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Measurement of low optical absorption in highly scattering media using the thermal lens effect

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Abstract. In this work we show that the thermal lens effect can be applied to highly scattering and weakly absorbing materials. We apply the thermal lens effect and the z-scan technique to estimate the effective absorption coefficient of a suspension of TiO_2 particles with a mean diameter of 220 nm at two wavelengths: 488 nm and 514 nm. From the effective absorption coefficient we estimate the absorption cross section of the particles.

1. INTRODUCTION

In this work we are interested in measuring the small absorption cross section of highly scattering particles by a thermal lens method [1-6]. As it is well know the laser beam propagating through an absorbing sample generate a heat flow that gives rises to a change in the refractive index and produces a self defocusing (or self focusing) effect in the output beam. This effect can be measured by using the z-scan technique [5]. This technique measures the self-defocusing effect by detecting the changes of the intensity of the transmitted optical field along the optical axis in the far field as a function of the sample position as illustrated in figure 1. A typical curve of the on axis intensity versus sample position is shown in the inset of figure 1. The absorption coefficient can be deduced from the difference between the maximum (peak) and the minimum (valley) intensities normalized to when the thermal lens signal is negligible, T_{pv}.

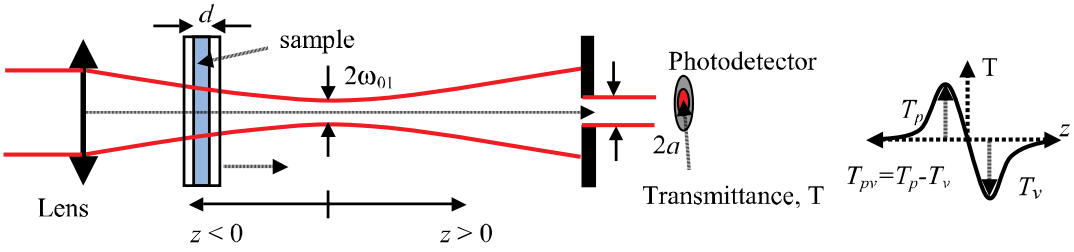


Figure 1. Schematic diagram of the z-scan technique and the transmittance signal vs. position at the photodetector.

2. THERMAL LENS IN GRANULAR MEDIA

When a laser beam is focused on a slab of a suspension of particles, part of it will be scattered away from the direction of the beam giving rise to a diffuse field which does not preserve the phase, and part of it will travel through the slab as it would do through an homogeneous medium with an effective refractive index [7]. This latter component of the total field is commonly denominated the coherent field. For a dilute particle suspension, the effective refractive index, n_{eff}, seen by the coherent field upon propagation through the particles suspension, consists of the refractive index of the matrix material plus a contribution due to the particles. This contribution is proportional to the volume fraction occupied by the particles and to their forward scattering amplitude [7]. The imaginary part of n_{eff} is referred to as the extinction coefficient, κ, and in general, it is due to scattering and to absorption. The attenuation of the intensity of the coherent component as a function of distance traveled through the turbid medium is given by Beer-Lambert's law,

$$I = I_0 \exp(-2k\kappa z) = I_0 \exp(-\alpha_{ext} z)\,, \tag{1}$$

where $\alpha_{ext} = \eta C_{ext}$, C_{ext} is the extinction cross section of the particles, and η is the particle number density. As is well known, when the matrix material is non-absorbing, C_{ext} can be split into the scattering and absorption cross sections: $C_{ext} = C_{sca} + C_{abs}$. Therefore we can write $\alpha_{ext} = \alpha_{sca} + \alpha_{abs}$ where $\alpha_{sca} = \eta C_{sca}$ and $\alpha_{abs} = \eta C_{abs}$. We may refer to these coefficients as the effective scattering and absorption coefficients. Now, the absorption of optical energy per unit volume within the slab of the particle suspension is $\dot{g} = \alpha_{abs} I$, where I is the light intensity. Clearly $\dot{g}$ depends differently on α_{abs} than on α_{sca}; and it should be possible to discern between them from photo-thermal measurements and an independent measurement of α_{ext}. Actually if $\alpha_{ext} d << 1$, where d is the thickness of the slab of particles, then I is constant through the slab and $\dot{g}$ is simply proportional α_{abs} and it does not depend on α_{sca}.

Therefore, in a suspension of scattering particles, if there is some optical absorption a thermal lens will form and the coherent field will respond to it. On the other hand, it is not difficult to see that the diffuse field will not respond significantly to the thermal lens effect. However, some of the diffuse light may reach the detector and contribute to the signal. This contribution will in general depend on the position of the sample and will give rise to a z-scan signal. When there is strong scattering and low absorption, this spurious signal will mask the thermal lens signal. Therefore, one must find a way to subtract from the signal the contribution of the diffuse field. We have accomplished this in the following way: at each position of the sample, the laser beam is applied at some time, t_1, and shut at a later time t_2. The photothermal signal is taken as the signal at time t_2 minus the signal at time t_1, when the thermal lens has not yet formed. Since only the coherent beam responds to the thermal lens effect, the contribution from the diffuse light is the same at t_2 and at t_1, and therefore it is removed. The period $t_2 - t_1$ must be large compared with the time of formation of the thermal lens which depends on the thermal diffusivity of the sample, D, and the beam radius.

From the parabolic lens model [4] we obtain the following expression for the effective absorption coefficient,

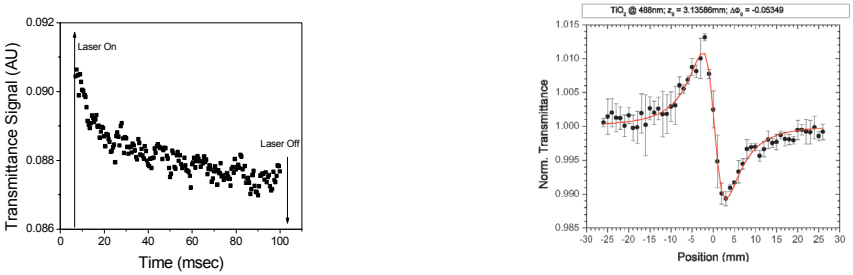
$$\alpha_{abs} = \frac{T_{pv}}{\dfrac{\partial n}{\partial T}\dfrac{P_0}{\pi\kappa} k_0 d}\,, \tag{2}$$

where $\partial n/\partial T$ is the thermo-optical coefficient, P_0 is the optical power incident to the sample, κ is the thermal conductivity of the sample, and k_0 is the wave-number in vacuum. The parabolic lens model assumes a 2D problem and therefore it neglects the flow of heat through the walls of the sample container; that is, along the z-axis. Then, it overestimates the strength of the thermal lens, and as a consequence, Eq. (2) underestimates the absorption coefficient Therefore, Eq. (2) actually gives a lower bound to the absorption coefficient. On the other hand, if one assumes that heat flows freely

into the container walls and outside, that is, one neglects the thermal interfaces; then, Eq. (2) is modified. In this case one would underestimate the strength of the thermal length and obtain an upper bound to the absorption coefficient. We have derived an expression for such an upper bound which we use below for interpreting our experimental results below. Details of this upper bound will be published elsewhere.

3. RESULTS

Our purpose was to estimate the absorption cross section of TiO_2 particles used in the paint industry. We dispersed TiO_2 particles of mean diameter of 220 nm and a volume fraction of 0.31% in an optical adhesive. A 100 μm thick slab-cavity was formed with two parallel 1mm thick glass slabs and filled with the particle suspension and then cured to form a static particle suspension. The Z-scan setup is illustrated in Fig. 1. It consists of an air cooled Ar^+ laser, a lens of 75 mm focal distance, an electro-mechanical shutter, a motorized linear stage to displace the sample, and a photodetector with a 0.5 mm radius aperture placed at about 2 m from the focal point. The shutter, data acquisition, and sample displacement was controlled by a personal computer. The displacement of the sample was done in steps of 1 mm. In Fig. 2a we show the transmittance as a function of time after opening the laser shutter and closing it 90 msec later. The sample position was 4 mm before the focal point, that is at negative z, and the laser power incident to the sample was 60 mW. Now, to obtain a z-scan curve at every position of the sample, the shutter was opened and closed 100 msec later and the transmittance was registered as a function of time during the time interval the shutter was open. The normalized transmittance, corrected from the diffuse contribution, was calculated from the first and final value of the transmittance. This operation was repeated 50 times at each position z of the sample and then averaged. In Fig. 2b we show the result of averaging three z-scan curves taken consecutively and using a wavelength of 488 nm. A fit with the parabolic lens model to the experimental data is also shown in the figure. The curve gives a value of the T_{pv} of 0.022. A similar curve was obtained for a wavelength of 514 nm and a T_{pv} of 0.032 was obtained. Curves at both wavelengths for a similar sample but without the embedded particles were obtained and no thermal lens signal was detected in neither case; indicating that the optical absorption of the cement itself was negligible. The values of the κ, D, $\partial n/\partial T$ were taken as those of the cement alone, that is 0.205 W m^{-1} $^oC^{-1}$, (0.1-0.2) x 10^{-6} m^2 s^{-1}, and 1.7 x 10^{-4} $^oC^{-1}$ respectively. α_{ext} was measured directly by transmission; and we obtained, α_{ext} = 0.01 μm^{-1}. Using these values in Eq. (1) and Eq. (2) give an effective absorption coefficient of the particles suspension of α_{abs} = (1.3 – 2.6) x 10^{-2} cm^{-1} for 488nm and of α_{abs} = (1.7 – 3.7) x 10^{-2} cm^{-1} for 514 nm. From the absorption coefficient and assuming all the particles are spherical with a diameter equal to the mean diameter of 220 nm we obtain an absorption cross section of C_{abs} = (2.3 – 4.7) x 10^{-6} μm^2 at 488 nm and of C_{abs} = (3.0 – 6.6) x 10^{-6} μm^2 at 514nm.



(a) (b)

Figure 2. (a) Transmittance as a function of time at $z \approx$ - 4 mm and P_0 = 60mW; (b) Thermal lens signal as a function of the sample position for λ = 488nm.

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