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# THIRD-HARMONIC GENERATION IN REGULAR DOMAIN STRUCTURES

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**Abstract**

The process of frequency conversion in regular domain structures is studied using the constant-intensity approximation. The investigations are carried out at values of complex amplitudes of the fundamental radiation and third harmonic at the output of each domain equal to the values of the corresponding complex amplitudes at the input of the subsequent domain. We show that the optimum length of each domain depends on the input pump intensity in the given domain. Thus, it is possible by choosing the optimum lengths of domains, phase mismatch, and pump intensity even at a low number of periods of nonlinear susceptibility modulation of the lattice to reach considerable values of conversion efficiency at the structure output in comparison with the traditional case of homogeneous nonlinear media.

**Keywords:** frequency conversion, constant-intensity approximation, regular domain structure.

## 1. Introduction

Generation of high harmonics is attractive for solving a number of applied problems in the UV and extreme-ultraviolet (EUV) ranges of the spectrum and obtaining photons of high energy. The gases transparent in the UV range supplement the class of transparent nonlinear crystals used for the third-order processes, and the direct process of third-harmonic generation (THG) is always allowed in contrast to the second-harmonic generation (SHG). With this highly efficient generation of harmonics, the phase adjustment between the pump and harmonic waves can be achieved. A prospective correcting scheme of undesirable shift of the phases between the interacting waves in nonlinear media is regular domain structures (so-called quasi-phase-matched schemes) [1, 2].

The current interest in regular domain structures is explained, in particular, by the successive elaboration of such structures using ferroelectrics [3, 4]. In the process of ferroelectric growth, under definite conditions a change in the susceptibility sign versus the boundary of layers is observed. For instance, if a crystal belongs to the centrosymmetric class of symmetry, the components of the third-order nonlinear susceptibility at the domain boundary do not change sign. Therefore, in TGS crystals the regular domain structure is not realized while seignette-salt crystals with similar structures are successfully employed. Crystallographic analysis of $LiNbO_3$ shows that, at regular domain structures in such crystals, the second-harmonic generation is efficient, but it is not the case for the third-harmonic generation. An

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analogous analysis for a $Pb_5Ge_3O_11$ ferroelectric provides the possibility to observe quasi-phase-matched THG in such crystals [5].

Ferroelectrics are also used for the quasi-phase-matched parametric conversion of the laser-radiation frequency [6–8]. The layers with quadratic nonlinearity are similar to the layers with cubic nonlinearity in their action when one accounts for the consecutive interactions at the boundaries of each layer [9–11]. In addition to solid crystals, quasi-phase-matched THG was observed in optical structures [12] and liquid crystals [13].

The quasi-phase-matched regime is an efficient way of converting the input pump beam inside nonlinear media into a beam of practically any wavelength within the crystal-transparency range. At present, the process of frequency conversion at quasi-phase-matched interactions was realized in the whole spectral range, from IR to EUV [2,9]. Quasi-phase-matched interactions were used for creating light sources of the three basic colors, red, green, and blue (RGB sources) and for determining the crystals' quality [14–18].

Analysis of the process of frequency tripling in regular domain structures at consecutive quasi-phase-matched interaction was considered in [7–11], where the constant-field approximation (CFA) was mainly used. Along with layers of quadratic nonlinearity, quasi-phase-matched THG with layers of cubic nonlinearity can be employed. For direct third-harmonic generation, KDP crystals ($LiNbO_3$, $LilO_3$, BBO, and LBO) are successfully used. In [19,20], the efficiency of conversion to the third harmonic of laser radiation was about 26% for a LBO crystal and 24% for a KDP crystal at a pump power of about 200 GW/cm$^2$, i.e., a substantial increase in the contrast of femtosecond pulses was reached at wavelength $\lambda = 1.24\ \mu$m. The quasi-phase-matched third-harmonic generation was demonstrated using a simple silica structure of six modulation periods in [21], where it was shown that after a suitable development such devices might become practical and inexpensive alternative sources of blue or near ultraviolet radiation, if the usual pulsed near infrared solid-state lasers are used.

Investigations of quasi-phase-matched interactions of the fundamental radiation in the constant-intensity approximation (CIA) [19, 20] allows one to discover a number of new effects that have not been observed in CFA [21, 22]. The analysis of nonlinear quasi-phase-matched wave interaction in CIA in the SHG case showed that, by selecting the optimum length of domains, it is possible to achieve phase mismatch between the interacting waves, and with the same pump intensity to increase the efficiency of conversion in comparison with the traditional case of homogeneous nonlinear media [23].

In this study, we analyze in the CIA the phase effects of the fundamental radiation in the process of direct tripling of the laser-radiation frequency in regular domain structures created from the crystalline layers with cubic nonlinearity. We assume the complex-amplitude values of the fundamental radiation and the third harmonic at the output of each domain to be equal to the input values of the corresponding complex amplitudes at the input of the subsequent domain. In view of the theory elaborated, we provide concrete recommendations concerning the factors restricting the efficiency of conversion to the third harmonic.

## 2. Theory

We consider the dynamics of energy exchange between the waves of the fundamental radiation and third harmonic in the domains, each representing a layer with cubic susceptibility. Let us assume that the regular domain structure under study consists of consecutive layers with periodically changing susceptibility sign.

We produce consecutive calculations of the quasi-phase-matched interaction from layer to layer in the structure. In the general case of dissipating nonlinear media, the process of direct third-harmonic generation in a separately taken domain is described by the corresponding reduced equations. The method of calculations assumes a solution of the problem with appropriate boundary conditions where the output parameters of the preceding layer are the input parameters of the subsequent layer.

We investigate the process of frequency tripling in the first domain. The reduced equations describing THG in the first domain, taking into account the medium losses, read as follows [24]:

$$\frac{dA_1}{dz} + \delta_1 A_1 = -i\gamma_1 A_3 A_1^* \exp(i\Delta z), \qquad \frac{dA_3}{dz} + \delta_3 A_3 = -i\gamma_3 A_1^3 \exp(-i\Delta z), \tag{1}$$

where $A_{1,3}$ are the complex amplitudes of the pump wave and third harmonic at frequencies $\omega_{1,3}$ ($\omega_3 = 3\omega_1$), $\delta_j$ and $\gamma_j$ are, respectively, the absorption coefficients and nonlinear coefficients of interacting waves at corresponding frequencies $\omega_j$ ($j = 1, 3$), $n_{1,3}$ are the refractive indices at frequencies $\omega_{1,3}$, $\lambda_1$ is the wavelength of the pump radiation, and $\Delta = k_3 - 3k_1$ is the phase mismatch of the waves within the boundaries of each domain.

First, we solve the problem in the first domain when at the input of the regular domain structure there exists only the pump wave and there is no third-harmonic wave, namely,

$$A_1(z=0) = A_{10}\exp(i\varphi_{10}), \qquad A_3(z=0) = 0, \tag{2}$$

where $z = 0$ corresponds to the first-domain input and $\varphi_{10}$ is the initial phase of the pump wave at the first-domain input.

Using the standard procedure of solving the system of equations (1) taking into account the condition (2) in the CIA, the expression for the complex amplitude of the third harmonic at the first domain output ($z = l_1$) reads [25]

$$A_3(l_1) = -i\gamma_3 A_{10}^3 l_1 \operatorname{sinc} \lambda_1' l_1 \exp[2i\varphi_{10} - (\delta_3 + 3\delta_1 + i\Delta)l_1/2], \tag{3}$$

where

$$\lambda_1'^2 = 3\Gamma_1^2 - (\delta_3 - 3\delta_1 - i\Delta)^2/4, \qquad \Gamma_1^2 = \gamma_1\gamma_3 I_{10}^2, \qquad \operatorname{sinc} x = \frac{\sin x}{x}, \qquad I_j = A_j A_j^*.$$

From (3) it follows that the third-harmonic amplitude depends on the length periodically. First, there occurs a transfer of the fundamental radiation energy to the third harmonic at a distance of the optimum length (coherent length). Then reverse pumping over the energy takes place. At the second-harmonic generation, the optimum length of interaction $l_{\rm opt}^{(2)}$ in the first domain at small $\Delta$ is inversely proportional to $\sqrt{I_{10}}$ ($l_{\rm opt}^{(2)} \sim 1/\sqrt{2\gamma_1\gamma_2 I_{10} + \Delta^2/4}$) [22]. At the third-harmonic generation, the optimum length of interaction $l_{\rm opt}^{(3)}$, as follows from (3), is inversely proportional to the first degree of the pump intensity $I_{10}$ ($l_{\rm opt}^3 \sim 1/\sqrt{3\gamma_1\gamma_3 I_{10}^2 + \Delta^2/4}$ ). Thus, at the third-harmonic generation, the optimum length of interaction depends more substantially on the pump intensity. Hence, under definite conditions, the third harmonic may reach the maximum conversion before it takes place for the second harmonic, i.e., at $l_{\rm opt}^{(3)} < l_{\rm opt}^{(2)}$.

The optimum length of the first domain, at which the third-harmonic amplitude reaches its maximum, is determined by the formula ($\delta_3 = 3\delta_1$) [25]

$$l_{1,\rm opt} = \lambda_1^{-1} \arctan(\lambda_1/\delta_3), \tag{4}$$

where

$$\lambda_1^2 = 3\Gamma_1^2 + \Delta^2/4.$$

Hence one can see that if one uses the CIA (in contrast to the CFA), the coherent length depends not only on the phase mismatch but also on the pump intensity $I_{10}$ and the media losses. With increase in the pump intensity, the optimum length decreases.

Now we consider generation of the third harmonic in the second domain. If the conditions of the quasi-phase-matched regime at the transition from the first domain to the second one is fulfilled, the tensors of cubic susceptibility of the first and second domains differ in sign. This is equivalent to changing the sign of nonlinear coefficients at the transition from layer to layer [1,7,9]. The complex amplitudes in this case are also described by the system of equations (1), but the nonlinear coefficients in the second domain are denoted as $\gamma'_{1,3}$, and the initial values of the complex amplitudes of the fundamental radiation and third harmonic are determined by the values at the output of the first layer, i.e., the boundary conditions are as follows:

$$A_{1,3}(z=0) = A_{1,3}(l_1)\exp[i\varphi_{1,3}(l_1)]. \tag{5}$$

Here $\varphi_{1,3}(l_1)$ is the wave-phase change at the transition from the first to the second domain at frequencies $\omega_{1,3}$, respectively, and $z = 0$ again corresponds to the input but of the second domain.

In the CIA, solving the system of equations (1) taking into account condition (5) for the complex amplitude of the third harmonic at the output of the second domain ($z = l_2$), we arrive at

$$\begin{aligned} A_3(l_2) &= A_3(l_1)\left\{\cos\lambda'_2 l_2 - \left[i\gamma'_3\frac{A_1^3(l_1)}{A_3(l_1)}e^{i[3\varphi_1(l_1)-\varphi_3(l_1)]} - \frac{3\delta_1-\delta_3+i\Delta}{2}\right]\frac{\sin\lambda'_2 l_2}{\lambda'_2}\right\} \\ &\quad\times\exp\left[i\varphi_3(l_1) - (\delta_3+3\delta_1+i\Delta)l_2/2\right], \end{aligned} \tag{6}$$

where

$$\lambda_2'^2 = 3\Gamma_2^2 - (\delta_3 - 3\delta_1 - i\Delta)^2/4, \qquad \Gamma_2^2 = \gamma'_1\gamma'_3 I_1^2(l_1).$$

Inserting (3) in the system of equations (1), we obtain the expression for the complex amplitude of the fundamental radiation $A_1(l_1)$. Hence, taking into account (3), the ratio $A_1^3(l_1)/A_3(l_1)$ reads

$$\frac{A_1^3(l_1)}{A_3(l_1)} = \frac{i}{\gamma_3}\left(\lambda'_1\cot\lambda_1{}'l_1 + \frac{\delta_3-3\delta_1-i\Delta}{2}\right)\exp(i\Delta l_1). \tag{7}$$

As a result, for the complex amplitude of the third harmonic, taking into account (7), one obtains $\delta_3 = 3\delta_1$ from (6)

$$\begin{aligned} A_3(l_2) &= A_3(l_1)\left\{\cos\lambda_2 l_2 + \left[(\lambda_1\cot\lambda_1 l_1 - \frac{i\Delta}{2})\frac{\gamma'_3}{\gamma_3}e^{i\psi} + \frac{i\Delta}{2}\right]\frac{\sin\lambda_2 l_2}{\lambda_2}\right\} \\ &\quad\times\exp\left[i\varphi_3(l_1) - \delta_3 l_2 - i\Delta l_2/2\right], \end{aligned} \tag{8}$$

where

$$\psi = 3\varphi_1(l_1) - \varphi_3(l_1) + \Delta l_1, \qquad \lambda_2^2 = 3\Gamma_2^2 + \Delta^2/4.$$

To simplify the results, we assume that the initial values of the wave phases at the input to each domain are equal to zero, $\varphi_{1,3} = 0$. However, these initial values will be taken into consideration below in analyzing the conversion process in regular domain structures.

The efficiency of conversion from (8) to the third harmonic after passing through two domains $\eta_3(l_2) = I_3(l_2)/I_{10}$ is determined by the following expression:

$$\eta_3(l_2) = \eta_3(l_1)\left[\left(\cos\lambda_2 l_2 + c_1\frac{\sin\lambda_2 l_2}{\lambda_2}\right)^2 + b^2\frac{\sin^2\lambda_2 l_2}{\lambda_2^2}\right]\exp\left(-2\delta_3 l_2\right), \tag{9}$$

where

$$c_1 = \frac{\gamma_3'}{\gamma_3}\left(\lambda_1\cot\lambda_1 l_1\cos\psi - \frac{\Delta}{2}\sin\psi\right), \qquad b = -\frac{\gamma_3'}{\gamma_3}\left(\frac{\Delta}{2}\cos\psi + \lambda_1\cot\lambda_1 l_1\sin\psi\right) + \frac{\Delta}{2}.$$

Here $\eta_3(l_1) = I_3(l_1)/I_{10}$ is the efficiency of conversion to the third harmonic after the wave passes through the first domain; it is determined from (3).

At the optimum length of the first domain $l_{1,\mathrm{opt}} = \pi/2\lambda_1$, the efficiency $\eta_3(l_2)$ is

$$\eta_3(l_2) = \eta_3(l_1)\left[\left(\cos\lambda_2 l_2 + \frac{\gamma_3'}{\gamma_3}\frac{\Delta}{2}\sin\psi\right)^2 + \Delta^2\left(\frac{\gamma_3'}{\gamma_3}\cos\psi - 1\right)^2\frac{\sin^2\lambda_2 l_2}{4\lambda_2^2}\right]\exp\left(-2\delta_3 l_2\right).$$

In the case $\psi = 2\pi n$, $n = \pm1, \pm2, \ldots$, the expression is simplified and we arrive at

$$\eta_3(l_2) = \eta_3(l_1)\left[\cos^2\lambda_2 l_2 + \Delta^2\left(1 - \frac{\gamma_3'}{\gamma_3}\right)^2\frac{\sin^2\lambda_2 l_2}{4\lambda_2^2}\right]\exp\left(-2\delta_3 l_2\right). \tag{10}$$

One can see that in order to obtain an increase in the frequency-conversion efficiency, the best is the case where the values in round brackets are summed up. This can be realized for the nonlinear coefficients that differ in sign, i.e., at $\gamma_3'/\gamma_3 = -1$.

From (9), one can obtain the optimum value of the second-layer length ($\delta_3 = 0$)

$$l_{2,\mathrm{opt}} = 0.5\,\lambda_2^{-1}\arctan\left[-2q/(q^2 + p - 1)\right],$$

where

$$q = \frac{\gamma_3'}{\gamma_3}\frac{\lambda_1}{\lambda_2}\cot\lambda_1 l_1, \qquad p = \left(\frac{\gamma_3'}{\gamma_3} - 1\right)^2\Delta^2/4\lambda_2^2.$$

From (9) also follows that there exist optimum values of the pump intensity and phase mismatch $\Delta$ under which the conversion efficiency is maximum.

To determine the complex amplitude of the fundamental radiation at the output of the second domain, we insert solution (8) into the system of equations (1). As a result, we obtain the following expression:

$$\begin{aligned} A_1(l_2) \;=\; & A_{10}\left\{l_1\frac{\gamma_3}{\gamma_3'}\mathrm{sinc}\,\lambda_1 l_1\left[c_1\cos\lambda_2 l_2 - \lambda_2\sin\lambda_2 l_2 + b\frac{\Delta}{2}\frac{\sin\lambda_2 l_2}{\lambda_2} + i\left(-c_2\frac{\Delta}{2} + b\cos\lambda_2 l_2\right)\right]\right\}^{1/3} \\ & \times\exp\left[-\delta_3(l_1 + l_2)/3 - i(l_1 - l_2)\Delta/6\right], \end{aligned} \tag{11}$$

where

$$c_2 = \cos\lambda_2 l_2 + c_1\frac{\sin\lambda_2 l_2}{\lambda_2}.$$

Our next step is to consider the process of third-harmonic generation in the third domain. To fulfill the conditions of quasi-phase-matched interactions, the signs of nonlinear coefficients of the first and third domains must coincide. The boundary conditions at the input of the third domain are the following:

$$A_{1,3}(z=0) = A_{1,3}(l_2), \tag{12}$$

where $z = 0$ corresponds to the input of the third domain.

In the process of frequency tripling in the third domain, $\gamma_{1,3}$ are the nonlinear coefficients in the system of equations (1) in the third domain. Solving the system of equations (1) for complex amplitudes of the third harmonic in a standard way taking into account (12), we obtain

$$A_3(l_3) = A_3(l_2)\left\{\cos\lambda_3 l_3 - i\left(\gamma_3 \frac{A_1^3(l_2)}{A_3(l_2)} e^{i[3\varphi_1(l_2)-\varphi_3(l_2)]} - \frac{\Delta}{2}\right)\frac{\sin\lambda_3 l_3}{\lambda_3}\right\}\exp\left(-\delta_3 - i\,\Delta/2\right) l_3, \tag{13}$$

where $\varphi_{1,2}(l_2)$ is the wave-phase change at the transition from the second to the third domain at frequencies $\omega_{1,3}$ respectively,

$$\lambda_3^2 = 3\Gamma_3^2 + \Delta^2/4, \qquad \Gamma_3^2 = \gamma_1\gamma_2 I_1^2(l_2).$$

The ratio $\dfrac{A_1^3(l_2)}{A_3(l_2)}$ can be found, in view of expressions (8) and (11). From (13), we obtain

$$A_3(l_3) = A_3(l_2)\left\{\cos\lambda_3 l_3\left[\frac{c_4 + ic_5}{c_2 + ic_3}\exp\left(i\Delta l_2\right) - i\frac{\Delta}{2}\right]\frac{\sin\lambda_3 l_3}{\lambda_3}\right\}\exp\left(-\delta_3 - i\frac{\Delta}{2}\right) l_3, \tag{14}$$

where

$$c_3 = b\frac{\sin\lambda_2 l_2}{\lambda_2}, \qquad c_4 = c_1\cos\lambda_2 l_2 - \lambda_2\sin\lambda_2 l_2 + c_3\frac{\Delta}{2}, \qquad c_5 = -c_2\frac{\Delta}{2} + b\cos\lambda_2 l_2.$$

The expression for $A_1(l_3)$ can be obtained solving the second equation of (1) for the complex amplitude of the fundamental radiation taking into consideration (14).

From (14), we obtain the efficiency of the harmonic conversion at the output of the third domain as follows:

$$\eta_3(l_3) = \eta_3(l_2)\left[\left(\cos\lambda_3 l_3 + c_8\gamma_2\frac{\sin\lambda_3 l_3}{\lambda_3}\right)^2 + (c_9\gamma_2 - \frac{\Delta}{2})^2\frac{\sin^2\lambda_3 l_3}{\lambda_3^2}\right]\exp(-2\delta_3 l_3), \tag{15}$$

where

$$c_6 = c_2\cos\Delta l_2 + c_3\sin\Delta l_2, \qquad c_7 = -c_2\sin\Delta l_2 + c_3\cos\Delta l_2,$$

and

$$c_8 = \frac{c_4c_6 + c_5c_7}{\gamma_3\prime(c_2^2 + c_3^2)}, \qquad c_9 = \frac{c_4c_7 - c_5c_6}{\gamma_3\prime(c_2^2 + c_3^2)}.$$

Now we consider the case where the frequency conversion takes place in the fourth domain. From the quasisynchronism conditions follows that nonlinear coefficients of the fourth domain coincide in sign with nonlinear coefficients of the second domain. Therefore, in the system of equations (1), if the tripling process is considered in the fourth domain, the nonlinear coefficients are denoted as $\gamma'_{1,3}$. The boundary conditions at the input of the fourth domain are as follows:

$$A_{1,3}(z=0) = A_{1,3}(l_3), \tag{16}$$

where $z=0$ again corresponds to the domain input.

Solving the system of equations (1) with the boundary conditions (16) for the third-harmonic generation in the fourth domain, we obtain the following expression for the complex amplitude of the third harmonic at the output of this domain

$$A_3(l_4 = A_3(l_3)\left\{\cos\lambda_4 l_4 - i\left(\gamma'_3\frac{A_1^3(l_3)}{A_3(l_3)} - \frac{\Delta}{2}\right)\frac{\sin\lambda_4 l_4}{\lambda_4}\right\}\exp\left(-\delta_3 l_4 - i\frac{\Delta l_4}{2}\right), \tag{17}$$

where

$$\lambda_4^2 = 3\Gamma_4^2 + \Delta^2/4, \qquad \Gamma_4^2 = \gamma'_1\gamma'_3 I_1^2(l_3).$$

After some algebra, we arrive at the expression for the ratio $A_1^3(l_3)/A_3(l_3)$

$$\frac{A_1^3(l_3)}{A_3(l_3)} = \frac{c_{14}\cos\Delta l_3 - c_{15}\sin\Delta l_3 + i\left(c_{15}\cos\Delta l_3 + c_{14}\sin\Delta l_3\right)}{c_{10}^2 + c_{11}^2}, \tag{18}$$

where

$$c_{10} = \cos\lambda_3 l_3 + c_8\gamma_3\frac{\sin\lambda_3 l_3}{\lambda_3}, \qquad c_{11} = \left(c_9\gamma_3 - \frac{\Delta}{2}\right)\frac{\sin\lambda_3 l_3}{\lambda_3},$$

$$c_{12} = c_8\cos\lambda_3 l_3 - \left[\frac{\Delta}{2\lambda_3}\left(c_9 - \frac{\Delta}{2\lambda_3}\right) + \frac{\lambda_3}{\gamma_3}\right]\sin\lambda_3 l_3,$$

$$c_{13} = c_9\cos\lambda_3 l_3 + c_8\frac{\Delta}{2\lambda_3}\sin\lambda_3 l_3, \qquad c_{14} = -c_{10}c_{13} - c_{11}c_{12}, \qquad c_{15} = c_{10}c_{12} - c_{11}c_{13}.$$

From (17), in view of (18), we obtain

$$A_3(l_4) = A_3(l_3)\left\{\cos\lambda_4 l_4 - \left[c_{19}\gamma'_3 + i\left(c_{18}\gamma'_3 - \frac{\Delta}{2}\right)\right]\frac{\sin\lambda_4 l_4}{\lambda_4}\right\}\exp\left(-\delta_3 l_4 - i\frac{\Delta l_4}{2}\right), \tag{19}$$

where

$$c_{16} = c_{15}\cos\Delta l_3 + c_{14}\sin\Delta l_3, \qquad c_{17} = c_{14}\cos\Delta l_3 - c_{15}\sin\Delta l_3, \qquad c_{18} = \frac{c_{17}}{c_{10}^2 + c_{11}^2}, \qquad c_{19} = \frac{c_{16}}{c_{10}^2 + c_{11}^2}.$$

According to (19), the efficiency of the third harmonic after propagating through all four consecutive domains is determined by the formula

$$\eta_3(l_4) = \eta_3(l_3)\left[\left(\cos\lambda_4 l_4 + c_{19}\gamma'_3\frac{\sin\lambda_4 l_4}{\lambda_4}\right)^2 + \left(c_{18}\gamma'_3 - \frac{\Delta}{2}\right)^2\frac{\sin^2\lambda_4 l_4}{\lambda_4^2}\right]\exp\left(-2\delta_3 l_4\right). \tag{20}$$

Carrying out the same procedure in the case of $n$ domains, one obtains for the amplitude of the third harmonic

$$A_3(l_n) = A_3(l_{n-1}) \left\{ \cos\lambda_n l_n + i\left(\frac{\Delta}{2} \mp \gamma_3^{(n)} \frac{A_1^3(l_{n-1})}{A_3(l_{n-1})}\right) \frac{\sin\lambda_n l_n}{\lambda_n} \right\} \exp\left(-\delta_3 l_n - i\frac{\Delta}{2} l_n\right), \tag{21}$$

where $\lambda_n^2 = 3\Gamma_n^2 + \Delta^2/4$ and $\Gamma_n^2 = \gamma_1^{(n)}\gamma_3^{(n)} I_1^2(l_{n-1})$, $I_1(l_{n-1})$ is the radiation intensity at the fundamental frequency at the input of the $n$th domain. The superscript corresponds to the odd number of domains ($\gamma_3^{(n)} = \gamma_3$), and the subscript corresponds to the even number of domains ($\gamma_3^{(n)} = -\gamma_3$).

The efficiency of conversion to the third harmonic after $n$ domains is determined by the following formula:

$$\eta_3(l_n) = \eta_3(l_{n-1}) \left[ \left(\cos\lambda_n l_n + c_a\gamma_3 \frac{\sin\lambda_n l_n}{\lambda_n}\right)^2 + \left(c_b\gamma_3 - \frac{\Delta}{2}\right)^2 \frac{\sin^2\lambda_n l_n}{\lambda_n^2} \right] \exp\left(-2\delta_3 l_n\right), \tag{22}$$

where the coefficients $c_a$ and $c_b$ are determined through the parameters that we have derived for the previous domains, such as the domain lengths $l_j$, phase mismatch $\Delta$, coefficients $\gamma_{1,3}$ ($\gamma'_{1,3}$), losses $\delta_{1,3}$), and intensity of the interacting waves. We do not present here the expressions for $c_{a,b}$ because of their cumbersomeness. The formulas obtained are similar in their structure to analogous formulas we obtained for the case of second-harmonic generation, but there are also differences, such as, for example, the expressions for $\lambda_n$ and $\Gamma_n$.

In this section, while analyzing the process of frequency conversion in regular domain structures within the CIA framework, we took into account two facts. First, the pump intensity was assumed to be constant inside one domain only, but it changed from domain to domain, as one can see from the boundary conditions (2), (5), (12), and (16). Second, as follows from our results, the optimum (coherent) lengths of the domains were not the same.

The analysis of the expressions for $\eta_3(l_2)$ (9), $\eta_3(l_3)$ (15), $\eta_3(l_4)$ (20), and $\eta_3(l_n)$ (22) obtained in the CIA showed that the optimum values of the lengths of the second, third, fourth, and fifth domains depend on the pump intensity. The synchronism-curve zeros obtained in the CIA in contrast to the CFA are not equidistant [19]; this fact is very important for calculating the synchronism width in regular domain structures.

## 3. Results and Discussion

Below, we present the results of our study of the quasi-phase-matched interaction, within the CIA framework, at THG in regular domain structures. Numerical calculation of the equations for the efficiency of conversion is presented in Figs. 1–5. Curves are plotted for the optimum lengths of the domains.

The dynamics of conversion into harmonics is shown in Fig. 1 in the presence and absence of losses in the medium under study. The dependences of the conversion efficiency $\eta_2(l_1)$ on the domain lengths $\Gamma_1 l_j$ are shown for two values of the phase mismatch $\widetilde{\Delta} = \Delta/2\Gamma_1$. One can observe an obvious maximum on the curves. During the pump-wave propagation in the regular domain structure, a gradual increase in $\eta_2$ from zero at the structure input to the maximum value at the structure output takes place. Because of

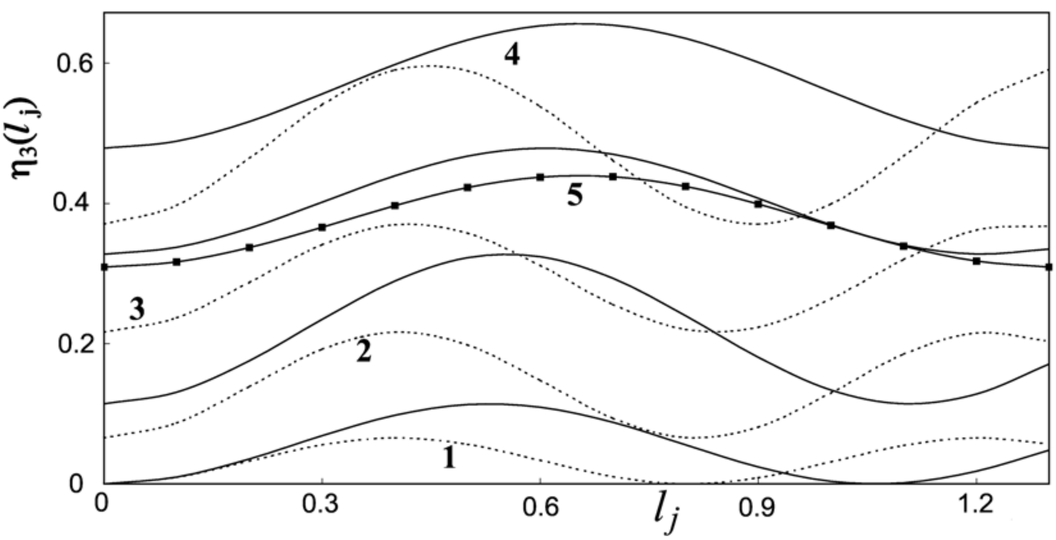


**Fig. 1.** Dependences of the efficiency of conversion $\eta_3(l_1 l_2 l_3 l_4)$ on the domain lengths $l'_j$ ($l'_j = \Gamma_1 l_j$) $j = 1 - 4$, respectively, calculated in the CIA at $\widetilde{\delta}_{1,2,3} = 0$ (solid and dotted curves 1–4) and 0.15 (curve with small squares 5) for $\Delta/2\Gamma_1 = 2.4$ (solid curves 1–4) and 3.5 (dotted curves 1–4). Here $\Gamma_1 l_{1,\rm opt} = 0.5307$, $\Gamma_1 l_{2,\rm opt} = 0.5544$, and $\Gamma_1 l_{3,\rm opt} = 0.6072$ (solid curves) and $\Gamma_1 l_{1,\rm opt} = 0.4022$, $\Gamma_1 l_{2,\rm opt} = 0.4077$, and $\Gamma_1 l_{3,\rm opt} = 0.4212$ (dotted curves).

optimum lengths of the domains, as soon as the efficiency reaches the optimum value in the first domain on the coherent length $\widetilde{l}_{1,\rm opt} = l_{1,\rm opt}/\Gamma_1 = 0.5307$ ($\widetilde{\Delta} = 2.4$), the harmonic wave passes to the second domain. Here, growth of $\eta_2$ continues until the coherent length $\widetilde{l}_{2,\rm opt} = 0.5544$. Then the wave enters the third domain where, as soon as the efficiency reaches its maximum value at $\widetilde{l}_{3,\rm opt} = 0.6072$, the harmonic wave, in turn, begins to propagate in the fourth domain. Finally, in the fourth domain, the efficiency $\eta_{3\max}$ reaches the maximum value $\eta_{3\max} = 0.6542$ ($\widetilde{\Delta} = 2.4$) at the coherent length $\widetilde{l}_{4,\rm opt} = 0.6950$, and so on. Hence, choosing the lengths of the domains with respect to the corresponding values of the coherent lengths, it is possible to obtain high efficiency of conversion at the output of the regular domain structure even with a few number of domains.

It is obvious from the comparison of the corresponding curves ($\Delta/2\Gamma_1 = 2.4$) (solid line) and ($\Delta/2\Gamma_1 = 3.5$) (dotted line) that, with increase in reduced phase mismatch from 2.4 to 3.5, both the efficiency $\eta_3$ and optimum lengths of the domains decrease. The latter leads to a shift of the corresponding dotted curves to the left with respect to the solid curves. In addition, it is also obvious that, with increase in the phase mismatch, the period of the spatial beatings of the third-harmonic intensity decreases. This fact was also reported in [26, 27], in view of accurate calculations of the system of reduced equations for the case of the second-harmonic generation. With increase in the losses in nonlinear media, on the one hand, the optimum domain length decreases (the curves' maxima shift to the left with respect to the maximum of curve 4), and this agrees with (3). On the other hand, just as predicted, the efficiency of conversion decreases (compare curves 4 and 5).

In addition to the effects described, the efficiency of conversion to the third harmonic is also affected by the values of nonlinear coefficients $\gamma_{1,3}$ themselves, i.e., their ratio $\gamma_3/\gamma_1$ also affects the efficiency of conversion [11].

In Fig. 2, we present the efficiency of conversion after light propagation of one and two domains

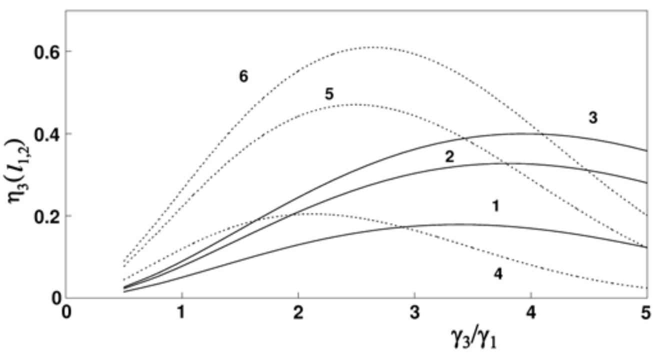


**Fig. 2.** Dependences of $\eta_3(l_{1,2})$ on the ratio $\gamma_3/\gamma_1$ for $\delta_j = 0$, $\gamma_1 I_{10} l_1 = 0.53$, and $\gamma_1 I_{10} l_2 = 0.55$ at $\widetilde{\Delta} = \Delta/2\gamma_1 I_{10} = 2.8$ (curves 3 and 6), 3 (curves 2 and 5), and 3.5 (curves 1 and 4). Dependences of $\eta_3(l_1)$ (solid curves 1–3) and $\eta_3(l_2)$ (dotted curves 4–6).

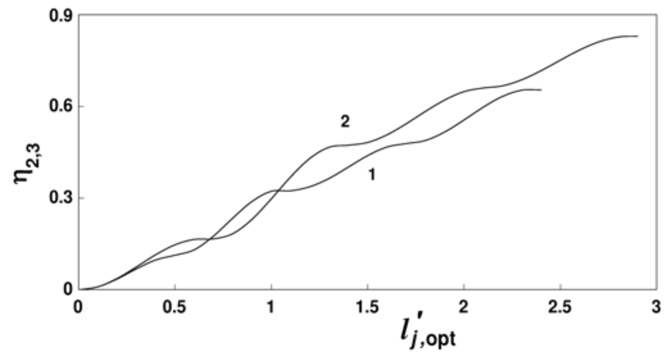


**Fig. 3.** Dependences of the efficiency of the third-harmonic generation $\eta_3(l_j)$, $j = 1-4$ on the optimum length of a regular domain structure $l'_{\rm opt}$ ($l'_{\rm opt} = \sum_{j=1}^{n} \Gamma_1 l_{j,\rm opt}$, where $n$ is the number of domains at $\Delta/2\Gamma_1 = 2.4$ and $\delta_{1,2} = 0$, respectively, for $\Gamma_1 l_{1,\rm opt} = 0.5307$, $\Gamma_1 l_{2,\rm opt} = 0.5544$, $\Gamma_1 l_{3,\rm opt} = 0.6072$, and $\Gamma_1 l_{4,\rm opt} = 0.6950$.

$\eta_3(l_{1,2})$ versus the value of $\gamma_3/\gamma_1$ for three different values of the phase mismatch $\widetilde{\Delta} = \Delta/\gamma_1 I_{10}$. One can observed a maximum, which testifies to the existence of an optimum value $(\gamma_3/\gamma_1)_{\rm opt}$. With decrease in the phase mismatch, the curve maxima shift to the right, i.e., the value $(\gamma_3/\gamma_1)_{\rm opt}$ increases.

Analyzing the behavior of the curves presented in Figs. 1 and 3, one can draw the conclusion that, with increase in the number of domains, the optimum values of the domain lengths increase, $l_{1,\rm opt} < l_{2,\rm opt} < l_{3,\rm opt} < l_{4,\rm opt}$.

In Fig. 4, dependences of the the conversion efficiency $\eta_3$ on the pump intensity $\widetilde{I}_{10}$ are presented. The observed maximum testifies to the existence of an optimum value of the pump intensity, and this is confirmed by the fact that the behavior of the experimental dependence $\eta_3(\widetilde{I}_{10})$ [28] differs qualitatively from the results obtained in the CFA, where the efficiency of conversion grew monotonically with increase in pump intensity. With increase in the medium losses, the optimum value $\widetilde{I}_{10,\rm opt}$, at which the efficiency reaches the maximum value, decreases.

The synchronism curves after one, two, three, and four domains in the presence and absence of losses are shown in Fig. 5. It follows from the observed maximum of the efficiency of conversion that there exists an optimum value of the phase mismatch. An increase in a number of domains leads (similar to the SHG case) to the increase in both the efficiency of conversion into the third harmonic and optimum values of the phase mismatch. The behavior of the dotted and solid curves shows that in dissipative media a shift of the maxima of the synchronism curves to greater values of the phase mismatch is observed.

In conclusion, we consider the example of a direct quasi-phase-matched generation of the third harmonic of a Nd:YAG laser ($\lambda = 1.064\ \mu$) in crystals with periodic domain structure. As is known, KDP crystals are destroyed by subnanosecond laser pulses at $S_{10} \sim 10^{10}$ W/cm$^2$, by picosecond pulses at $10^{12}$ W/cm$^2$, and by 30 fs pulses at $\sim 10^{14}$ W/Cm$^2$ [28]. $LiNbO_3$ crystals are damaged in fields three to four times smaller [28, 30].

Note that the results obtained above in the CIA for the case of stationary interaction are applicable also to the case of nonstationary interaction including picosecond pulses (applicability conditions for approximation in this case were considered in detail in [22]).

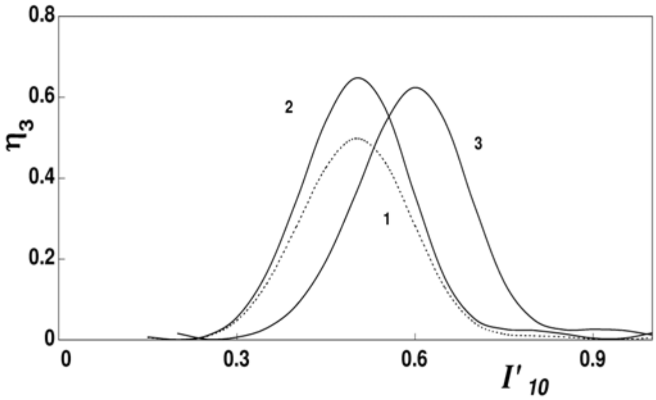


**Fig. 4.** Dependences of the efficiency of conversion $\eta_3(l_4)$ on the pump intensity in the case of optimum domain lengths $\Delta/2\Gamma_1 = 2.5$ (curves 1 and 2) and 2 (curve 3) for $\delta_2/\Gamma_1 = 0$ (curves 2 and 3), and 0.1 (curve 1).

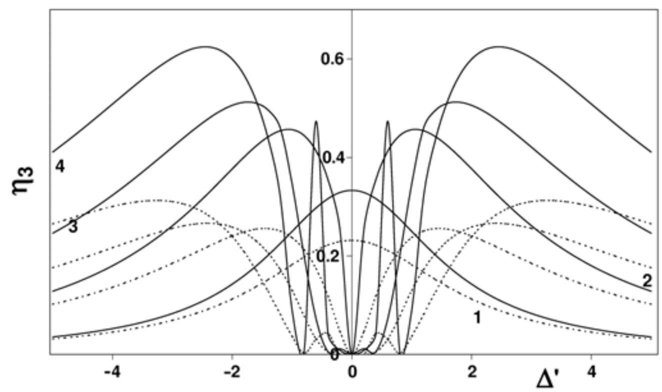


**Fig. 5.** Synchronism curves for $\delta_2/\Gamma_1 = 0$ (dotted curves 1–4) and 0.2 (dashed curves 1–4) calculated for the corresponding optimum lengths of the domains, namely, $\Gamma_1 l$ for $n = 1$, $\Gamma_1 l = \Gamma_1 l_1^{\rm opt}$, for $n = 2$, $\Gamma_1 l = \Gamma_1 l_2^{\rm opt} + \Gamma_1 l_1^{\rm opt}$, for $n = 3$, $\Gamma_1 l = \Gamma_1 l_1^{\rm opt} + \Gamma_1 l_2^{\rm opt} + \Gamma_1 l_3^{\rm opt}$, and for $n = 4$, $\Gamma_1 l = \Gamma_1 l_1^{\rm opt} + \Gamma_1 l_2^{\rm opt} + \Gamma_1 l_3^{\rm opt} + \Gamma_1 l_4^{\rm opt}$. Here $\Gamma_1 l_1^{\rm opt} = 0.4736$, $\Gamma_1 l_2^{\rm opt} = 0.4777$, $\Gamma_1 l_3^{\rm opt} = 0.4889$, and $\Gamma_1 l_4^{\rm opt} = 0.5023$.

Now we evaluate the maximum efficiency of conversion in a nonabsorbing KDP crystal with periodic domain structure at a pump power of $I_{10} = 100$ GW/cm$^2$. From the numerical calculations performed it follows that, by choosing the optimum parameters, it is possible to achieve maximum efficiency of conversion to the third harmonic in KDP crystals at the optimum length of the first domain $l_{1,\rm opt} = 0.27$ mm equal to $\eta_3 = 11.54\%$, which agrees with the experimentally obtained value of the efficiency of conversion for a KDP crystal at a pump power of 100 GW/cm$^2$ [19]. At the output of the second domain, $\eta_3(l_{2,\rm opt} = 0.292 \text{ mm}) = 33.78\%$, at the output of the third domain, $\eta_3(l_{3,\rm opt} = 0.32 \text{ mm}) = 50.29\%$, and at the output of the fourth domain, $\eta_3(l_{4,\rm opt} = 0.369 \text{ mm}) = 53.69\%$. From the above analysis of the expression for optimum lengths follows that the length of each domain at which the efficiency of conversion in a regular domain structure is maximum depends on the pump intensity at the domain input. During the laser-radiation propagation in a structure, its intensity gradually decreases due to the energy transfer to the third harmonic. In the CIA, we can take into account this effect considering each domain separately, which permits one to carry out a more strict analysis of nonlinear interactions in the process of frequency conversion. Hence, with increase in the number of the domains, the pump intensity at the each subsequent domain input decreases, and the optimum length of domains increases, as observed in our case. The optimum length of the suggested periodic domain structure based on a KDP crystal (for four domains) appears to be equal to 1.262 mm.

The same efficiency of conversion efficiency was obtained in a $LiNbO_3$ crystal at pump power $I_{10} = 50$ GW/cm$^2$ and optimum lengths of the domains ($l_{1,\rm opt} = 0.13$ mm, $l_{2,\rm opt} = 0.137$ mm, $l_{3,\rm opt} = 0.15$ mm, and $l_{4,\rm opt} = 0.174$ mm). The smaller pump power and optimum lengths of the domains are connected with the higher cubic nonlinearity of $LiNbO_3$ crystals in comparison with KDP crystals [31]. The whole size of the periodic domain structure based on $LiNbO_3$ (for four domains) is equal to 0.592 mm. When the absorption in the domains ($\delta_{1,2,3} l_j$=0.15) is taken into account, the efficiency of conversion to the third harmonic at the output of the structure decreases by 20%.

Finally, in the case of nonabsorbing BBO crystal with only one domain at pump power $I_{10} = 50\ \mathrm{GW/cm^2}$ (a passively mode-locked Nd: phosphate glass laser, $\lambda = 1.054\ \mu$m), the maximum efficiency of conversion can be obtained at the crystal optimum length $l_{1,\mathrm{opt}} = 0.284$ cm, and it is higher than the experimentally observed efficiency by 0.8% (at crystal length 0.72 cm) [31].

## 4. Conclusions

Thus, by choosing optimum lengths of the domains, phase mismatch, and pump intensity, it is possible even at a low number of periods of nonlinear susceptibility to modulate the structure to achieve high efficiency of conversion at the output of regular domain structures. The length of each domain at which the efficiency of conversion frequency is maximum depends on the pump intensity at the input of a given domain.

With increase in the number of domains, the pump intensity at the domain input decreases, and the optimum length of the domains increases. Under certain conditions, when intense laser fields are applied to the domain input, cases are possible where the third harmonic reaches the maximum efficiency of conversion before it takes place for the second harmonic.

Our analysis of the processes of direct frequency tripling in regular domain structures, developed here, can be applied to the efficiency of generation of difference and sum frequencies as well as to the investigation of the parametric interaction of nonlinear optical waves in such structures.

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