

## STATIC COMPONENT OF PHOTOTHERMAL RESPONSE IN NON-TRANSPARENT SAMPLES \*

Zlatan Šoškić<sup>1</sup>, Slobodanka Galović<sup>2,3</sup>, Nebojša Bogojević<sup>1</sup>,  
Slobodan Todosijević<sup>1</sup>

<sup>1</sup>Faculty of Mechanical and Civil Engineering in Kraljevo, University of Kragujevac

<sup>2</sup>Institute of Nuclear Sciences "Vinča", University of Belgrade, Belgrade, Serbia

<sup>3</sup>Joint Institute for Nuclear Research, Bogoliubov Laboratory for Theoretical Physics,  
Dubna, Russia

**Abstract:** *The paper presents the analysis of the static component of temperature distribution in non-transparent samples during photothermal measurements. Analytical expressions for static part of temperature distribution in the irradiated sample and in its surroundings are determined using one dimensional model of heat transfer in a typical photothermal environment. It is established that the dominant factors that influence the shape and the mean value of the temperature distribution are optical absorption coefficient and thermal conductances of the sample and the surroundings. Important special cases are described and analytical expressions for temperatures of the front and the back side of the sample are derived.*

**Key words:** *photothermal effect, non-destructive characterization*

### 1. INTRODUCTION

Photothermal (in the following text abbreviated as PT) effect comprises generation of heat in a sample irradiated by light. Measurement techniques based on PT effect are being studied and applied as an alternative non-destructive tool for measurement of thermal, optical and other related physical properties, especially when standard methods are not applicable [1-4]. Besides, the PT measurement techniques are increasingly used for investigations of subsurface structure and macroscopic defects [1-6].

PT methods are based on direct or indirect recording of the surface temperature variations, which are caused by generation and transfer of heat that is produced as consequence of absorption of light. Usual sources of light are harmonically modulated beams of ordinary light sources or pulse laser beams [1-14], and the resulting temperature variation,

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**Corresponding author:** Zlatan Šoškić

Faculty of Mechanical and Civil Engineering in Kraljevo, University of Kragujevac • E-mail: soskic.z@mfkv.kg.ac.rs

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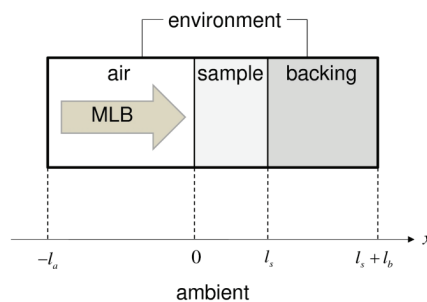
which represents the response of the system, consists of two components: a static component and a time-dependent component.

Vast majority of research efforts is focused to analyses of the time-dependent component of the PT response [7-14] for two reasons: the first reason is that the time-dependent component is relevant for determination of dynamic thermal properties of samples, and the second reason is lower influence of noise to direct or indirect detection of the time-dependent component [15]. However, this paper deals with the static component of the PT response, which is of relevance for the setup of instrumental configurations for measurements, as well as for proper interpretation of results of some photothermal methods [7].

To this purpose, the paper presents a model of heat propagation in a typical PT measurement setup, which results in a system of linear differential equations with the respective boundary conditions. The paper considers only optically non-transparent samples, because they cannot be studied by conventional optical techniques, and on the other hand, they are more convenient for PT measurements because of the higher heat generation [1-14]. The system of equations is solved in an analytic form, and the obtained results are discussed.

## 2. MODEL

A typical configuration for PT measurements is schematically presented in Fig. 1. A solid sample with a flat front surface and the length  $l_s$  is mounted on a backing with the length  $l_b$ , and its front surface is exposed to a light beam, which passes through an air column with the length  $l_a$ . The air and the backing present the environment of the sample, and the ambient temperature, which is initial temperature of the sample and the environment is  $T_{amb}$ . As a consequence of absorption of light by the sample, it is heated, and due to transfer of the heat, temperature distributions in the sample  $T^{(s)}$ , the air column  $T^{(a)}$ , and the backing  $T^{(b)}$  change. Temperature variations due to PT heating, defined as  $T - T_{amb}$ , are proportional to the PT response signal.



**Fig. 1** A typical setup for PT measurements (MLB stands for the modulated optical beam)

Two assumptions are usual [7-14] in the analyses of a typical PT response:

- that the light intensity is uniform over the irradiated surface of the sample, and
- that the sample is homogenous over all cross-sections.

Under these assumptions, for symmetry reasons, the temperature distribution over the cross-sections of the sample is uniform, and the temperature varies only along the direction of the incident light beam. Hence, the heat transfer may be considered one dimensional. For the description of the heat transfer and the PT response there is selected an axis with the direction and orientation of the light beam, and with the origin at the surface of the sample, denoted as the  $x$ -axis in Fig. 1. Therefore, the PT response may be represented as a function of only two variables,  $T - T_{amb} = T(x,t) - T_{amb}$ . The samples for PT measurements are prepared so that the assumptions are valid in the vast majority of practical cases. Since no further assumptions regarding the setup of the measurement are introduced, the following analysis has wide applicability.

The common approach to determination of the temperature variations due to the heat generation and transfer is based on the energy conservation law and a constitutive equation that relates heat flux to the temperature variations. The usual approach assumes application of the Fourier's constitutive equation [17], but, as briefly reported in [18], it is not appropriate approximation in many modern applications, including PT methods [19, 20]. The very interesting discussion about generalization of heat conduction equation may be found in papers [18-26]. Therefore, the temperature variations of PT response in one dimensional case are described by the hyperbolic differential equation [18-21]:

$$\frac{\partial^2 T(x,t)}{\partial x^2} - \frac{1}{D} \left[ \frac{\partial T(x,t)}{\partial t} + \tau \frac{\partial^2 T(x,t)}{\partial t^2} \right] = -\frac{1}{k} \left[ S(x,t) + \tau \frac{\partial S(x,t)}{\partial t} \right] \quad (1)$$

where the transfer of the heat through a medium is described by the heat conductivity  $k$ , the relaxation time of thermal processes  $\tau$  and the thermal diffusivity of the medium  $D = k/C$ , with  $C$  standing for the volumetric heat capacity of the medium. The generation of the heat is described by volumetric heat generation rate  $S(x,t)$ , defined later.

Since the heat transfer in PT measurements in the investigated setup occurs through the air column, the sample and the backing, three differential equations of type (1), with three different sets of material characteristics  $D$ ,  $\tau$  and  $k$  are needed to describe the heat flow in PT experiments. Besides, the right-hand sides of the equations differ, because it can be assumed that the heat generation due to irradiation by the light beam occurs only in the sample. For typical PT experiment setups with non-transparent samples the following assumptions can be adopted:

- that the absorption of the light in the air column is negligible in comparison to the absorption by the sample, and
- that almost all light energy transmitted to the sample is absorbed within the sample, so that reflections from the backing, absorption of the light by backing and multiple reflections of light beam may be neglected.

Then the differential equations of the model are:

$$\frac{\partial^2 T^{(a)}(x,t)}{\partial x^2} - \frac{1}{D_a} \left[ \frac{\partial T^{(a)}(x,t)}{\partial t} + \tau_a \frac{\partial^2 T^{(a)}(x,t)}{\partial t^2} \right] = 0, \quad x < 0 \quad (2a)$$

$$\frac{\partial^2 T^{(s)}(x,t)}{\partial x^2} - \frac{1}{D_s} \left[ \frac{\partial T^{(s)}(x,t)}{\partial t} + \tau_s \frac{\partial^2 T^{(s)}(x,t)}{\partial t^2} \right] = -\frac{1}{k_s} \left[ S(x,t) + \tau \frac{\partial S(x,t)}{\partial t} \right], \quad 0 < x < l_s \quad (2b)$$

$$\frac{\partial^2 T^{(b)}(x,t)}{\partial x^2} - \frac{1}{D_b} \left[ \frac{\partial T^{(b)}(x,t)}{\partial t} + \tau_b \frac{\partial^2 T^{(b)}(x,t)}{\partial t^2} \right] = 0, x > l_s \quad (2c)$$

The solution of the system of differential equations has to satisfy physical conditions of continuity of the temperature variations and the flux on interfacial surfaces between the air column, the sample and the backing:

$$T^{(a)}(x=0,t) = T^{(s)}(x=0,t) \quad (3a)$$

$$q^{(a)}(x=0,t) = q^{(s)}(x=0,t) \quad (3b)$$

$$T^{(s)}(x=l_s,t) = T^{(b)}(x=l_s,t) \quad (3c)$$

$$q^{(s)}(x=l_s,t) = q^{(b)}(x=l_s,t) \quad (3d)$$

where the dependences between the temperatures  $T^{(i)}$  and heat fluxes  $q^{(i)}$  are given by the respective constitutive equations [18-21]:

$$q^{(i)}(x,t) + \tau^{(i)} \frac{\partial q^{(i)}(x,t)}{\partial t} = -k_i \frac{\partial T^{(i)}(x,t)}{\partial x}, i = a,s,b \quad (3e)$$

To be solved, a system of differential equations needs certain boundary conditions. In the case of the considered model, boundary conditions should define the temperatures at the ends of the model domain, so at the beginning of air column ( $x = -l_a$ ) and at the end of the backing ( $x = l_s + l_b$ ). In all practical PT measurements, the lengths of the air layer and the backing layer are sufficient to neglect the temperature variations far from the sample:

$$T^{(a)}(x = -l_a, t) = T_{amb} \quad (4a)$$

$$T^{(b)}(x = l_s + l_b, t) = T_{amb} \quad (4b)$$

As it was already mentioned, in a PT experiment, intensity of the light beam is modulated, and it can be expressed as  $I(t) = I_0 f(t)$ , with  $I_0$  standing for the amplitude of the intensity of the light beam, and  $f(t)$  for the intensity modulation function. The usual modulations are the harmonic modulation, with  $f(t) = (1 + \cos \omega t)/2$ , where  $\omega$  represents the circular frequency of the harmonic modulation, and the pulse modulation,  $f(t) = [h(t) - h(t-T)]$ , with  $h(t)$  being the Heaviside (step) function, and  $T$  the duration of the excitation pulse. If  $R$  is optical coefficient of reflection of the surface of the sample, than the intensity of light transferred to the sample is  $(1-R) \cdot I(t)$ . Due to light absorption in the sample, its intensity decreases, and can be expressed as  $I(x,t) = (1-R) \cdot I(0,t) \cdot \exp(-\beta x)$ , with  $\beta$  standing for the coefficient of absorption of light in the sample. One of the consequences of light absorption is the heating of the sample, and if the quantum efficiency of conversion of light into heat in the sample is  $\eta$ , then the volumetric heat generation rate in the sample during PT measurements is:

$$S(x,t) = -\frac{dI}{dx} = I_0 (1-R) \eta \beta \exp(-\beta x) f(t) \quad (5a)$$

In the case of harmonic modulation, the expression for the volumetric heat generation has the form

$$S(x, t) = S_0 \exp(-\beta x)(1 + \cos \omega t) \quad S_0 = \beta \eta (1 - R) \frac{I_0}{2} = \beta q_0 \quad (5b)$$

and in the case of the pulse modulation it has the form

$$S(x, t) = S_0 \exp(-\beta x)(h(t) - h(t - T)) \quad (5c)$$

where  $S_0$  represents volumetric heat generation rate at the surface of the sample, and  $q_0$  the static component of the incident heat flux transferred from the sample to the surroundings. Equations (2)–(5) represent the model that will be used in this paper. Similar models have been already used for various investigations of photothermal and photoacoustic phenomena [19, 20, 27-32].

### 3. RESULTS

If the modulation of the light beam is harmonic, the excitation in PT measurements can be represented as a sum of two components, one of them being static, while the other is time-dependent:

$$S(x, t) = S_{stat}(x) + S_t(x, t) \quad S_{stat}(x) = S_0 \exp(-\beta x) \quad S_t(x, t) = S_0 \exp(-\beta x) \cos \omega t \quad (6a)$$

In the case of pulse modulation, the excitation in PT measurements can be represented by static component during the interval  $t_c < t < T$ , where  $t_c$  stands for the time constant of the sample [33]:

$$S(x, t) = S_{stat}(x) \quad S_{stat}(x) = S_0 \exp(-\beta x) \quad t_c < t < T \quad (6b)$$

Since the differential equations in the model for the PT response are linear, the PT response also has to have two components, one static and the other time-dependent:

$$T(x, t) - T_{amb} = \theta(x) + \vartheta(x, t) \quad (7)$$

The static component  $\theta(x)$  is the response to the static component of excitation  $S_{stat}(x)$ , and the time-dependent component is the response to the time-dependent component of excitation  $S_t(x)$ .

As explained in the introduction, the goal of this paper is to present calculation of the static component  $\theta(x)$  that determines the mean temperature of the surface of the sample, which is of relevance for the setup of photothermal measurements, but also influences the indirect component of the photoacoustic response.

By inserting the expression (7) into the differential equations of the model (2), conditions of continuity (3) and boundary conditions (4), and following separation of static and time-dependent components, one obtains a set of differential equations for the static component of the PT response:

$$\frac{\partial^2 \theta^{(a)}(x)}{\partial x^2} = 0, \quad x < 0 \quad (8a)$$

$$\frac{\partial^2 \theta^{(s)}(x)}{\partial x^2} = -\frac{1}{k_s} S_0 \exp(-\beta x), \quad 0 < x < l_s \quad (8b)$$

$$\frac{\partial^2 \theta^{(b)}(x)}{\partial x^2} = 0, x > l_s \quad (8c)$$

with conditions of continuity being

$$\theta^{(a)}(x=0) = \theta^{(s)}(x=0) \quad (9a)$$

$$q^{(a)}(x=0) = q^{(s)}(x=0) \Rightarrow \frac{1}{k_a} \frac{d\theta^{(a)}(x)}{dx} \Big|_{x=0} = \frac{1}{k_s} \frac{d\theta^{(s)}(x)}{dx} \Big|_{x=0} \quad (9b)$$

$$\theta^{(s)}(x=l_s) = \theta^{(b)}(x=l_s) \quad (9c)$$

$$q^{(s)}(x=l_s) = q^{(b)}(x=l_s) \Rightarrow \frac{1}{k_s} \frac{d\theta^{(s)}(x)}{dx} \Big|_{x=l_s} = \frac{1}{k_b} \frac{d\theta^{(b)}(x)}{dx} \Big|_{x=l_s} \quad (9d)$$

and boundary conditions

$$\theta^{(a)}(x=-l_a) = 0 \quad (10a)$$

$$\theta^{(b)}(x=l_s+l_b) = 0 \quad (10b)$$

Differential equations (10) determine the form of the solution for the PT response  $\theta(x)$  as

$$\theta^{(a)}(x) = A_0^{(a)} + A_1^{(a)} \cdot x, x < 0 \quad (11a)$$

$$\theta^{(s)}(x) = A_0^{(s)} + A_1^{(s)} \cdot x - \theta_\beta \cdot \exp(-\beta x), 0 < x < l_s \quad (11b)$$

$$\theta^{(b)}(x) = A_0^{(b)} + A_1^{(b)} \cdot x, x > l_s \quad (11c)$$

with  $\theta_\beta = S_0/(k_s \beta^2)$ , and  $A_0^{(i)}, A_1^{(i)}$  ( $i = a, s, b$ ) being six constants that should be determined on the basis of conditions of continuity and boundary conditions given by six linear equations (9) and (10). This model is equivalent to the model presented in [7], meaning that the common and the generalized model of heat conduction give the equivalent results in the static regime.

Combination of the continuity condition (9a) and the boundary condition (10a) enables one to express the static component of PT response in air,  $\theta^{(a)}(x)$ , in dependence of the temperature of the front (irradiated) surface of the sample  $\theta_a = \theta_s(0)$ :

$$\theta^{(a)}(x) = \theta_a \left( 1 + \frac{x}{l_a} \right), x < 0 \quad (12a)$$

while the combination of the continuity condition (9c) and the boundary condition (10b) enables one to express the static component of PT response in backing,  $\theta^{(b)}(x)$ , in dependence of the temperature of the back (opposite to the irradiated) surface of the sample  $\theta_b = \theta_s(l_s)$ :

$$\theta^{(b)}(x) = \theta_b \left( 1 - \frac{x-l_s}{l_b} \right), x > l_s. \quad (12b)$$

By setting the values  $x=0$  and  $x=l_s$  into the (11b) it is possible to express the constants  $A_1^{(s)}$  and  $A_2^{(s)}$ , as well as the temperature distribution in the sample  $\theta^{(s)}(x)$ , also using the temperatures of the front and the back surface of the sample  $\theta_a$  and  $\theta_b$ :

$$\theta^{(s)}(x) = \{\theta_a + \theta_b[1 - \exp(-\beta x)]\} - \{(\theta_a - \theta_b) + \theta_b[1 - \exp(-\beta l_s)]\} \frac{x}{l_s}, 0 < x < l_s. \quad (12c)$$

The front and the back surface temperatures of the sample  $\theta_a$  and  $\theta_b$  may be now calculated from the (12), conditions of the continuity of the heat flux (9b) and (9d):

$$\theta_a = \frac{[1 - \exp(-\beta l_s)] + g_b \left\{ 1 - \frac{1}{\beta l_s} [1 - \exp(-\beta l_s)] \right\}}{g_a + g_b + g_a g_b} \frac{q_0}{(k_s / l_s)}$$

$$\theta_b = \frac{[1 - \exp(-\beta l_s)] - g_a \left\{ \exp(-\beta l_s) - \frac{1}{\beta l_s} [1 - \exp(-\beta l_s)] \right\}}{g_a + g_b + g_a g_b} \frac{q_0}{(k_s / l_s)} \quad (13)$$

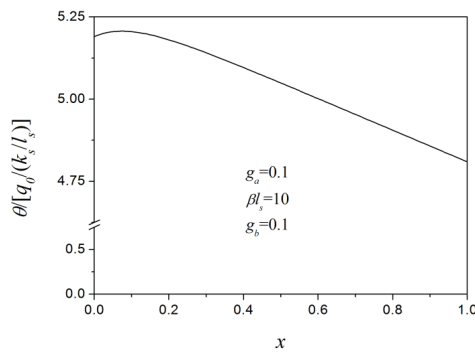
where  $g_i = (k_i / l_i) / (k_s / l_s)$  ( $i = a, b$ ) represent parameters of each PT measurement setup, since they depend on material and dimensions of the selected sample, the selected backing and the length of air column. The physical meaning of the parameters  $g$  for the considered one-dimensional case is that  $g_i$  represents relative thermal conductance of the respective medium (the air column for  $g_a$  and the backing for  $g_b$ ), i.e. the ratio between the thermal conductance of the medium and the thermal conductance of the sample.

#### 4. ANALYSIS

The equations (12) and (13) describe the general solution of the considered model of static component of PT response. The solutions of PT response in the air column and in the backing are linear, with the temperature decreasing with distance from the sample. The solution in the sample consists of two parts: an exponential part and a linear part. The graph of temperature distribution that illustrates temperature distribution in a PT sample is shown in Fig. 2. The temperature in the sample does not change monotonously, having the maximal value at the distance

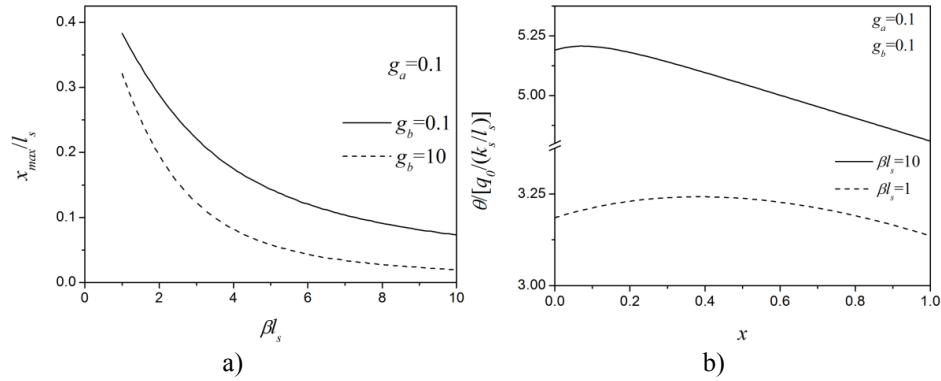
$$x_{\max} = \frac{1}{\beta} \ln \frac{g_a + g_b + g_a g_b}{g_b + \frac{g_a}{\beta l_s} \{ \exp(-\beta l_s) + g_b [1 - \exp(-\beta l_s)] \}} \quad (14)$$

from the irradiated surface.



**Fig. 2** Normalized temperature distribution for a PT setup described by parameters shown in the graph

The responses in PT experiments are proportional to the front and/or the back surface temperatures  $\theta_a$  and  $\theta_b$ , and as (13) shows, the surface temperatures of the sample depend on relative thermal conductances of the air column ( $g_a$ ) and backing ( $g_b$ ), as well as on characteristics of light absorption in the sample, measured by the product  $\beta l_s$ .



**Fig. 3** Influence of optical absorption coefficient of sample on: a) position of maximum of temperature distribution b) temperature distribution in a sample. The parameters of the PT setups are shown in graphs

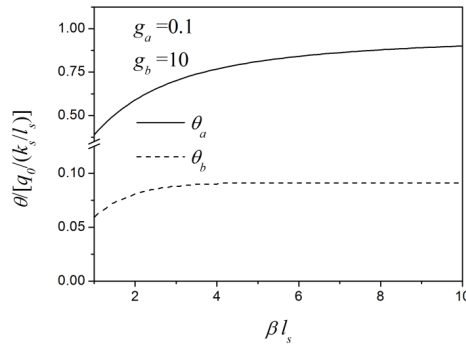
#### 4.1 The influence of absorption of the light in the sample

Optical absorption properties of the sample, expressed through the coefficient of absorption  $\beta$ , influence spatial distribution of the heat generated in the sample, and consequently, the temperature distribution within the sample. Larger value of  $\beta l_s$  means that the absorption of the light energy occurs in thinner layer behind the front surface of the sample. If absorption occurs close to the irradiated surface of the sample, than the maximum of the temperature arises closer to the surface, as it is shown in Fig. 3a, and the overall shape of the temperature distribution is closer to linear, as it is shown in Fig. 3b.

Coefficient of absorption also influences the surface temperatures of a sample, increasing the difference between temperatures of the front and the back side of a sample, as it is shown in Fig. 4. Temperature of the front surface of a PT sample increases with increase of the coefficient of absorption because of the increased amount of the generated heat and because the higher values of coefficient of absorption mean that temperature maximum is closer to the front surface of the sample. At sufficiently high values of optical absorption coefficient, the temperature maximum occurs so close to the surface of the sample that the further increase of optical absorption coefficient does not cause significant increase of the front surface temperature, and it reaches a saturation plateau.

The temperature of the back surface of the sample shows similar behaviour, but it shows saturation at lower values of optical absorption coefficient, because of the increased distance between the maximum of the temperature distribution and the back surface of the sample. Therefore, relative change in distance between the maximum of the temperature distribution and the back surface of the sample became negligible at smaller values of optical absorption coefficient.

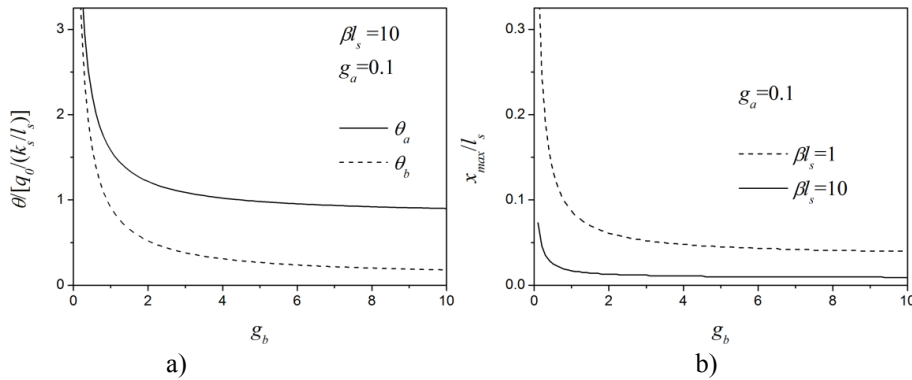




**Fig. 4** Influence of optical absorption coefficient of sample on temperature of surfaces of a sample. The parameters of the PT setups are shown in graphs

**4.2 The influence of thermal conductance of surroundings**

Thermal conductance of the air and the backing determine heat fluxes transferred on front and back side of the sample. Therefore, the conductances influence the temperature distribution in the sample, reducing the temperatures in the sample with increase of the conductances of surroundings. Such behaviour is illustrated in Fig. 4a that shows influence of thermal conductance of backing on the front and back temperature of a sample.



**Fig. 5** Influence of thermal conductance of backing on: a) surface temperature of a sample b) position of maximum of temperature distribution. The parameters of the PT setups are shown in graphs

On the other hand, thermal conductances of surroundings generally do not affect significantly the shape of thermal distribution in the sample (the position and value of maximal temperature) in PT measurements, which is illustrated in the Fig. 4b that shows influence of thermal conductance of backing on position of cross-section with maximal temperature in the sample. Thermal conductances affect the shape of thermal distribution only if the sample is transparent.

### 4.3 Samples with surface absorption

A very important practical case of non-transparent samples are opaque samples, which completely absorb light in a narrow region close to the irradiated surface, which is described by the condition  $\beta l_s \gg 1$ . In that case, the expressions for static components of temperatures of the front and the back side of the sample, given by equations (13) simplify, have a symmetric form:

$$\theta_a = \frac{1 + g_b}{g_a + g_b + g_a g_b} \frac{q_0}{(k_s / l_s)} \quad \theta_b = \frac{1 + g_a}{g_a + g_b + g_a g_b} \frac{q_0}{(k_s / l_s)} \quad (15)$$

The obtained expressions for the PT response may be further simplified for important practical cases. Since the air is a heat insulator with thermal conductivity considerably smaller than the thermal conductivities of the sample ( $k_a \ll k_s$ ), and that the length of air column in PT experiments is usually considerably larger than the length of the sample ( $l_a \gg l_s$ ), it holds  $g_a \ll 1$ . The expressions for the temperatures of the surfaces of an opaque sample in PT measurements are further reduced to the form:

$$\theta_a = (g_b + 1)\theta_b \quad \theta_b = \frac{1}{g_a + g_b} \frac{q_0}{(k_s / l_s)} \quad (16)$$

Thermal conductance of backing, expressed through  $g_b$ , varies depending on thermal properties of the material that is used, and the material of the backing depends on the experimental setup.

The lowest practical values of the thermal conductance of the backing are obtained in the cases when the backing is air ( $g_b = g_a$ ), which happens in various experimental setups when the measured response depends on temperature of the back side of the sample. Therefore, in the case of an opaque sample with air on both sides, the expressions for the temperatures of the surfaces of a sample in PT measurements have the form:

$$\theta_a = \frac{1 + g_a}{2} \frac{q_0}{(k_a / l_a)} \quad \theta_b = \frac{1}{2} \frac{q_0}{(k_a / l_a)} \quad (17)$$

The highest values of the thermal conductance of the backing are obtained in the cases when the backing is metal, when it can be adopted that  $g_b \gg 1 \gg g_a$ . Therefore, in the case of an opaque sample on a metal backing, the expressions for the temperatures of the surfaces of a sample in PT measurements have the form:

$$\theta_a = \frac{q_0}{(k_s / l_s)} \quad \theta_b = \frac{q_0}{(k_b / l_b)} \quad (18)$$

## 5. CONCLUSION

The paper presents an analysis of a static component of PT response based on one dimensional model of heat transfer in typical PT measurement configuration. The solution of the model determines spatial distribution of temperature in PT sample and its surroundings, and leads to an analytical expression, given by (13), for temperatures of the front side and back side of the sample.

It is concluded that the temperature distribution and the temperatures of the front and the back side of the sample depend on the optical absorption coefficient of the sample and on the ratios between thermal conductances of surroundings to thermal conductance of the sample. The dependences of the static component of PT response on these parameters were analysed, and it is shown that the optical absorption coefficient has dominant influence on the shape of the distribution of the temperature within PT sample, while the ratios of thermal conductances of the surroundings to the thermal conductance of the sample have dominant influence on the mean value of the temperature of the sample.

A very important special case of an opaque sample is discussed further and a very simple expression for temperatures of the front and the back side of the PT sample, given by (16), is derived. The expressions are further simplified for the limiting cases of an opaque sample with air in backing, given by (17) and an opaque sample with metal backing, given by (18).

The obtained expressions for temperatures of the sample, given by equations (13), (16), (17) and (18), are not influenced by dynamic thermal properties and can be applied in classic or generalized approximation of photothermal generated heat conduction.

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