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Experimental determination and the theoretical model of an equivalent temperature of nonlinear optical crystals interacting with high-power laser radiation

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Abstract. **We present the results of the use of resonant ultrasound spectroscopy to study changes in temperature of crystals during their interaction with high-power laser radiation. To measure the nonuniform temperature of a crystal, use is made of the frequency (calibrated at uniform temperature θ) of the nth mode of the piezoelectric resonance $\mathrm{Rf}_n(\theta)$ and its change $\Delta\mathrm{Rf}_n(P_{\mathrm{in}})$ upon crystal irradiation by laser light with an average power P_{in}. A spatially inhomogeneous distribution of thermodynamic temperature in the crystal at a fixed laser power P_{in} is set in correspondence with equivalent temperature $\theta_{\mathrm{eq}}(\mathrm{P_{in}})$, defined in this paper. A mathematical model is proposed, justifying the correctness of its introduction. This parameter can be used to study any piezoelectric crystals interacting with laser radiation.**

Keywords: *high-power laser radiation, nonlinear optical crystals, equivalent temperature.*

1. Introduction

Nonlinear crystals are widely used to control both the temporal and spatial parameters of radiation, as well as to convert laser radiation frequency in laser and optoelectronic devices. High pulse and average radiation powers obtained recently make it urgent to study the interaction of radiation with these crystals. In this case, it is necessary to examine both changes in the properties of the medium and characteristics of the process, such as nonlinear optical frequency conversion, for which the phase-matching condition is fulfilled in a limited temperature range determined by the phase-matching temperature width [1].

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The process of radiation propagation in material media is always accompanied by radiation absorption and, consequently, by changes in the medium temperature; therefore, the analysis of the nature and rate of change of the temperature can provide valuable information about the characteristics of changes and development of the interaction process. However, precise methods enabling temperature control of the crystals interacting with high-power laser radiation have not been developed so far. The absence of the possibility of noncontact measurement of temperature of a nonlinear optical crystal exposed to high-power radiation significantly limits the ability to control the optical power absorbed by the crystal. The uncontrolled increase in the absorbed power under such conditions makes it difficult not only to study the mechanisms of formation and development of defects, but also to determine functionally (depending on the incident power) the optical damage threshold of crystals. For more than forty years of studies of mechanisms of laser damage in such crystals as quartz [2 – 4], potassium dihydrogen phosphate [5 – 7], lithium niobate [8 – 12], potassium titanyl phosphate [13 – 17] and others [18 – 24], it was found that the degradation of any of these crystals is inevitably accompanied by its nonuniform heating, nonlinearly increasing with increasing laser power [2 – 24].

For the experimental investigation of laser damage threshold of nonlinear optical crystals, use is made of damage detection methods [25]. But, obviously, the end result of the crystal damage during its operation can be determined by the nature of changes in properties of the medium, in particular the rate of change of temperature.

The noncontact temperature measurement proposed for the study of both traditional and new nonlinear optical crystals will make it possible in some cases to carefully monitor the phase-matching conditions, and in other cases – to perform early diagnostic tests of the starting point of crystal damage. All the more, when changing any parameter of laser radiation (power, intensity, pulse duration, wavelength, etc.), the study of crystal damage is repeated many times (statistical method) [25]. The first step in this direction was made in [24], which shows the results of noncontact temperature measurement of solid samples (including DKDP crystals). It should be noted that the experiment [24] was carried out under extreme conditions in which the central region of the crystal was heated by laser radiation to extremely high (6000 – 12000 K) temperatures. Naturally, at this temperature in this region the crystal atoms are ionised and the ions and electrons form a plasma column.

Precision measurement of temperature in the interaction of laser radiation with a nonlinear optical crystal is necessary for another, equally important, reason. The calorimetric method for measuring the optical absorption coefficients of the crystals (ISO 11551 [26]) is based on traditional measurement of nonuniform temperature of air [27–32], surrounding a nonuniform heated crystal. The accuracy of this method is limited to changes in air temperature at a given spatial point and the sensitivity of a sensor (thermocouple, thermistors, etc.). In these measurements, it is needed to know the coordinates of the sensor, which is located near the heated crystal, as well as to choose correctly the model of heat transfer between the crystal and the ambient air. Obviously, the accuracy of the calorimetric method will significantly increase in the measurement of the temperature of both the crystal and the ambient air. Moreover, with increasing laser power incident on the crystal, there appears the effect of nonlinear optical absorption. Without measuring the temperature of the crystal under these conditions, the calculated absorption coefficients and coefficients of heat transfer, as well as their dependence on the laser power become inaccurate.

It is well known that nonlinear optical crystals have piezoelectric properties. In such crystals due to the inverse piezoelectric effect the radiofrequency (RF) field can excite acoustic vibrations. When the external RF field frequency (f_g is the generator frequency) coincides with the eigenfrequency of the nth mode of the acoustic oscillations ($f_g = \mathrm{Rf}_n$) piezoelectric resonance is observed. To record piezoelectric resonances of nonlinear optical crystals under laser irradiation, we have developed a test bench, which controls the temperature of the crystal and the ambient air with an error of less than 10 mK [33–35]. We have found that the resonance frequencies of nonlinear optical crystals are sensitive to absorption of even very weak optical radiation. It should be noted that in the 1990s attempts were made to use high sensitivity of piezoelectric resonances to optical radiation to determine small absorption coefficients of crystals [36, 37]. Unfortunately, hasty conclusions of paper [37] restricted the applicability of the piezoresonance method for measuring the temperature of the crystal irradiated by extremely low power output (less than 30 mW). Our studies show that the possibility to accurately measure the temperature of nonlinear optical crystals by the acoustic resonance methods under laser irradiation is limited only by the powers, leading to crystal damage. After preliminary studies we have found an opportunity to develop acoustic resonance methods for studying laser interaction with crystals. Moreover, there has appeared a real possibility of precision noncontact diagnostics of degradation of crystals interacting with high-power laser radiation [33, 38].

2. Experiment

A simplified scheme of the test bench is presented in Fig. 1. We have experimentally investigated SiO_2, $KTiOPO_4$, KH_2PO_4, $LiNbO_3$, LiB_3O_5 crystals, which confirmed the possibility of using the bench and the developed method for media with different point symmetry groups.

Detailed description of the bench and the method of stationary studies are given in [34, 35]. The nonlinear optical crystal C_r^m under study was placed in the centre of a capacitor made of flat metal plates. To control the temperature of the electrodes and the air temperature inside the capacitor, we placed near the electrodes additional QT1 and QT2 quartz thermal resonators with small transverse dimensions (Fig. 1, right). Piezoelectric resonances of the main crystal and thermal resonators were determined by the spectral characteristics of the impedance of the circuit comprising a capacitor connected in series to the load resistance generator $R_{load} = 47\ \Omega$. The investigated quartz crystal had the shape of a cuboid with polished faces (parallel to the crystallographic axes) with the following dimensions: $L_x = 2.9$ mm, $L_y = 2.9$ mm, and $L_z = 11.4$ mm; the dimensions of the thermal quartz resonators were as follows: $L_x = 1.0$ mm, $L_y = 1.0$ mm, and $L_z = 11.0$ mm.

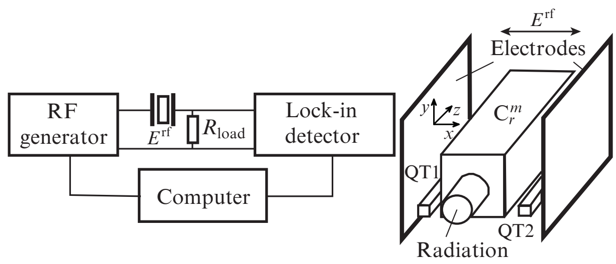

Figure 1. A simplified scheme of the experimental setup: on the left is the electric circuit for measuring the radio frequency admittance (impedance) of a nonlinear optical crystal (E^{rf} is the electric field strength in a flat capacitor), on the right is the scheme for measuring nonuniform heating of nonlinear optical crystal C_r^m by laser radiation (QT1 and QT2 are quartz thermal resonators).

Figure 2 shows the RF spectrum of the voltage module $|U_{ad}|$ on the load resistance R_{load}, measured at an ambient temperature of $\theta_a = 20\,°C$ for the main quartz crystal and thermal resonators. The spectrum contains a number of pronounced high-Q resonances; however, in this range (1.4–2.0 MHz) only a few lines belong to the C_r^m crystal under study, one of which, with the resonance frequency Rf_9 = 1527594 Hz, is investigated in more detail. The selected frequencies of the additional thermal resonators are as follows: Rf_1(QT1) = 1772000 Hz, Rf_1 (QT2) = 1865000 Hz. It should be noted that the information content of the spectra depends not only on the resonant frequency, but also on the spectral line shape $|U_{ad}(f_g)|$.

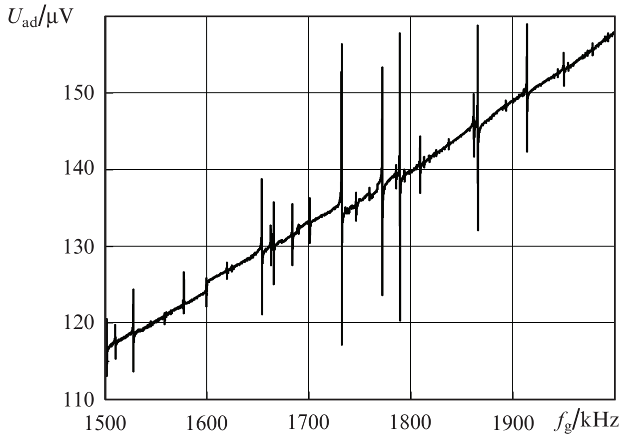

Figure 2. RF spectrum of the voltage U_{ad}, proportional to the admittance of the capacitor with a quartz crystal C_r^m and quartz thermal resonators QT1 and QT2, measured at uniform temperature of ambient air, θ_a.

3. Calculation of equivalent crystal temperature

Highly temperature sensitive piezoelectric resonances of the crystal and thermal resonators are preliminary calibrated during uniform heating (without exposure to laser radiation). For all these crystals the dependence of the shift of the resonance frequency Rf_n on the temperature exhibited linear behaviour in the range 290–420 K. During the temperature calibration the piezoelectric resonance thermal coefficients $K_n^{\mathrm{prt}} = \partial\mathrm{Rf}_n/\partial\theta$ of the main and auxiliary crystals are determined [33]. In the investigated temperature range the piezoelectric resonance thermal coefficients were as follows: $K_9^{\mathrm{prt}}(\mathrm{C}_r^m) = -47.5$ Hz K^{-1}, $K_1^{\mathrm{prt}}(\mathrm{QT1}) = +36$ Hz K^{-1}, $K_1^{\mathrm{prt}}(\mathrm{QT2}) = +15$ Hz K^{-1}.

Under the action of laser radiation with power P_{in}, the resonant frequencies of the main and auxiliary crystals $[\mathrm{Rf}_9(\mathrm{C}_r^m), \mathrm{Rf}_1(\mathrm{QT1}), \mathrm{Rf}_1(\mathrm{QT2})]$ are changed. The functional dependence of the piezoelectric resonance frequency on the power P_{in} is used to determine peizoresonance optical coefficients $K_n^{\mathrm{pro}} = \partial\mathrm{Rf}_n/\partial P_{\mathrm{in}}$. The coefficient K_n^{pro} of the main crystal depends primarily on the optical absorption coefficient of the crystal, α, and on the conditions of heat transfer between the crystal and air [33]. At high powers P_{in} in the case of nonlinear conversion the coefficient K_n^{pro} may depend on the laser power. The coefficients K^{pro} of additional sensors QT1 and QT2 are dependent on their proximity to the main crystal. The accuracy of determining the temperature of nonuniformly heated air with their help increases with decreasing size of the sensor. It was found that the spectral dependence of the line shapes of piezoelectric resonances in quartz during uniform heating in the absence of laser radiation and during nonuniform heating by laser radiation in the investigated range of powers (0–12 W) are functionally identical [34]. Therefore, we propose to characterise nonuniform heating of the crystal by laser radiation with the help of equivalent heating temperature $\Delta\theta_{\mathrm{eq}}(P_{\mathrm{in}})$, defined as follows [33]:

$$\Delta\theta_{\mathrm{eq}}(P_{\mathrm{in}}) = \frac{\Delta\mathrm{Rf}_n(P_{\mathrm{in}})}{K_n^{\mathrm{prt}}}. \qquad (1)$$

The equivalent temperature of the crystal is

$$\theta_{\mathrm{eq}}(P_{in}) = \theta_{\mathrm{a}} + \Delta\theta_{\mathrm{eq}}(P_{\mathrm{in}}). \qquad (2)$$

The temperature distribution obeys the heat conduction equation

$$\kappa\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\theta + \alpha I(x,y) = 0, \qquad (3)$$

where $\kappa(x, y)$ is the thermal conductivity of the medium [crystal (κ_{c}) or air (κ_{a})]; $I(x, y)$ is the spatial distribution of the radiation intensity. We do not take into account here the tensor nature of the thermal conductivity, which, in particular, is associated with a significant scatter in the values of various components of the tensor for these crystals found in the literature. It should be noted that this does not affect the general conclusions of the work, and the tensor nature of the thermal conductivity can be easily accounted for, since equation (3) is solved numerically.

In a standardised method of laser calorimetry the heating model used is based on the convective heat transfer at crystal–air interface [27–32]. In this (simplified) model, we also use the convective heat transfer. The crystal is assumed long enough so that we can neglect the effects of temperature on the ends and consider the temperature distribution along the z axis to be uniform. Equation (3) is supplemented by a convective boundary condition at the crystal–air interface [33]

$$\kappa_{\mathrm{a}}\frac{\partial\theta}{\partial n} = h^{\mathrm{T}}(\theta_{\mathrm{in}} - \theta_{\mathrm{out}}), \qquad (4)$$

where θ_{in} is the crystal temperature near the interface; θ_{out} is the air temperature near the interface; n is the normal to the crystal–air interface; h^{T} is the heat transfer coefficient. In addition, at the outer boundary of the computational domain, which usually coincides with the inner area of the capacitor, the boundary condition of the first type $\theta = \theta_0$ is applied.

For the past 50 years calculations of the resonance frequencies of piezoelectric crystals have been an independent problem of mathematical physics. Fundamental works in this area belong to the 1970s. First, we should mention the work of Eer Nisse [39], Tiersten [40], Holland [41], Demarest [42] and Ohno [43]; a review of variational methods for such problems is given elsewhere [44]. For the numerical analysis use is made of the variational formulation of the problem. Tiersten offers a method of elimination of initial and boundary conditions by adding the existing constraints (written in the form of equations), multiplied by the undetermined Lagrange multipliers. This method with some modifications is often used for such tasks. Below it is presented in abbreviated form (a detailed computational procedure of the piezoelectric resonance frequencies and the temperature dependence of these frequencies under conditions of uniform heating is given in [38, 45] and PhD theses [46, 47]). The eigenmodes of the crystal piezoelectric resonances are found by varying the Lagrangian of the system [43]

$$L_0(u_i,\varphi) = \iiint\Big[\frac{1}{4}c_{ijkl}(u_{i,j} + u_{j,i})(u_{k,l} + u_{l,k}) + 2e_{ijk}(u_{i,j} + u_{j,i})\varphi_{,k} - \varepsilon_{ij}\varphi_{,i}\varphi_{,j} - \rho\omega^2\sum u_i^2\Big]\mathrm{d}\Omega. \qquad (5)$$

Here c_{ijkl} is the tensor of elastic constants of the sample; e_{ijkl} is the tensor of piezoelectric modules; ε_{ij} is the dielectric tensor; ρ is the density of the sample; $\omega = 2\pi\mathrm{Rf}_n$ is the intrinsic vibration frequency of the sample points that is determined; $u_i(x, y, z)$ are the components of mechanical displacement of the sample points; φ is the electric potential; Ω is the crystal volume. $F_{,j} = \partial F/\partial x_i$ is the most commonly used method of writing the derivatives. To solve numerically problem (5), the unknown functions $u_i(x, y, z)$, $\varphi(x, y, z)$ must be expanded in some set of basis functions $\{\psi_p\}$. As a result, the Lagrangian is dependent on the expansion coefficients C_p^i, and the equations for the eigenmodes are obtained by its differentiation in the components:

$$\begin{cases} C^{\varphi} = -D^{-1}\Pi^{\mathrm{T}}C^u, \\ (\Gamma - \Pi D^{-1}\Pi^{\mathrm{T}})C^u = \omega^2 I C^u, \end{cases}$$

$$\Gamma_{pq}^{ik} = \iiint_{\Omega} c_{ijkl}\frac{\partial\psi_p}{\partial x_j}\frac{\partial\psi_q}{\partial x_l}\mathrm{d}\Omega,\ \Pi_{pq}^{i} = \iiint e_{ijl}\frac{\partial\psi_p}{\partial x_j}\frac{\partial\psi_q}{\partial x_l}\mathrm{d}\Omega, \qquad (6)$$

$$D_{pq} = \iiint_\Omega \varepsilon_{jl} \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_l} \mathrm{d}\Omega, \; I_{pq} = \iiint_\Omega \rho \psi_p \psi_q \mathrm{d}\Omega.$$

After finding the eigenfrequencies and spatial distributions of the functions u_i, φ, the piezoresonance thermal coefficients are calculated at a known temperature dependence of the elastic constants.

The calculation of the piezoelectric resonance frequency during nonuniform heating of the crystal requires additional approximations (constraints). We will consider this problem in more detail. The nonuniform temperature distribution inside the crystal leads to an additional shift of its resonance frequencies, because this heating leads to the spatial dependence of the elastic constants. Let

$$c_{ijkl}(x,y,z) = c^0_{ijkl} + c^1_{ijkl} \delta\theta(x,y,z), \tag{7}$$

where the superscript '0' denotes the value of the parameter at a uniform temperature θ_0, and the superscript '1' denotes the temperature derivative of the corresponding parameter;

$$\delta\theta(x,y,z) = \theta(x,y,z) - \theta_0. \tag{8}$$

The elastic tensor in (6) can be written as

$$\Gamma^{ik}_{pq} = \Gamma^{ik,0}_{pq} + c^1_{ijkl} \iiint_\Omega \delta\theta(\boldsymbol{r}) \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_l} \mathrm{d}\Omega. \tag{9}$$

We can calculate that in the case of a maximum order of the decomposition polynomials $N = 20$, the number of basis functions is about 1800. In this case, for each pair of subscripts j, l, the number of integrals to be calculated in (9), is more than three millions, which is practically hard to implement.

We expand the calculated temperature distribution in the basis functions, which are assumed to be orthonormal:

$$\delta\theta(x,y) = \sum_{r_x r_y} \delta\theta_{r_x r_y} \psi_{r_x}(x) \psi_{r_y}(y),$$

$$\delta\theta_{r_x r_y} = \int_{-L_x/2}^{L_x/2} \int_{-L_y/2}^{L_y/2} \delta\theta(x,y) \psi_{r_x}(x) \psi_{r_y}(y) \mathrm{d}y \mathrm{d}x. \tag{10}$$

Then, the addition of the right-hand side of (9) has the form

$$(\delta\Gamma^{ik}_{pq})_{\mathrm{opt}} = c^1_{ijkl} \iiint_\Omega \delta\theta_{r_x r_y} \psi_{r_x}(x) \psi_{r_y}(y) \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_l} \mathrm{d}\Omega$$

$$= c^1_{ijkl} \delta\theta_{r_x r_y} V^{jl}_{pqr}, \tag{11}$$

$$V^{jl}_{pqr} = \iiint_\Omega \psi_{r_x}(x) \psi_{r_y}(y) \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_l} \mathrm{d}\Omega. (11)$$

Let us also denote

$$B^{jl}_{pq} = \iiint_\Omega \frac{\partial \psi_p}{\partial x_j} \frac{\partial \psi_q}{\partial x_l} \mathrm{d}\Omega. \tag{12}$$

After determining Γ, we calculate (for example, by perturbation theory) the resonance frequency shift corresponding to the given radiation power P_{in}. This shift can be associated with a uniform temperature change, which gives the same shift in magnitude. This temperature change is what we call an equivalent temperature of crystal heating. Calculations using perturbation theory give the expression for the equivalent temperature of heating:

$$\Delta\theta_{\mathrm{eq}}(P_{\mathrm{in}}) = \frac{c^1_{ijkl} \delta\theta_{r_x r_y}(P_{\mathrm{in}}) C^i_{p,n} V^{jl}_{pqr} C^k_{q,n}}{c^1_{ijkl} C^i_{p,n} B^{jl}_{pq} C^k_{q,n}}. \tag{13}$$

The possibility of calculating the equivalent crystal temperature leads to an algorithm determining the coefficients α, h^{T} (initially unknown). To find them, it is necessary to solve the inverse problem of successive refinement of the coefficients yielding a temperature distribution in the crystal and the ambient air, at which the equivalent temperatures of the main and auxiliary crystals coincide (with the required accuracy) with those measured. It must be emphasized that the solution of the inverse problem is more of academic interest. In practice, the coefficients α, h^{T} are obtained from kinetic measurements of the equivalent crystal temperature by calorimetric methods (our experiments on the kinetics of the equivalent temperature of the nonlinear optical crystals under conditions of unsteady heating are described in [47]).

We calculated the equivalent temperature for different modes of the same main quartz crystal (at different focusings of laser radiation) and for other nonlinear optical crystals. Calculations show that the equivalent heating temperature calculated by formula (13) and the equivalent temperature itself, $\theta_{\mathrm{eq}} = \theta_{\mathrm{a}} + \Delta\theta_{\mathrm{eq}}$ is always between the minimum ($\theta_{\min}$) and maximum ($\theta_{\max}$) values of the thermodynamic temperature of the main quartz crystal (Figs 3, 4). The difference (observed in Fig. 4) in the plots of the equivalent heating temperature of thermal resonators QT1 and QT2 is determined

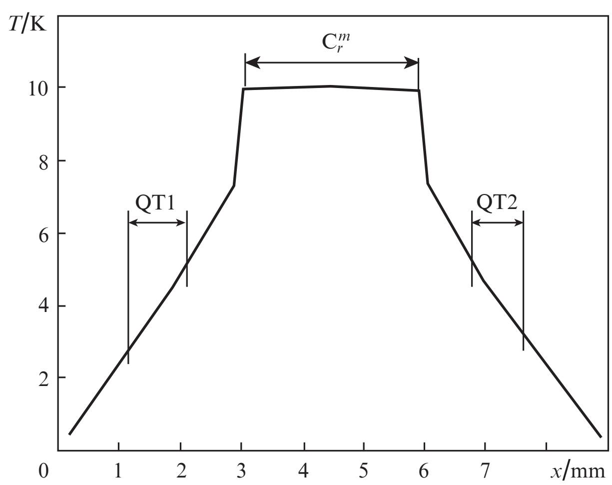

Figure 3. Thermodynamic (calculated) nonuniform temperature of the main crystal C_r^m and temperature of the air surrounding the crystal. Calculations are carried out for radiation of a collimated laser beam [diameter 1.5 mm, parameter $M^2 = 1$ (Gaussian distribution)].

by the impossibility of their exact (symmetric) installation near the main crystal, which once again proves the necessity of measuring the equivalent temperature of the main crystal. For the crystal under study the found values of α, h^{T} in the range $P_{\mathrm{in}} = 0 - 12$ W do not depend on P_{in} and are as follows: $\alpha = 0.7\times10^{-3}$ cm^{-1}, $h^{\mathrm{T}} = 55$ W m^{-2} K^{-1}. Traditional calorimetric measurement of these coefficients (within experimental errors) gave similar results.

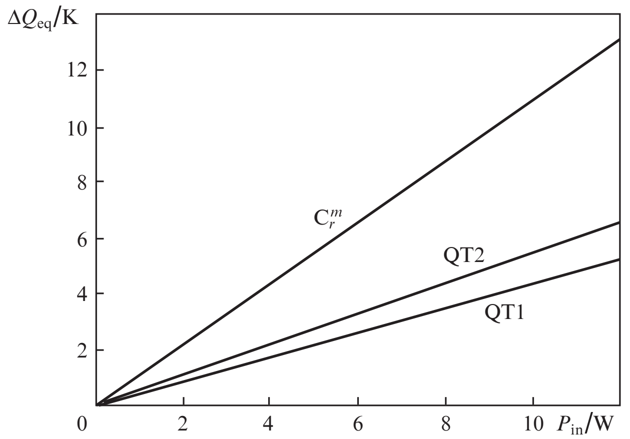

Figure 4. Linear growth of equivalent heating temperature of the main crystal C_r^m and thermal resonators QT1 and QT2 with the power P_{in} of laser radiation incident on the crystal.

4. Conclusions

Thus, the experimentally measured change in the piezoelectric resonance frequency of a nonlinear optical crystal makes it possible to characterise the nonuniform temperature of the crystal exposed to high-power laser light. The equivalent temperature found by us, as shown by calculations, lies in a narrow range between the maximum and minimum values of the thermodynamic temperature. It should be emphasized that the error in the values of the parameters obtained experimentally is determined not by accuracy of measuring the piezoelectric resonance frequency, which remains extremely high, but by the accuracy of determining the shape, dimensions and some (necessary) physical parameters of the sample. Unfortunately, in the literature the necessary parameters of a quartz crystal (not to mention other nonlinear optical crystals) are not accurate enough ($\lesssim 1\%$).

It should be emphasized that many (well-known from the literature) mechanisms of degradation of nonlinear optical crystals, leading to their damage [2–24], define the extraordinary complexity and perhaps impossibility to construct a general, common for all crystals, theoretical model of nonuniform temperature distribution inside each heated crystal. At the same time, there is a principle possibility of searching for such piezoresonance modes of a nonuniformly heated crystal (with a large temperature gradient within it) that would allow this gradient to be experimentally measured. Our experiments show that in piezoelectric crystals, there are plenty of both volume and surface acoustic modes. If these modes are successfully selected and properly identified [39], it will be possible to choose those that perform the averaging of the thermodynamic temperature only in a limited spatial region of the crystal.

Note also that the field of application of the research results presented in this paper is not limited to scientific and academic problems, such as studies of the mechanisms of laser damage of the crystals. For a number of practical applications, the obtained results aimed at determining optical absorption and heat transfer coefficients between the crystal and ambient air in real time are used in some devices of nonlinear frequency conversion of laser radiation [46, 47].

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