Title
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DISCLAIMER

A NOVEL METHOD FOR THE STUDY OF OPTICAL PROPERTIES OF SURFACES

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Abstract

A new technique, photothermal displacement spectroscopy, for probing the optical properties of surfaces is presented. Optical spectra are obtained directly, and surface and bulk contributions are easily and unambiguously differentiated. The technique is well suited for in situ ultra-high vacuum experiments.

The optical properties of electronic surface states are of significant importance from fundamental and technological viewpoints. A variety of optical spectroscopic techniques have been used to investigate such properties.[1] Most prominent among them are reflectance and ellipsometric methods. However, to determine the contribution of surface states, the difference between spectra of clean and intentionally contaminated surfaces must be measured.

We have recently developed a new sensitive technique, photothermal displacement spectroscopy,[2] for directly measuring the optical and thermal properties of surfaces and thin films. The technique, which is relatively easy to implement, is particularly suited for studies requiring wide ranges of pressures and temperatures.

The physical principle underlying photothermal displacement spectroscopy is that when a modulated beam of electromagnetic radiation (pump beam) is absorbed, heating of the absorbing medium will occur. As the illuminated surface expands due to the optical heating, it buckles and is displaced. The magnitude of the displacement is related quantitatively to the optical absorption coefficient.[2]

The surface displacement h is given by

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

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

An important characteristic parameter in photothermal spectroscopy is the thermal diffusion length, L_{th}, which defines the depth within the sample 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_{th} is the thermal conductivity of the material. The frequency dependence of the thermal diffusion length is what we exploit to differentiate between surface and bulk contributions to the photothermal signal. It can be readily seen from Eq. (2) that by increasing the modulation frequency of the pump beam one is able to enhance the ratio of surface to bulk contributions.

Fig. (1) shows two possible experimental configurations 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 Fig. (1.a) we give the details of the beam deflection scheme. The time-dependent change in the slope of the photo-induced surface buckling is measured as a time-dependent change in the deflection angle of a weak probe beam (typically a He Ne laser) as detected by a position sensor.

The interferometric scheme is shown in Fig. (1.b). The sample of interest serves as one arm of a Michelson interferometer. The position of mirror in the other arm is modulated to overcome thermally induced drifts and the effect mechanical vibrations on the interferometers.

Both schemes have comparable sensitivities ($\alpha \ell \sim 10^{-6}$); however, the beam deflection approach is easier to implement.

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 at 20 m Torr. For reference, we give the absorption spectrum in this wavelength region as obtained by conventional transmission methods, thus establishing that photothermal displacement spectroscopy indeed measures the optical absorption.

The ability of photothermal deflection spectroscopy to differentiate between surface and bulk absorptions is demonstrated in Fig. (3.a). A 50 Å gold film was evaporated on 2 mm thick didymium glass. The absorption spectra of gold only (50 Å film on a transparent substrate) and of didymium only are given in Fig. (3.b). In the wavelengths region of 700-800 nm, approximately 25% of the incident light is absorbed by the gold film; with didymium having a strong absorption peak of $\sim$6.5 cm^{-1} around 740 nm. As shown in Fig. (3.a), at 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 by a factor of 2, while the didymium peaks are decreased by an additional factor 2-3. Higher modulation frequencies will clearly result in a further reduction of the substrate (bulk) contribution to the photothermal signal.

In conclusion, we have presented a new and sensitive photothermal technique[3] which directly measures the optical absorption of surfaces and obviates the need for the subtraction of spectra necessary in the case of conventional optical absorption spectroscopy. The technique is

readily suited for UHV studies, requires no mechanical or electrical contacts, and is linear over a wide range of absorption coefficients.

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References

(1) See, for example, H. Lüth, Appl. Phys. 8, 1 (1975).

(2) M.A. Olmstead, N.M. Amer, S. Kohn, D. Fournier, and A.C. Boccara, submitted to Appl. Phys. A.

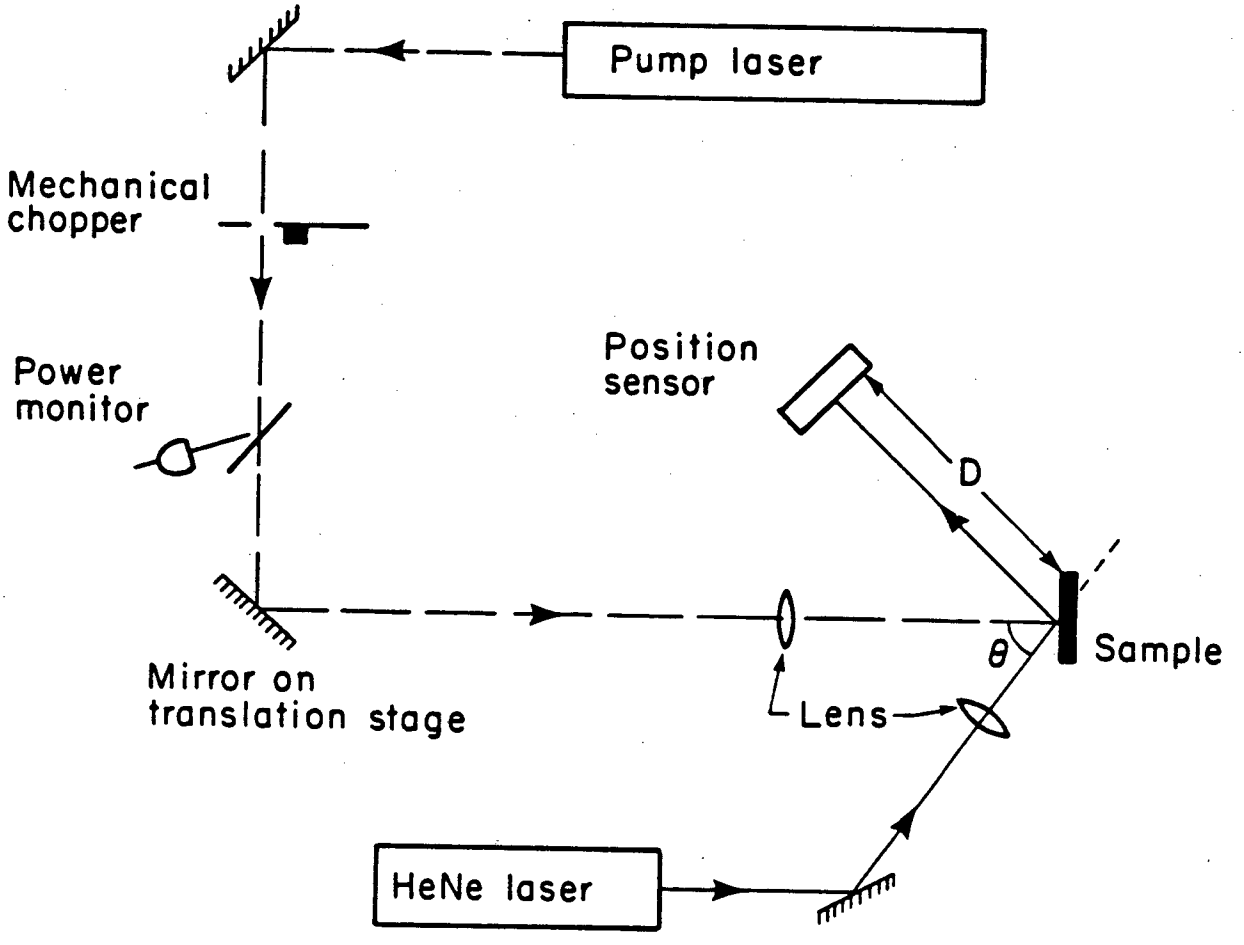
(3) Other photothermal techniques include photoacoustic spectroscopy (see, for example, Optoacoustic Spectroscopy and Detection, Y.-H. Pao, Ed., Academic Press, 1977; W.B. Jackson and N.M. Amer, J. Appl. Phys. 51, 3343 (1980)), and photothermal deflection spectroscopy (W.B. Jackson, N.M. Amer, A.C. Boccara, and D. Fournier, Appl. Opt. 20, 1333 (1981)). However, these techniques are not suited for experiments which require UHV or wide range of temperatures.

Figure Captions

Fig. 1: Experimental Configuration: a) beam deflection scheme, b) interferemetric scheme.

Fig. 2: Absorption spectra of didymium as obtained by photothermal displacement spectroscopy and by conventional transmission techniques.

Fig. 3: a) Surface vs. bulk differentiation, b) spectra of neat gold (50 Å film on transparent substrate), and of neat didymium glass (2 mm thick).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Mechanical chopper
Power monitor
Position sensor
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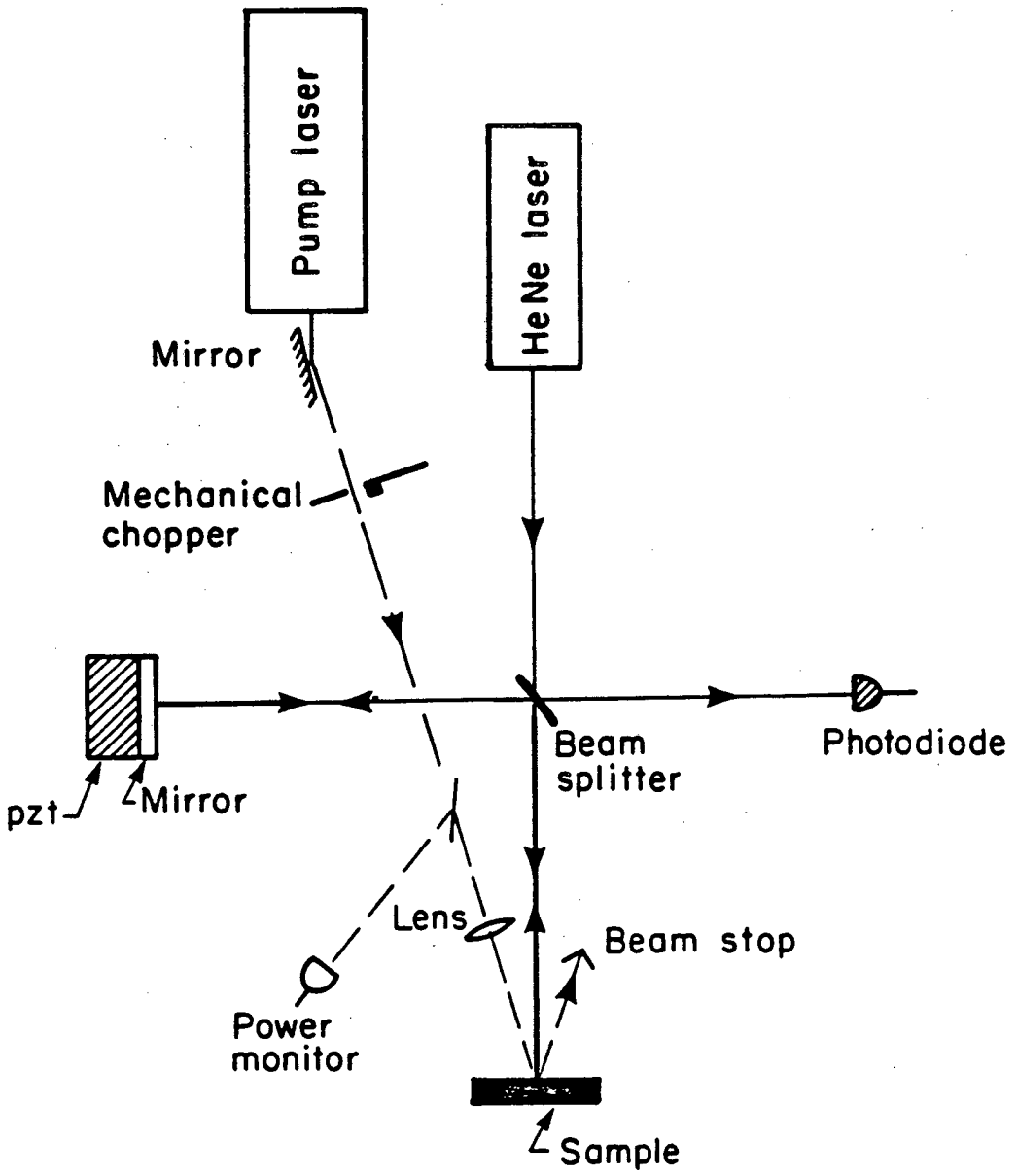
Sample
θ
Mirror on translation stage
Lens
HeNe laser
XBL 831 - 22

Fig. (1.a)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He Ne laser
Mirror
Mechanical chopper
pzt
Mirror
Beam splitter
Photodiode
Lens
Beam stop
Power monitor
Sample
XBL 832-7779

Fig. (1.b)

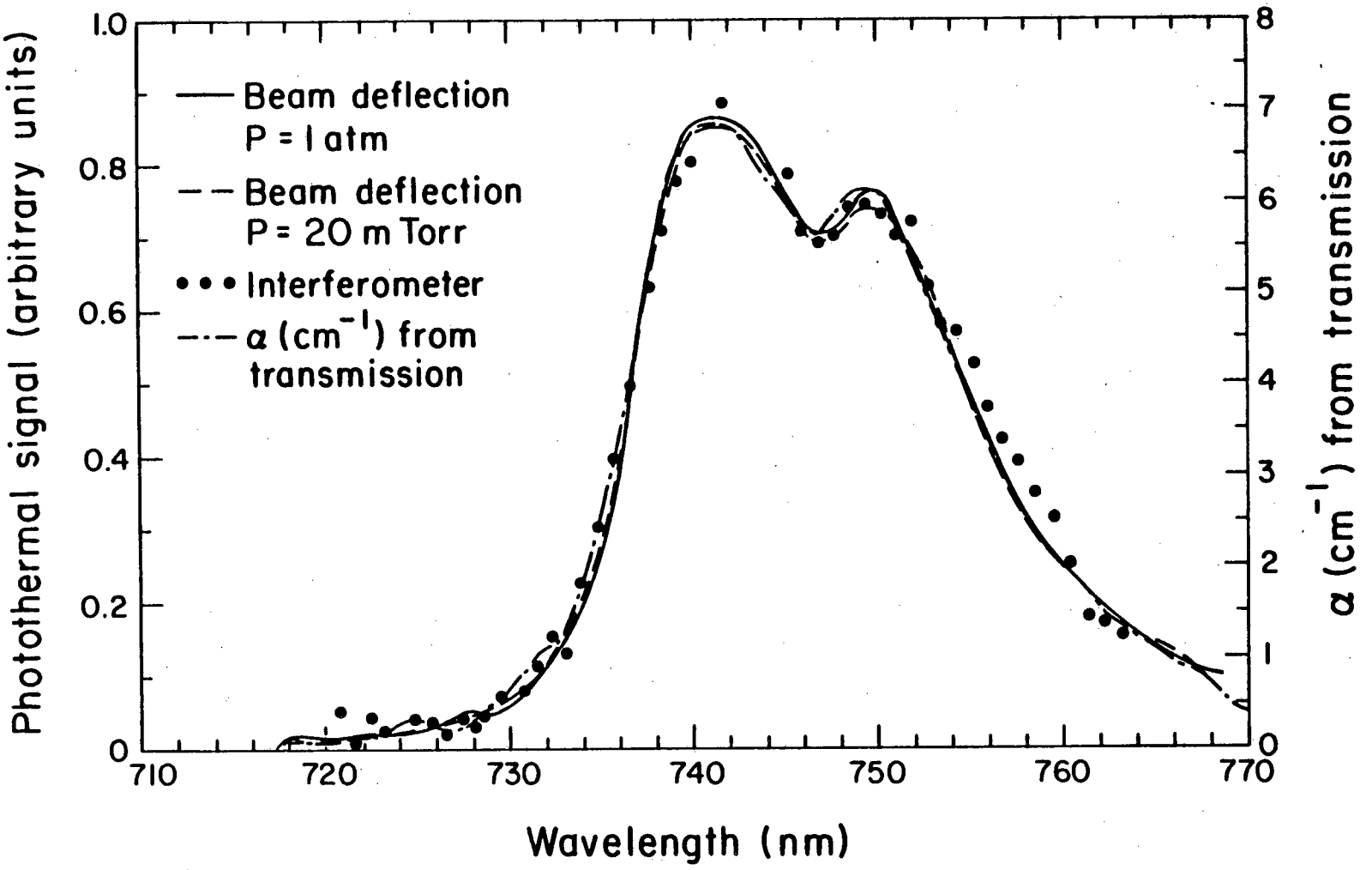

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Fig. (2)

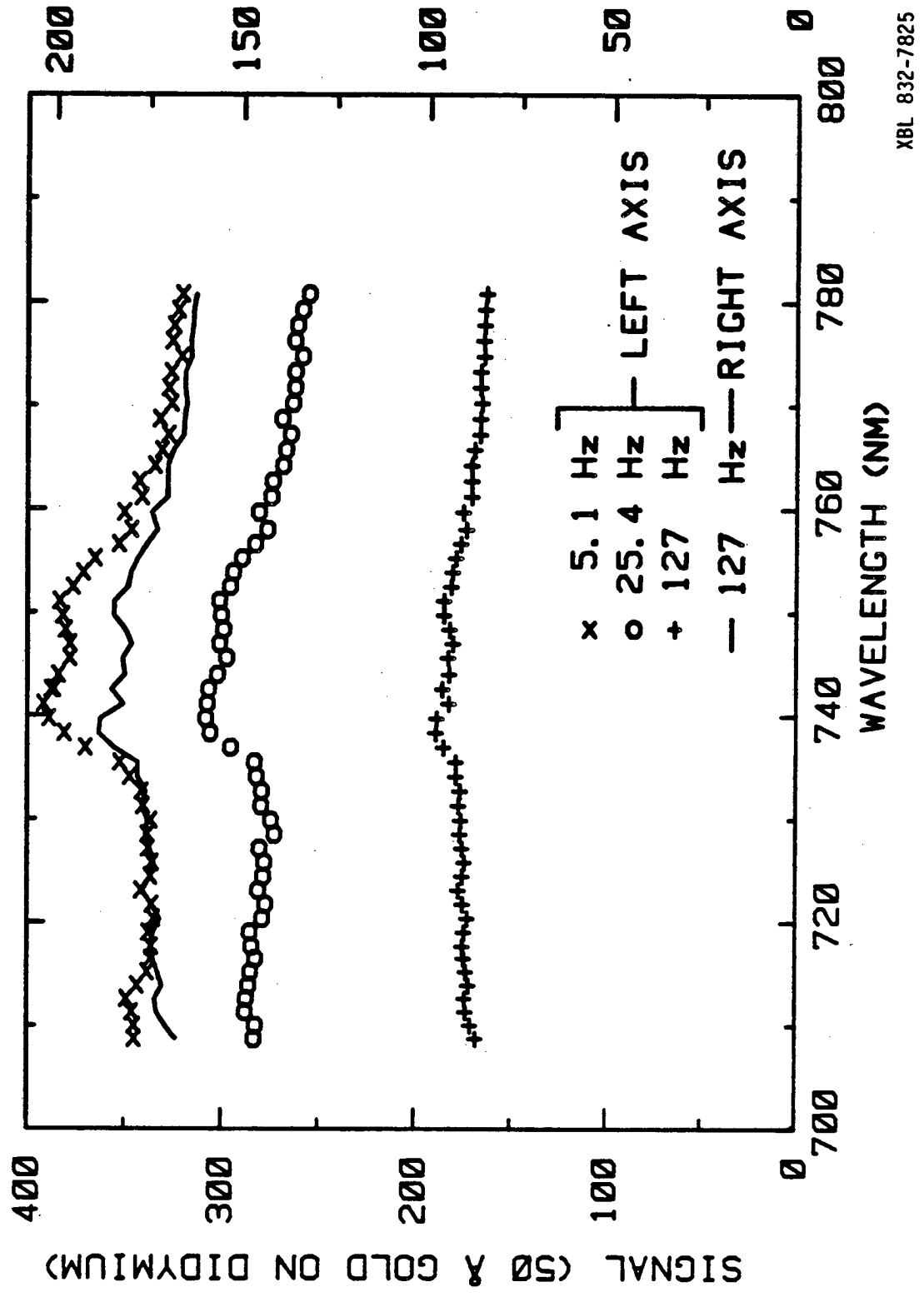

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Fig. (3.a)

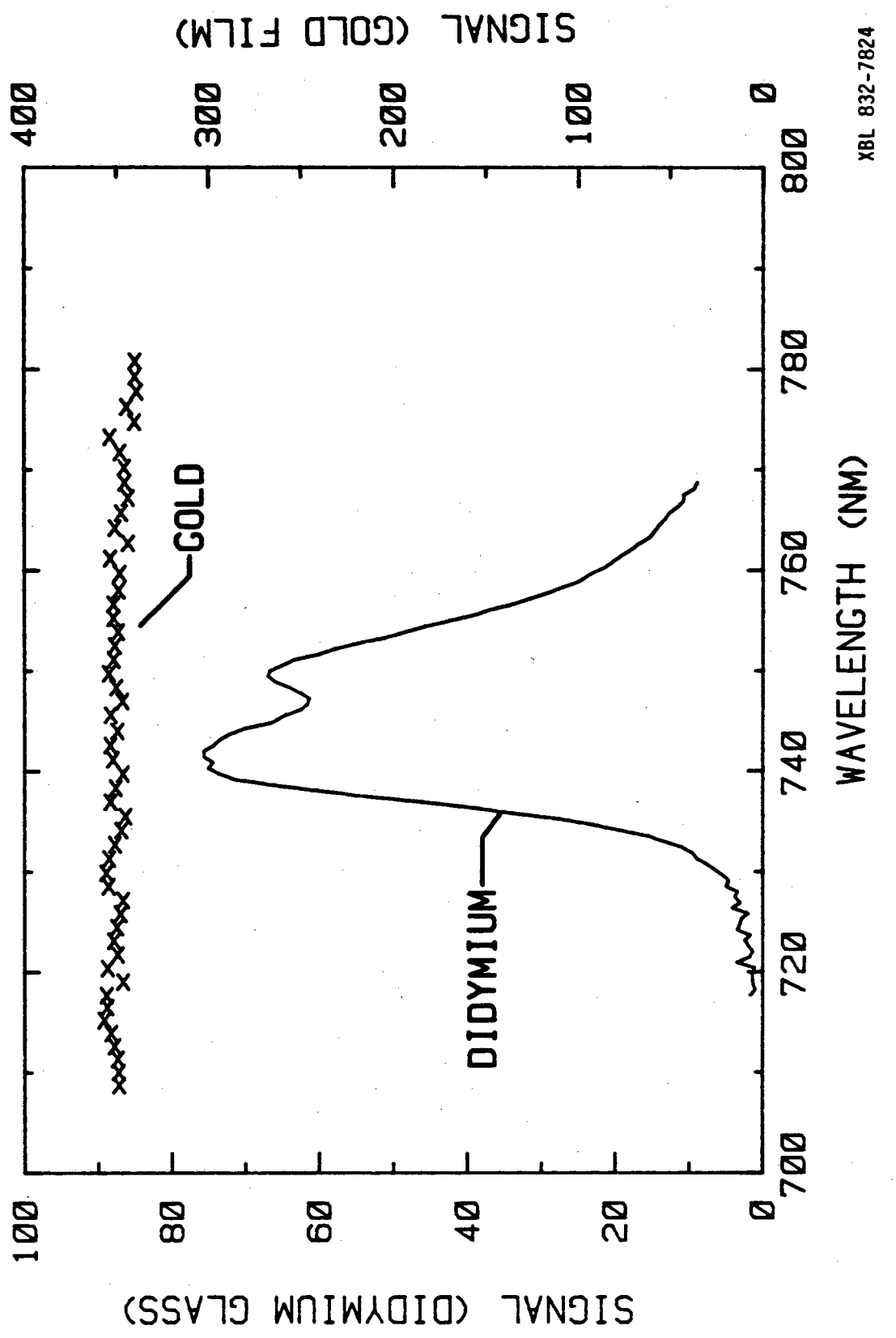

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Fig. (3.b)

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