied by a change in the polarization ellipses, which experience pure rotation. In the remaining cases, the interaction between counter-propagating waves [see Wabnitz and Gregori[105] (1986) and Kaplan[4] (1983)] leads to periodic, multistable, and chaotic spatial distributions of polarization in each beam [see also Maĭer[106] (1984) and Daino *et al.*[107] (1985)]. A fundamental step in the investigation of polarization instabilities was taken by Gaeta, Boyd, Ackerhalt and Milonni[5] (see Gauthier *et al.*[160]). They showed that, in an isotropic medium with finite nonlinear response time constants, the "frozen" static spatial polarization distributions that arise as a result of the interaction between counter-propagating waves with time-independent initial intensity cease to be temporally stable. In particular, two important special cases can be defined for the interactions between two waves with the same linear polarization, which, as indicated above, constitute a pair of nonlinear eigenpolarizations of the stationary problem (equivalently to the case of an infinitely "fast" nonlinearity). If the nonlinear

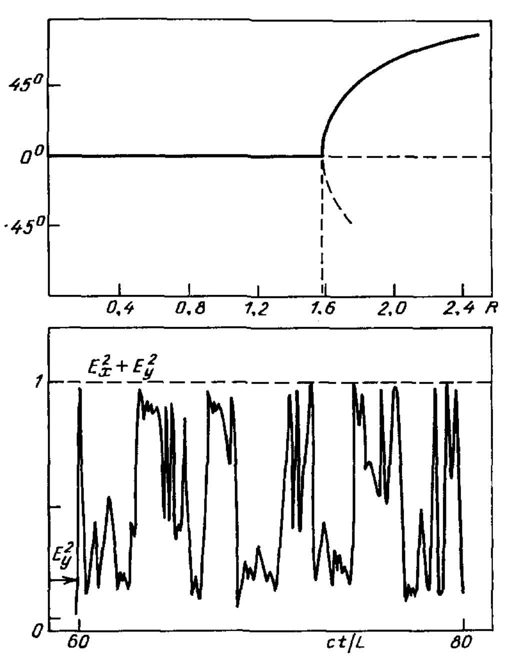

FIG. 14. Spontaneous symmetry breaking during the interaction between two counter-propagating equally linearly polarized waves in an isotropic medium with a finite nonlinear response time constant τ. When the combined intensity of the interacting waves is above the threshold value $R = (|E_f|^2 + |E_b|^2)k\chi^{(3)}_{1111}L \simeq 1.6$, the polarization of each of the waves begins to rotate in the direction determined by the fluctuation "seed". The angle of rotation is shown as a function of R. When the intensity threshold is substantially exceeded, the distribution of polarization within the body of the sample becomes chaotic. In the case of the fast nonlinear response $\tau \ll L/c$ (L is the length of the sample), chaos arises only under asymmetric excitation. The polarization components of one of the waves (E_f) are shown in the figure for $R = 6$ as functions of time (based on Ref. 5).

response time constant τ is close to the time Ln/c taken by light to traverse the nonlinear crystal of length L, the temporal chaotization of polarization occurs for any intensity ratio of the counter-propagating waves. This temporal chaotization of polarization is accompanied by an effective transfer of energy to the polarization component perpendicular to the original polarization, and back. The temporal instability has a threshold and develops as the spontaneous symmetry breaking process (Fig. 14). When the nonlinearity time constant is $\tau \ll Ln/c$, the temporal polarization instability develops only for asymmetric excitation for which the intensities of the counter-propagating waves are initially unequal.

The significance of the results is broader than the specific manifestation that we have discussed.[5] It would appear that in many cases of a finite nonlinearity time constant that is commensurate with the time taken by light to cross the nonlinear sample, the temporal polarization instability that is similar to the "McCall instability" in nonlinear systems which has two characteristic relaxation times,[12] can appear in the system.

Tratnik and Sipe[109] (1987) have introduced the idea of the polarization soliton when they considered the case of counter-propagating pulses with arbitrary distribution of intensity and polarization in a nondissipative isotropic medium with cubic nonlinearity of the strictional type. They noted a class of solutions for which the interaction between the counter-propagating pulses reduced to the rotation of the polarization distribution as a whole. They found the conditions that had to be satisfied by the "pulse area" for two important cases, i.e., when the interaction led to the rotation of the entire polarization distribution by 2π (i.e., the pulses are restored and we have "polarization transparency") and when the polarization switched to the orthogonal direction (this case is important for the development of devices for controlling light by light).

The phrase *polarization domain* was introduced by Zakharov and Mikhailov[110] (1987) in relation to the interaction between counter-propagating waves in isotropic media and in media with a fourfold symmetry axis. The polarization domain is understood to be a region with a stable polarization state. The polarization-switching region constitutes the domain walls, and it was shown that the rate of displacement and the size of the domain walls depended on the amplitudes and frequencies of the interacting waves.

Xuan *et al.*[111] (1983) have observed the polarization instability of elliptically polarized picosecond pulses (wavelength 0.532 μm) in nitrobenzene, CS_2, and a number of other liquids for intensities of 100–2000 MW/cm^2, but below the threshold for stimulated Raman scattering. One of the possible mechanisms for the polarization instability can, in our view, be the interaction between the elliptically polarized pump wave and the counter-propagating waves reflected from the walls of the chamber, i.e., the chaotic polarization distribution that is produced in this case within the liquid in the "time overlap" zone of the incident and reflected waves should be very sensitive to small changes in the pump-beam intensity, even when the polarization parameters integrated over the pulse are measured.

Leaving on one side fiber lightguides, in which self-rotation of the polarization ellipse is difficult to investigate because of growth and strain birefringence (Sec. 7), we note that the most convenient model object for the investigation of the interaction between elliptically polarized beams is provided by liquid crystals in which threshold polarization effects can be observed at low levels of optical interaction.[158]

7. POLARIZATION INSTABILITY IN BIREFRINGENT MEDIA

From the practical point of view, the weakly birefringent fiber lightguide is an important object in which nonlinear-optics polarization effects can be investigated. Birefringence in fiber lightguides may be random, due to fabrication imperfections or strains produced during assembly in a particular device, but it can also be introduced deliberately, e.g., to preserve the polarization of radiation. The question of polarization instability in birefringent media was purely academic only three or four years ago. Sala[58] (1984) was the first to draw attention to the "trigger" behavior of the polarization of light in a birefringent crystal. Winful[112,113] (1985) and (1986), respectively, Wabnitz *et al.*[114,115] (1986), and Roman *et al.*[116] (1986) have formulated the criteria for, and the basic manifestations of, polarization instability in this system. The dependence of the refractive index on the wave intensity leads to a loss of stability along the "fast" birefringence axis (Fig. 15). The effect has a threshold (hard spontaneous polarization symmetry breaking), i.e., when the intensity threshold has been reached, a slight change in the linear polarization parallel to the "fast" birefringence axis leads to large changes in the polarization parameters at exit from the anisotropic medium. The threshold intensity can be estimated from the formula

$$I_s \sim n n_2^{-1} \Delta n$$

where n and n_2 are the linear and nonlinear refractive indices, respectively, and Δn is the birefringence of the medium. We note that the above condition for I_S actually shows that the instability arises for intensities that produce a nonlinear change in the refraction of In_2 of the order of the linear birefringence Δn. Consequently, the effect is significant in media with small birefringence (large natural birefringence will "stabilize" the eigenwaves).

In our view, the most interesting objects for the observation of spontaneous polarization symmetry breaking in the form of the loss of stability along the "fast" birefringence axis are crystals with an "isotropic point," i.e., crystals for which the curves representing the frequency dependence of the refractive index for O and E waves are found to cross. Many of the materials employed in nonlinear optics ZnO, ZnS, CdSe, SnO_2, MgF_2, Al_2O_3, $AgGaS_2$, and CdS have an "isotropic point" in the transparency region. When CdS is employed at room temperature, the "isotropic" wavelength is 522 nm, and the dispersion of birefringence is

$$\eta = \frac{\partial n}{\partial \nu} = 3{,}4 \cdot 10^{-5}\ \text{cm}.$$

We can hypothesize that the threshold for the observation of spontaneous symmetry breaking can be reduced as much as desired by approaching the isotropic point as closely as necessary in frequency. Assuming that the most important limitation is the spectral width $\Delta\nu$ of the laser radiation, we obtain the following realistic estimate:

$$I_s \sim n\eta\Delta\nu \cdot n_2^{-1},$$

which, for a spectrum with a width of 100 MHz ($\Delta\nu \sim 3\cdot 10^{-3}$ cm^{-1}), gives $I_S = 80$ kW/cm^2. (Data on nonlinear refraction in CdS were taken from Ref. 117.)

Trillo *et al.*[118] (1987) have observed the polarization instability in single-mode fiber lightguides with a core diameter of 4.5 μm and a length of 53 cm. The spatial period of beats determined by the natural birefringence of the fiber was 90% of its length. The light-wave power necessary for bifurcation was of the order of 100 W (Fig. 3). The fact that hard spontaneous polarization symmetry breaking can be observed in a medium with weak birefringence does not mean (as in any other case) that spatial polarization chaos becomes possible: the number of integrals of motion is insufficient in this case for the complete integrability of the equations.

An interesting situation arises when birefringence and gyrotropy are combined. Gyrotropy can arise in a fiber lightguide as a result of helical strain, e.g., when the fiber is coiled, or when it is wound on a rod or twisted. It has been shown experimentally that gyrotropy of up to some hundreds of radians per meter of the fiber is equivalent[119] in magnitude to helical deformation.[10]

Matera and Wabnitz[120] (1986) have shown that the variation in the polarization parameters of a powerful light wave in a system of this kind is exceedingly complicated. For example, the evolution of the component S_3 of the Stokes

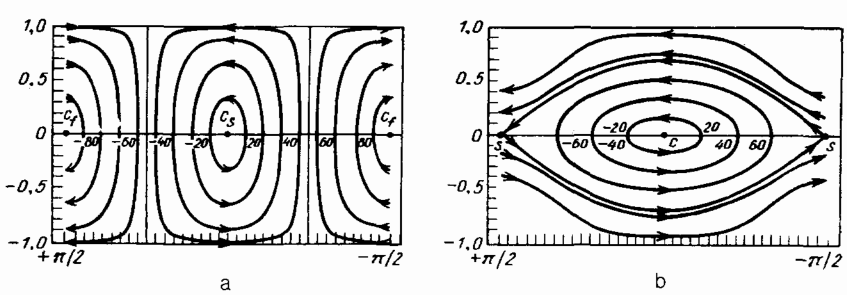

FIG. 15. Spatial instability of polarization in birefringent crystals: phase trajectories for the polarization parameters of radiation propagating in a birefringent crystal.[114] The Stokes parameter S_3/S_0 is shown along the vertical axis ($S_3/S_0 = 1$ for right circularly polarized wave and $S_3/S_0 = -1$ for left circularly polarized wave; $S_3/S_0 = 0$ for linearly polarized light), and the angle $\theta = \text{arctg}(E_f/E_S)$, is plotted along the horizontal axis and defines the orientation of the principal axis of the polarization ellipse relative to the slow birefringence axis. a—"Linear optics"; $|E|^2 \ll \Delta n/n\chi^{(3)}$; polarizations parallel to the fast and slow axes are at the centers of the phase trajectory; b—"nonlinear optics"; $|E|^2 \gg \Delta n/n\chi^{(3)}$; the polarization ellipse oscillates around the direction of the slow birefringence axis; the fast axis transforms into a saddle, and the polarization along this axis becomes unstable.

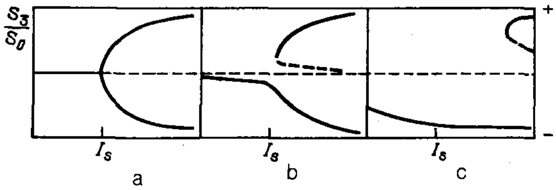

FIG. 16. Polarization instability in a weakly birefringent and gyrotropic medium.[120] The ratio of the components of the Stokes vector of the electromagnetic wave, S_3/S_0, at exit from the fiber lightguide is shown as a function of the light intensity $|E|^2$. The incident polarization is linear; a—lightguide without torsion; when $|E|^2 = |E_S|^2 \sim \Delta n/\chi^{(3)}$; spontaneous polarization symmetry breaking is observed; b—weak helical deformation (one complete twist per 200 beat lengths) and polarization bistability with soft excitation of right circular polarization is observed; c—highly twisted fiber; helical strain—one twist per beat length $L = 2\pi\lambda/\Delta n$.

vector was reduced to an equation similar to the equation of motion of a unit mass in a central potential described by a fourth-degree polynomial. It is well known that finite trajectories are not closed in polynomial potentials (see, for example, Ref. 1), i.e., the variation in the polarization parameters is spatially nonperiodic. The question therefore is: what are the consequences of gyrotropy in hard spontaneous polarization symmetry breaking in birefringent media? It is found that even slight torsion of the fiber lightguide leads to the "softening" of the bifurcation curve, i.e., at exit from the medium, and as the intensity increases, the polarization parameters rapidly reach "circular" values (Fig. 16; cf. chiral selectivity in stereochemistry, discussed in Sec. 3). The stabilization of polarization states by torsion in fiber lightguides with linear, nonlinear, and random coupling between polarization modes was investigated by Vatarescu[121] (1987).

We note that the equations describing the twisted, slightly birefringent, fiber lightguide are analogous to the set of relations between the polarization parameters and the intensity of a light wave in a birefringent crystal with chiral coupling between the O and E modes near the isotropic point at which the frequency dispersion curves for the ordinary and extraordinary refractive indices are found to cross.[122] In crystals, the strong frequency and temperature dependence of birefringence ensures that the spectrum of possible nonlinear-optics effects is much richer, i.e., the spectral-polarization instability and the temperature instability due to the self-heating of the crystal by the laser radiation (by analogy with the effect discussed in Sec. 3) become possible.

The nonlinear optics of birefringent fiber lightguides with a periodic modulation of the refractive index has been discussed by Wabnitz[123] (1987), Mecozzi *et al.*[124] (1987), and Caglioti *et al.*[125] (1987). Polarization chaos is possible in these systems in a number of cases. A chaotic distribution appears when the intensity approaches its threshold for spontaneous symmetry breaking. By analogy with the nonlinear oscillator, we can show that periodic modulation of birefringence is analogous to a periodic external force that stimulates the oscillator to chaotic motion. Polarization switching in a periodically twisted fiber lightguide has been observed,[126] using a tunable picosecond dye laser. A significant effect was achieved for a peak power of about 1 kW.

The above hard spontaneous polarization symmetry breaking in a weakly birefringent medium is a clear example of spatial instability. Blow *et al.*[127] (1987) have extended the formulation of the problem, and being interested in the propagation of solitons in slightly birefringent fibers, they tackled the problem from the space-time point of view. The actual analysis was performed by direct numerical solution of a pair of coupled nonlinear Schröedinger equations. Strong analogy with the stationary state was found for the case of soliton propagation. If the length of beats associated with birefringence is small in comparison with the soliton period, both soliton modes are stable. For large beat lengths (weak birefringence), the fast soliton mode is unstable and the energy transfers to the slow mode during the propagation process. We see here the emergence of an important difference as compared with the stationary case, when the loss of stability leads to a change in the phase relations between the eigenmodes and, as a consequence, to a change in the polarization states of light at exit from the fiber without a monodirectional energy transfer between the orthogonal components. This exclusion of monodirectional energy transfer between the orthogonal modes (when only the linear coupling is taken into account) is due to the existence of an integral of the motion. In the soliton problem, there is an additional degree of freedom that decouples the integral of the motion. It arises when dispersion is taken into account: in the case of weak birefringence, the energy is monodirectionally "pumped" into the slower mode. When dispersion is taken into account, a nearly chaotic transfer of energy is possible under certain particular conditions between the initial polarization soliton modes: by observing the pulse amplitude in one of the modes at different points in the fiber (in a gedanken experiment), we can see the continuous distributions over the spatial frequencies. (In this connection, see also the paper by Wabnitz,[128] who shows that, when frequency dispersion is taken into account, this leads to a loss of polarization stability along the "slow" birefringence axis of the fiber optical lightguide, as well.) Vatarescu[129] (1986) has established that energy transfer between modes is also possible in the stationary case without frequency dispersion when the nonlinear coupling between the eigenstates is taken into account.

The chaotic behavior of polarization in the case of counter-propagating waves in a birefringent medium is not limited by the integrals of the motion. This fact was established by Tratnik and Sipe[62] (1986). Vatarescu[130] (1986) has noted the possibility of spatially nonperiodic energy transfer between the two polarization modes in the case of the counter-propagating light waves in a highly birefringent optical fiber. Trillo and Wabnitz[131] (1987) carried out a numerical simulation and found a chaotic distribution of polarization for two counter-propagating waves. Moreover, chaos was possible within the limits of one order of magnitude of intensity near the threshold for spontaneous polarization symmetry breaking, i.e., for low intensities, intrinsic birefringence will stabilize polarization, thus allowing only regular beats and, contrariwise, for high intensities, the nonlinear increment on the refractive index is sufficient to give rise to chaotic motion. Polarization instability in birefringent lightguides and its practical applications to devices for controling light by light are discussed in Refs. 132–135.

In the case of large birefringence, it is probably impossible to observe spontaneous polarization symmetry breaking in the form of a loss of stability along the "fast" birefringence axes in crystals, but a nonlinear birefringent crystal placed between polarizing prisms is an interesting optical object.

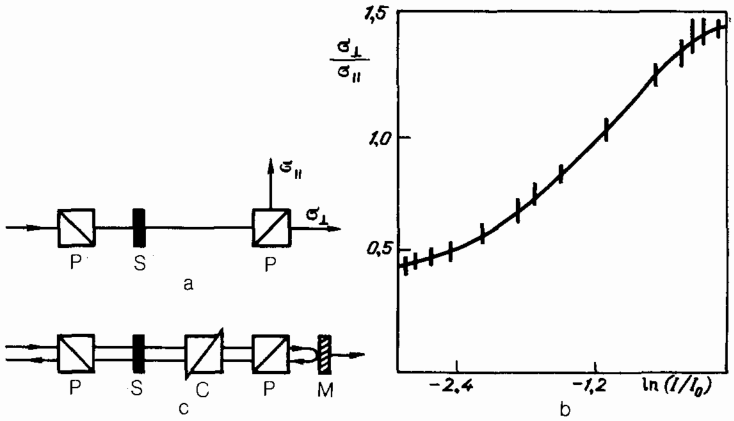

FIG. 17. Polarization devices for controlling light by light, based on Wood's filter:[136] a—optical arrangement of the filter, P—polarizing prisms, S—sample of birefringent crystal; b—energy ratio for pulses with orthogonal polarizations at exit from the nonlinear crystal as a function of the intensity of exciting radiation; crystal length 1.2 mm, pulse length 30 ps, wavelength $\lambda_0 = 532$ nm; there is a clear variation in the ratio of the intensities of orthogonal components with increasing pump power density ($I = 800\ \mathrm{MW/cm^2}$); the departure of the curve from the $\cos^2[(I/I_0) + \varphi_0]$ law is due to the nonlinear absorption of radiation and the averaging of the effect over the packet of picosecond pulses;[137] c—optical arrangement of the bistable polarization device without a resonator and incorporating a birefringent nonlinear crystal; *M*— mirror, *C*—Babinet compensator, whose setting selects the state of the bistable element.[136]

We recall that, if the optic axis of a birefringent crystal is perpendicular to the beam, and set at 45° to the polarizer in front of the crystal and to the analyzer after it, the system is known as Wood's filter. It is clear that the transmission spectrum will be transformed when high-intensity light beams are employed because crystal refraction is very intensity-dependent. This question has been examined in detail by Otsuka *et al.*[136] (1985). The transmission of the filter is given by the simple formula

$$T = \cos^2\left(\frac{I}{I_0} + \varphi_0\right);$$

where I is the incident intensity. The modulation of the transmission coefficient is significant when the intensity becomes

$$I \sim I_0 \sim (n_2^{(E)} - n_2^{(0)})\,\lambda L^{-1},$$

where $n_2^{(0)}$, $n_2^{(E)}$ are the nonlinear refractive indices for the O and E modes, respectively. Figure 17 shows the results obtained by Aleksandrovskiĭ *et al.*[137] (1984) on the effect of incident radiation power on the parameters of Wood's filter.

Otsuka, Yumoto, and Song[136] were the first to show that, when a fraction of the transmitted radiation was reflected back into the crystal, it was possible to achieve an optical polarization-bistable device demonstrating the hysteresis states for pump intensities in the region of I_0.

The view of the present author is that optic-fiber devices without resonators, which exploit polarization switching in weakly birefringent systems, will find actual practical applications in the near future in communication devices with polarization data coding. The next step will be the transition to compact polarization switches, using highly nonlinear gyrotropic and nongyrotropic crystals with an isotropic point. The advances achieved in recent years in the technology of semiconducting materials will ensure that the composition of solid solutions can be adjusted by displacing the isotropic point toward the lines of popular laser sources. The tuning of an optical filter incorporating the $CuAlSe_2$ gyrotropic crystal with an isotropic point and the polarization modulation of radiation at 0.532 μm have recently been observed by Zheludev, Makovetska, Popov, Semenikhin, and Tarasenko (1988).

8. SOME REMAINING QUESTIONS AND FUTURE PERSPECTIVES

We shall now briefly enumerate the questions that have not been discussed in detail in this review. Zartov, Panajotov, and Peyeva[138,139] (1986 and 1987, respectively) have examined a hybrid metastable optical polarization device in an electrooptic modulator used in the Fabry-Perot interferometer with electrooptic feedback, which is controled by the intensity or polarization of the pump. They have also considered a multistable polarization device based on an electrooptic modulator without a resonator[140] (1987). Korpel and Lohmann[141] (1986) have introduced a configuration based on Fabry-Perot resonators and anisotropic polarization elements, e.g., full-wave plates. The first experiments on polarization bistability in the Fabry-Perot resonator containing a birefringent element and a polymeric film with a strong thermal nonlinearity were carried out by Cush and Kirkby[142] (1986) [see also Indebetouw[143] (1988)].

Zon and Kupershmidt[144] (1984) have carried out a theoretical analysis of bistability in the magnetization of a gas of free electrons or a semiconducting crystal in the case of the inverse Faraday effect. Since this bistability can be probed optically by observing the circular magnetic dichroism or Faraday rotation of a low-intensity probe wave, it is possible to develop a birefringent optical scheme for recording optical polarization instability.

In our view, the stability of light-wave polarization in two-beam systems deserves attention. Zheludev, Ruddock, and Illingworth[87] (1987) were the first to investigate experimentally the chaotic response in a solid-state system without a resonator. The system was based on a semiconducting cluster glass in a two-beam scheme, and exploited the competition between two nonlinear mechanisms and optical excitation by modulated radiation. Remarkably, the polarization-sensitive effect of thermally induced birefringence was used in this experiment to detect the orthogonal component of the probe beam. This meant that it was possible to observe the different regimes of subharmonic and chaotic response with enhanced contrast, i.w., chaos could be observed in both amplitude and polarization of light.

An interesting series of investigtions involves the study of two-beam systems in media with nonlinear gyrotropy. The starting point here are the investigations of Harris[145] (1976), Tinoco[146] (1976), Belyĭ, Serdyukov and Bokut'[147] (1975), Golubkov and Makarov[148] (1976), and Ananasevich, Zheludev, and Dovchenko[149] (1986).

It has not been our aim in this paper to provide a complete review of publications on polarization instability in active systems, i.e., in laser amplifiers and oscillators. We have confined our attention to noting the overall properties of active and passive systems, namely, the possibility of polarization *multistability and oscillatory and stochastic regimes.* Nasyrov[150] (1982) has examined the propagation of an electromagnetic wave in the active medium of a gas laser. He finds that, in the amplifying medium, the polarization of radiation tends to the linear state for $J \to J \pm 1$ transitions, and to the circular state for the transition with the conservation of total angular momentum ($J \to J$). The opposite situation occurs in absorbing media. When counter-propagating waves interact in the double-transit amplifier, this results in polarization oscillations with a period equal to the time for two complete transits of radiation through the active system. The necessary condition for the observation of the effect is that the intensity gain must be of the order of a few tens.

Krivoshchekov *et al.*[151] (1982) have investigated polarization instability in the gas laser. Semiconductor lasers have been considered by Chan and Liu[152] (1987) and by Sapia *et al.*[153] (1987).

We have ignored the polarization instability of liquid crystals, i.e., media with exceptionally high optical nonlinearities. This question has been reviewed in a recent paper by Arakelyan[154] (1987).

As far as the practical applications of polarization instability and multistability are concerned, we note that, in polarization devices for controling light by light, the coding of data (signal) relies on the polarization state of light, i.e., on phase modulation. In optics, polarization modulation often has important advantages as compared with amplitude modulation. These advantages are, first, the greater contrast of data signal variation in switching from one stable state to another; second the analogy with the phase multistabilities[11] of a symmetric many-level system for coding and counting, e.g., 1, 0, -1 (right ellipse, linear polarization, left ellipse); third, polarization switching does not involve loss of intensity, which means that it will be possible to develop complex cascade logic devices that do not require intermediate light amplifiers [Ref. 155 (1985)].

In the case of local nonlinearity, polarization multistability can be observed, as noted above, for light intensities $I_S \sim n_2^{-1}$. The interference gain in intensity is of the order of $Q\lambda/d$ and arises when the nonlinear medium is placed in the resonator (d is the resonator length and Q is its Q-factor). Spatial polarization instability in birefringent media with feedback and spontaneous polarization symmetry breaking in the form of loss of stability by "fast" polarization occur for intensities $I_S \sim \Delta n n_2^{-1}$. An intensity criterion for the temporal polarization chaos in the interaction between counter-propagating waves in a medium with a "noninstantaneous" nonlinearity is also quite realistic: ($I_1 + I_2 \sim n_2\lambda/L$) ($L$ is the length of the nonlinear medium). It is therefore clear that, in many cases, the gain as compared with local non-linearity can be 10^4–10^6, which ensures favorable conditions for the observation of polarization instability in resonator and nonresonator distributed system. This offers a considerable margin as far as the light-beam power is concerned, when we take into account limitations on the self-focusing threshold and optical breakdown.

Polarization chaos is often a purely classical effect, similar to the chaotic behavior of nonlinear dynamic systems with several degrees of freedom. At this point, and as part of our discussion, it is appropriate to introduce Lipkin's viewpoint[156] (1977), which, in our opinion, is not well founded. He considers that the existence of unpolarized (solar) light *is one of the arguments in favor of corpuscular theory:* "the existence of unpolarized light already gives an indication of the quantum nature of light. Unpolarized light cannot be described as a single simple classical monochromatic wave or as any linear combination of such waves. Unpolarized light can be described classically as a series of very rapid short bursts or pulses of light, each having a different polarization, with no correlation between the polarizations of different pulses.If these pulses and the interval between them are very short compared to the characteristic working times of the measuring apparatus, they will be detected as a continuous beam and any polarization measurement will give an average of the polarizations of the individual pulses." This definition of unpolarized or partially polarized light can equally well be looked upon as an intuitive definition of polarization chaos (partially polarization-randomized radiation). On the other hand, is there *really* an obvious physical difference between these two concepts? Whether the researcher observes a mixture of uncorrelated wave trains of different polarization or temporal polarization chaos that is a consequence of the stochastization of the nonlinear response of the dynamic system, he is actually concerned at each instant of time with the electric (or magnetic) field vector of the wave. In both cases, the recorded parameters are the intensities, e.g., of the mutually orthogonal field components between which there is no constant phase difference in either case. A difference can appear only in quantitative parameters, namely, in the duration and shape of the corresponding mutual correlation functions for the orthogonal intensity components. Thus, the existence of unpolarized light cannot, in itself, be a sufficient condition for the demonstration of its quantum nature. On the other hand, it has recently been shown by Kennedy and Wabnitz[157] (1988) that polarization instability in nonlinear system can be the reason for the formation of "squeezed" (subquantum or noiseless) states of different polarization components of radiation.

The detailed statistical properties of polarization chaos constitute an unsolved problem. Here, we must pass from the analysis of realizations produced by numerical simulation to a search for regular procedures for calculating global parameters, e.g., one-dimensional and multidimensional probability distributions for the Stokes parameters, and to a study of the transition to polarization chaos as a phase transition.

Even the very earliest papers[13] noted the analogy between spontaneous polarization symmetry breaking in atomic transitions and butterfly-type topological singularities in catastrophe theory. The results of a number of other investigations are being formulated within the framework of catastrophe theory. For example, the topological cuspidal-edge singularity corresponds to the polarization bistability in a cubic crystal placed in a Fabry-Perot resonantor.

A completely new range of investigations into the polarization instabilities in the plane perpendicular to the beam in nonlinear optical systems with coherent feedback is emerging.[108] The problem here is to examine two-dimensional polarization distributions over the cross section of the light beam, including both highly symmetric polarization structures and polarization chaos over the cross section of the beam and (or) in time.

9. CONCLUSION

Studies of polarization multistability, which began in the early 1980's with two completely dissimilar and independent papers (Refs. 13 and 77, 1981 and 1982, respectively) have now merged into one of the most dynamic branches of nonlinear optics, which has attracted the attention of at least twenty major laboratories across the world. Quite unexpectedly, it now occupies a central position in many topical areas of scientific enquiry, ranging from the chiral purity of the biological world to ultrahigh-speed data transmission, the theory of dynamic chaos in nonlinear systems, and fundamental problems in classical and quantum statistics of light. Striking analogies are emerging between instability in nonlinear optics and well-known traditional problems in mechanics, such as the dynamics of the nonlinear oscillator, the motion of cosmic objects in the field of mutual attraction, the stability of helical and longitudinal deformations, and the dynamics of a propeller-driven airplane and a rigid body with one fixed point.

Research into polarization instabilities has now emerged from the phase of theoretical prediction and numerical simulation, and has entered the stage of experimental investigation, indicating possible serious practical applications to optical data processing.

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1)The role of parity nonconserving weak interactions in spontaneous symmetry breaking will be discussed in Sec. 4.

2)We note that the theory of Kitano *et al.*, based on the equation of balance, has a considerable heuristic value and predicts correctly the basic features of optical tristability, but is not valid for the intensity values necessary for the effect to be observed. A more adequate theory was developed later by Savage *et al.*[14] (1982) and is discussed in detail by Hamilton[15] (1983).

3)When second-order effects due to spatial dispersion are taken into account, weak birefringence is possible in cubic crystals and has been seen experimentally (see Ref. 40 for further details). First-order dispersion effects are examined in Sec. 4.

4)A detailed review of the factors leading directly to the nonlinear anisotropy is outside our scope here. The details can be found in Refs. 42, 48, and 50.

5)The transformation of the linear polarization is also found to occur on reflection from a cubic crystal. However, here, $\mathrm{Re}\Delta\chi^{(3)}$ determines the self-induced rotation and $\mathrm{Im}\Delta\chi^{(3)}$ the self-induced ellipticity. The effect has been confirmed experimentally by investigating reflection from the ⟨001⟩ plane of the GaAs crystal [see Ref. 52 (1988)].

6)The Stokes 4-vector formalism is a possible and very convenient method of describing the polarization of light (see, for example, Born and Wolf[56] (1973) and Rozenberg[57] (1977). If the field components of the electromagnetic wave are given in the Cartesian system, the Stokes 4-vector is given by $S_i = \mathbf{E}\hat{\sigma}_i\mathbf{E}^*$, where $\hat{\sigma}_i$ are the Pauli spinor matrices

$$\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}, \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}, \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix}, \begin{pmatrix}0 & -i\\ i & 0\end{pmatrix}.$$

7)Classical oscillator models of a nonlinear medium that can be used to describe amplitude effects in intrinsic bistability have been used by a number of authors (see, for example, Flitzanis and Tang[78] (1980), Goldstone and Garmire[79,80] (1984), and Golubkov, Makarov, and Matveeva[81] (1987).

8)The symmetry of other optical electromagnetic polarization effects is discussed in detail by Baranova *et al.*[82] (1977), whose terminology we employ here.

9)We recall a relatively close classical analogy from the theory of elasticity[2] (1965), i.e., that of a rod of circular cross section subjected to torsion. When the torsional moment reaches a certain critical value, the rectilinear rod becomes unstable (an example is provided by the fully wound rubber band in a toy airplane with a rubber-band driven motor, which tend to "kink". If we continue this analogy, we can compare the polarization instability in gyrotropic media with the torsional strain of a coiled telephone cable. The coil "kink" is an example of the instability of a chiral object. We note that an ensemble of microscopic coils is a possible model of both natural gyrotropy and of nonlinear optical activity[103] (1979).

10)This is a surprising example of a system that is completely free from nonlinear gyrotropy, i.e., the dependence of the rotatory power of the medium on intensity. A twisted fiber lightguide can be well simulated by a stack of wave plates turned clockwise relative to one another in the direction of propagation of light.

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