

Self-Action of Second Harmonic Generation and Longitudinal Temperature Gradient in Nonlinear-Optical Crystals

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Abstract. Model of second harmonic generation with thermal self-action was developed. Second harmonic generation temperature phase matching curves were measured and calculated for periodically polled lithium niobate crystal. Both experimental and calculated data show asymmetrical shift of temperature tuning curves with pump power.

1. Introduction

Despite rapid development of laser physics and quantum optics in last decades, problem of generation spectrum broadening of high-power coherent radiation sources is still vitally important in nowadays. One of the most efficient ways to deal with this issue is application of nonlinear-optical crystals for conversion of pump laser radiation into multiple harmonics [1]. Requirement of coherence conservation specifies fulfillment of phase matching condition for pump and generated photons along nonlinear-optical crystal length [2]. Wave detuning from phase matching is linear function of refractive indices of interacting harmonics. Crystal temperature change in course of laser frequency conversion is the main factor responsible for refractive indices variations. As follows, accurate and noncontact methods of nonlinear-optical crystal temperature control are needed in case of high-power (tens or hundreds Watts) pump radiation nonlinear conversion [3].

In present paper we introduce novel method for noncontact temperature measurement of nonlinear-optical crystals during its interaction with laser radiation. Here radiofrequency (RF) impedance spectroscopy plays the key role. As every nonlinear-optical crystal possess piezoelectric properties then its response to the applied ac electric field strongly varies at frequencies that correspond to internal vibration modes of the sample. It is well known that in first approximation piezoelectric resonance frequencies linearly depend on temperature. Earlier we introduced application of impedance spectroscopy technique for precise temperature measurement of crystal heated by laser radiation [4]. Experimental determination of equivalent temperature of nonlinear-optical crystal in course of laser radiation frequency conversion was also demonstrated [5]. Moreover piezoelectric laser calorimetry was successfully applied for determination of optical absorption coefficients of crystals in wide spectral range [6, 7].

We developed mathematical model that proves validity of equivalent temperature concept, which suggests direct temperature measurement of nonuniformly heated crystal using its temperature calibrated piezoelectric resonances [8]. Temperature calibration of resonance frequencies is performed



in uniform equilibrium heating conditions of the crystal. However, in initial theoretical approach, used for determination of resonance frequencies temperature dependence, just two-dimensional temperature distribution in crystal cross-section was considered. Temperature gradient along radiation propagation direction (third dimension) was assumed to be negligibly small due to very low optical absorption. In some experimental cases such simplification can be rough enough and definitely it cannot be relied on when considering nonlinear-optical processes of laser radiation frequency conversion. This is evident in case optical absorption coefficient at generated frequency is much higher than that at the pump frequency. For correct interpretation of obtained experimental data the 3D heat conduction problem with distributed heat source, conditioned by energy absorption of two light waves, ought to be solved. It should be taken into account that these waves have different values of optical absorption coefficient and besides its powers change along crystal length.

2. Theoretical model

For the interpretation of our second harmonic generation (SHG) experiments three physical processes should be considered. These are second harmonic generation, non-uniform crystal heating due to optical absorption, and piezoelectric (acoustic) vibrations of the crystal. However, as crystal vibrates in different frequency domain compared to frequencies of interacting light waves, only first two processes significantly affect each other. In addition, variation of crystal optical properties caused by its acoustic vibrations is negligibly small. So that it will be sufficient to consider optical second harmonic generation together with accompanying thermal effects and independently characteristic piezoelectric vibrations of the crystal.

2.1. Coupled equations for second harmonic generation and crystal laser heating.

Three-dimensional temperature distribution of the nonlinear-optical crystal in process of second-harmonic generation can be obtained by solving equations that describe nonlinear conversion process. We shall consider a dielectric medium with a non-zero component of the quadratic optical susceptibility. When electromagnetic waves with electric field strength E propagate in such dielectric a nonlinear polarization arises:

$$P_{NL} = \epsilon_0 \chi^{(2)} EE. \quad (1)$$

Here ϵ_0 – dielectric permittivity of vacuum, and $\chi^{(2)}$ - quadratic susceptibility tensor. If we consider interaction of both first and second harmonic waves, then total electric field inside crystal can be represented as:

$$E(x_3, t) = \frac{1}{2} e_p^1 [A_1(x_3, t) \exp(j(\omega t - kx_3)) + c.c.] + \frac{1}{2} e_p^2 [A_2(x_3, t) \exp(j(2\omega t - Kx_3)) + c.c.]. \quad (2)$$

Here, A_1 and A_2 are complex envelopes of the first and the second harmonic respectively, k and K are corresponding wave vectors $k = n_\omega \omega / c$, $K = n_{2\omega} 2\omega / c$, e_p^1 and e_p^2 - unit vectors specifying directions of polarizations of the interacting waves. By substituting equation (1) into (2) we can obtain components of the nonlinear polarization oscillating at the fundamental and doubled frequencies. Equation describing the propagation of electromagnetic waves in a nonlinear dielectric can be obtained using Maxwell equations:

$$\nabla^2 E - \frac{1}{\epsilon_0 c^2} \frac{\partial^2 D}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 P_{NL}}{\partial t^2}. \quad (3)$$

Here $D = \epsilon_0 (1 + \chi^{(1)}) E$ is electric displacement vector, c is speed of light. Performing differentiation with respect to space and time, in assumption that medium is both weakly nonlinear and low absorbing, and also neglecting the second derivatives of complex envelopes we can obtain following equation that describe interaction between the first and second harmonics [2]:

$$\begin{cases} \frac{dA_1}{dx_3} + \alpha_1 A_1 = -j\sigma_1 A_1^* A_2 \exp(-j\Delta k x_3), \\ \frac{dA_2}{dx_3} + \alpha_2 A_2 = -j\sigma_2 A_1^2 \exp(j\Delta k x_3). \end{cases} \quad (4)$$

Here value $\Delta k = K - k = 2\omega n_{2\omega}/c - \omega n_{\omega}/c$ is so-called wave detuning. Following designations were used:

$$\begin{aligned} \alpha_1 &= k e_p^1 \frac{\text{Im}(\varepsilon(\omega)) : e_p^1}{2n_{\omega}^2}, & \alpha_2 &= K e_p^2 \frac{\text{Im}(\varepsilon(2\omega)) : e_p^2}{2n_{2\omega}^2}, \\ \sigma_1 &= \frac{1}{2} k e_p^1 \frac{\chi^{(2)}(\omega) : e_p^1 e_p^2}{n_{\omega}^2}, & \sigma_2 &= \frac{1}{4} K e_p^2 \frac{\chi^{(2)}(2\omega) : e_p^1 e_p^1}{n_{2\omega}^2}. \end{aligned} \quad (5)$$

From equation (4) we can conclude that process of laser radiation nonlinear conversion occurs most efficiently when Δk value equals to zero. And in those regions where Δk is nonzero nonlinear conversion efficiency decreases due to nonzero phase difference between interacting waves. As it is mentioned above, the temperature of the crystal can strongly affect wave detuning, since the refractive index of these waves depend on crystal temperature. Thus for adequate modelling it is important to consider not only the "average" crystal temperature but also its distribution in 3D space.

In order to calculate three-dimensional temperature distribution inside nonlinear-optical crystal in process of SHG it is necessary to solve three-dimensional stationary heat conduction problem with a heat source represented by absorption of the first and second harmonic energy.

$$\begin{cases} \kappa_{cr} \nabla^2 \theta_{cr} + \alpha_1 I_{\omega}(x_1, x_2, x_3) + \alpha_2 I_{2\omega}(x_1, x_2, x_3) = 0, \\ -\kappa_{cr} \frac{\partial \theta_{cr}}{\partial n} \Big|_{\partial \Gamma} = h^T (\theta_{cr} - \theta_{air}) \Big|_{\partial \Gamma}. \end{cases} \quad (6)$$

Here κ_{cr} is thermal conductivity of the crystal, $I(x_1, x_2, x_3)$ is radiation intensity distribution; θ_{cr} and θ_{air} are temperatures of the crystal and air respectively; n is the normal vector to the crystal interface; h^T is heat transfer coefficient; $|\partial \Gamma$ denotes values at the interface.

To solve separately system of equations (6), one can use the Rayleigh-Ritz variation principle. It allows us to search extremum of the specific functional instead of solving differential equations directly [9, 10]. According to variation principle the solution is sought in the form of a linear combination of basis functions, which in our case can be represented by following polynomials:

$$\psi_{m,n,l} = \left(\left(\frac{x_1}{L} \right)^{2m} + \frac{2m}{\xi L} - 1 \right) * \left(\left(\frac{x_2}{L} \right)^{2n} + \frac{2n}{\xi L} - 1 \right) * \frac{\left(\left(\frac{x_3}{L_3} \right)^l + \frac{x_3}{L_3} \frac{l - \xi L_3}{\xi L_3 - 1} \right), \text{for } l \text{ odd}}{\left(\left(\frac{x_3}{L_3} \right)^l + \frac{l}{\xi L} - 1 \right), \text{for } l \text{ even}}. \quad (7)$$

Here $\xi = h^T / \kappa_{cr}$. Degree of each basis function $2m+2n+l$ does not exceed the natural number N that was set equal to 12. Every polynomial in this set satisfies boundary conditions at every facet of the crystal. In more details derivation of equations (4) and approach for solving the three-dimensional heat conduction problem (6) was introduced in our previous paper [11]. Intensities of first and second harmonic, as well as the length of the wave detuning are unknowns here. It was demonstrated that for second harmonic generation the longitudinal temperature gradient is much more substantial than transvers one. It means that the last one can be neglected. General physical considerations suggest that there is certain equilibrium state of the system when overheating of the crystal is compensated by second harmonic power decrease, conditioned by increase of phase detuning. Thus, systems (4) and (6) can be solved using iterative procedure where the calculated solutions of one system are taken as

input parameters for another one. It was ascertained that several dozens of iterations are sufficient to obtain consistent solution.

2.2. Calculation of eigenfrequencies of piezoelectric crystals

Internal acoustic vibration spectrum of any elastic body can be calculated using Hamilton's principle of least action. Vibration pattern and eigenfrequencies of crystal modes are obtained by minimization of Lagrangian L of the system [12, 13]. When we deal with piezoelectric material other terms should be included into Lagrangian functional. These are electric and piezoelectric parts of potential energy [14, 15]

$$L = \iiint \left(\frac{1}{2} \rho \dot{u}_i^2 - \frac{1}{8} c_{ijkl} (u_{i,j} + u_{j,i}) (u_{k,l} + u_{l,k}) - \frac{1}{2} e_{ijk} \varphi_{,i} (u_{j,k} + u_{k,j}) + \frac{1}{2} \varepsilon_{ij} \varphi_{,i} \varphi_{,j} \right) d\Omega. \quad (8)$$

Here $F_{,j} = \partial F / \partial x_j$ denotes derivative; ρ is density of the crystal; $u_i(x_1, x_2, x_3)$ are the components of mechanical displacement of the sample points; c_{ijkl} , d_{ijk} , ε_{ij} are elastic, piezoelectric and dielectric tensors respectively; φ is electric potential; Ω is the crystal volume. Lagrangian minimization can be performed using variation principle and the Rayleigh-Ritz approximation method. However, in case crystal is in the form of rectangular parallelepiped ($L_x \times L_y \times L_z$) it is more convenient to use other kind of expansion functions ψ_m - normalized Legendre polynomials $P_m(x, y, z)$ [14, 15]:

$$\psi_m(x, y, z) = \left(\frac{(2m_x + 1)(2m_y + 1)(2m_z + 1)}{L_x L_y L_z} \right)^{1/2} P_{m_x} \left(\frac{2x}{L_x} \right) P_{m_y} \left(\frac{2y}{L_y} \right) P_{m_z} \left(\frac{2z}{L_z} \right). \quad (9)$$

Applying separation of variables, every basis function can be represented as a product of Legendre polynomials each depending on the individual coordinate. Thus, we can expand unknown functions, which are three shift functions in different directions and electric potential function. By substituting these expansions into Lagrangian, variation integral problem is reduced to eigenvalue problem of specific matrix. Eigenvectors give values of corresponding expansion coefficients and the eigenvalues represent piezoelectric resonance frequencies. Figure 1 shows calculation results of typical resonance mode vibration amplitude distribution.

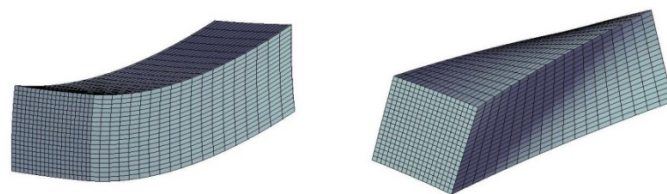


Figure 1. Calculated amplitude distribution of two piezoelectric resonances of the crystal

3. Second harmonic generation in PPLN crystal

Block scheme of experimental setup for crystal piezoelectric resonance measurement in course of second harmonic generation is shown in Fig.2. Periodically polled lithium niobate crystal (PPLN) in the form of rectangular parallelepiped with dimensions $4.9 \times 2.6 \times 1$ mm³ was used in experiments. Polling axis was directed along 1 mm direction.

Crystal is placed inside capacitor that is connected in series with load resistor R. Crystal response to the applied ac voltage from RF generator is analyzed in wide radiofrequency range by measuring voltage on R via lock-in amplifier. Both capacitor plates are made of sets of interconnected metallic pins allocated at the radial arc. Such configuration helps to minimize uncontrollable heating of capacitor plates caused by absorption of scattered radiation. Piezoelectric thermoresonators, made of quartz crystals with small cross-section, are placed at certain distance above and under the PPLN crystal in order to control air temperature.

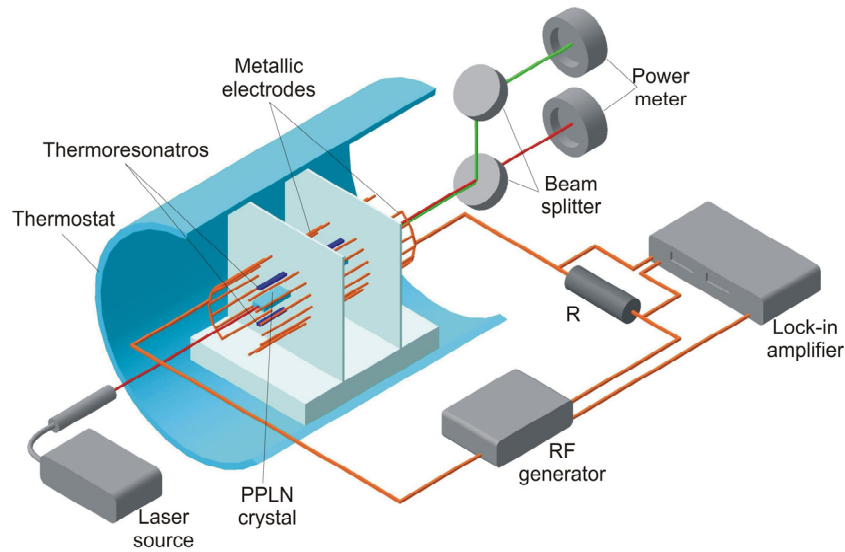


Figure 2. Block scheme of experimental setup

Temperature calibration of piezoelectric resonance frequencies $Rf_i(T)$ was performed when crystal was uniformly heated. Thus piezoelectric resonance thermal coefficients K_i^{prt} were determined. As follows crystal equivalent temperature change due to heating by laser radiation of P power can be obtained by measuring resonance frequency shift (see Ref. [4] for details):

$$\Delta\Theta_{\text{eq}} = \Delta Rf_i(P) / K_i^{\text{prt}}. \quad (10)$$

Equivalent temperature can be regarded as the true crystal temperature because in this case crystal itself acts as thermal probe.

SHG experiments were carried out using CW single-mode polarized ytterbium fiber laser ($\lambda=1064$ nm, $\Delta\lambda=0.1$ nm) with up to 25 W output power as a pump source. Phase matching equivalent temperature curves of PPLN were measured at fixed pump levels when wave detuning value between first and second harmonic was changed via thermostat temperature adjusting (see Fig. 3).

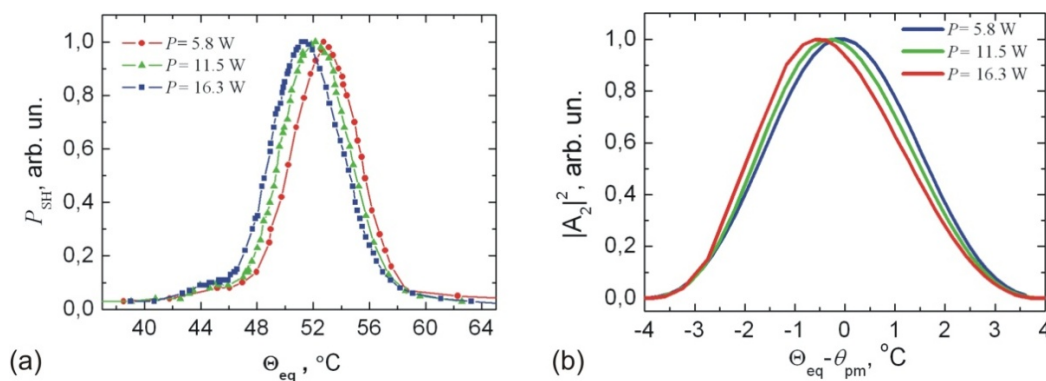


Figure 3 Measured (a) and calculated (b) equivalent temperature tuning curves of PPLN crystal at different pump levels.

Phase matching temperature that corresponds to maximum second harmonic output power P_{SH} shifts to lower temperatures with power $d\theta_{\text{pm}}/dP = -0.11$ °C/W. Experimental results reveal that symmetry distortion of temperature tuning curves occurs at higher pump levels. Calculated curves exhibit the same behavior. The main reason for this is presence of the longitudinal temperature gradient inside crystal. However, characteristic width $\Delta\theta_{\text{pm}}=6$ °C of the curve remains almost unchanged.

4. Conclusions

In present paper we have introduced theoretical approach that supposes consideration of SHG problem in terms of thermal self-action. Predicted by this model shift and distortion of temperature tuning curves with pump power were verified experimentally. Further development of mathematical model of nonlinear-optical crystal equivalent temperature in course of SHG should include consideration of piezoelectric (acoustical) vibrations of 3D solid in condition of nonuniform transverse and longitudinal temperature distribution.

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