Pis'ma v ZhETF, vol. 75, iss. 4, pp. 206 – 210 © 2002 February 25

Optical chaos in nonlinear photonic crystals

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Submitted 9 January 2001

We examine the spatial evolution of lightwaves in a nonlinear photonic crystal with a quadratic nonlinearity when simultaneously a second harmonic and a sum-frequency generation are quasi-phase-matched. We find the conditions for a transition to Hamiltonian chaos for different amplitudes of lightwaves at the boundary of the crystal.

PACS: 05.45.Ac, 42.65.–k, 42.70.Mp

Wave-mixing in nonlinear optical materials is a basis of modern optical sciences and technologies. Cascading several wave-mixing processes in the same low-loss material one can in principle achieve a high efficiency using a large value of the lowest-order optical nonlinearity. The theoretical investigations of cascading of several scalar optical three-wave-mixing processes in the bulk materials with $\chi^{(2)}$ nonlinearity has a long history [1]. In particular, Akhmanov and co-workers have found the efficiency of third harmonic generation (THG) via cascading of a second harmonic generation (SHG) and a sum-frequency mixing (SFM) in a quadratic medium [2], while Komissarova and Sukhorukov have described an efficient parametric amplification at a high-frequency pump in the same system [3]. Obviously the observation of these nonlinear effects demands a simultaneous satisfaction of phase-matching conditions for several parametric processes as perfectly as possible. On other hand, it has been shown later that the systems, for which several wave-mixing processes can be simultaneously phase-matched, are in general nonintegrable; therefore a competition of two (or more) parametric processes can often result in a chaotic spatial evolution of lightwaves [4, 5]. However, until nowadays, it was unclear how to achieve a phase-matching for several processes in *homogeneous medium* employing traditional techniques, such as using a birefringence in ferroelectric crystals.

The solution of this problem has been found rather recently [6 – 8]; it consists in an introduction of the different types of *artificial periodicity* of a nonlinear medium, which results in a formation of nonlinear 1D and 2D superstructures termed *optical superlattices* [9] or *nonlinear photonic crystals* (NPCs) [10]. In NPCs there is a periodic (or quasiperiodic) spatial variation of a nonlinear susceptibility tensor while a linear susceptibility tensor is constant.

In these engineering nonlinear materials a phase mismatch between the interacting lightwaves could be compensated by the Bragg vector of NPC. The idea of such kind of *quasi-phase-matching* (QPM) was introduced by Bloembergen and co-workers many years ago [11]. However, only recently the rapid progress in fabrication of high quality ferroelectric crystals with a periodic domain inversion made the QPM method very popular [9, 12]. We should stress that the conditions for QPM may be fulfilled for several wave-mixing processes simultaneously; the QPM also has an advantage as using of largest nonlinear coefficient.

Nowadays there are several experiments on an observation of third and fourth harmonics in different periodically or quasiperiodically poled ferroelectric crystals with $\chi^{(2)}$ nonlinearity [7, 13, 14], which clearly demonstrate an importance of multiple-mixing in NPCs for the potential applications. Modern theoretical activities on the nonlinear lightwaves interactions in NPCs are mainly focused on the studies of strong energy interchange between the waves [12] (this is a development of the earlier activities [2, 3]), as well as on the formation of spatial optical solitons [15].

In this work we describe a novel for the physics of NPCs effect of Hamiltonian optical chaos. Namely, we show that spatial evolution of three light waves participating simultaneously in SHG and SFM in the conditions of QPM is chaotic for many values of complex amplitude

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of the waves at the boundary of $\chi^{(2)}$-NPC. There also exists an integrable limit when the evolution of waves is always regular regardless of absolute values of their complex amplitudes. The integrable limit corresponds to the particular values of two combinations of wave phases at the boundary of nonlinear medium. In particular, the problem of THG belongs to the integrable limit, therefore in the conditions of recent experiments [7, 13, 14], nonlinear light dynamics should be always regular. However, even a rather small change in amplitudes and phases of waves at the boundary of crystal, in respect to those considered in [7, 13, 14], should result in a transition to chaos.

We consider a spatial evolution of three co-propagating plane waves

$$E = \frac{1}{2}\sum_{j=1}^{3} A_j \exp\left[i(j\omega t - k_j z)\right] + \text{c.c.}, \quad k_j = k(j\omega)$$

in a periodically poled crystal in the conditions when simultaneously SHG, $\omega+\omega \to 2\omega$, and SFM, $\omega+2\omega \to 3\omega$ take place. Equations of motion for the slowly varying complex amplitudes A_l ($l = 1, 2, 3$) of the waves are [12, 9]

$$\begin{aligned} \frac{dA_1}{dz} &= -i\beta_3 g(z) A_3 A_2^* e^{-i\Delta k_3 z} - i\beta_2 g(z) A_2 A_1^* e^{-i\Delta k_2 z}, \\ \frac{dA_2}{dz} &= -i2\beta_3 g(z) A_3 A_1^* e^{-i\Delta k_3 z} - i\beta_2 g(z) A_1^2 e^{i\Delta k_2 z}, \quad (1) \\ \frac{dA_3}{dz} &= -i3\beta_3 g(z) A_1 A_2 e^{i\Delta k_3 z}, \end{aligned}$$

where $g(z)$ is a function that equals to $+1$ (or -1) in a single positive (negative) polarization domain of the ferroelectric crystal. In this work for the sake of simplicity we consider only a periodic alternative domain superlattice with a spatial period Λ. However, $g(z)$ can be a quasiperiodic function in the case of nonlinear quasicrystals [8, 9]. Note that we consider a typical situation $\lambda \ll \Lambda$, where λ is a wavelength [9, 12, 14].

The coupling constants between waves, β_2 and β_3, are defined as

$$\beta_{2,3} = \omega d_{\text{eff}}/cn_{2,3},$$

where $d_{\text{eff}} = 2\pi\chi^{(2)}$ and $n_j \equiv n(j\omega)$ ($j = 1, 2, 3$) are the refractive indexes for the different waves. Of course $n_1 \neq n_2 \neq n_3$ due to a light dispersion. However, it may be shown that $\Delta n/n \simeq \lambda/\Lambda \ll 1$ in the conditions of QPM, therefore in what follows we will take $\beta_2 = \beta_3 \equiv \beta$. Finally, the phase mismatches involved in Eqs. (1) are $\Delta k_2 = k_2 - 2k_1$ and $\Delta k_3 = k_3 - k_2 - k_1$. Let both these mismatches be compensated by a reciprocal lattice vector of NPC, that is

$$\Delta k_2 = 2\pi m_1/\Lambda, \quad \Delta k_3 = 2\pi m_2/\Lambda, \qquad (2)$$

where $m_j = \pm1, \pm3, \pm5, \ldots$ The methods of achievement of QPM for several parametric processes in a single NPC have been recently discussed in Refs. [6, 8, 10] (theory) and Refs. [7, 13, 14] (experiment).

The dynamical system (1) together with the initial conditions, which in our case are the values of complex amplitudes at the boundary of NPC, $A_j(z = 0)$, completely determine the nonlinear spatial evolution of waves. Before specification of these initial conditions, we can further simplify the equations of motion. First, we introduce new scaled amplitudes $a_l = A_l/\sqrt{l}A_0$, where $l = 1, 2, 3$ and $A_0 \equiv \max(|A_1(0)|, |A_2(0)|, |A_3(0)|)$. Second, we make the Fourier expansion of the function $g(z)$

$$g(z) = \sum_{n=1}^{\infty} \frac{4}{\pi n} \sin\left(\frac{2\pi n z}{\Lambda}\right),$$

where index n takes only odd values. Now we substitute this expansion into Eqs. (1), take into account the QPM conditions (2) and make an averaging of resulting motion equations over "the short characteristic spatial scale" $2\pi/\Lambda$. We have the following basic equations

$$\begin{aligned} \dot{a}_1 &= -a_2 a_1^* - \xi a_3 a_2^*, \\ \dot{a}_2 &= 0.5 a_1^2 - \xi a_3 a_1^*, \\ \dot{a}_3 &= \xi a_1 a_2, \end{aligned} \qquad (3)$$

where $\xi = \sqrt{3} m_2/m_3$ (m_j are the quasi-phase matching orders, see Eq. (2); we assume that $m_3 \geq m_2$). Overdot in Eqs. (3) means the derivative in respect to z/l_{nl} with a characteristic nonlinear length l_{nl}, defined as

$$l_{nl} = \frac{\pi m_2}{2\sqrt{2}}\frac{1}{\beta A_0}. \qquad (4)$$

In the derivation of motion equations (3) we removed all fastly varying terms performing averaging over $2\pi/\Lambda$. It can be shown that such a procedure is correct if $l_{nl} \gg \Lambda$ [16].

The equations (3) can be presented in the canonical form with the Hamiltonian function

$$H = \left[-i\left(\xi a_1^* a_2^* a_3 + \frac{1}{2} a_1^{*2} a_2\right)\right] + \text{c.c.}, \qquad (5)$$

$$i\dot{a}_l = \frac{\partial H}{\partial a_l^*}, \quad i\dot{a}_l^* = -\frac{\partial H}{\partial a_l}.$$

Additionally to the energy of wave interaction $E \equiv H$, (Eq. (5)), the dynamical system (3) has the integral of motion

$$|a_1|^2 + 2|a_2|^2 + 3|a_3|^2 = 1 \qquad (6)$$

corresponding to the conservation of energy of noninteracting waves. In a general case the system (3) does not have other global integrals of motion, thus it is *non-integrable* and should demonstrate *chaotic dynamics* for many initial conditions $a_l(0)$ [17, 18]. However, for some values of ξ and some specific initial conditions an additional local integral of motion can arise. Let us list these cases because they include the physically important situations.

First, if one of the parametric processes, either SHG or SFM, is dominant ($\xi \ll 1$ or $\xi \gg 1$), then an additional integral of motion arises, which is of the Manley-Rowe type [12]. Second, nonlinear dynamics is strongly dependent on the initial values of two "resonant phases" $\psi_2(0)$ and $\psi_3(0)$, where

$$\psi_2 = 2\theta_1 - \theta_2, \quad \psi_3 = \theta_1 + \theta_2 - \theta_3 \qquad (7)$$

and θ_j ($j = 1, 2, 3$) are the lightwave phases, i.e. $a_j = |a_j| \exp(-i\theta_j)$. We found that *for* $\psi_2(0) = \psi_3(0) = 0$ *dynamics is always regular*. Moreover, using approaches of Refs. [3, 19], it is possible to show that an additional local motion integral exists in this case [20]. In particular, the problem of THG ($a_1(0) = 1$, $a_2(0) = a_3(0) = 0$) belongs to this class of initial conditions. Therefore, the spatial dynamics of lightwaves at THG is regular (cf Ref. [21], where an analytic solution has been found).

We perform an intensive search of chaotic trajectories solving the equations of motion (3) numerically for two characteristic values of control parameter ξ that correspond to the experimental situations described in works [7] and [13], correspondingly:

Set I: The QPMs of first order for both processes, $m_1 = m_3 = 1$, $\xi = \sqrt{3} \approx 1.73$;

Set II: The QPMs of ninth and 33d orders, $m_1 = 9$, $m_3 = 33$, $\xi = 3\sqrt{3}/11 \approx 0.472$.

We consider several types of initial conditions, which cover practically all physically interesting cases (note that all these initial conditions satisfy to the restriction arising from the integral of motion (6)):

Problem 1: $a_1(0) = \alpha$, $a_2(0) = \left[1 - \alpha^2\right]^{1/2} \cdot 2^{-1/2} \exp(-i\phi)$, $a_3(0) = 0$, where the real parameters ϕ and α vary in the ranges $-\pi \le \phi < \pi$ and $0 \le \alpha \le 1$, correspondingly. Obviously here $|\psi_2(0)| = |\psi_3(0)| = |\phi|$.

Problem 2: $a_1(0) = \left[1 - 3\alpha^2\right]^{1/2} 3^{-1/2} \exp(-i\theta_1)$, $a_2(0) = \left[1 - 3\alpha^2\right]^{1/2} 3^{-1/2} \exp(-i\theta_2)$, $a_3(0) = \alpha \exp(-i\theta_3)$, where $-\pi \le \theta_j < \pi$ ($j = 1, 2, 3$) and $0 \le \alpha \le 3^{-1/2} \approx 0.57735$.

Problem 3: $a_1(0) = \alpha \exp(-i\theta_1)$, $a_2(0) = 0$, $a_3(0) = \left[1 - \alpha^2\right]^{1/2} 3^{-1/2} \exp(-i\theta_3)$, $-\pi \le \theta_j < \pi$ ($j = 1, 3$) and $0 \le \alpha \le 1$.

We start our analysis with problem 1. This set of initial conditions describes, in particular, the THG at $\alpha = 1$ ($\phi = 0$) and the parametric amplification with a low-frequency pump at $\alpha \ll 1$ [12]. In order to increase the efficiency of energy transformation from a basic wave of frequency ω to a wave of frequency 3ω, it was suggested recently to mix some nonzero signal at the frequency 2ω with a basic beam [22]. Such kind of initial conditions correspond to $\alpha \to 1$ (but $\alpha \ne 1$) with different values of phase ϕ.

To distinguish regular and chaotic dynamics we compute the maximal Lyapunov exponent $\lambda_{\max}$ for the different values of initial lightwave's amplitudes, α, and phases, ϕ. For chaos $\lambda_{\max} > 0$, in contrast $\lambda_{\max} = 0$ for a regular motion [18]. The dependence of $\lambda_{\max}$ on α for the first order QPMs (set I) is depicted in Fig.1. For $\phi = 0$ the initial values of resonant phases, $\psi_2(0)$ and $\psi_3(0)$, are zero corresponding to the integrable limit with $\lambda_{\max} = 0$ independently on the value of α (not shown in Fig.1). However, even a small deviation from the integrable limit, $|\psi_2(0)| = |\psi_3(0)| = |\phi| = 0.01$, results in a chaotic motion for a quite wide range of initial conditions (dashed line). Further increase in the value of $|\phi|$ makes chaos more strong (dotted line, $|\phi| = 0.1$); the most strong chaos arises for $|\phi| = \pi/2$ (solid line) corresponding to the initial values of resonant phases $|\psi_{2,3}(0)|$ most distant from the integrable limit.

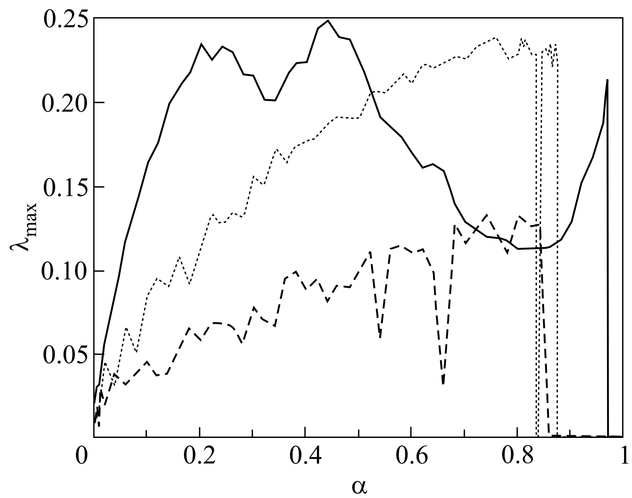

Fig.1. Dependence of the value of maximal Lyapunov exponent on the amplitude of first wave at the boundary of optical superlattice α and for the different phases: $\phi = -\pi/2$ (solid line), $\phi = -0.1$ (dotted line) and $\phi = -0.01$ (dashed line). The first order QPMs (problem 1, set I)

The motion is always regular for the standard THG ($\alpha = 1$), as well as for some range of α in the vicinity of $\alpha = 1$ (see the right hand side of Fig.1). A regular spatial evolution of lightwaves for $\alpha = 1$ is shown in the upper subplot of Fig.2. However, for $|\phi| = \pi/2$ a strong chaos exists already for $\alpha \approx 0.95$, i.e. for $a_1(0) = 0.95$, $a_2(0) \approx 0.22i$, $a_3(0) = 0$, see lower subplot of Fig.2. Thus the possibility of transition to chaos must be taken into account in the application of an additional pump of

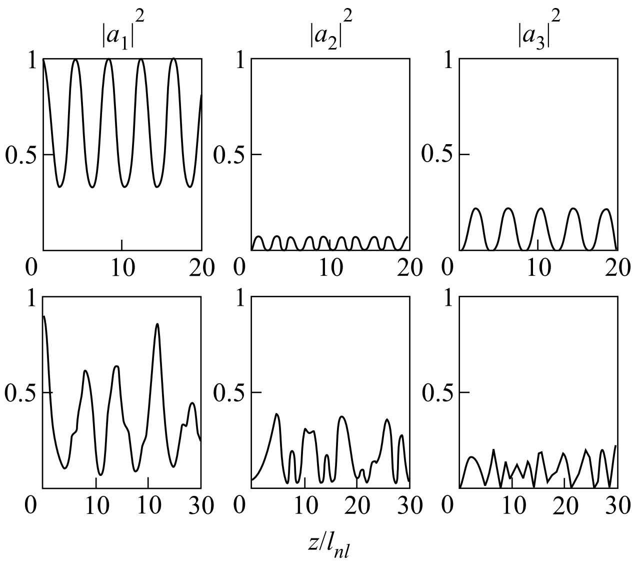

Fig.2. Regular (upper) and chaotic (lower) spatial evolutions of scaled intensities of lightwaves at the first order QPMs. For the upper subplot $\alpha = 1$ and $\phi = 0$, while for the lower subplot $\alpha = 0.95$ and $\phi = \pi/2$

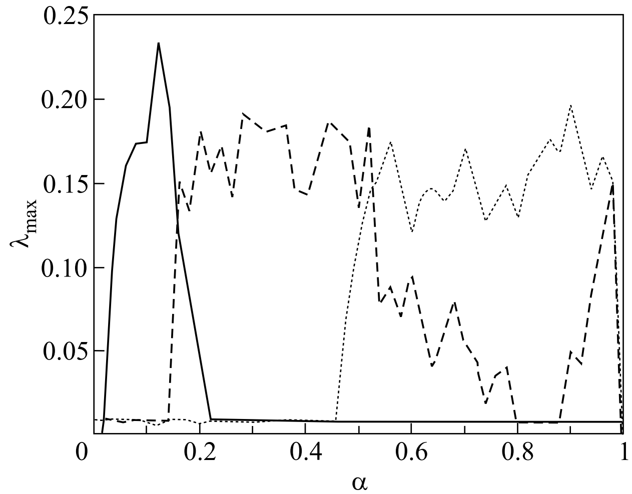

Fig.3. Same as in Fig.1 but for the high order QPMs (problem 1, set II): $\phi = -\pi/2$ (solid line), $\phi = -0.1$ (dashed line) and $\phi = -0.01$ (dotted line)

frequency 2ω in order to increase an efficiency of THG [22].

We consider now the situation corresponding to the left hand side of Fig.1 with $\alpha \ll 1$. This is the parametric amplification with a low-frequency pump [12]. In this case our analysis demonstrates that the evolution of waves is weakly chaotic for $|\psi_{2,3}(0)|$ distant from the integrable limit. In this regime, the Lyapunov exponent has some very small but yet positive value, therefore it is very difficult to distinguish a weak chaos from a regular motion. In practical terms it means that one needs to have a very long sample to see the differences between regular and weakly chaotic spatial evolutions of light waves.

Now we turn to the consideration of nonlinear dynamics using the second set of QPM parameters but same set of initial conditions (set II, problem 1). Main results on the transition to chaos are depicted in Fig.3. Again, as in Fig.1, $|\psi_2(0)| = |\psi_3(0)| = |\phi| = 0$ results in a regular motion, while a motion is chaotic for many initial conditions if $|\phi| > 0$. However the absolute values of Lyapunov exponent are small: Really, $\max \lambda_{\max} \simeq 0.1$ in Fig.1, but $\max \lambda_{\max} \simeq 0.01$ in Fig.3. Therefore we conclude that multiple interaction of waves employing high order QPMs is more stable against a transition to chaos in comparison with the case of first order QPMs.

We consider now a nonlinear dynamics in the case when some portion of energy is presented at $z = 0$ in each of interacting waves (Problem 2). We present our findings in Fig.4. A strong chaos arises as soon as one of the resonant phases becomes different from the integrable limit $|\psi_{2,3}(0)| = 0$ ($|\psi_2(0)| = \pi$ and $|\psi_3(0)| = \pi/2$ for a solid line, $|\psi_2(0)| = \pi/2$ and $|\psi_3(0)| = 0$ for a dashed line). We should note that for the parameters corresponding to a solid curve in Fig.4 strong chaos exists for almost all values of initial wave amplitudes α. Chaos is sufficiently weaker for the high order QPMs in comparison with the case of first order QPMs: *cf* a dashed line with a dashed and dotted line that correspond to the same values of phases θ_j but to the different sets of QPM-parameters.

Finally, we analyze the set of initial conditions termed as Problem 3. In particular, it includes the down conversion [3, 12] or, in other words, the fractional conversion $\omega \to (2/3)\omega$ [19] in the case $\alpha \ll 1$. For this set of initial conditions we did not find visible regions of chaotic dynamics.

In order to reliably distinguish regular and chaotic spatial evolutions of lightwaves in conditions of an experiment, one needs to have many enough characteristic nonlinear lengths, l_{nl}, on the total length of the crystal L: $L/l_{nl} \gtrsim 10$ [4, 5]. Importantly, it appears possible to meet this condition in the typical NPCs. Really, for a periodically poled lithium niobate with a period $\Lambda = 30\,\mu$m, a crystal length $L \simeq 1$ cm, a nonlinear coefficient $d_{33} = 34$ pm/V [7, 13] and a light intensity $A_0^2 = 0.76$ GW/cm^2 ($\lambda = 1.064\ \mu$m) [23], we have $L/l_{nl} \simeq 100$. Moreover, chaos should be more easier observable in the GaAs optical superlattice with $d_{14} \geq 90$ pm/V [24].

In summary, we have shown that a simultaneous multiwavelength generation in the typical nonlinear photonic crystals is often chaotic. This fact must be taken into

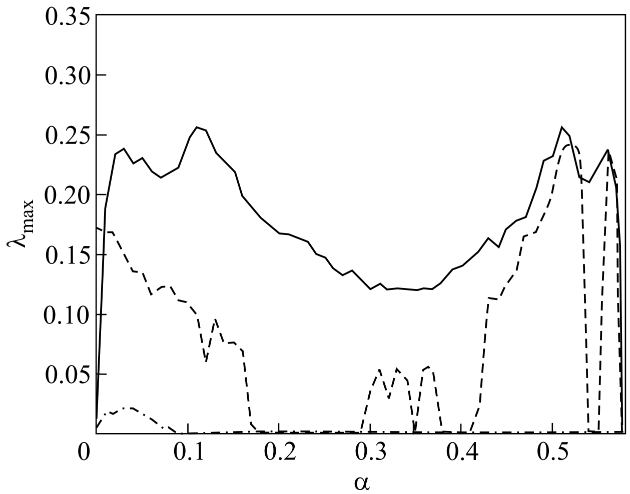

Fig.4. Dependence of the value of maximal Lyapunov exponent on the amplitude of third wave at the boundary of optical superlattice, α, and for the different phases and the QPM orders (problem 2, sets I and II): $\theta_1 = \pi/2$, $\theta_2 = 0$, $\theta_3 = \pi$, first order QPMs (solid line); $\theta_1 = \theta_2 = \theta_3 = -\pi/2$, first order QPMs (dashed line); $\theta_1 = \pi/2$, $\theta_2 = 0$, $\theta_3 = \pi$, high order QPMs (dashed and dotted line)

an account in realization of compact laser multicolor sources for printers, scanners, and color displays based on the quasi-phase-matched harmonics generation.

We should distinguish our results from a recent paper [25], where nonlinear spatial field dynamics and chaos have been studied in a quadratic media with a periodic Bragg grating.

We thank Andreas Buchleitner for discussions and support, Martin Fejer for useful comments, and Pekka Pietiläinen for critical reading of the manuscript. KNA is also greatful to co-authors of his works [4, 5] for a creative cooperation. This research was partially supported by the Academy of Finland (grants # 163358 and # 100487) and the MPI PKS.

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