The theory of optical bistability and self-oscillations in a CuCl crystal

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The theory of optical bistability (OB) and self-oscillations in condensed media involving excitons and biexcitons is elaborated. A CuCl crystal, where convincing experimental evidences for biexciton existence are available, is chosen as a model. Equations of states of the steady-state OB theory, differing substantially from the analogous equations in the model of two-level atoms and exciton spectrum domain, are derived. Stability of steady states is studied, and self-oscillations are shown to appear in unstable sections of OB curves. Either regular or stochastic self-pulsations with formation of complicated limit cycles and strange attractors in the phase space are possible, depending on the system parameters. A scenario for the transition to the dynamic chaos mode is found. The dynamic OB is also studied, and deformation of a pulse transmitted through a crystal is shown. The occurrence of spatial turbulence in a system of coherent excitons, photons, and biexcitons in crystals is predicted.

1. INTRODUCTION

At present, the phenomenon of optical bistability (OB) has become an object of numerous theoretical and experimental investigations and is, in essence, an independent area of nonlinear optics.[1] Semiconductors possessing high values of nonlinear susceptibilities are the most promising objects for OB.[2] A fairly complete review of OB in semiconductors is given in Refs. 3 and 4. Of special interest is the investigation of OB in the case of resonant excitation of excitons and biexcitons due to "giant" nonlinearities[4] and the short relaxation times of quasiparticles.

OB in the exciton spectrum was first studied theoretically in the works by Elesin and Kopaev.[5] The work by Kochelan *et al.*[6,7] treats the same range of problems. In our work,[8–13] a theory of optical bistability, switching, regular and stochastic self-pulsations in a system of coherent excitons of high density with allowance for exciton-exciton interaction was constructed on the basis of the Keldysh equations.[14] This phenomenon was revealed experimentally in the works by Dneprovskii *et al.*[15,16]

The notion of a biexciton, whose existence was predicted by Moskalenko[17] and Lampert,[18] is widely invoked to interpret new absorption and fluorescence bands in semiconductors. At present, convincing experimental proofs of the existence of biexcitons in CuCl crystal are available. These proofs are based on the observation of the *M*-band resulting from biexciton radiative recombination[19–21] and two-photon excitation of biexciton from the crystal ground state.[22,23] In the work of Gogolin, Rashba, and Hanamura,[24–27] the corresponding transitions are shown to feature giant oscillator strengths favoring the most pronounced manifestation of nonlinear phenomena in this frequency range.

In a CuCl crystal, the biexciton bond energy is of the order of $I_{\text{biex}} \sim 40$ meV, and the exciton absorption band and the *M*-band of biexciton recombination are well separated from each other in essential contrast to the situation in CdS and CdSe crystals where the biexciton bond energy is small, $I_{\text{biex}} \sim 3$ meV. Hence in studies of nonlinear optical phenomena in crystals of the CdS type, photons of the same pulse can excite excitons from the ground crystal state and convert them to biexcitons in the case when the spectral width of the incident radiation is $\hbar\Delta\omega \sim I_{\text{biex}}$. In a CuCl crystal, the photon frequency resonant with the exciton transition frequency has a large resonance detuning with respect to the transition in the region of the fluorescence *M*-band due to the large bond energy.

On the other hand, it is known that the observation of coherent nonlinear effects such as nonlinear nutation,[28–30] self-induced transparency,[31,32] and OB with the participation of excitons and biexcitons requires the prior preparation of a coherent exciton system. Then the excitons can convert optically to biexcitons under the action of a pulse resonant with the exciton-biexciton transition frequency. Therefore, a correct study of the OB phenomenon in a CuCl crystal necessitates simultaneous action of two independent optical pulses each being in resonance with a definite transition. Note that OB of excitons and biexcitons was studied in Refs. 33–38. We have shown in our work[33] that both amplitude and frequency hysteresis are possible in a system of coherent excitons, photons, and biexcitons. The prediction that a bistable dependence of polariton density on incident light intensity can exist in crystal two-photon excitation was made in Ref. 35. In Ref. 36, an intensity-dependent dielectric function exhibiting bistable behavior was calculated using the density matrix formalism. The OB phenomenon in a system of excitons and biexcitons in a Fabry–Perot cavity was investigated in Ref. 37 with allowance for the Kerr correction for the exciton dielectric function. The OB phenomenon in a system of coherent excitons and biexcitons for various quantum transitions was studied theoretically in Ref. 38. The common disadvantage of these works is in the fact that propagation effects are not included, stability of steady-state curves of the OB theory is not examined, switching times between

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OB branches are not studied, and self-pulsations appearing in a system due to the hysteresis curve instability are not investigated. In Ref. 39 we studied steady-state and transient OB and mutilstabilities involving excitons and biexcitons for crystals of the CdS type. In this paper, we construct the theory of OB, switching times, regular and chaotic self-pulsations in a CuCl crystal with the participation of coherent excitons and biexcitons.

2. THE HAMILTONIAN OF THE PROBLEM AND THE BASIC EQUATIONS

In the most general case, the coherent interaction of resonant laser radiation with excitons and biexcitons differs essentially from the two-level atom model. The point is that a system of excitons and biexcitons differs from a disordered combination of atoms or impurity centers in method of preparation and organization of the initial state. Excitons and biexcitons are transient crystal excitations, whereas a system of two-level atoms can be in the ground state as long as desired. Therefore, as noted earlier, for the OB phenomenon to occur the exciton system should be prepared in advance using a laser source with photon energies $\hbar\omega_1 = E_g - I_{\rm ex}$, where E_g is the band gap and $I_{\rm ex}$ is the exciton bond energy. The photons of the second laser pulse causing exciton-biexciton conversion have an energy $\hbar\omega_2 = E_g - I_{\rm ex} - I_{\rm biex}$.

For simplicity, we shall use the three-level model, as applied to a copper chloride crystal, in which the energies of the "crystal ground state-exciton" and "exciton-biexciton" transitions differ by the biexciton bond energy. Suppose that photons of the first pulse are in resonance with a transition in the exciton spectral range, and photons of the second pulse are in resonance with the region of the fluorescence M-band of a CuCl crystal. We consider one macrofilled mode of coherent (in the Bogolyubov sense) excitons and biexcitons and one macrofilled mode of coherent photons of every pulse. The full Hamiltonian of the problem consists of a sum of Hamiltonians of free excitons, biexcitons, and fields, as well as the interaction Hamiltonian which in the model adopted has the form

$$H_{\rm int} = -\hbar g(aE_1^+ + a^+E_1^-) - \hbar gG(a^+bE_2^- + ab^+E_2^+), \quad (1)$$

where $a^+(b^+)$ is the creation operator of exciton (biexciton), g is the constant of exciton-photon interaction, G is the exciton-biexciton optical conversion coefficient and $E_j^{+(-)}$ is the positive (negative)-frequency component of the electric field of electromagnetic wave of the jth pulse.

The equations of motion for the exciton and biexciton amplitudes have the form

$$i\frac{\partial a}{\partial t} = \omega_{\rm ex}a - i\gamma_{\rm ex}a - gE_1^+ - gGbE_1^-, \quad (2)$$

$$i\frac{\partial b}{\partial t} = \omega_{\rm biex}b - i\gamma_{\rm biex}b - gGE_2^+a, \quad (3)$$

where $\hbar\omega_{\rm ex}$ and $\hbar\omega_{\rm biex}$ are the energies of exciton and biexciton formation, respectively, and $\gamma_{\rm ex}$ and $\gamma_{\rm biex}$ are the decay constants of excitons and biexcitons determining the rate of quasiparticle escape from coherent modes to incoherent. These constants were inserted phenomenologically into the equations of motion. Note that these equations can be derived rigorously in terms of the quantum theory of fluctuations and decays from the flow part of the corresponding Fokker–Planck equation.[40] The equations for the positive-frequency field component have the form

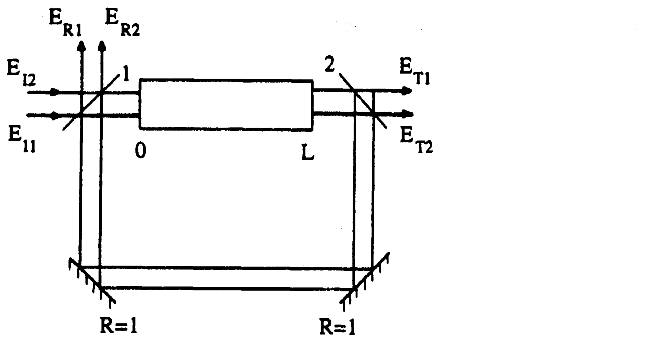

FIG. 1. Ring cavity diagram. $E_{I,j}$, $E_{R,j}$, and $E_{T,j}$ are the amplitudes of incident, reflected, and transmitted fields, respectively.

$$c_1^2\frac{\partial^2 E_1^+}{\partial z^2} - \frac{\partial^2 E_1^+}{\partial t^2} = 4\pi\hbar g\frac{\partial^2 a}{\partial t^2}, \quad (4)$$

$$c_1^2\frac{\partial^2 E_2^+}{\partial z^2} - \frac{\partial^2 E_2^+}{\partial t^2} = 4\pi\hbar gG\frac{\partial^2(a^+b)}{\partial t^2}. \quad (5)$$

Here, c_1 and c_2 are the velocities of field propagation in the medium.

Let us represent the solution of Eqs. (2)–(4) in the form of a product of slowly-varying envelopes and rapidly oscillating components with carrier frequencies ω_1 and ω_2 and wave vectors k_1 and k_2:

$$\begin{aligned} a &= \tilde{A}\exp(-i\omega_1 t + ik_1 z), \\ E_1^+ &= e_1^+\exp(-i\omega_1 t + ik_1 z), \\ b &= \tilde{B}\exp(-i(\omega_1+\omega_2)t + i(k_1+k_2)z), \\ E_2^+ &= e_2^+\exp(-i\omega_2 t + ik_2 z). \end{aligned} \quad (6)$$

Let us consider further the OB theory in the ring cavity geometry. Let a sample of length L be placed between the input and the output cavity mirrors which are characterized by a transmission coefficient T. The two remaining mirrors are considered to be totally reflecting (see Fig. 1). The boundary conditions for the ring cavity have the form

$$\begin{aligned} E_j^+(0,t) &= T^{1/2}E_{I,j} + Re^{\beta_{0,j}}E_j^+(L,t-\Delta t) \\ E_{T,j}(t) &= T^{1/2}E_j^+(L,t), \end{aligned} \quad (7)$$

where $E_{T,j}$ and $E_{I,j}$ are the amplitudes of the fields incident on the cavity input mirror and transmitted through the cavity, $R = 1 - T$ is the reflection coefficient of the cavity mirrors *1* and *2*, $\Delta t = (2l + L)/c_0$ is the retardation time introduced by the feedback, c_0 is the velocity of light *in vacuo*, and $\beta_{0,j}$ is the phase increment in the cavity. Substituting (6) in Eqs. (2)–(5) in the slowly-varyingenvelope approximation[1,8] and the mean-field approximation[1,9] with allowance for the boundary conditions (7), we get

$$\frac{\partial X_1}{\partial\tau}=\sigma_1(-X_1+2C_1A+Y_1), \tag{8}$$

$$\frac{\partial X_2}{\partial\tau}=\sigma_2(-X_2+2C_2AB+Y_2), \tag{9}$$

$$\frac{\partial A}{\partial\tau}=-dA-d(X_1+X_2B), \tag{10}$$

$$\frac{\partial B}{\partial\tau}=-B+X_2A, \tag{11}$$

where X_j, Y_j, A, B are the normalized field amplitudes, τ is the dimensionless time, and C_j are the constants of the OB theory defined by the expressions

$$X_j=\frac{e_{T,j}}{e_s},\quad Y_j=\frac{e_{I,j}}{e_s},\quad A=i\frac{\tilde{A}}{A_s},\quad B=\frac{\tilde{B}}{B_s},$$
$$e_s=\frac{\sqrt{T\gamma_{\rm ex}\gamma_{\rm biex}}}{gG}\quad A_s=\frac{\sqrt{\gamma_{\rm ex}}}{\sqrt{\gamma_{\rm biex}}}G^{-1},\quad B_s=G^{-1},$$
$$\tau=\gamma_{\rm biex}t,\quad d=\frac{y_{\rm ex}}{\gamma_{\rm biex}},\quad C_j=\frac{\alpha_jL}{4T},\quad \sigma_j=\frac{c_jT}{\gamma_{\rm biex}L}, \tag{12}$$
$$\alpha_j=\frac{4\pi\hbar g^2\omega_j}{\gamma_{\rm ex}c_j},\quad j=1,2.$$

Equations (8)–(11) describe temporal evolution of coherent excitons, biexcitons, and electromagnetic fields in the ring cavity and are the basis for studying OB.

In the steady-state case, $\dot{X}_1=\dot{X}_2=\dot{A}=\dot{B}=0$, we get coupled equations of states of the OB theory in a CuCl crystal:

$$Y_1=X_1\left(1+2C_1\frac{1}{(1+X_2^2)}\right), \tag{13}$$

$$Y_2=X_2\left(1+2C_2\frac{X_1^2}{(1+X_2^2)}\right). \tag{14}$$

These equations express the relation between the normalized wave amplitudes at the input (Y_1 and Y_2) and the output (X_1 and X_2) of the cavity and are in essence the equations for a nonlinear optical quadrupole with two input and two output channels.[38] The variation of one or both parameters provides flexible variation of the quadrupole output parameters. Optical bistability is possible in both channels. For instance, it is easily seen from (13)–(14) that at a fixed Y_1 value, nonlinear dependence of the amplitude X_2 of radiation leaving the cavity in the second channel is determined not only by the bistability parameter in the second channel C_2 but by the bistability constant of the first channel and the pumping amplitude in the first channel as well. This fact presents a unique possibility of controlling the nonlinear behavior in the second channel by the pumping in the first channel. The criterion for the existence of OB in the second channel is the inequality

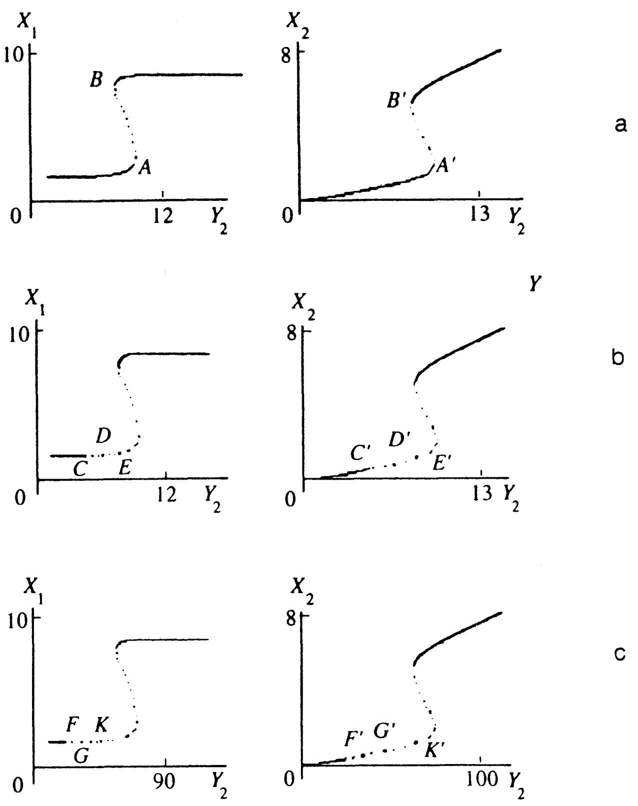

FIG. 2. Steady-state OB. X_1, $X_2(Y_2)$ in the ring cavity at various values of the parameters: a) $Y_1=10$, $C_1=5$, $C_2=4$, $d=0.1$; b) $Y_1=10$, $C_1=5$, $C_2=4$, $d=0.7$; c) $Y_1=40$, $C_1=20$, $C_2=19$, $d=0.7$.

$$Y_1^2>Y_k^2=2\frac{(1+2C_1)^2}{C_2}, \tag{15}$$

which differs fundamentally from the conditions of OB occurrence in two-level systems[1,34] and in the exciton spectrum range.[9,12,13] The nonlinear dependence of the output radiation amplitudes X_1 and X_2 on the input radiation amplitudes under the conditions such that OB occurs is shown in Fig. 2 for various values of the parameters.

3. COMPUTER EXPERIMENT

It is of great interest to investigate the stability of the OB curve in connection with the possible development of self-pulsations in unstable sections of the OB curves. The investigation of stability of optical hysteresis steady states with respect to small perturbations is determined by the characteristic equation for the Jacobian of the system:

$$|J-\lambda E| = \begin{vmatrix} -\sigma_1-\lambda, & 0, & 2C_1, & 0 \\ 0, & -\sigma_2-\lambda, & 2C_2B, & -2C_2A \\ d, & dB, & -d-\lambda, & -dX_2 \\ 0, & A, & X_2, & -1-\lambda \end{vmatrix},$$

where E is the unit matrix. If the real parts of all the roots of the characteristic equation are negative, the correspond-

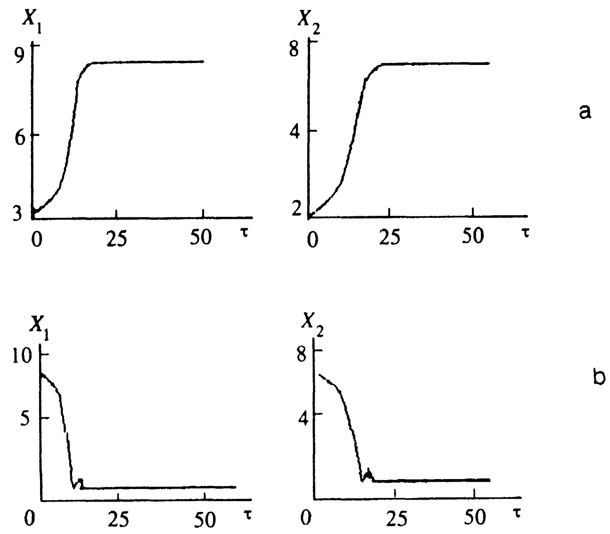

FIG. 3. Optical switching from the lower to the upper branches and from the upper to the lower branches (the evolution of points A, B in Fig. 2a, respectively).

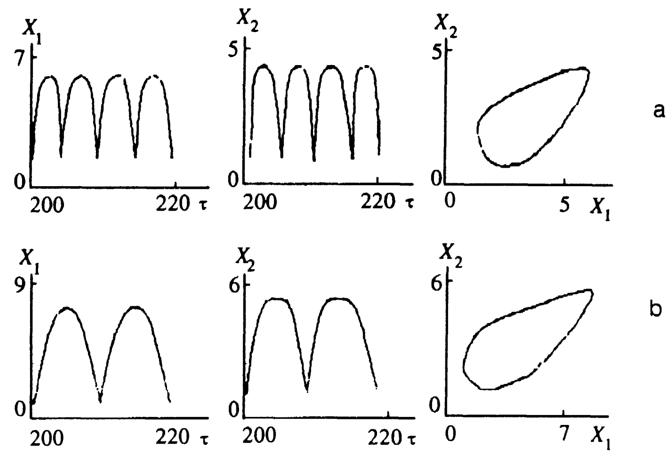

FIG. 4. Temporal evolution of the system at $Y_1=10$, $C_1=5$, $C_2=4$, $d=0.7$. a) evolution of point C, Fig. 2b; b) evolution of point D, Fig. 2b.

ing steady states are stable with respect to small perturbations. Using the Routh-Hurwitz criterion, stability of steady states at various values of the parameters was investigated. For $C_1=5$, $C_2=4$, $d=0.1$, $Y_1=10$, both the upper and the lower branches of the OB curves are stable (Fig. 2a). It is of interest to study switching times between them. The basis for the investigation of the switching times is the set of equations (8)–(13). At present, there is no standard algorithm for solving nonlinear differential equations of general form and it is a difficult problem to obtain analytical solutions of Eqs. (8)–(13). In this connection, we performed a computer experiment. The initial conditions are taken in such a way that they correspond to the Y_2 value near the "up-down" switching threshold. At time $t=0$, a discontinuous change the pumping Y_2 is prescribed such that $Y_2+\Delta Y_2$ lies to the other side of the corresponding switching threshold. Switching from the lower OB branches to the upper ones and from the upper branches to the lower ones is shown in Fig. 3. As seen from the figure, the switching times are of the same order and are $\tau \cong (10-15)\gamma_{ex}^{-1}$ s. This differs essentially from the corresponding switching times in the model of two-level systems where up and down switching times are essentially different. Since the relaxation times of excitons and biexcitons in CuCl are $t \sim 10^{-11}$ s (Ref. 41), optical switching times in a system of coherent excitons lie in the picosecond range. At certain parameter values, a part of the OB curves becomes unstable. The nonlinear dependences of X_1, $X_2(Y_2)$ are plotted in Fig. 2b for $Y_1=10$, $C_1=5$, $C_2=4$, and $d=0.7$. The upper parts of the OB curves are stable as before, whereas a part of the lower branches becomes unstable. In this case, nonlinear self-pulsations develop in the system. As one moves from the point $C(C')$ to the point $D(D')$, bifurcation of oscillation period doubling is observed in the system, and the phase trajectory goes into a limit cycle (Fig. 4). At the point E, the system switches to the upper branches of the OB curves. The dynamic evolution of the system becomes more complicated as the parameters increase ($C_1=20$, $C_2=19$, $d=0.7$). Near the point F (Fig. 2c) corresponding to the initial point of instability regions, nonlinear periodic self-oscillations appear in the system, and the phase trajectory goes into a limit cycle in the shape of a figure eight curve. As the imaging point moves towards the window center, the oscillations become more complicated, new harmonics appear in their spectrum, and finally, stochastic self-oscillation and an optical turbulence mode develops in the middle part of the window [the point $G(G')$]. The stochastic self-modulation process and the corresponding projections of the phase trajectories are shown in Fig. 5b. In contrast to the famous Lorenz chaos,[42–44] where stochastic oscillations and strange attractor creation are associated with jumps between the corresponding equilibrium states, in our case, stochasticity is related to the creation of a chaotic attractor in the four-dimensional phase space which is filled with phase trajectories in a complicated manner. As the imaging point moves further to the right, regular nonlinear oscillations develop in the system again, and the phase trajectories go into a limiting cycle of complicated structure (Fig. 5c) after which the system makes a jump to the upper stable branches of the OB curves. In experimental investigations of OB, not static but dynamic OB is often observed, which results from the comparison of time-dependent external pumping with the corresponding system response. OB of such type was first considered by Bischofberger and Shen.[45] The behavior of a nonlinear Fabry–Perot interferometer filled with a Kerr medium under the action of pulses of various shapes has been studied theoretically and experimentally. The authors obtained a very good agreement between theory and experiment. As for dynamic OB in a system of excitons, photons, and biexcitons, this problem has not been solved yet. We performed a computer experiment in which the set of nonlinear differential equations (8)–(11) describing the dynamics of coherent excitons, photons, and biexcitons was solved numerically with

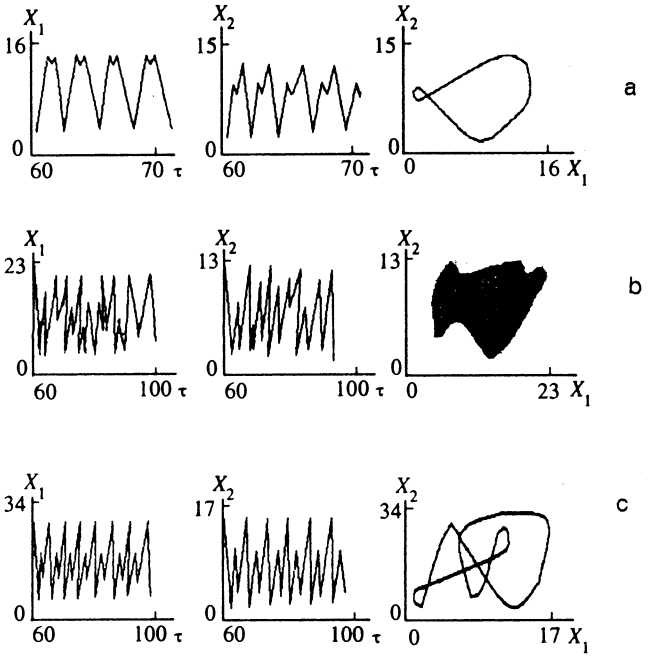

FIG. 5. Temporal evolution of the system at $Y_1=40$, $C_1=20$, $C_2=19$, $d=0.7$. a) evolution of point F, Fig. 2c; b) evolution of point G, Fig. 2c; c) evolution of point K, Fig. 2c.

allowance for the initial conditions for the ring cavity. The external pumping $Y(\tau)$ was a parabolic function of time. The results of the experiment performed are presented in Fig. 6. In Fig. 6a, the shape of the incident pulse of length $\tau=100$ and the amplitude of field leaving the cavity are presented for the case when both the upper and the lower branches of the OB curves are stable. As a result, OB with the counterclockwise motion is present in both channels. In Fig. 6b, hysteresis curves with counterclockwise motion are also presented. However, in the lower branch there are sections with self-pulsations of large amplitude exhibiting both regular and chaotic behavior. This is associated with the fact that a system of coherent excitons, photons, and

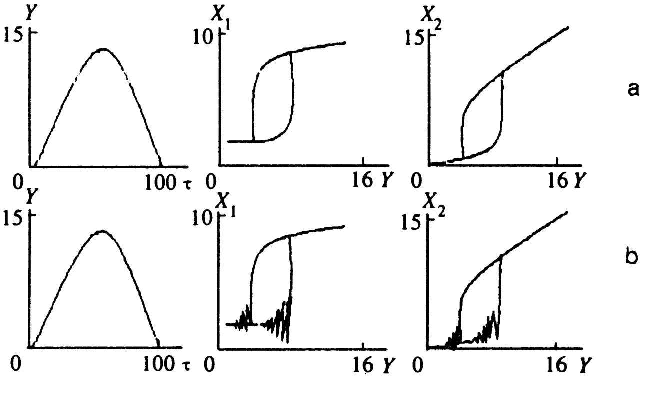

FIG. 6. Dynamic bistability for external pulse of the parabolic shape of length a) $\tau=100$, $C_1=5$, $C_2=4$, $d=0.1$; b) $\tau=100$, $C_1=5$, $C_2=4$, $d=0.7$.

biexcitons falls into the instability region at the corresponding values of the parameters. Note in conclusion that the self-oscillations studied arising due to instability of the steady states are one more example of the occurrence of temporal structures in nonlinear dynamic systems. Meanwhile, the input equations are nonlinear partial differential equations describing the space-time evolution of coherent quasiparticles in condensed media. A theory for the evolution of spatial turbulence has been developed for equations of this type.[46] A new class of transitions of the "order-chaos" type in the form of moving transition fronts was found. Analogous phenomenon may occur in a system of excitons, photons, and biexcitons as well. Along with dynamic optical turbulence, the development of spatial turbulence and the occurrence of structures of the "order-chaos" and "chaos-order" types are possible.

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