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Model of resonant acoustic spectroscopy of interaction of high power single-mode laser radiation with crystals

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Abstract. Every nonlinear optical laser frequency conversion process is accompanied by the crystal heating due to the absorption of some part of the radiation energy. Optical absorption coefficients and internal crystal temperature distribution during interaction with the high-power laser radiation are directly determined from modelling the experimentally obtained stationary and kinetics data of the radiation-induced frequency shifts of the crystal piezoelectric resonances.

1. Introduction
Any process of nonlinear frequency conversion or parametric oscillation of the high-power laser radiation leads to the crystal temperature increase. Heating of the nonlinear-optical crystal is caused by both linear and nonlinear absorption of laser radiation. Optical calorimetry is the most widespread method used to measure absorptance of a crystal. Usually it exploits weak optical powers and determines the temperature of the sample surface using various temperature measuring devices. We introduce a method for the determination of the internal crystal temperature during its interaction with high-power laser radiation. It includes both the analysis of the piezoelectric resonances frequency shifts, which are related to the equivalent temperature of the crystal, and the numerical heating model, that allows us to determine real crystal temperature distribution from the measured kinetics of the piezoelectric resonance frequency.

2. Piezoelectric resonance modelling
Every nonlinear optical crystal sample has its own set of the intrinsic acoustic vibration modes. Well-established procedure based on the Rayleigh-Ritz method [1] enables one to calculate these eigenmodes for the certain temperature provided that the elastic constants of the crystal are known at this temperature.

Free vibrations of the nonlinear-optical KTiOPO4 (KTP) crystal, which is assumed to be a rectangular parallelepiped with the dimensions $L_x \times L_y \times L_z$ ($L_y = L_z = 3$ mm, $L_x = 30$ mm), are considered. Crystallographic axes are parallel to the parallelepiped edges: crystal $c$-axis is parallel to $z$, crystal $a$, $b$ axes are in $x$-$y$ plane. When the external electric field of the radiofrequency $\omega_{rf}$ is applied to the crystal, it can excite some of its intrinsic vibration modes, provided that $\omega_{rf}$ coincides with one
of the crystal eigenfrequencies. Equations defining these eigenfrequencies can be obtained by searching the stationary point of the Lagrangian:

\[
L_0 = \iiint \left( \frac{1}{4} c_{ijkl} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \left( \frac{\partial u_l}{\partial x_k} + \frac{\partial u_k}{\partial x_l} \right) + 2 e_{ijk} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \frac{\partial \rho}{\partial x_k} - E_{ij} \frac{\partial \phi}{\partial x_j} \frac{\partial \phi}{\partial x_i} - \rho \omega_{rf}^2 \sum_{i} u_i^2 \right) dV \tag{2.1}
\]

This Lagrangian can be found in literature, for example, in [2]. It incorporates elastic energy (first term), piezoelectric and dielectric as well as kinetic energy. \( u_i \) is the mechanical displacement vector as function of spatial coordinates, \( \phi \) is the electric potential. Analytical solution of this task cannot be obtained [1]. To obtain a numerical solution, one should choose a set of the basis functions \( \psi_p \) and expand unknown displacement and potential into this set.

\[
u_i = C^i_p \psi_p, \quad \phi = C^\phi_p \psi_p, \quad i = 1, 2, 3, \quad p = (p_1, p_2, p_3)
\tag{2.2}
\]

After substituting expansions (2.2) Lagrangian (2.1) takes the form

\[
L_0 = C^i_p C^\phi_p \Gamma_{pq} + 2C^i_p C^\phi_p \Pi_{pq} - C^i_p C^\phi_p D_{pq} - \rho \omega_{rf}^2 C^i_p C^\phi_p I_{pq}
\tag{2.3}
\]

where

\[
\Gamma_{pq} = \iiint c_{ijkl} \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_i} dV, \quad \Pi_{pq} = \iiint e_{ijl} \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_i} dV, \quad D_{pq} = \iiint e_{ijl} \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_i} dV, \quad I_{pq} = \iiint \psi_p \psi_q dV.
\tag{2.4}
\]

When the temperature and the other external parameters are held constant, all material parameters \( (c_{ijkl}, e_{ijkl}, \epsilon_{ij}, \rho) \) are independent on the coordinates, and they can be taken outside the integral sign in (2.4). Equations for the eigenmodes are obtained by differentiating (2.3) with respect to the unknown coefficients \( C^i_p, C^\phi_p \). This leads to the following equations:

\[
\begin{cases}
\left( \Gamma - \Pi D^{-3} \Pi^T \right) C^{1-3} = \rho \omega_{rf}^2 I C^{1-3} \\
C^\phi = -D^{-3} \Pi C^{1-3}
\end{cases}
\tag{2.5}
\]

This is the generalized eigenvalue problem. From (2.5) it can be seen that the piezoelectric effect leads to the correction of the \( \omega_{rf} \). For the KTP crystal we have \( c \sim 10^{11} \text{Pa}, e \sim 1 \text{C/m}^2, \epsilon \sim 15 \epsilon_0 \). The value of this correction is estimated as follows: \[ \| \Pi D^{-3} \Pi^T \| / \| \Gamma \| \sim e^2 / (\epsilon \cdot c) \] and is about 7%.

One of the most convenient bases is the set of the Legendre polynomials. More specifically,

\[
\psi_p(x, y, z) = \psi_{p_1}(x) \psi_{p_2}(y) \psi_{p_3}(z)
\]

\[
\psi_{p_1} = \sqrt{\frac{2p_1 + 1}{L_x} \frac{2x}{L_x}}
\tag{2.6}
\]

From the properties of the Legendre polynomials it follows that matrix \( I \) is unity matrix. In the Fig. 1 an example of the eigenmode calculation for the KTP crystal is shown.

Fig. 1. Calculated eigenmode of the KTP crystal sample for the \( f_{t0} = 832.5 \text{ kHz} \) piezoelectric resonance (\( T_0 = 294K \)).

3. Resonance frequency shift at homogeneous heating.
Elastic constants of any material depend on its temperature. So, if the temperature of the sample changes, its resonance frequencies shift. The major contribution to the frequency shift is produced by the elastic constants. Small temperature shift can be treated as the disturbance of the initial system. Let’s denote perturbation operator (matrix) as \( \delta \Gamma \). From the first-order stationary perturbation theory
we can obtain correction $\delta f_r$ to the resonance frequency $f_r$ (and the corresponding eigenvector $C_r$) as follows:

$$\delta f_r = \frac{C_r^T \delta \Gamma C_r}{8\pi^2 \rho f_r^2 C_r^T IC_r} \quad (3.1)$$

The frequency shifts of the KTP piezoelectric resonances were measured experimentally during both uniform heating without laser radiation and the non-uniform heating caused by the interaction with single-mode high-power laser radiation. Crystal was placed between two electrodes in unclamped manner (a 100 mkm layer of air is present between the crystal and electrodes). More detailed description of the experiment can be found in [3] or in the other ICPPP 15 conference proceedings of the corresponding authors accepted for the publication in the Journal of Physics: Conference Series.

An example of the calculation of the piezoelectric resonance frequencies and its temperature coefficients is presented for our KTP crystal in the Table 1 together with the experimental results obtained for the uniform heating. As it can be seen from the Table 1, average frequency error is about 0.2%. This accuracy is better than the estimated earlier 7% piezoelectric contribution. Thus piezoelectric and dielectric terms in (2.1) by all means should be taken into account.

Table 1. Piezoelectric resonance frequencies and temperature coefficients $df_r/dT$ for the KTP.

| Resonance number | Resonance frequency $f_{THz}$ kHz | Temperature coefficient $df_r/dT$, Hz/K |
|------------------|----------------------------------|--------------------------------------|
|                  | Calculated | Measured | Calculated | Measured |
| 1                | 808.32     | 808.5     | -56.6      | -50.6     |
| 2                | 808.32     | 810.5     | -53.4      | -50.7     |
| 3                | 808.32     | 817.4     | -40.7      | -52.3     |
| 4                | 808.32     | 823.9     | -55.6      | -52.5     |
| 5                | 808.32     | 831.7     | -56.2      | -54.8     |
| 6                | 808.32     | 832.5     | -58.3      | -53.5     |

The average frequency shift is $\left( \frac{1}{f} \frac{df_T}{dT} \right)_{elast} \approx 7 \cdot 10^{-5} K^{-1}$, while frequency shift due to the thermal expansion is $\left( \frac{1}{f} \frac{df}{dT} \right)_{exp} \approx \frac{1}{2} \alpha_T = 4 \cdot 10^{-6} K^{-1}$, where $\alpha_T$ is the thermal expansion coefficient of the KTP crystal. So the effect of thermal expansion can indeed be neglected.

4. **Inhomogeneous crystal heating by the laser radiation**

When the KTP crystal is heated by the laser radiation the non-uniform temperature distribution inside the crystal volume can be also determined exploiting piezoelectric resonances. In our experiment laser source was ytterbium CW polarized single-mode fiber laser operating at the wavelength $\lambda = 1064$ nm with the laser power up to 14 W and the beam diameter 1 mm. Radiation propagates inside the crystal along its $x$-axis. It is assumed that convective heat transfer takes place on the outer sides of the crystals:

$$\kappa \frac{dT}{dn} + \mu (T - T_e) = 0 \quad (4.1)$$
Here $\mu$ is the heat transfer coefficient, $\kappa$ – thermal conductivity; $T_a$ is the air temperature near the outer crystal facets. Heat conduction equation in the presence of the laser radiation of the $I(x,y,z)$ intensity ($\omega$ is the optical frequency) can be written as follows:

$$\rho C_{sp} \left( \frac{\partial T}{\partial t} \right) = \kappa \Delta T - \frac{\partial I^\omega(x,y,z)}{\partial x}$$  \hspace{1cm} (4.2)

Here $C_{sp}$ is the crystal specific heat capacity; $\rho$ is crystal density. When the temperature distribution is non-uniform, material constants in integrals (2.4) could not be taken outside the integral sign. These integrals however still can be estimated if the temperature distribution can be factorized into functions depending on only one spatial coordinate. To calculate the kinetics of piezoelectric resonance frequency shift one should first solve equation (4.2), then substitute two-dimensional temperature distribution into (2.4) and then determine frequency shift. In the Fig. 2 (a) typical stationary calculated temperature distribution is presented. Both calculated and measured kinetics for the 832.5 kHz KTP resonance are shown in Fig. 2 (b).

Following parameters were obtained to best fit measured kinetics: optical absorption coefficient $\alpha = 0.12m^{-1}$, heat transfer coefficient $\mu = 0.7W/(m^2K)$. To ensure the parameters are determined uniquely one has to analyze several resonances.

The method introduced in this paper allows determining temperature distribution inside a nonlinear-optical crystal during its interaction with high-power laser radiation. This is important in laser frequency conversion applications, as phase matching condition is strongly dependent on the crystal temperature.

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