Dynamic optical chaos of coherent excitons and biexcitons in semiconductors

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We predict the possibility (in principle) of ultrashort dynamic chaos appearing in a system of coherent excitons, photons, and biexcitons in semiconductors. We show that, depending on the value of the exciton–biexciton conversion constant and on the initial quasiparticle concentration, both regular and stochastic oscillations are possible in the system. The dynamic stochasticity of coherent quasiparticles is caused by breakdown of integrals of motion of the system.

1. INTRODUCTION

At present attention is focused on studies of nonlinear coherent interaction of laser radiation with matter. A special place among these phenomena is occupied by the problem of dynamic chaos in both dissipative and Hamiltonian dynamical systems.

In view of the large values of nonlinearities in semiconductors in the excitonic part of the spectrum, the problem of nonlinear cooperative phenomena in a system of coherent excitons, photons, and biexcitons is of considerable interest. Among such phenomena are self-induced transparency and nonlinear optical nutation.[1–7] A theory of dynamic chaos and formation of strange attractors of excitons and biexcitons in solids, which allows for dissipation processes, has been built by a number of researchers.[8–14] They have shown, for instance, that dynamic evolution of coherent excitons, photons, and biexcitons in condensed media, with allowance for dissipation processes, is described by a generalized system of Lorenz equations in a four-dimensional phase space. Basing their reasoning on Keldysh's equations generalized to the case of coherent pumping and damping, the authors of Refs. 15–18 show how in principle it is possible for dynamic chaos to emerge in a system of coherent excitons and photons.

Developing methods for generating and forming light pulses with lengths up to 10^{-15} s (femtosecond) has become one of the most striking achievements of laser physics in recent times.[19] This makes it possible to lift all restrictions on the characteristic times of the dynamics of a system of coherent excitons and biexcitons in semiconductors, the relaxation times of the latter being of the order of 10^{-9}–10^{-11} s, and to continue studying the physical process when ultrashort laser pulses act on matter. The interest is due, on the one hand, to the prediction and investigation of new nonlinear effects when ultrashort laser pulses act on matter, and on the other to the use of these effects for ultrafast data processing, where chaotic instability is undesirable. Hence, fixing values in the parameter space at which dynamic chaos sets in is of great practical importance, too.

Dynamic chaos in optical Hamiltonian systems was studied in the model of two-level atoms in Refs. 20–23. But similar phenomena in the excitonic part of the spectrum have still to be studied.

This paper is devoted to a new cooperative nonlinear phenomenon, the ultrashort dynamic chaos of coherent excitons, photons, and biexcitons in condensed media. In contrast to dissipative dynamic chaos studied in Refs. 8–18 and caused by the production of strange attractors in a diminishing volume of the phase space, when the system of photons, excitons, and biexcitons is open (the length of laser pulses is of the order of, or greater than, the relaxation time), ultrashort optical chaos evolves in times shorter than the relaxation time, when the system of coherent quasiparticles is Hamiltonian. In this case the volume of the phase space occupied by the system is invariant and dynamic chaos appears because integrals of motion of the system break down and a stochastic layer appears in the phase space near the separatrix.

2. STATEMENT AND HAMILTONIAN OF PROBLEM. DYNAMIC EQUATIONS

Let us examine the ultrashort temporal evolution, without allowing for dissipation processes, of a spatially homogeneous system of coherent (in the sense of Bogolyubov) excitons, photons, and biexcitons in the event of optical conversion of a biexciton into an exciton in the vicinity of the M luminescence band of a semiconductor. There is ample experimental proof at present of the existence of biexcitons in crystals, based on observations of the M band caused by radiative recombination of biexcitons.[23–26] This band is shifted toward the long-wave part of the spectrum in relation to the excitonic light-absorption band by a quantity equal to the biexciton's binding energy $\hbar\omega_M = E_g - I_a - I_b$, where ω_M is the boundary frequency of the M band, E_g the band gap, and I_a and I_b the binding energies of an exciton and a biexciton, respectively. For CuCl crystals, $I_b \sim 40$ meV (Refs. 23, 26, and 27). Hence, the photon energy $\hbar\omega_M$ possesses considerable detuning from the resonance in the transition between the ground and excitonic states of the crystal. In view of this we will not consider such transitions here. Moreover, as shown in Refs. 28 and 29, the processes of optical conversion of excitons into biexcitons and of radiative recombination of biexcitons are characterized by an

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enormous oscillator strength, which facilitates a stronger manifestation of nonlinear coherent phenomena in this region of the spectrum. Because the binding energy in CuCl and CuBr crystals is high enough, we also ignore processes of two-photon production of a biexciton from the ground state of the crystal in view of the large detuning from resonance. The relaxation times for excitons and biexcitons in CuCl are equal, respectively, to 1.38×10^{-10} s and 1.38×10^{-11} s (Ref. 30). Since, on the one hand, the temporal width of the excitation pulse must be smaller than the relaxation times, observing ultrashort dynamic chaos in a system of coherent excitons and biexcitons requires laser pulses with picosecond and subpicosecond lengths, which presently feasible ultrashort laser pulses. As shown below, exciton concentrations at which observation of ultrashort dynamic chaos in crystals of the CuCl type is possible are of the order of 10^{16}–10^{17} cm^{-3}. Thanks to the small exciton radius in CuCl ($a_a\sim 7$ Å), high exciton concentrations ($n_a\sim 10^{19}$ cm^{-3}) can be created in CuCl (Refs. 24, 31, and 32).

Our problem contains two types of nonlinearity: optical nonlinearity caused by exciton–biexciton conversion, which occurs for all levels of excitation of the crystal, and, generally speaking, the nonlinearity caused by the interaction between quasiparticles, the importance of which increases with the level of excitation of the crystal. In Refs. 26, 33, and 34 it is shown that allowing for exciton–exciton interaction leads to a shift of the excitonic level, both under conditions when the Bose–Einstein condensation of excitons is realized and under conditions of biexciton production

$$E(n_a)=\hbar\omega_a+\nu(0)n_a,\quad \nu(0)=\frac{26\pi}{3}I_a a_a^3,$$

where $\hbar\omega_a$ is the energy of exciton production, and $\nu(0)$ the Fourier transform of the exciton interaction energy. The nature of the coupling constant $\nu(0)$ depends strongly on the symmetry of the wave functions of the interacting quasiparticles. A shift of the excitonic level in a CuCl crystal has been observed in experiments.[35,36] For concentrations $n_a\sim 10^{16}$–10^{17} cm^{-3} in CuCl this shift amounts to 0.018–0.18 meV. Hence, in what follows we do not explicitly take the nonlinearity caused by exciton–exciton coupling into account, assuming that it renormalizes the quasiparticle-production energy. More than that, Khadzhi[37] has shown that in ultrashort nonlinear phenomena such as self-induced transparency and nonlinear nutation in exciton–biexciton conversion the interparticle interaction contributes little to formation of nonlinear processes because of the huge oscillator strength of the optical conversion of excitons into biexcitons.

The above estimates suggest that all the necessary criteria for experimental observation of ultrashort chaos in a system of coherent excitons and biexcitons are quite achievable. The model considered is shown schematically in Fig. 1.

The starting point in a theoretical investigation of ultrashort dynamic chaos in a system of coherent excitons, photons, and biexcitons, restricted to pulse lengths shorter

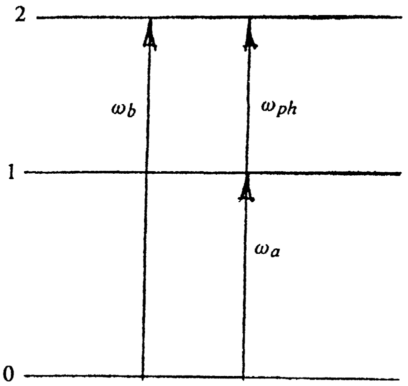

FIG. 1. Energy diagram of one-photon conversion of an exciton into a biexciton: 0 is the ground state of the system, *1* the excitonic level, *2* the biexcitonic level, $\omega_{\rm ph}$ the excitation frequency, ω_a the exciton transition frequency, and ω_b the biexciton transition frequency.

than the quasiparticle relaxation time, is the interaction Hamiltonian in the second quantization representation. Only one macrofilled mode of coherent excitons, photons, and biexcitons characterized by fixed wave vectors is considered. This makes it possible to introduce amplitudes and phases for the respective quantities.

The Hamiltonian of the problem consists of the Hamiltonian of free excitons, biexcitons, and photons and the Hamiltonian of the interaction of the radiation field with the system of coherent excitons and biexcitons:

$$\begin{aligned}H=&\hbar\omega_a a_p^+a_p+\hbar\omega_b b_{k+p}^+b_{k+p}+\hbar\omega_{\rm ph}c_k^+c_k\\&+i\sigma(k,p)g_k\hbar(a_p^++a_{-p})(c_k^++c_{-k})b_{k+p}\\&-i\sigma(k,p)g_k\hbar(a_p+a_{-p}^+)(c_k+c_{-k}^+)b_{k+p}^+,\end{aligned}\qquad(1)$$

where $a_k^+(a_k)$, $b_k^+(b_k)$, and $c_k^+(c_k)$ are the operators of creation (annihilation) of an exciton, biexciton, and photon, respectively, with a wave vector k; the quantities $\hbar\omega_a$ and $\hbar\omega_b$ are the renormalized (owing to the interparticle interaction) energies of production of an exciton and biexciton; $\hbar\omega_{\rm ph}$ is the photon energy; σ is the constant of optical conversion of an exciton into a biexciton and is defined in Ref. 6; and g_k is the dipole moment of the transition from the ground state of the crystal to the excitonic. In what follows we put $\hbar=1$. The interaction of light with excitons and biexcitons contains both a resonance part and an antiresonance part. The resonance interaction leads to coherent nutation of excitons, photons, and biexcitons studied in Refs. 1 and 6. As shown below, allowing for antiresonance terms in the Hamiltonian leads to destruction of bound states of the system of excitons and biexcitons interacting with the radiation field and to formation of dynamic ultrashort chaos.

In what follows it is expedient to go over to amplitude and phase variables:

$$a_p=\sqrt{n}\exp\{-i\varphi_a+ipx\},\quad a_p^+=\sqrt{n}\exp\{i\varphi_a-ipx\},$$

$$a_{-p}=\sqrt{n}\exp\{-i\varphi_a-ipx\},\quad a_{-p}^+=\sqrt{n}\exp\{i\varphi_a+ipx\},$$

$$b_{k+p}=\sqrt{N}\exp\{-i\varphi_b+i(k+p)x\},$$

$$b^{+}_{k+p}=\sqrt{N}\exp\{i\varphi_b-i(k+p)x\}, \quad (2)$$

$$c_k=\sqrt{f}\exp\{-i\varphi_{\rm ph}+ikx\},\quad c^{+}_k=\sqrt{f}\exp\{i\varphi_{\rm ph}-ikx\},$$

$$c_{-k}=\sqrt{f}\exp\{-i\varphi_{\rm ph}-ikx\},$$

$$c^{+}_{-k}=\sqrt{f}\exp\{i\varphi_{\rm ph}+ikx\},$$

where n and φ_a are the number and phases of excitons, and f and $\varphi_{\rm ph}$ and N and φ_b are similar quantities for photons and biexcitons, respectively. In these variables the system Hamiltonian assumes the form

$$H=\omega_a n+\omega_b N+\omega_{\rm ph}f+\sigma g\sqrt{nNf}[\sin(\varphi_b-\varphi_a-\varphi_{\rm ph})+\sin(\varphi_b-\varphi_a+\varphi_{\rm ph})+\sin(\varphi_b+\varphi_a-\varphi_{\rm ph})+\sin(\varphi_b+\varphi_a+\varphi_{\rm ph})]. \quad (3)$$

The temporal evolution of coherent excitons, photons, and biexcitons is described by the following system of equations:

$$\frac{dn}{dt}=-\frac{\partial H}{\partial\varphi_a},\quad \frac{\partial N}{\partial t}=-\frac{\partial H}{\partial\varphi_b},\quad \frac{\partial f}{\partial t}=-\frac{\partial H}{\partial\varphi_{\rm ph}},$$
$$\frac{d\varphi_a}{dt}=\frac{\partial H}{\partial n},\quad \frac{d\varphi_b}{dt}=\frac{\partial H}{\partial N},\quad \frac{d\varphi_{\rm ph}}{dt}=\frac{\partial H}{\partial f}. \quad (4)$$

Note that Eqs. (4) can also be obtained from the Heisenberg equations of motion.

We introduce the following phase variables: $\psi=\varphi_b-\varphi_a-\varphi_{\rm ph}$, $\varphi_1=\varphi_b-\varphi_a+\varphi_{\rm ph}$, $\varphi_2=\varphi_b+\varphi_a-\varphi_{\rm ph}$, and $\varphi_3=\varphi_b+\varphi_a+\varphi_{\rm ph}$. In this case we get from Eqs. (4)

$$\dot n=\frac{\lambda}{4}\sqrt{nNf}[\cos\psi+\varepsilon(\cos\varphi_1-\cos\phi_2-\cos\phi_3)],$$

$$\dot N=-\frac{\lambda}{4}\sqrt{nNf}[\cos\psi+\varepsilon(\cos\varphi_1+\cos\phi_2+\cos\phi_3)],$$

$$\dot n=\frac{\lambda}{4}\sqrt{nNf}[\cos\psi+\varepsilon(-\cos\varphi_1+\cos\phi_2-\cos\phi_3)],$$

$$\dot\psi=\Delta+\frac{\lambda}{8}\left(\sqrt{\frac{nf}{N}}-\sqrt{\frac{Nf}{n}}-\sqrt{\frac{nN}{f}}\right)\left(\sin\psi+\varepsilon\sum_{i=1}^{3}\sin\varphi_i\right), \quad (5)$$

$$\dot\varphi_1=\Delta+2+\frac{\lambda}{8}\left(\sqrt{\frac{nf}{N}}-\sqrt{\frac{Nf}{n}}+\sqrt{\frac{nN}{f}}\right)\times\left(\sin\psi+\varepsilon\sum_{i=1}^{3}\sin\varphi_i\right),$$

$$\dot\varphi_2=\Delta+\frac{2\omega_a}{\omega_{\rm ph}}+\frac{\lambda}{8}\left(\sqrt{\frac{nf}{N}}+\sqrt{\frac{Nf}{n}}-\sqrt{\frac{nN}{f}}\right)\times\left(\sin\psi+\varepsilon\sum_{i=1}^{3}\sin\varphi_i\right),$$

$$\dot\varphi_3=\Delta+2+\frac{2\omega_a}{\omega_{\rm ph}}+\frac{\lambda}{8}\left(\sqrt{\frac{nf}{N}}+\sqrt{\frac{Nf}{n}}+\sqrt{\frac{nN}{f}}\right)\times\left(\sin\psi+\varepsilon\sum_{i=1}^{3}\sin\varphi_i\right),$$

where $\lambda=8\sigma g/\omega_{\rm ph}$ and $\Delta=(\omega_b-\omega_a-\omega_{\rm ph})/\omega_{\rm ph}$, and a dot denotes a derivative with respect to dimensionless time $\tau=t\omega_{\rm ph}$. The parameter ε is introduced in such a manner that $\varepsilon=0$ when the antiresonance terms are ignored, while at $\varepsilon=1$ the system of equations (5) is equivalent to (4). In the resonance approximation ($\varepsilon=0$) the system has three integrals of motion,

$$f+N=C,\quad N+n=C_1,\quad n-f=C_2. \quad (6)$$

Combining this with (5) readily yields

$$\dot\psi=\frac{\partial P}{\partial N}=\Delta+\frac{\lambda}{8}\left(\sqrt{\frac{(C_1-N)(C_N)}{N}}-\sqrt{\frac{(C-N)N}{C_1-N}}-\sqrt{\frac{(C_1-N)N}{C-N}}\right)\sin\psi, \quad (7)$$

$$\dot N=-\frac{\partial P}{\partial\psi}=-\frac{\lambda}{4}\sqrt{N(C-N)(C_1-N)}\cos\psi,$$

$$P=N\Delta+\frac{\lambda}{4}\sqrt{N(C-N)(C_1-N)}\sin\psi,$$

where $P=\omega_{\rm ph}^{-1}(H-C_1\omega_a-C\omega_{\rm ph})$ is an additional integral of motion and assumes the role of the Hamiltonian in the parameter space (N,ψ). In the resonance approximation the system of coherent excitons, photons, and biexcitons nutates in time with a frequency that strongly depends on the initial quasiparticle concentrations. The phase trajectories in this case are closed curves that depend on the integrals of motion and on the resonance detuning.

At $P=C\Delta$ and $C_1=C$ the phase trajectory becomes a separatrix, which corresponds to an aperiodic oscillation regime. As Belobrov, Zaslavskiĭ, and Tartakovskiĭ have shown,[21,22] arbitrarily small perturbations destroy the trajectories near the separatrix, with the motion of excitons, photons, and biexcitons becoming stochastic.

3. BREAKDOWN OF INTEGRALS OF MOTION AND STOCHASTIZATION OF PHASE TRAJECTORIES

Since from the very beginning the states of excitons, photons, and biexcitons are assumed to be macrofilled, we cannot consider the initial values N_0, n_0, and f_0 to be zero.

However, in what follows we extrapolate cases of the type $n_0 \gg N_0$ to the initial conditions of the type $n_0=0$, $N_0=0$, etc.

Introducing the notation $\bar{\lambda} = \lambda\sqrt{C_1}$, $\bar{n}=n/C_1$, $\bar{f}=f/C_1$, $\bar{N}=N/C_1$, and $\bar{C}=C/C_1$, going over to the variables N and $\dot{N}$, and dropping the bar, we obtain in from (7) the unperturbed case

$$\dot{N}^2=\frac{\lambda^2}{16}N(1-N)(C-N)-(P-N\Delta)^2. \tag{8}$$

Generally, the solution to Eq. (8) has the form

$$N=N_3+N_{23}sn^2\left(\frac{\sqrt{N_{13}}}{8}\lambda\tau+F(\varphi_0),K\right). \tag{9}$$

Here F is the elliptic integral of the first kind, $K = \sqrt{N_{23}/N_{13}}$ the modulus of the elliptic function, $\varphi_0 = \sin^{-1}\sqrt{(N_0-N_3)/N_{23}}$, and $N_{ik}=N_i-N_k$, with $N_3<N_2<N_1$ the roots of the equation

$$\frac{\lambda^2}{16}N(1-N)(C-N)-(P-N\Delta)^2=0. \tag{10}$$

At $C=1$ and $P=\Delta$ we have

$$N=\frac{16\Delta^2}{\lambda^2}+\left(1-\frac{16\Delta^2}{\lambda^2}\right)\tanh^2\left(\frac{1}{8}\left(1-\frac{16\Delta^2}{\lambda^2}\right)\lambda\tau+\ln\left(\tan\frac{\varphi_0}{2}+\frac{\pi}{4}\right)\right), \tag{11}$$

that is, all excitons and photons are converted into biexcitons, which finishes the evolution of the system.

We note that P and Δ impose restrictions on the initial conditions. The solution of Eq. (8) has physical meaning when $N_0 \geq N_3$. We also note that for nonzero Δ aperiodic motion is possible, while periodic oscillations can also occur at $\Delta=0$. These solution classes were not discussed in our earlier papers.

The variation, by a perturbation, of the integral of the motion P with allowance for canonical variables N and ψ is described by the equation

$$\dot{P}=\left\{\frac{\lambda^2}{32}[3N^2-2N(1+C)+C]\sum_{i=1}^{3}\sin(\varphi_i-\psi)-\frac{\lambda\Delta}{4}\sqrt{N^3-(1+C)N^2+CN}\sum_{i=1}^{3}\cos\varphi_i\right\}\times\left[1+\frac{P-N\Delta}{4(C-N)}+\frac{\omega_{\rm ph}(P-N\Delta)}{4\omega_a(1-N)}\right]^{-1}. \tag{12}$$

Below we are interested in the system's motion near the separatrix, where $P\to\Delta$ and $C\to 1$. In this case we have for the characteristic roots the expressions:

$$N_{1,2}=C\pm\frac{P-C\Delta}{\lambda\sqrt{C}/4\pm\Delta},\quad N_3=\frac{16P^2}{\lambda^2+32P^2}. \tag{13}$$

The quantity $N(t)$ varies almost from 0 to 1, and at a turning point of the hyperbolic type its period tends to infinity. In this case we have instead of (12)

$$\begin{aligned}\dot{P}&=\frac{\lambda^2}{32}A(\tau)\sum_{i=1}^{3}\sin\theta_i,\\ \dot{\theta}_1&=\Delta+2+O(\lambda),\\ \dot{\theta}_2&=\Delta+\frac{2\omega_a}{\omega_{\rm ph}}+O(\lambda),\\ \dot{\theta}_3&=\Delta+2\left(1+\frac{\omega_a}{\omega_{\rm ph}}\right)+O(\lambda),\end{aligned} \tag{14}$$

where $A(\tau)$ is a periodic function with a period $2\pi/\omega(P)$, a height of the order of unity, and a width $2\pi/\omega_0$, with $\omega(P)$ the nonlinear oscillation frequency and ω_0 the frequency of small oscillations of the system equal to $\lambda/4$ as $C\to 1$. If we now change from (14) to a system of discrete transformations, we get

$$\begin{aligned}P_{m+1}&=P_m+\bar{\Delta}P,\quad \bar{\Delta}=\frac{\lambda^2}{32}\int A(\tau)\sum_{i=1}^{3}\sin\theta_i d\tau,\\ \theta_{im+1}&=\theta_{im}+\frac{4\pi}{\omega(P_{m+1})}\\ &=\theta_{im}+\frac{4\pi}{\omega(P_m)}-\frac{4\pi}{\omega^2P_m}\frac{d\omega(P_m)}{dP_m}\bar{\Delta}P.\end{aligned} \tag{15}$$

The nature of the solution of Eqs. (15) is determined by a quantity M (Refs. 20 and 21):

$$M=\frac{4\pi}{\omega^2(P)}\left|\frac{d\omega(P)}{dP}\right|\bar{\Delta}P. \tag{16}$$

When $M \ll 1$, the system performs conditionally periodic oscillations, and when $M \gg 1$, the motion becomes stochastic with a phase-correlation splitting time

$$R(\tau)=\left\langle\exp i\sum_{k=1}^{3}[\theta_k(\tau_1)-\theta_k(\tau_1+\tau)]\right\rangle\sim\exp\left(-\frac{3\tau}{\tau_c}\right), \tag{17}$$

where $\tau_c=[\omega(P)\ln M]^{-1}$.

The boundary of the stochastic layer is determined from the condition that $M(P_0,C,H)$ be of the order of unity, or

$$P_0=\Delta+96\pi\exp\left(-\frac{8\pi\alpha}{\lambda}\right), \tag{18}$$

with constant α of the order of unity.

The growth rate of the nutation decay in the stochastic layer is

$$\gamma_c=\tau_c^{-1}=\frac{\pi\lambda\ln M}{8\ln[(\lambda^2-16\Delta^2)/4\lambda(P-\Delta)]}. \tag{19}$$

4. COMPUTER SIMULATION

In the most general case Eqs. (5) have one integral of motion (the energy of the system) and a region of motion in phase space which is a five-dimensional hypersurface

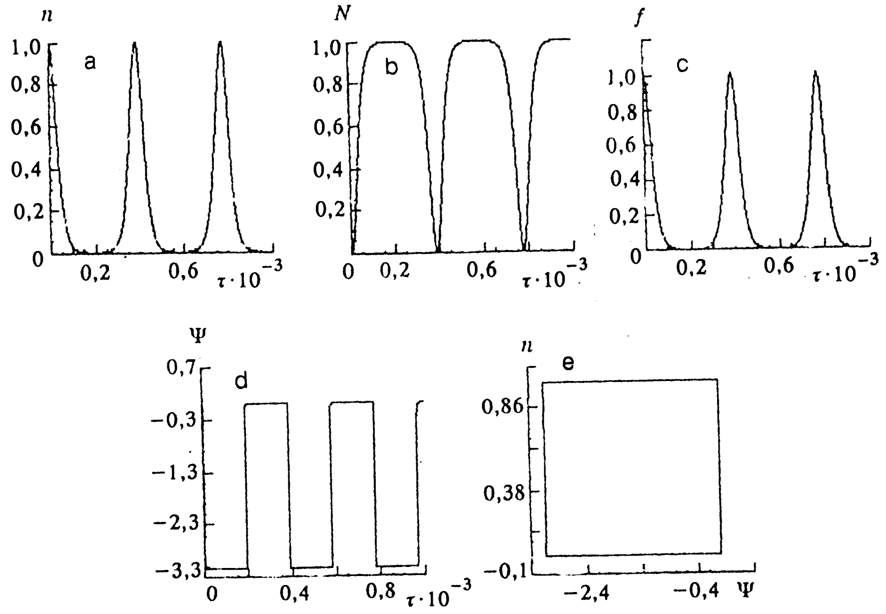

FIG. 2. Temporal evolution of the number of (a) coherent excitons, (b) biexcitons, and (c) photons, and (d) of phase, and (e) the projection of the phase trajectory on the exciton-concentration-phase plane at $C=1.001$, $p=-1.58\times10^{-9}$, and $\lambda=0.2$.

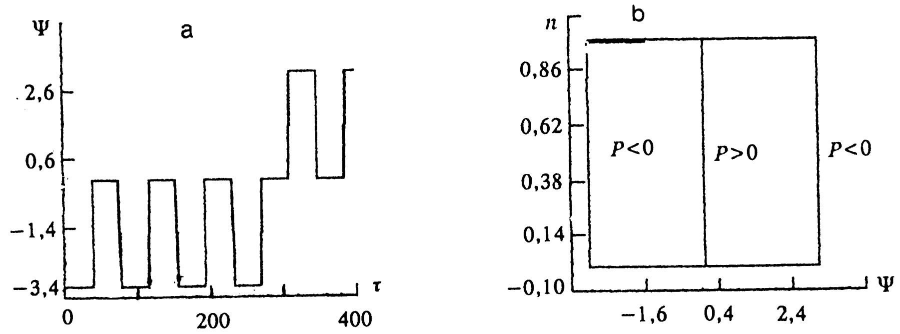

FIG. 3. (a) The time dependence of the phase, and (b) projections of the phase trajectory on the exciton-concentration-phase plane in the event of a small unit perturbation at $C=1.001$, $p=-1.58\times10^{-10}$, and $\lambda=1$.

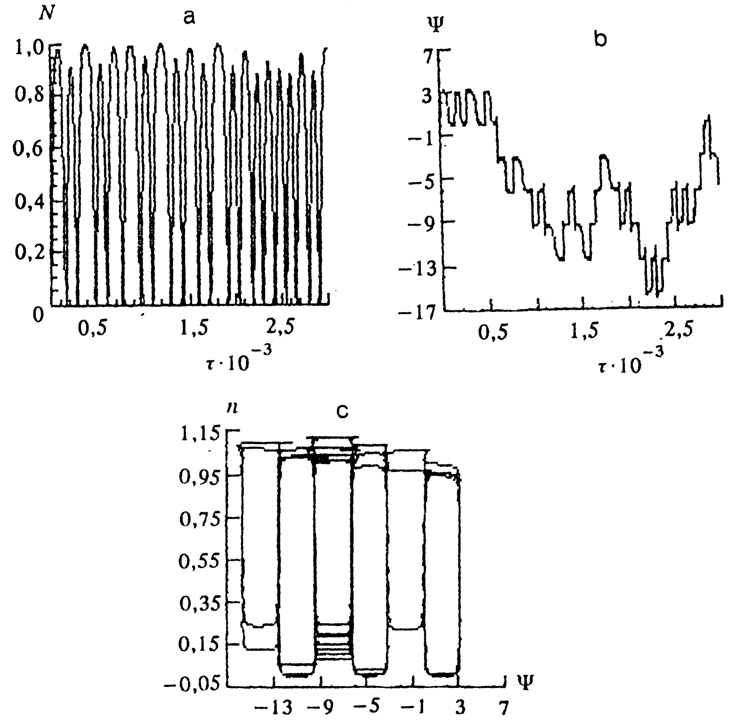

FIG. 4. Temporal evolution of the number of (a) coherent biexcitons and (b) of phase, and (c) the phase portrait projection in the exciton-concentration-phase plane at $n_0=f_0=1$, $N_0=10^{-8}$, $\varphi_{a0}=\varphi_{b0}=\varphi_{ph}=10^{-8}$, and $\lambda=0.3$.

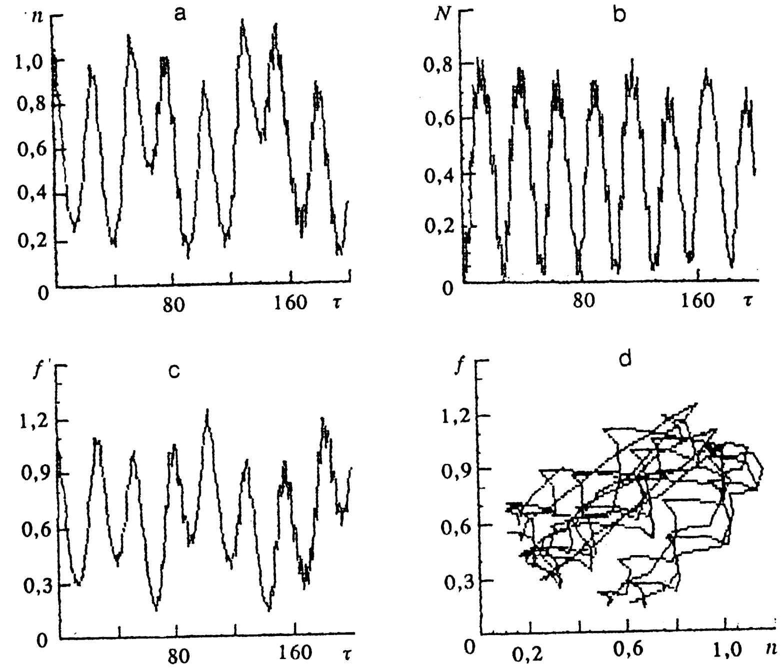

FIG. 5. The time dependence of the number of (a) excitons, (b) biexcitons, and (c) photons, and (d) the projection of the phase trajectory onto the (f,n) plane at $n_0=f_0=1$, $N_0=10^{-8}$, $\varphi_{a0}=\varphi_{ph}=1.571$, $\varphi_{b0}=10^{-8}$, and $\lambda=0.3$.

defined by (1) in a six-dimensional phase space. Ignoring the antiresonance terms leads to the appearance of additional integrals of motion, C and P.

The nature of the motion of the system of excitons, photons, and biexcitons depends on the coupling constant λ. For $\lambda \ll 1$ the motion is conditionally periodic. If $C \to 1$ and $P \to \Delta$, the phase trajectories move closer to the separatrix and land in the stochastic layer, which grows bigger as λ increases. Since at present there is no standard algorithm for solving nonlinear differential equations of the general type, and obtaining analytic solutions of the system of equations (5) is an extremely complex problem, we performed a computer simulation of the particular case $\Delta=0$.

Figure 2 shows the temporal evolution of coherent quasiparticles and of the phase, and also the projection of the phase trajectory on the exciton-concentration-phase plane for unperturbed motion. At $P=0$ the phase trajectory is a rectangle. The number of particles changes from minimal to maximal at a constant phase $\psi=n\pi$ ($n=0,1,2,\ldots$), and when an extremum is reached the phase suddenly changes: $\psi_{t+0}=\psi_{t-0}\pm\pi$. Such a phase trajectory is highly unstable and can be forced into another π-region of the phase space by an arbitrarily small unit perturbation (Fig. 3). This does not alter the nature of the temporal dependence of the number of quasiparticles. The entire phase space consists of a sequence of such rectangles with $P=0$; inside of these there are regions where $P>0$ and $P<0$ alternately. At constant values of λ the growth in $|P|$ makes the phase trajectories circular and smaller, so that trajectories with larger values of $|P|$ are "internal" with respect to trajectories with smaller $|P|$.

Figure 4 depicts the time dependence of the number of biexcitons, the resonance phase, and the projection of the phase trajectory onto the (n,ψ) plane in the event of a perturbation. The reader can see that small perturbations caused by the antiresonance terms in the Hamiltonian increase the size of the phase-variation region, owing to sudden transitions to different π-regions in the phase space and to the appearance of new harmonics in the oscillations in of number of quasiparticles, and also because the phase portrait of the respective oscillations becomes more complicated. The motion is conditionally periodic. When the coupling constant λ reaches its unity, critical value the system performs complicated nonlinear oscillations, which indicate the beginning of stochastization of the motion (Fig. 5). A further increase in λ makes the system completely stochastic, and the stochasticity region fills the entire admissible phase space (Fig. 6).

Figure 7 illustrates the development of local instability for different values of λ. The distance between two initially close trajectories is

$$D=\left[(n_1-n_2)^2+(N_1-N_2)^2+(f_1=f_2)^2 + \left(\frac{\psi_1-\psi_2}{2\pi}\right)^2+\sum_{i=1}^{3}\left(\frac{\varphi_{i1}-\varphi_{i2}}{2\pi}\right)^2\right]^{1/2},$$

and the decay rate γ_c can be found from the equation

$$D=D_0\exp(\gamma_c\tau).$$

The decay rate γ_c increases with λ; for instance, $\gamma_c=0.014$ at $\lambda=0.6$, $\gamma_c=0.029$ at $\lambda=1$, and $\gamma_c=0.183$ at $\lambda=2.1$.

Note that different nonresonance terms of the Hamiltonian of the interaction of the electromagnetic field with the system of coherent excitons and biexcitons are responsible for the breakdown of different integrals of motion. If the detuning from resonance Δ is nonzero, there can also be a breakdown of the integrals of motion, but at larger values of λ.

In conclusion we give numerical estimates of λ for the

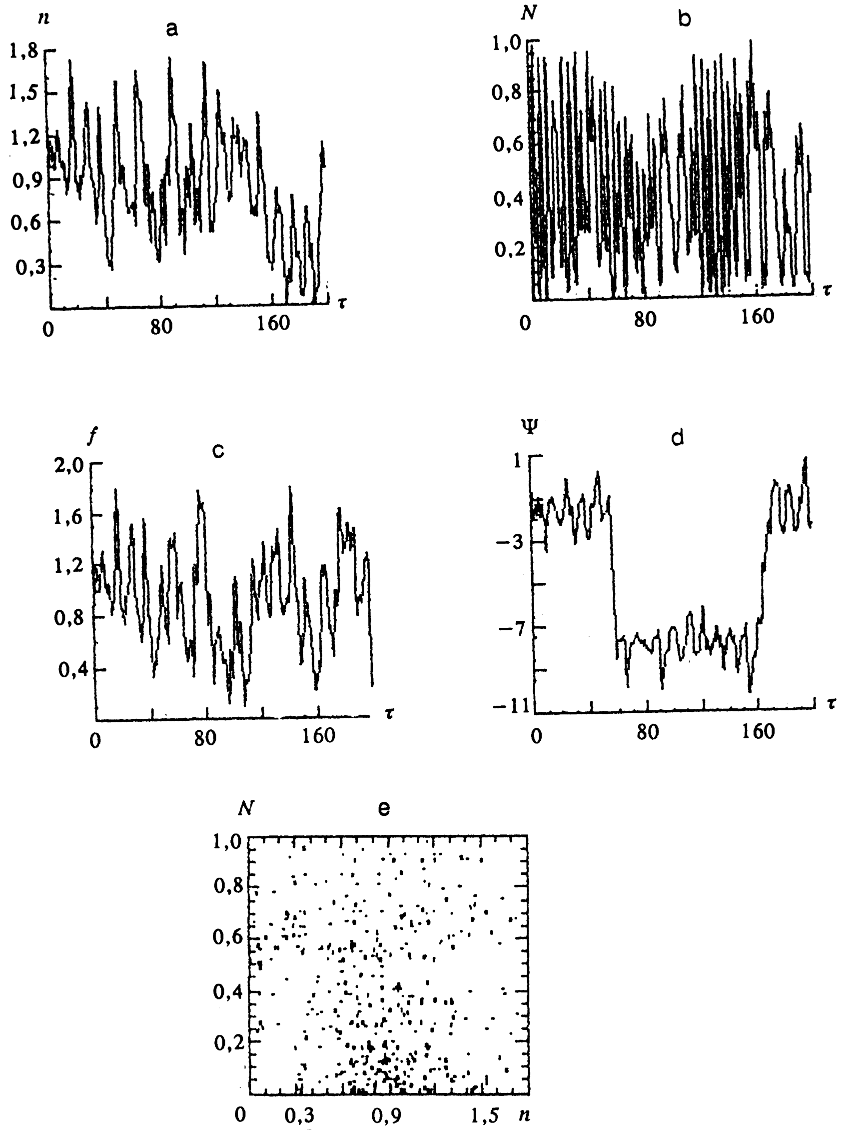

FIG. 6. Expanded stochastic mode of oscillations of (a) coherent excitons, (b) biexcitons, (c) photons, and of (d) the phase, and (e) the phase portrait in the (n,N) plane at $n_0=f_0=1$, $N_0=10^{-8}$, $\varphi_{a0}=\varphi_{ph}=1.571$, $\varphi_{b0}=10^{-8}$, and $\lambda=2$.

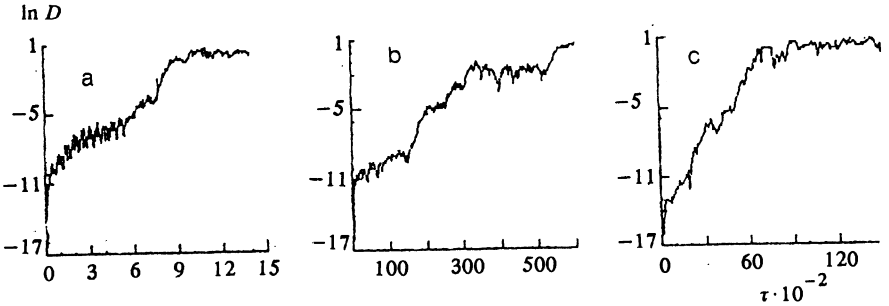

FIG. 7. The time dependence of the distance between initially close trajectories at $n_0=f_0=1$, $N_0=10^{-8}$, $\varphi_{a0}=\varphi_{ph}=1.571$, and $\varphi_{b0}=10^{-8}$ for (a) $\lambda=0.6$, (b) $\lambda=1$, and (c) $\lambda=2.1$.

CuCl crystal. At $\sigma=1.25\times10^{-8}$ cm$^{3/2}$, $g=5\times10^{13}$ s^{-1}, $\omega_{ph}=4\times10^{15}$ s^{-1}, and $n=5\times10^{16}$ cm^{-3} we get $\lambda = 8\sigma g\sqrt{n_0}/\omega_{ph} = 0.35$. For $n_0\geqslant5\times10^{17}$ cm^{-3} we get $\lambda\geqslant1$. Thus, for exciton concentrations of the order of 10^{17} cm^{-3}, ultrashort optical dynamic chaos may manifest itself in the system of excitons, photons, and biexcitons in crystals of the CuCl type in the event of exciton–biexciton conversion.

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