

Photopyroelectric measurement of the thermal diffusivity of solids

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A novel and simple approach using a lateral heating source for the sample is proposed as an alternative method for the photopyroelectric measurement of the thermal diffusivity of solids. The method is experimentally tested with use of a Si sample.

It is by now well established that the modern photothermal spectroscopies, such as photoacoustic spectroscopy (PAS),^{1,2} photothermal radiometry,^{3,4} beam deflection spectroscopy,⁵⁻⁷ and photodisplacement spectroscopy,^{8,9} provide not only a sensitive means for measuring the optical absorption spectra but also exhibit the unique feature of allowing us to perform depth profile¹⁰⁻¹² as well as the measurement of the thermal properties¹³⁻¹⁸ of a sample. The basic principle of these thermal spectroscopies consists of probing the temperature fluctuation in the sample as a result of the non-radiative deexcitation processes that take place following the absorption of a modulated light beam. In particular, for determining the thermal diffusivity, one of the most widely used methods is the two-signal photoacoustic method of Yasa and Amer.¹⁸ This method consists of measuring the photoacoustic signal S_2 , obtained when the light beam is incident from the rear side, and the signal S_1 when the beam is incident on the front side. For a highly absorbing sample the ratio S_2/S_1 is given by¹⁸

$$S_2/S_1 = [\cosh^2(\pi f l^2/\alpha)^{1/2} - \sin^2(\pi f l^2/\alpha)^{1/2}]^{-1/2}, \quad (1)$$

where α is the thermal diffusivity, f is the modulation frequency, and l is the sample thickness. The fitting of the data, as a function of the modulation frequency, to Eq. (1) allows the determination of the thermal diffusivity. The disadvantage of this approach is that one has to measure the amplitude of two photoacoustic signals, on different sides of the sample, at the same surface conditions. This difficulty is usually not easily overcome, so that additional errors are introduced in the normalization process.

In this paper we describe an alternative single-signal approach for measuring the thermal diffusivity by use of the recently developed photopyroelectric spectroscopy (PPS).¹⁹⁻²¹ This new spectroscopy technique consists of using a thin pyroelectric (e.g., polyvinylidene difluoride, PVF₂) film in intimate contact with a solid sample on which a monochromatic light beam whose intensity is sinusoidally modulated at a frequency ω is incident. Following the absorption of the incident light, the nonradiative deexcitation processes within the solid causes the sample temperature to fluctuate and, through heat diffusion to the surrounding pyroelectric film, the sample-pyroelectric film-interface temperature fluctuates. As a result of this temperature fluctuation a pyroelectric voltage is produced in the film given by

$$V = \frac{p l_d}{\epsilon} \Delta T,$$

where p is the pyroelectric coefficient of the film, l_d is the film thickness, ϵ is the film dielectric constant, and ΔT is

the temperature rise in the pyroelectric thin film. This novel photopyroelectric approach for measuring the thermal diffusivity is schematically shown in Fig. 1. The light from a chopped laser beam is focused by means of a cylindrical lens along one face (normal to the y axis; cf. Fig. 1) of the sample, thereby generating at a lateral distance x_0 from the pyroelectric film-sample border ($x=0$) a localized periodic heat source. As a result of the localized heating at $x=x_0$ a thermal wave is set in the back face (at the $y=0$ plane) of our sample. This thermal wave diffuses along the x direction and eventually reaches the PVF₂ detector at $x=0$, attenuated exponentially as $\exp(-a_s x_0)$, where $a_s = (\pi f/\alpha)^{1/2}$ is the thermal diffusion coefficient. Thus, by measuring the pyroelectric signal, at a fixed modulation frequency, as a function of the heating beam offset x_0 , the thermal diffusivity α is readily obtained from the coefficient of x_0 in the exponential. To ensure that along the $x=x_0$ plane the sample is being uniformly heated one has two possibilities. One of them is to use a heating laser wavelength such that the sample is optically transparent. In this case, only a fraction βt_s (β is the bulk optical absorption coefficient at the incident laser wavelength and t_s is the sample thickness) of the incident light intensity is converted into heat, and the

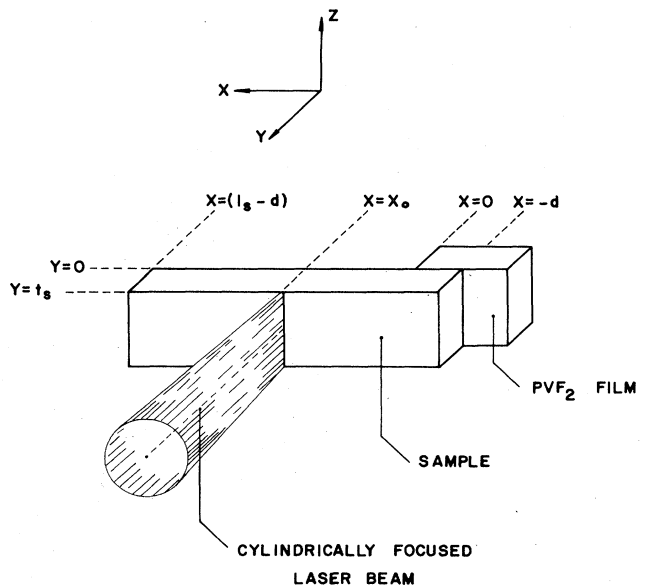


FIG. 1. Schematic configuration for the photopyroelectric lateral-heating method for measuring the thermal diffusivity.

heat deposition in the $x = x_0$ plane may be assumed uniform. If, however, the sample is optically opaque at the heating laser wavelength, which is the more likely situation for the cases of metallic and semiconducting samples and the common heating lasers available, the uniform heating may be achieved by working with thermally thin samples such that the thermal diffusion length $(\alpha/\pi f)^{1/2}$ is much longer than the sample thickness t_s . This important case of optically opaque samples is the one we address ourselves to in what follows. For a thermally thin sample it can be shown that the periodic temperature fluctuation ϕ_s in the $x = x_0$ plane is given by

$$\phi_s(x_0, t) = \frac{\beta' I_0}{t_s k_s \sigma_s^2} e^{j\omega t}, \quad (2)$$

where β' is the surface absorption coefficient of the sample, I_0 is the heating laser intensity, k_s is the sample thermal conductivity, and $\sigma_s = (1+j)a_s$, where $a_s = (\pi f/\alpha_s)^{1/2}$. With use of Eq. (2) as a boundary condition in the heat-flow equation along the x direction and neglecting the heat diffusion into the surrounding air one gets

$$\phi_s(x, t) = \frac{\beta' I_0}{t_s k_s \sigma_s^2} \left(\frac{e^{\sigma_s(d+x)} + e^{-\sigma_s(d+x)}}{e^{\sigma_s(d+x_0)} + e^{-\sigma_s(d+x_0)}} \right) e^{j\omega t}, \quad (3)$$

where d is the length of the thermal-contact region between the PVF₂ film and our sample, as shown in Fig. 1. The temperature ΔT rise in the PVF₂ film is obtained by taking the average temperature fluctuation in the detector-sample region ($-d < x < 0$), namely,

$$\Delta T = \frac{\beta' I_0}{t_s k_s \sigma_s^3 d} \left(\frac{e^{\sigma_s d} - e^{-\sigma_s d}}{e^{\sigma_s(d+x_0)} - e^{-\sigma_s(d+x_0)}} \right) e^{j\omega t},$$

which for $d\sigma_s \gg 1$ (large detector-sample thermal-contact region) reduces to

$$T \approx \frac{\beta' I_0}{t_s k_s \sigma_s^3 d} e^{-x_0 \sigma_s} e^{j\omega t}. \quad (4)$$

Equation (4) together with Eq. (2) tells us that the pyroelectric signal should decay exponentially as a function of the beam offset x_0 , with the coefficient of x_0 in the exponential being the thermal diffusion coefficient $a_s = (\pi f/\alpha_s)^{1/2}$.

To test the above method for determining a_s we have measured the photopyroelectric signal of a silicon wafer (6 mm long, 4 mm wide, and 185 μm thick) as a function of x_0 . The heating laser beam was provided by 4 mW He-Ne laser (Spectra Physics Model 185-01). The laser beam

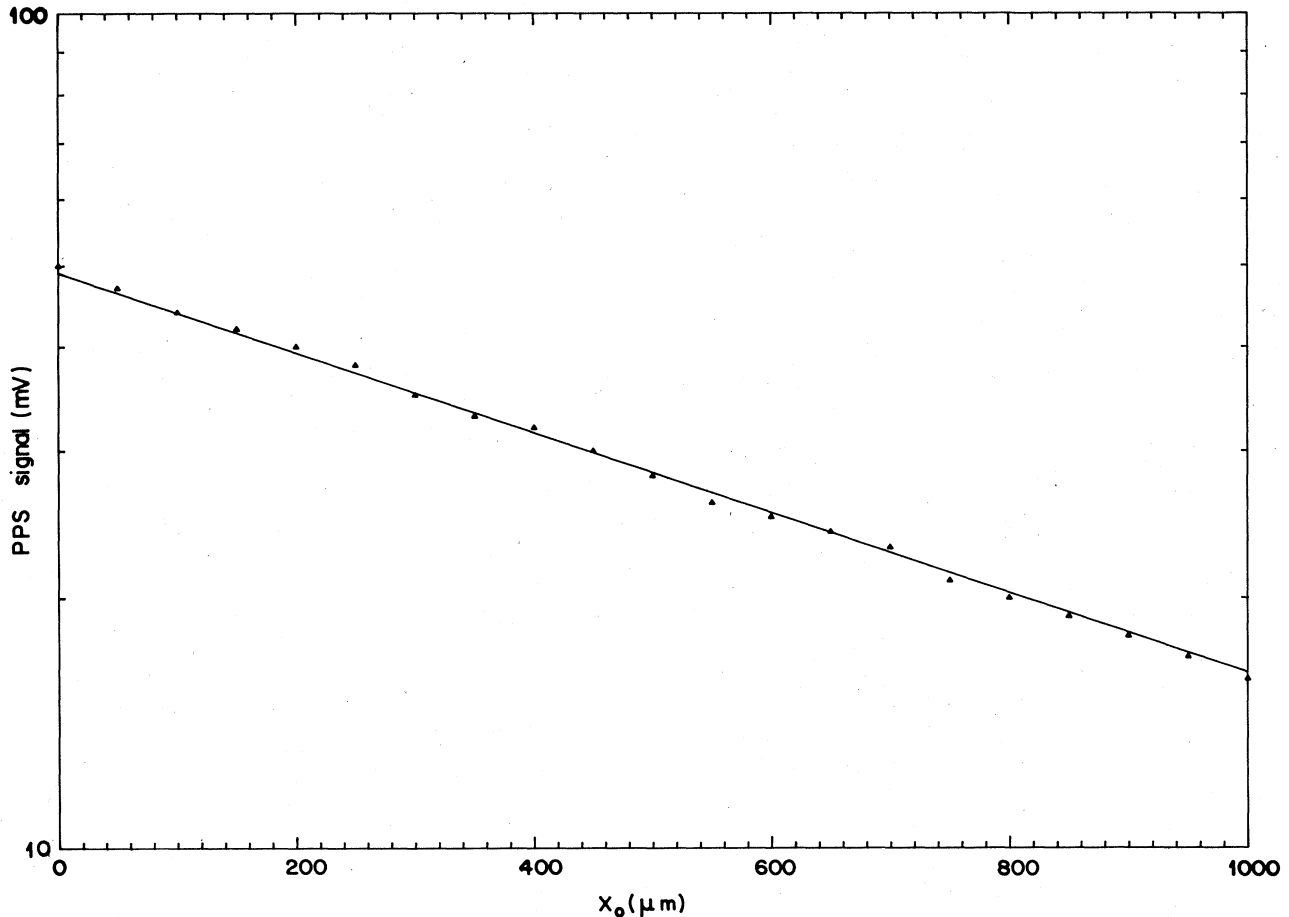


FIG. 2. Semilogarithmic plot of the photopyroelectric signal (PPS) of a silicon wafer, 185 μm thick, as a function of the illumination position x_0 , with use of a He-Ne laser modulated at 29 Hz. The triangles refer to the experimental data, whereas the solid curve is the least-square fitting of the data to an expression of the form $S = S_0 \exp(-a_s x_0)$.

modulated by a variable frequency chopper was focused on the 4-mm-wide face of the silicon wafer by means of a cylindrical lens which provided a focused strip 50 μm wide. The sample was in thermal contact with a 28 μm -thick Pennwalt PVF₂ film (5.5 mm long and 4 mm wide) supported by a printed circuit plate. The length d of the sample-PVF₂ thermal-contact region was 3.2 mm. To optimize the sample-PVF₂ thermal contact we have used a few microns thin layer of thermally conducting grease (Thermal Compound No. 128-8, Wakefield Engineering, Inc.). The output voltage from the PVF₂ detector was connected to a preamplifier and later fed into a Princeton Applied Research lock-in amplifier. The whole setup was mounted on a micrometer positioner so that it could be moved back and forth along the x direction. In this way we could vary the heating region position x_0 . In Fig. 2 we plot the amplitude of the pyroelectric signal as a function of x_0 at a modulation frequency of 29 Hz. Here we note that, because of the finite waist of the focused laser beam the illumination position is defined relative to the beam waist; i.e., we have subtracted the beam waist ($\approx 50 \mu\text{m}$) in the illumination position

scale. The least-square fitting of the experimental data to an expression [cf. Eqs. (2) and (4)]

$$S = S_0 e^{-a_s x_0},$$

yielded the coefficient $a_s = (\pi f / \alpha_s)^{1/2}$, from which the thermal diffusivity was deduced to be $\alpha_s = 0.83 \text{ cm}^2/\text{s}$. This value of α_s is in very good agreement with the literature value²² of $0.88 \text{ cm}^2/\text{s}$.

In conclusion, we have demonstrated in this paper the potential usefulness of the photopyroelectric spectroscopy for measuring the thermal diffusivity of solids. The fixed-frequency method using a lateral illumination configuration was tested for the important case of an optically opaque sample. In this context it is complementary to the previous photoacoustic method¹⁶ where only the case of an optically transparent sample was considered. The extreme simplicity, sensitivity, as well as its adaptability to practical restrictions imposed by experimental system requirements suggest that the present PPS method is a potentially more useful technique than the corresponding PAS method.

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