ON THE THEORY OF FREQUENCY DOUBLING IN TWO NONLINEAR CRYSTALS

Sh.Sh. AMIROV[1,2,3], A.I. HASANOVA[1], N.A. MAMMADOVA[1]
[1]*Department of Medical and Biological Physics, Azerbaijan Medical University, 167 S.Vurgun str., Az-1022, Baku, Azerbaijan*
[2]*Faculty of Physics, Baku State University, 23 Z.Khalilov str., Az-1148, Baku, Azerbaijan*
[3]*Department of Physics and Electronics, Khazar University, 41 Mahsati str., Az-1096, Baku, Azerbaijan*
[*]*Corresponding author: e-mail: phys_med@mail.ru*

The process of frequency doubling in two different nonlinear crystals in series is presented in the constant intensity approximation. The frequency doubling efficiency is sufficiently determined by the phase relationship between interacting waves. Analytical expressions for optimum values of crystal length, phase mismatch and phase relation are obtained in this approximation. It was shown that by choosing the phase mismatch, nonlinear coefficient of coupling for the second crystal unlike from those at the first crystal, as well as the optimum phase condition one can significantly increase the efficiency of frequency doubling in comparison with the identical crystals.

Keywords: frequency doubling, constant intensity approximation, phase effects, crystals in series.
DOI:10.70784/azip.1.2025128

1. INTRODUCTION

One of the effective methods of development of powerful sources of coherent radiation in the UV region is the multiplication of laser frequencies. Full conversion of the energy of fundamental wave to the energy of excited one is impossible even in phase matching conditions [1]. To increase coherency length at which occurs the pumping of the energy of exciting wave into the energy of harmonic, in addition to Fabry-Perrot resonators the special schemes including successively placed nonlinear crystals are used [2]. In the fixed field approximation the changes in phases of interacting waves is independent on the intensity of fundamental wave and therefore accurate analysis of the process of nonlinear interaction between waves is impossible. Previously, the constant intensity approximation was applied by us for analysis the second harmonic generation in Fabry-Perrot resonator [3], third harmonic generation in two crystals arranged in series [4] and cascade generation of the third harmonic [5].

2. DISCUSSION AND RESULTS

In this paper we have carried out detailed analysis of the frequency doubling in two series arranged different nonlinear crystals by employing the constant intensity approximation [6].Nonlinear crystals of different lengths are separated with air gap. A pump wave with frequency ω_1 excites the wave with doubled frequency $\omega_2 = 2\omega_1$ in the first crystal. After emergence from the first crystal those waves freely propagate in the air gap and are normally incident onto the surface of the second crystal, where the interaction of propagating waves occurs. One can consider the frequency conversion in each crystal individually. First we write the truncated equations [4,5] describing nonlinear interaction of waves

$$\frac{dA_1}{dz} + \delta_1 A_1 = -i\gamma_1 A_2 A_1^* e^{i\Delta_1 z}$$
$$\frac{dA_2}{dz} + \delta_2 A_2 = -i\gamma_2 A_1^2 e^{-i\Delta_1 z} \tag{1}$$

where $A_j\,(\mathrm{j}=1,2)$ – are the complex amplitudes of a pump and second harmonic waves, waves respectively, δ_j – are the absorption coefficients of those waves, $\Delta_1 = k_2 - 2k_1$ – is the difference in wave numbers (the phase mismatch parameter characterizing deviation from phase matching condition), γ_1, γ_2 – are the coefficients of nonlinear coupling.

Applying boundary conditions $A_1(z=0) = A_{10}$, $A_2(z=0) = 0$ to solve above set of equations in the TCA (most frequently termed as Tagiyev-Chirkin approximation)[6] for a complex amplitude of doubled wave at the exit of the first crystal yields

$$A_2(l_1) = -i\gamma_2 A_{10}^2 l_1 \sin c\lambda_1 l_1 \cdot \exp\left[-\left(\delta_2 + 2\delta_1 + i\Delta_1\right)l_1/2\right] \tag{2}$$

where $\lambda_1 = \left[2\Gamma_1^2 - \frac{\left(\delta_2 - 2\delta_1 - i\Delta_1\right)^2}{4}\right]^{1/2}$, $2\Gamma_1^2 = \gamma_1\gamma_2 I_{10}$, $I_{10} = A_{10}\cdot A_{10}^*$, $\sin cx = \frac{\sin x}{x}$

131, H.Javid ave, AZ-1073, Baku
Institute of Physics
E-mail: jophphysics@gmail.com

A similar expression can be obtained for the complex amplitude of a pump wave at the output of the first crystal

$$A_1(l_1) = A_{10}\left(\cos\lambda_1 l_1 + \frac{\delta_2 - 2\delta_1 - i\Delta_1}{4}\sin\lambda_1 l_1\right)^{1/2} \cdot \exp\left[-\left(\delta_2 + 2\delta_1 + i\Delta_1\right)l_1/4\right] \tag{3}$$

We can obtain from this expression the optimum length $l_1^{opt.}$ of the first crystal at which the harmonic intensity reachs its maximum value:

$$l_1^{opt.} = \frac{arctg(\lambda_1/\delta_2)}{\lambda_1} \tag{4}$$

where $\lambda_1^2 = 2\Gamma_1^2 + \frac{\Delta_1^2}{4}$

The boundary conditions for conversion to second harmonic in the second crystal are given by

$$A_1(z=0) = A_1(l_1)e^{i\varphi_1(d)},\ A_2(z=0) = A_2(l_1)e^{i\varphi_2(d)} \tag{5}$$

where $d-$ is the thickness of air gap between nonlinear crystals, $\varphi_{1,2}(d)$ – are the phase shifts of waves, $z=0$ corresponds to the input of the second crystal.

Analysis of frequency doubling in the second crystal also is carried out by the set of equations (1). However, here we substitute different coefficients for losses, nonlinear coupling as well as difference in wave numbers. Solving new set of equations with boundary conditions (5) for the complex amplitude of second harmonic yields

$$A_{2,out} = A_2(l_1)\left[a - c + (b+u)e^{i\psi} + i(g - fe^{i\psi})\right]\cdot\exp\left[-(\delta_2' + 2\delta_1' + i\Delta_2)z/2\right] \tag{6}$$

where

$$a = \cos\lambda_2 z,\ b = \beta\frac{\lambda_1}{\lambda_2}\cot\lambda_1 l_1 \sin\lambda_2 z,\ c = \frac{\delta_2' - 2\delta_1'}{2\lambda_2}\sin\lambda_2 z,\ u = \frac{\delta_2 - 2\delta_1}{2\lambda_2}\sin\lambda_2 z,$$

$$f = \beta\frac{\Delta_1}{2\lambda_2}\sin\lambda_2 z,\ g = \beta\frac{\Delta_2}{2\lambda_2}\sin\lambda_2 z,\ \beta = \frac{\gamma_2'}{\gamma_2}$$

and $\Psi = \Delta_1 l_1 + 2\varphi_1(d) - \varphi_2(d) -$ is the common phase shift between the pump and second harmonic waves in crystals and the air gap.

It is seen from (6) that the amplitude of second harmonic wave is the function of phase shift between Ψ waves. Variation in Ψ is equivalent to the change in signs of coefficients of nonlinear coupling γ_2 and γ_2' in the second crystal. Hence, depending on the phase mismatch Δ_2 and coefficients of nonlinear coupling of the second crystal one can distinguish four types of interaction.

Frequency conversion efficiency is given by

$$\eta = \gamma_2\Gamma_1^2(\gamma_1\rho_1)^{-1}(A+B)(\sin^2 x_1\cosh^2 y_1 + \sinh^2 y_1\cos^2 x_2)\times\exp\left[-\left(\delta_2 + 2\delta_1\right)l\right]$$

where

$$A = \left[\cos x_2\cosh y_2 + (Q_5\cos\Psi - Q_6\sin\Psi - \frac{Q_1}{2\sqrt{\rho_2}})\sin x_2\cosh y_2 - (Q_6\cos\Psi + Q_5\sin\Psi + \frac{Q_4}{2\sqrt{\rho_2}})\sinh y_2\cos x_2\right]^2$$

$$B = \left[\left(Q_6\cos\Psi + Q_5\sin\Psi + \frac{Q_4}{2\sqrt{\rho_2}}\right)\sin x_2\cosh y_2 + \left(Q_5\cos\Psi - Q_6\sin\Psi - \frac{Q_1}{2\sqrt{\rho_2}}\right)\sinh y_2\cos x_2 - \sin x_2\sinh y_2\right]^2$$

$$Q_1 = \left(\delta_2' - 2\delta_1'\right)\cos\xi_2/2 - \Delta_1\sin\xi_2/2,\ Q_2 = \left(\delta_2 - 2\delta_1\right)\cos\xi_2/2 - \Delta_1\sin\xi_2/2$$

$$Q_3 = \left(\delta_2 - 2\delta_1\right)\sin\xi_2/2 - \Delta_1\cos\xi_2/2,\ Q_4 = \left(\delta_2' - 2\delta_1'\right)\sin\xi_2/2 - \Delta_1\cos\xi_2/2$$

$$Q_5 = \beta\left(\sqrt{\frac{\rho_1}{\rho_2}}M + \frac{Q_2}{2\sqrt{\rho_2}}\right),\quad Q_6 = \beta\left(\sqrt{\frac{\rho_1}{\rho_2}}L - \frac{Q_2}{2\sqrt{\rho_2}}\right),\ \beta = \gamma_2'/\gamma_1'$$

$$M = K_1 \cos\theta + K_2 \sin\theta\,,\ L = K_1 \sin\theta + K_2 \cos\theta\,,\ K_1 = \frac{\tan x_1 / \cosh^2 y_1}{\tan^2 x_1 + \tanh^2 y_1}$$

$$K_2 = \frac{\tanh y_1 / \cos^2 x_1}{\tan^2 x_1 + \tanh^2 y_1}\,,\ x_{1,2} = \sqrt{\rho_{1,2}}\, l \cos\xi_{1,2}/2\,,\ y_{1,2} = \sqrt{\rho_{1,2}}\, l \sin\xi_{1,2}/2\,,$$

$$\rho_{1,2}^2 = \left[2\Gamma_{1,2}^2 + \frac{\Delta_{1,2}^2}{4} - \frac{(\delta_2 - 2\delta_1)^2}{4}\right]^2 + \frac{\Delta_{1,2}^2}{4}(\delta_2 - 2\delta_1)^2\,,\ \Psi = 2n\pi\,, \tag{7}$$

$$\xi_{1,2} = \tan^{-1}\left[\Delta_{1,2}(\delta_2 - 2\delta_1)/2\right] / \left[2\Gamma_{1,2}^2 + \frac{\Delta_{1,2}^2}{4} - \frac{(\delta_2 - 2\delta_1)^2}{4}\right],\ \theta = (\xi_1 - \xi_2)/2$$

Here we apply condition $\delta_2 = 2\delta_1 = \delta$ for losses. Intensity of second harmonic in accordance with (6) can be expressed by

$$I_{2,output} = I_2(l)\left\{a^2 + b^2 + f^2 + g^2 + 2\left[(ab - gf)\cos\Psi + (af + gb)\sin\Psi\right]\right\} \cdot e^{-2\delta l} \tag{8}$$

As can be seen harmonic intensity is a function of parameter Ψ expressing the phase relationship between interacting waves. Efficiency of frequency conversion reaches its maximum, when the following condition is fulfilled:

$$\Psi_{\max} = \pi\, n + \tan^{-1}\left[\frac{f(a-c) + g(b+u)}{(a-c)(b+u) - gf}\right],\ n = 0,1,2,... \tag{9}$$

Analysis of formula (9) for the optimum value of length of the second crystal yields

$$l_2^{opt.} = \arcsin \lambda_2^{-1}\left(1 + \frac{s}{\sqrt{4 + s^2}}\right)/2 \tag{10}$$

where $\Psi = 2n\pi$ and $s^2 = (q^2 + p - 1)/q$, $q = \beta\frac{\lambda_1}{\lambda_2}\cot\lambda_1 l_1$, $p = \frac{(\sigma_2 - \beta\sigma_1)^2}{2 + \sigma_2^2}$, $\sigma_{1,2} = \frac{\Delta_{1,2}}{2\Gamma_1}$.

The phase mismatch parameter receives its optimum value under optimum length of the first crystal:

$$\Delta_2^{opt.} = 4\gamma_1' \Gamma_1 \left(\gamma_1 \sqrt{2 + \sigma_1^2}\right)^{-1} \tag{11}$$

As can be seen from (11) the optimum value of phase mismatch in the second crystal depends not only on the phase mismatch in the first one, but also on the intensity of fundamental wave and coefficients of nonlinear coupling. Taking into account (12) yields to maximum value of harmonic efficiency

$$\eta_{\max.}(\sigma_1) = \frac{1}{2 + \sigma_1^2}\left(\frac{\gamma_2}{\gamma_1} + \frac{1}{2}\frac{\gamma_2'}{\gamma_1'}\sigma_1\sqrt{2 + \sigma_1^2}\right) \tag{12}$$

From (12) it follows that to obtain maximum efficiency of second harmonic the phase mismatch Δ_1 in the first crystal should receive definite $(\Delta_1 \neq 0)$ value. Further increase in phase mismatch Δ_1 leads to saturation in harmonic efficiency.

3. CONCLUSION

Analysis of frequency doubling in the Tagiyev-Chirkin approximation has allowed us to consider influence of the phases of backward waves on the phase of pumping wave as well as the linear losses of the interacting waves in the nonlinear medium. At the optimum crystal length the frequency doubling efficiency reaches its maximum value depending on both the intensity and the linear losses of the interacting waves. Optimim values of parameters such as crystal length, phase mismatch and phase relationship obtained in the mentioned approximation are the functions of intensity of fundamental wave. With the purpose to increase efficiency of frequency doubling more suitable type of conversion is considered to be with the same signs of nonlinear coupling coefficients and opposite signs of phase mismatch parameters. Thus by choosing the optimum values of problem parameters and compensating undesired phase shifts of interacting waves one can increase efficiency of conversion upon second harmonic generation in different nonlinear crystals arranged in series.

[1] *J.F. Rayntjes*. Nonlinear Optical Parametrical Processes in Liquids and Gases (in Russian) Moscow, Mir, 1987 , 512 p.

[2] *H. Huang, H. Wang, Sh. Wang and D. Shen.* Designable cascaded nonlinear optical frequency conversion integrating multiple nonlinear interactions in two $KTiAsO_4$ crystal. Optics express. 2018, V.26, issue 2, p.642-650.

[3] *Z.A. Tagiev, R.J. Kasumova and Sh.Sh. Amirov*. Theory of Intracavity Second Harmonic Generation in the Fixed-Intensity Approximation (in Russian). Opt. Spectrosk.1993, v.75, issue 4, pp.908-913.

[4] *Z.A.Tagiev and Sh.Sh. Amirov.* Third Harmonic Generation in nonlinear crystals arranged in series (in Russian) . Opt.Spectrosk.1992, v.73, issue 3 , pp.578-582.

[5] *Sh.Sh. Amirov, Z.A. Tagiev, M.G. Shakhtakhtinsky*. On the Theory of Cascade Third Harmonic Generation (in Russian).Quantum Electronics 1991, v.18, issue 6, pp.715-717.

[6] *Z.A. Tagiev and A.S. Chirkin.* Fixed intensity approximation in the thory of nonlinear waves id dispersed media (in Russian). Zh.Eksp. Teor.Fiz. 1977, v.73, issue 4, pp.1271-1282.

Received: 30.01.2025