NONLINEAR-OPTICAL PHENOMENA

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Nonlinear pulse compression in inhomogeneous photonic crystals upon backward second harmonic generation

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***Abstract*.** **Frequency doubling of phase-modulated laser pulses, which is caused by a quasi-synchronous interaction of counterpropagating waves, is studied theoretically in crystals with an aperiodic domain structure. The simultaneous influence of the change in the domain period and the phase-modulation depth of fundamental radiation on the formation of a second-harmonic pulse is analysed in the nonstationary regime. It is shown that there exists an optimal relation between chirps in an aperiodic crystal and the phase modulation of fundamental radiation at which the maximum nonlinear compression of the second-harmonic pulse duration is possible.**

Keywords: *backward second harmonic generation, quasi-synchronous interactions, crystals with an aperiodic domain structure, nonlinear compression of laser pulses.*

Nonlinear-optical crystals with a quadratic susceptibility can be used for nonlinear compression of ultrashort laser pulses in different types of parametric interaction of waves in the nonstationary regime. Compression means a decrease in the pulse duration of radiation generated in nonlinear-optical processes compared to the pulse duration of input radiation. Therefore, compression of this type is often called the 'nonlinear pulse compression'. For example, during sum-frequency generation, the efficient pulse compression at the sum frequency can be achieved in the case of the optimal relation between the group-velocity mismatch and intensity [1] or in the case of the inclined phase fronts [2, 3] of incident wave beams in the nonstationary regime. This pulse compression is also observed during the nonstationary second harmonic generation (SHG) upon the type II phase matching if mutually perpendicular polarised pulses of fundamental radiation are directed into the nonlinear crystal with an optimal time delay with respect to each other [4 – 6].

Note that the most spectacular nonlinear compression of phase-modulated laser pulses occurs during the nonstationary SHG in quadratic nonlinear-opticalal pulses with an aperiodic domain structure (ADS) in which the inverse domain period changes linearly along the longitudinal coordinate (the so-called ADS crystals with a linear chirp) [7 – 10]. It was shown that the effective pulse compression of fundamental radiation with some depth of quadratic phase modulation should be observed at the optimal quantity of the ADS-crystal linear chirp $D_{\rm opt} = -\upsilon_0^2/C$ and the optimal crystal length $L_{\rm opt} = |(\sqrt{2}\upsilon_0)/(\tau_0 D_{\rm opt})|$, where $\upsilon_0 = 1/V_1 - 1/V_2$; V_1 and V_2 are the group velocities of fundamental radiation and second harmonic (SH) pulses, respectively; $C = \tau_0(\tau_1^2 - \tau_0^2)^{1/2}$ is the parameter determining the phase-modulation depth of the fundamental radiation pulse; τ_0 and τ_1 are the durations of transform-limited and phase-modulated pulses of fundamental radiation, respectively. The nonlinear compression physics in this case consist in the following: different spectral components of the fundamental radiation pulse are efficiently transformed along the crystal into the corresponding SH pulse components so that different spectral SH components leave the nonlinear crystal simultaneously. As a result there arise optimal conditions for nonlinear compression of the SH pulse. For example, the use of this scheme allowed 150-fold nonlinear compression of a 17-ps fundamental radiation pulse to be experimentally achieved upon frequency doubling of laser radiation in the $LiNbO_3$ ADS crystal [8].

The aim of this paper is also to study the nonlinear compression mechanism of short laser pulses during the nonstationary SHG. Unlike previous papers, we consider the case when the quadratic nonlinear-optical medium generates a SH wave directed towards fundamental radiation. This type of frequency conversion is called the backward SHG [11]. At present, this and other types of parametric frequency conversion can be successfully realised in practice due to the improvement of the crystal growth technology providing the quasi-synchronous interaction of waves in media with ultrashort domain dimensions [12].

In the nonstationary regime the processes of the backward SHG of the phase-modulated fundamental radiation pulse in ADS crystals are described by a system of equations, which in the moving coordinate system has the form [13]

$$\frac{\partial A_1}{\partial z} = -\mathrm{i}\Gamma A_1^* A_2 \exp(-\mathrm{i}D_1 z^2),$$
$$\frac{\partial A_2}{\partial z} - \upsilon\frac{\partial A_2}{\partial t} = \mathrm{i}\Gamma A_1^2 \exp(\mathrm{i}D_1 z^2) \tag{1}$$

with the boundary conditions

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$$A_1(z,t)|_{z=0} = \frac{A_0}{(\tau_0^2 + \mathrm{i}C)^{1/2}} \exp\left[-\frac{t^2}{2(\tau_0^2 + \mathrm{i}C)}\right], \tag{1a}$$

$$A_2(z,t)|_{z=L} = 0.$$

Here, A_1 and A_2 are the complex amplitudes of fundamental radiation and second harmonic, respectively; A_0 is the maximum real amplitude of fundamental radiation at the input to the nonlinear medium; Γ is the nonlinear coupling coefficient; L is the nonlinear medium length; $\upsilon = 1/V_1 + 1/V_2$.

In (1) we used the Fourier expansion of the sign-alternating quadratic-susceptibility function for the case, when the inverse domain period changes along the direction z and the linear phase detuning is compensated for only at the crystal centre [7]:

$$\delta(z) = \frac{2\pi}{m} \exp[-\mathrm{i}(\Delta k z + D_1 z^2 + D_2 z^3 + ...)]. \tag{2}$$

Here, $\Delta k = 2k_1 + k_2$; k_1 and k_2 are the wave numbers of fundamental radiation and second harmonic, respectively; m is the quasi-synchronism order; D_1 and D_2 are the Fourier expansion coefficients of the aperiodic lattice.

To solve the linear part of the system of differential equations with partial derivative (1), we used the standard method of the fast Fourier transform and to solve the nonlinear part, we used the fourth-order Runge–Kutta method. Then, the results of these methods were joined together in accordance with the scheme of 'symmetric splitting' of steps, which was developed to analyse the interaction of wave packets in dispersive media [14, 15]. Note that because of the 'nonuniformity' of boundary conditions (1a), the solution of system (1) requires additional iterative algorithms. In particular, algorithms based on the 'shooting' method [13] or Lagrange multipliers [16] refer to such iterative algorithms.

In this paper, we developed a special iterative algorithm, based on the Lagrange multiplier method, to determine the unknown SH pulse profile at the input to the nonlinear medium ($z = 0$). Note that this method was used in [16] for determining the unknown time profile of the fundamental radiation intensity distribution in order to obtain the specified SH intensity distribution profile during the nonstationary SHG in homogeneous nonlinear-optical media with the quadratic nonlinearity.

Unlike [16], we used the following boundary conditions for the functions of the Lagrange multipliers: $\lambda_1(z,t)|_{z=L} = 0$, $\lambda_2(z,t)|_{z=L} = -A_2(z,t)|_{z=L}$, where $\lambda_1(z,t)$ and $\lambda_2(z,t)$ are the functions of the Lagrange multipliers for fundamental radiation and second harmonic, respectively. The sequence of our algorithm work corresponds to the sequence of algorithm work in [16]. (In calculations we assumed that the transverse dimensions [beam radius] of fundamental radiation are much smaller than the transverse dimensions of the ADS crystal and quasi-synchronous interaction of waves occurs only along the coordinate z.)

Based on the numerical solution of (1), we analysed the effect D_1 (in calculations we assumed that only the linear chirp is present in the ADS crystal: $D_1 \neq 0$, $D_2 = 0$) on the backward SHG efficiency and the SH pulse compression coefficient σ ($\sigma = \tau_2/\tau_1$; τ_2 is the SH pulse duration at $z = 0$, which was determined as a root-mean-square half-width of the intensity distribution at the 1/e level from the maximum one [17]). In calculations we used dimensionless quantities $A_0 = 1$, $\tau_0 = 1$, $\upsilon = 1$, $C = 50$, $\Gamma = 1$ and $L = L_{\mathrm{opt}}$. Let us compare the characteristic interaction length $L_{\mathrm{nl}} = 1/(A_0\Gamma)$ and the quasi-static interaction length $L_\upsilon = \tau_0/\upsilon$ with the length L of the nonlinear crystal under our conditions in order to estimate the effect of the group velocity dispersion and intensity of fundamental radiation on the backward SHG process. In this case, the condition $L \gg L_{\mathrm{nl}} \gg L_\upsilon$ is fulfilled, because $L \approx 7$, $L_{\mathrm{nl}} = 1$ and $L_\upsilon = 0.1$. This means that the influence on frequency conversion process of group velocity detuning compared to other effects is decisive. Preliminary estimates show that under these conditions the decrease in the fundamental radiation intensity during the nonstationary SHG does not exceed a few percent. In this connection, this nonlinear-optical process was analysed under conditions of a weak energy exchange (i.e. in the specified-field approximation) by neglecting the distortion of the SH pulse shape caused by the depletion of fundamental radiation. Note that the developed approach can be used only in the case of a strong energy exchange of interacting waves. However, the theoretical analysis of the mentioned problem is beyond the scope of this paper.

The results of calculations are presented in Fig. 1 which shows the dependences of the SH pulse compression coefficient σ (solid curve) and the backward SHG energy efficiency η (dashed curve) on the normalised quantity of the ADS-crystal spatial chirp D_1/D_{opt}. One can se that when D_1 increases, the SH pulse duration, after some growth, decreases. At $D_1 = D_{\mathrm{opt}}$, maximum compression of the SH pulse duration is observed. Thus, our calculations show that the maximum compression during the backward SHG is achieved for the optimal values of the length and the linear spatial chirp of the ADS crystal, which are equal to their corresponding optimal values during the forward SHG.

Note that the maximum energy efficiency of the backward SHG in the case under study is observed at $D_1 \neq 0$ and not at $D_1 = D_{\mathrm{opt}}$ or $D_1 = 0$. One can see it well from the dependence of the backward SHG efficiency plotted with the help of a dashed curve. (However, calculations show that the maximum value of the SH pulse peak intensity is

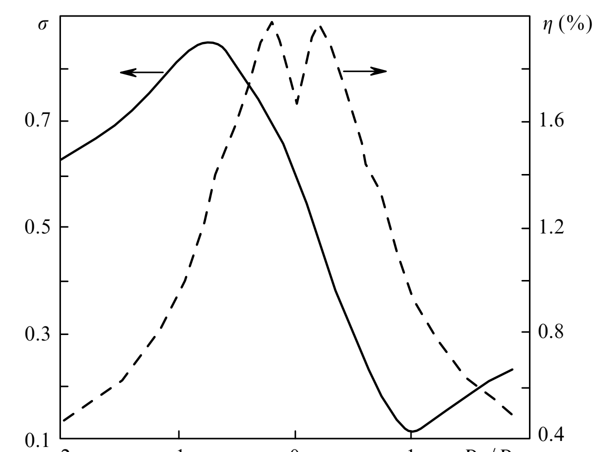

Figure 1. Dependences of the compression coefficient σ (solid curve) and the energy conversion efficiency η (dashed curve) on the spatial chirp quantity D_1/D_{opt} of the ADS crystal.

observed under optimal conditions of compression for the forward SHG at $D_1 = D_{opt}$.) It follows from the analysis of the figure that the energy efficiency of the backward SHG at $D_1 = D_{opt}$ compared to the case of a periodic polarised crystal ($D_1 = 0$) decreases by approximately twice. This relatively small decrease in the efficiency is caused by the fact that the spectral width of the ADS-crystal synchronism is larger than that of a periodic crystal [18]. Recent theoretic investigations of the forward SHG [19] and the degenerate parametric amplification of light [20, 21] also demonstrated a similar behaviour of the frequency conversion efficiency of phase-modulated laser pulses in ADS crystals. Note that as we assumed in calculations, the maximum deviation of the domain period from its exact value, at which the quasi-synchronous interaction condition is fulfilled for the central frequency of the fundamental radiation pulse, does not exceed the quasi-synchronism order m.

Figure 2 shows the time profiles of the normalised SH intensity at the ADS-crystal input for different D_1. It follows from calculations that at $D_1 = D_{opt}$ the maximum intensity is approximately 8 or 14 times higher than that at $D_1 = 0$ and $D_1 = -D_{opt}$, respectively. At the same time the energy efficiency η of the backward SHG at $z = 0$ was 1.6 %, 1.05 % and 0.8 % for $D_1 = 0$, $-D_{opt}$ and D_{opt}, respectively (see Fig. 1).

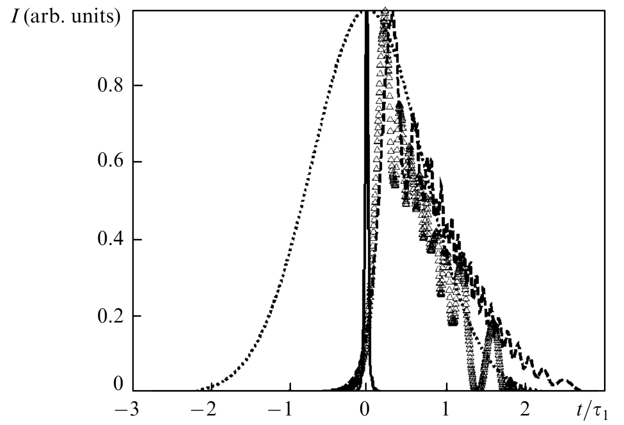

Figure 2. Normalised time profiles of the SH intensity for $D_1 = D_{opt}$ (solid curve), 0 ($\triangle$) and $-D_{opt}$ (dashed curve) at $z = 0$. The intensity profile of the input phase-modulated pulse of fundamental radiation (dotted curve) is presented for comparison.

Note also that the experimental realisation of different types of the parametric frequency conversion, including the backward SHG in periodically polarised nonlinear crystals with extremely small domains, can be achieved in practice, because the growth technology of such crystals is well developed at present [22]. For example, paper [12] demonstrated experimentally the parametric amplification of light in periodically polarised $LiNbO_3$ crystals with a 800-nm period for the amplification of an idler wave counter-propagating with respect to the pump beam. It is shown in [13] that the backward SHG of femtosecond laser pulses in the ADS crystals can be used for optimal production of femtosecond SH pulses as is in the case of the conventional SHG [23].

Consider the specific experiment: frequency doubling of radiation at 1.56 μm, with the maximum intensity $I_0 = 10$ GW cm^{-2}, the duration $\tau_1 = 10.5$ ps and the phase modulation depth $C = 1.4$ ps^2 upon the ee – e-type interaction in the $LiNbO_3$ crystal of length $L = 0.1$ cm with the minimum and maximum domain period $\Lambda_{min} = 2.52$ μm and $\Lambda_{max} = 2.88$ μm, respectively. In this case, $\upsilon \approx$ 150 ps cm^{-1} and the domain period Λ_0, at which quasi-synchronous backward SHG of the 15th order is possible, is 2.7 μm [thus, $D_1 = \pi(\Lambda_{min} - \Lambda_{max})/(L\Lambda_{max}\Lambda_{min}) \approx D_{opt} \approx 1.55$ mm^{-2}]. At these parameters, the varying period range of the ADS-crystal domains lies between the quasi-synchronism orders $m = 14$ and 16 for the backward SHG while the ADS-crystal chirp quantity and the phase modulation depth of the input fundamental radiation pulse satisfy the optimal conditions for the effective compression of the SH pulse. The results of calculations are presented in Fig. 3. Figure 3a shows the change in the energy efficiency of the backward SHG along the ADS-crystal length, while Fig. 3b – the normalised (by the maximum intensity of an optimally compressed SH pulse) time profiles of the SH intensity at the nonlinear medium input. One can see that the change in the sign of the ADS-crystal chirp strongly affects the time profile of the SH intensity. The SH pulse (solid curve) elongates at the opposite sign of the ADS-crystal chirp with respect to its optimal value. Under the optimal conditions, the SH pulse is efficiently compressed ($\sigma \approx 0.05$ is the dashed curve in Fig. 3b). Thus, in this case the SH pulse is compressed approximately by 20 times.

Note that in the general case, the conditions for the quasi-synchronism of different orders can be fulfilled for different types of the three-frequency interaction. In this case, it is necessary to take into account all associated types of the frequency-conversion processes. However, in the case considered here, no such analysis is necessary. Calculations

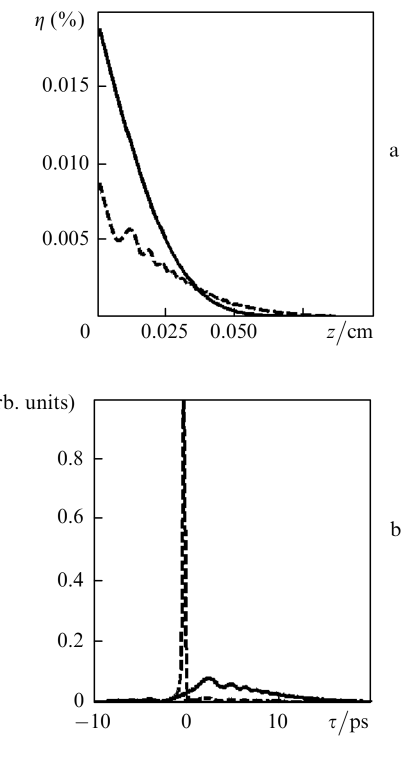

Figure 3. Dependence of the energy efficiency of the backward SHG along the ADS-crystal length (a) and the SH intensity profiles at the nonlinear medium input (b); solid curves correspond to the calculations obtained at $D_1 = -D_{opt}$, dashed curves – at $D_1 = D_{opt}$.

show that in the range of variations in the dimensions of lithium niobate domains and in the spectral region, the quasi-synchronism condition of the 25th order (the domain dimension is ~ 2.73 μm) is fulfilled only for the backward third harmonic generation. Taking into account the decrease in the frequency conversion efficiency with increasing the synchronism order, the effect of the backward third harmonic generation in the case under study should be insignificant.

Note also that in this case, the energy efficiency of the backward SHG is relatively small, which is explained by the choice of the quasi-synchronism order for which the production of the domains with the required thickness is possible at present. The use of a lower quasi-synchronism order in the case of the corresponding decrease in the domain thickness can lead to a significant increase in the efficiency.

Thus, in this paper we have studied theoretically the backward SHG of phase-modulated laser pulses in ADS crystals and nonlinear pulse compression. We have considered the case, when the inverse domain period changes linearly along the propagation direction of interacting waves. The simultaneous influence of the change in the domain period and the group velocity detuning has been studied. It has been shown that there exists an optimal relation between the quantities of the ADS-crystal chirps and the phase modulation of fundamental radiation at which the maximum compression of the SH pulse duration compared to the input pulse duration is possible in the nonstationary regime. The results of this paper can be used for the compression of ultrashort laser pulses in practice.

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