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Publication Date

1983



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Invited talk presented at the Third International Meeting on Photoacoustic and Photothermal Spectroscopy, Paris, France, April 5-8, 1983

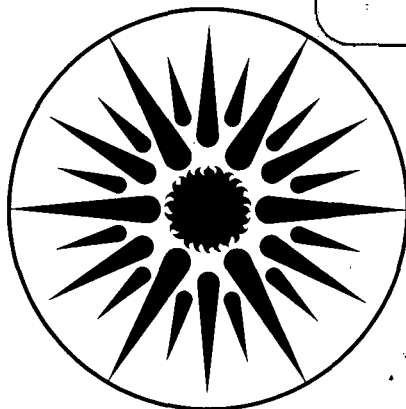
PHOTOTHERMAL DISPLACEMENT SPECTROSCOPY OF SURFACES AND THIN FILMS

N.M. Amer, M.A. Olmstead, D. Fournier, and A.C. Boccara

January 1983

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PHOTOTHERMAL DISPLACEMENT SPECTROSCOPY OF
SURFACES AND THIN FILMS

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We have developed a new technique, photothermal displacement spectroscopy, for studying the optical and thermal properties of surfaces.

Photothermal displacement spectroscopy is based on optical detection of the thermal expansion of a sample as it is heated by absorption of electromagnetic radiation. Details of the experimental configuration are shown in Fig. 1. An optical absorption spectrum is generated as an intensity modulated, tunable light beam (pump beam) is focussed onto the sample, and, depending on the optical cross section, some fraction of the radiation is absorbed. As the excited electrons decay non-radiatively, the sample is heated. The illuminated surface is then displaced as the sample expands. A probe beam reflected from the surface is deflected by the slope of the surface displacement.

This deflection is measured by a position sensitive photodiode whose output is then amplified by a phase sensitive lock-in amplifier referenced to the pump beam modulation. Thermal information can be obtained by measuring the shape and phase of the photothermal displacement relative to the illumination as a function of the modulation frequency [1].

The absence of any electrical or mechanical contact with the sample makes this technique especially well suited for ultrahigh vacuum studies, including experiments when a wide temperature variation is necessary. Photothermal displacement spectroscopy can also be performed using interferometry to measure the height of the surface displacement, although long term stability requirements make the beam deflection method more suitable for most experimental situations [1].

The ability of photothermal displacement spectroscopy to distinguish between surface and bulk absorptions on the same sample is demonstrated in figure 2. A 50 Å gold film was evaporated on didymium glass. In the wavelength region investigated, about 1/4 of the incident light is absorbed by the Au film; didymium glass has a strong absorption band with a peak value of 6.4 cm^{-1} (see Fig. 2b). For the Au coated didymium glass, the photothermal signal is the sum of the structureless surface (thin film) absorption due to the gold and the peaked substrate absorption. As the frequency is increased by a factor of 25, the signal due to the Au (in the region $< 730 \text{ nm}$, e.g., all the signal is due to the Au) is decreased by a factor of about 2; the signal due to the glass (the bump at the center of the spectrum) has decreased by an additional factor of 2 or 3. By examining the change in the spectrum as a function of frequency, it is possible to distinguish absorptions occurring at the surface from the more uniform bulk absorption.

The increased sensitivity of photothermal displacement spectroscopy to surface absorptions over bulk can be seen by noting the relative magnitudes of the two signals. Using published values for the index of refraction of evaporated gold films, the percentage of the incident light reflected, transmitted and absorbed at 750 nm can be calculated using the three layer Fresnel equations to be 15%, 59% and 26%, respectively. The 6.4 cm^{-1} peak absorption of the 0.25 cm thick didymium glass absorbs 80% of the light transmitted through the Au film. Thus about twice as much light is absorbed in the glass as on the surface, but the signal due to the didymium at 5.1 Hz is only about 1/7 of the

signal due to the 50 Å film. Even if the effective length of the didymium is taken to be the thermal length, the signal is still heavily weighted by the surface heating. At 5.1 Hz the thermal length of the glass is about 160 microns, so 10% of the transmitted light (6% of the incident light) is absorbed in the first thermal length. Thus the didymium signal would be expected to be about 1/4 that of the Au signal, which is clearly not the case.

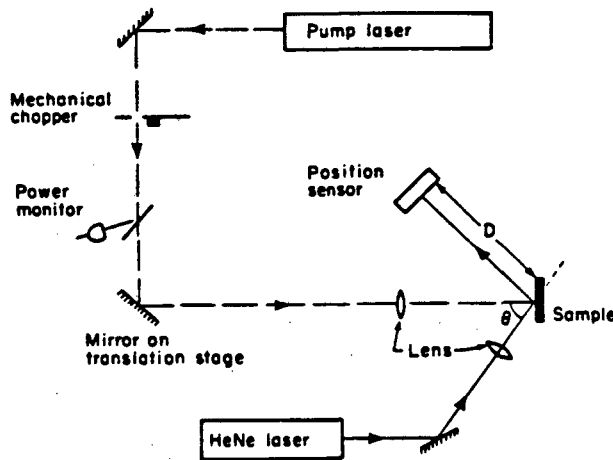


Fig. (1)

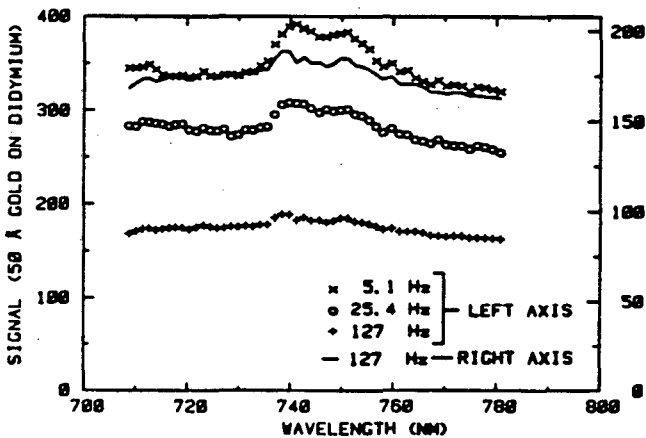


Fig. (2a)

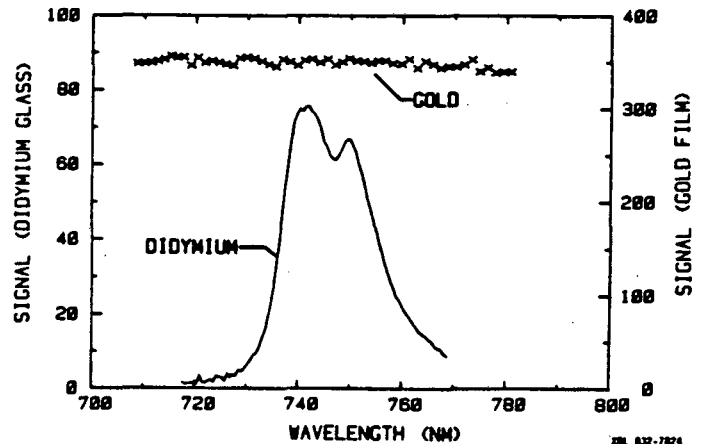


Fig. (2b)

References: 1) M. Olmstead, N. M. Amer, D. Fournier and A. C. Boccara, to appear in Appl. Phys. A.

This work was supported by the Office of Energy Research, Physical and Technological Research Division of the US Department of Energy under contract No. DE-AC03-76SF00098, and by DARPA Contract N. 3343.

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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