On the Use of the Thermal Lens Effect as a Thermo-Optical Spectroscopy of Solids

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Abstract. A theoretical model for the thermal lens effect due to the absorption of modulated light by a solid is presented. The discussion includes an analysis of the modulation frequency response of the temperature rise in the solid. It is suggested that this effect may be used as a new technique for studying the thermo-optic properties of solids.

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In recent years a number of thermo-optical spectroscopic techniques have been suggested especially for measuring weak absorptions of solutions. Amongst these techniques we include thermal lensing [1-3], interferometry [4-6], photothermal beam deflection [7], photothermal radiometry [8, 9], and photoacoustic spectroscopy [10]. Using solutions with low background absorption, absorptivities ranging from 10^{-5} to 10^{-7} cm^{-1} have been measured with interferometry [4, 5] and thermal lensing [3, 11]. In the case of solids, such as semiconductors, one is often facing the opposite situation, namely, that of optically opaque samples. Of the above techniques, the best suitable for dealing with these opaque solids are in our opinion the photothermal radiometry and the photoacoustic spectroscopy, since, even in the optical opaqueness condition, their signal remain proportional to the optical absorption coefficient of the sample.

In this paper we present a detailed theoretical discussion of the thermal lens effect (TLE) in solid samples aiming the understanding of its dependence on the optical and thermal properties of the sample. In particular, we show that the TLE may be successfully used as an alternative technique for measuring the absorption coefficient of highly opaque samples with some advantages, for instance, over conventional photoacoustic spectroscopy. The thermal lens effect was first reported by Gordon et al. [1] and its basic principle consists essentially in the observation of the changes induced in the refractive index of a sample as it is heated by the absorption of light, as follows. Consider an intensity-modulated laser beam with a Gaussian profile propagating along the z-axis and incident upon a sample, as shown in Fig. 1. The energy absorbed from the beam by the sample is converted, in part or in whole, into heat by nonradiative de-excitation processes within the solid. The periodic temperature distribution thus created in the solid produces, in turn, a spatially dependent change in the refractive index n proportional to \(\frac{dn}{dT}\).

This spatially dependent refractive index turns the medium into a lens for the light beam. In the case of liquids and gases the refractive index changes because of a decrease in density with increasing temperature. In these cases \(\frac{dn}{dT}\) is negative, and the thermal lens is a diverging lens. Thus, as the laser penetrates the sample, the development of the thermal lens causes a spread of the beam and a drop in its intensity, so that for a transparent sample, by measuring the deflection as well as the magnitude of the emerging beam with a small photodetector placed at the other end of the sample, the thermo-optic properties of the sample can be studied. In the cases of solids, however, \(\frac{dn}{dT}\) is usually positive, and one is eventually interested in studying optically opaque samples. This means that alternative detection techniques must be used. In what follows, after presenting the theory of the thermal lens effect in Sect. 1, we discuss in Sect. 2 the use of two...
different detection techniques, namely, reflectance spectroscopy and interferometry. Finally, in Sect. 3 a summary of our finding is presented.

1. Theory

1.1. Outline of the Theory

Let us consider the geometry shown in Fig. 1, where the solid of length $l$ [cm] has a bulk optical absorption coefficient $\beta$ [cm$^{-1}$]. A sinusoidally chopped monochromatic light beam with wavelength $\lambda$ is incident on the solid with intensity $I(r,t) = I_0(r)(1 + \cos \omega t)/2$, where $\omega = 2\pi f$ is the modulation frequency of the laser beam, and $I_0(r)$ [W/cm$^2$] is the Gaussian intensity distribution of the laser, namely,

$$I_0(r) = \frac{P}{\pi r_0^2} e^{-r^2/r_0^2},$$

where $r_0$ is the laser beam waist and $P$ is the incident power. For the sake of simplicity, we assume the solid to be an semi-infinite isotropic medium. This approximation is justifiable since a focused laser beam is typically much smaller than the sample dimensions. Furthermore, we shall be mainly interested in the case of optically opaque samples, where the optical penetration depth $\beta^{-1}$ is much smaller than the sample length.

Under these conditions, one then adopts the following model. The absorption of modulated light causes periodic heating in the sample. The temperature fluctuation (relative to the ambient), $T(r,z,t)$, in the sample is then determined by solving the thermal diffusion equation

$$\nabla^2 T - \frac{1}{\alpha} \frac{\partial T}{\partial t} = -\frac{Q}{K},$$

where $x = K/\rho c$ is the thermal diffusivity [cm$^2$/s] of the sample, $K$ [W/cm°C] is the thermal conductivity, $\rho$ [g/cm$^3$] the density, $C$ [J/g°C] the specific heat, and $Q$ is the thermal energy source due to the absorption light. In terms of the incident intensity and the optical absorption coefficient, the heat density produced at a point $r$ in the sample is given by

$$Q(r,t) = \frac{1}{2} \beta I_0(r) e^{-\beta z}(1 + \cos \omega t).$$

Knowing the temperature distribution $T(r,z,t)$, the periodically varying refractive index induced in the sample is then obtained from

$$n(r,z,t) = n_0 + \frac{dn}{dT} T(r,z,t),$$

where $n_0$ is the refractive index at the ambient temperature, and $dn/dT$ is the rate of change of the refractive index with temperature, which we assume to be an a priori known parameter. Assuming that both $T(r,z,t)$ and $Q(r,t)$ have a time dependence $\exp(j\omega t)$ (with the real parts of the solutions representing the physical variables, as usual), (2) reduces to, after suppressing the time dependence,

$$\nabla^2 T - \frac{j\omega}{\alpha} T = -\frac{\beta P}{2\pi r_0^2 K} e^{-\beta z} e^{-j\omega t^2}.$$  

where

$$P^2 = \int \left( \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) + \frac{\partial^2}{\partial z^2} \right).$$

Using the laser beam waist $r_0$ as unit of length, let us introduce the following dimensionless variables: $R = r/r_0$, $Z = z/r_0$, $W = \beta r_0$, $\mu^2 = j\omega r_0^2/\alpha$, and $B = \beta P/2\pi K$. In terms of these dimensionless variables, (5) can be written as

$$\frac{1}{R} \frac{\partial}{\partial R} \left( R \frac{\partial T}{\partial R} \right) + \frac{\partial^2 T}{\partial Z^2} - \mu^2 T = -Be^{-\mu^2} e^{-R^2}.$$  

1.2. Temperature Distribution

To solve (7), we have to find a particular solution $T_p$ and a homogeneous solution $T_{hom}$. The particular solution may be found by letting $T_p(R,Z) = 0$ and $T_{hom}(R,Z)$.

$$T_p(R,Z) = \int_0^\infty d\lambda \lambda \mu I_0(\lambda R) e^{-\lambda Z}.$$  

Substituting (8) into (7) and writing $\exp(-R^2)$ as

$$\exp(-R^2) = \int_0^\infty d\lambda \lambda F(\lambda) I_0(\lambda R),$$

where $F(\lambda) = \frac{1}{2} e^{-\lambda^2/2}$,