Optical bistability caused by self-induced anisotropy of light absorption in cubic crystals

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It is shown that the presence of unstable directions of the polarization plane of intense radiation may lead to a new type of optical bistability (OB) (the instability of specific directions arises in nongyrotropic crystals from the self-action of radiation propagating, for example, along a fourfold axis). The case of resonant nonlinear absorption of light is discussed in detail. An analysis is given for the symmetry-caused extrema of the dependence of the field strength at which new stationary states arise on the orientation of the plane of polarization. Near an extremum, this dependence may be both analytic and nonanalytic, corresponding to different mechanisms of OB. The stability of stationary states is discussed by taking the field inhomogeneity into consideration. The results are applied to specific mechanisms of nonlinearity, and in particular, to two-photon resonant absorption.

The dependence of the absorption of light on its intensity is a well-known nonlinear optical effect. Nonlinear absorption has been observed in noncrystalline substances as well as in crystals (see for example Ref. 1). In crystals, this absorption is generally anisotropic. It is significant that such anisotropy, i.e., self-induced dichroism (SID), is present even in cubic crystals (linear not circular dichroism is considered everywhere below; spatial dispersion is not considered). Since cubic crystals are isotropic in weak fields, they are suitable for an experimental study of SID.[2–4]

As in dichroism in linear optics, in SID there are isolated limiting directions toward which the plane of polarization turns as radiation propagates in the crystal. It is significant, however, that for intense radiation propagating along an axis of symmetry of order higher than second, there are several such limiting directions. In fact, the directions obtained by rotation about a symmetry axis of the crystal or by reflection in a plane passing through the axis are equivalent, and if one of them is a limiting direction, the others are also limiting. The rotation to one or the other of the equivalent limiting directions is determined by the initial orientation of the plane of polarization.[2]

Obviously, under the indicated conditions, there are always directions which separate the regions of "attraction" to different limiting directions. They are polarization-unstable: slight deviations of the plane of polarization from them increase with the thickness of the crystal (during fluctuation of polarization, however, they generally do not increase). In the presence of a resonator, when the transmitted radiation returns to the crystal, the fluctuational deviation from the unstable direction, as will be shown below, can increase in time. As a result, in the steady state, the plane of polarization at the resonator output turns out to be deflected through a finite angle toward either one or the other limiting direction, i.e., bistability takes place.

The dichroic bistability (DOB) described is due to the self-induced change of radiation polarization in a nonlinear medium. The polarization effects in general enrich substantially the picture of optical bistability. Interesting examples of systems with a specific type of nonlinearity, where these effects are appreciable, have been discussed recently,[5–7] and the optical bistability predicted in Ref. 5 and due to the degeneracy of the atomic energy levels in a gas has already been observed experimentally.[8]

In cubic crystals, the role of polarization effects is particularly important because of the presence of unstable directions of the polarization plane (in the presence of nonlinearity of both light absorption[2] and refraction[9]). As a result, qualitatively new mechanisms of bistability arise, in particular, DOB, and the threshold value of the radiation intensity sharply decreases in many cases.

The analysis of bistability in cubic crystals can be successfully performed in a very general form, without specifying the nonlinearity model, by studying the polarization dependence of the field intensity at which new stationary states arise. As a result of symmetry, the corresponding function has singular points and extrema, the behavior in the vicinity of which determines the bistability mechanism. Such an analysis is given below for radiation propagating along a fourfold axis. The mechanism of optical nonlinearity is considered to be resonant, and it is assumed that the absorption depends on the intensity, and that the nonlinearity of refraction for linearly polarized radiation may be neglected. A ring cavity is chosen as the resonator model.

Section 1 discusses the condition of appearance of several stationary states of the field in the cavity (the equation of the bifurcation surface has been derived, and in the case of linearly polarized radiation, the equation of a bifurcation curve). Section 2 gives a general analysis of self-induced dichroism and establishes the symmetry properties of bifurcation curves. Section 3 shows that in the vicinity of symmetry-caused extrema bifurcation curves can be both analytic and nonanalytic (the second case corresponds to the appearance of DOB). In Sec. 4, the results obtained form the basis of

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a discussion of bistability for two specific models of nonlinear absorption. Section 5 discusses the stability of stationary states. Section 6 contains concluding remarks.

1. CONDITIONS OF APPEARANCE OF SEVERAL STATIONARY STATES

The field $\mathbf{E}(0)$ of the forward edge of the crystal in a ring cavity is made up of two quantities: the field ε of incident radiation which has entered the cavity and the field $\mathbf{E}(d)$ of radiation reflected by the mirror system at the output of the crystal:

$$\mathbf{E}(0)=\boldsymbol{\varepsilon}+R\exp(i\varphi_R)\mathbf{E}(d). \tag{1}$$

Here R is the resultant reflectance of the mirrors, and φ_R is the phase shift resulting from the reflections and propagation of light through the cavity. Equation (1) is written for the stationary case and neglects the oppositely traveling wave in the cavity.

The relationship between the field at the output $\mathbf{E}(d)$ and input $\mathbf{E}(0)$ of the crystal is determined by material equations, and is very complex when nonlinearity is considered. In the absence of spatial dispersion for radiation propagating along the [001] axis (z axis), this relationship can be described phenomenologically by means of the two transmittances T_x and T_y:

$$E_\varkappa(d)=T_\varkappa E_\varkappa(0), \quad \varkappa=x, y \tag{2}$$

[the x, y axes in the (001) plane are chosen along symmetry axes of type $\langle 100\rangle$ or $\langle 110\rangle$]. The transmittances T_x, T_y depend on the field $\mathbf{E}(0)$ incident on the crystal and on the crystal thickness d. Let us emphasize that relation (2) covers the case of arbitrary, including strong, optical nonlinearity and reflects the fact that because of symmetry $E_\varkappa(d)=0$ when $E_\varkappa(0)=0$. In strong fields, $E_\varkappa(d)$ generally depends on all the components of vector $\mathbf{E}(0)$, and as a result of self-induced optical anisotropy $T_x\neq T_y$.

Equations (1) and (2) relate the field of radiation transmitted through the cavity [proportional to $\mathbf{E}(d)$] to the field of radiation incident on the cavity (proportional to ε). The relationship of $\mathbf{E}(d)$ to ε can be nonunique. The bifurcation values of the field at which two branches of solutions (1), (2) merge (or appear) are determined from the condition

$$\partial(\varepsilon_x, \varepsilon_y)/\partial(E_x(d), E_y(d))=0. \tag{3}$$

If the dependence of $T_{x,y}$ on $\mathbf{E}(0)$ is unique (i.e., there is no local optical bistability of the crystal), the criterion (3) assumes the form

$$D=\frac{\partial(\varepsilon_x,\varepsilon_y)}{\partial(E_x(0),E_y(0))}=0, \quad \varepsilon_\varkappa=E_\varkappa(0)\,(1-RT_\varkappa\exp(i\varphi_R)),$$
$$\varkappa=x, y. \tag{4}$$

The solution of Eqs. (4) describes a surface in the space ε, Ψ, Φ, where Ψ is the slope of the major semiaxis of the polarization ellipse of the incident radiation relative to the x axis, and Φ is the phase difference of the ε_x, ε_y components. Its form is very complex. Below we shall examine the relatively simple case of linearly polarized incident radiation ($\Phi=0$), which is of independent physical interest. The radiation in the cavity may also be assumed linearly polarized. In the presence of self-induced anisotropy, this radiation can only take place if the nonlinearity of the refraction in the crystal is negligible. This in turn takes place in the presence of strictly resonant optical nonlinearity (for example, if the light frequency ω or its overtone lies at the center of the extrinsic absorption band), when

$$\mathrm{Im}(T_y/T_x)=0 \quad \text{at} \quad \mathrm{Im}(E_y(0)/E_x(0))=0. \tag{5}$$

With (5) taken into account $T_\varkappa$ may be represented in the form

$$T_\varkappa=t_\varkappa\exp(i\varphi_d), \quad t_\varkappa=t_\varkappa(E,\psi),$$
$$E\equiv E(0)=[\,|E_x^2(0)|+|E_y^2(0)|\,]^{1/2}, \tag{6}$$
$$\psi\equiv\psi(0)=\mathrm{arctg}\,[E_y(0)/E_x(0)\,],$$

where the quantities $t_x(E,\psi)$, $t_y(E,\psi)$ are real and determine the transmission amplitude, and the phase φ_d depends only on the crystal thickness d [and not on $E(0)$].

If the cavity is tuned to resonance ($\varphi_R+\varphi_d=2\pi n$) and the incident radiation is linearly polarized, (4) reduces to the condition

$$D\equiv D(E,\psi)=0,$$
$$D(E,\psi)=-\left(1-Rt_x-RE\frac{\partial t_x}{\partial E}\right)\times\left[R\frac{\partial t_y}{\partial\psi}\sin\psi-(1-Rt_y)\cos\psi\right]\cos\psi$$
$$+\left(1-Rt_y-RE\frac{\partial t_y}{\partial E}\right)\left[R\frac{\partial t_x}{\partial\psi}\cos\psi+(1-Rt_x)\sin\psi\right]\sin\psi, \tag{7}$$
$$t_{x,y}=t_{x,y}(E,\psi), \quad \varepsilon_\varkappa=E_\varkappa(1-Rt_\varkappa).$$

These conditions describe the relationship between the bifurcation value of the field intensity on the front face of the crystal $E\equiv E(0)$ and the inclination ψ of its polarization vector, as well as the relationship between the bifurcation values of the corresponding parameters of the external field, ε and Ψ. When the bifurcation curve crosses $\varepsilon_B(\Psi)$ the number of stationary solutions (1) with linear polarization changes by a factor of two. Equation (7) also determines the region of the parameters of the system (crystal thickness, etc.) in which bistability for linearly polarized radiation is possible.

2. SELF-INDUCED ROTATION OF THE PLANE OF POLARIZATION AND SYMMETRY PROPERTIES OF BIFURCATION CURVES

The bistability mechanism is determined by the form of the transmittances t_x, t_y. These coefficients as functions of the angle ψ between the field intensity vector $\mathbf{E}\equiv\mathbf{E}(0)$ and the x axis satisfy certain relationships which follow from the symmetry properties of a cubic crystal:

$$t_\varkappa(E,\psi)=t_\varkappa(E,\psi+n\pi)=t_\varkappa(E,-\psi)$$
$$(\varkappa=x, y), \quad t_x(E,\psi)=t_y(E,\pi/2-\psi) \tag{8}$$

[in Eq. (8), use was made of the fact that the x,y axes were chosen along symmetry directions in the (001) plane].

The simplest angular dependence of $t_{x,y}$, which satisfies the conditions (8) and takes into account and actually determines the self-induced dichroism, is shown in Fig. 1 (in particular, Fig. 1 describes the microscopic models analyzed in Sec. 4). Let us see how the polarization plane of the radiation

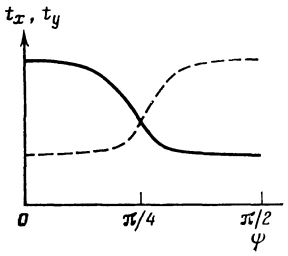

FIG. 1. Characteristic dependence of transmittances t_x, t_y on the slope angle of the radiation polarization vector on the front face of the crystal relative to the axis of symmetry (see text).

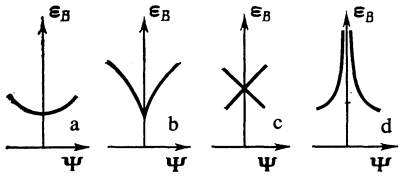

FIG. 2. Angular dependence of the bifurcation value of the amplitude of the field ε incident on the cavity at small angles Ψ between the polarization plane and the axis of symmetry of the crystal: (a) extremum corresponding to absorptive optical bistability; (b) extremum corresponding to dichroic bistability; (c) self-intersection; (d) tendency to infinity.

rotates in the crystal as a result of SID. We will initially assume that $t_x(E,\psi)$ in Fig. 1 is described by a solid curve, and $t_y(E,\psi)$, by a dashed curve, i.e., $t_x > t_y$ when $0 < \psi < \pi/4$. It is then obvious from Eqs. (2) and (6) that

$$E_x(d)/E_y(d) > E_x(0)/E_y(0), \quad \text{if} \quad E_x(0)/E_y(0) > 1,$$

i.e., $\psi(d) < \psi(0)$ if $0 < \psi \equiv \psi(0) < \pi/4$. Similarly, if $\pi/2 > \psi(0) > \pi/4$, then $\psi(d) > \psi(0)$. Hence the polarization vector in this case rotates toward the closer of the axes x, y, and the directions forming an angle $\pi/4$ with the axes are unstable for the polarization plane.

On the other hand, if the solid curve in Fig. 1 describes $t_y(E,\psi)$, and the dashed curve describes $t_x(E,\psi)$, then the unstable directions for polarization are the axes x, y, and the limiting stable directions are the axes rotated through $\pi/4$. Let us note that when the coordinate system is rotated through $\pi/4$, this case actually reduces to the preceding one.

Thus if a given symmetry axis (x axis) corresponds to the unstable direction, then $t_y > t_x$ when $|\Psi| \to 0$. In addition, t_y may exceed unity: a small field increment perpendicular to $\mathbf{E}(0)$ [and when $\psi = 0$ the vector $\mathbf{E} \equiv \mathbf{E}(0)$ is obviously directed along the x axis] may be amplified in the crystal. At the same time, in the absence of additional pumping $t_x(E,0) < 1$. If however the x axis corresponds to the limiting direction of the polarization plane, then $1 > t_x > t_y$ on the axis.

It is evident from Eqs. (7) and (8) that the function $D(E, \psi)$ satisfies the equations

$$D(E, \psi) = D(E, -\psi) = D(E, \psi + n\pi),$$
$$D(E, \psi) = D(E, \pi/2 - \psi).$$

Therefore the bifurcation curves $E_B(\psi), \varepsilon_B(\Psi)$ describing the solution of Eq. (7) are symmetric with respect to the values of ψ and Ψ which are multiples of $\pi/4$ (i.e., relative to the axes of symmetry):

$$E_B\left(\frac{n\pi}{4} + \psi\right) = E_B\left(\frac{n\pi}{4} - \psi\right),$$
$$\varepsilon_B\left(\frac{n\pi}{4} + \Psi\right) = \varepsilon_B\left(\frac{n\pi}{4} - \Psi\right), \quad n = 0, \pm 1, \ldots. \tag{9}$$

3. FORM OF BIFURCATION CURVES NEAR THE SYMMETRY-CAUSED EXTREMA AND MECHANISMS OF OPTICAL BISTABILITY

Since the field at the output of the crystal is continuously dependent on the field at the entrance, the transmittances t_x, t_y are continuous and differentiable with respect to E, ψ. Therefore, the bifurcation curves $E_B(\psi)$ are smooth. By vitue of the symmetry properties of Eqs. (9), the points $\psi = n\pi/4$ are shown on the $E_B(\psi)$ curves (symmetric directions). In the most general case, when $\partial D/\partial E \neq 0$ for $E = E_B$, $\psi = n\pi/4$, the function $E_B(\psi)$ at these points has an extremum and is described by a parabola in the vicinity of the latter.

On the bifurcation curve for the external field $\varepsilon_B(\Psi)$ the points $\Psi = n\pi/4$ are also shown, and if $E_B(\psi)$ has an extremum on a given axis of symmetry, the extremum $\varepsilon_B(\Psi)$ corresponds to this extremum. It is significant, however, that in contrast to $E_B(\psi)$, the function $\varepsilon_B(\Psi)$ near the extremum can be described not only by a parabola [Fig. 2(a)], but also by a nonanalytic curve, shown in Fig. 2(b).[1] The type of behavior of $\varepsilon_B(\Psi)$ near the extremum (for the sake of argument, near $\Psi = \psi = 0$) is determined by the mechanism of nonlinear absorption. Formally, it depends on which of the two factors in the first term in $D(E, \psi)$ in Eq. (7) becomes zero when $\psi = 0$ [when $\psi = 0$ the second term in $D(E,\psi)$ becomes identically equal to zero].

We will first analyze the case $1 - Rt_y(E_B, 0) \neq 0$. It is known to occur if the x axis is a limiting stable direction of polarization (then $t_y < y_x < 1$ and $Rt_y < 1$ when $|\psi| \ll 1$). The extremal values of $E_B(0)$ and $\varepsilon_B(0)$ are determined in this case according to (7) from the equations

$$1 - Rt_x^{(0)} - RE_0 \frac{\partial t_x^{(0)}}{\partial E_0} = 0, \quad E_0 \equiv E_B(0),$$
$$\varepsilon_0 \equiv \varepsilon_B(0) = E_0(1 - Rt_x^{(0)}); \tag{10a}$$
$$t_x^{(0)} \equiv t_x(E_0, 0).$$

Near $\Psi = 0$ the bifurcation curve $\varepsilon_B(\Psi)$ is described by a parabola:

$$\delta\varepsilon_B(\Psi) = {}^1/_2 A_1 \varepsilon_0 \Psi^2, \quad \delta\varepsilon_B(\Psi) \equiv \varepsilon_B(\Psi) - \varepsilon_B(0), \quad |\Psi| \ll 1;$$
$$A_1 = 1 - \left(\frac{1 - Rt_x^{(0)}}{1 - Rt_y^{(0)}}\right)^2 - R\frac{1 - Rt_x^{(0)}}{(1 - Rt_y^{(0)})^2}\left\langle\frac{\partial^2 t_x}{\partial \psi^2}\right\rangle_0, \tag{10b}$$
$$t_{x,y}^{(0)} \equiv t_{x,y}(E_0, 0),$$
$$(\partial^2 t_x/\partial\psi^2)_0 \equiv (\partial^2 t_x/\partial\psi^2)_{E=E_0, \psi=0}.$$

A different type of extremum arises if when $\psi = 0$

$$1 - Rt_y(E_0, 0) = 0, \quad E_0 = E_B(0). \tag{11a}$$

In this case, on the $\varepsilon_B(\Psi)$ curve the point $\Psi = 0$ is a turning point[10] (spinodal point), and the dependence of ε_B on Ψ is nonanalytic:

$$\delta\varepsilon_B(\Psi)=A_2\varepsilon_0\Psi^{2/3}, \quad |\Psi|\ll 1;$$

$$A_2=\frac{3}{2}a_1^{2/3}a_3^{-1}(a_1a_4-a_2a_3)^{1/3},$$

$$a_1=1-aE_0\frac{\partial t_x^{(0)}}{\partial E_0}, \quad a_2=1+a\left(\frac{\partial^2 t_x}{\partial\psi^2}\right)_0, \tag{11b}$$

$$a_3=-aE_0\frac{\partial t_y^{(0)}}{\partial E_0}, \quad a_4=a\left(\frac{\partial^2 t_y}{\partial\psi^2}\right)_0, \quad a=R(1-Rt_x^{(0)})^{-1}.$$

The form of curves (10b) and (11b) is shown in Fig. 2 (a) and (b). For specificity, the parameters A_1 and A_2 were chosen positive.

The two indicated types of extrema of $\varepsilon_B(\Psi)$ correspond to two types of optical bistability in the vicinity of symmetric directions. The condition (10a) determines the threshold of so-called[11] "absorptive" bistability. The latter arises if the differential transmission of the crystal $\partial t_x/\partial E$ is positive and sufficiently high, so that the increase of the field at the entrance to the crystal $E(0)$ leads to an increase in transmission providing for an even greater increase of $E(0)$ in the presence of feedback (longitudinal instability for the symmetric direction of polarization). In the vicinity of the extremum point $(\varepsilon_0, 0)$ when

$$b(\delta\varepsilon-\delta\varepsilon_B(\Psi))<0, \quad \delta\varepsilon\equiv\varepsilon-\varepsilon_0, \quad b=2\frac{\partial t_x^{(0)}}{\partial E_0}+E_0\frac{\partial^2 t_x^{(0)}}{\partial E_0^2} \tag{10c}$$

there are two stationary states of the radiation in the cavity. On the bifurcation curve $\delta\varepsilon=\delta\varepsilon_B(\Psi)$ they merge, and when the sign of $b[\delta\varepsilon-\delta\varepsilon_B(\Psi)]$ changes, both states disappear.

Absorptive bistability is known for isotropic media. It can be observed in crystals thanks to the self-induced anisotropy of absorption, not only by changing the intensity of the external field ε, but also by rotating its polarization plane (by changing Ψ while ε is fixed). Depending on the sign of A_1b, the angular dependence of the field change at the cavity output $\delta\varepsilon_{\text{out}}(\Psi)$ in the vicinty of $|\Psi|\ll 1$, $|\delta\varepsilon|\ll\varepsilon_0$ is described either by an ellipse (when $A_1b<0$) or by two hyperbolas (when $A_1b>0$).

The condition (11a) defines a new type of threshold—of dichroic bistabilty. For DOB, it is necessary that the condition $t_y(E,0)>1$ be satisfied, i.e., that for specified symmetric directions (x axis) of the polarization plane, the transverse fluctuations of field intensity E be amplified by the crystal and by the cavity as a whole (transverse instability). Obviously, DOB can only arise if the symmetry axis considered is an unstable direction of polarization [the instability condition $t_y(E,0)>t_x(E,0)$ is known to be satisfied when $t_y(E,0)>1$].

The dependence of the field at the cavity output on angle Ψ for $|\delta\varepsilon|\ll\varepsilon_0$, $|\Psi|\ll 1$ in the case of DOB is shown in Fig. 3. At the turning point ($\varepsilon=\varepsilon_0$, $\Psi=0$), three stationary states of the system merge.[10] For a small deviation from this point, the amplitude and polarization of the emerging radiation can have either three or one value when $A_2[\delta\varepsilon_B(\Psi)-\delta\varepsilon]$ is negative and positive, respectively. Let us note that an S shape arises in DOB on the angle vs angle curve, whereas in the case of absorptive bistability, the dependence of the field at the output on the field at the input is S-shaped.[11]

Since it is precisely near the extremum of $\varepsilon_B(\Psi)$ that the cause of the instability is manifested in pure form, their analysis makes it possible to classify the bistability mechanisms. It follows from the results cited that in crystals with nonlinear absorption, there are two main types of bistability—absorptive and dichroic.

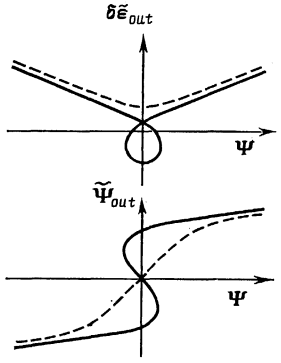

FIG. 3. Angular dependence of amplitude increase $\delta\varepsilon_{\text{out}}$ of the field at the cavity output and of the angle Ψ_{out} between its polarization plane and the symmetry axis of the crystal in the vicinity of the point of closest approach determined by symmetry: $\delta\tilde{\varepsilon}_{\text{out}}=a_1a_2^{-1}\delta\varepsilon_{\text{out}}$, $\tilde{\Psi}_{\text{out}}=-a_1a_3A_2\Psi_{\text{out}}$. Solid curves: $A_2[\varepsilon-\varepsilon_B(0)]>0$, dashed curves $-A_2[\varepsilon-\varepsilon_B(0)]<0$ (the signs of $\delta\tilde{\varepsilon}_{\text{out}}$ at the minima of the solid and dashed curves are opposite).

4. BIFURCATION CURVES FOR SPECIFIC MECHANISMS OF NONLINEAR ABSORPTION. DICHROIC BISTABILITY IN THE CASE OF CUBIC NONLINEARITY

In order to establish on the basis of the results of the preceding sections whether a specific mechanism of resonant nonlinear absorption can lead to bistability and what type of bistability takes place, it is necessary to calculate explicitly the parameters of light transmission by the crystal, t_x and t_y. The calculation of $t_{x,y}$ amounts to solving Maxwell's equations. For slowly changing field envelopes $E_{x,y}(z)$, these equations in the steady state are

$$\partial E_\varkappa(z)/\partial z=i\xi P_\varkappa(z), \quad \xi=2\pi\omega/cn, \quad \varkappa=x, y, \tag{12}$$

where

$$P_\varkappa(z)=i\frac{\alpha}{\xi}E_\varkappa(z)+P_\varkappa^{(r)}(z). \tag{13}$$

The term $P_\varkappa^{(r)}(z)$ has been isolated in Eq. (13), for the polarization of a crystal. It describes the contribution of resonant processes causing nonlinear absorption of light. The parameters α and n in Eqs. (12) and (13) respectively describe the linear absorption and refraction of light related to other types of processes.

Let us first consider one of the simplest mechanisms of optical nonlinearity—absorption saturation; it will be assumed that the resonant absorption of the x component of the field is independent of the y component (and vice versa), and can be described in terms of the two-level model. Resonance polarization is then given by[12]

$$P_\varkappa^{(r)}=iCE_\varkappa[1+g|E_\varkappa|^2]^{-1}, \quad \varkappa=x, y. \tag{14}$$

An example of the physical system described by Eq. (14) is provided by two-level impurity centers having several equivalent positions (orientations) in the unit cell of a cubic crystal. If the impurities (for example, linear molecules) are ori-

ented in directions of type ⟨100⟩ and the dipole moment of the impurity transition resonantly excited by the radiation is parallel to the axis of symmetry, the x, y axes in Eq. (14) are obviously directed along the [100], [010] axes; if however the impurities are oriented in the ⟨111⟩ directions, the x, y axes in Eq. (14) are directed along [110], [1$\bar{1}$0] (for more detail, see Ref. 2). For the impurity mechanism of nonlinearity, the parameter C in Eq. (14) is proportional to the concentration of impurities, and g is determined by the times of longitudinal and transverse relaxations and can be very large. At exact resonance $\mathrm{Im}C = \mathrm{Im}g = 0$.

From Eqs. (12)–(14), taking Eqs. (2) and (6) into account, we find the transcendental equation for the transmission parameters t_x:

$$t_x = \tilde{t}(E_x(0)),$$
$$\tilde{t}^2(E) = [(1+\eta^{-1}\tilde{t}^2(E)gE^2)/(1+\eta^{-1}gE^2)]^{1-\eta}\cdot\exp(-2\alpha\eta d), \qquad \eta = (\alpha+\xi C)/\alpha. \tag{15}$$

It is evident from Eq. (15) that optical nonlinearity (14) leads to self-induced dichroism ($t_x \neq t_y$). The dependence of t_x, t_y on the inclination of the polarization plane $\psi = \psi(0)$, according to Eq. (15), is qualitatively described by Fig. 1 (t_x being the solid curve, and t_y, the dashed curve). The directions $\psi = n\pi/2$ (axes x, y) are the limiting stable directions of the polarization plane, and $\psi = (2n+1)\pi/4$ are the unstable directions.[2]

It can readily be ascertained that there are four solutions for the equation of the bifurcation curve (7) with Eq. (15) taken into account, as follows:

$$E_B^{(1,2)}(\psi) = E_a^{(1,2)}|\cos\psi|^{-1}, \quad E_B^{(3,4)}(\psi) = E_a^{(1,2)}|\sin\psi|^{-1}, \tag{16}$$

where $E_a^{(1,2)}$ (with $E_a^{(2)} > E_a^{(1)}$) are the solutions of the equation

$$1 - R\tilde{t}(E_a) - RE_a\frac{\partial\tilde{t}(E_a)}{\partial E_a} = 0. \tag{17}$$

Expressions (16) and (17) have a simple physical meaning; indeed, in the model (14), different components of the field do not affect each other. Therefore, bistability arises independently for each of them and has an absorptive nature [an analysis using the model (14) in the presence of a single field component is given, for example, in Ref. 11].

Consideration of self-induced dichroism substantially enriches the picture of optical bistability. When $\varepsilon_B^{(2,1)}(\Psi) < \varepsilon < \varepsilon_B(\Psi)$, the radiation in the cavity has six stationary states. The extrema and singularities of the bifurcation curves are described by curves 2, (a), (c), (d). At the point $\Psi = 0$, the curves $\varepsilon_B^{(1,2)}(\Psi)$ have a minimum and are described near it by the parabola $\delta\varepsilon_B^{(1,2)}(\Psi) = \frac{1}{2}\varepsilon_B^{(1,2)}(0)\Psi^2$ (which corresponds to absorptive bistability; see Sec. 3). When $\Psi \to \pm\pi/2$, the functions $\varepsilon_B^{(1,2)}(\Psi) \to \infty$. The curves $\varepsilon_B^{(3,4)}(\Psi)$ are obtained from the curves $\varepsilon_B^{(1,2)}(\Psi)$ by reflection relative to the axis $\psi = \pi/4$, and therefore the curves $\varepsilon_B^{(i)}(\Psi)$ and $\varepsilon_B^{(i+2)}(\Psi)$ $(i = 1,2)$ intersect at the points $\Psi = (2n+1)\pi/4$. Let us note that in the region of bistability, the crystal amplifies the counterradiation with the same wavelength, and this may complicate the picture.

Another well-known mechanism of resonant nonlinear absorption is two-photon absorption (see for example Ref. 13). It is described by terms cubic in the field $E(z)$ in the resonance polarization of the crystal $\mathbf{P}^{(r)}$

$$P_x^{(r)} = i(\gamma_1 E_x|\mathbf{E}|^2 + \gamma_2 E_x^*\mathbf{E}^2 + \gamma_3 E_x|E_x|^2), \quad E_x \equiv E_x(z). \tag{18}$$

In exact resonance, the parameters $\gamma_{1,2,3}$ are real. From the condition of absence of light amplification by the crystal $(\partial|E|^2/\partial z < 0)$, allowing for (12), (13) and (18) it follows that

$$\gamma_1 + {}^1/_2\gamma_3 \geqslant 0, \quad \gamma_1 + \gamma_2 + {}^1/_2\gamma_3 \geqslant 0, \quad \gamma_1 + \gamma_2 + \gamma_3 \geqslant 0. \tag{19}$$

We will hereinafter assume that $\gamma_2 \leqslant 0$ (the phase difference of the field components E_x, E_y does not increase with the crystal thickness[9]).

Since according to (12), (13), (18) and (19) the radiation absorption increases with the intensity, it is obvious that absorptive bistability in the model (18) is impossible. At the same time, dichroic bistability may arise under certain conditions. Self-induced dichroism is caused by the last term in Eq. (18) (for isotropic media $\gamma_3 = 0$). When $\gamma_3 < 0$, the radiation polarization plane is rotated in the crystal toward the nearer of the axes x, y, and when $\gamma_3 > 0$, to the closer of the axes rotated through $\pi/4$ relative to axes x,y.[9] Generally, as is evident from Eqs. (12), (13), and (18), the case $\gamma_3 < 0$ is reduced to the case $\gamma_3 > 0$ by rotating the coordinates through $\pi/4$ and simultaneously replacing $\gamma_{1,2,3}$ by $\tilde{\gamma}_{1,2,3}$, where

$$\tilde{\gamma}_1 = \gamma_1 + \gamma_3, \quad \tilde{\gamma}_2 = \gamma_2 + {}^1/_2\gamma_3, \quad \tilde{\gamma}_3 = -\gamma_3. \tag{20}$$

The angular dependence of transmittances $t_{x,y}$ is described by Fig. 1 (when $\gamma_3 > 0$, the solid curve is t_y and the dashed curve is t_x, and when $\gamma_3 < 0$, vice versa).

It follows from the results of Sec. 3 that the criterion of appearance of DOB is determined by the transmission of the crystal near the unstable direction of the polarization plane [see (11a)], i.e., near $\psi = n\pi/2$ in the case $\gamma_3 > 0$ and near $\psi = \pi(2n+1)/4$ for $\gamma_3 < 0$. The transmittances $t_{x,y}(E,\psi)$ near the symmetric directions can be calculated explicitly on the basis of Eqs. (12), (13) and (18). As a result, using Eq. (11a) in the case $\gamma_3 > 0$, we obtain for the threshold field $\varepsilon_0 = \varepsilon_B(0)$ the expression

$$\varepsilon_B(0) = \varepsilon_c[1-(Rt_0)^{1-\gamma}][(Rt_0)^{2\gamma}-1]^{1/2} \quad (\gamma_3 > 0),$$
$$\varepsilon_c = \left(\frac{\alpha}{\xi}\right)^{1/2}(1-t_0^2)^{-1/2}(\gamma_1+\gamma_2+\gamma_3)^{-1/2}, \quad \gamma = \frac{\gamma_1+\gamma_2+\gamma_3}{\gamma_1+\gamma_2}, \quad t_0 = \exp(-\alpha d). \tag{21a}$$

Since the low-field transmittance t_0 as well as the feedback factor R are smaller than unity, it follows from (21a), allowing for (19), that DOB is possible [$\varepsilon_B(0)$ being a real number] only when $\gamma < 0$, i.e.,

$$\gamma_1 + \gamma_2 < 0 \quad (\gamma_3 > 0). \tag{22a}$$

When $\gamma_3 < 0$, the expression for the threshold field $\varepsilon_B(\pi/4)$ is also given by Eq. (21a) if in the latter one replaces ε_c and γ by $\tilde{\varepsilon}_c$ and $\tilde{\gamma}$, where in accordance with Eq. (20)

$$\tilde{\varepsilon}_c = (\alpha/\xi)^{1/2}(1-t_0^2)^{-1/2}(\gamma_1+\gamma_2+{}^1/_2\gamma_3)^{-1/2},$$
$$\tilde{\gamma} = \frac{\gamma_1+\gamma_2+{}^1/_2\gamma_3}{\gamma_1+\gamma_2+{}^3/_2\gamma_3} \quad (\gamma_3 < 0). \tag{21b}$$

When $\gamma_3 < 0$, the existence criterion for DOB becomes

$$\gamma_1+\gamma_2+{}^3/_2\gamma_3<0 \quad (\gamma_3<0). \tag{22b}$$

It follows from (21a) and (21b) that the threshold field ε_0 decreases with increasing feedback factor R, and changes nonmonotonically (has a minimum) with crystal thickness $d=\alpha^{-1}|\ln t_0|$. This is easy to understand, since a thin crystal causes a comparatively slight rotation of the plane of radiation polarization, and in a very thick crystal almost all of the radiation is absorbed, and the effectiveness of the feedback is slight.

Under DOB conditions in the optical nonlinearity model (18), the bifurcation curve $\varepsilon_B(\Psi)$ near the unstable directions of the polarization plane and the angular dependence of the field at the cavity output have the shape shown in Figs. 2(b) and 3. A complete analysis of the bifurcation curves for arbitrary $\gamma_{1,2,3}$ can be performed numerically.

The relationship between the parameters γ_1, γ_2, γ_3 in two-photon resonance absorption is determined by the manner of interpreting the transformation of the states between which the transition takes place (see for example Ref. 13). As an example, we shall consider the case in which

$$\gamma_2=0, \quad \gamma_3=-\gamma_1, \quad \gamma_1>0. \tag{23}$$

Expression (23) describes a whole series of transitions in cubic crystals [in particular, Eqs. (18) and (23) can readily be obtained in a standard manner[14] for a weakly damped oscillator with frequency $\omega_0=2\omega$, which is transformed according to a vectorial representation of group T_d, including cubic anharmonicity of type $Vxyz$]. Obviously, relation (22b) follows from (23), i.e., DOB is possible in the vicinity of ⟨110⟩ type directions in this case.

It is a simple matter in the case of (23) to solve Eqs. (12), (13), and (18) analytically and obtain the parameters $t_x(E,\psi)$, $t_y(E,\psi)$ in explicit form (they are expressed in terms of elementary functions, with the field E entering into the combinations $E/\tilde{\varepsilon}_c$ then to study the bifurcation curve $\varepsilon_B(\Psi)$. The form of $\varepsilon_B(\Psi)/\tilde{\varepsilon}_c$ is determined by the unique parameter Rt_0 and is shown for different Rt_0 in Fig. 4. As is evident from the latter, DOB exists in a finite range of angles $\Delta\Psi$:

$$\varepsilon_B(\Psi)\to\infty \quad \text{for} \quad \Psi\to\pi/4\pm\Delta\Psi,$$
$$\Delta\Psi=\arctan\left[R^{-2}t_0^{-2}\left(1-(1-R^2t_0^2)^{1/2}\right)^2\right]. \tag{24}$$

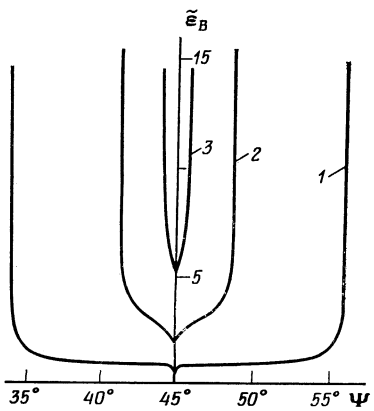

FIG. 4. Angular dependence of reduced bifurcation field $\tilde{\varepsilon}_B=\varepsilon_B(\Psi)/\tilde{\varepsilon}$ for two-photon absorption in the model (18), (23). Curves 1–3 correspond to $Rt_0=0.75, 0.5, 0.25$.

According to Eq. (24), the DOB region expands quickly with increasing Rt_0 (cf. Fig. 4).

5. STABILITY OF STATIONARY STATES

The dynamics of a cavity containing a nonlinear crystal is characterized by two times: the time τ_R taken by the radiation to complete a circuit of the cavity, and the polarization relaxation time τ_r of the medium (with $\tau_r, \tau_R \gg \omega^{-1}$). The value of τ_r ranges from 10^{-7} to 10^{-12} s (for optically oriented tunnel centers, τ_r can be as long as ~1 s or longer), and $\tau_R \sim 10^{-8}$–10^{-10} s. One can therefore distinguish two limiting mechanisms of instability of stationary states in a cavity which correspond to $\tau_r \ll \tau_R$ and $\tau_r \gg \tau_R$.[11]

The instability condition in the case where the response of the crystal is instantaneous, $\tau_r \ll \tau_R$, can easily be obtained if the delay (finiteness of the velocity of light) is considered in Eqs. (1) and (2). The fluctuational contribution to the field at the crystal output $\delta\mathbf{E}(d,t)$ obviously consists of the sum of the fluctuation field $\delta\mathbf{E}(d,t-\tau_R)$, reflected by the mirrors and transformed by the crystal, and fluctuation field $f(t,\tau_R)$, present at other points of the cavity (in the range from $t-\tau_R$ to t) and arriving at point d at time t:

$$\delta E_\varkappa(d,t)=\sum_{\varkappa'}\frac{\partial E_\varkappa(d)}{\partial E_{\varkappa'}(0)}R\exp(i\varphi_R)\,\delta E_{\varkappa'}(d,t-\tau_R)+f_\varkappa(t,\tau_R) \tag{25}$$

[$\mathbf{E}(0)$ and $\mathbf{E}(d)$ being the stationary values of the field]. It is clear from Eqs. (25) and (2) that a stationary state with given $E(0)$ is stable if the principal values $\gamma_{1,2}$ of tensor $\hat{\Lambda}$

$$\Lambda_{\varkappa\varkappa'}=R\exp(i\varphi_R)\,\partial(E_\varkappa(0)\,T_\varkappa)/\partial E_{\varkappa'}(0), \tag{26}$$

satisfy the inequality

$$|\lambda_{1,2}|<1. \tag{27}$$

Let us note that the values of $E(0)$ for which $\lambda=1$ are bifurcation values: as is evident from Eq. (4), $D=0$ for such $E(0)$.

In the case of linearly polarized radiation and purely dissipative nonlinearity (5), the potential $\hat{\Lambda}$ is real. Then the inequality (27) will have to break down, and the state of the radiation in the cavity will turn out to be unstable if

$$D(E,\psi)=1-\tilde{\Lambda}_{xx}-\tilde{\Lambda}_{yy}+\tilde{\Lambda}_{xx}\tilde{\Lambda}_{yy}-\tilde{\Lambda}_{xy}\tilde{\Lambda}_{yx}<0,$$
$$\tilde{\Lambda}_{\varkappa\varkappa'}=R\partial(E_\varkappa t_\varkappa)/\partial E_{\varkappa'}, \quad E_\varkappa\equiv E_\varkappa(0) \tag{28}$$

[$D,(E,\psi)$ was defined in Eq. (7)].

If the external field ε passes through the bifurcation value ε_B [intersects the curve $\varepsilon_B(\Psi)$], so that two new stationary states appear, then as is evident from Eq. (7), the function $D(E,\psi)$ for them has a different sign. It therefore follows from Eq. (28) that one of them in the case of a "lag-free" crystal is unstable.

The analysis of the case of a slowly relaxing crystal, $\tau_r \gg \tau_R$, is substantially more complex. It essentially amounts to the analysis of the stability of the field distribution in the crystal; this field is inhomogeneous, and as the thickness changes, there is a change not only in the amplitude but also in the direction of the plane of polarization. Polarization fluctuations P of the crystal are the source of fluctuations of the field, and it is the polarization relaxation

that determines the kinetics of the system.

A very general approach to the analysis is of stability that makes it possible to examine a large number of systems will be illustrated with a simple but important model in which polarization fluctuations are described by the local equation

$$(\partial/\partial t)\delta\mathbf{P}(z,\ t)=-\hat{M}\delta\mathbf{P}(z,\ t)+\hat{N}\delta\mathbf{E}(z,\ t), \tag{29}$$

where the tensors $\hat{M}$ and $\hat{N}$ are dependent on the stationary value of the field $\mathbf{E}(z)$. The real parts of the principal values of $\hat{M}$ are assumed to be positive for all z, so that locally the fluctuations do not increase, and instability can only be due to feedback. Slow fluctuations of the field and polarization are related by the Maxwell equation [cf. Eq. (12)]

$$(\partial/\partial z)\delta\mathbf{E}(z,\ t)=i\xi\delta\mathbf{P}(z,\ t) \tag{30}$$

and satisfy the boundary condition [cf. Eqs. (1), (6), (12)]

$$\begin{aligned}&\delta\mathbf{E}(0,\ t)=\bar{R}\delta\mathbf{E}(d,\ t),\\&\bar{R}=R\exp[i\varphi_R+i\tilde{\varphi}_d],\quad \tilde{\varphi}_d=n\omega d/c.\end{aligned} \tag{31}$$

In Eqs. (30) and (31), we neglected the delay in the cavity and crystal, this being valid for $\tau_r\gg\tau_R$, $(\alpha c)^{-1}$.

Substituting (30) into (29) and performing a Laplace transformation, we obtain for the quantities

$$\delta E_\varkappa(z,s)=\int_0^\infty dt e^{-st}\delta E_\varkappa(z,t) \tag{32}$$

an inhomogeneous system of two first-order equations. Its solution may be represented in the form

$$\delta\mathbf{E}(z,s)=\mathbf{w}(z)+\sum_{i=1,2}c_i\mathbf{v}_i(z,s), \tag{33}$$

where $\mathbf{w}$ is the inhomogeneous part, and the $\mathbf{v}_i$ are the solutions of the homogeneous system

$$(s\hat{I}+\hat{M})\frac{\partial\mathbf{v}}{\partial z}-i\xi\hat{N}\mathbf{v}=0 \tag{34}$$

($\hat{I}$ being a unit tensor). It is convenient to choose these solutions so that

$$v_{i\varkappa}(0,\ s)=\delta_{i\varkappa}\quad (i=1,\ 2;\ \ \varkappa=x,\ y\equiv 1,\ 2). \tag{35}$$

The coefficients c_i in (33) are detemined from the boundary condition (31). The system of equations for c_i has a unique solution if

$$\mu(s)=|\delta_{i\varkappa}-\bar{R}v_{i\varkappa}(d,\ s)|\neq 0. \tag{36}$$

If (36) is satisfied for all $\mathrm{Re}\,s\geqslant 0$, then, as is evident from Eq. (32), the field fluctuations attenuate with time, i.e., the state of the system is stable. It is easy from Eq. (36) to obtain the sufficient condition of instability in the purely dissipative resonance case ($R=\bar{R}$, $\mathrm{Im}\hat{M}=\mathrm{Im}\hat{N}=0$), when $\mu(s)$ is real with $\mathrm{Im}\ s=0$. According to (34) and (36), in this case $v(z,s)=$ constant when $s\to\infty$ and $\mu(s)\to(1-R)^2>0$. At the same time, as is evident from Eqs. (34), (29) and (30), when $s=0$ the relationship of $\mathbf{v}(d,s)$ to $\mathbf{v}(0,s)$ is determined by the response of the crystal to the increase in field $\mathbf{E}\equiv E(0)$:

$$v_{i\varkappa}(d,0)=\sum_{\varkappa'}v_{i\varkappa'}(0,0)\,\partial(E_\varkappa t_\varkappa)/\partial E_{\varkappa'}. \tag{37}$$

It follows from Eqs. (36) and (37) that $\mu(0)=D(E,\psi)$ when $D(E,\psi)$ is defined in Eq. (7). If $D(E,\psi)<0$, the function $\mu(s)$ for a certain s becomes zero, i.e., $D(E,\psi)<0$ is a sufficient condition of instability.

Thus as when $\tau_R\gg\tau_r$ in the case of $\tau_r\gg\tau_R$ one of the two states with linear polarization which are formed at the bifurcation point is unstable. It can be shown that this is also the case for models more general than Eqs. (29) and (30) [for example, when the terms $\sim\partial\delta E(z,t)/\partial t$ are considered in Eqs. (29) and (30), which is essential, for example, for systems with long longitudinal relaxation times]. Since the function $D(E,\psi)$ changes sign only at the bifurcation points, a solution that is unstable near a bifurcation point is unstable in the entire region of its existence [as well as for systems described by ordinary differential equations and undergoing bifurcation of codimension 1 (Ref. 10)]. Let us note, however, that the condition $D(E,\psi)>0$, which is satisfied for the second of the solutions generated at a bifurcation point, does not guarantee its stability. The solution of this problem requires a complete calculation of the determinant $\mu(s)$.

Such a calculation is trivial in the absence of anisotropy in the "one-dimensional" case $\delta\mathbf{E}(z,t)\|\mathbf{E}(z)\|\mathbf{E}(0)$, $\mathrm{Im}E_xE_y^*=0$:

$$\mu(s)=1-\bar{R}\exp\left[i\xi\int_0^d dz\,N/(M+s)\right]. \tag{38}$$

If the parameters N, M, $\bar{R}$, $i\xi$ are real and N is a function of constant sign, it follows from Eqs. (36) and (38) that in the presence of absorption bistability, one of the solutions generated at the bifurcation point is unstable, and the other is stable relative to slow fluctuations of the polarization of the medium.

6. CONCLUSION

It follows from the results of this work that the self-induced anisotropy of nonlinear absorption of light by cubic (and tetragonal) crystals can lead to a new type of bistability, i.e., dichroic optical bistability. The latter arises when for specific symmetric orientations of the polarization plane, the transverse fluctuations of a strong field are amplified in the crystal. Such amplification is characteristic of a series of resonance mechanisms of nonlinearity (see Sec. 4, as well as Ref. 2). Self-induced resonance dichroism is particularly pronounced for crystals with reorienting centers having several equivalent positions in the unit cell. Experimentally self-induced rotation of the polarization plane was observed in Ref. 3 in KCl crystals containing F_A(Li) centers, even in nonlaser fields. It can be shown that in this case DOB has practically no field intensity threshold at low temperatures.

As a result of self-induced anisotropy in the presence of optical bistability, multivaluedness is manifested in both the intensity and polarization of the radiation at the cavity output. It can be observed by changing both the intensity of the incident radiation and the orientation of the polarization plane relative to the crystal axes.

As a rule, because of self-induced anisotropy, it is not possible in complex real systems to describe analytically the propagation of radiation with arbitrary polarization. It is essential therefore that in order to establish the type of bista-

bility and threshold value of the radiation intensity, if suffices in the majority of cases to limit oneself to symmetric directions and to study the form of the bifurcation curves near the symmetry-determined extrema (cf. the examples in Sec. 4). Such analysis also makes it possible to establish the instability of certain stationary states.

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[1)] Generally speaking, the branches of bifurcation curves can intersect (or have singular points of higher order), and for certain ψ the function $E_B(\psi)$ [and correspondingly $\varepsilon_B(\Psi)$] may tend to infinity. Both types of singularities can be attributed to symmetry properties. They are then located on the axes of symmetry and are not shifted along ψ (or along Ψ) when there is a finite change in the parameters of the system [see Fig. 2 (c), (d) and the example in the next section]. Let us note that the self-intersection of $\varepsilon_B(\Psi)$ on the axis [Fig. 2(c)] is not necessarily related to the self-intersection of $E_B(\psi)$. This occurs, for example, if one of the functions $1 - Rt_\varkappa(E_B\psi)$ ($\varkappa = x,y$) becomes zero when $\psi \neq n\pi/4$.

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