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Optical Chaos in Nonlinear Photonic Crystals[1]

K. N. Alekseev[1, 2, 3, *] and A. V. Ponomarev[3, 4]

[1] Department of Physical Sciences, University of Oulu FIN-90014, Finland

*e-mail: Kirill.Alekseev@oulu.fi

[2] Max-Planck-Institute für Physik komplexer Systeme, D-01187 Dresden, Germany

[3] Theory of Nonlinear Processes Laboratory, Kirensky Institute of Physics, Russian Academy of Sciences, Krasnoyarsk, 660036 Russia

[4] Department of Physics, Krasnoyarsk State University, Krasnoyarsk, 660041 Russia

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We examine the spatial evolution of lightwaves in a nonlinear photonic crystal with a quadratic nonlinearity, when a second harmonic and a sum-frequency generation are simultaneously quasi-phase-matched. We find the conditions for a transition to Hamiltonian chaos for different amplitudes of lightwaves at the crystal boundary. *© 2002 MAIK "Nauka/Interperiodica".*

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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 bulk materials with $\chi^{(2)}$nonlinearity has a long history [1]. In particular, Akhmanov and coworkers 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 described an efficient parametric amplification at a high-frequency pump in the same system [3]. Obviously, the observation of these nonlinear effects demands the simultaneous fulfillment of phase-matching conditions for several parametric processes as perfectly as possible. On the other hand, it was shown later that the systems for which several wave-mixing processes can be simultaneously phase-matched are in general nonintegrable; therefore, the competition of two (or more) parametric processes can often result in the chaotic spatial evolution of lightwaves [4, 5]. However, until recently, it was unclear as to how one can achieve phase-matching for several processes in a *homogeneous medium* employing traditional techniques, such as the use of birefringence in ferroelectric crystals.

The solution of this problem has been found rather recently [6–8]; it consists in the introduction of different types of *artificial periodicity* of a nonlinear medium, which results in the 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 the nonlinear susceptibility tensor, while the linear susceptibility tensor is constant.

In these engineered nonlinear materials, a phase mismatch between the interacting lightwaves could be compensated by the Bragg vector of NPC. The idea of this kind of *quasi-phase-matching* (QPM) was introduced by Bloembergen and coworkers many years ago [11]. However, only recently, the rapid progress in the fabrication of high-quality ferroelectric crystals with a periodic domain inversion has 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 of using the largest nonlinear coefficient.

Nowadays, there are several experiments on the observation of third and fourth harmonics in different periodically or quasiperiodically poled ferroelectric crystals with $\chi^{(2)}$ nonlinearity [7, 13, 14], which clearly demonstrate the importance of multiple mixing in NPCs for 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 the effect of Hamiltonian optical chaos novel for the physics of NPCs. Namely, we show that spatial evolution of three light waves participating simultaneously in SHG and SFM under the conditions of QPM is chaotic for many values of the complex amplitude of the waves at the boundary of $\chi^{(2)}$-NPC. There also exists an integrable limit, where the evolution of waves is always regular regardless of the absolute values of their complex amplitudes. The inte-

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grable 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, under the conditions of recent experiments [7, 13, 14], nonlinear light dynamics should always be regular. However, even a rather small change in amplitudes and phases of waves at the boundary of crystal, with respect to those considered in [7, 13, 14], should result in a transition to chaos.

We consider a spatial evolution of three copropagating plane waves

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

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

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

where $g(z)$ is a function equal 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

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

where $d_{\text{eff}} = 2\pi\chi^{(2)}$ and $n_j \equiv n(j\omega)$ ($j = 1, 2, 3$) are the refractive indices for the different waves. Of course, $n_1 \neq n_2 \neq n_3$ because of light dispersion. However, it can be shown that $\Delta n/n \simeq \lambda/\Lambda \ll 1$ under 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

$$\Delta k_2 = 2\pi m_1/\Lambda, \quad \Delta k_3 = 2\pi m_2/\Lambda, \tag{2}$$

where $m_j = \pm1, \pm2, \pm5, \ldots$. The methods of achieving QPM for several parametric processes in a single NPC were recently discussed in [6, 8, 10] (theory) and [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 series 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 the resulting equations of motion over the short characteristic spatial scale $2\pi/\Lambda$. We have the following basic equations

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

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

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

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

Equations (3) can be represented in the canonical form with the Hamiltonian function

$$\begin{aligned}H &= \left[-i\left(\xi a_1^* a_2^* a_3 + \frac{1}{2} a_1^{*2} a_2\right)\right] + \text{c.c.},\\ i\dot{a}_l &= \frac{\partial H}{\partial a_l^*}, \quad i\dot{a}_l^* = -\frac{\partial H}{\partial a_l}.\end{aligned} \tag{5}$$

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

$$|a_1|^2 + 2|a_2|^2 + 3|a_3|^2 = 1 \tag{6}$$

corresponding to the conservation of energy of noninteracting waves. In the general case, the system (3) does not have other global integrals of motion; thus, it is *nonintegrable* 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

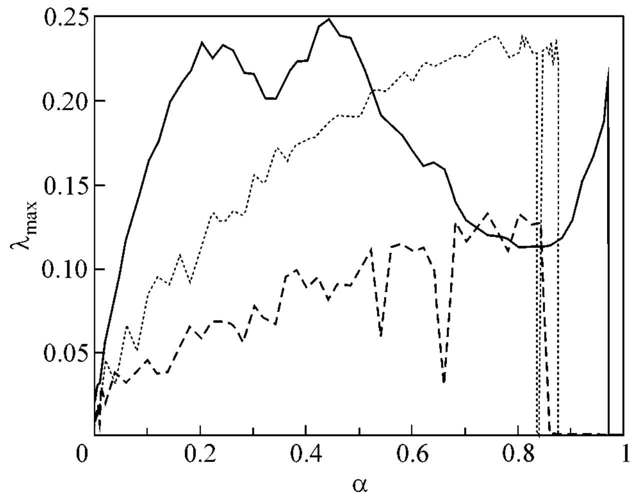

Fig. 1. Dependence of the value of the maximal Lyapunov exponent on the amplitude of the first wave at the boundary of optical superlattice α and for different phases: $\phi = -\pi/2$ (solid line), $\phi = -0.1$ (dotted line), and $\phi = -0.01$ (dashed line). The first-order QPMs (problem 1, set I).

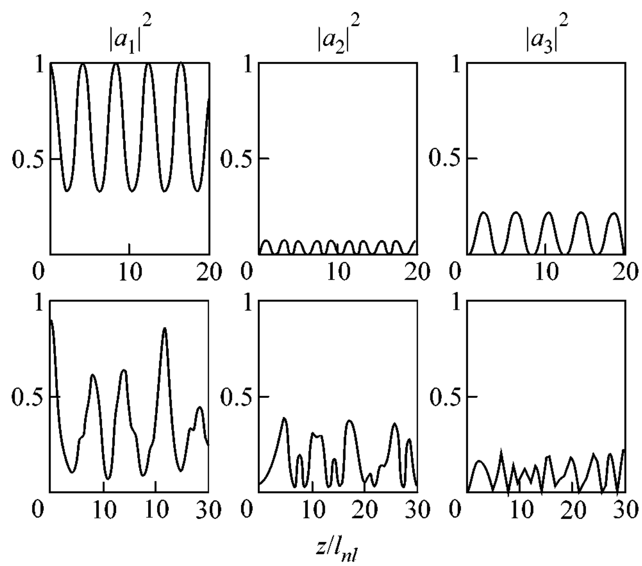

Fig. 2. Regular (upper) and chaotic (lower) spatial evolutions of scaled intensities of lightwaves at the first-order QPMs. For the upper subplot, $\alpha = 1$ and $\phi = 0$, while for the lower subplot, $\phi = 0.95$ and $\phi = \pi/2$.

list these cases, because they include 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, the nonlinear dynamics strongly depends on the initial values of the 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 $\psi_2(0) = \psi_3(0) = 0$, *the dynamics is always regular*. Moreover, using approaches [3, 19], it is possible to show that an additional local integral of motion 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. [21], where an analytic solution has been found).

We performed 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 [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 the 9th and 33rd 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 the restriction arising from the integral of motion (6)]:

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

Problem 3: $a_1(0) = \alpha\exp(-i\theta_1)$, $a_2(0) = 0$, $a_3(0) = [1 - \alpha^2]^{1/2} \times 3^{-1/2}\exp(-i\theta_3)$, $-\pi \le \theta_j < \pi$ ($j = 1, 3$) 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 that some nonzero signal at the frequency 2ω be mixed with a basic beam [22]. This kind of initial condition corresponds to $\alpha \longrightarrow 1$ (but $\alpha \ne 1$) with different values of phase ϕ.

To distinguish between regular and chaotic dynamics, we compute the maximal Lyapunov exponent λ_{max} for different values of initial lightwave amplitudes, α, and phases, ϕ. For chaos $\lambda_{max} > 0$, in contrast $\lambda_{max} = 0$ for a regular motion [18]. The dependence of λ_{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 chaotic motion for a quite wide range of initial conditions (dashed line). A further increase in the value of $|\phi|$ makes chaos more strong (dotted line, $|\phi| = 0.1$); the strongest chaos arises for $|\phi| = \pi/2$ (solid line), corresponding to the initial values of resonant phases $|\psi_{2,3}(0)|$ that are most distant from the integrable limit.

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 side of Fig. 1). A regular spatial evolution of lightwaves for $\alpha = 1$ is shown in the upper subplot in Fig. 2. However, for $|\phi| = \pi/2$, strong chaos exists already for $\alpha \approx 0.95$, i.e., for $a_1(0) = 0.95$, $a_2(0) \approx 0.22i$, and $a_3(0) = 0$; see lower subplot in Fig. 2. Thus, the possibility of transition to chaos must be taken into account in the application of an additional pump of frequency 2ω in order to increase the efficiency of THG [22].

We now consider the situation corresponding to the left 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 yet positive value; therefore, it is very difficult to distinguish between weak chaos and 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.

We now turn to the consideration of nonlinear dynamics using the second set of QPM parameters but the same set of initial conditions (set II, problem 1). The 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 motion is chaotic for many initial conditions if $|\phi| > 0$. However, the absolute values of the Lyapunov exponent are small: really, $\lambda_{max} \simeq 0.1$ in Fig. 1, but $\lambda_{max} \simeq 0.01$ in Fig. 3. Therefore, we conclude that the 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 now consider a nonlinear dynamics in the case when some portion of the energy is presented at $z = 0$ in each of the interacting waves (Problem 2). We present our findings in Fig. 4. 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 the 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.

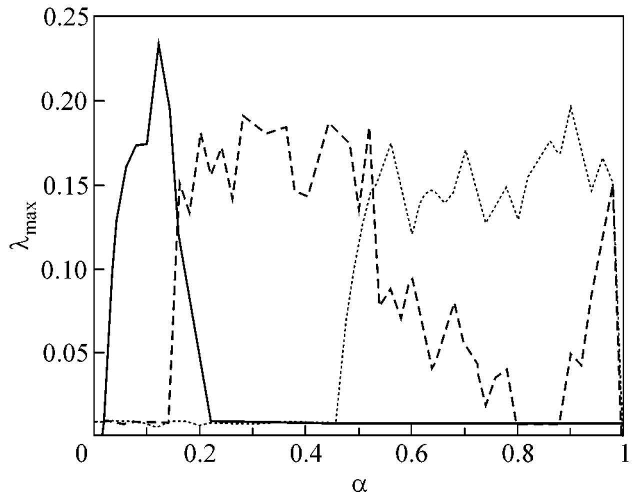

Fig. 3. The same as in Fig. 1 but for the high-order QPMs (problem 1, set II): $\phi = -\pi/2$ (solid line), $\phi = -0.1$ (dashed line), and $\phi = -0.01$ (dotted line).

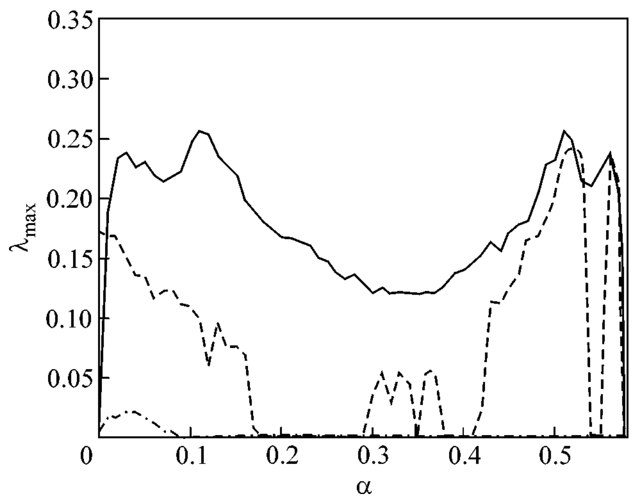

Fig. 4. Dependence of the value of the maximal Lyapunov exponent on the amplitude of the third wave at the boundary of optical superlattice α and for different phases and QPM orders (problem 2, sets I and II): $\theta_1 = \pi/2$, $\theta_2 = 0$, $\theta_3 = \pi$, first order QPMs (solid line); $\theta_1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

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 $\omega \longrightarrow (2/3)\omega$ [19] in the case of $\alpha \ll 1$. For this set of initial conditions, we did not find visible regions of chaotic dynamics.

In order to reliably distinguish between regular and chaotic spatial evolutions of lightwaves in conditions of an experiment, one needs to have many 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. In actuality, for a periodically poled lithium niobate with a period Λ = 30 μm, 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 (λ = 1.064 μm) [23], we have $L/l_{nl} \simeq 100$. Moreover, chaos should be more easily observable in the GaAs optical superlattice with $d_{14} \geq 90$ pm/V [24].

In summary, we have shown that simultaneous multiwavelength generation in typical nonlinear photonic crystals is often chaotic. This fact must be taken into an account for the realization of compact laser multicolor sources for printers, scanners, and color displays based on quasi-phase-matched harmonics generation.

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

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