ANALYSIS OF PHOTOTHERMAL RESPONSE OF THIN SOLID FILMS BY ANALOGY WITH PASSIVE LINEAR ELECTRIC NETWORKS

by

Slobodanka P. GALOVIĆ, Zlatan N. ŠOŠKIĆ, and Marica N. POPOVIĆ

Original scientific paper
UDC: 620.1:681.785.42/.43
DOI: 10.2298/TSCI0904129G

The paper presents conditions that should be met in order to make the photothermal induced temperature variations of a solid sample analogous to the voltage variations of the electric network with passive linear elements. Further analysis shows that such analogy enhances experimental determination of the thermal properties of thin solid layers by photothermal frequency method.

Key words: *photothermal response, thermal wave, transmission-line theory*

Introduction

Photothermal (PT) measurement techniques are being intensively developed and applied with increased success to the measurement of thermal, optical, and other related physical properties, as well as for the investigation of subsurface structure and macroscopic defects of various samples [1-6].

PT methods are based on direct or indirect recording of surface temperature variations that arise from the generation and transfer of heat produced as a consequence of the absorption of laser radiation of modulated intensity by a sample. The resulting signal depends on the amount of generated heat (depending on the coefficients of optical absorption and the efficiency of the heat-to-light conversion of the sample) and on the heat transfer process (hence on the sample's thermal conductivity, coefficient of thermal diffusivity and other thermophysical properties). Therefore, PT methods have broad capabilities as tool for non-destructive characterization of various materials.

In order to determine the physical properties of the investigated structure, it is necessary, as the first step, to develop a mathematical model that sufficiently well describes physical processes leading from the optical excitation to the thermal response (direct problem), and then to solve the inverse problem of determining the physical properties of the system once the optical excitation, thermal response and model are known. The inverse problem is usually solved by application of curve-fitting. However, non-linear fitting being a rather complex procedure, quite often gives ambiguous results [7, 8]. There fore it is worthy of effort to investigate approximate solutions of the problem that enable determining the physical properties of the system without non-linear curve-fitting, by analyzing only certain ranges of PT signal in function of the modulation frequency.

This paper deals with one such solution of the inverse problem in analysis of the PT response, the case when a mathematical model of the PT induced surface temperature variations is analogous to the model of the flow of electric current through an electric network consisting of passive linear elements (resistors, capacitors, and coils). In that case, already developed methods of solving the inverse problem in passive linear electric networks may also be applied to solve the inverse problem of characterization of materials by PT methods.

The first part of the paper presents an analysis of a model of the PT heat propagation in order to determine conditions under which the investigated analogy holds. After that, the analytic expression for the PT response under those conditions is determined, and finally, a method of solving the inverse problem in the case under consideration is presented, and its potentials for application for characterization of material properties are discussed.

Model

Propagation of the PT induced heat through the sample

The analysis presented in the paper considers a typical PT configuration schematically presented in fig. 1. A solid sample of length d_s, mounted on backing of length d_b, is exposed to radiation of optical beam passing through air layer with length d_a. Air and backing present the environment of the sample surrounded by ambient with temperature T_{amb}. As a consequence of the absorption of light by the sample, the sample is heated, and due to heat transfer, temperatures of the sample (T_s), air layer (T_a), and backing (T_b) are changed. Temperature variations due to time-dependent PT heating, defined as $\vartheta \triangleq T - T_{dc} \quad T_{amb}$, are proportional to the PT response [5]. T_{dc} is the steady-state temperature variation.

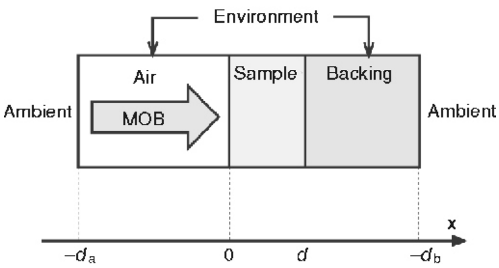

Figure 1. Typical PT experiment setup
MOB stands for modulated optical beam

As the heating of the sample's surface can be considered uniform over the entire heated surface, it can be assumed that temperature is uniform over all cross-sections and that it varies only along the direction of the incident light beam (x-axis in fig.1.). Hence, the heat transfer may be considered one-dimensional, and the validity of this assumption is already recognized [5]. Therefore, the PT response should be represented as a function of only two variables, $\vartheta = \vartheta \;\, x, t$.

Temperature variations arise due to the transfer of heat generated by the absorbed radiation, and they are, in one-dimensional case, described by energy balance equation:

$$C_v \frac{\partial T(x,t)}{\partial t} \quad S(x,t) \quad \frac{\partial q(x,t)}{\partial x} \tag{1}$$

where C_v stands for the volumetric specific thermal capacity of the medium, $S(x, t)$ – for the volumetric heat generation rate, and $q(x, t)$ – for the heat flux. Taking into account thermal memory and heat conduction effects, the heat flux depends on the temperature gradient, and this fact is expressed by the following equation [5, 9-11]:

$$q(x,t) \quad \tau \frac{\partial q(x,t)}{\partial \tau} \quad k \frac{\partial \vartheta(x,t)}{\partial x} \tag{2}$$

where k is the heat conductivity, and τ – the relaxation time of thermal processes in the medium.

In the case of a homogeneous and opaque sample, it can be assumed that the incident light is absorbed by a very small region of the sample, so that the heat generation rate can be expressed as $S(x, t) = S(t)\,\delta(x)$, and thus thermal variations and heat flux may be described by a set of hyperbolic homogeneous differential equations of the second order:

$$\frac{\partial^2 \vartheta_i(x,t)}{\partial x^2} \quad \frac{1}{D_i}\frac{\partial^2 \vartheta_i(x,t)}{\partial t} \quad \frac{\tau_i}{D_i}\frac{\partial^2 \vartheta_i(x,t)}{\partial t^2} \tag{3}$$

$$\frac{\partial^2 q_i(x,t)}{\partial x^2} \quad \frac{1}{D_i}\frac{\partial^2 q_i(x,t)}{\partial t} \quad \frac{\tau_i}{D_i}\frac{\partial^2 q_i(x,t)}{\partial t^2} \tag{4}$$

where the index i denotes part of the considered system ($i = a, s$, and b stands for air, sample, and backing, respectively) and D stands for thermal diffusivity of the medium ($D = k/C$).

To finalize the model for the description of the PT response, eqs. (3) and (4) are to be completed with homogeneous initial conditions:

$$\vartheta_a(x, t = 0) = 0 \text{ and } q_a(x, t = 0) = 0, \tag{5}$$

homogeneous boundary conditions:

$$\vartheta_a(x \quad -d_a, t) \quad 0 \quad \text{and} \quad \vartheta_b(x \quad d \quad d_b, t) \quad 0, \tag{6}$$

and conditions of continuity of temperature and heat flux on interfacial surfaces:

$$\begin{gathered}\vartheta_a(x \quad 0,t) \quad \vartheta_s(x \quad 0,t) \quad \text{and} \quad q_a(x \quad 0,t) \quad q_s(x \quad 0,t) \quad S(t) \\ \vartheta_s(x \quad d,t) \quad \vartheta_b(x \quad d,t) \quad \text{and} \quad q_s(x \quad d,t) \quad q_b(x \quad d,t)\end{gathered} \tag{7}$$

Analogy with flow of electric current through lines

Analogies between heat conduction and electric conduction processes were already investigated and applied to solving thermal problems [6, 11-16], but presenting models neither includes passive linear electrical network analogy nor conditions that should be met in order to make this analogy. In the considered case of the model of the PT response presented in the previous chapter, an analogy with a model of electric current flow through homogeneous lines may easily be derived. The voltage between the lines, $u(x, t)$, and the electric current passing through them, $i(x, t)$, satisfy the following equations:

$$c\frac{\partial u(x,t)}{\partial t} \quad \frac{\partial i(x,t)}{\partial x} \tag{8}$$

$$\frac{\partial u(x,t)}{\partial x} \quad ri \quad l\frac{\partial i}{\partial t} \tag{9}$$

where r stands for distributed resistance ($r = \mathrm{d}R/\mathrm{d}x$), c – for distributed capacitance ($c = \mathrm{d}C/\mathrm{d}x$), and l for distributed inductance ($l = \mathrm{d}L/dx$) of the line.

From eqs. (8) and (9), hyperbolic homogeneous differential equations of the second order describing voltage and electric current, the so-called telegraphy equations, may easily be derived:

$$\frac{\partial^2 u(x,t)}{\partial x^2} \quad rc\frac{\partial u(x,t)}{\partial t} \quad lc\frac{\partial^2 u(x,t)}{\partial t^2} \tag{10}$$

$$\frac{\partial^2 i(x,t)}{\partial x^2} \quad rc\frac{\partial i(x,t)}{\partial t} \quad lc\frac{\partial^2 i(x,t)}{\partial t^2} \tag{11}$$

An analogy between the differential equations describing the PT response – eqs. (3) and (4), and the flow of electric current through lines – eqs. (8) and (9), is established by introducing the following relationships:

$$u \quad \vartheta, i \quad q, r \quad \frac{1}{k}, l \quad \frac{\tau}{k}, c \quad C_v \tag{12}$$

However, by introducing analogy (12), it is stated only that heat conduction through one homogeneous layer (air, sample or backing) can be described by an electric current flow through homogeneous lines [6, 11-16].

Since in PT frequency methods the excitation of the system (light beam) is modulated by amplitude, the generated heat can be described as $S(t) = S_0\cos(\omega t)$, and it is suitable to employ the Fourier transform to analyze the problem. Symbol ω signifies modulation frequency. Temperature variation and heat flux can be represented by their complex representatives $\tilde{\vartheta}_i(x)$ and $\tilde{q}_i(x)$:

$$\vartheta_i(x,t) \quad \sqrt{2}\vartheta_i(x)\cos[\omega\tau + \theta_i(x)] \quad \mathrm{Re}\{\sqrt{2}\tilde{\vartheta}_i(x)\} \tag{13}$$

$$q_i(x,t) \quad \sqrt{2}q_i(x)\cos[\omega\tau + \psi_i(x)] \quad \mathrm{Re}\{\sqrt{2}\tilde{q}_i(x)\} \tag{14}$$

The analogous problem is solved by application of complex representatives for voltage and current, $\tilde{U}$ and $\tilde{I}$, respectively:

$$u(x,t) \quad \sqrt{2}U(x)\cos[\omega t \quad \theta(x)] \quad \mathrm{Re}\{\sqrt{2}\tilde{U}(x)\} \tag{15}$$

$$i(x,t) \quad \sqrt{2}I(x)\cos(\omega t \quad \psi_i(x)) \quad \mathrm{Re}\{\sqrt{2}\tilde{I}(x)\} \tag{16}$$

The symbols θ and ψ signify the phase lag of dynamic temperature $\vartheta(x,t)$, – or voltage $u(x, t)$ and heat flux $q(x, t)$ – or electric current $i(x,t)$, from dynamic source.

Then, the telegraph equations become ordinary linear differential equations of the second order in the complex domain (j is imaginary unit):

$$\frac{d^2\tilde{U}(x)}{dx^2} \quad (j\omega rc \quad \omega^2 lc)\tilde{U}(x) \tag{17}$$

$$\frac{d^2\tilde{I}(x)}{dx^2} \quad (j\omega rc \quad \omega^2 lc)\tilde{I}(x) \tag{18}$$

It is convenient to write linear differential equations of the second order in the following form:

$$\frac{d^2\tilde{U}(x)}{dx^2} \quad \tilde{\sigma}^2\tilde{U}(x) \quad \textit{and} \quad \frac{d^2\tilde{I}(x)}{dx^2} \quad \tilde{\sigma}^2\tilde{I}(x) \tag{19}$$

and it follows that $\tilde{\sigma}(x)$ is given by:

$$\tilde{\sigma}_i \quad \sqrt{\tilde{z}\tilde{y}}, \quad \tilde{z} \quad r \quad j\omega l, \quad \tilde{y} \quad j\omega c \tag{20}$$

$\tilde{\sigma}(x)$ is called complex transmission coefficient, while $\tilde{z}$ and $\tilde{y}$ are the distributed impedance and admittance of the line.

The general solution of eqs. (17) and (18), taking into account the relation between voltage and electric current, is given by expressions:

$$\tilde{U}(x) \quad A_1\exp(\quad \tilde{\sigma}_i x) \quad A_2\exp(\tilde{\sigma}_i x) \tag{21}$$

$$\tilde{I}(x) \quad \frac{A_1}{\tilde{Z}_c}\exp(\quad \tilde{\sigma}_i x) \quad \frac{A_2}{\tilde{Z}_c}\exp(\quad \tilde{\sigma}_i x) \tag{22}$$

where $\tilde{Z}_c$ stands for the characteristic impedance of the line:

$$\tilde{Z}_c \quad \sqrt{\frac{\tilde{z}}{\tilde{y}}} \tag{23}$$

while A_1 and A_2 are constants which are to be determined from the boundary conditions.

Model of the environment and surface thermal sources

In order to develop an electric network that will be the analog of the PT induced system under consideration, the boundary and interfaces conditions – eqs. (6) and (7), should be employed. The boundary conditions, eq. (6), essentially mean that the layers of air and backing are much longer than the layer of the sample, suggesting that the environment of the sample should be modeled as very long (semi-infinite) lines.

In the case of very long (semi-infinite) lines, the constant A_2 has to be zero, to provide finite values for voltage and current when $x \to \infty$. Therefore, for very long lines there holds the equation (independent of x)

$$\frac{\tilde{U}(x)}{\tilde{I}(x)} \quad \tilde{Z}_c \quad \text{const.} \tag{24}$$

showing that a very long line can be modeled as electric element with impedance equal to the characteristic impedance of the line. Besides, the fact that $A_2 = 0$ also implies that near the end of very long lines voltage and electric current tend to zero.

The derived conclusion may be used to develop an analogous electric model for analysis of PT phenomena in opaque samples, which is presented in fig. 2. Layers of air and backing, from the point of view of their interactions with the sample, may be modeled with impedances and that have values:

$$\tilde{Z}_{ca} \quad \sqrt{\frac{1 \quad j\omega\tau_a}{j\omega k_a C_a}}, \quad \tilde{Z}_{cb} \quad \sqrt{\frac{1 \quad j\omega\tau_b}{j\omega k_b C_b}} \tag{25}$$

The sample is modeled by electric lines of length d and by the distributed impedance and admittance:

$$\tilde{z} \quad \frac{1}{k_s}(1 \quad j\omega\tau_s), \quad \tilde{y} \quad j\omega C_s \tag{26}$$

The generated heat at $x = 0$ is modeled by ideal current source giving electric current S_0, thus satisfying boundary conditions eq. (7). The values of voltages and electric currents in this circuit are equal to the values of temperature variations and heat flux at the respective points of the sample.

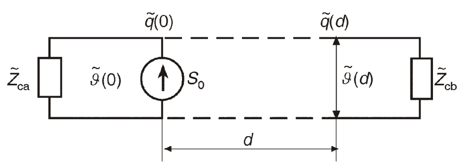

Figure 2. Analogous electric circuit where sample is modeled by electric lines

Model of the sample

PT applications to characterization and imaging of materials imply the direct or indirect measurement of temperature variations of one surface of the sample. Therefore, from the point of view of the analysis of the PT response, a relevant model of the sample should be able to describe temperature variations on the sample's surfaces, while temperature variations within the sample are not of particular interest. Therefore, the simplest relevant model of the sample should describe only the relations between surface temperature variations and heat fluxes, *i. e.* the simplest relevant analogous electric circuit can be described only by voltages and electric currents at the ends of the line.

Putting $x = 0$ and $x = d$ into eqs. (21) and (22), it can be easily shown that the voltages and the electric currents at the ends of the line of finite length d satisfy the following matrix equation:

$$\begin{bmatrix} \tilde{U}(0) \\ \tilde{I}(0) \end{bmatrix} = \begin{bmatrix} ch(\tilde{\sigma}d) & \tilde{Z}_{\mathrm{c}} sh(\tilde{\sigma}d) \\ \frac{1}{\tilde{Z}_{\mathrm{c}}} sh(\tilde{\sigma}d) & ch(\tilde{\sigma}d) \end{bmatrix} \begin{bmatrix} \tilde{U}(d) \\ \tilde{I}(d) \end{bmatrix} \tag{27}$$

It is usual in the theory of electric circuits to formally introduce total line impedance $\tilde{Z} = \tilde{z}_{\mathrm{s}} d$ and total line admittance $\tilde{Y} = \tilde{y}_{\mathrm{s}} d$, which can be used to rewrite the previous matrix equation in the form where only those parameters are used:

$$\begin{bmatrix} \tilde{U}(0) \\ \tilde{I}(0) \end{bmatrix} = \begin{bmatrix} ch(\sqrt{\tilde{Z}\tilde{Y}}) & \sqrt{\frac{\tilde{Z}}{\tilde{Y}}} sh(\sqrt{\tilde{Z}\tilde{Y}}) \\ \sqrt{\frac{\tilde{Y}}{\tilde{Z}}} sh(\sqrt{\tilde{Z}\tilde{Y}}) & ch(\sqrt{\tilde{Z}\tilde{Y}}) \end{bmatrix} \begin{bmatrix} \tilde{U}(d) \\ \tilde{I}(d) \end{bmatrix} = \begin{bmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{bmatrix} \begin{bmatrix} \tilde{U}(d) \\ \tilde{I}(d) \end{bmatrix} \tag{28}$$

Relation (28) is known in the theory of electric circuits as the representation of the electric network by *a*-parameters. It is also established that, for any given matrix of *a*-parameters, it is possible to compose a four-terminal electric network consisting of elements with concentrated parameters, which is described by the given matrix. Therefore, it is possible to compose an electric network consisting of the elements with concentrated (*not anymore distributed*) parameters, which has the same relations between the voltages and electric currents at the ends, described by eq. (28), as the considered line. The scheme of the network is presented in fig. 3, while the values of the impedance and admittance in the network are:

$$\tilde{Z}_{\mathrm{eq}} = \tilde{Z} \frac{th\left(\frac{\sqrt{\tilde{Z}\tilde{Y}}}{2}\right)}{\frac{\sqrt{\tilde{Z}\tilde{Y}}}{2}} \quad \text{and} \quad \tilde{Y}_{\mathrm{eq}} = \tilde{Y} \frac{sh\sqrt{\tilde{Z}\tilde{Y}}}{\sqrt{\tilde{Z}\tilde{Y}}} \tag{29}$$

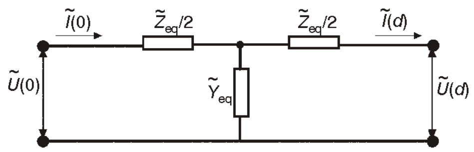

Figure 3. Electric network modeling the PT induced sample

As the considered line of finite length represents an electric model of the PT induced sample, the electric network from fig. 3 is an electric model of temperature variations and heat fluxes at the surfaces of the sample.

The impedance and the admittance representing the sample in fig. 3 should have the values:

$$\tilde{Z}_{eq} = \frac{d}{k}(1 + j\omega\tau) \frac{th\left(\frac{1}{2}\frac{d}{\sqrt{D\tau}}\sqrt{-(\omega\tau)^2 + j\omega\tau}\right)}{\frac{1}{2}\frac{d}{\sqrt{D\tau}}\sqrt{-(\omega\tau)^2 + j\omega\tau}}, \quad \tilde{Y}_{eq} = j\omega C \frac{sh\left(\frac{d}{\sqrt{D\tau}}\sqrt{-(\omega\tau)^2 + j\omega\tau}\right)}{\frac{d}{\sqrt{D\tau}}\sqrt{-(\omega\tau)^2 + j\omega\tau}} \tag{30}$$

The special case considers multi-layer structures, where each layer has different thermal characteristics. However, since the boundary conditions for the heat transfer between the layers consist of continuity of heat flux and temperature on interfacial surfaces, while the boundary conditions between the electric lines consist of continuity of voltage and electric current, multi-layer structures may be modeled by complex electrical network consisting of several stages, each stage representing one layer of multi-layer structure by electrical network in fig. 3. Such electric model for analysis of the PT response of a structure with n layers is presented in fig. 4.

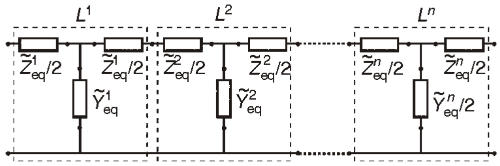

Figure 4. Analogous electrical network for the PT induced sample with *n* layers

Each layer L^m is represented by impedances and admittance that have values as determined by eq. (30).

While the circuit in fig. 3 has a rather simple structure, the impedance and the admittance cannot be represented by real electric elements (resistors, capacitors, and coils); so, the obtained analogous circuit is not suitable for application to standard methods of analysis of electric circuits.

Analogy with passive linear networks

Expressions for the equivalent impedance and admittance, eq. (29), of the PT induced sample may be expressed using the following series expansions:

$$\frac{sh(\sqrt{\tilde{Z}\tilde{Y}})}{\sqrt{\tilde{Z}\tilde{Y}}} \quad 1 \quad \frac{\tilde{Z}\tilde{Y}}{6} \quad \frac{\tilde{Z}^2\tilde{Y}^2}{120} \quad \ldots \text{ and} \tag{31}$$

$$\frac{th\dfrac{\sqrt{\tilde{Z}\tilde{Y}}}{2}}{\dfrac{\sqrt{\tilde{Z}\tilde{Y}}}{2}} \quad \frac{1 \quad \dfrac{\tilde{Z}\tilde{Y}}{24} \quad \dfrac{\tilde{Z}^2\tilde{Y}^2}{1920} \quad \ldots}{1 \quad \dfrac{\tilde{Z}\tilde{Y}}{8} \quad \dfrac{\tilde{Z}^2\tilde{Y}^2}{384} \quad \ldots} \tag{32}$$

Therefore, when the condition:

$$\frac{\tilde{Z}\tilde{Y}}{6} \ll 1 \tag{33}$$

is satisfied, it holds that:

$$\tilde{Z}_{eq} \quad \tilde{Z} \quad d(r \quad j\omega), \quad \tilde{Y}_{eq} \quad \tilde{Y} \quad j\omega Cd \tag{34}$$

In that case, the analogous electric network from fig. 3 may be presented by using only passive linear elements (resistors, capacitors, and coils), as it is shown in fig. 5.

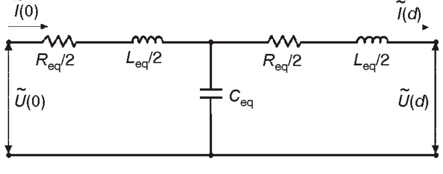

Figure 5. Analogous passive linear network model of the PT induced sample

The elements in a passive linear network (in further text also denoted by the abbreviation PLN) that is the model of a surface temperature variations of a PT induced sample has the following values:

$$R_{\mathrm{eq}} \quad \frac{d}{k}, \quad L_{\mathrm{eq}} \quad d\frac{\tau}{k}, \quad C_{\mathrm{eq}} \quad dC_v \tag{35}$$

When the PT response can be modeled by PLN, the matrix equation (28) reduces to:

$$\begin{matrix} \tilde{U}(0) \\ \tilde{I}(0) \end{matrix} \quad \begin{matrix} 1 \quad \dfrac{\tilde{Z}\tilde{Y}}{2} & \tilde{Z} \quad 1 \quad \dfrac{\tilde{Z}\tilde{Y}}{4} \\ \tilde{Y} & 1 \quad \dfrac{\tilde{Z}\tilde{Y}}{2} \end{matrix} \quad \begin{matrix} \tilde{U}(d) \\ \tilde{I}(d) \end{matrix} \tag{36}$$

The special case considers multi-layer structures, where each layer satisfies the condition – eq. (33). These structures may be modeled by PLNs consisting of several stages, each stage representing one layer of multi-layer structure by PLN in fig. 5.

Discussion

Conditions for modeling PT response by passive linear networks

It is already established that the condition to model analogous electric circuit by passive linear elements is expressed by eq. (33). Rewritten by electric line parameters, the relations take the form:

$$d^2(-\omega^2 lc + j\omega rc) \ll 6 \tag{37}$$

Electric line parameters may be used to express condition for the modeling in terms of the properties of the PT induced sample:

$$d^2 \quad \omega^2 \frac{t_{\mathrm{s}}}{6D_{\mathrm{s}}} \quad j\omega \frac{1}{6D_{\mathrm{s}}} \quad \ll 1 \tag{38}$$

which, being expressed by complex numbers, may be turned into conjunction of two conditions expressed by real numbers:

$$\frac{\omega^2}{\omega_{\mathrm{D}}\omega_{\mathrm{r}}} \ll 1 \quad \text{and} \quad \frac{\omega}{\omega_{\mathrm{D}}} \ll 1 \tag{39}$$

where the following symbols are introduced:

$$\omega_{\mathrm{r}} \quad \frac{1}{\tau_{\mathrm{s}}} \quad \text{and} \quad \omega_{\mathrm{D}} \quad \frac{6D_{\mathrm{s}}}{d^2} \tag{40}$$

The conditions given by eq. (39) can also be expressed as:

$$\frac{\omega}{\omega_\tau} \ll \frac{\omega_{\mathrm{D}}}{\omega_\tau} \quad \text{and} \quad \frac{\omega}{\omega_\tau} \ll \sqrt{\frac{\omega_{\mathrm{D}}}{\omega_\tau}} \tag{41}$$

Introducing critical thickness d_τ,

$$d_\tau \quad \sqrt{6D_{\mathrm{s}}\tau_{\mathrm{s}}} \tag{42}$$

defined so that:

$$\frac{\omega_{\mathrm{D}}}{\omega_\tau} \quad \frac{d_\tau}{d}^{\;2} \tag{43}$$

the condition for modeling the PT response by PLN – eq. (39), reduces to:

$$\omega \ll \omega_{\mathrm{t}} \; \frac{d_\tau}{d}^{\;n}, \quad \text{where} \quad \omega_\tau \;\; \frac{1}{\tau_s}, \quad d_\tau \;\; \sqrt{6D_{\mathrm{s}}\tau_{\mathrm{s}}}, \quad n \;\; \begin{matrix} 1, d \;\; d_\tau \\ 2, d \;\; d_\tau \end{matrix} \tag{44}$$

The derived condition (44) shows that the PT response of any sample can be modeled by PLN for sufficiently low frequencies. For a sample with thickness d_τ, the limiting frequency is ω_τ; for samples thinner than d_τ, the limiting frequency increases inversely proportionally to the thickness of the sample; for samples thicker than d_τ the limiting frequency decreases inversely proportionally to the square of the thickness of the sample. At the same time, it should be noted that when the modeling of the sample by PLN is possible, the samples thicker than d_τ can be modeled by networks consisting only of resistor and capacitor (RC networks), because thermal memory effects may be neglected at frequencies much lower than ω_τ.

The condition (44) is graphically presented in fig. 6: in order to apply the modeling by PLN, the modulation frequency and depth of the PT induced sample should describe a point deep within the shadowed area.

Table 1 presents a list of typical representatives of various classes of materials, together with respective critical thicknesses d_τ and frequencies ω_τ, calculated on the basis of bulk material properties.

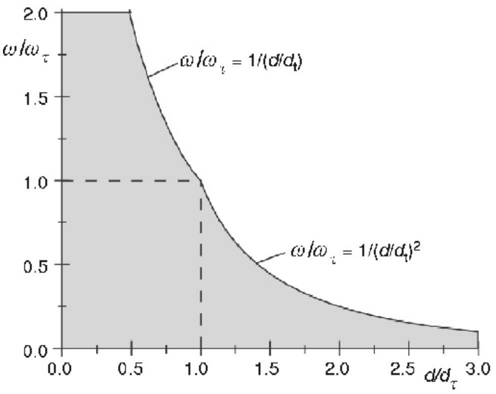

Figure 6. Graphical presentation of conditions for modeling PT induced sample by linear passive electric network

Table 1. A list of the critical thickness and the limiting frequencies

Material (representative)	d_τ [m]	ω_τ [s^{-1}]
Metal (aluminum)	$2.4\cdot10^{-8}$	10^{12}
Semiconductor (silicon)	$2.3\cdot10^{-6}$	10^{8}
Polymer (PVC)	$2.2\cdot10^{-5}$	10^{3}
Porous (dry sand)	$4.1\cdot10^{-3}$	10^{-1}
Organic (wood)	$3.6\cdot10^{-2}$	10^{-2}

The table suggests that thin layers (up to the order of micrometer) of crystalline materials may be modeled by PLN for all experimentally achievable modulation frequencies (of the order of 10 Hz to 100 MHz), while the thicknesses of the samples of non-crystalline materials, which may be modeled by PLN, are of the order of millimeter.

Characterization of PT sample properties by analogy with passive linear networks

This chapter discusses the possibilities of application of the developed analogy in the determination of the properties of PT induced samples. The analogy will be applied by using methods for frequency analysis of PLN to analysis of frequency dependence of the PT response.

In order to characterize the material properties of thin layers, frequency response of differential PT response of the so-called free standing sample is measured. The differential PT response is the ratio of temperature variations on the sample's surfaces, $\vartheta(d)/\vartheta(0)$, and free-standing sample is a sample that is surrounded by air or some other gas (ideally vacuum). Heat conductivities of gases are very low compared with solids; so, assuming that k_{air} 0, it can be considered that the characteristic impedances of air and backing in analogous electric model, $\tilde{Z}_{\mathrm{ca}}$ and $\tilde{Z}_{\mathrm{cb}}$, are infinite, so that electric currents passing through them are equal to zero.

The theory of PLN shows that frequency dependence of any voltage may be expressed in the complex domain by rational functions of frequency, and analyzed by applying analysis of zeroes and poles of the functions, as it will be presented later in the text.

In the case of the free-standing sample, $\tilde{I}(d)$ in the analogous electric circuit from fig. 5 equals to zero, and thus eq. (36) gives result:

$$\tilde{U}(0) \quad 1 \quad \frac{\tilde{Z}\tilde{Y}}{2} \quad \tilde{U}(d) \tag{45}$$

Therefore applying the analogy between the electric circuit and the PT induced sample, and the already established relations (12), (33), (35), (40), and (42), it can be concluded that:

$$\frac{\tilde{\vartheta}(d)}{\tilde{\vartheta}(0)} \quad G(j\omega) \quad \frac{1}{3 \quad \frac{d}{d_\tau}^{2} \quad j\frac{\omega}{\omega_\tau}^{2} \quad 3 \quad \frac{d}{d_\tau}^{2} \quad j\frac{\omega}{\omega_\tau} \quad 1} \quad \frac{\omega_{\mathrm{D}}\omega_\tau}{3s^2 \quad 3\omega_\tau s\omega_{\mathrm{D}}\omega_\tau} \tag{46}$$

where $s = j\omega$. The function $G(s)$ is, in fact, the transfer function of the PT induced sample if the temperature variation at the illuminated surface $\vartheta(0)$ is considered to be the system input, and the temperature variation on the opposite side of the PT sample $\vartheta(d)$ the output of the investigated system. $G(s)$ is a transfer function of the second order, and its behavior depends on the roots of its characteristic polynomial – which is the polynomial in denominator of $G(s)$ [17].

The physical interpretation of $G(j\omega)$ is that its modulus $|G(j\omega)|$ shows frequency dependence of the ratio of amplitudes of temperature variations $\vartheta(d)/\vartheta(0)$, and its argument $\arg[G(j\omega)]$ shows frequency dependence of the phase difference between the temperature variations $\vartheta(d)$ and $\vartheta(0)$. The expressions for $|G(j\omega)|$, which is the amplitude-frequency characteristic of the system, and $\arg[G(j\omega)]$, which is the phase-frequency characteristic of the system, are:

$$\frac{\vartheta(d)}{\vartheta(0)} \quad |G(j\omega)| \quad G(\omega) \frac{1}{\sqrt{1 \quad 3 \quad \frac{d}{d_\tau}^{2} \quad \frac{\omega}{\omega_\tau}^{2\;2} \quad 9 \quad \frac{d}{d_\tau}^{4} \quad \frac{\omega}{\omega_\tau}^{2}}} \tag{47}$$

$$\theta(d) \quad \theta(0) \quad \arg G(j\omega) \quad \mathrm{arctg}\frac{1}{\frac{1}{3} \quad \frac{d_\tau}{d}^{2} \quad \frac{\omega_\tau}{\omega} \quad \frac{\omega}{\omega_\tau}} \tag{48}$$

The usual method of analyzing PLN is application of Bode plots, asymptotic log-log plots of the amplitude-frequency characteristic. If the thickness of the PT sample satisfies the condition:

$$d \quad d_\tau \frac{2}{\sqrt{3}} \tag{49}$$

then the roots of the characteristic polynomial are real, and the amplitude-frequency characteristic of the transfer function can be approximately presented by the Bode plot in fig. 7.

Therefore, the log-log graph of amplitude of the experimentally measured differential PT spectra has two distinct features, frequencies ω_1 and ω_2, where the graph changes the slope. The easiest way to determine those frequencies from the recorded spectra could be to draw the low-frequency asymptote of the graph (which is parallel to abscissa), the high-frequency asymptote of the graphic (having a slope of –40 dB) and the medium range asymptote (with a slope of –20 dB); then, the intersection of the low-frequency asymptote and the medium range asymptote has the abscissa ω_1, and the intersection of the high-frequency asymptote and the medium range asymptote has abscissa ω_2.

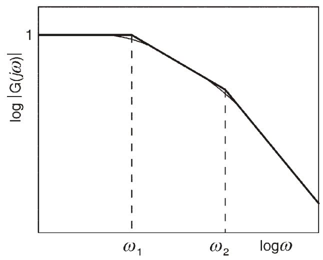

Figure 7. Bode plot (thick line) and log-log graph (thin line) of amplitude of differential PT response for PT induced sample that satisfies eq. (49) where ω_1 and ω_2 are roots of the characteristic polynomial of *G*(*s*)

Frequencies ω_1 and ω_2 can easily be calculated to be:

$$\omega_1 \quad \frac{\omega_\tau}{2}\; 1 \quad \sqrt{1 \quad \frac{4}{3} \quad \frac{d_\tau}{d}^{\;2}}\;, \quad \omega_2 \quad \frac{\omega_\tau}{2}\; 1 \quad \sqrt{1 \quad \frac{4}{3} \quad \frac{d_\tau}{d}^{\;2}} \tag{50}$$

showing that the frequency ω_1 decreases from $\omega_\tau/2$ to zero, while the frequency ω_1 increases from $\omega_\tau/2$ to ω_τ with the increase of sample thickness. Taking into consideration that the PLN model of the PT sample can be applied only to frequencies satisfying the condition described by eq. (44), the conclusion about the values of ω_1 and ω_2 means that the Bode plot analysis may be applied only in the determination of ω_1, since the PLN model is not valid for the frequency range to which ω_2 belongs.

Nevertheless, by determining ω_1 from the experimental data for sufficiently thick PT samples, it is possible to establish one analytical relation between the material parameters ω_τ and d_τ (and consequently τ and D). It means that the fitting procedure for the determination of the material parameters may be significantly simplified by application of PLN model, thus improving reliability of the data obtained by fitting.

If the thickness of the PT sample does not comply with the condition (49), the characteristic polynomial has double real, or complex conjugate, roots. The Bode plot in that case has one distinctive feature, frequency ω_0 where the Bode plot rapidly changes its slope from 0 dB to –40 dB.

This frequency could easily be determined from log-log graphics of the experimental differential PT spectra as the intersection of their low-frequency and high-frequency asymptotes. However, by analyzing eq. (46), it can easily be shown that the following equation holds:

$$\omega_0 \quad \frac{1}{\sqrt{3}} \frac{d_\tau}{d} \omega_\tau \tag{51}$$

where from it can be concluded that in the case when the condition expressed by eq. (49) is not satisfied, the high-frequency asymptote belongs to a frequency range where modeling by PLN is not applicable, and consequently, Bode plot analysis is not useful for thin samples.

However, it is well known in the theory of electric circuits that a transfer functions of the form (46), which can be rewritten in the form:

$$G(s) \quad \frac{\omega_D \omega_\tau}{3s^2 \quad 3\omega_\tau s \quad \omega_D \omega_\tau} \quad \frac{1}{T^2 s^2 \quad 2\xi T s \quad 1}, \quad T \quad \sqrt{\frac{3}{\omega_D \omega_\tau}}, \quad \xi \quad \frac{1}{2}\sqrt{3\frac{\omega_\tau}{\omega_D}} \quad \frac{\sqrt{3}}{2}\frac{d}{d_\tau} \tag{52}$$

has the resonant maximum [15]:

$$G_{\max.} = \frac{1}{2\xi\sqrt{1-\xi^2}} \text{ at frequency } \omega_{\max.} = \omega_0\sqrt{1-2\xi^2} \approx \omega_0 \tag{53}$$

when the condition $\xi < 0.5$ is statisfied, as it is shown in fig. 8

Considering eq. (52), it means that PT spectra of thin samples, satisfying condition $d/d_\tau < 1/3^{1/2}$, exhibit a peak at a frequency $\omega_{\max.}$, which can still be approximately considered to belong to the frequency range where PLN model is applicable, because the validity of the model for thin samples is determined by condition (44).

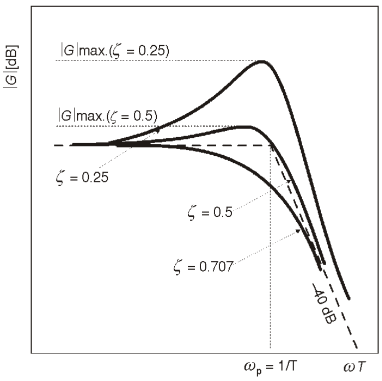

Figure 8. Spectra of differential PT response (eq. 53) for three different values of ξ

Therefore, while the Bode plot analysis cannot be applied for thin samples, the PLN model provides the possibility of determining ω_0 and ξ of thin samples from the experimental data without application of the fitting procedure. Equations (44), (52), and (53) enable further determination of the values of the sample parameters D_s and τ_s, thus providing the possibility of performing characterization of a sample without fitting.

Besides, it should be noted that the expression given by eq. (46), derived by application of the PLN modeling, is suitable for non-linear curve fitting. Thus, even in the cases when conventional methods of analysis of frequency analysis of electric circuits are not applicable, the modeling of the PT response by PLN simplifies the characterization of PT induced samples.

The special case that will be discussed is the case of the free-standing multi-layer PT sample. In order to model the PT response of such structure by linear passive electric network presented in fig. 4, it is necessary that the condition (44) be satisfied for each of the layers. If that is the case, a PT induced sample with n layers may be modeled by linear passive electric network that, based on eq. (36), can be represented by a-parameters as follows:

$$\begin{bmatrix}\tilde{U}(0)\\ \tilde{I}(0)\end{bmatrix} = \underline{T_1}\,\underline{T_2}\cdots\underline{T_n}\begin{bmatrix}\tilde{U}(d)\\ \tilde{I}(d)\end{bmatrix} = \underline{T_a}\begin{bmatrix}\tilde{U}(d)\\ \tilde{I}(d)\end{bmatrix} \tag{54}$$

where $\underline{T_m}$ $(m = 1, 2,\ldots,n)$ stands for the matrix of a-parameters of m-th layer.

In the case of the free-standing multi-layer PT induced sample, similar to the case of the free-standing single layer sample, the differential PT response $\vartheta(d)/\vartheta(0)$ can be calculated by analysis of the analogous network in the case when $\tilde{I}(d) = 0$. From eq. (54) then follows that:

$$\tilde{U}(0) = (\underline{T})_{11}\,\tilde{U}(d) \tag{55}$$

Considering the form of matrix $\underline{T_m}$, it can be concluded that the elements of the matrix $\underline{T_a}$ are polynomial expressions of the general form:

$$(\underline{T})_{11} = \sum_{u=0}^{n}\sum_{v=0}^{u}\beta_{uv}\tilde{Z}^{u-v}\tilde{Y}^{v} = \sum_{k=0}^{2n}\alpha_k s^k \tag{56}$$

where α_k and β_{uv} are real constants. Hence, the differential PT response of the multi-layer sample can be expressed in the following form:

$$\frac{\tilde{\vartheta}(d)}{\tilde{\vartheta}(0)} = G(s) = \frac{1}{\sum_{k=0}^{2n}\alpha_k s^k} \tag{57}$$

Therefore, the recommended non-linear fitting procedure for determining thermal properties of multi-layer samples is to apply modeling by linear passive electric network in order to find the dependence of coefficients a_k on the thermal properties of the materials of layers, and then to perform non-linear fitting of square of the inverse of the differential PT response spectra to polynomial of order 4n, as:

$$\left[\frac{\vartheta(0)}{\vartheta(d)}\right]^2 = \frac{1}{|G(j\omega)|^2} = \left[\sum_{k=0}^{n}(-1)^{k+1}\alpha_{2k}\omega^{2k}\right]^2 + \left[\sum_{k=1}^{n}(-1)^{k}\alpha_{k}\omega^{2k-1}\right]^2 \qquad (58)$$

An example of modeling of a three-layer structure (metal-polymer-metal) by electric network of the second order is presented in paper [16]. It is also of interest to point out that the expressions (52) and (53) present theoretical basis for Pade's approximate procedures for the determination of thermal properties of bipolar transistors presented in [18].

Conclusions

This paper presents an analysis of the possibility and potentials of modeling of the PT response of a thin sample by linear passive electric network. It is established that such a possibility depends on the modulation frequency of PT excitation and the thickness of the sample. If this condition is satisfied, the environment of the sample may be modeled by impedances and the sample itself by linear passive network presented in fig. 5.

If the free-standing PT induced sample can be modeled by passive linear network, its differential PT response is determined and it is shown that thermal properties of the material of the sample can be determined by the analysis of the peak in PT spectra (for the samples thinner than $d_\tau/3$), the combination of the Bode plot analysis and non-linear fitting (for the samples thicker than d_τ) or by linear fitting of polynomial expression (for the thin samples of arbitrary thickness).

Moreover, by analysis of the case of the multi-layer thin film sample, it has been shown that the modeling of the PT response by electric network may be useful even when the obtained network is not linear or not simple, since the expressions for the PT response may be more easily derived and analyzed, offering easier insight into the phenomenon compared with other models.

It can be concluded that modeling of the PT response by linear passive network is an easy-to-use and useful tool for the analysis of PT signal in function of modulation frequencies, since the derived models can significantly simplify the analysis of the PT response. Besides, obtained results could be employed in extraction of thermal properties by other lock-in thermography methods.

Acknowledgments

This work was supported by Ministry of Science and Technology Development of the Republic of Serbia (project no. 141013).

References

[1] Mandelis, A., Diffusion-Wave Laser Radiometric Diagnostic Quality-Control Technologies for Materials, *NDT&E International, 34* (2001), 4, pp. 277-287
[2] Vargas, H., Miranda, L. C. M., Photoacoustic and Related Photothermal Techniques, *Phys. Rep., 16* (1988), 2, pp. 45-101

[3] Rosencwaig, A., Gersho, A., Theory of the Photoacoustic Effect with Solids, *J. Appl. Phys., 47* (1976), 1, pp. 64-69

[4] Todorović, D. M., *et al*., Thermoelastic and Electronic Strain Contributions to the Frequency Transmission Photoacoustic Effect in Semiconductors, *Phys. Rev., B 55* (1997), 23, pp. 15631-15642

[5] Galovic, S., Kostoski, D.,Photothermal Wave Propagation in Media with Thermal Memory, *J. Appl. Phys., 93* (2003), 3, pp. 3063-3070

[6] Chen, G., Hui, P., Pulsed Photothermal Modeling of Composite Samples Based on Transmission-Line Theory of Heat Conduction, *Thin Solid Films, 339* (1999), pp. 58-67

[7] Hansen, P. C., Numerical Tools for Analysis and Solution of Fredholm Integral Equations of the First Kind, *Inverse Problems, 8* (1992), pp. 849-872

[8] Nagasaka, Y., Thermophysical Properties Measurement of Liquids and Solids by Photoacoustic and Photothermal Techniques, *Proceedings* (Eds. F. Scudieri, M. Bertolotti), 10th International Conference of Photoacoustic and Photothermal Phenomena, Rome, Italy, 1998, AIP Press, Woodbury, New York, USA, 1998, Vol. 463, pp. 289-294

[9] Tzou, D. Y., An Engineering Assessment to the Relaxation Time in Thermal Wave Propagation, *Int. J. Heat Mass Transfer, 36* (1993), 7, pp. 1845-1851

[10] Novikov, I. A., Harmonic Thermal Waves in Materials with Thermal Memory, *J. Appl. Phys., 81* (1997), 3, pp. 1067-1072

[11] Ulbrich, C. W., Exact Electric Analogy to the Vernotte Hypothesis, *Phys. Rev., 123* (1961) 6, pp. 2001-2002

[12] Suszynski, Z., Thermal Model Based on the Electrical Analogy of the Thermal Processes, *Proceedings* (Eds. F. Scudieri, M. Bertolotti), 10th International Conference of Photoacoustic and Photothermal Phenomena, Rome, Italy, 1998, AIP Press, Woodbury, New York, USA, 1998, Vol 463, pp. 197-199

[13] Hui, P., Tan, H. S., A Transmission-Line Theory for Heat Conduction in Multilayer Thin Films, *IEEE Trans. Comp. Pack. Manufact. Tech.-Part B, 17* (1994), 3, pp. 426-434

[14] Hui, P., Tan, H. S., Modelling of Thermal Diffusivity Measurement of Diamond Thin Films Using a Pulsed Laser Technique, *Surf. Coat. Technology, 62* (1993), pp. 361-366

[15] Kaminski, W., Hyperbolic Heat Conduction Equation for Materials with a Nonhomogeneous Inner Structure, *ASME J. Heat Transfer, 112* (1990), pp. 555-560

[16] Galović, S., Šoškić, Z., Photothermal Characterization of Polymer-Based Three-Layered Nanoelectronic Structures, *Proceedings*, ISSE, Budapest, 2008, pp. 172-177

[17] Stojić, M., Continuous System of Automatic Control (in Serbian), Naučna knjiga, Belgrade, 1990

[18] Nenadovic, N., *et al*., Extraction of Thermal Network Function for Bipolar RF Devices and ICs, *Proceedings*, XLVII ETRAN Conference, Herceg-Novi, Serbia and Montenegro, 2003, Vol. 4, pp. 225-228

Author´s affiliations:

S. P. Galović (**corresponding author**)
Laboratory for Radiation Chemistry and Physics,
Vinča Institute of Nuclear Sciences
P. O. Box 522, 11001 Belgrade, Serbia
E-mail: bobagal@vinca.rs

Z. N. Šoškić
Faculty of Mechanical Engineering Kraljevo,
University of Kragujevac, Serbia

M. N. Popović
Laboratory for Thermal Engineering and Energy,
Vinča Institute of Nuclear Sciences
Belgrade, Serbia

Paper submitted: July 14, 2009
Paper revised: November 6, 2009
Paper accepted: November 17, 2009