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Thermal diffusivity measurements in solids by means of the phase analysis of the photoacoustic signal

A. Calderón, R. A. Muñoz Hernández and S. A. Tomas
CINVESTAV-IPN, Depto. de Física. Apdo. Postal 14-740, 07000 México City, México.
E-mail: tonio@fis.cinvestav.mx

A. Cruz-Orea and F. Sánchez Sinencio
PMCATA, CINVESTAV-IPN. Apdo. Postal 14-740, 07000 México City, México

We presented a new method for determining the thermal difusivity in opaque solids by means of the phase analysis of the photoacoustic signal at low modulation frequencies. This method is tested by means of the determination of the thermal diffusivity in some solids and we show its importance in materials with high diffusivity values as metals, some semiconductors and materials whose thickness is small.

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Introduction

Photothermal phenomena in solids materials are generated by a combination of thermal expansion, thermal diffusion, and thermoelastic bending effects [1-5]. One of these mechanisms may predominate depending on the particular material and/or the experimental conditions. A possible way of distinguishing among these effects is the use of photoacoustic (PA) techniques, in particular by studying the PA signal dependence on the modulated chopping frequency of the incident light beam *f*.

From the presentation of the open photoacoustic cell (OPC) applied to the thermal characterisation in solids [6,7], this has been widely used in the measurement of the thermal properties in a variety of materials, and in composite systems, among other things [8-11]. This is a minimal volume PA detector which consist of mounting the radiation absorbing material directly onto a commercial electret microphone [12], using the front chamber of the microphone itself as the usual gas chamber of conventional photoacoustics. Its advantages over the conventional photoacoustics detectors are the use of a minimal gas chamber with no further transducer medium needed, no cell machining required, and low cost.

Up until now, the analysis of the PA signal detected with the OPC to determinate the thermal diffusivity is realised with the amplitude or phase dependence on the modulated chopping frequency of the incident light beam *f* in the thermally thick regimen [6-11]. However, for materials with great capacity of heat diffusion as metals, some semiconductors and others, as well as for many samples whose thickness is small, the thermally thick regime can occur in hundred or several thousands of Hz. This present the next inconvenients: *i*) due to the fact that the PA signal decreases exponentially with the modulation frequency, the noise-signal reason increase quickly, which implies lost reliability in the analysis. It can be used a light source of greater power to increase the PA signal, and consequently decreasing the noise-signal reason, with the drawback that the light can be sufficiently intensive as for modify the characteristics of the sample in study. *ii*) With the increase of the modulation frequency, the thermoelastic mechanism of generation of the PA signal is manifested, with an intensity depending on the particular material and/or the experimental conditions [3, 6]. This compels to use highest modulation frequencies to accomplish the analysis of the PA signal, for those which the thermoelastic mechanism will be dominant, due to the fact that in the mixture region of the diffusion and thermoelastic mechanisms the complexity in the mathematical expressions difficult the analysis [3, 6].

In this work we present a new method to determine the thermal diffusivity in opaque solids by means of the phase analysis of the PA signal working at low modulation frequencies. It is tested by means of the determination of the thermal diffusivity in some metals and semiconductors.

Theory

Applying the one-dimensional thermal diffusion model of Rosencwaig and Gersho [14] one gets for optically opaque samples that the amplitude and phase difference of the OPC signal are given, respectively, by

$$A = C_0 \frac{1}{f\sqrt{\cosh(2a_s l_s) - \cos(2a_s l_s)}} \tag{1}$$

$$\Delta\varphi = -atan\left(\frac{tan(a_s l_s)}{tanh(a_s l_s)}\right) - \pi/2 \tag{2}$$

where,

$$C_0 = \sqrt{2\alpha_s \alpha_g}\, V_0 I_0 / (T_0 l_g k_s \pi)$$

In this expressions a_i, l_i, k_i and α_i are the thermal diffusion coefficient, thickness, thermal conductivity and thermal diffusivity of material *i*, respectively. Here the subscript *i* denotes the sample (*s*) and gas (*g*) media, respectively. T_0 is the room temperature, I_0 is the incident beam intensity and V_0 is a quantity dependent only on the microphone characteristics.

The thermal diffusivity can be obtained from the high-modulation-frequency behaviour of either the signal amplitude or its phase. For the case of the signal amplitude, and if the sample is thermally thick, namely $x = a_s l_s = \sqrt{f/f_c} \gg 1$, the equation (1) reduces to,

$$A = \sqrt{2}C_0 \frac{1}{f} e^{-\sqrt{f/f_c}} \tag{3}$$

That is, the amplitude of the PA signal decreases exponentially with the modulation frequency in the form $(1/f)\exp(-\sqrt{f/f_c})$, were

$$f_c = \frac{\alpha}{\pi l_s^2} \tag{4}$$

This parameter is denominated "cut frequency" and represent the modulation frequency value for which the thermal diffusion length is equal to the sample thickness. Then α_s is obtained from the experimental data fitting coefficient $c = f_c^{-1/2}$ in the argument of the exponential.

For the case of the signal phase at the same regime, equation (2) reduces to,

$$\Delta\varphi = -\sqrt{\frac{f}{f_c}} + \pi/2 \tag{5}$$

Thus, the PA signal phase decreases linearly with $f^{1/2}$. Then α_s is obtained from the experimental data fitting the phase slope $c = f_c^{-1/2}$.

For many materials any of the two previously mentioned procedures are appropriate to determine α_s whereas the fit frequency interval do not correspond to frequencies so high that the reason noise-signal is considerable or the thermoelastic binding is present. However, for materials with great capacity of heat diffusion as metals, some semiconductors and others, as well as for many samples whose thickness is small, the thermally thick regime can occur in hundred or several thousands of Hz.

For saving this difficult is desirable to analyse the PA signal for low modulation frequencies. In the thermally thin regime, namely if $x = l_s a_s \ll 1$, the PA signal amplitude given in equation (1) is reduced to

$$A = \frac{C_0}{l_s}\sqrt{\frac{\alpha}{\pi}}\frac{1}{f^{3/2}} \tag{6}$$

In this case, the amplitude of the PA signal decreases as $f^{-3/2}$ with the increasing of the modulation frequency and is not possible to obtain the α_s by fitting.

In the other part, if $f / f_c \le (\pi/2)^2$ the phase equation (2) is closely equal to the expression,

$$\Delta\varphi = -\frac{1}{\pi f_c} f - 3\pi/4 \tag{7}$$

and the relative error in this approximation is smaller that 1.2 % [15]. Moreover, the time of heat diffusion through the sample τ_s is related with α_s and l_s by means of: $a_s = l_s^2 / \tau_s$, hence of the equation (4) is obtained that

$$\tau_s = 1/\pi f_c \tag{8}$$

This is precisely the slope in equation (7), which can be writing as

$$\Delta\varphi = -\tau_s f - 3\pi/4 \tag{9}$$

In this form, the PA signal phase lineally decrease with the modulation frequency when this vary from 0 to $(\pi/2)^2 f_c$. Hence, by means of the fitting of the experimental dates of the PA signal phase to the lineal equation (9) can be obtained the time of heat diffusion through the sample τ_s, and then, of the expressions (8) and (4) the cut frequency f_c and the thermal diffusivity α_s of the same, respectively.

Experimental Results

The experimental set-up, shown in Fig. 1, consisted of a 40 *mW* Argon laser whose beam is modulated using a variable speed mechanical chopper (SRS-model 540) and focused onto the sample. The sample is placed on top of the condenser microphone. The microphone output voltage is measured using a lock-in (SRS-model 850). All the data acquisition was micro-computer controlled. The samples studied are given in table I, together with their thickness l_s.

Table I. Samples types and their thickness l_s.

	Sample	l_s (μm)
1.	Ag (high purity)	440 ± 1
2.	Ag (high purity)	335 ± 1
3.	Al (high purity)	181 ± 1
4.	Si crystalline	291 ± 1
5.	GaAs crystalline	447 ± 1

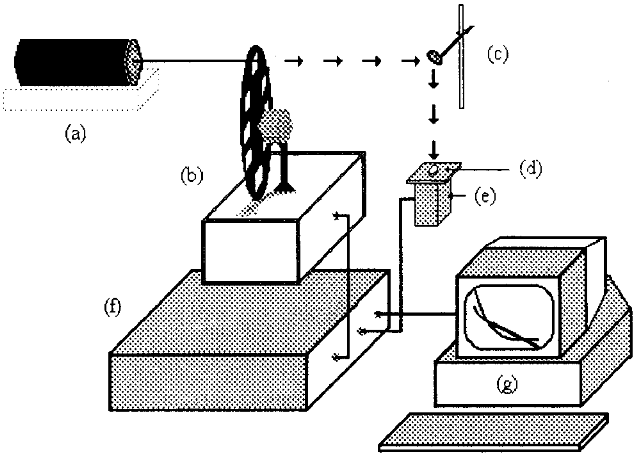

Fig. 1 Schematic experimental arrangement. (a) laser, (b) mechanical chopper, (c) mirror, (d) sample, (e) OPC, (f) lock-in amplifier and (g) computer.

In Fig. 2 we show the dependence of the PA signal phase on the modulation frequency for the silver samples 1 and 2. The sample 2 was obtained from the sample 1 through polished. Since these defer only in their thickness is of be waited that its times of heat diffusion τ_s will be differents and consequently its cut frequency too. However, we should obtain a same value for the thermal diffusivity in both samples by means of the analysis of the PA signal phase.

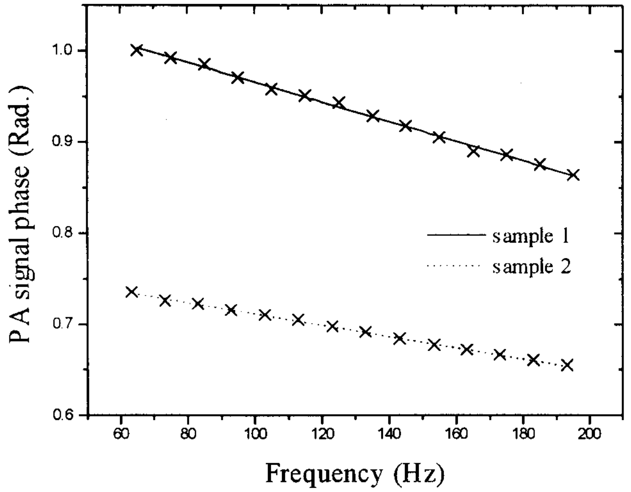

Fig. 2 PA signal phase vs frequency for the samples 1 and 2, and fit function.

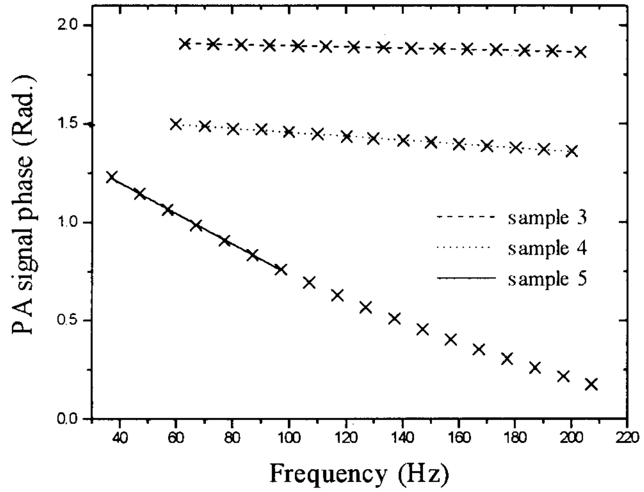

Fig. 3 PA signal phase vs frequency for the samples 3, 4 and 5, and fit function.

The Fig. 3 show the dependence of the PA signal phase on the modulation frequency for the samples 3, 4 and 5. These are of differents materials, hence we should obtain a different value for the thermal diffusivity by means of the analysis of the PA signal phase.

In the curves of the PA signal phase given in the Figs. 2 and 3 has been subtracted of microphone response. In all the cases, the phase present a lineal dependence with the modulation frequency, which is of to sane to the equation (9). The time of heat diffusion τ_s was obtained from the fitting of the PA phase data to low modulations frequencies, to the equation (9). Then, using the expressions (8) and (4) we obtained the cut frequency f_c and the thermal diffusivity α_s, respectively. The results are given in table II, together with values quoted in the literature [16]

Table II. Comparison of the thermal diffusivity values obtained with values quoted in the literature [16]

Sample	τ_s (*ms*)	f_c (Hz)	α_s (*cm*^{2}/*s*) Measured	α_s (*cm*^{2}/*s*) Literature
1.	1.068	294 ± 5	1.79 ± 0.04	1.74 ±0.07
2.	0.605	513 ± 25	1.81 ± 0.09	1.74 ±0.07
3.	0.298	1061 ± 106	1.09 ± 0.12	0.97 ±0.07
4.	1.001	318 ± 13	0.85 ± 0.04	0.88 ±0.06
5.	7.764	41 ± 2	0.26 ± 0.01	0.24 *

* *Reported value without experimental error .*

We observed that only the curve corresponding to the sample 5 not presents a linear behaviour in all the interval 40-200 Hz. It is linear until approximately 100 Hz and after is diverted increasingly from this behaviour while increases the frequency. This is thoroughly of agreement with the equation (9), which indicate that the phase is lineal with f in the interval (0, $(\pi/2)^2 f_c$), and since for the sample 5 we have f_c = 41 Hz, then $(\pi/2)^2$ (41 Hz) = 101.1 Hz.

Conclusions

The good agreement between the observed values of the thermal diffusivity and the literature values shows that the new method presented is accurate for obtaining the thermal diffusivity in solids opaque. Its advantage over the other methods is that this is realised to low modulations frequencies, hence, the noise-signal reason is not important and the PA signal it should be alone to the mechanism of thermal diffusion.

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