Concept of equivalent temperature of the nonlinear-optical crystal interacting with nonuniform laser radiation

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Abstract. Experimental criterion was determined for application of equivalent temperature of nonlinear-optical crystal heated both uniformly and nonuniformly by laser radiation with known spatial distribution of intensity. Novel coefficients are introduced in laser physics that characterize thermal response of crystal to the laser radiation propagating through it. Theoretical model is proposed for calculation of these coefficients. Physical parameters of nonlinear-optical crystals were determined that have significant influence upon correspondence between calculated and measured values of introduced coefficients.

1. Introduction
Generation of radiation in solid state lasers is always accompanied by the conversion of some part of optical pump power into heat leading to the active medium temperature rise. In condition of continuous generation of radiation the thermodynamic characterization of laser medium is based on the exploitation of nonuniform and at the same time nonequilibrium temperature.

There is one important class of thermodynamic problems in laser physics that refers to the laser radiation frequency conversion in nonlinear-optical crystals. It is essential that efficient harmonics generation and parametric conversion of laser radiation is governed by phase matching conditions that are fulfilled only in certain temperature range of the crystal [1]. However during nonlinear-optical process it is typical that optical absorption coefficients of both pump radiation and converted one change with pump power. Moreover heat exchange coefficient between crystal and air also depends on crystal temperature. These facts lead to uncontrollable rise of crystal temperature with pump power and can result in irreversible crystal degradation (optical damage). Precise nonlinear-optical crystal temperature control is crucial in such circumstances.

Another class of thermodynamic problems in optics is connected with laser calorimetry [2]. At present laser calorimetry method is international standard for determination of optical absorption coefficients of solid optical media. This technique supposes irradiation of the test sample by low level optical power (usually several Watts) and the last one is not heated significantly provided that its optical absorption coefficient is not high.

Present work is dedicated to the elaboration of theoretical model together with experimental realization, based on acousto-resonant spectroscopy technique, of precise temperature measurement of the nonlinear-optical crystals nonuniformly heated by laser radiation [3-9]. Such fact that all nonlinear-optical crystals possess piezoelectric properties admits direct noncontact excitation and
detection of acoustical vibrations in crystal using external radiofrequency (RF) electric field. When one of the intrinsic vibration modes frequencies $R_{fn}$ of the crystal sample coincides with the probe electric field frequency $f$ the piezoelectric resonance occurs. Equivalent heating temperature during crystal interaction with laser radiation of power $P$ can be directly determined basing on experimentally measured piezoelectric resonance frequency shift $\Delta R_{fn}(P)$ provided that resonance frequency dependence on temperature $R_{fn}(\theta)$ for the case of crystal uniform heating was determined.

2. Equivalent temperature concept

As it was clarified propagation of laser radiation through the crystal results in its nonuniform heating due to optical absorption. For example single-mode laser radiation intensity has Gaussian distribution in cross section to the propagation direction. In this case crystal temperature distribution $\theta_{cr}(x_1, x_2)$ can be obtained by solving nonstationary heat conduction equation with appropriate boundary condition at the crystal-air interface. In assumption of weak optical absorption and crystal convective air cooling it can be written as follows

$$\left\{ \begin{array}{l}
\rho_{cr} c_{cr} \frac{\partial \theta_{cr}}{\partial t} = \kappa_{cr} \left( \frac{\partial^2 \theta_{cr}}{\partial x^2} + \frac{\partial^2 \theta_{cr}}{\partial y^2} \right) + \alpha(\lambda)I(x_1, x_2), \\
\rho_{a} c_{a} \frac{\partial \theta_{a}}{\partial t} = \kappa_{a} \left( \frac{\partial^2 \theta_{a}}{\partial x^2} + \frac{\partial^2 \theta_{a}}{\partial y^2} \right), \\
-\kappa_{cr} \frac{\partial \theta_{cr}}{\partial n} = h^T (\theta_{a} - \theta_{cr}).
\end{array} \right. \hspace{1cm} (1)$$

Here subscripts “cr” and “a” denote reference of values to the crystal and air respectively; $\rho$ and $c$ are density and specific heat capacity; $\kappa$ is thermal conductivity; $\alpha(\lambda)$ is crystal optical absorption coefficient, which is wavelength dependent; $I(x_1, x_2)$ is radiation intensity spatial distribution; $\theta_{cr}$ and $\theta_{a}$ are temperatures at the crystal-air interface; $n$ is the normal vector to the interface; $h^T$ is the heat transfer coefficient. At the outer boundary air temperature is $\theta_0$. Due to relatively low crystal overheating its cooling, conditioned by mechanism of spontaneous radiation, is not considered.

Mathematically simplified solution of problem (1) is used in laser calorimetry technique, which is standardized method for measuring optical absorption and heat transfer coefficients [2]. In case laser radiation of power $P$ at $\lambda$ wavelength propagates along crystal length $L_3$ during time interval $0 < t < t_1$ then crystal temperature change caused by optical absorption of laser radiation can be expressed as:

$$\theta_{cr}(t) - \theta_0 = \begin{cases} 
\frac{\alpha L_3 P \tau}{mc_{cr}} [1 - \exp(-t/\tau)], & 0 < t < t_1 \\
\frac{\alpha L_3 P \tau}{mc_{cr}} [\exp(-t/\tau) - \exp(-(t-t_1)/\tau)], & t > t_1.
\end{cases} \hspace{1cm} (2)$$

Here $m$ is crystal mass. Equation (2) is obtained by solving combined nonstationary heat conduction equations (1) taking into account several simplifying assumptions such as that temperature distribution inside crystal volume is uniform during interaction with laser radiation and crystal overheating is relatively small in comparison with ambient air temperature. Characteristic time $\tau$ describes crystal temperature loss.

$$\tau = \frac{mc_{cr}}{h^TS} \hspace{1cm} (3)$$

where $S$ is overall area of crystal surface. Its value can be obtained by fitting of the experimental data of crystal heating ($0 < t < t_1$) and cooling ($t > t_1$) by the function of the following type

$$\theta_{cr}(t) = \theta_{cr}^0 + \Delta \theta_{cr} \exp(-t/\tau) \hspace{1cm} (4)$$

In case crystal parameters and laser power $P$ are known then optical absorption $\alpha$ and heat transfer $h^T$ coefficients can be calculated using fitting parameters $\tau$ and $\Delta \theta_{cr}$ as follows
Kinetics of the sample temperature is measured during heating by laser radiation and subsequent cooling after the laser is switched off. However, in this case sample temperature or to say more precisely air temperature in the vicinity of the sample surface is measured indirectly using external thermal detectors.

Equivalent temperature concept has been recently employed in calorimetry technique. In case of piezoelectric crystals its temperature is directly measured using piezoelectric resonance frequency temperature dependence. It was shown that in case of piezoelectric crystal uniform temperature $\theta_{cr}$ change its piezoelectric resonance frequencies $R_{fn}$ in a first approximation linearly shift with temperature \([3-9]\)

$$R_{fn}(\theta_{cr})=R_{fn}(\theta_0)+K_{n}^{\text{pr}}(\theta_{cr} - \theta_0).$$  \(6\)

Here $K_{n}^{\text{pr}}$ is the piezoelectric thermal coefficient of the particular $n$-th vibration mode and $\theta_0$ is initial crystal temperature. Obviously crystal temperature $\theta_{cr}$, which in this case is equal to surrounding air temperature, can be determined if resonance frequency shift is known

$$\theta_{cr} = \theta_0 + \frac{R_{fn}(\theta_{cr}) - R_{fn}(\theta_0)}{K_{n}^{\text{pr}}}. \quad (7)$$

It was also demonstrated that in case of linear optical absorption the piezoelectric resonance frequencies linearly depend on laser power $P$

$$R_{fn}(P) = R_{fn}(0) + K_{n}^{\text{po}} P.$$  \(8\)

Here $K_{n}^{\text{po}}$ are piezoelectric resonance optical coefficients and $R_{fn}(0)$ corresponds to resonance frequency at $P=0$.

Nonuniform temperature of the crystal can be precisely determined using notion of crystal equivalent temperature $\Theta_{eq}$. Crystal equivalent temperature is expressed as follows

$$\Theta_{eq}(P) = \theta_0 + \Delta\Theta_{eq}(P).$$  \(9\)

Temperature $\theta_0$ corresponds to $P=0$ and $\Delta\Theta_{eq}(P)$ is crystal equivalent heating temperature determined directly from the $n$-th piezoelectric resonance frequency shift

$$\Delta\Theta_{eq}(P)=\frac{R_{fn}(P)-R_{fn}(0)}{K_{n}^{\text{pr}}}.$$  \(10\)

In linear case of $R_{fn}$ dependence on $P$ equation (10) transforms in

$$\Delta\Theta_{eq}(P)=\frac{K_{n}^{\text{po}}}{K_{n}^{\text{pr}}} P = \beta P.$$  \(11\)

Here $\beta$ is piezoelectric resonance optothermal coefficient, which value depends mostly on crystal optical absorption $a(\lambda)$, and heat exchange conditions.

True thermodynamic temperature of the crystal interacting with laser radiation can be expressed as

$$\theta_{cr}(x_1,x_2,x_3,P) = \theta_0 + \Delta\Theta_{eq}(P) + \delta\theta(x_1,x_2,x_3,P).$$  \(12\)

Our calculations revealed that always the crystal overall temperature rise greatly exceeds nonuniform part. It means that thermodynamic temperature of the crystal heated by laser radiation can be successfully replaced by equivalent temperature. In many interesting cases such as nonlinear-optical frequency conversion the crystal is heated nonlinearly with laser power. Still its equivalent temperature can be determined using equation (9) - (11) \([8]\).

3. Experimental results

3.1. Experimental setup

Simplified block-scheme of the experimental setup for the nonlinear-optical crystal’s electrical impedance measurements during interaction with laser radiation is shown in figure 1 (a). Crystal
sample is placed between strip metallic electrodes. Two electrodes form a capacitor and function as an antenna that excites crystal vibration and simultaneously detects its radiofrequency response. Voltage of the frequency \( f \) from the radiofrequency generator is applied to the capacitor that is connected in series with the load resistor \( R \). Radiofrequency field is a probe field and its power that affects the crystal is very low (\( 10^{-3} - 10^{-6} \) W). Lock-in amplifier is used for measuring complex voltage \( U_R \) on \( R \) that is proportional to the electrical current flowing through the crystal. Thermostat is used for the crystal temperature \( \theta_{cr} \) stabilization.

Figure 1 (b) shows dependencies of the \( U_R \) voltage amplitude on frequency \( f \) measured for two mutually perpendicular orientations of quartz crystal in respect to the probe electric field \( \mathbf{E}_R \). Specimen of \( \alpha \)-quartz is rectangular parallelepiped \( 2.9 \times 2.9 \times 11.2 \) mm\(^3\) with polished facets. At certain frequencies \( f_{np} \) piezoelectric resonances between the external electric field and crystal internal vibration modes are observed. Figure 1 (b) inset shows spectrum at piezoelectric resonance \( f_R = 1.52768 \) MHz (at \( \theta_{cr} = 18 \) °C) that was selected for further investigations.

3.2. Temperature calibration and crystal heating by laser radiation

For investigation of the crystal interaction with laser radiation the temperature calibration of the selected piezoelectric resonance frequencies is made. For precise measurement of crystal equivalent temperature sensitive piezoelectric resonances, i.e. with high piezoelectric resonance thermal coefficient are preferable. Figure 2 (a) shows dependence of the selected resonance frequency on crystal temperature during its uniform heating. Linear approximation (6) gives piezoelectric resonance thermal coefficient \( K_{prt} = -53.6 \) Hz/K of the selected mode.

Equivalent temperature of the crystal heated by laser radiation of power \( P \) can be directly determined using relations (9) and (10) by measuring piezoelectric resonance frequency shift \( R(f(P)) \) of the calibrated piezoelectric resonance. Figure 2 (b) shows dependence of the selected resonance frequency on laser power. Laser source is continuous wave single-mode fiber laser that generates polarized radiation at wavelength \( \lambda = 1064 \) nm. Collimated Gaussian beam of 1.4 mm diameter (beam quality factor \( M^2 \) is close to 1) was directed along crystal length \( L_s = 11.2 \) mm. Values of \( R(f(P)) \) at each power \( P \) were measured after reaching stationary temperature state of the quartz crystal with surrounding air.

Linear approximation (8) gives value of piezoelectric resonance optical coefficient \( K^{opt} = -42.5 \) Hz/W of selected resonance. Then optothermal coefficient \( \beta \), which is independent on resonance selection, can be obtained using (11). Its value is \( \beta = 0.79 \) K/W.
3.3. Crystal temperature kinetics measurement during and after laser irradiation

Crystal piezoelectric resonance frequency kinetics is measured during interaction with laser radiation. Then both optical absorption and heat transfer coefficients can be obtained replacing $\theta_{cr}$ in (2) and (4) by the value of $\Theta_{eq}$ represented by (9). Figure 3 shows experimental results of our crystal heating by laser radiation of 10.5 W power at 1064 nm wavelength and subsequent cooling.

Exponential fittings of quartz crystal heating and cooling kinetics using equation (4) give characteristic temperature kinetics times $\tau_h = 53.18$ s and $\tau_c = 53.19$ s respectively. Corresponding values of optical absorption at $\lambda = 1064$ nm and heat transfer coefficients are calculated using equations (3) and (5): $\alpha = 2.4 \cdot 10^{-3}$ cm$^{-1}$ and $\dot{h} = 23.5$ Wm$^{-2}$K$^{-1}$ respectively.

Experimental results reveal that both heating and cooling events gives almost equal values of characteristic temperature kinetic time ($\tau_h \approx \tau_c$). This is the main criteria for validity of equivalent temperature concept of the crystal nonuniformly heated by laser radiation. When laser radiation affects the crystal its temperature is nonuniform and can be calculated using equation (1). It is essential that after switching off laser radiation the temperature distribution inside crystal rapidly becomes uniform due to the high value of its thermal conductivity. It means that in the last case equivalent temperature of the crystal is identical to the crystal thermodynamic temperature.

4. Equivalent temperature model

4.1. Stages of generalized model

Modeling of equivalent temperature of nonlinear-optical crystal nonuniformly heated by laser radiation should contain following stages:

- Determination of crystal elastic and piezoelectric tensors temperature dependence;
• Measurement of crystal impedance (admittance) spectrum for finding piezoelectric resonances [3];
• Measurement of piezoelectric resonance thermal coefficients $K_{pr}$ from crystal uniform heating experiment [3];
• Theoretical calculation of crystal internal vibration modes spectrum and identification with experimentally observed piezoelectric resonances [9];
• Measurement of piezoelectric resonance optical coefficients $K_{pr}$ from crystal nonuniform laser heating experiment [3];
• Determination of optothermal coefficient $\beta$ for the crystal equivalent temperature $\Theta_{eq}(P)$ determination at given laser power $P$ [3];
• Measurement of laser radiation intensity distribution;
• Measurement of optical absorption $\alpha(\lambda)$ and heat transfer $hT$ coefficients [4, 6].
• Theoretical calculation of the nonuniform temperature distribution $\theta_{cr}(x_1, x_2, x_3, P)$ in crystal heated by laser radiation by solving heat conduction equation (1). Determination of $\theta_{cr}(x_1, x_2, x_3, P)$ maximum $\theta_{max}(P)$ and minimum $\theta_{min}(P)$ [5, 9];
• Theoretical calculation of crystal equivalent temperature $\Theta_{eq}(P)$ for the laser power $P$ at $\lambda$ wavelength in compliance with elastic and piezoelectric coefficients distribution that corresponds to calculated nonuniform temperature distribution [5, 9];
• Measured and calculated equivalent temperature should lie between calculated values $\theta_{min}(P)<\Theta_{eq}(P)<\theta_{max}(P)$ [5, 9].

4.2. Basic equations and boundary conditions
The vibration of the sample is characterized by the vector of mechanical displacement $u_i(x_1, x_2, x_3)$ and electric potential $\phi(x_1, x_2, x_3)$. Equations describing motion of the sample can be written as

\[
\begin{align*}
\rho \ddot{u}_i &= T_{ij, j} + F_i \\
D_{ij, j} &= 0 \\
T_{ij} &= c_{ijkl} S_{kl} - d_{ik} E_k \\
D_i &= \varepsilon_i E_j + d_{ik} S_{jk}
\end{align*}
\]  

(13)

Here point above the variable means time derivative and $G_{ij} = \partial G/\partial x_j$ denotes spatial derivative; indices $i, j, k, l = 1, 3$; $\rho$ is sample density; $T_{ij}$ and $S_{ij} = 0.5(u_{ij, j} + u_{ij, j})$ are stress and strain tensors respectively; $F_i$ are components of body force vector; $D_i$ and $E_i = -\phi_j$ are components of electric induction and electric field vectors respectively; $c_{ijkl}$, $d_{ik}$, $\varepsilon_i$ are elastic, piezoelectric and permittivity tensors respectively. Region occupied by the sample is denoted by $\Omega$, its boundary is denoted $\partial\Omega = \Gamma$. In general system (13) should be supplemented by initial and boundary conditions. For our purposes only boundary conditions have to be considered. These are split into mechanical and electrical boundary conditions of the first or second kind. They can be represented as follows:

\[
\begin{align*}
\mathbf{u}_i(\mathbf{r}) &= \mathbf{u}_j, \mathbf{r} \in \Gamma_u; & T_{ij}(\mathbf{r}) n_j &= \mathbf{F}_{ij}, \mathbf{r} \in \Gamma_b; & \phi(\mathbf{r}) &= \phi_j, \mathbf{r} \in \Gamma_\phi; & D_i(\mathbf{r}) n_j &= \mathbf{D}_i, \mathbf{r} \in \Gamma_D.
\end{align*}
\]  

(14)

Here $\mathbf{r}$ is the radius vector. Boundaries are $\Gamma = \Gamma_u \cup \Gamma_b$ and $\Gamma = \Gamma_\phi \cup \Gamma_D$. For the determination of resonance frequencies for bodies of different geometry the common approach is to use Lagrange-Hamilton variation principle.

4.3. Variation principle
As was noted by many authors (e.g., see [10-12]) variational formulation is very useful tool for numerical solution of the problems with unspecified boundary conditions. Tiersten [10] proposed to use this method in such a way that no constraints are imposed on the variations. His idea was to add to the main functional the constraints multiplied by undetermined Lagrange multipliers. Following Tiersten one can obtained functional for piezoelectric medium in the following form:
Here $n$ is normal vector to the surface and $dA$ is area element. By finding stationary points of (15) using natural boundary conditions $\Gamma_u = \Gamma_\varphi = \emptyset$, $\vec{F}_i = \vec{D} = 0$ we will obtain the set of eigenmodes of the sample. The process of obtaining stationary solutions is described, for example, by Demarest [12] for the case $\varepsilon_{ij} = d_{ijk} = 0$. It may be divided into two stages: separation of time dependence and variation of spatial part of the functional. The latter stage was described by Tiersten and Holland [10, 11]. The most important conclusion concerning the time dependence is that it is harmonic with frequency $w$. So later on we consider only spatial part of the functional [9].

\[
J_S = \sum_{\Omega} \left( \rho w^2 \sum_{i} \varepsilon_{ij} - c_{ijkl} S_{ij} S_{kl} + \varepsilon_{ij} E_j + 2 d_{ijkl} S_{ij} E_k \right) d\Omega.
\]  

(16)

In order to solve numerically problem (16) the Rayleigh-Ritz method is usually used. In this method unknown functions of displacement and potential are expanded using some set of orthonormal basis functions $\{\psi_p\}$ of the total number $N_b$ ($p = 1, N_b$). It means that equations for eigenmodes can be obtained by substitution series expansions $u_i = C_n^u \psi_p$ and $\varphi = C_n^\varphi \psi_p$ in Lagrangian (16) and subsequent differentiation with respect to each expansion coefficient $C_n$ [9]. Extremum of the functional (16) corresponds to the condition of equal to zero derivatives $\partial J_S / \partial C = 0$. In matrix form equations for eigenmodes can then be written as

\[
\begin{bmatrix}
C_n^u \\
(Y - \Pi V^{-1} \Pi^\top) C_n^u
\end{bmatrix}
= w^2 W C_n^u.
\]

(17)

Matrices $W$, $Y$, $\Pi$ and $V$ are ($p = 1, N_b$, $q = 1, N_b$)

\[
W_{pq} = \int_{\Omega} \rho \psi_p \psi_q d\Omega, \quad Y_{pq}^{ik} = \int_{\Omega} c_{ijkl} \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_i} d\Omega,
\]

\[
\int_{\Omega} d_{ijkl} \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_i} d\Omega, \quad V_{pq} = \int_{\Omega} \varepsilon_{ij} \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_i} d\Omega.
\]

(18)

After finding the eigenfrequencies $w_n$ and pertinent spatial distributions of the displacement and potential the piezoelectric resonance thermal coefficients $K_n^{\text{pm}} = \frac{1}{2\pi} \frac{d w_n}{d \theta}$ can be calculated if temperature dependence of the elastic constants $c_{ijkl}(\theta)$ is known. Detailed calculation can be found elsewhere [6, 9]. As it was estimated the contribution to resonance frequency shift of other parameters (e.g. density $\rho$) variation with temperature is not determinative. It was also demonstrated that for common values of material piezoelectric and dielectric coefficients the contribution of piezoelectric effect to intrinsic vibration mode frequencies doesn’t exceed 0.5% [13]. So for simplification we shall neglect $\Pi V^{-1} \Pi^\top$ term in (17). Using perturbation theory the resonance frequency shift can be expressed as:

\[
\frac{d w_n}{d \theta} = \frac{C_n^{\text{pm}}}{2 \rho w_n} \frac{d N_{n}^{ik}}{d \theta} C_n^{\text{pm}}.
\]

(19)
During interaction with laser radiation the nonuniform temperature distribution inside the crystal leads to an additional shift of its eigenmode frequencies, because here unlike the uniform case elastic constants will have spatial dependence. If we consider small uniform temperature perturbation \(d\theta\) then resulting elastic coefficients \(c_{ijkl}^d\) can be represented by

\[
c_{ijkl}^d = c_{ijkl}^0 + \frac{dc_{ijkl}}{d\theta} d\theta.
\]

Here \(c_{ijkl}^0\) corresponds to the initial uniform temperature \(\theta_0\). By substituting (20) into equation (18) for matrix \(Y\) we obtain using relation (19) the following expression for piezoelectric resonance thermal coefficients:

\[
K_{n}^{\text{pt}} = \frac{d}{d\theta} c_{ijkl}^a B_{pq}^\beta C_{kp,n}^a.
\]

Here \(C_{kp,n}^a\) are expansion coefficients of mechanical displacement that corresponds to the \(n\)-th vibration mode. And matrix \(B\) is

\[
B_{pq}^\beta = \int \int \int \frac{\partial \psi_{p}}{\partial x_j} \frac{\partial \psi_{q}}{\partial x_j} d\Omega.
\]

5. Comparison of experimental and theoretical results

Eigenfrequencies of quartz in rectangular parallelepiped form with dimensions \(L_1 \times L_2 \times L_3\) can be found as solution of eigenvalue problem (17) using Legendre polynomials \(P_{p,i}(x_i)\) as basis functions [9, 12]:

\[
\psi_{p}(x_1, x_2, x_3) = \prod_{i=1}^{3} \left[ \frac{2p_i + 1}{L_i} \right]^{1/2} P_{p,i} \left( \frac{2x_i}{L_i} \right).
\]

Such basis is convenient to use here because for such sample shape it forms orthogonal system:

\[
\int \int \int \psi_{p} \psi_{q} d\Omega = \delta_{p_1, q_1} \delta_{p_2, q_2} \delta_{p_3, q_3}.
\]

Here \(p_i\) is polynomial degree specified by \(i=1,3\) and \(p-1, N\) where \(N\) is total number of basis functions. Vibration spectrum \(w_n\) is calculated from the known sample dimensions, its mass, and a set of surmised elastic constants. The main problem is that number of calculated modes in narrow frequency range greatly exceeds number of experimentally observed ones. Special identification procedure should be performed for finding correct correspondence between certain measured mode and one of the calculated modes. Recently we have introduced novel simple methods of modes identification. First one relies on comparison of measured and calculated piezoelectric resonance thermal coefficients (for more details see [9]) and second one is based on comparison of resonance dimensional coefficients, i.e. mode frequency shift with the change of sample size [7]. These coefficients can be experimentally measured and also calculated using expression (21).

Figure 4 (a) shows calculated and measured resonance spectra of our quartz crystal (2.9×2.9×11.2 mm\(^3\)) in frequency range from 0.9 MHz to 1.5 MHz. Initial set of quartz crystal elastic constants together with its dependence on temperature were taken from literature [14]. By variation of elastic constants (about 1\%) and its temperature derivatives (about 2\%) values we have identified calculated mode \((Rf_{\text{calc}} = 1527700 \pm 100 \text{ Hz at } 18 \degree \text{ C}; \ (K^{m}_{\text{calc}})^{\text{calc}} = -54 \pm 1 \text{ Hz/K})\) with experimentally measured mode \((Rf_{\text{exp}} = 1527681 \pm 1 \text{ Hz at } 18 \degree \text{ C}, \ (K^{m}_{\text{exp}})^{\text{exp}} = -53.6 \pm 0.1 \text{ Hz/K})\), which was selected for detailed investigations (see section 3). It can be seen that there are few experimental modes that do not match well enough with calculated ones. Disagreement can be attributed to the imperfection of crystal form and errors of sample sizes determination.
Figure 4. (a) Calculated (0.5 level) and measured (1 level) resonance spectra of quartz crystal (rectangular parallelepiped 2.9×2.9×11.2 mm³). Arrow indicates resonance mode that was identified. (b). Calculated stationary temperature distribution in quartz crystal, irradiated by 10.5 W laser power, and ambient air in respect to the thermostat temperature [9].

Stationary temperature distribution inside crystal heated by laser radiation and surrounding air can be calculated using equation (1) with equal to zero time derivatives. Figure 4 (b) shows calculated stationary temperature distribution for our quartz crystal (2.9×2.9×11.2 mm³) irradiated by laser radiation of power $P = 10.5$ W at $\lambda = 1064$ nm. Laser radiation intensity has Gaussian distribution with beam diameter $d = 1.4$ mm at 1/e² level:

$$I(x_1, x_2) = \frac{2P}{\pi(d/2)^2} \exp\left(\frac{-2x_1^2 + x_2^2}{(d/2)^2}\right).$$  (25)

Values of optical absorption and heat transfer coefficients: $\alpha(\lambda) = 2.4 \times 10^{-3}$ cm⁻¹ and $h_T = 23.5$ Wm⁻²K⁻¹ were measured in calorimetric experiment (see section 3.3). Other necessary physical parameters of quartz crystal and air were taken from literature.

Crystal overall temperature rise is about $8^\circ$C and maximum difference of temperature inside crystal is of $\Delta\theta_c = 10^{-2}$ C order. Temperature gradient in the surrounding air is relatively high in comparison with crystal volume. Also there is a gap of about $1^\circ$C between crystal and air temperatures at crystal interface. These results reveal that for characterization of crystal response to the propagating laser radiation it is necessary to use equivalent temperature concept.

Calculation of crystal equivalent temperature is more of academic rather than practical interest. From practical point of view it is necessary to calculate:

- Temperature distribution inside volume of the crystal heated by laser radiation with given spatial intensity distribution. Nonuniform temperature distribution is not measured experimentally
- Frequencies of internal vibration modes of the crystal when its temperature is uniform;
- Dependence of all vibration mode frequencies on uniform crystal temperature and comparison of calculated piezoelectric resonance thermal coefficients with measured ones.

Identification of crystal vibration modes gives opportunity to amend accuracy of experimental values of elastic constants together with its temperature derivatives. These are key parameters of nonlinear-optical crystal that specify its threshold values of optical fluence, which leads to optical damage of crystals.

Frequency shifts of resonances caused by absorption of laser radiation of power $P$ can be calculated, using e.g. perturbation theory. This frequency shift can be associated with the crystal uniform temperature change that gives the same shift value. Thus determined temperature change is called an equivalent heating temperature of the crystal. Implementation of perturbation theory gives the following expression for the crystal equivalent heating temperature [9]:

$$...$$
\[
\Delta \Theta_n(P) = \frac{\partial C_{ijkl} \partial \theta_n(P) C^{\alpha}_{ip,a} Z^{\beta}_{pqsl} C^{\gamma}_{kj,a}}{\partial \theta}, \quad (26)
\]

Where matrix \( B \) is expressed by equation (22) and matrix \( Z \) is
\[
Z^{\beta}_{pqsl} = \int_{\Omega} \psi_i(x_1) \psi_i(x_2) \frac{\partial \psi_i}{\partial x_i} \frac{\partial \psi_j}{\partial x_j} d\Omega. \quad (27)
\]

Expression (26) contains temperature derivatives of elastic constants. Values \( B^{ij}_{pq}, Z^{ij}_{pqsl} \) depend only on sample geometry. Vector \( C_{ip,a}^{\alpha} \) characterizes given \( n \)-th resonance. Coefficients \( \partial \theta_n(P) \) characterize the temperature distribution of the irradiated crystal with transverse dimensions \( L_1 \) and \( L_2 \).
\[
\frac{L_1}{2} \leq x_1 \leq \frac{L_2}{2}, \quad \frac{L_1}{2} \leq x_2 \leq \frac{L_2}{2},
\]

Temperature dependence on the coordinate \( x_3 \) along the radiation propagation direction \( L_3 \) is not considered due to low optical absorption. Crystal temperature perturbation \( \partial \Theta \), caused by interaction with laser power, can be expanded using our orthonormal basis functions \( \{ \psi_p \} \):
\[
\partial \Theta(x_1, x_2) = \sum_{r,s} \partial \theta_{rs} \psi_r(x_1) \psi_s(x_2) dx_1 dx_2. \quad (28)
\]

Calculations performed for several resonance vibration modes of crystal in frequency range 20 kHz to 180 kHz revealed that difference between measured value of equivalent heating temperature and calculated one using equation (26) is less than 0.01% [9]. Higher accuracy cannot be achieved because of values of elastic constants and its temperature derivatives as well as sample sizes are determined with lower precision.

6. Main conclusions

Important applications of equivalent temperature concept is determination of optical absorption \( \alpha(\lambda) \) and heat transfer coefficients \( h^T \) in laser calorimetry technique [6]. Classical laser calorimetry, which is standardized method for optical absorption measurement, is based on finding of the correspondence between the solution of nonstationary heat conduction equation and experimentally measured crystal temperature kinetics when it is heated and cooled during and after laser irradiation respectively. Classical approach, which supposes indirect temperature measurement via external thermal detectors, lacks accuracy and crystal equivalent temperature kinetics should be measured instead. Despite that degree of crystal temperature nonuniformities in cases of heating and cooling differ from each other the characteristic kinetic times, measured in each case, have close values due to both low optical absorption and application of low-power laser radiation. However, when high-power radiation is involved especially in nonlinear-optical interactions the application of equivalent temperature concept plays crucial role. It has been already successfully used for determination of crystal temperature in course of nonlinear-optical frequency conversion of laser radiation. Second harmonic generation experiment using nonlinear-optical periodically polled lithium niobate (PPLN) crystal was recently performed [8]. It was observed that when second harmonic power is low the PPLN equivalent temperature linearly depend on pump laser power. However, when PPLN temperature is close to phase matching temperature and second harmonic is efficiently generated, the crystal equivalent temperature exhibit substantially nonlinear dependence on pump power.

Reference

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