Crystal equivalent temperature model in process of nonlinear conversion of laser radiation

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Abstract. Theoretical modeling was performed for estimation of influence of thermal effects in nonlinear-optical crystals on process of second harmonic generation of laser radiation. Due to absorption of both pump and generated radiation the temperature distribution inside crystal is substantially nonuniform as in cross-section as along its length. Three dimensional heat conduction problem was solved considering two types of heat sources, conditioned by exponential decay and parabolic growth of laser radiation intensity along crystal length. Longitudinal temperature gradient inside crystal is detrimental for process of laser radiation conversion.

1. Introduction

Currently due to rapid growth of laser physics and technology one of the urgent goals is development of laser sources that can generate high-power radiation at wavelengths in broad spectrum range. As a rule, generation wavelengths of modern laser sources are specified by the active media and cannot be widely tuned. Promising solution of spectral broadening is conversion of laser radiation wavelength in nonlinear-optical crystals [1]. Nonlinear-optical processes include parametric oscillation as well as multiple harmonic generation of laser radiation. These processes are coherent i.e. phase difference or phase velocity relation between interacting electromagnetic waves plays crucial role. As refractive indices of the crystal and therefore phase velocities of waves to a considerable extent depend on crystal temperature the last one should be accurately measured and controlled for efficient radiation conversion. Recently noncontact impedance spectroscopy method has been proposed for precise temperature measurement of nonlinear-optical crystals interacting with laser radiation [2-4]. It is well known that all nonlinear-optical crystals are piezoelectrics. Each crystal sample has its own set of intrinsic vibration modes. Piezoelectric resonances are observed in nonlinear-optical crystal response to the applied radiofrequency electric field when frequency of excitement corresponds to any intrinsic mode frequency. It was demonstrated that with high accuracy piezoelectric resonance frequencies Rf_n (n – mode index) linearly depend on crystal uniform temperature θ with piezoelectric resonance thermal coefficients K_n^{prt}. When we consider interaction with laser radiation the crystal is heated due to optical absorption and commonly its temperature is nonuniformly distributed in correspondence with laser intensity profile. It was proved experimentally [2, 3] and theoretically [4] that for the last case nonuniform temperature distribution can be characterized using notion of crystal equivalent temperature. Crystal equivalent temperature is directly determined from measured resonance

frequency shift $\Delta Rf_n(P)$ caused by laser irradiation of P power taking into account K_n^{prt} value of the resonance. Equivalent temperature concept has been already successfully implemented in well-known laser calorimetry technique for determination of optical absorption and heat transfer coefficients of nonlinear-optical crystals [5].

Nonlinear-optical crystal temperature measurement is of great interest directly in course of nonlinear-optical conversion of laser radiation [6-8]. In this case crystal is heated nonuniformly along propagation direction of laser radiation as far as optical absorption coefficients of pump and converted wavelengths can differ significantly. For theoretical interpretation of experimental results obtained in [6-8] it is necessary to elaborate model of acoustical vibration of bulk crystal sample under condition of nonuniform transverse and longitudinal crystal temperature distribution. First step to resolving of this problem is finding the solution of three dimensional heat conduction equation with spread heat source specified by the absorption of pump and generated laser radiation, which powers vary along propagation distance. Second harmonic generation (SHG) of single-mode laser radiation in nonlinear-optical crystal will be considered further.

2. Model of second harmonic generation in nonlinear-optical crystals

In order to find out temperature distribution inside nonlinear-optical crystal in course of SHG it is necessary to solve equations that describe nonlinear-optical SHG process. Dielectric crystal with nonzero second-order nonlinear-optical susceptibility is considered for this purpose. Nonlinear polarization P_{NL} of dielectric occurs when light wave with electric field E goes through it:

$$P_{\text{NL}} = \varepsilon_0 \chi^{(2)} : EE \,. \tag{1}$$

Here colon denotes tensor sum, ε_0 is permittivity of vacuum and $\chi^{(2)}$ is third rank tensor of second-order nonlinear-optical susceptibility. Overall electric field space-time dependence will be the sum result of first and second harmonic waves propagating along x_3 can be represented as

$$E(x_3,t) = \frac{1}{2}\mathrm{e}_1\left[A_1(x_3,t)\exp\left(i(\omega t - k_1 x_3)\right) + c.c.\right] + \frac{1}{2}\mathrm{e}_2\left[A_2(x_3,t)\exp\left(i(2\omega t - k_2 x_3)\right) + c.c.\right]. \tag{2}$$

Here index ‘1’ and ‘2’ refers to the first (ω) and second (2ω) harmonic respectively; ‘*c.c.*’ means complex conjugation; A denotes complex envelope; ω is frequency; e_i are unit vectors representing electric field polarization; k_i are wave vectors that depend on the refractive indices n_ω and $n_{2\omega}$ that are complex values in general case:

$$k_1 = \frac{n_\omega \omega}{c}, \quad k_2 = \frac{n_{2\omega} 2\omega}{c}, \tag{3}$$

where c is light velocity in vacuum. By Substituting (1) into (2) we can write in explicit form components of nonlinear polarization that oscillate at ω and 2ω frequencies:

$$P_{\text{NL}} = \frac{1}{4}\chi_\omega^{(2)} : \mathrm{e}_1\mathrm{e}_1 A_1^2(x_3)\exp\left[i(2\omega t - 2k_1 x_3)\right] + \frac{1}{2}\chi_{2\omega}^{(2)} : \mathrm{e}_1\mathrm{e}_2 A_1^*(x_3)A_2(x_3)\exp\left[i(\omega t + k_1 x_3 - k_2 x_3)\right] + c.c., \tag{4}$$

Here ‘*’ denotes complex conjugation; subscript of $\chi_\omega^{(2)}$ indicates on its dispersion properties.

Wave equation that describes evolution of electromagnetic wave in dielectric can be written as [9]

$$\nabla^2 E - \frac{1}{\varepsilon_0 c^2}\frac{\partial^2 D}{\partial t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2 P_{\text{NL}}}{\partial t^2}, \tag{5}$$

Here $D = \varepsilon_0(1+\varepsilon)E$ is electric displacement vector where ε is dielectric permittivity of the crystal. By substitution (2) and (4) in equation (5) we receive the following system:

$$\begin{cases} \left(\nabla^2 - \dfrac{1}{\varepsilon_0 c^2}\dfrac{\partial^2}{\partial t^2}\varepsilon :\right)\mathrm{e}_1 A_1(x_3)\exp\left[i(\omega t - kx_3)\right] = \dfrac{2}{\varepsilon_0 c^2}\dfrac{\partial^2}{\partial t^2}\chi_\omega : \mathrm{e}_1\mathrm{e}_2 A_1^*(x_3)A_2(x_3)\exp\left[i(\omega t + k_1 x_3 - k_2 x_3)\right] \\ \left(\nabla^2 - \dfrac{1}{\varepsilon_0 c^2}\dfrac{\partial^2}{\partial t^2}\varepsilon :\right)\mathrm{e}_2 A_2(x_3)\exp\left[i(2\omega t - k_2 x_3)\right] = \dfrac{4}{\varepsilon_0 c^2}\dfrac{\partial^2}{\partial t^2}\chi_{2\omega} : \mathrm{e}_1\mathrm{e}_1 A_1^2(x_3)\exp\left[i(2\omega t - 2k_1 x_3)\right]. \end{cases} \tag{6}$$

For simplification we assume that dielectric medium is low-loss and low-nonlinear and neglecting the second derivatives with respect to coordinates of complex envelopes (slow varying amplitude approximation). Then after performing differentiation in (6) we obtain following equation that describe evaluation of first and second harmonics:

$$\begin{cases} \dfrac{dA_1}{dx_3} + \alpha_1 A_1 = -i\sigma_1 A_1^* A_2 \exp\left(-i\Delta k x_3\right), \\ \dfrac{dA_2}{dx_3} + \alpha_2 A_2 = -i\sigma_2 A_1^2 \exp\left(i\Delta k x_3\right). \end{cases}, \tag{7}$$

Here $\Delta k = k_2 - k_1$ is wave detuning and following designations are used:

$$\begin{aligned} \alpha_1 &= k_1 \mathrm{e}_1 \frac{\mathrm{Im}\left(\varepsilon_\omega\right) : \mathrm{e}_1}{2n_\omega^2}, \qquad \alpha_2 = k_2 \mathrm{e}_2 \frac{\mathrm{Im}\left(\varepsilon_{2\omega}\right) : \mathrm{e}_2}{2n_{2\omega}^2}, \\ \sigma_1 &= \frac{1}{2} k_1 \mathrm{e}_1 \frac{\chi_\omega^{(2)} : \mathrm{e}_1 \mathrm{e}_2}{n_\omega^2}, \qquad \sigma_2 = \frac{1}{4} k_2 \mathrm{e}_2 \frac{\chi_{2\omega}^{(2)} : \mathrm{e}_1 \mathrm{e}_1}{n_{2\omega}^2}. \end{aligned}, \tag{8}$$

It can be concluded from the system (7) that SHG process has the highest efficiency when wave detuning Δk is close to zero. As it was mentioned earlier crystal temperature is the key parameter that affects the Δk value because refractive indices n_ω and $n_{2\omega}$ are temperature dependent. In those points of the dielectric media where Δk is nonzero conversion efficiency decreases. Thus besides crystal average temperature it is also important to know temperature distribution inside crystal volume.

3. Three dimensional model of crystal heating

Stationary 3D heat conduction problem have to be solved with heat source, conditioned by absorption of first and second harmonic, in order to obtain 3D temperature distribution of nonlinear-optical crystal in process of SHG.

$$\begin{cases} \kappa_{\mathrm{cr}} \nabla^2 \theta_{\mathrm{cr}} + \alpha(\omega) I\left(x_1, x_2\right) f(x_3) = 0, \\ -\kappa_{\mathrm{cr}} \left.\dfrac{\partial \theta_{\mathrm{cr}}}{\partial n}\right|_{\partial\Gamma} = h^{\mathrm{T}} \left(\theta_{\mathrm{cr}} - \theta_{\mathrm{a}}\right)\Big|_{\partial\Gamma}. \end{cases}, \tag{9}$$

Here κ_{cr} is thermal conductivity of the crystal, $\alpha(\omega)$ is optical absorption coefficient, which depends on frequency; $I(x_1,x_2)$ is radiation intensity transverse distribution; θ_{cr} and θ_{a} are temperatures of the crystal and air respectively; n is the normal vector to the interface; h^{T} is the heat transfer coefficient; $|\partial\Gamma$ denotes values at the interface. Function $f(x_3)$ represents longitudinal dependence of radiation intensity along propagation direction.

Generally speaking systems of equations (7) and (9) are coupled because distributions of harmonic intensities change the crystal temperature distribution and the last one in turn affects the harmonic intensities through the wave detuning.

System (9) can be solved employing variational principle. It states that for equation type $\hat{L}u(x_1,x_2,x_3) = F(x_1,x_2,x_3)$, where $\hat{L}$ is differential operator, which is real, linear, self-conjugate and positive-definite, the solution in region D is equivalent to such function $u(x_1,x_2,x_3)$ that minimizes the following functional [10, 11]

$$\Phi(u) = \left\langle \hat{L}u, u \right\rangle - 2\left\langle u, F \right\rangle. \tag{10}$$

In our case $\hat{L} = -\nabla^2$ and function $u(x_1,x_2,x_3)$ represents θ_{cr} distribution. Triangular brackets denote the scalar product of square-integrable functions ξ in region D:

$$\left\langle \xi_i, \xi_j \right\rangle = \iiint_{\mathrm{D}} \xi_i(x_1,x_2,x_3)\xi_j(x_1,x_2,x_3)dx_1dx_2dx_3. \tag{11}$$

Rayleigh-Ritz method is popular and convenient approach for solving differential equations. For minimization of (10) we select basis functions $\{\psi\}$ that produce full set in D region. For simplicity of

calculations we chose basis functions that satisfy boundary conditions of the given problem (9). Then unknown function $\theta_{cr}(x_1, x_2, x_3)$ is expanded using selected basis functions.

$$\theta_{cr}(x_1, x_2, x_3) = \theta_a + \sum_i a_i \psi_i \,. \tag{12}$$

Here a_i are expansion coefficients. Air temperature θ_a is constant and for simplification of basis function type it can be treated separately.

Substituting (12) into (10) and using condition of minimum for function of several variables, which is equality to zero of function derivatives with respect to variables at the point of minimum, we obtain so-called Rayleigh system:

$$\left\langle -\nabla^2 \psi_i, \psi_j \right\rangle a_j = \frac{\alpha(\omega)}{\kappa_{cr}} \left\langle I(x_1, x_2) f(x_3), \psi_i \right\rangle . \tag{13}$$

In case explicit forms of basis functions and distributed heat source are known then scalar products can be calculated. As follows equation (13) becomes system of linear equations with unknown expansion coefficients a_j. So that it can be solved using standard methods of linear algebra.

4. Experimental results

Experiment of crystal equivalent temperature measurement in course of SHG was conducted using nonlinear-optical periodically poled lithium niobate (PPLN). Crystal is a rectangular parallelepiped with dimensions 2.6×1×4.9 mm^3, polled along x_2. Block-scheme of experimental setup used for observation of piezoelectric resonances in crystals is introduced in Figure 1 (a). PPLN crystal is placed inside the capacitor, formed by two strip electrodes. Voltage from the radiofrequency (RF) generator is applied to this capacitor. Voltage frequency f is swept while its low value amplitude is fixed. Crystal response is analyzed using lock-in amplifier by measuring voltage U_R on the load resistor R, which is connected in series with the capacitor.

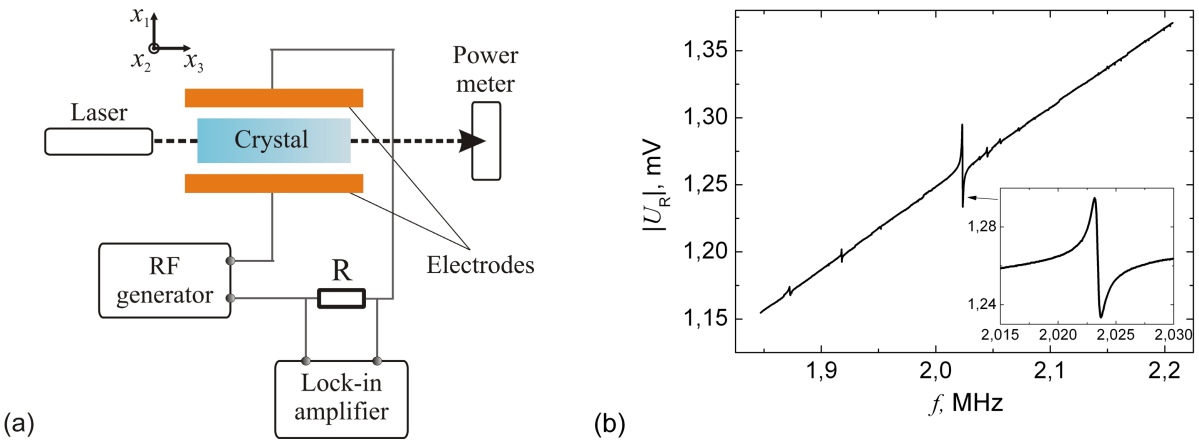

Figure 1. (a) Simplified block-scheme of experimental setup; (b) Frequency dependence of the voltage amplitude measured on load resistor R for PPLN crystal, inset shows dependence of the response near PPLN piezoelectric resonance.

Piezoelectric resonances are clearly observed in measured spectrum. Piezoelectric resonance frequencies Rf_n are precisely measured due to high quality factors. Piezoelectric resonance thermal coefficients K_n^{prt} are determined by measuring linear resonance frequency shift with temperature in crystal uniform heating calibration experiment [2, 3]:

$$\Delta Rf_n = K_n^{prt} \Delta\theta_{cr} \,. \tag{14}$$

When laser radiation goes through the crystal its nonuniform temperature distribution is characterized by *crystal equivalent temperature* Θ_{eq}. This temperature is obtained directly by measuring resonance

frequency shift, caused by influencing laser power, in respect to the frequency at crystal initial temperature θ_{cr}^{0} at zero power.

$$\Theta_{eq}(P_0) = \theta_{cr}^{0} + \frac{\Delta R f_n(P_0)}{K_n^{prt}}. \quad (15)$$

Experiment of SHG in PPLN crystal was performed using as a pump source the continuous wave polarized single-mode ytterbium fiber laser (λ=1064 nm) [7, 8]. Figure 2 a shows dependence of crystal equivalent temperature on pump power measured for two cases of pump polarization e_1 direction. When $e_1 \| x_1$ second harmonic is not generated and crystal is linearly heated with pump power with 1.35 K/W rate. For the $e_1 \| x_2$ case at first PPLN equivalent temperature linearly changes with pump power when second harmonic power P_{sh} is low, i.e. wave detuning Δk is far from zero. However in this case substantial nonlinear behavior of PPLN equivalent temperature is observed at phase matching when second harmonic generation efficiency is much higher.

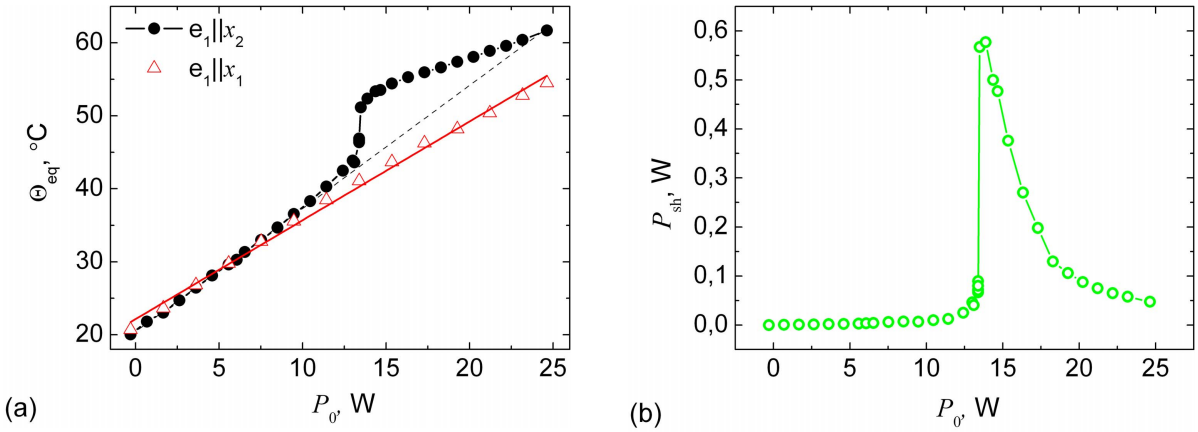

Figure 2. Dependencies on pump power of PPLN crystal equivalent temperature for two cases of pump polarization e_1 directions (a) and generated second harmonic power for the $e_1 \| x_2$ case (b).

5. Calculation results

For modeling temperature distribution of nonlinear-optical crystal, heated by laser radiation, the convective heat exchange at crystal ends and lateral surfaces should be taken as boundary conditions for solving heat conduction problem (9). This is due to experimental procedure of crystal equivalent temperature determination that supposes unclamped boundaries of the crystal.

For single-mode beam with radius r and power P_0 laser radiation intensity has Gaussian profile of transverse distribution

$$I(x_1, x_2) = \frac{2P_0}{\pi r^2} \exp\left(-2\frac{x_1^2 + x_2^2}{r^2}\right), \quad (16)$$

Estimation of temperature effects will be considered for two different cases of longitudinal dependencies of laser intensity:

$$f_1(x_3) = \exp(-\alpha x_3), \quad f_2(x_3) = \left(\frac{x_3}{L_3}\right)^2 \exp(-\alpha x_3), \quad (17)$$

Here L_3 is crystal dimension along x_3, and P_0 is pump laser power. First function describes exponential decay of pump radiation in the absence of second harmonic and another one is the solution of (7) in assumptions of $\Delta k = 0$ and low conversion degree.

Also for simplicity we shall seek the solution of (9) in the cuboid crystal with equal dimensions along x_1 and x_2 axes, i.e. $L_1=L_2\equiv L$, L_3 dimension along x_3 direction and assume $\alpha_1=\alpha_2\equiv\alpha$. Taking into account boundary conditions and symmetry of the problem it is convenient to use following basis:

$$\psi_{mnl}=\left\{\left[\left(\frac{x_1}{L}\right)^{2m}+\frac{2m}{\mu L}-1\right]\left[\left(\frac{x_2}{L}\right)^{2n}+\frac{2n}{\mu L}-1\right]+\left[\left(\frac{x_1}{L}\right)^{2n}+\frac{2n}{\mu L}-1\right]\left[\left(\frac{x_2}{L}\right)^{2m}+\frac{2m}{\mu L}-1\right]\right\}\cdot$$
$$\cdot\begin{bmatrix}\left(\frac{x_3}{L_3}\right)^{l}+\frac{x_3}{L_3}\frac{l-\mu L_3}{\mu L_3-1} & \text{for } l \text{ odd}\\ \left(\frac{x_3}{L_3}\right)^{l}+\frac{l}{\mu L}-1 & \text{for } l \text{ even}\end{bmatrix} \quad . \qquad (18)$$

Here $\mu=h^{\mathrm{T}}/\kappa_{\mathrm{cr}}$ and total degree $2m+2n+l$ of each basis function does not exceed N. For calculations it was chosen N=12. Each polynomial in (18) satisfies boundary conditions at all crystal facets and due to symmetry doesn't depend on permutation of subscripts m and n. Functions contain polynomials only with even degree of x_1 and x_2 due to the symmetry in respect to x_1x_3 and x_2x_3 planes (see figure 1).

System (13) was solved using following parameters of lithium niobate crystal: L=2 mm; L_3=10 mm; κ_{cr}=4.6 $\mathrm{WK^{-1}m^{-1}}$; r=0.5 mm; P_0=3 W; $\alpha=10^{-2}$ $\mathrm{cm^{-1}}$; h^{T}=43 $\mathrm{Wm^{-2}K^{-1}}$, θ_{a}=0 °C. Calculated temperature distributions at lateral facet of the PPLN are shown in figure 3 for two cases given by (17).

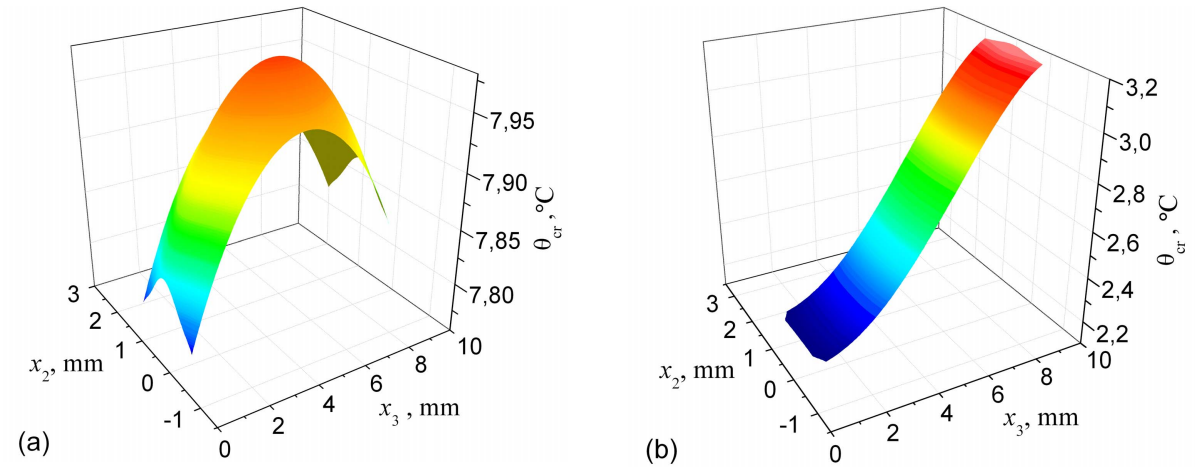

Figure 3. Calculated temperature distributions at lateral facet (x_2x_3) of the PPLN crystal for exponential $f_1(x_3)$ (a) and quadratic $f_2(x_3)$ (b) heat source distributions.

It can be seen that from figure 3 (a) that longitudinal temperature distribution is not monotonous for the case of intensity exponential decay. At both ends of the crystal the convective heat exchange occurs resulting in lower temperature at crystal ends in comparison with its centre as far as heat source is almost independent on x_3 coordinate. Here maximum temperature difference inside crystal is small (about 0.2 °C) in comparison with crystal average heating.

However if intensity growth is parabolic (see figure 3 (b)) the temperature field distribution differs from previous case. Maximum temperature difference in transverse direction is low. Still in longitudinal direction it is comparable with crystal average temperature rise. Calculations also reveal that in this case crystal temperature change has rather linear tendency along propagation direction of light (x_3) because thermal conductivity somehow flattens temperature distribution. It means that thermal effects influence on SHG process can be estimated using linear approximation of crystal longitudinal temperature change.

It should be notified that system (9) is linearly dependent on laser power P_0 and optical absorption coefficient $\alpha(\omega)$, which is assumed to be small enough. Thereby temperature distribution inside crystal

linearly depends on variation of these parameters, i.e. absolute values of crystal temperature alter while relative temperature distribution remains unchanged.

Figure 4 shows first and second harmonic power distributions along crystal length as a result of numerical calculation of system (7). Figure 4 (a) represents ideal case of uniform crystal temperature distribution and wave detuning Δk=0.

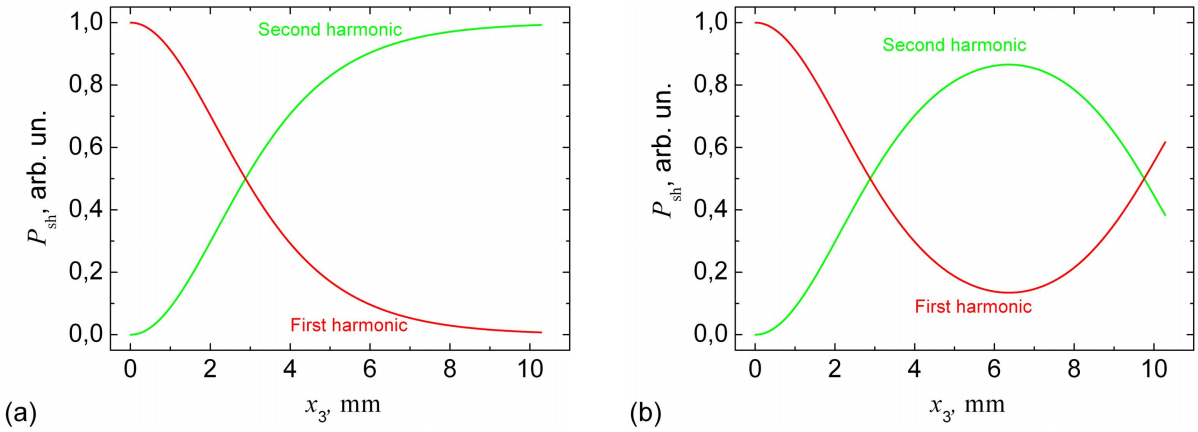

Figure 4. Calculated distributions of harmonics power along PPLN length for the case of uniform temperature distribution and zero wave detuning (a), for the case of linear temperature gradient 1°C/cm along crystal length (b).

Results, shown in figure 4 (b), are obtained when linear longitudinal temperature gradient inside crystal is considered. Here wave detuning difference from zero $\Delta k \neq 0$ is caused by temperature difference of 1°C between the input and output crystal facets. In the beginning conversion process is similar to the ideal case but at some crystal length the process of inverse energy conversion from second harmonic to the first one initiates due to Δk distinction from zero. It is clear that second harmonic conversion efficiency strongly decreases longitudinal temperature gradient inside crystal is presented.

6. Discussion and conclusions

Three dimensional temperature distribution inside nonlinear-optical crystal was calculated for two different cases of laser intensity longitudinal dependence. First one is exponential decay and the second is quadratic rise of intensity along crystal length. Figure 3 (a) represents nonuniform crystal temperature distribution when second harmonic is not generated. In turn figure 3 (b) corresponds to crystal heating in course of second harmonic generation assuming uniform heating due to absorption of pump radiation. It is revealed that in case of absorption alone the crystal maximal temperature difference is much less than its average temperature change. However, in course of SHG the transverse temperature gradient of the crystal remains small. And yet longitudinal temperature difference is high enough and can be of the same order as crystal average temperature rise. This is important because even small temperature difference of 1°C along crystal length (10 mm) may substantially impede nonlinear-optical conversion process.

Two situations introduced in figure 3 (a) and (b) can be easily realized in experiment by variation of single parameter that is laser polarization direction. PPLN crystal equivalent temperature change with laser power measured for two orthogonal directions of pump polarization $e_1 \| x_1$ and $e_1 \| x_2$ (see figure 2) reveals instances with absence of SHG and presence of SHG respectively. Crystal equivalent temperature linearly depends on pump power when $e_1 \| x_1$ (no SHG) and calculated crystal temperature is almost uniform in this case. However in course of efficient SHG when $e_1 \| x_2$ the equivalent temperature nonlinearly changes with pump power and corresponding longitudinal (along x_3) temperature distribution is substantially nonuniform. Thus experimentally measured equivalent temperature of the crystal heated by laser radiation is some sort of average crystal temperature.

Evidently averaging depends on piezoelectric resonance selection. Model of crystal equivalent temperature for the case of uniform temperature distribution has been already introduced [4]. For accurate description of crystal real temperature averaging in course of SHG it is necessary to elaborate model of piezoelectric resonance frequency shift under strong longitudinal temperature gradient inside crystal. Besides problems of SHG (7) and crystal heating (9) should be solved conjointly. These are the goals of future investigation.

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