Kinetics of the acoustic resonances in nonlinear-optical crystals during the interaction with the single-mode high-power laser radiation

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Abstract. Resonant acoustic spectroscopy is applied for determination of KTP crystal optical absorption coefficient $\alpha$ at the $\lambda=1.064$ $\mu$m. Kinetics of the KTP crystal different resonant acoustical modes under high-power laser radiation influence were measured. We suppose that the discrepancy $\alpha$ values obtained is due to acoustical resonances sensitivity to inhomogeneous heating. Resonant acoustic spectroscopy can probably enable a determination of the inhomogeneous crystal temperature distribution.

1. Introduction
Conversion of laser radiation in nonlinear-optical crystals is always accompanied by inhomogeneous crystal heating due to linear and nonlinear absorption of laser radiation. In case of the high-power laser applications optical absorption in crystals may lead to the crystal optical damage [1]. Most of the nonlinear-optical crystals belong to noncentrosymmetric classes and may exhibit piezoelectric properties, making it possible to excite acoustic resonance in crystal by the external electric field. Every crystal sample has its own set of the acoustical vibration modes of different frequencies and different spatial configurations. Crystal piezoelectric resonance can be observed when the frequency $f$ of an external electric field is equal to $i$-th crystal acoustical vibration mode resonance frequency $f_i$. Measurement of the effect of the high power radiation on different spatial acoustical modes resonances can enable us to investigate the interaction of high power laser radiation with nonlinear optical crystals.

2. Experiment
A KTiOPO\textsubscript{4} (KTP) crystal was selected for the investigation. The crystal sample is polished to a rectangular slab with dimensions 30×3×3 mm\textsuperscript{3}. Crystal $c$-axis and $b$-axis are directed along the 3 mm sides and $a$-axis is parallel to the 30 mm side. KTP crystal density is $\rho = 3.03$ g/cm\textsuperscript{3} [1], the specific heat is $c = 727$ J/(kg*K) [1].
We measured the optical absorption coefficient of the KTP crystal at $\lambda=1.064$ µm wavelength. Classical laser calorimetry is the predominant method for the optical materials absorptance testing [2]. It is based on the temperature measurement near the crystal surface heated by laser radiation. Optical absorption coefficient $\alpha$ is obtained from the matching the experiment with the heating model suggested. However, the internal temperature of the thermal sensor is measured, rather than the temperature of the crystal surface. Resonant acoustic spectroscopy technique gives an opportunity to measure the crystal temperature $T_c$ during both linear optical interaction [3] and nonlinear-optical interaction [4] with the laser radiation. Here it is convenient to characterize the inhomogeneous crystal heating during the high-power laser radiation influence by certain average temperature. We introduce an equivalent crystal temperature $T_{eq}$ which can be directly determined from the piezoelectric resonance frequency $f_i$ provided that the dependence of the resonant frequency on temperature $f_i(T)$ is known for the case of the homogeneous crystal heating without laser radiation influence. In this case the temperature distribution inside the crystal $T_c(x,y,z,t,P)$ can be represented as follows:

$$T_c(x,y,z,t,P) = T_{eq}(t,P) + \Delta T(x,y,z,t,P) \quad (1)$$

In (1) $\Delta T$ is a spatial temperature nonuniformity inside the crystal, $P$ is laser power, $t$ is time. Equivalent temperature $T_{eq}$ is some kind of average crystal volume temperature and it depends on the absorbed laser radiation energy. Time dependence of the $T_{eq}(t)$ can be determined from the piezoelectric resonance frequency kinetics data: $f_i(t)$.

The block-scheme of the experimental setup is shown in Fig. 1a. Electric field from a radio-frequency (RF) generator is applied to the KTP crystal located between two silver electrodes forming a capacitor. The capacitor is connected in series with a small load resistor $R$. External electric field $E_{rf}$ of the frequency $f_{rf}$ is parallel to the crystal $b$-axis. Amplitude and phase of the voltage $U_R$ on the load resistor $R$ are measured by a lock-in amplifier. From the measurement data the electric admittance $Y(f)$ of the KTP is determined. Equivalent electrical scheme of the electro-mechanical resonator formed by the electrodes with the piezoelectric crystal inside is shown in the Fig. 1b. Typical dependencies of the admittance amplitude $|Y(f)|$ and phase $\phi(f)$ on the frequency $f$ around the crystal piezoelectric resonance are shown in the Fig. 1c and Fig. 1d respectively.

Figure 1. (a) Experimental setup. (b) Equivalent electrical scheme of the piezoelectric resonator. Admittance amplitude (c) and admittance phase (d) dependencies on frequency around the crystal piezoelectric resonance.

The RF generator is synchronized with the lock-in amplifier via PC. Capacitor formed by the KTP crystal is placed in the thermostate with the temperature regulation in the 290 – 360 K range. The laser source is a polarized single-mode CW fiber laser operating at $\lambda=1.064$ µm. The maximum output laser power is 14 W. Laser radiation propagates along the KTP crystal a-axis.
3. Results

Four piezoelectric resonances of the KTP crystal, which were selected for the kinetics measurements are shown in Fig. 2a. The coefficients of each piezoelectric resonance frequency shift $df_i/dT$ during the homogeneous heating without the laser radiation were measured. For the 290-360 K temperature range the piezoelectric resonance frequencies exhibit linear dependence on temperature. The example of the $f_i(T)$ dependence and the linear approximation are shown in Fig. 2b for the 832 kHz resonance. The values of the measured $df_i/dT$ coefficients are listed in Table 1 with $f_{0i}$ values corresponding to 305 K.

| Resonance Frequencies Temperature Derivatives. |
|---------|---------|---------|---------|---------|
| $f_{0i}$, [kHz] | 832 | 831 | 823 | 817 |
| $df_i/dT$, [Hz/K] | -53.1±0.2 | -54.2±0.2 | -52.5±0.5 | -52.8±0.3 |

Each kinetics measurement was performed with a constant laser power $P$. The external electric field frequency $f_{el}$ was fixed near the piezoelectric resonance frequency $f_i$ (see Fig. 3a). At this frequency the derivative $d|Y(f)|/df$ is at its maximum and the measurement system is the most sensitive to the piezoelectric resonance shift.

At the $t=0$ the shutter was closed and the lock-in amplifier started to record the amplitude and the phase of the voltage $U_R$ on the load resistor $R$ with the 512 Hz rate. At some instant $t_0 > 0$ the shutter is opened. The portion of the laser power absorbed inside the crystal volume was small due to the low optical absorption coefficient $\alpha$ value at the laser wavelength used. Laser radiation leads to the inhomogenous crystal heating resulting in the piezoelectric resonance frequency shift and the admittance amplitude $|Y(f)|$ changes accordingly. The piezoelectric resonance positions at the succesive time moments $t_1$ and $t_2$ after the shutter opening are shown in Fig. 3a for the 832 kHz resonance. The kinetics of the piezoelectric resonance frequency shift $\Delta f_i(t) = f_i(t) - f_i(0)$ is shown on the Fig. 3a inset. It was obtained from the time dependence of admittance $|Y(t)|$ through the admittance dependence on frequency $|Y(f)|$. The equivalent crystal temperature change can be calculated as follows:

$$\Delta T_{eq}(t) = \frac{\Delta f_i(t)}{df_i/dT}$$

An example of the KTP equivalent crystal temperature change with time for three laser powers $P$ is introduced in the Fig. 3b inset. The optical absorption coefficient $\alpha$ can be determined from the known $\Delta T_{eq}(t)$ derivative at the shutter opening moment $t_0$. 

![Figure 2](image-url)
\[ k(P) = \frac{dT_{eq}(t)}{dt} \bigg|_{t=t_0} \quad (3) \]

The simplified heat conduction equation for the KTP crystal at the instant time \( t_0 \) can be written as follows:

\[ \alpha \cdot l \cdot P \cdot dt = m \cdot c \cdot dT_{eq} \quad (4) \]

Here \( \alpha \) [cm\(^{-1}\)] is the KTP crystal optical absorption coefficient, \( l \) is the crystal length along the crystal a-axis (30 mm), \( P \) is the laser power, \( m \) and \( c \) are the crystal mass and specific heat capacity, \( dt \) and \( dT_{eq} \) are the small increments of time and the equivalent crystal temperature. It is assumed that the \( \alpha l \ll 1 \) condition is true for the specified laser wavelength. The expression (5) for the crystal absorption coefficient \( \alpha \) can be obtained from (4) using (3).

\[ \alpha(P) = \frac{mc}{l} \cdot \frac{(dT_{eq}(t)/dt)}{P} \bigg|_{t=t_0} = \frac{mc}{l} \cdot \frac{k(P)}{P} \quad (5) \]

Kinetics of the piezoelectric resonance frequencies \( f_i(t) \) was measured for the different laser powers. Experiments were carried out for the two laser polarization \( P_L \) directions along the crystal c-axis \( (P_L||c) \) and along the crystal b-axis \( (P_L||b) \).

Dependencies of the \( k_i(P) \) coefficients of the four KTP piezoelectric resonances are shown in Fig. 3b for the case of the laser polarization directed along the crystal c-axis \( (P_L||c) \). Assuming linear optical absorption of the laser radiation inside the crystal volume, the linear approximation of the \( k(P) \) dependencies was made for each KTP crystal resonance. The optical absorption coefficient \( \alpha \) was calculated using (5) (see Table 2 for reference).

From Fig. 3b and Table 2 it can be seen that the discrepancy between \( \alpha \) values, calculated from different resonances measurement, cannot be explained by experimental inaccuracy. We suggest that the various crystal acoustical vibration modes are sensitive to the inhomogeneous crystal heating in different ways. We believe this can present an opportunity for determination of the inhomogeneous crystal temperature distribution inside the crystal volume during interaction with the high-power laser radiation.

### Table 1. KTP optical absorption coefficient \( \alpha \).

| \( f_0 \) (kHz) | 832 | 831 | 823 | 817 |
|----------------|-----|-----|-----|-----|
| \( \alpha_i \times 10^{-3} \) cm\(^{-1}\), \( (P_L||b) \) | 0.84±0.02 | 1.04±0.02 | 0.77±0.02 | 0.89±0.02 |
| \( \alpha_i \times 10^{-3} \) cm\(^{-1}\), \( (P_L||c) \) | 0.88±0.02 | 1.02±0.02 | 0.77±0.02 | 0.87±0.02 |
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