Quantum Electronics **38** (3) 258 – 262 (2008)

ULTRASHORT LIGHT PULSES

PACS numbers: 42.65.Ky;42.65.Re; 42.70.Mp
DOI: 10.1070/QE2008v038n03ABEH013700

Formation of subfemtosecond laser pulses in aperiodically poled nonlinear-optical crystals

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***Abstract*. The method of synthesis of ultrashort laser pulses in nonlinear aperiodically poled crystals based on the simultaneous generation of several higher optical harmonics is considered. The interaction of four waves with multiple frequencies involving three mutually coupled nonlinear three-frequency processes is studied. It is shown that by introducing intense laser radiation into a crystal, pulses of duration of the order of a few hundreds of attoseconds can be produced at the crystal output.**

Keywords: *quasi-phase-matched interactions, aperiodic variation of nonlinearity, coupled processes, multifrequency interactions, synthesis of ultrashort pulses.*

1. Introduction

The possibility of generating few-cycle laser pulses of duration $10^{-15} - 10^{-17}$ s is being extensively studied at present (see, for example, [1]). The generation of such pulses is of interest for various fields in science, technology, and medicine. Modern methods proposed for generating few-cycle pulses are mainly reduced to the problem of phasing equidistant high-order harmonics. This principle is conceptually close to the principle of mode-locking, which is often used in laser systems [2, 3]. In this case, the pulse duration is determined by the width of the spectrum and their repetition rate – by the intermode beat frequency.

The possibility of generating attosecond pulses by the summation of higher radiation harmonics generated in gas was pointed out in review [4]. Beats of attosecond duration upon excitation of two frequencies were observed in experiments with a sodium vapour [5]. A few-cycle light pulse was produced by generating SRS in gases upon excitation of five frequencies [6]. Modern theoretical methods of obtaining attosecond pulses were mainly developed for gases (see [6] and references therein and [7 – 9]).

It seems that Hänsh was the first to consider the use of the phasing of equidistant optical frequencies, obtained by frequency conversion in nonlinear-optical crystals, for generating subfemtosecond light pulses [10]. He discussed the synthesis of a femtosecond pulse by the superposition of six frequencies: the radiation frequencies from two lasers and the sum and difference frequencies excited by lasers in nonlinear crystals.

The aim of this paper is to call attention to the fact that several higher optical harmonics can be simultaneously efficiently generated in an aperiodically poled nonlinear-optical crystal in comparatively moderate laser fields. Therefore, subfemtosecond pulses can be synthesised by using one laser and only one nonlinear-optical crystal. This circumstance allows one to exclude a number of technical aspects which should be overcome in the method proposed in [10]. In particular, the phase-matching condition in our method is fulfilled automatically.

2. Wave interactions in an aperiodically poled crystal

It is known that to realise nonlinear-optical interactions of light waves in a crystal, the phase-matching conditions should be fulfilled. At present along with traditional phase-matching interactions, the quasi-phase-matching interactions are widely used [11] in which the wave detuning of the interacting waves is compensated by the reciprocal nonlinear lattice vector of a periodically poled crystal, providing thereby the efficient energy exchange between the waves. Quasi-phase-matching interactions allow one to use small nonlinear coefficients and realise nonlinear-optical processes when usual phase-matching conditions are not fulfilled. Moreover, in periodically poled crystals the so-called consecutive coupled processes can be performed, when quasi-phase-matching conditions are fulfilled simultaneously for two nonlinear-optical processes (at different quasi-phase-matching orders). However, the dispersion properties of periodically poled crystals allow one to realise in the collinear-interaction geometry simultaneously only two coupled three-frequency processes.

At the same time, several nonlinear-optical processes can be realised in crystals in which the nonlinear susceptibility coefficient changes aperiodically with distance [12]. This approach is based on the following principle. The factor characterising a periodic dependence of the nonlinear coupling coefficient of waves in a periodically poled crystal can be written in the form

$$g(z) = \operatorname{sign}\left(\sin\frac{2\pi z}{\Lambda}\right), \tag{1}$$

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Received 9 July 2007; revision received 31 August 2007
Kvantovaya Elektronika **38** (3) 258 – 262 (2008)
Translated by M.N. Sapozhnikov

where Λ is the modulation period of the nonlinear coefficient.

The method of producing an aperiodic structure in a crystal studied in [12] generalises relation (1) to the case of an arbitrary number of simultaneously proceeding nonlinear-optical processes:

$$g(z) = \mathrm{sign}\left(\sum_{j=1}^{N} a_j \sin\left(\frac{2\pi z}{\Lambda_j} + \varphi_j \right) \right), \tag{2}$$

where a_j and φ_j are the amplitude and phase of modulating harmonics, respectively; N is the number of three-frequency processes simultaneously proceeding in the crystal; $\Lambda_j = 2\pi/|\Delta k_j|$ is the period of the periodically poled crystal for realisation of the jth process with the wave detuning Δk_j. The analysis of (2) showed that the spectrum of the aperiodic function $g(z)$ contains the components $2\pi/\Lambda_j$ of the reciprocal lattice vector compensating the wave detunings for the jth process in the first quasi-phase-matching order. Note that a change in phases φ_j does not affect the nonlinear conversion efficiency, whereas the ratio of amplitudes a_j plays a considerable role. In fact, a_j determines the specific weight of the jth process. A change of the relation between amplitudes a_j is equivalent to a change in the efficiency of the jth nonlinear process with respect to other processes.

3. Coupled three-frequency interactions in a $LiNbO_3$ crystal

Consider a lithium niobate crystal with the aperiodically modulated nonlinear coupling coefficient of waves. (Such crystals can be produced by the after-growth polarisation switching method [13].) We will study the possibility of realisation of two types of coupled four-wave interactions in an aperiodically poled $LiNbO_3$ crystal.

The first type of interactions proceeding in the field of the intense pump wave with frequency ω (type I) consists of the processes

$$\omega \to \omega/2 + \omega/2, \tag{3.1}$$

$$\omega + \omega/2 \to 3\omega/2, \tag{3.2}$$

$$\omega/2 + 3/2\omega \to 2\omega. \tag{3.3}$$

Type I includes parametric frequency down-conversion (3.1) and two sum-frequency generation processes (3.2) and (3.3).

The second type of interactions (type II) includes the processes

$$\omega \to \omega/2 + \omega/2, \tag{4.1}$$

$$\omega + \omega/2 \to 3\omega/2, \tag{4.2}$$

$$\omega + \omega \to 2\omega. \tag{4.3}$$

One can see that, unlike the type I interaction, radiation at frequency 2ω is the result of the pump-wave frequency doubling rather than of frequency mixing. Below, we will use for convenience the notation: $\omega_1 = \omega$, $\omega_2 = \omega/2$, $\omega_3 = 3/2\omega$ and $\omega_4 = 2\omega$. The wave detunings for these processes are

$$\Delta k_1 = k_1 - 2k_2, \tag{5.1}$$

type I

$$\Delta k_2 = k_3 - k_2 - k_1, \tag{5.2}$$

$$\Delta k_3 = k_4 - k_2 - k_3; \tag{5.3}$$

$$\Delta k_1 = k_1 - 2k_2, \tag{6.1}$$

type II

$$\Delta k_2 = k_3 - k_2 - k_1, \tag{6.2}$$

$$\Delta k_3 = k_4 - 2k_1. \tag{6.3}$$

Here, k_j is the wave number for the wave at frequency ω_j. These mismatches determine the corresponding periods of the domain structure in the first quasi-sphase-matching order by the relation $\Lambda_j = 2\pi/|\Delta k_j|$.

We calculated the wave mismatches and corresponding modulation periods Λ_j by using dispersion data for a $LiNbO_3$ crystal presented in [14]. It was assumed that the domain walls lie in the zy plane and the reciprocal nonlinear lattice vector is parallel to the x axis, which is possible by producing the domain structure by the polarisation switching method [13]. Periods Λ_j for the wavelength $\lambda_1 = 2\pi c/\omega = 1.064$ μm are

$$\Lambda_1 = 20.7\ \mu\mathrm{m}, \tag{7.1}$$

type I

$$\Lambda_2 = 14.2\ \mu\mathrm{m}, \tag{7.2}$$

$$\Lambda_3 = 8.0\ \mu\mathrm{m}; \tag{7.3}$$

$$\Lambda_1 = 20.7\ \mu\mathrm{m}, \tag{8.1}$$

type II

$$\Lambda_2 = 14.2\ \mu\mathrm{m}, \tag{8.2}$$

$$\Lambda_2 = 14.2\ \mu\mathrm{m}. \tag{8.3}$$

Figure 1 shows the function $g(z)$ (2) for coupled quasi-phase-matching interactions, which contains modulation periods of type I.

The four-wave processes under study are described by the following systems of truncated equations for the complex amplitudes of interacting waves:
for type I interactions (3),

$$\frac{\mathrm{d}A_1}{\mathrm{d}z} = \mathrm{i}g(z)\left[\beta_{11}A_2^2 \mathrm{e}^{\mathrm{i}\Delta k_1 z} + \beta_{21}A_2^* A_3 \mathrm{e}^{-\mathrm{i}\Delta k_2 z}\right],$$

$$\frac{\mathrm{d}A_2}{\mathrm{d}z} = \mathrm{i}g(z)\left[\beta_{12}A_1 A_2^* \mathrm{e}^{-\mathrm{i}\Delta k_1 z}\right.$$

$$\left. + \beta_{22}A_3^* A_1 \mathrm{e}^{-\mathrm{i}\Delta k_2 z} + \beta_{32}A_3^* A_4 \mathrm{e}^{-\mathrm{i}\Delta k_3 z}\right], \tag{9}$$

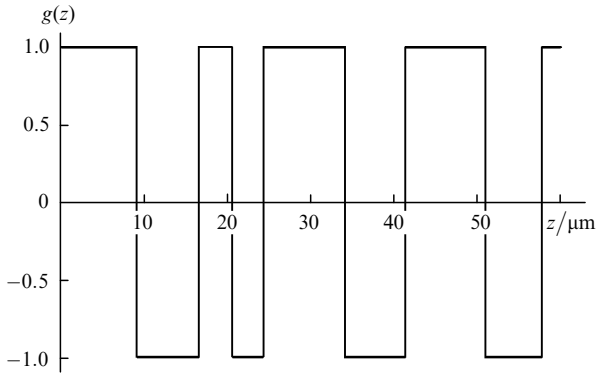

Figure 1. Function $g(z)$ characterising the aperiodic variation of the sign of the nonlinear coefficient upon the interaction of light waves with multiple frequencies in a $LiNbO_3$ crystal (the fundamental radiation wavelength is 1.064 μm).

$$\frac{dA_3}{dz} = ig(z)\left[\beta_{23}A_1A_2e^{i\Delta k_2 z} + \beta_{33}A_2^*A_4e^{-i\Delta k_3 z}\right],$$

$$\frac{dA_4}{dz} = ig(z)\left[\beta_{34}A_3A_2e^{i\Delta k_3 z}\right];$$

for type II interactions (4)

$$\frac{dA_1}{dz} = ig(z)\left[\beta_{11}A_2^2e^{i\Delta k_1 z} + \beta_{21}A_2^*A_3e^{-i\Delta k_2 z} + \beta_{31}A_1^*A_4e^{-i\Delta k_3 z}\right],$$

$$\frac{dA_2}{dz} = ig(z)\left[\beta_{12}A_1^*A_2e^{-i\Delta k_1 z} + \beta_{22}A_3^*A_1e^{-i\Delta k_2 z}\right], \qquad (10)$$

$$\frac{dA_3}{dz} = ig(z)\left[\beta_{23}A_1A_2e^{i\Delta k_2 z}\right],$$

$$\frac{dA_4}{dz} = ig(z)\left[\beta_{34}A_1^2e^{i\Delta k_3 z}\right].$$

Here, A_j is the complex amplitude of the wave at frequency ω_j; β_{ij} are the nonlinear coupling coefficients satisfying the relations (see [15], p. 291):

type I $\quad \beta_{12} = \beta_{11},\ \beta_{23} = \beta_{21} + \beta_{22},\ \beta_{34} = \beta_{32} + \beta_{33};$

type II $\quad \beta_{12} = \beta_{11},\ \beta_{23} = \beta_{21} + \beta_{22},\ \beta_{34} = \beta_{31}.$

The system of equations (9), (10) was solved numerically. It was assumed that pumping was performed at 1.064 μm. The pump intensity was assumed to be ~ 100 MW cm^{-2}; the characteristic nonlinear length for lithium niobate corresponding to this pump intensity is $L_{nl} = (\beta_{11}|A_1(0)|) \approx 0.1$ cm, where A_1 is the pump-wave amplitude.

Figure 2 presents the dependences of the interacting waves on the crystal length for type I and type II processes. The system of equations was solved for complex amplitudes A_j normalised to $|A_1(0)|$. The boundary conditions for both systems of equations were

$$\mathrm{Re}A_1(z=0) = 1, \quad \mathrm{Re}A_2(z=0) = 10^{-3}, \quad \mathrm{Re}A_3(z=0) = \mathrm{Re}A_4(z=0) = 0, \quad \mathrm{Im}A_j(z=0) = 0, \quad j = 1-4.$$

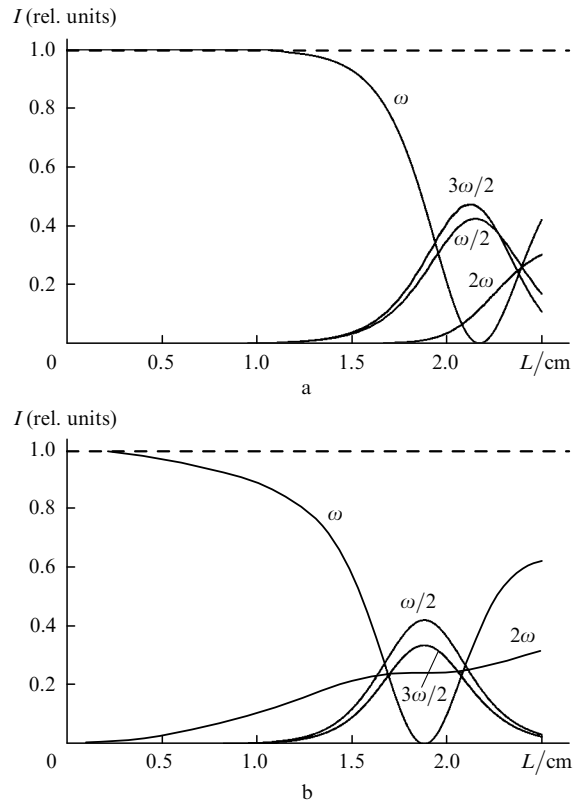

Figure 2. Dependences of the wave intensities on the interaction length for the type I process for the spatial amplitudes of harmonics $a_1 = 13.9$, $a_2 = 12.1$, $a_3 = 7$ (a) and for the type II process for $a_1 = 20$, $a_2 = 18$, and $a_3 = 1.4$ (b).

4. Discussion of results

It follows from Fig. 2 that a considerable energy exchange between the interacting waves occurs at the interaction lengths of ~ 2 cm, which corresponds in our case to $20L_{nl}$. From the point of view of generation of multiple frequencies, the type II interaction is preferable because second harmonic generation occurs due to the pump-wave frequency doubling, whereas the type I interaction requires intense fields at frequencies $\omega/2$ and $3\omega/2$. Note also that type II has two interaction lengths (see Fig. 2b) at which the wave intensities are almost equal. The dynamics of energy exchange between the waves presented in Fig. 2 takes place at some nonlinear coupling coefficients of the waves, which, as mentioned above, are determined by the amplitudes a_j of modulating harmonics in (2).

We studied the radiation field at the output of a crystal of length L, which is a superposition of waves with multiple frequencies

$$E(t, L) = \sum_{j=1}^{4} A_j(L)e^{i(\omega_j t - k_j L)}. \qquad (11)$$

Our analysis has shown that the behaviour of the 'instantaneous field intensity' $S(t, z) = (cn/8\pi)|E(t, z)|^2$ (the light energy flux density) is complicated. It depends on the interaction length, which determines both the intensities of interacting waves and their phases. To form a regular train of pulses, two conditions should be fulfilled: the phases of the waves should be equal and the amplitudes of the harmonics should be almost equal to each other. Our

numerical calculations have shown that these conditions can be fulfilled by selecting properly the crystal length. In this case, the phasing of the waves at some interaction length is provided by the dispersion of the crystal itself.

Figure 3 shows the typical time dependences of the radiation intensity at the crystal output in this case. One can see that the radiation intensity changes periodically. The different values of the peak intensities of pulses for different interaction lengths are caused, in our opinion, by the incomplete phasing of generated frequencies. The pulse repetition period determined by the frequency $\omega/2$ is ~ 7.1 fs, the pulse duration being dependent on the number of harmonics. According to Fig. 3, it slightly exceeds 700 as. The peak intensity is more than three times greater than the fundamental radiation intensity. Note that pulses in Fig. 3 differ from traditional few-cycle laser pulses because they do not contain the modulation internal structure, i.e. represent video pulses [4].

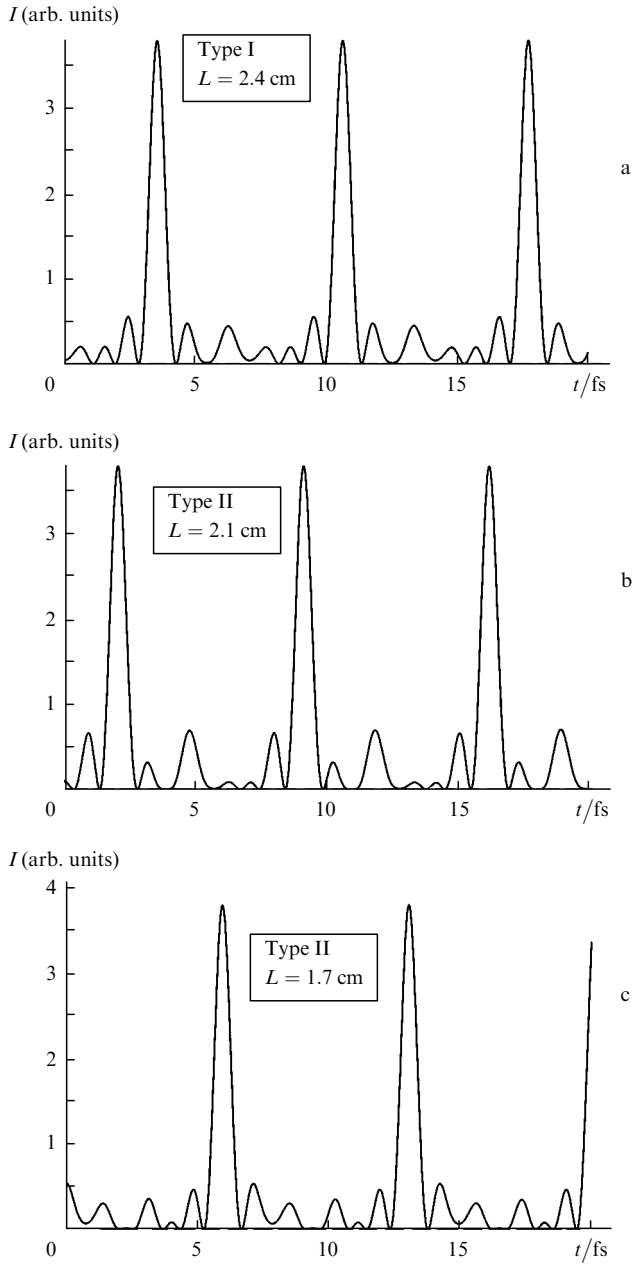

Figure 3. Time dependences of the radiation intensity for the type I process for the crystal length $L = 2.4$ cm (a) and the type II process for $L = 2.1$ (b) and 1.7 cm (c).

To realise the interactions under study in real nonlinear-optical crystals, which can have lengths of a few centimetres, intensities ~ 100 MW cm^{-2} are required. Such intensities are provided by 100-ps pulses of a 1.064-μm Nd : YAG laser with an average output power of 1 W. However, one should bear in mind that the theory developed above concerns the stationary interaction of the waves, while in the case of short pulses, the group delay effects can be manifested in the nonlinear process. Table 1 presents the group-velocity mismatch in a $LiNbO_3$ crystal for the waves under study. The group delay is characterised by the length $l_{gr} = \tau/v_{jk}$, here $v_{jk} = 1/u_j - 1/u_k$ is the group delay and u_q is the group mismatch of the wave of length λ_q.

Table 1. Group-velocity mismatch v_{jk} (10^{-12} s cm^{-1}) in $LiNbO_3$.

λ (μm)	2.128	1.064	0.709	0.532
2.128	0	−0.15	−2.8	−8.14
1.064	0.15	0	−2.65	−7.99
0.709	2.8	2.65	0	−5.34
0.532	8.14	7.99	5.34	0

Then, according to Table 1, the minimal l_{gr} for $\tau = 100$ ps is ~ 12 cm. Therefore, group-delay effects are insignificant in this case, and theoretical results obtained in the paper can be used to analyse the interaction of nanosecond laser pulses.

5. Conclusions

We have studied the method of synthesis of subfemtosecond laser pulses in nonlinear-optical crystals with the aperiodically modulated nonlinear susceptibility. Two four-wave processes involving three simultaneously proceeding nonlinear processes with multiple frequencies have been investigated for a $LiNbO_3$ crystal with the aperiodically modulated nonlinear coefficient. It has been shown that pulses of duration of a few hundreds of attoseconds can be generated by selecting properly the crystal length. To generate shorter pulses, it is necessary to use the entire transparency region of the nonlinear crystal, which is possible upon pumping at longer wavelengths. This will provide the generation of a larger number of harmonics and an increase in the pulse repetition period.

The results presented in the paper demonstrate the principal possibility of generating attosecond pulses in nonlinear crystals with the aperiodically changing nonlinear coefficient.

Acknowledgements. The authors thank G.D. Laptev for discussions of the results of the paper and P.G. Kryukov for useful comments. This work was partially supported by the Russian Foundation for Basic Research (Grant No. 05-02-17017) and Grant No. MK-4708.2006.2 of the President of the Russian Federation.

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