Threshold-free optical bistability due to giant nonlinearity of resonance absorption in crystals containing F_A centers

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An analysis is made of a polarization optical bistability in crystals containing F_A centers and placed in a resonator. The bistability is due to the possibility of amplification of fluctuations of the polarization of radiation in such crystals and in practice has no threshold in respect of the radiation intensity. An analysis is made of the ranges of values of the parameters of a doped crystal and of a resonator in which this bistability can be observed. The analysis is carried out for ring and Fabry–Perot resonators.

INTRODUCTION

Several optical nonlinearity mechanisms lead to a polarization instability of linearly polarized radiation, i.e., they are responsible for the growth of fluctuations of the polarization of such radiation as it propagates in a nonlinear medium. If this medium is inside a resonator, then the growth of a polarization instability in a system may give rise to a polarization optical bistability (POB). The specific nature of the POB is manifested most clearly when the incident radiation is polarized in a symmetry plane of a crystal. The transmitted radiation is then polarized elliptically and the polarization ellipse has one of two orientations symmetric relative to the initial reflection of the vector **E**. An optical bistability considered in Refs. 1–3 belongs to the POB class.

An example of a medium in which the polarization instability appears at extremely low radiation intensities is an alkali halide crystal containing F_A centers. Each of these centers is a complex consisting of an F center and a positively charged substitutional impurity present in the same unit cell. These centers have six equivalent orientations (along axes of the ⟨100⟩ type—see Ref. 4). The optical spectrum of an F_A center usually has two bands F_{A1} and F_{A2} corresponding to the absorption of light polarized along and across the axis of the center.

The mechanism of the optical nonlinearity of the F_A centers is associated with the anisotropy of the resonance absorption by single centers and with the ease of transfer of the excited F_A centers from one equivalent orientation to another. In view of these properties of the centers, polarized resonance radiation orients the centers preferentially along a specific axis so that a crystal becomes optically anisotropic. The intensities necessary for the effective optical orientation of the centers are governed by the ratio of the reorientation probabilities in the ground and excited states, and at low temperatures these intensities are low. In the case of some systems, such as the F_A(Li) centers in KCl, the probability of reorientation in the ground state is negligible and the resonance radiation intensity determines simply the kinetics of the optical orientation process, but not the steady-state anisotropic distribution of the centers. Systems of this kind will be considered below.

An analysis of the nonlinear response of a doped crystal is based on the solution of the rate equations for the populations of the various orientations of the F_A centers. In the specific case when the radiation propagates in a crystal along the [001] axis the component of the polarization **P** determined by these centers can be represented in the form[5]:

$$P_\varkappa = \frac{nc}{2\pi i\omega} p_\varkappa, \quad p_\varkappa = i\zeta_2 E_\varkappa + i\frac{1}{2}\mu(\zeta_1-\zeta_2)\{2E_\varkappa(\mu|\mathbf{E}|^2 + (1-\mu)|E_\varkappa|^2) - (1-\mu)E_\varkappa^*\mathbf{E}^2\}|\mathbf{E}|^2\Big[\mu(2+\mu)|\mathbf{E}|^4 + \frac{1}{4}(1-\mu)^2|E_x^2-E_y^2|^2\Big]^{-1}, \quad \varkappa = x, y \tag{1}$$

(the X and Y axes have the ⟨110⟩ orientations). The parameters ζ_1 and ζ_2 in Eq. (1) are proportional to, respectively, the polarizabilities of the F_A centers along and across their axes and, moreover, they are proportional to the concentration of the centers; these parameters depend resonantly on the frequency of light ω. The ratio of the absorption cross sections of light polarized along and across the axis of the centers is

$$\mu = \zeta_2''/\zeta_1'' \quad (\zeta_{1,2} \equiv \zeta_{1,2}' + i\zeta_{1,2}''). \tag{2}$$

If $\mu = 1$, the anisotropy of the absorption by a single center and, consequently, the optical orientation disappear and we have $p_x = (1/3)i(\zeta_1 + 2\zeta_2)E_x$.

Expressions such as Eq. (1) were used in Ref. 5 to describe a strong self-induced change in the polarization of the radiation observed in KCl crystals containing F_A(Li) centers. It follows from Eq. (1) that under certain conditions (see below) the fluctuations of the polarization of the radiation in a crystal are enhanced close to the [110] direction.

1. CRITERION OF APPEARANCE OF THE POLARIZATION OPTICAL BISTABILITY

A complete analysis of the optical bistability in a resonator containing a crystal with F_A centers requires solution of equations for the field allowing for Eq. (1) and for the corresponding boundary conditions. However, we can find the values of the parameters corresponding to the onset of the optical bistability and obtain quantitative information on its manifestations by solving a much simpler problem. This follows from the results of a general analysis of the POB given in Ref. 6. It should be noted that since in the systems under discussion the radiation intensity does not affect the steady-state (strongly nonlinear) properties of the systems, as demonstrated by Eq. (1), it follows that the usual "intensity" optical bistability (absorptive or refractive) is then im-

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possible. According to Ref. 6, the threshold parameters for the onset of the POB can be found from the condition

$$\partial(\psi_t,\ \chi_t)/\partial(\psi_t,\ \chi_t)=0, \quad \psi_t\to 0,\ \chi_t\to 0. \tag{3}$$

Here, ψ_i (ψ_t) is the angle between the major axis of the polarization ellipse of the radiation incident on (transmitted by) the system and the polarization-unstable symmetric direction in a nonlinear nongyrotropic medium; the parameter χ determines the degree of polarization of the radiation [$\cos 2\chi = (I_\parallel - I_\perp)(I_\parallel + I_\perp)^{-1}$] and the direction of rotation of the vector $\mathbf{E}(t)$ (see Ref. 7). It follows from the condition (3) that the relationship between the increments in the polarization parameters of the incident and transmitted radiation at the POB threshold is strongly nonlinear [according to the linear theory, fluctuations of the polarization of the radiation in a system grow without limit, $|\partial(\psi_t,\chi_t)/\partial(\psi_i,\chi_i)|\to\infty$].

We can calculate the Jacobian (3) by solving the field equations assuming obviously that the radiation is polarized almost along the polarization-unstable symmetric direction (we then have $|\psi_{i,t}|,\ |\chi_{i,t}|\ll 1$). We shall correspondingly assume $|E_x|\gg|E_y|$ in Eq. (1) and linearize the expression for the polarization along E_y:

$$p_x \approx {}^1/_2 i\alpha E_x, \qquad p_y \approx {}^1/_2 i(\alpha-\beta)E_y - {}^1/_2 i\beta E_y^* E_x (E_x^*)^{-1}, \tag{4}$$

where

$$\alpha \equiv \alpha' + i\alpha'' = 2\zeta_2 + 4\mu(\zeta_1-\zeta_2)/(1+5\mu);$$
$$\beta \equiv \beta' + i\beta'' = 4(\zeta_1-\zeta_2)\mu(1-\mu)/(1+\mu)(1+5\mu).$$

We can see from Eq. (4) that the nonlinear polarization of the F_A centers has a certain important feature: if the components of the field E_y and E_x are phase-shifted by $\pi/2$ (which corresponds to $\psi=0$, and $\chi = -\mathrm{Im}E_y/E_x$), then $p_y/E_y = p_x/E_x$, i.e., the investigated crystal is "isotropic" in the XY plane. This property is associated physically with the fact that the anisotropy of the nonlinear response is due to the anisotropy of the absorption by the F_A centers and appears because of a difference between the intensities of the field components which are longitudinal and transverse to the ⟨100⟩ axes of the centers; if $\arg E_y E_x^{-1} = \pm\pi/2$ these intensities are equal in the XY plane: $({}^1/_2)|E_x+E_y|^2 = ({}^1/_2)|E_x-E_y|^2$. Consequently, if $\arg E_y E_x^{-1} = \pm\pi/2$ then the polarization in a crystal does not change, so that we have $\partial\psi_i/\partial\chi_t = 0$, and the condition (3) reduces to the simpler form:

$$\partial\psi_t/\partial\psi_t = 0,\ \psi_t\to 0,\ \chi_t = 0. \tag{5}$$

If a crystal is placed in a single-mode ring resonator, the radiation emerging from the resonator is determined uniquely by the field $E(0)$ on the front face of a crystal. This is related to the field $\vec{\mathscr{E}}$ penetrating the resonator by

$$\mathbf{E}(0) = \vec{\mathscr{E}} + R e^{i\varphi_R}\mathbf{E}(d). \tag{6}$$

Here R^2 and φ_R are the resultant values of the reflection coefficient and of the phase shift in the resonator; $E(d)$ is the field on the rear face of the investigated crystal.

If we use Eq. (6), we find that the condition for the POB given by Eq. (5) consists of the following form in the case of a ring resonator:

$$\mathrm{Re}\left[\left(1-Re^{i\varphi_R}E_y(d)/E_y(0)\right)\left(1-R\bar{e}^{i\varphi_R}E_x^*(d)/E_x^*(0)\right)\right]=0, \tag{7a}$$

$$\arg E_y(0)/E_x(0) = 0. \tag{7b}$$

We are using here the fact that $\psi_i = \mathrm{Re}\mathscr{E}_y/\mathscr{E}_x$ and $\psi_t \propto \mathrm{Re}E_y(0)/E_x(0)$ if $|E_y(0)/E_x(0)|\ll 1$.

If a crystal is placed in a Fabry–Perot resonator, the field inside this resonator can be represented conveniently by a superposition of waves traveling to the "right" and "left":

$$\mathbf{E}(z) = \mathbf{E}_r(z)\exp(ikz) + \mathbf{E}_l(z)\exp(-ikz),\ k=\omega n/c \tag{8}$$

(and the corresponding equation for the magnetic field is similar). Then, on the front ("left") face of the crystal we have

$$\mathbf{E}_r(0) = \vec{\mathscr{E}} + \mathbf{R}_l\exp(i\varphi_{Rl})\mathbf{E}_l(0), \tag{9}$$

and on the rear ("right") face we find that

$$\mathbf{E}_l(d) = R_r\exp(i\varphi_{Rr})\mathbf{E}_r(d) \tag{10}$$

(R_l^2 and R_r^2 are the reflection coefficients of the left- and right-hand mirrors). The condition for the appearance of the POB given by Eq. (5) is now modified to the following expression if we use Eq. (9):

$$\mathrm{Re}\left[\left(1-R_l e^{i\varphi_{Rl}}E_{ly}(0)/E_{ry}(0)\right)\right.$$
$$\left.\times\left(1-R_l e^{-i\varphi_{Rl}}E_{lx}^*(0)/E_{rx}^*(0)\right)\right]=0, \tag{11a}$$

$$\arg E_{ry}(0)/E_{rx}(0) = 0. \tag{11b}$$

2. RANGE OF MANIFESTATION OF THE POLARIZATION OPTICAL BISTABILITY IN A RING RESONATOR

Explicit determination of the relationships between the parameters of a system ensuring the appearance of the POB can be obtained by substituting in the system (7) or (11) the solution of the Maxwell equations for a nonlinear crystal. In the case of a ring resonator, this solution can be obtained analytically. Then, the equations for the envelopes of the field have the form

$$\frac{dE_x}{dz} = -\frac{1}{2}\gamma_0 E_x + p_x, \tag{12}$$

where γ_0 is the reciprocal of the absorption length of the crystal; p_x are the quantities defined in Eq. (4). Substituting the solution of equations of the (12) type into the system (7) and finding extrema with respect to φ_R (which corresponds to selection of the resonator tuning optimal for the POB), we find the relationships between the parameters ensuring the POB threshold are

$$(1-R^2e^{-\nu d})(1-R^2e^{-\xi d})$$
$$-\nu^2R^2e^{-\nu d}\{1-\exp[{}^1/_2(\xi+\gamma)d]\}^2 = 0, \tag{13a}$$

$$\gamma = \alpha'' + \gamma_0,\ \xi = 2\beta'' - \alpha'' - \gamma_0,$$
$$\nu = \beta'/\beta'' \equiv (\zeta_1'-\zeta_2')/(\zeta_1''-\zeta_2''). \tag{13b}$$

It is clear from Eq. (13) that if $\xi>0$, i.e., in the same range of frequencies and the F_A-center concentrations where $2\beta''-\alpha''>\gamma_0$, the POB in a ring resonator is possible for any feedback coefficient R^2; in the range $R^2\ll 1$, the minimum thickness of the crystal must satisfy $d_{\min}\approx -\xi^{-1}\ln[R^2(1+\nu^2)]$ [an allowance is made for the fact that according to Eqs. (4) and (13), we have $\gamma>0$ and $\gamma-\xi>0$]. As R increases, the necessary thickness de-

creases and in the limit $R \to 1$ we have

$$d_{min}=(1-R^2)[(\xi+\gamma)(1+\nu^2)^{1/2}+\xi-\gamma]^{-1} \quad (1-R^2 \ll 1). \qquad (14)$$

It is clear from Eq. (14) that the POB in a ring resonator appears if the following material inequality is obeyed:

$$(\xi+\gamma)(1+\nu^2)^{1/2}+\xi-\gamma>0. \qquad (15)$$

This inequality may be satisfied even when $\xi<0$, which corresponds—as can be seen from Eqs. (4) and (12)—to a negative gain for a small component of the field E_y in the case of linearly polarized radiation. Amplification of the polarization fluctuations (i.e., amplification of $|E_y|$) in the $\xi<0$ case is possible only in a certain range of values of the difference between the phases of the components $\varphi_y-\varphi_x$ and then only if the inequality (15) is satisfied, i.e., it is possible only in the case of a sufficiently strong anisotropy of the real part of the susceptibility of the F_A centers:

$$|\nu|=(\zeta_1'-\zeta_2')/(\zeta_1''-\zeta_2'')|>2|\xi\gamma|^{1/2}/(\xi+\gamma).$$

Since in the $\nu\neq 0$ case the difference between the faces varies with the thickness of a crystal, then on the whole the amplification of the fluctuation polarizations is possible for $\xi<0$ only in a limited range of crystal thicknesses $d<d_{max}$. Consequently, the POB then exists in a range of thicknesses with upper and lower limits, and only when the feedback coefficient R is sufficiently large. It should be noted that the criterion of Eq. (15) is known to be violated at very low concentrations of the F_A centers when we have $\beta''[1+(1+\nu^2)^{1/2}]-\alpha''<\gamma_0$.

3. POLARIZATION OPTICAL BISTABILITY IN A FABRY–PEROT RESONATOR

In a crystal placed inside a Fabry–Perot resonator the field has a complex structure which is governed by the interference of opposite waves with the envelopes $\mathbf{E}_r$ and $\mathbf{E}_l$ described by Eq. (8). The equations for the envelopes of $\mathbf{E}_p$ and $\mathbf{E}_l$ are obtained from the Maxwell equations by averaging the total polarization of a crystal [multiplied by $\exp(-ikz)$ or $\exp(ikz)$, respectively] over rapidly oscillating factors of the $\exp(\pm 2ikz)$ type; if we allow from Eqs. (1) and (4), these equations become

$$\frac{dE_{rx}}{dz}=\left(\frac{1}{2}i\alpha-\gamma_0\right)E_{rx},\quad \frac{dE_{lx}}{dz}=-\left(\frac{1}{2}i\alpha-\gamma_0\right)E_{lx},$$

$$\frac{dE_{ry}}{dz}=\left[\frac{i}{2}(\alpha-\beta)-\gamma_0\right]E_{ry}-\frac{i\beta}{2}(E_{rx}^*)^{-2}[E_{ry}^*(|E_{rx}|^2 \qquad (16a)$$

$$-|E_{lx}|^2)+(E_{ly}E_{rx})^*E_{lx}], \qquad (16b)$$

$$\frac{dE_{ly}}{dz}=-\left[\frac{i}{2}(\alpha-\beta)-\gamma_0\right]E_{ly}+\frac{i\beta}{2}E_{ry}^*(E_{rx}^*)^{-1}E_{lx}. \qquad (16c)$$

We note the asymmetry of the equations for E_{ry} and E_{ly}; this is due to the fact that the field $E_{lx}\exp(ikz)+E_{lx}\exp(-ikz)$, influencing, because of the optical nonlinearity, the value of E_y is a superposition of a standing wave (which naturally does not contribute an asymmetry) and a wave traveling to the right ($|E_{rx}|>|E_{lx}|$).

The system of equations (16) subject to the boundary conditions of Eqs. (10) and (11) will now be solved analytically for a high-Q resonator on condition that $1-R_rR_l \ll 1$. The expression for the minimal thickness of a crystal d_{min} necessary for the POB in the case of optimal tuning of the resonator (optimal values of φ_{Rl}) are given by Eq. (14), where R^2 should be replaced with R_rR_l. Hence, it is clear that the POB in both Fabry–Perot and ring resonators can appear only if the condition of Eq. (15) is satisfied.

The range of values of the feedback coefficients necessary for the POB in a Fabry–Perot resonator is much narrower than for a ring resonator; even when $\xi>0$, there is a minimum value of R_rR_l below which the POB does not appear. This is due to the fact that the growth increment of the polarization fluctuations may be positive, demonstrated by the system (16), only for a wave traveling "to the right." Therefore, the ratio $|E_{ly}(0)/E_{ry}(0)|$ considered as a function of the crystal thickness d reaches its maximum value at some finite d; the maximum value of $|E_{ly}(0)/E_{ry}(0)|$ limits R_lR_r from below, as becomes clear on examination of Eqs. (10) and (11).

In general, the range of the parameters corresponding to the POB can be found by solving the boundary-value problem described by Eqs. (16), (10), and (11). Such calculations were carried out for a symmetric resonator ($R_r=R_l=R$). The dependence of the threshold (for the POB) crystal thickness d on R is shown in Fig. 1. This figure includes also the corresponding dependence for a ring resonator. For convenience, the ordinate gives the transmission of the field component E_x which is governed directly by the value of d:

$$t=\exp(-\gamma d)\equiv\exp\{-d[\gamma_0+6\zeta_2''(1+\mu)(1+5\mu)^{-1}]\}. \qquad (17)$$

It is clear from Eqs. (4), (13), and (16) that the dependence of t on R^2 is governed by the dimensionless parameters $\mu=\zeta_2''/\zeta_1''$, $\nu=(\zeta_1'-\zeta_2')(\zeta_1''-\zeta_2'')^{-1}$, and γ_0/ζ_2''. In turn, μ and ν for the specific F_A centers with a known resonance susceptibility spectrum are governed by the frequency of light, and the ratio γ_0/ζ_2'' is determined by the concentration of the centers. The curves plotted in Fig. 1 were calculated for $\mu=0.141$, $\nu=-1.98$, and $\mu=1099$, $\nu=1.20$; in the case of the F_A(Li) centers in KCl these values correspond to $\lambda=608$ and 569 nm, respectively.

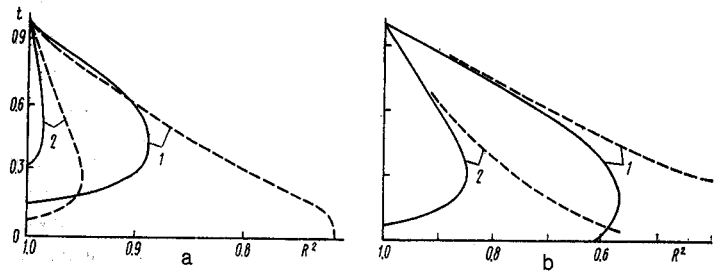

FIG. 1. Dependences of the transmission coefficient of a crystal for radiation polarized along the [110] axis, which ensures the occurrence of the polarization optical bistability, on the reflection coefficient of the mirrors R^2 in a symmetric Fabry–Perot resonator (continuous curves) and in a ring resonator (dashed curves): a) wing of the F_{A1} band ($\mu=0.141$, $\nu=-2$) for $\gamma_0/\zeta_2''=0$ (1) and 0.29 (2); b) wing of the F_{A2} band ($\mu=1099$, $\nu=1.2$) for $\gamma_0/\zeta_2''=0$ (1) and 0.22 (2).

It is clear from Fig. 1 that the values of R^2 necessary for the appearance of the POB are readily attained experimentally if μ and ν have the values given above. From the point of view of realization of the threshold-free POB it is important also that the interval of the radiation frequencies optimal for the POB corresponds to a specific relationship between the absorption coefficient γ_0 of the crystal matrix and the concentration of the centers multiplied by the oscillator strength of the resonance transition. The limits of this interval are set by the condition (15).

It is also clear that in the case of crystals containing the F_A centers and placed inside the resonator there may be practically threshold-free POB. The range of existence of the optical bistability in a Fabry–Perot resonator has upper and lower limits to the crystal thickness, and it is also limited from below in respect of the reflection coefficient of the mirrors R^2; in the case of a ring resonator there may be no upper limit to d or lower limit to R^2. There is again an optimal interval of the radiation frequencies where the POB appears most strongly.

In conclusion, it should be noted that in the case of crystals containing the F_A centers the time in which a steady-state nonlinear susceptibility is established is relatively long (it exceeds 10^{-8} sec). However, it is important to note that this time is inversely proportional to the intensity of the incident radiation[8] and can therefore be controlled easily within wide limits. It follows that in bistable devices of this type we can vary the switching times within wide limits.

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Optical parametric oscillation in $LiNbO_3$, CsH_2AsO_4, and $Ba_2NaNb_5O_{15}$ crystals pumped synchronously by a picosecond $La_2Be_2O_5$:Nd laser

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An investigation was made of the energy, time, and spectral characteristics of resonator picosecond optical parametric oscillators made of $LiNbO_3$, CsH_2AsO_4, and $Ba_2NaNb_5O_{15}$ crystals. The maximum conversion efficiencies in these crystals were 5.3, 7.2, and 12.5%, respectively. The output wavelength was tunable within the range 0.65–3 μ.

We shall report a study of optical parametric oscillators (OPOs) made of $LiNbO_3$, CsH_2AsO_4, and $Ba_2NaNb_5O_{15}$ crystals pumped synchronously using the second harmonic of a lanthanum beryllate laser with passive mode locking.[1] The duration of the pump pulses was 10 psec. Earlier investigations of resonator picosecond OPOs had been carried out using pump pulses of duration in excess of 30 psec (Refs. 2 and 3) or 3–5 psec (Refs. 4 and 5).

We determined the energy, time, and spectral characteristics of OPOs utilizing $LiNbO_3$, CsH_2AsO_4, and $Ba_2NaNb_5O_{15}$ crystals of lengths 2.5, 4, and 1 cm, respectively. The output energy of the laser acting as a master oscillator was 10–12 mJ. Its second harmonic was generated in a KDP crystal 2-cm long with an efficiency of 25%. The repetition frequency of the pulses was 5 Hz. The OPOs were pumped by a train of 10–20 pulses whose energy was 2.5 mJ. The duration of an ultrashort pulse was 10 psec and the width of the spectrum was 7.5 cm^{-1}. Each OPO consisted of two plane mirrors with a crystal between them. The reflection coefficient of one (nontransmitting) mirror was 100% in the range 0.65–3 μ, whereas the reflection coefficient of the exit mirror at the OPO emission wavelength could be varied within the range 25–100%. The optical lengths of the resonators were matched by continuous displacement of the exit mirror placed on a micrometer stage. The radiation wavelength was tunable by variation of the temperature of a crystal. The results were recorded and analyzed statistically using a computer-based system.[6] The spectra were recorded with an MDR-3 monochromator and the pulse duration was determined using an Agat SF-3 image-converter camera. The energy characteristics were measured employing an ILD-2 laser dosimeter.

Ultrashort pulses were generated in these OPOs for any of the exit mirrors employed in this study. The radiation energy was measured in a beam which did not pass through the resonator. Figure 1 shows the dependences of the energy

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