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ABSOLUTE MEASUREMENT OF OPTICAL ATTENUATION USING PHOTOTHERMAL LASER-BEAM-DEFLECTION SPECTROSCOPY

G.C. Wetsel, Jr. and S.A. Stotts

*Department of Physics, Southern Methodist University, Dallas,
Texas 75275, U.S.A.*

Résumé - On présente une méthode photothermique de mesure absolue du coefficient d'absorption. Son domaine d'application est défini par un modèle théorique.

Abstract - A method of absolute measurement of optical attenuation using photothermal spectroscopy has been developed. Its range of applicability is characterized by a theoretical model.

We have discovered that laser-beam-deflection spectroscopy (LBDS) can be used for the absolute measurement of wave or particle-beam attenuation.¹ In this paper we report: 1) the successful application of LBDS to the absolute measurement of optical attenuation, β , in $U^{3+}:\text{CaF}_2$, 2) comparison of the LBDS results to determination of β by enclosed-cell-microphone-detection photoacoustic spectroscopy (ECMD PAS) and by measurement of transmitted and reflected light, and 3) the results of a theoretical model developed to explain the experiment and characterize its range of applicability.

An exponential decrease of the intensity of a wave or particle beam with distance (z) along the direction of wave propagation, $I(z) = I_0 e^{-\beta z}$, where I_0 is the intensity at $z = 0$, is characteristic of any loss process for which the spatial rate of change of intensity is linearly proportional to the intensity. If the photothermal laser-beam deflection signal is linearly proportional to $I(z)$, then a semi-logarithmic plot of that signal versus z will be a straight line the slope of which is equal to $-\beta$. Such a measurement provides a direct, absolute value of attenuation -- by the definition of attenuation. This method contrasts to indirect methods in which transmitted intensity is measured or in which intensity-dependent signals are compared to a reference signal (relative measurements), and to absolute-attenuation measurements that require precise theory and knowledge of material parameters to infer β from the data.

The application of laser-beam-deflection measurements to spectroscopy or photothermal laser-beam-deflection spectroscopy (PTLBDS), was first reported by Boccara, et al.², who measured relative optical absorption as a function of wavelength for several solid samples. In subsequent studies PTLBDS was utilized to measure the optical absorption coefficient in weakly absorbing gaseous³ and condensed⁴ samples. The deflection signal was calibrated in the former study using a gaseous mixture of known β and in the latter study using solid and liquid samples that had previously been characterized with a spectrophotometer. Photothermal laser-beam deflection has also been employed to measure thermal images of surface⁵ and subsurface⁶ structure.

The concept of absolute-attenuation measurement using LBDS was tested using a chopped Ar^+ laser "pump" beam and a He-Ne laser "probe" beam. The pump beam at a wavelength of 514 nm was directed at normal incidence into a $0.7 \times 0.7 \times 1$ cm $U^{3+}:\text{CaF}_2$ crystal.⁷ The probe beam at 633 nm, a wavelength for which the sample is weakly absorbing, was directed perpendicular to the pump beam through the region of refractive-index gradient produced by the heat of the absorbed pump beam. Both

parallel (z) and perpendicular (y) deflection signals were detected by a two-dimensional position sensor. As the sample was scanned parallel to z using a stepping motor, the amplitude and phase of the deflection signal were measured by a lock-in-analyzer. The experiment was controlled and data automatically acquired using a computer.

The position-sensor signal corresponding to the component of the probe-beam deflection parallel to the direction of pump-beam propagation is shown in Fig. 1 as a function of z for $f = 25$ Hz. The data range extends from a point near the pump-beam entrance surface to a point in the bulk of the sample. The semilog plot of the data is well characterized by a straight line except near $z = 0$ (near the input surface). A least-squares fit of these data to a straight line yields the slope, and hence, β . Similar results were obtained for repeated measurements of the parallel component of deflection and for measurements of the component of deflection perpendicular to the direction of pump-beam propagation. Based on these various measurements, the most probable value of β obtained by the PTLBDS method is 10 cm^{-1} for the $\text{U}^{3+}:\text{CaF}_2$ sample at 514 nm. It should be mentioned that a position-sensor voltage of $100 \text{ } \mu\text{V}$ corresponds to a deflection angle of 5×10^{-7} radians.

Measurements on the same sample were made using two other independent methods for purposes of comparison with the PTLBDS result. Measurements of incident, reflected, and transmitted power at 514 nm were made using a photodetector and a calibrated optical attenuator. With the assumption that there is no surface attenuation for this sample, a value of 11 cm^{-1} was inferred from the reflection-transmission measurements. Previously, the attenuation in this sample had been determined from ECMD-PAS experiments.⁸ The ECMD-PAS signal was measured as a function of chopping frequency and the data were fit to the theory of McDonald and Wetsel⁹ using a computer search procedure and a provision for surface attenuation. Assuming a value for surface absorption extrapolated from the data of Hordvik and Skolnik¹⁰ for CaF_2 , the value estimated for β was about 12 cm^{-1} . A theoretical model has been developed that describes the temperature gradients produced by photothermal conversion of the

Fig. 1. Position-sensor amplifier output voltage vs. z for U^{3+} : $f = 25$ Hz, wavelength = 514 nm, $\text{U}^{3+}:\text{CaF}_2$.

pump beam and the deflection of the probe beam for the case where the probe beam propagates in the sample perpendicular to the pump beam. The theoretical procedure is similar to that for the cases of photothermal imaging of surface and subsurface structure^{6, 11}, and for PTLBDS when the probe beam is outside the sample^{11, 12}, or propagating inside the sample parallel to the pump beam.¹² The thermal diffusion equation with an axially-symmetric heat-source term characteristic of absorption from a harmonically-modulated Gaussian pump beam was first solved. Then, the Euler differential equations for the optical path, derived from Fermat's principle, were integrated to obtain the equations for the parallel and perpendicular deflection angles. For small angles, the expression for the angle corresponding to deflection parallel to the direction of pump-beam propagation is given by:

$$\Theta_{||} = -A\beta e^{-\beta z} \int_0^{\infty} \{1 - \exp[-(\sqrt{\lambda^2 + \mu^2} - \beta)z]\} \cos(y_0 \lambda) \exp(-r^2 \lambda^2 / 8) d\lambda, \\ \frac{[(\beta^2 - \mu^2) - \lambda^2]}{[(\beta^2 - \mu^2) - \lambda^2]},$$

where y_0 = initial position of probe beam relative to center of pump beam, $A = (\alpha \beta I_0^2 / 4 \kappa n_0) (\partial n / \partial T)_0$, α = fraction of absorbed light converted into heat, r = Gaussian pump-beam radius, κ = thermal conductivity, n_0 = equilibrium refractive index, $(\partial n / \partial T)_0$ = equilibrium refractive-index gradient, $\mu = (1+j)a\sqrt{f}$, $a = (\mu_p C_v / \kappa)^{1/2}$, ρ = density, C_v = specific heat, and f = chopping frequency. A similar expression is obtained¹ for $\Theta_{\perp}$.

The most important feature of $\{1\}$ vis-a-vis the absolute measurement of optical attenuation is the factor, $e^{-\beta z}$, outside the integral. If the value of the integral were independent of z or weakly dependent on z , then the functional dependence of $\Theta_{||}$ on z would be exponential or nearly exponential, respectively. The range of applicability of PTLBDS for determining β is then given by the range of parameters such that the second term in curly brackets of (1) is small compared to unity. Examination of that term reveals that $\Theta_{||} \propto e^{-\beta z}$ when $\mu > \beta$ ($\mu = \text{Re} \mu$) and $z > z_c$ where $z_c = 2.3/(\mu - \beta)^{-1}$. Numerical integration of (1) for a wide range of appropriate parameters verifies that the above inequalities provide a good measure of applicability of the method.

Calculations of $\Theta_{||}$ as a function of z using (1) for $z < z_c$ give a deviation from a straight line on a semilog plot of the same general shape as that of the data in Fig. 1; however, part of the rollover in the data near $z=0$ is due to the finite diameter of the probe beam ($\sim 0.8 \text{ mm}$). Comparison of experimental and theoretical values of $\Theta_{\perp}(z)$ involve considerations¹ similar to those for $\Theta_{||}(z)$, above.

As further evaluations of the model, $\Theta_{||}$ and $\Theta_{\perp}$ were measured as a function of y_0 . The results for the amplitude of $\Theta_{||}$ are shown in Fig. 2 along with the predictions of (1). The agreement is excellent considering that the pump beam is probably not exactly Gaussian. The experimental and theoretical variations of the phase of $\Theta_{||}$ with y_0 were also in good agreement except for $|y_0| > r$. Similarly, experimental and theoretical variations of $\Theta_{\perp}$ with y_0 are in excellent agreement.¹

Whereas the frequency dependence of $\Theta_{||}$ as indicated by (1) appears to be complicated, it is dominated by a f^{-1} dependence, as is most photothermal phenomena. The deflection angle increases with increasing β until saturation occurs; the predictions of (1) for CaF_2 are shown in Fig. 3.

In summary, we have developed a method of absolute measurement of wave or particle-beam attenuation using laser-beam-deflection spectroscopy. The experimental method and its range of applicability have been characterized by a theoretical model of PTLBDS that is in excellent agreement with the data for the test sample, a crystal of $\text{U}^{3+}:\text{CaF}_2$. The value of β obtained by PTLBDS was compared with the values obtained from two independent methods. The value of β inferred from transmission-reflection measurements is subject to errors due to surface attenuation and inhomogeneity in the sample, and to calibration errors in the optical attenuators. The value of β inferred from ECMD-PAS measurements^{8,9,13}, is subject to errors due to surface attenuation and inhomogeneity in the sample, imprecise knowledge of the material parameters of the sample, and some uncertainty about the

Fig. 2. Normalized $\Theta_{||}$ vs. y_0 for $z = 1.0$ mm, $f = 25$ Hz (+). Solid curve is calculated using eq. (1) with $\beta = 10$ cm $^{-1}$, $r = 1.0$ mm, and CaF $_2$.

Fig. 3. Theoretical $\Theta_{||}$ vs. β : $f = 25$ Hz, $z = \beta^{-1}$, $y_0 = 0$, $I_0 = 1.0$ W/m 2 , $r = 1.0$ mm, CaF $_2$.

validity of the theory and computer data fitting. The LBDS technique permits the determination of attenuation by direct probing of the pump-beam intensity, $I(z)$; only the distance (z) calibration is required. It is applicable to samples that are nearly transparent at some (probe-beam) wavelength and for which some fraction of the absorbed energy is converted into heat; yet, the measured attenuation factor, β , is characteristic of all processes that remove energy from the pump beam. The method is limited on the low- β end of the range of applicability by noise and by the sample length required ($\sim \beta^{-1}$); it is limited on the high- β end by the chopping-frequency dependence of the signal and hence eventually noise, and by the availability of sufficiently-small z - step size and probe-beam diameter. We expect that this range is from transparent to quite opaque for typical materials.

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