J. Phys. D: Appl. Phys. 21 (1988) S14–S16/ECOOSA '88. Printed in the UK

Photothermal deflection applied to thermal diffusivity measurements of ceramic (ferrite) materials

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Received 4 May 1988

Abstract. The photothermal deflection method for the measurement of the thermal diffusivity of solid materials is discussed. Measurements on ferrites of different compositions are presented.

1. Introduction

Photothermal deflection is a powerful and sensitive method of measuring the thermal diffusivity of solid samples [1–3]. This method, in its tranverse configuration, consists essentially in the detection of the deflection caused on a probe laser beam travelling in the air layer near the sample surface by a gradient in the air refractive index produced by heating the sample with a pumped laser beam (figure 1(*a*)).

The deflection angle ϕ is related to the temperature gradient perpendicular to the probe beam, $\nabla_\perp T$, by

$$\phi = \frac{1}{n}\frac{\mathrm{d}n}{\mathrm{d}T}\int_{\text{path}} \nabla_\perp T(r, t)\,\mathrm{d}s \qquad (1)$$

where n is the refractive index of the air. Equation (1) can be solved for the case of a sinusoidally modulated Gaussian pumped beam if the optical (absorption coefficient and reflectivity) and thermal (conductivity and diffusivity) parameters are known.

Usually integration of equation (1) is rather tedious and in most cases computer calculations are needed; these limit the use of the method if absolute values of the parameters involved are wanted.

If one is interested in the measurement of thermal diffusivity alone, however, it is possible to show that the measurement can be made in such a way as to avoid explicit solution of equation (1), and the thermal diffusivity obtained without any knowledge of other parameters. For this purpose the amplitude of ϕ is measured as a function of the offset between the probe beam and the centre of the pumped beam (figure 1(*b*)). When some simplifying assumptions are made, it will be shown below that the thermal diffusion length l_T of the material can be derived, and from this the thermal diffusivity χ is obtained, once the chopper frequency ω is known, by the relation

$$l_T = (2\chi/\omega)^{1/2}.$$

The method has been applied to the measurement of the thermal diffusivity of some ferromagnetic garnets.

2. Method of thermal diffusivity measurement

The principle of the method consists in measuring the probe deflection by successively displacing it with respect to the centre of the heating beam by a quantity y (figure 1(*b*)). If the thermal diffusivity of the sample is much lower than that of air ($\chi = 0.19\,\mathrm{cm^2\,s^{-1}}$), the presence of the air can be neglected and, by taking into account only the conduction over the sample, an expression for the deflection angle ϕ can be obtained of the form [3, 4]

$$\phi = \phi_0 \exp(-y/l_T) \exp[\mathrm{j}(\omega t - y/l_T)] \qquad (2)$$

where ϕ_0 is the maximum deflection at $t = 0$ at the pumped laser centre ($y = 0$).

Equation (2) corresponds to the solution of the unidimensional problem of a sample heated by a point source sinusoidally modulated with the probe beam grazing at the sample surface. Clearly, because experimentally the pump beam has a finite size w, equation (2) is only valid for $y > w$.

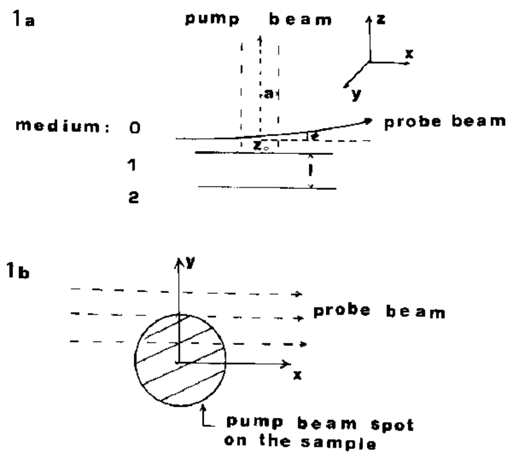

Figure 1. Geometry of the photothermal deflection method.

Simple inspection of equation (2) shows now that, for example, the in-phase signal nullifies as a function of y when $y_0 = (\pi/2)l_T$, which allows one to find the thermal diffusivity, by measuring y, from the simple formula

$$\chi = (2\omega/\pi^2)y_0^2.$$

A computer simulation of equation (1) has been performed in the range of parameters for which equation (2) is expected to be valid; the results are shown in figure 2. It is seen that the zero points of the exact and approximate formulae nearly coincide for $z_0 = 0$.

A closer examination of the problem has shown however that equation (2) is a good approximation when the vertical offset z_0 of the probe beam, which corresponds to the mean distance of this beam from the sample surface (see figure 1(a)), is nearly equal to zero or at least the condition $z_0 \ll l_G$ (l_G is the thermal diffusion length of air) is fulfilled. This is simply because equation (2) is based on integration of the thermal gradient at the sample surface ($z_0 = 0$). Again a computer simulation shows that for high values of z_0 the approximation loses its validity, and the zero-crossing point obtained by equation (2) gives a lower value of the thermal diffusivity (figure 2).

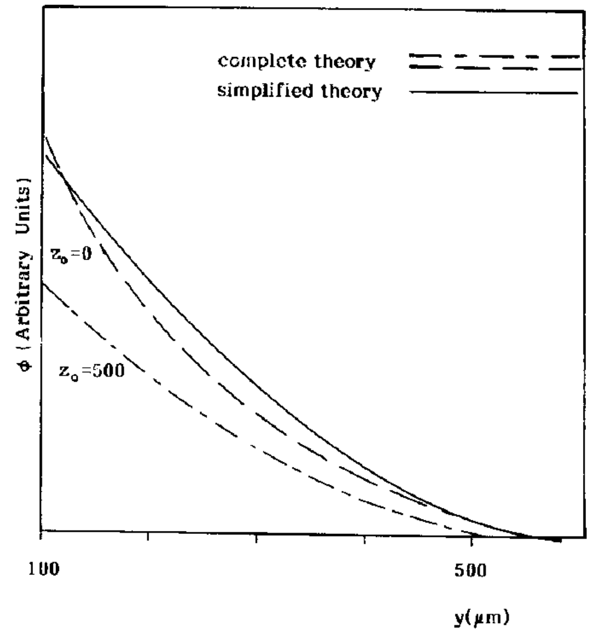

Figure 2. Photothermal deflection ϕ versus offset y for different z_0 values. The full curve is from equation (2).

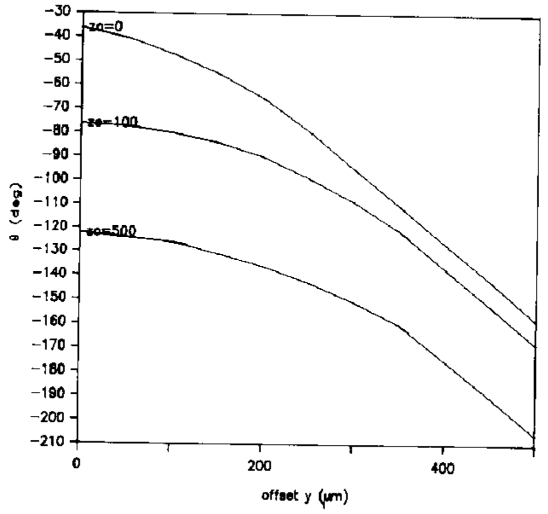

Figure 3. Computer calculation of θ versus y for different z_0 values. Numerical data were $R = 0.1$, $\alpha = 1000\,\mathrm{cm}^{-1}$, $\chi = 0.07\,\mathrm{cm}^2\,\mathrm{s}^{-1}$, $\omega = 20$ Hz.

A better approximation is found by using a different expression for ϕ given by

$$\phi = R(y)\exp[\mathrm{j}(\omega t + \theta(y))] \qquad (3)$$

where the probe phase $\theta(y)$ now has the more general form

$$\theta(y) = -y/l_T + f(z_0)$$

f being an arbitrary function depending on a large number of parameters, and in particular on the vertical offset z_0.

Figure 3 shows $\theta(y)$ versus y for different values of z_0. It can be seen that for large values of y, $\theta(y)$ displays a linear behaviour. The function f has two fundamental effects: it shifts θ vertically for increasing values of z_0, and causes the linear behaviour to start at different y values for different z_0 values. This behaviour makes it possible to derive χ from the slope α of the curve, irrespective of the value of the function f, simply as

$$\chi = \omega/2\alpha^2.$$

3. Experimental results

The samples examined were ceramic ferrites of the garnet type. A CO_2 laser was used as the pumping beam, while a He–Ne beam was used as the probe.

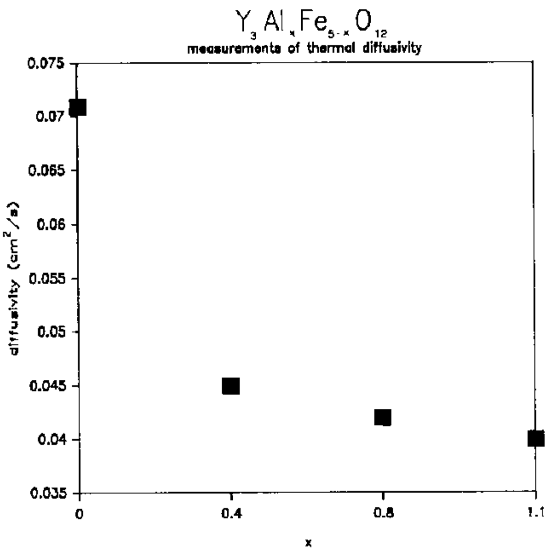

Figure 4. Measured diffusivity as a function of Al content in $Y_3Al_xFe_{5-x}O_{12}$.

Two different sets of experiments have been performed. In the first set substitution of Al in YIG has been considered. Figure 4 shows that χ decreases as x increases in the formula $Y_3Al_xFe_{5-x}O_{12}$. This result can be understood on the basis of the increased disorder due to the introduction of Al.

In the second set of experiments a complex compound of formula $Y_{0.5}Gd_{2.5}Fe_{4.3}In_{0.5}Al_{0.2}O_{12}$ has been studied as a function of the content of added Be out of stoichiometry. In this case (figure 5) there is first a decrease in χ on increasing the Be content, but on further increasing the mole fraction of Be, χ increases. This behaviour is probably connected with the different positions of the Be atoms in the garnet lattice as the molar concentration of Be increases.

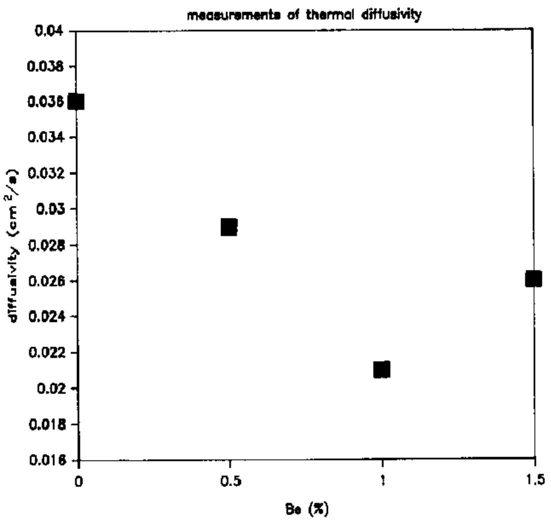

Figure 5. Measured diffusivity of $Y_{0.5}Gd_{2.5}Fe_{4.3}In_{0.5}Al_{0.2}O_{12}$ as a function of added Be.

4. Conclusions

The photothermal deflection method in its transverse version can be adapted to obtain simple measurements of the thermal diffusivity of solid opaque samples.

The results obtained for some ferrites may help in understanding the effect of substitutions in such materials.

References

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