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Method for measurement of the thermal diffusivity in solids: Application to metals, semiconductors, and thin materials

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We present a method for determining the thermal diffusivity in opaque solids by means of an analysis of the photoacoustic phase signal at low modulation frequencies using the open-cell photoacoustic technique. We show for $f \leqslant (\pi/2)^2 f_c$, where f_c is the modulation frequency at which the thermal diffusion length matches the sample thickness, the photoacoustic phase signal can be written in linear form with the modulation frequency f. Then, obtaining the proportionality coefficient by fitting the experimental data, the thermal diffusivity of the sample can be determined. The advantage of this method is that it is realized in a range of modulation frequencies below those normally used, hence, the photoacoustic signal should be alone attributed to the mechanism of thermal diffusion. Moreover, the signal-to-noise ratio will be more reliable. This method was tested in some samples and it is also shown to be important in solids with high diffusivity values and thin materials. © *1998 American Institute of Physics.* [S0021-8979(98)04922-6]

I. INTRODUCTION

Photothermal phenomena in solid materials result from a combination of thermal expansion, thermal diffusion, and thermoelastic bending effects.[1–4] One of these mechanisms may predominate depending on the particular material and/or the experimental conditions. A way to distinguish these effects is the use of photoacoustic (PA) techniques, in particular by studying the PA signal dependence on the modulated chopping frequency f of the incident light beam. The open photoacoustic cell (OPC), developed for thermal characterization of solids,[5] has been widely used in the measurement of the thermal properties of a large variety of materials, ranging from metals and semiconductors, to polymers and foodstuffs.[6–9] In the OPC method the sample under study is mounted directly onto a commercial electret microphone[10] using the front chamber of the microphone as the gas chamber of conventional photoacoustics. Its advantage over conventional photoacoustic cells is the use of a minimal gas chamber with no further transducer medium needed, no cell machining required, and low cost.

The thermal diffusivity α_s, of a sample with thickness l_s, can be determined by the OPC method by analyzing the signal amplitude or the signal phase dependence on the modulation frequency f of the incident light beam in the thermally thick regime,[5–9] in which $l_s \gg \mu_s$, where μ_s $(=\sqrt{\alpha_s/\pi f})$ is the thermal diffusion length in the sample for the frequency f. However, for materials with a high thermal diffusivity, such as metals and some semiconductors, as well as for many samples whose thickness is very small, the thermally thick regime can only be reached at modulation frequencies of hundreds or several thousands of Hz. This has two main disadvantages: (i) since the PA signal decreases exponentially with the modulation frequency, the signal-to-noise ratio decreases quickly, resulting in lack of reliability in the analysis. Although this problem can be overcome by using a high intensity light beam, this might not be appropriate because the light could be intense enough to modify the characteristics of the sample under study. (ii) By increasing the modulation frequency, the thermoelastic mechanism of generation of the PA signal is manifested. Since the mathematical expressions that take into account both the heat diffusion and thermoelastic bending effect are complex, it is necessary to work at higher modulation frequencies where the thermoelastic contribution is the dominant effect.[3,5]

In this work we present a new method to determine the thermal diffusivity of opaque solids by analyzing the PA signal phase at low modulation frequencies. The predominant PA signal generating mechanism is then thermal diffusion. This method is tested in some metals and semiconductors, including high purity Ag and Al, and crystalline Si and GaAs.

II. EXPERIMENTAL SETUP

In our OPC experimental setup,[6] a 40 mW Ar^+ ion laser (Omnichrome 543-200 MA) is used and its monochromatic light beam intensity is modulated at a frequency f by a variable speed mechanical chopper SR-540 (Stanford Research Systems) before it normally impinges on the surface of the sample. The sample is placed in contact with the front chamber of an electret microphone.[10] The PA voltage signal obtained is detected by a lock-in amplifier SR-850 (Stanford Research Systems) interfaced to a personal computer, which permits recording the amplitude and phase of the signal as a function of the modulation frequency.

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TABLE I. Comparison of the measured values for the thermal diffusivity with those quoted in the literature.

Sample	l (μm)	f_c (Hz)	α_s (cm^2/s)	α_s (cm^2/s)
			Measured	Literature[a]
1. Ag	440±1	294± 5	1.79±0.04	1.74±0.07
2. Ag	335±1	513± 25	1.81±0.09	1.74±0.07
3. Al	181±1	1061±106	1.09±0.12	0.97±0.07
4. Si	291±1	318± 13	0.85±0.04	0.88±0.06
5. GaAs	447±1	41± 2	0.26±0.01	0.24[b]

[a]Reference 12.
[b]Reported value without experimental error.

The studied samples are listed in Table I. Samples 1 and 2 consist of high purity silver sheets with different thickness. Sample 2 was obtained from sample 1 by polishing. Sample 3 is a high purity aluminum sheet and 3 and 4 are crystalline semiconductors Si and GaAs, respectively.

III. THEORETICAL MODEL

From the one-dimensional thermal diffusion model of Rosencwaig and Gersho[1] it is seen that the amplitude and phase difference of the OPC signal for optically opaque samples are given, respectively, by

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

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

where

$$C_0 = \frac{\sqrt{2\alpha_s \alpha_g} V_0 I_0}{T_0 l_g k_s \pi}.$$

In these expressions α_i, l_i, k_i, and a_i are the thermal diffusivity, thickness, thermal conductivity, and thermal diffusion coefficient ($a_i = \sqrt{\pi f/\alpha_i}$) of material i, respectively. Here the subscript i denotes the sample (s) and gas (g) regions. T_0 is the ambient temperature, I_0 is the incident beam intensity, and V_0 is a quantity dependent on the microphone characteristics.

By means of expansions in power series in terms of the adimensional variable $x = a_s l_s = \sqrt{f/f_c}$, we obtained from Eq. (2), in the interval $f/f_c \leqslant (\pi/2)^2$, that $\Delta\varphi$ can be reduced to next linear dependence with f

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

In this approximation there is an error smaller than 1.2%.[11] The characteristic frequency $f_c = \alpha_s/\pi l_s^2$ represents the modulation frequency at which the thermal diffusion length ($\mu_s = a_s^{-1}$) matches the sample thickness. From Eq. (3) we observe that the PA phase signal decreases linearly with the modulation frequency in this range with a slope equal to $-1/\pi f_c$. Hence, by fitting Eq. (3) to the experimental PA phase signal the characteristic frequency f_c can easily be obtained. Then, the thermal diffusivity α_s of the sample is obtained if we know its thickness l_s.

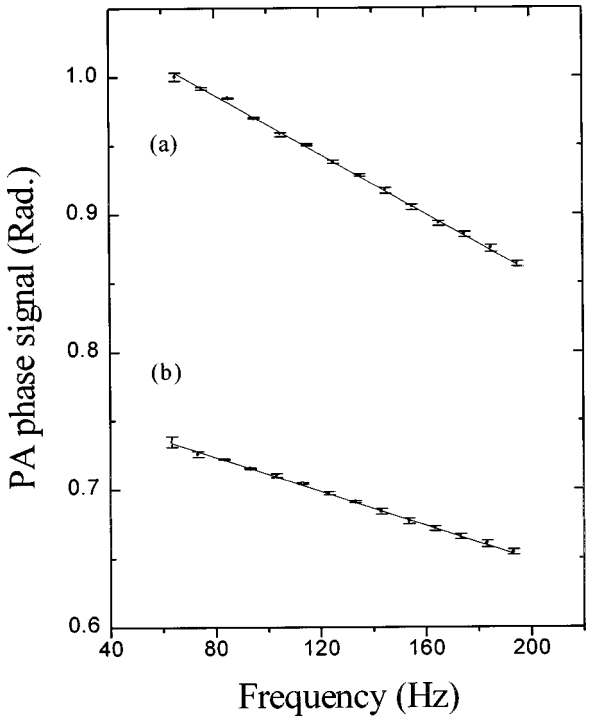

FIG. 1. Photoacoustic signal phase dependence on the modulation frequency for silver samples 1 and 2 [graphs (a) and (b), respectively). The lines indicate the best fit to Eq. (3).

IV. EXPERIMENTAL RESULTS AND DISCUSSION

In Fig. 1 is shown the PA phase signal dependence on the modulation frequency for the silver samples 1 and 2. Since sample 2 was obtained from sample 1 by polishing, these differ only in their thickness and it would be expected that their characteristic frequencies will be different. However, we should obtain the same value for the thermal diffusivity of both samples by the analysis of the PA phase signal. In Fig. 2 the PA phase signal is shown as a function of modulation frequency for the samples 3, 4, and 5, corresponding to Al, Si, and GaAs samples, respectively.

The curves of the PA signal phase given in Figs. 1 and 2 have been corrected by subtracting of the microphone response characteristic. In all cases, the phase presents a linear dependence with the modulation frequency, which is in accordance with Eq. (3). The curve for sample 5 is linear until approximately 100 Hz, after it diverges from this behavior with increased frequency. Linear behavior occurs in the frequency range where Eq. (3) is valid, since for this sample $f_c = 41$ Hz and then $(\pi/2)^2(f_c) = 101.1$ Hz. For all samples, the characteristic frequency f_c was obtained from fitting the PA phase data to Eq. (3), for the modulation frequencies indicated in Figs. 1 and 2. In these figures, the solid lines represent the fittings. Then, we obtained the thermal diffusivity $\alpha_s (= \pi l_s^2 f_c)$ as from the values of l_s and f_c. The results are given in Table I, along with values quoted in the literature.[12]

V. CONCLUSIONS

The agreement between the observed values of the thermal diffusivity and the literature values shows that the

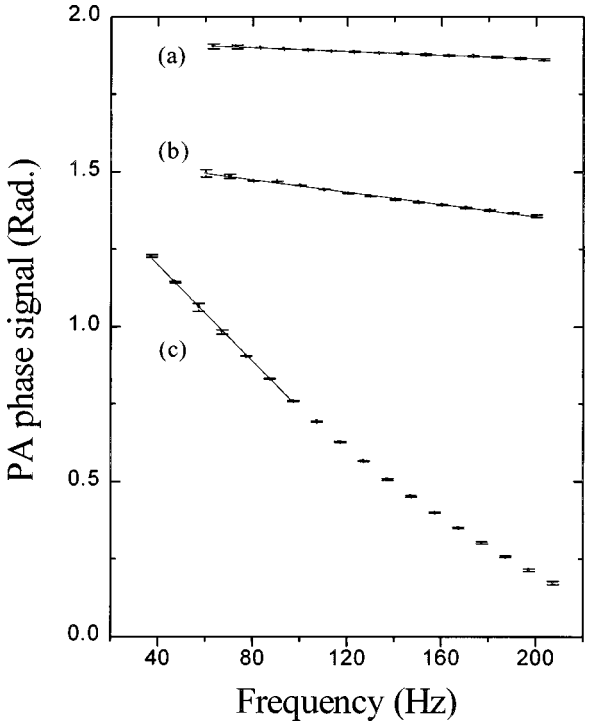

FIG. 2. Photoacoustic signal phase dependence on the modulation frequency for samples 3, 4, and 5 [graphs (a), (b), and (c), respectively). The lines indicate the best fit to Eq. (3).

method here presented for analyzing the PA phase signal is accurate for obtaining the thermal diffusivity in opaque solids. Its advantage over other methods is that it is realized in the range of modulation frequencies below those normally used for the thermally thick regime, hence, the signal-to-noise ratio will be more reliable and the PA signal should be only attributed to the mechanism of thermal diffusion.

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