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PHOTOTHERMAL DISPLACEMENT SPECTROSCOPY OF SURFACES AND THIN FILMS

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Résumé -

Abstract - A new spectroscopic technique for probing the optical and thermal properties of solids and thin films is presented. Surface and bulk contributions can be readily differentiated, and a sensitivity of $\alpha\ell\sim10^{-6}/W$ is demonstrated.

We have recently developed a new spectroscopic technique, photothermal displacement spectroscopy /1/, for measuring the optical and thermal properties of surfaces, thin films, and bulk solids. The technique, which is relatively easy to implement, is particularly suited for studies requiring a wide range of pressures and temperatures.

The physical basis of photothermal displacement detection is that when an intensity-modulated beam of electromagnetic radiation (pump beam) is absorbed, heating will ensue. As the illuminated surface expands due to the optical heating, it buckles and is displaced. The magnitude of the displacement can be related to the optical absorption coefficient in a quantitative manner /1/.

The surface displacement h is approximately given by

$$h \simeq \alpha_{th}\, \beta P/(2Af\rho C) \qquad (1)$$

where α_{th} is the thermal expansion coefficient of the solid, β is the fraction of absorbed light, P is the incident power, f is the modulation frequency, A is the optically heated area, ρ the density, and C the heat capacity.

An important characteristic parameter in photothermal spectroscopy is the thermal diffusion length L_{th} which defines the depther within the solid from which the photothermal signal is generated. L_{th} is given by

$$L_{th} = (K_{th}/\pi f \rho C)^{\frac{1}{2}} \qquad (2)$$

where K is the thermal conductivity of the material. It can be seen from Eq. (2) that by increasing the modulation frequency of the bump beam, one is able to enhance the ratio of the surface to bulk contribution to the signal.

Fig. (1) shows two possible experimental arrangements for performing photothermal displacement spectroscopy: a beam deflection scheme which measures the slope of the photo-induced displacement, and an interferometric scheme which measures the displacement itself. In the beam deflection scheme (Fig. 1.a), the buckling is measured as a time-dependent change in the deflection angle of a weak probe beam as detected by a position sensor. In the case of interferometric detection (Fig. 1.b), the sample serves as an arm of a Michelson interferometer. The position of the

mirror in the other arm is modulated to eliminate thermally-induced drifts and the effects of mechanical vibrations. Both schemes yield comparable sensitivities ($\alpha l \sim 10^{-6}$); however, the beam deflection approach is easier to implement.

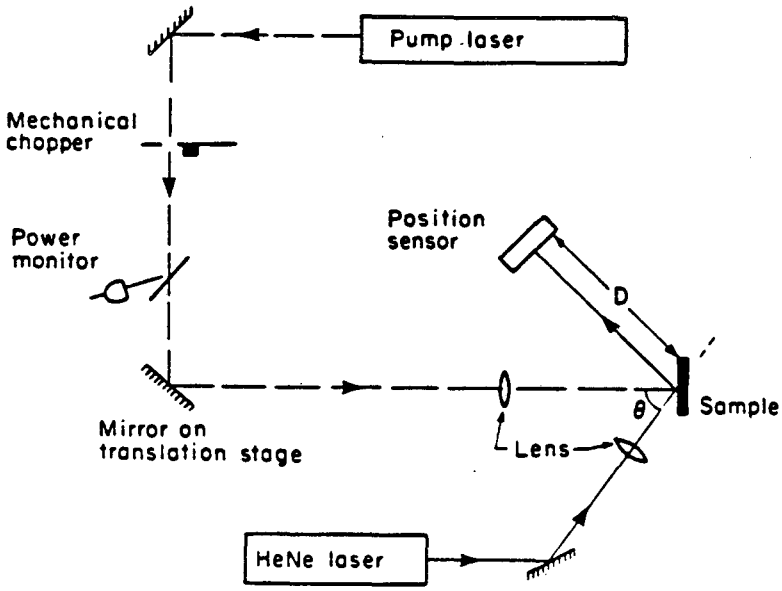

Fig. (1.a)

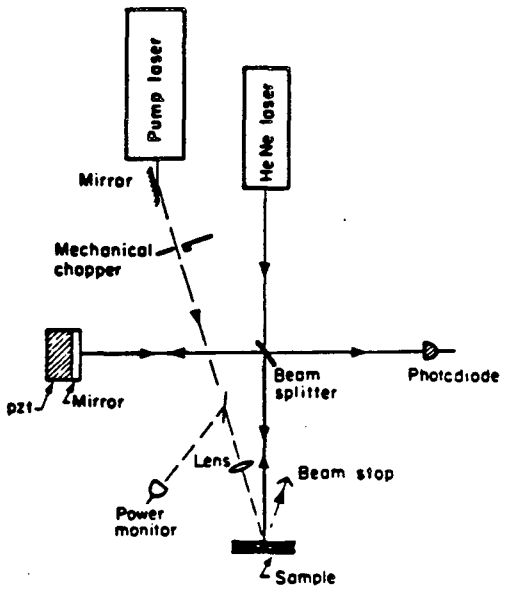

Fig. (1.b)

Fig. 1: The Experimental Arrangement.

To demonstrate the spectroscopic feasibility of this technique, we measured the absorption spectrum of didymium glass in the 700-800 nm range (Fig. 2). Spectra were obtained at atmospheric pressure and 20 m torr. For reference, we give the spectrum as obtained by conventional transmission methods. That the spectra are identical establishes that photothermal displacement spectroscopy indeed measures the optical absorption.

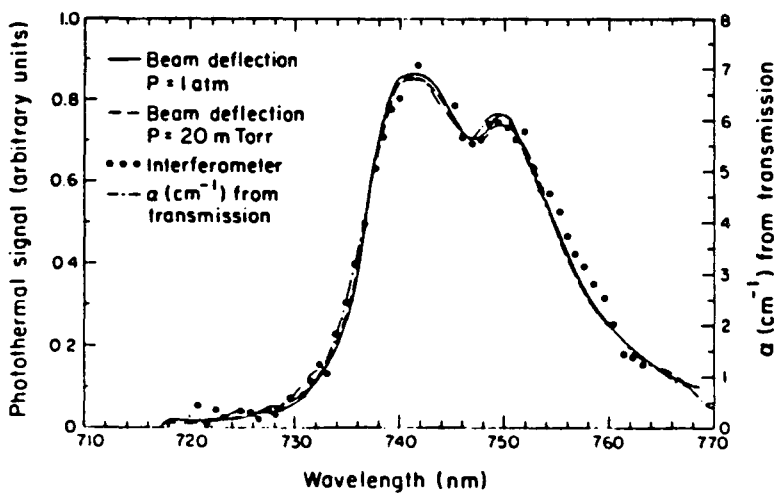

Fig. 2: Absorption Spectra of Didymium as Obtained by Photothermal and Conventional Methods.

The ability of photothermal detection to differentiate between surface and bulk or substrate absorptions is demonstrated in (Fig. 3.a). A 50 Å gold film was evaporated on a 2 mm thick didymium glass. [The absorption spectra of the gold (50 Å film on a transparent substrate), and of pristine didymium (without the gold film) are shown in Fig. (3.b)].

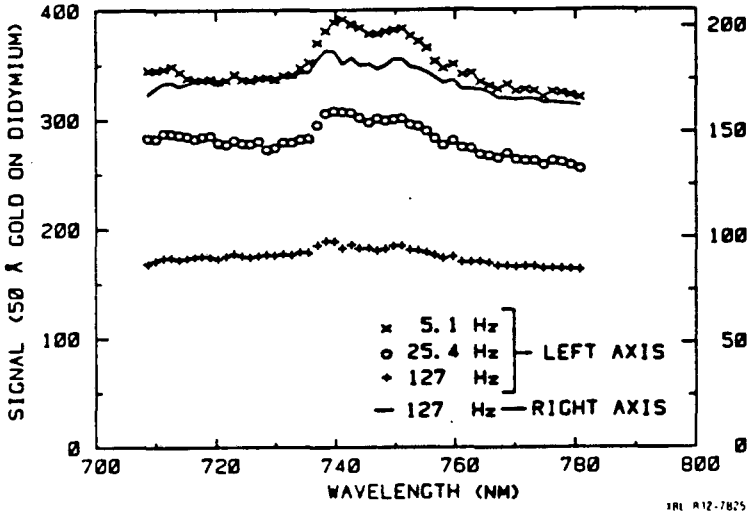

Fig. (3.a)

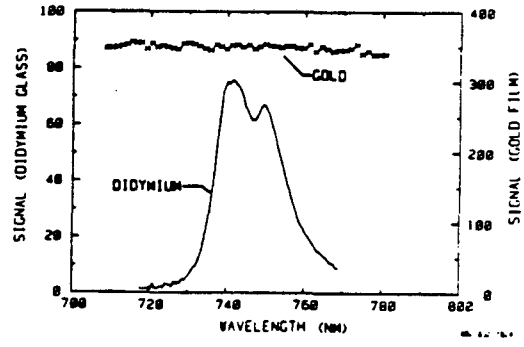

Fig. (3.b)

In the region of 700-800 nm, approximately 25% of the incident light is absorbed by the gold film; with the didymium having a strong absorption coefficient of ~ 6.5 cm around 740 nm. As shown in Fig. (3.a), at a modulation frequency of 5 Hz, the photothermal signal of the gold-coated didymium is the sum of the featureless gold absorption plus the peaks of the didymium absorption band. As the modulation frequency is increased to 127 Hz, the gold signal is decreased only by a factor of 2, while the didymium peaks are decreased by an additional factor of 2-3. Clearly, higher modulation frequencies will result in a further reduction of the substrate (or the bulk) contribution to the photothermal signal.

In conclusion, we have presented a new and sensitive photothermal spectroscopic tool which is particularly suited for in situ characterization of materials under a wide variety of experimental conditions. It requires no mechanical or wetted contacts and is particularly suited for research which requires ultra-high vacuum.

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References

(1) OLMSTEAD, M.A., AMER, N.M., KOHN, S.E., FOURNIER, D., and BOCCARA, A.C., submitted to Appl. Phys. A.

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