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Minimum Absorption Coefficient Available for Measurements Using Time-resolved Photothermal Common-path Interferometry on the Example of Synthetic Crystalline Quartz

[1], [*] Ksenia VLASOVA, [1] Alexandre MAKAROV, [1] Nikolai ANDREEV, [2] Alexei KONOVALOV

[1] Institute of Applied Physics of the Russian Academy of Sciences,
46 Ulyanova St., 603950 Nizhny Novgorod, Russia
[2] Quartz Technology, LLC., 11 Sovetskaya Sq., 391500, Ryazan region,
Industrial Township Shilovo, Russia
[1] Tel.: +7(831) 436-34-77
[*] E-mail: vlasova.kseniya.1@gmail.com

Abstract: Using the example of an absorption measurement in synthetic crystalline quartz, which has specific ratios of the physical parameters, it was possible to observe the competition among various effects using a time-resolved photothermal common-path interferometry scheme, namely, the thermo-optical effect, nonlinearity of the refractive index in the heating beam field, and heat diffusion. This competition masks the contribution of the thermo-optical effect weakened by stresses in the heating region, and determines the minimum absorption coefficient that can still be correctly calculated from the measurement results. The time-resolved photo-thermal common-path interferometry scheme facilitates separating the contributions to the measured signal of these effects and increases the measurement range in the direction of smaller absorption values. The amount of absorption in various materials at which such competition occurs depends on the ratio of its physical constants, which is $\leq 5 \times 10^{-7}$ cm^{-1} for crystalline quartz, $\leq 2 \times 10^{-8}$ cm^{-1} for quartz glass and $\leq 10^{-5}$ cm^{-1} for crystalline silicon. The problems of using experiment measurements of small absorption values in samples of synthetic crystalline quartz are discussed using an example crystal grown through the hydrothermal method at the Russian company Quartz Technologies.

Keywords: Minimum measurable absorption, Refractive index nonlinearity, Thermo-optical parameter, Heat diffusion, Synthetic quartz crystal.

1. Introduction

The development of technology for producing ultra-pure quartz glasses brings up the question of reliable methods for ultra-low $\alpha \sim 10^{-7}$ cm^{-1} absorption measurements within the relevant optical range. In addition, in the technology used for growing ultra-pure

silicon, methods for absorption measurements at the level of $\alpha \sim 10^{-11}$ cm^{-1} in the near infrared wavelength range [1] are and in demand.

At present, various indirect measurement schemes based on the change in refractive index during the local heating of samples through laser radiation [2-6] have become common. Such schemes differ in

sensitivity depending on the specific implementation of the method used for fixing the change in the refractive index, as well as on the various technical details of their execution.

The most sensitive schemes for measuring the absorption coefficient in quartz glasses, claiming, according to their authors, to measure the absorption coefficient of $\sim 10^{-8}$ cm^{-1} are PCI [2-3], LID [4-5], Shack-Hartmann [6]. However, as shown through control bulk absorption measurements in quartz glasses with an OH group content not exceeding 1–2 ppm and with absorption guaranteed to be within the range of 10^{-6} cm^{-1} to 10^{-7} cm^{-1}, the level of 10^{-6} cm^{-1} for the listed schemes is the detection limit [1, 7].

We proposed a new, more sensitive scheme called a time-resolved photothermal common-path (TPCI) [8], which is also based on the photothermal effect; however, unlike the schemes listed above, TPCI achieves a temporal resolution to the process of changing the refractive index that occurs when heating and cooling a sample.

The temporal resolution realised in the scheme was associated with the heated region characteristic cooling time, which occurs when the temperature distribution width increases owing to the thermal conductivity. This time varies widely and depends on the sample material and heating beam width. For our conditions, it was 700 μs for quartz glass and 120 μs for crystalline quartz. For silicon under similar conditions, this time can be estimated as ≤10 μs. The heating pulse duration is selected based on the cooling times.

In [9-10], absorption measurements at $\alpha \sim 10^{-7}$ cm^{-1} were demonstrated in ultra-pure quartz glasses (Ohara and Heraeus) and synthetic crystalline quartz (Quartz Technology) using the new scheme. A signal-to-noise ratio of 50:1 was found. In addition, this scheme has a constructive potential for increasing the sensitivity for measurements in quartz glass.

At the same time, the question of limiting the absorption value available for measurements using photothermal methods remains open. One of the aspects fundamentally limiting such measurements, as mentioned in the literature, is related to the nonlinear dependence of the refractive index on the magnitude of the heating laser radiation field [1, 3].

In our earlier study [8-9] when measuring absorption using TPCI in quartz glasses with a low OH group content, the nonlinear refractive index response was observed within the pulse of the heating beam field and was about 1/6 of thermo-optical signal. In this study, using the TPCI scheme, we observed the competition of various effects, namely the thermo-optical effect, nonlinearity of the refractive index in the heating beam field, and a temperature variation from the thermal conductivity, when we measured the absorption in synthetic crystalline quartz. These effects mask the contribution of the thermo-optical effect, which is weakened by stresses in the heating region, thereby reducing the thermal deformations. In some conditions, this may lead to the impossibility of

calculating the absorption values from the measurement results.

In addition, the experiment showed the relationship between the parameters of heating radiation and the physical parameters of crystalline quartz at which the observed competition corresponds to the measurement of the absorption coefficient close to the limit. This makes it possible to estimate the value of the absorption coefficient available for measurement using the TPCI scheme in various transparent dielectrics. The design features of the TPCI scheme, which distinguish it from its prototype, i.e. the PCI scheme, are presented. These features make it possible to achieve a sensitivity in which one can observe the competition between various physical processes that distort the results of measurements in synthetic crystalline quartz.

2. Main Features of the TPCI Scheme

The so-called photothermal common-path interferometry (PCI) [2-3] scheme is widely used to measure the low absorption of transparent dielectrics. Fig. 1 shows a simplified installation of photothermal common-path interferometry (PCI). The specificity of the PCI scheme is such that, for calibration, samples should be used that are completely identical to those measured, with the exception of the absorption value, which in the control sample should be sufficiently large to be measured using conventional methods. This moment greatly complicates the measurement, and in certain cases, makes it impossible [8].

In [8], we proposed a new time-resolved photothermal common-path (TPCI) scheme, which is a modification of the PCI scheme. Fig. 2 shows a simplified setup of the TPCI scheme. Both schemes are based on the same principle, namely registration of the absorbed energy with the help of the refractive index response from the induced thermal deformations.
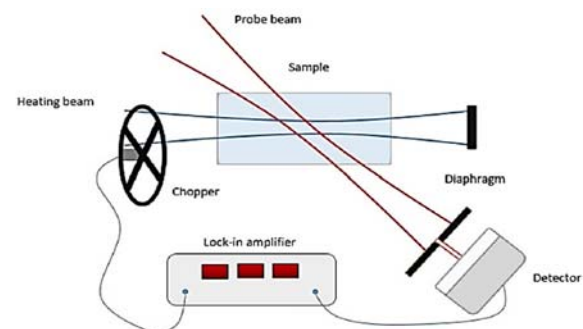

Fig. 1. PCI setup.

Here, we provide a brief description of the principle of both schemes. In both schemes, a sample was heated using the radiation ($\lambda = 1071$ nm) of a Yb fiber laser. In this study, the radiation was polarised and the output laser energy was varied from 1 to 40 mJ

by the laser control unit. The heated region of the sample in the TPCI scheme was illuminated coaxially using a linearly polarized probe beam of a single-mode continuous-wave laser. (λ = 630 nm; output power, 2 mW; and noise power, 0.1 % of the output power). In the PCI scheme, the beams intersected at an angle. The probe beam radiation was scattered through a time-varying spatial distribution of the refraction index, leading to the appearance of a time-varying component of the probe beam power, which was registered using a system comprising a detector (photodiode), diaphragm, diffuse plate, and projecting lens (not shown in Fig. 1).
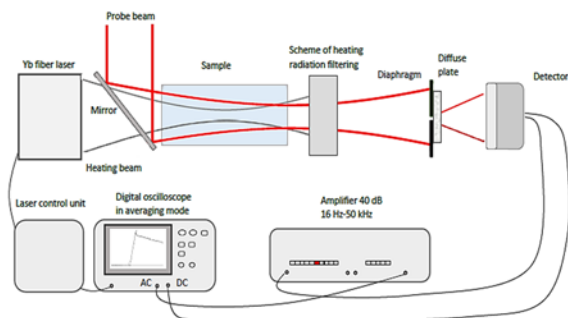

Fig. 2. TPCI setup.

The main differences between the PCI and TPCI schemes are the propagation of the heating and probe laser beams at an angle, the heating beam intensity modulation through a mechanical chopper, and the registration of the scattered component using the so-called synchronous detection method. In the synchronous detection method, the spectral component of a repetitively pulsed signal is measured at the modulation frequency of the heating beam power given based on the chopper characteristics.

The purpose of creating a new TPCI scheme was to use the calibration method, in which samples with known absorption value, thermoelastic and photothermal parameters are used for calibration, the values of which are guaranteed through the metrological procedures of their measurements. Here, we used a standard Russian K8 glass for such a sample [11]. The presence of such a sample opens the possibility of carrying out reliable absolute measurements, provided that there exists an appropriate theory that relates a signal obtained for a calibration isotropic sample with a signal generally obtained for a non-isotropic measured sample. To implement this approach, the axisymmetric configuration of the experimental scheme using coaxial heating and probe beams was chosen, which allowed solving the rather simple problem of probe beam diffraction by deformations caused by non-uniform heating taking into account the resulting stresses.

A detailed description of the solution to this problem for isotropic dielectrics is given in our previous study [8].

The proposed scheme has numerous design features that increase the sensitivity and information content, which is particularly significant when measuring the minimum measurable absorption in synthetic quartz.

Firstly, it is obvious that this refers to the coaxiality of the heating and probe beams, which became possible during the experiment as the result of applying an effective spectral filtering of the heating radiation (1071 nm) from the probe radiation (630 nm) with a suppression ratio of ~10^{-8}.

Secondly, the TPCI scheme uses an electronic control circuit. This control circuit has a simple and fast setting of the duration and repetition rate of the generated pulse of the heating laser. A periodic sequence of pulses is formed owing to the external modulation of the radiation power of a fiber laser. With this modulation method, the start of a digital oscilloscope sweep is conducted with a slight jitter. This allows the use of a digital oscilloscope in averaging mode over numerous events (~10^5), leading to noise suppression and an increasing resolution of the ADC from 8 to 11 bits [12]. In this case, when measuring an extremely low absorption of ~10^{-7} cm^{-1}, the signal-to-noise ratio increases to an acceptable level ($\approx$50:1).

Thirdly, the new scheme has a temporary resolution of the process of heating a sample with laser radiation and its cooling. The characteristic duration of the heating pulse ranges from 10 μs to 3 ms. The repetition rate is from 10 to 100 Hz

The sensitivity of our circuit was based on the sensitivity of the diode (300 mV per 1 mW at the probe laser wavelength of 630 nm), the gain of the amplifiers and was limited by the final (after averaging) noise of the electronic circuit, the main contribution to which was made through probe He-Ne laser noise (10^{-3} of 2 mW of radiation power).

In [8-9], we experimentally demonstrated the possibility of measuring the values of the absorption coefficients at $\alpha \sim 10^{-7}$ cm^{-1} in quartz glasses (QG) and synthetic crystalline quartz (SCQ). As a result, at a sufficiently low absorption of $\sim 2.5 \times 10^{-6}$ cm^{-1}, a nonlinear response of the refractive index was observed in the field of the heating beam.

Taken from [8] Fig. 3 shows the pulse of the variable component of the power of the probe beam, observed in Suprasil 311 glass, with a rectangular shape the heating pulse. Within the heating pulse, the inertia-less response of the probe beam variable component is clearly visible. This response was due to the nonlinear dependence of the refractive index on the heating beam field. As the calculation shows, the nonlinear response corresponds to the sum of two mechanisms responsible for the nonlinear dependence of the refractive index on the heating beam field, namely electronic nonlinearity and striction. The contribution of the thermo-optical component was measured when the rectangular heating pulse ended, when the contribution of the nonlinear component was zero, and the thermo-optical signal changed little when the heated region cooled.

Thus, the use of a rectangular heating pulse and temporal resolution made it possible to separate the contribution to the change in the refractive index of two components: first, the nonlinear dependence of the refractive index on the heating beam field, and second, the thermal contribution associated with thermal deformations.
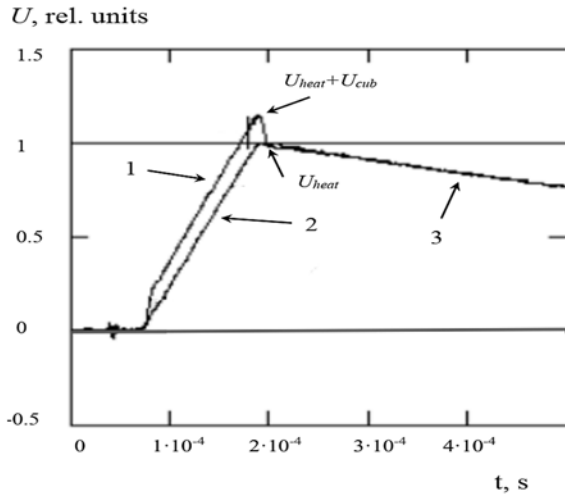

Fig. 3. Waveforms of the variable component of the probe beam with a pulse of 100 W and a duration of 100 μs: 1 – in Suprasil 311 quartz glass with an absorption of 2.6×10^{-6} cm^{-1}; 2 – in KU-1, quartz glass absorption of 10^{-5} cm^{-1} and 3 – time dependence of the signal during cooling of the samples.

To reliably measure the absorption in our scheme, we can increase the duration of the heating pulse, while maintaining its power, which increases the thermal contribution and leaves the nonlinear response unchanged. This technique is made possible owing to the operational control of the temporal characteristics of the heating laser using an electronic control circuit and the observation in time of the process of heating and cooling the sample in a pulsed periodic mode. This procedure was used by our group in [9] when measuring the absorption coefficient of 2×10^{-7} cm^{-1} in SK-1310 quartz glass (Ohara), in which the duration of the heating pulse was increased to 1 ms.

In turn, with an increase in the duration of the heating pulse, a spreading of the temperature profile during the heating by the laser beam may appear. The temporal resolution of the waveform distortion caused by this process allows the use of a theoretical calculation to make an amendment when calculating the absorption coefficient. As demonstrated later, this moment is fundamental in measurements of extremely low absorptions available for measurement.

3. Experiment

The above-described action using the proposed scheme makes it possible to apply a successful measurement of the absorption values of ultra-pure

QG at $\sim 10^{-7}$ cm^{-1} with sufficient accuracy (determined using a signal-to-noise ratio of 50:1).

However, the absorption values obtained were not the limiting ones possible for measuring with the help of the thermo-optical effect, although they were close to the maximum sensitivity of the scheme when measured in quartz glasses. A possible modernisation of the scheme to increase its sensitivity currently does not make sense owing to the lack of quartz glass samples of $\alpha \leq 10^{-7}$ cm^{-1}.

As it turns out, it is possible to observe the effects associated with the masking of the thermooptic effect by other physical effects when measuring the absorption coefficient in SCQ.

To demonstrate the problems associated with such measurements, Fig. 4 shows characteristic waveforms of the probe radiation variable component, obtained through absorption measurements in SCQ.

As a sample absorption measurement, we used a plate of SCQ with a crystallographic axis C perpendicular to its plane. The plate thickness was 1.8 cm. The transverse size along the growth axis (crystallographic axis a_1) X was 8 cm, and along the perpendicular axis Y was 10 cm. The crystal was grown using the hydrothermal method developed by the Russian company Quartz Technologies, LLC, and had a total impurity concentration including alkali and gas–liquid and mineral elements not exceeding 2–3 ppm. The concentration of the OH group was approximately 80 ppm.

The waveforms in Fig. 4 were obtained when the heating beam propagated across the crystallographic axis C along a_1 axis. The crystal length in this direction was 8 cm. This is greater than the heating beam waist length, which is equal to 3.5 cm.
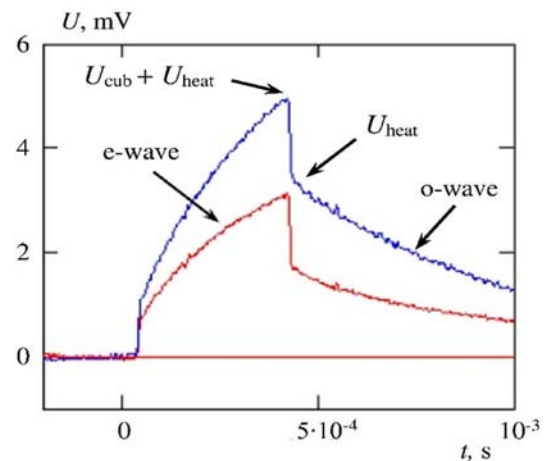

Fig. 4. Waveforms of the pulse response of the measuring system averaged over 10^5 events. The x-axis represents the time in seconds. The y-axis represents the voltage in millivolts of the variable component of the signal on the photodiode after passing through the amplifier.

Measurements along the C-axis were impossible owing to the small thickness of the quartz plate (1.8 cm) because, in our scheme, the signal was

masked by a signal related to the absorption of ambient air [8]. This was due to the coaxial propagation of the focused heating and probe beams along a considerable distance through the air outside the sample, due to which a strong component appeared in the observed signal associated with the heating of the surrounding air.

In the process of studying the absorption, it turned out that the signal caused by the thermo-optical effect in SCQ is much weaker than when measured in quartz glass, although their absorptions are comparable. In this case, the contribution of the nonlinear dependence of the refractive index to the signal begins to dominate. In the theoretical part of this study, it is shown that this occurs from the effect of stresses arising in the local heating region and a reduction of the thermal deformations.

The heating pulse with a power of up to 100 W, not shown in the oscillogram, had a rectangular shape with fronts of ~2 μs and duration of $\tau_{imp} \approx 400$ μs. The magnitude of the absorption was measured at the point U_{heat}, at which the field of the heating pulse decreases to zero and the magnitude of the nonlinear response is zero. The pulse parameters, as shown in the figure, are easily measurable. The signal-to-noise ratio was ~50:1.

The decrease in the system response to the thermal effects in the experiment led to an increase in the observed signal component associated with the cubic nonlinear dependence of the refractive index of the material on the heating beam field, which prevented an absorption measurement. For a reliable measurement of the absorption, it was necessary to increase the duration of the heating pulse, while maintaining its power, which increased the thermal contribution and left the nonlinear response unchanged. However, as it turned out, when the duration of the heating pulse exceeded a certain value, the procedure became ineffective because a further increase led to a rapid saturation of the signal dependence on the pulse duration and a decrease in the reliability of the calculations.

The optimal pulse duration τ_{imp} was defined as 400 μs. We associate the observed effect with an increase in the transverse temperature distribution of the heated region owing to the thermal conductivity, which is described through the diffusion equation. During this process, it can be seen that the time τ_{diff}, during which the cylindrical temperature Gaussian spatial distribution in the transverse direction expands twofold, is $\tau_{diff} = \frac{C_p \rho b_h^2}{4 \Lambda_m}$. Here, $b_h = 48$ μm is the minimum radius of the heating laser beam in the sample at the level of 1/e intensity, $C_p = 0.71$ J/g×deg is the heating capacity, $\rho = 2.65$ g/cm³ is the density, $\Lambda_m = \frac{\Lambda_\parallel + \Lambda_\perp}{2}$ W/cm×deg is the average thermal conductivity when the heating beam spreads across the crystallographic axis ($\Lambda_m = 0.085$ W/cm×deg) and $\Lambda_\parallel = 0.107$ W/cm×deg and $\Lambda_\perp = 0.062$ W/cm×deg) are the thermal conductivity along and across the crystallographic axis, respectively. In our case, τ_{diff}

was 130 μs, and the ratio of a certain optimal pulse duration to the time of the thermal diffusion τ_{diff} can be expressed using the equality $\tau_{imp} \approx 3 \times \tau_{diff}$.

To correctly calculate the absorption coefficient, it is necessary to consider the effect of heat diffusion in the calculations. This procedure was carried out in the framework of the Gaussian spatial distribution approximation used in [8], and will be published in a separate paper. The correction to the measured signal U_{heat} for this waveform is approximately 2 when considering the saturation of the time dependence.

Such a ratio of time essentially distinguishes these measurements from measurements carried out in QG [8], as shown in Fig. 3, where $\tau_{imp} \approx 100$ μs with $\tau_{diff} \approx 700$ μs, i.e. $\tau_{imp} \ll \tau_{diff}$.

To calculate the absorption from the voltage measured at the point U_{heat}, it is necessary to know the value of the thermo-optical parameter P for a particular crystal orientation during the experiment. Discussion of this issue is contained in [10] as well as in the theoretical part of the present study. According to a preliminary estimate of P, the absorption coefficient corresponding to the pulses shown was $(6 \times 10^{-7} \pm 30\ \%)$ cm⁻¹.

As noted in the introduction, another important feature of our scheme is the suppression of noise masking the signal at the input of a digital oscilloscope. To understand the essence of this process, we consider the conditions of the experiment under which the signal waveform data are obtained. The digitisation quantum of our oscilloscope ADC in this implementation was 390 μV. In this case, the full amplitude of the noise at the ADC input was approximately 100 mV (i.e. the sweep sensitivity was 10 mV/div). Further, as follows from the figure, the amplitudes of the measured pulse signals after averaging over $N = 10^5$ events are 3 and 5 mV. The noise component level is estimated at 50 μV, which is 7-times smaller than a digitised quantum. In the averaging process, the noise level decreased in accordance with ~ $1/\sqrt{N}$, and the resolution of the oscilloscope's ADC increased from 8 to 11 bits. A more detailed explanation of this process can be found in [12].

4. Calculation of Thermo-optical Parameter

The conclusion that the sensitivity of the scheme is reduced when measuring the absorption in crystalline quartz has found theoretical confirmation when solving the problem of diffraction of a probe beam on the induced spatial-temporal heterogeneity of the refractive index caused by local thermal deformations of the sample.

This theory for isotropic samples is described in [8]. From the solution to the problem, it follows that the signal measured in our scheme is proportional to the product $P \times \alpha$, where P is the thermo-optical parameter, which is an analogue of *dn/dT*, taking into

account the effect of stresses during non-uniform heating. Therefore, following [8], the notation $P = \left(\frac{\partial n}{\partial T}\right)_{eff}$ can be used. In anisotropic media, the theory differs by expressions for thermal deformations, which lead to dependencies P on the values of the components of the photoelastic tensor and stiffness tensor for different orientations of the sample being measured relative to the focused laser beam.

In this study, theoretical analyses were carried out for media with a trigonal symmetry of class 32, 3 m, $\overline{3}\,\overline{m}$. Crystalline quartz belongs to class 32.

To obtain the dependence of the coefficient P on the physical parameters of the crystal, we obtained solutions to the problem of thermo-elasticity for an infinite cylinder (the transverse size of the heated region is much smaller than the sample length) made from a crystal with a trigonal symmetry. We considered two cases, namely the cylinder generatrix along or perpendicular to the crystallographic axis C. The plane deformation approximation was used, and it was also believed that the characteristic distance of the heated region to the free boundary was much larger than the transverse size of this region. For this purpose, a special approach was developed to solve the problem, considering the specifics of our axially symmetric scheme. This decision will be described in detail in a separate publication. Herein, we present only the corresponding expressions P for two different experimental geometries calculated, for definiteness, using data on the physical constants given in [13-16].

1. With beam propagation along the growth axis X (crystallographic axis a₁) and perpendicular to the C-axis, the polarisation of the heating beam corresponds to an ordinary wave, and the probe beam has the polarisation of an extraordinary wave:

$$P_X = \left(\frac{\partial n_e}{\partial T}\right)_{\sigma=0} + \frac{n_e^3}{2}\left(\alpha_\perp^T (p_{11} + p_{13}) + \alpha_\parallel^T p_{12}\right) \\ - n_e^3 \frac{(c_{12} + c_{13})\alpha_\parallel^T + (c_{11} + 2c_{13} + c_{33})\alpha_\perp^T}{3c_{11} + 2c_{13} + 3c_{33} + 4c_{44}}(p_{11} \\ + p_{13} + p_{14}) \qquad (1)$$

2. With beam propagation along the C-axis, the polarisation of the heating and probe beams corresponds to an ordinary wave:

$$P_C = \left(\frac{\partial n_o}{\partial T}\right)_{\sigma=0} + \frac{n_o^3}{2}\left(\alpha_\perp^T (p_{11} + p_{12}) + \alpha_\parallel^T p_{13}\right) \\ - \frac{n_o^3}{4}\frac{c_{13}\alpha_\parallel^T + (c_{11} + c_{12})\alpha_\perp^T}{c_{11}}(p_{11} + p_{12}), \qquad (2)$$

where $\left(\frac{\partial n_{o,e}}{\partial T}\right)_{\sigma=0}$ is the derivative of the refractive index with respect to temperature for the o- or e-wave, measured with uniform heating and free expansion; n is the refractive index of an o- or e-wave; p_{11}, p_{12}, and p_{13} are the components of the photoelastic tensor; c_{11}, c_{12}, c_{13}, c_{33}, and c_{44} are the components of the stiffness tensor; and $\alpha_\parallel^T$ and $\alpha_\perp^T$ are the coefficients of the temperature linear expansion parallel and perpendicular to axis C, respectively.

Using the various values of the constants listed above for crystalline quartz taken from [13-16], we found that the possible value may be in the range $P_C = (1.5\text{-}2.5)\times 10^{-6}\ K^{-1}$. Such uncertainty is primarily associated with various data on the values of material constants in expression (4). For comparison, we give the values of P for QG from [8]: $P_{QG} = 9.3\times 10^{-6}\ K^{-1}$, that is, the relation $P_{QG} = (3\text{-}5.5)\times P_C$ takes place. Since the observed signal is proportional to the value of $P\times\alpha$ and, therefore, with equal values of absorption α, the observed signals in the TPCI scheme will decrease by 3–5.5 times when going from measuring absorption in cylindrical quartz glass samples to measuring absorption in cylindrical crystalline quartz samples.

Measurement of the absorption of ordinary waves in crystalline quartz for the propagation of beams along the C-axis in our scheme is unavailable. However, using the measurement results in the crystal, when the beams propagate across the C-axis (along the growth axis a₁), this value can be calculated because the absorption of an ordinary wave does not change under the propagation along and across the C axis: $\alpha = 6\times 10^{-7} \pm 30\ \%\ cm^{-1}$ (see the Experiment section). As mentioned above, uncertainty is related to the difference in the material constants cited in different sources.

5. Discussion

It follows from the above material that, in quartz glasses at an absorption of $\sim 10^{-7}\ cm^{-1}$, and when the detection scheme reaches a sufficient sensitivity, the contribution to the changes in the refractive index of its nonlinear dependence on the heating beam field can be observed. However, this contribution, given in our experiments in [9-10], was rather small, and with the temporal resolution of the process of changing the refractive index in the field of the heating beam with sufficient accuracy, is separated from the thermo-optical contribution.

However, already in this example, it can be seen that when registering changes in the refractive index using synchronous detection, an error will occur associated with different forms of the recorded pulse in the calibration pulse, which does not have a non-linear contribution, and in the measured sample. This is shown in Fig. 3, in which the pulse shape of the variable component of the test beam in KU-1 glass corresponds to the pulse shape in the calibration sample.

We are now interested in the magnitude of the limiting absorption coefficient available for measurement with sufficient accuracy in the TPCI scheme for quartz glasses, for crystalline silicon within the near IR range. We refer to this as α_{min}.

In this paper, the α_{min} value was obtained in synthetic crystalline quartz in the TPCI scheme, which

has sufficient sensitivity. This limit is associated with the competition of three effects.

The first is the thermo-optical effect, which is determined based on the joint dependence of the refractive index on the temperature at a constant density $(\frac{\partial n}{\partial T})_\rho$ and its dependence $(\frac{\partial n}{\partial \rho})_T$ on density ρ, which changes with the deformations depending on the stresses arising from the local heating. This effect is determined based on the energy density of the heating beam.

The second is the nonlinear dependence of the refractive index on the heating beam field, which has two components, striction and electron components [8]. The electronic nonlinearity at the temporal scale of the considered processes is practically inertia-less. The time required for establishing the stationary value of the striction component is related to the time for sound to travel along the waist of the heating radiation beam, which is ~5 μs [8]. Under certain circumstances, this contribution can significantly affect the procedure for the separation of nonlinear and thermo-optical contributions. This effect is determined by the power density of the heating beam.

In addition, owing to the thermal diffusion, there is a limiting duration of a heating pulse at which the thermo-optical contribution to the measured signal reaches the limit. In this case, the optimal ratio of the characteristic time of heat diffusion τ_{diff} and the duration of the heating pulse τ_{imp} is $\tau_{imp} \approx 3 \times \tau_{diff}$. With a longer duration, it becomes impossible to calculate the true signal, which should be observed in the absence of heat diffusion. Thus, the third competing effect is the heat diffusion of the temperature distribution in the field of the heating beam, which determines the magnitude of the limiting contribution of the thermo-optical effect.

In our scheme, when measured in the SCQ with an optimal heating pulse duration of 400 μs, the absorption coefficient $\alpha = 6 \times 10^{-7}$ cm^{-1} corresponds to a signal of 3 mV, and the signal-to-noise ratio is ~ 50:1. The accuracy when measuring the signal used later in the mathematical calculation of the absorption coefficient can be estimated as $\approx \pm 1$ %. The magnitude of such noises in our scheme is determined primarily by the noise of the probe laser and the averaging procedure of the waveforms over a sufficiently large number of pulses N of ~10^5, which leads to a decrease in the noise amplitude, and an increase in the ADC resolution. With a relatively small contribution to the signal of the nonlinearity of the refractive index, for example, in a CG with an absorption coefficient of $\geq 10^{-7}$ cm^{-1}, the absorption coefficient available for measurements with sufficient accuracy in the TPCI scheme is determined by the noise present on the waveform of the variable component of the test beam.

In addition, as shown in Fig. 4, with SCQ, the observed contribution of the nonlinear dependence of the refractive index on the heating beam field to the signal approximately corresponds to the thermo-optical contribution. In samples with a lower

absorption coefficient, even with an increase in the signal-to-noise ratio, the measurement accuracy of the thermo-optical contribution will begin to be determined based on the contribution of the establishment of the stationary value of the striction nonlinearity [8] arising at the trailing edge of the heating pulse. In addition, the measurements will be distorted by the finite time of the impulse response of the measuring path, which has a complex shape in its damped part.

In this regard, the level of the absorption coefficient at which the maximum thermo-optical contribution will be equal to the nonlinear response of the refractive index can be taken as the criterion at which all problems described arise, which is denoted as α_{min}.

Knowing the optimal ratio of the heating pulse duration and heat diffusion time $\tau_{imp} \approx 3 \times \tau_{diff}$, as well as the values of the thermo-optical parameters and coefficients of the cubic nonlinearity of the refractive index in QG and silicon, its α_{min} can be calculated. To do so, consider Formula (7) from [8] for the magnitude of the power P_{pulse} of the variable component of the probe radiation proportional to $U_{heat} + U_{cub}$ in the case of a rectangular heating pulse:

$$P_{pulse} = K \left[\frac{P_h * \tau_{pulse}}{\pi b_h^2 * c * \rho} \left(\frac{\partial n}{\partial T} \right)_{eff} \alpha_h + n_2 \frac{P_h}{\pi b_h^2} \right], \quad (3)$$

where P_h is the power, τ_{pulse} is the duration of the heating radiation pulse, and n_2 is the coefficient in the cubic dependence of the refractive index on the field of the heating beam. This coefficient depends on the orientation of the crystal. However, considering the estimated nature of our calculations, we do not take this dependence into account in crystalline quartz, and set the value of n_2 equal to its value in QG. In addition, the formula considers the linearity of the polarisations and the mutual orthogonality of the fields of the probe and heating beams. The coefficient K is determined using the parameters of the TPCI scheme [8].

Therefore, from the condition of equality of the maximum thermo-optical contribution, realized in accordance with the experiment using $\tau_{pulse} \approx 3 \times \tau_{eff}$ to the nonlinear contribution of the refractive index, α_{min} can be determined:

$$\alpha_{min} = \frac{0.8 * \Lambda_m * n_2}{b_{heat}^2 \left(\frac{\partial n}{\partial T} \right)_{eff}}, \quad (4)$$

where the square of the heating beam radius b_{heat}^2 is selected from the relation $(\frac{2\pi * b_{heat}^2}{\lambda_{heat}} \leq L_{sample}$ (where L_{sample} is the sample length). The characteristic value b for L_{sample} of ~5–10 cm is ~50 μm. Taking this into account, it follows from the above formula that α_{min} depends mainly on the physical parameters of the samples.

The Table 1 summarises the calculations of the most interesting parameters that determine α_{min}. For crystalline quartz, the calculation gives

$\alpha_{min}=4\times10^{-7}$ cm^{-1}, which corresponds to the experimental value of 6×10^{-7} cm^{-1}. The difference can be related to both the difference between the nonlinearity coefficient of a crystal and its value in quartz glass, and to the uncertainty of the calculated value of the thermo-optical parameter. The repetition rate of the heating pulses given in the table will determine the accumulation time required to obtain a value N of $\sim10^5$ when the signal is averaged using a digital oscilloscope. In the case of the QG, this will take more than 1 h, which will determine the requirements for the stability of the measurement conditions.

For crystalline silicon, the absorption coefficient limit, which can be measured using the photothermal method, can be estimated as $\sim 10^{-5}$ cm^{-1}.

That is, even with the uncertainty of our estimates, it can be said with confidence that the required level of measurement in crystalline silicon 10^{-11} cm^{-1} using the TPCI method in the presented configuration is unattainable. This statement applies even more to the PCI scheme. Finally, we can state that a measurable absorption level of 10^{-8} cm^{-1} is the limit for quartz glasses. At present, it seems that this level of absorption is unattainable in QG with modern production technologies.

In conclusion, it should also be noted that the described measurements are possible only with the resolution of the time dependence of the variable component of the power of the probing radiation. In the case of a synchronous detection scheme, the error of the measured absorption coefficient under the described conditions will differ many times from its actual value, and the α_{min} value determined above can be considered as the measurement limit using the synchronous detection method.

Table 1. Physical constants of various materials that determine the maximum low absorption available for measurement.

	Crystalline quartz $\lambda_{heat} =$ **1071 nm**	**Quartz glass** $\lambda_{heat} =$ **1071 nm**	**Silicon** $\lambda_{heat} =$ **1550 nm**
Λ_m (W/cm×K)	0.085	0.014	1.48
$n_2 \times 10^{16}$ (cm^2/W)	2.8	2.8	250 [17] 560 [18]
$\left(\frac{\partial n}{\partial T}\right)_{eff} \times 10^6$ (K^{-1})	1.5	8.4	215
α_{min} (cm^{-1})	4×10^{-7}	1.6×10^{-8}	0.58×10^{-5} 1.34×10^{-5}
τ_{imp} (µs)	400	2000	20
Repetition rate (Hz)	100	20	2000

6. Conclusions

In this paper, we experimentally demonstrated the main problems that arise when measuring the low absorption in crystalline quartz using a TPCI scheme.

1) A decrease in the sensitivity of the measuring scheme in a quartz crystal was found in comparison with quartz glass. The calculation of the thermo-optical parameter for crystalline quartz showed that this is due to a decrease in the temperature change of the refractive index during heating due to the stresses that appear.

2) With a heating pulse duration of 400 µs, the effect of the heat diffusion was found, which determines the limiting duration of the heating pulse. This is the duration that allows a correct calculation of the absorption coefficient from the measurement results.

3) Owing to the temporal resolution in the TPCI scheme, a nonlinear refractive index response in the heating beam field was observed, equal to the thermo-optical one, when we measured the absorption coefficient of 6×10^{-7} cm^{-1}. Thus, the incorrectness when measuring the reduced absorption coefficient using the synchronous detection method was demonstrated.

4) For quartz glass and crystalline silicon, maximum low absorption available for measurement and the optimal duration of the heating pulse during its measurement have been calculated.

5) In addition, our research showed that the absorption of our synthetic crystalline quartz with a total impurity concentration of 1-3 ppm corresponds to the best samples of quartz glass known: 2×10^{-7} cm^{-1} (Ohara SK-1310), 8×10^{-7} cm^{-1} (Suprasil 300) [9], which are used to make various elements for output stages of high-power CW lasers.

This leads to an important practical conclusion that by virtue of the ratio of thermo-optical constants for disk elements made of QG and SCQ [10], as well as the ratio of their thermal conductivities of 1:4.5, the transmitted CW radiation will experience at least an order of magnitude less distortion of wave front in SCQ compared to the QG. Thus, this material has great potential advantages over quartz glasses in the manufacture of flat elements installed at the output of high-power technological CW laser systems.

Acknowledgements

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