A self-consistent model of second harmonic generation of laser radiation in the periodically poled nonlinear-optical crystal conditioned by its nonuniform heating

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Abstract. We introduce a self-consistent model that describes the process of the second harmonic generation (SHG) of the laser radiation in a nonlinear-optical crystal in conditions of its nonuniform heating induced by optical absorption. A periodically poled crystal pumped by the pulse laser radiation focused into its center was considered. The iteration procedure of solving the SHG problem relies on the interrelated solution of two equation systems describing the temperature dependent SHG and the heat conduction processes. The heat sources are represented by the absorbed power of both pump and generated radiation. Along with the absorption of pump and generated waves the diffraction effect was also taken into account. Eventually the resulting distribution of the crystal temperature should correspond to the radiation intensity distributions of pump and second harmonic.

1. Introduction

Nowadays the frequency conversion of laser radiation in nonlinear-optical crystals is widely used for generation in visible and middle infrared spectral ranges. Interaction of laser radiation with nonlinear-optical crystals results in its nonuniform heating due to both linear and nonlinear absorption of pump and generated photons. Along with the violation of phase matching conditions followed by the decrease of the conversion efficiency the other one detrimental effect of the nonuniform heating is the formation of defects, which are precursors of crystal laser damage. Thus, the control of crystal temperature distribution during processes of nonlinear-optical frequency conversion is of great importance.

A theoretical model that correctly describes the SHG process will help to prevent the violation of phase matching conditions by compensating the induced temperature gradients in nonlinear-optical frequency conversion experiments. For example, in the paper [2] a significant increase of SHG conversion efficiency was achieved by measuring and conducting subsequent optimization of the temperature gradient in the periodically polled lithium niobate (PPLN) crystal.

2. Theoretical model of SHG

Well known equations can be used for the theoretical description of the SHG process taking into account diffraction and optical absorption [1]. In the case the pump radiation propagates along the $z$ axis it can be written as follows:
The phase mismatch \( \Delta k \neq 0 \) in the presence of diffraction and optical absorption the nonuniform crystal temperature distribution leads to the complex dependence of second harmonic power on pump power, crystal length and other parameters. As a rule, numerical methods are used for solving the system of equations (1) as in most cases it cannot be solved analytically.

2.1. Periodically poled crystals

Various periodically poled (PP) crystals are widely used for the quasi-phase-matched SHG of infrared pump radiation, especially at moderate power levels in single-pass schemes [2]. This is justified by the high conversion efficiency due to exploitation of much larger values of nonlinear-optical susceptibility in the case of PP crystals. The direction of optical axes in adjacent domains of PP structures is inverse, as follows the coefficients \( d_{\text{eff}} \) have opposite signs. The phase mismatch \( \Delta k \) can be compensated by choosing the appropriate length \( L_{\text{dom}} \) of the domains. In order to describe SHG in PP crystals using the equations (1) the following substitution should be made:

\[
d_{\text{eff}} \rightarrow d_{\text{eff}} \text{sign} \left( \cos \left( \pi \frac{z}{L_{\text{dom}}} \right) \right)
\]

2.2. Pulse pump

The efficiency of SHG of a relatively low average pump power can be substantially increased by using pulse pump with a high peak power. In this case the validity of the solution of the equations (1) considering the average radiation power or the average radiation power of the pulse is doubtful. The correct approach is to find the solution for a specific power at every moment of time. For the laser pulses having a relatively large duration, specifically \( \tau_{\text{pump}} \geq 1 \) ns [1], and a narrow spectrum width \( (\Delta \lambda \leq 0.1 \text{ nm}) \) the effects conditioned by the pump spectral composition and group velocity delay can be neglected.

2.3. Nonuniform heating

A nonuniform heating of nonlinear-optical crystals occurs during SHG due to the difference of absorption coefficients of the pump and generated radiation. The temperature dependence of the refractive index results in the dependence of the phase-mismatch on spatial coordinates. Therefore, when modelling it is necessary to integrate the phase mismatch from the input \( (z = 0) \) to the current coordinate

\[
\Delta k \cdot z \rightarrow \int_{0}^{z} \Delta k(x, y, z') \, dz'
\]

2.4. Heat conduction problem

The nonuniform heating of the crystal has a major impact on the increase of the phase-mismatch \( \Delta k \). Temperature distribution of the nonlinear-optical crystal can be obtained by finding the solution of the
heat conduction problem. Taking into account only linear absorption of pump $\alpha_t$ and second harmonic $\alpha_s$ the heat conduction equation for the crystal surrounded by air can be written as follows:

$$\begin{align*}
\kappa_n \nabla \cdot (c_n \nabla T_{cr}) + \alpha_t I_1(x,y,z) + \alpha_s I_2(x,y,z) &= 0 \\
\kappa_n \frac{\partial T_{cr}}{\partial n} &= h^T (T_{cr} - T_{air}) |_{\partial \Omega},
\end{align*}$$

(4)

here $\kappa_n$ is the heat conduction coefficient of the crystal; $I_1, I_2$ are the radiation intensities of pump and second harmonics respectively; $\nabla \cdot (c_n \nabla)$ denotes normal derivative to the surface $\partial \Omega$; $x, y, z$ – spatial coordinates; $h^T$ is the heat transfer coefficient; $T_{cr}, T_{air}$ are temperatures of the crystal and ambient air respectively. In the heat conduction problem (4) the absorbed radiations of pump and second harmonic act as the heat sources. The heat transfer coefficient is assumed to be independent of temperature.

2.5. A self-consistent model of SHG

Obviously, the solution of SHG problem (1) depends on the crystal temperature distribution. In turn, the solution of the heat conduction problem (4) depends on the distribution of radiation intensities. As follows, a self-consistent problem should be considered. Initially, SHG equations (1) are solved assuming a certain temperature distribution of the crystal, e.g. uniform and equal to air temperature. Then the heat conduction problem is solved using obtained intensity distributions. A resulting temperature distribution is used for the next iteration and so on. It is expected, that at a certain iteration the self-consistent problem will converge, i.e. the overall difference between the distributions obtained in two consequent iterations will be below the specified deviation.

3. Results

3.1. Problem formulation

We have considered SHG of the single-mode pulse radiation (the wavelength $\lambda$, the average power $P$, the pulse duration $\tau$, the pulse repetition rate $\nu$). Pump radiation (frequency $\omega$) is focused into the center of PP crystal with the poling period $L_{dom}$. The crystal is surrounded by air. A scattered radiation does not induce additional heating of the crystal and can be neglected.

The simulations are performed using the SHG equations (1) taking into account the relations (2) and (3). The resulting system of the SHG equations can be written as follows:

$$\begin{align*}
\frac{dA_1}{dz} + \alpha_t A_1 + \frac{\nabla^2 A_1}{2ik_1} &= -i \frac{\omega d_{eff}}{cn_1} A_2 \exp(-i \int_0^z \Delta k(x, y, z')dz') \cdot \text{sign} \left( \cos \left( \frac{2\pi z}{L_{dom}} \right) \right) \\
\frac{dA_2}{dz} + \alpha_s A_2 + \frac{\nabla^2 A_2}{2ik_2} &= -i \frac{\omega d_{eff}}{cn_2} A_1 \exp(i \int_0^z \Delta k(x, y, z')dz') \cdot \text{sign} \left( \cos \left( \frac{2\pi z}{L_{dom}} \right) \right),
\end{align*}$$

(5)

where $\nabla^2 = \partial^2 / \partial x^2 + \partial^2 / \partial y^2$, $\epsilon_0$ – an electric permeability of vacuum, $\mu_0$ – a magnetic constant, $c$ – a light velocity in vacuum. It is assumed, that there is no second harmonic at the input end of the crystal and the pump radiation intensity has the Gaussian profile with a beam waist diameter $w_0$.

A split-step Fourier method [3] was used for the numerical solution of the SHG problem. This method is widely used for solving the nonlinear partial differential equations, e.g. a nonlinear Schrödinger equation. A linear step is produced in the space of Fourier coordinates and a nonlinear step is produced in the space of real coordinates. The pump pulse was considered to have a Gaussian shape. It was divided
in time into equal intervals and the SHG problem was solved for a number of average input powers at each interval. Then the total power of the pulse was restored.

A finite element method was used for the numerical solution of the heat conduction problem (4). For an accurate description of the heat sources represented by absorbed radiation of pump and harmonic the maximal size of the element did not exceed the size of the splits along the axes used for solving the SHG problem.

As a rule, far from the phase matching temperature the self-consistent SHG problem is solved in 8–12 iterations with an error below 0.5%. However, near the phase matching temperature the problem converges very slowly due to the strong dependence of the second harmonic intensity distribution on the temperature distribution of the crystal and more than 200 iterations are required to obtain an error less than 3%.

3.2. Experiment and simulation results

For the verification of the introduced theoretical approach the SHG experiment was carried out using the PPLN crystal (1×3×40 mm³, Ldom=3.393 μm). The pump source was the single-mode pulse ytterbium-doped fiber laser operating at λ=1064 nm wavelength (Δλ<0.1 nm) with the pulse duration τ=1.4 ns and the pulse repetition rate ν=700 kHz. The average pump power was P=1 W.

The absorption coefficient at the pump wavelength αₚ=0.5 m⁻¹ and the convective heat transfer coefficient hₚ=28 W/m²/K were experimentally measured using an equivalent temperature concept described elsewhere [4, 5]. The other parameters of PPLN such as dₑₚ=30 pm/V, α₂=5 m⁻¹ and refractive indices were taken from literature [6]. The equivalent temperature of the PPLN was measured exploiting its temperature dependent piezoelectric resonance frequencies. Frequencies of the piezoelectric resonances, which can be excited noncontactly via an application of the probe radiofrequency electric field, are preliminary calibrated in uniform heating conditions.

The simulation results of longitudinal temperature distributions of PPLN crystal in the pump radiation propagation direction calculated for two temperatures of the ambient air are shown in Fig.1. A black horizontal line shows the equivalent temperature value measured at phase matching conditions, i.e. maximal second harmonic output power. SHG conversion efficiency was close to 30%. Nonmonotonic temperature distribution along the crystal length can be attributed to the inverse conversion process (2ω → ω) occurring due to violation of the phase matching condition.

![Fig. 1. Simulated longitudinal temperature distributions of PPLN crystal during SHG along the pump radiation propagation direction at two different ambient air temperatures; the black line shows experimentally measured equivalent temperature of PPLN.](image-url)
4. Discussion and conclusions

Every parameter used for solving the self-consistent SHG problem conditioned by nonuniform heating of the crystal was found to produce an impact on the simulation results. The major effects on the simulation results are produced by the variation of nonlinear-optical susceptibility and the beam waist diameter of the pump radiation. In order to perform an accurate modelling it is necessary to measure these parameters with the high accuracy and in most cases this is a challenging task.

Another way to verify the results of the introduced model is to measure the temperature gradient of the crystal in the process of SHG as it will help to eliminate the influence of uncertainties of the involved parameters. However, such measurements cannot be performed accurately using the conventional contact thermal sensors due to the presence of a strong radiation scattering that induces additional heating of the sensors and some other methods should be employed.

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