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THERMOTRANSMISSION MEASUREMENTS ON SELF-SUPPORTING METAL FILMS.

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M. Meuti, C. Coluzza[*], C. Quaresima[*], and R. Rosei[+]: THERMO-TRANSMISSION MEASUREMENTS ON SELF-SUPPORTING METAL FILMS[o]

ABSTRACT: - Thermotransmission measurements have been carried out on thin self sustained semitransparent gold films in the 1. 8 ÷ 3. 5 eV energy region. The results compare very favorable with the spectra of Rosei et al., taken on quartz backed samples, showing the feasibility of extending the techniques of modulation spectroscopy in the soft X-ray region.

It is shown also that the width of the lineshape is a factor of two smaller than that obtained on films evaporated on a quartz substrate.

The thermal time constant is in the range of a few milliseconds so that a veryhigh modulation efficiency can be obtained even operating at several hundred Hertz.

1. - INRODUCTION

Modulation spectroscopy has been one of the most powerful techniques in the last decade or so, for investigating the electronic structure of solids [1-4].

Several different techniques (piezo, electro thermo-modulation, etc.) have been useful in the study of metals, semiconductors and insulators, allowing an in depth view of the position and symmetry of energy levels.

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For many years this wealth of information has been within the reach of the experimenter only in a rather small energy window (0.5 ÷ 5.5 eV) and only in isolated efforts[5,6] the energy limits have been pushed to the transmission limit of LiF (~12 eV).

Quite recently, Piacentini et al.[7] and Aspnes et al.[8], have been able to extend substancially the energy region of modulation spectroscopy (up to 35 eV) with the help of the intense synchrotron radiation continuum emitted from the extremely stable beam in a storage ring[9].

Working on quartz supported gold films, Piacentini et al.[7], have been able to show that thermoreflectance is still a very useful techinique even if core levels or high lying states are involved[10].

Aspnes et al.[8,11], using electroreflectance measurements, have carried out a very detailed investigation of the conduction bands of the III-V semiconductors by exciting d-like core levels.

Virtually no modulation experiment[12] has been attempted beyond about 35 eV. The reason for this lies in the fact that above this energy, the reflectance of most solids drops to very low values, making impracticable any reflectance measurement (modulated or not). The only way to investigate the optical properties of solids in the soft X - ray range consists in performing transmission measurements on thin self-supported films[13]. This has been done quite successfully in the past few years[1]. The application of an external perturbation to produce a modulated optical response on these very fragile samples represents however a difficult technological problem.

In the following we describe an experiment of thermotransmission on thin selfsupported gold films performed in the 1.8 ÷ 3.5 eV photon energy region. The success of our effort shows the feasibility of the technique and the possibility of streightforward extension in the soft X-ray region.

2. - EXPERIMENTAL

Gold films of suitable thickness (200 ÷ 500 Å) have been evaporated on glass slides onto which a very thin film of NaCl had been predeposited.

Using a well developed technique[13], the films have been cut to size (approximately 5x12 mm^2) and floated off using distilled water with a few drops of Tensol. The films were then deposited on a thin slit (0. 5x8 mm^2) machined on a stumatite support (see Fig. 1). The stumatite was

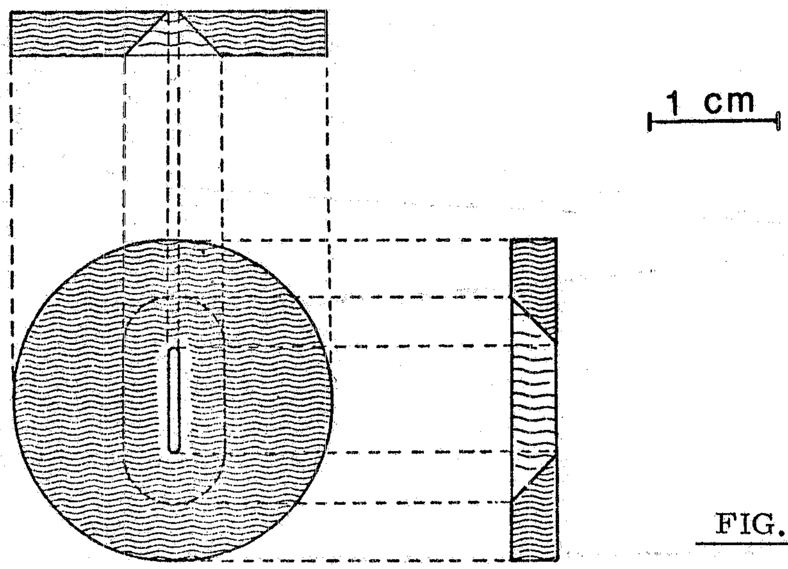


FIG. 1

chosen since it can be very easily machined to any size and shape. After keeping it for about 20 hours in an oven slowly heated up to 1175 ^oC, it becomes a very hard snd stable insulating material.

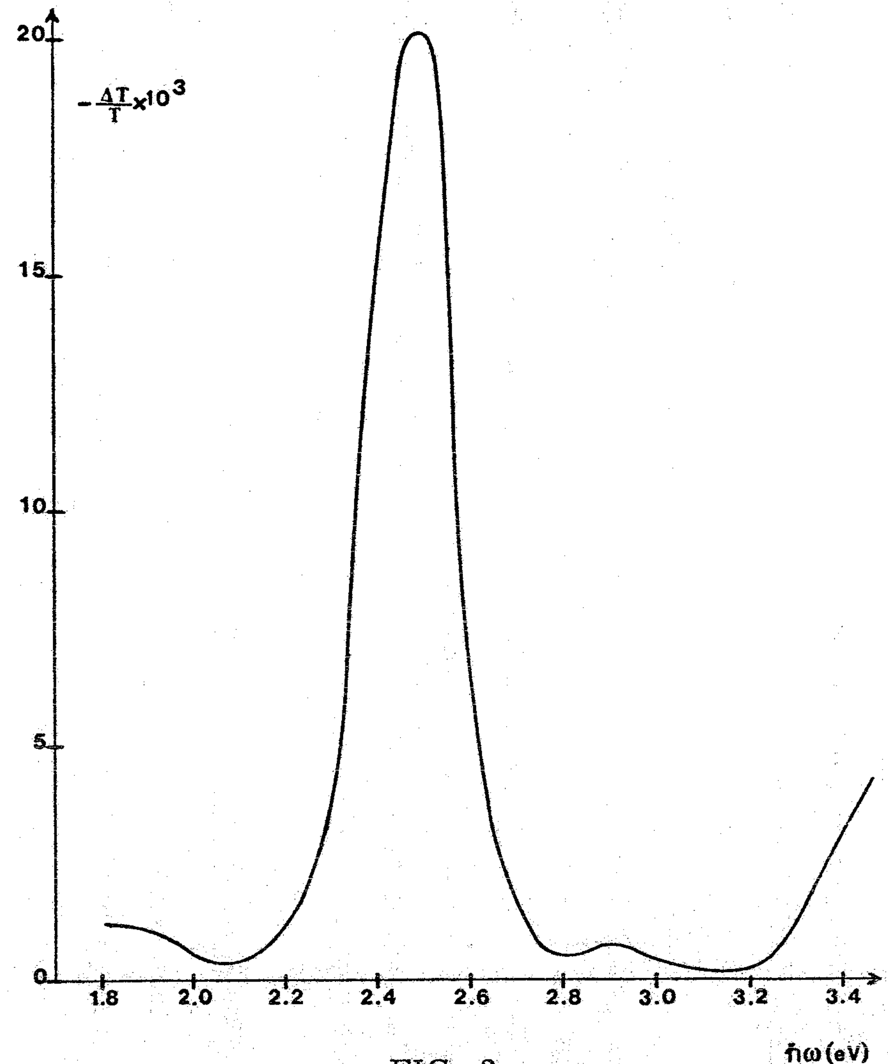
Electrical leads were indium soldered on gold contacts evaporated on the stumatite support. The electrical contact between films and leads was ensured through a thin layer of silver print. The support was then mounted on the cold finger of a cryostat (but the measurements were carried out at room temperature).

Square waves of different frequencies and power from a power amplifier provided the temperature modulation.

The optical layout has been described elsewhere[15] and the description will not be repeated here.

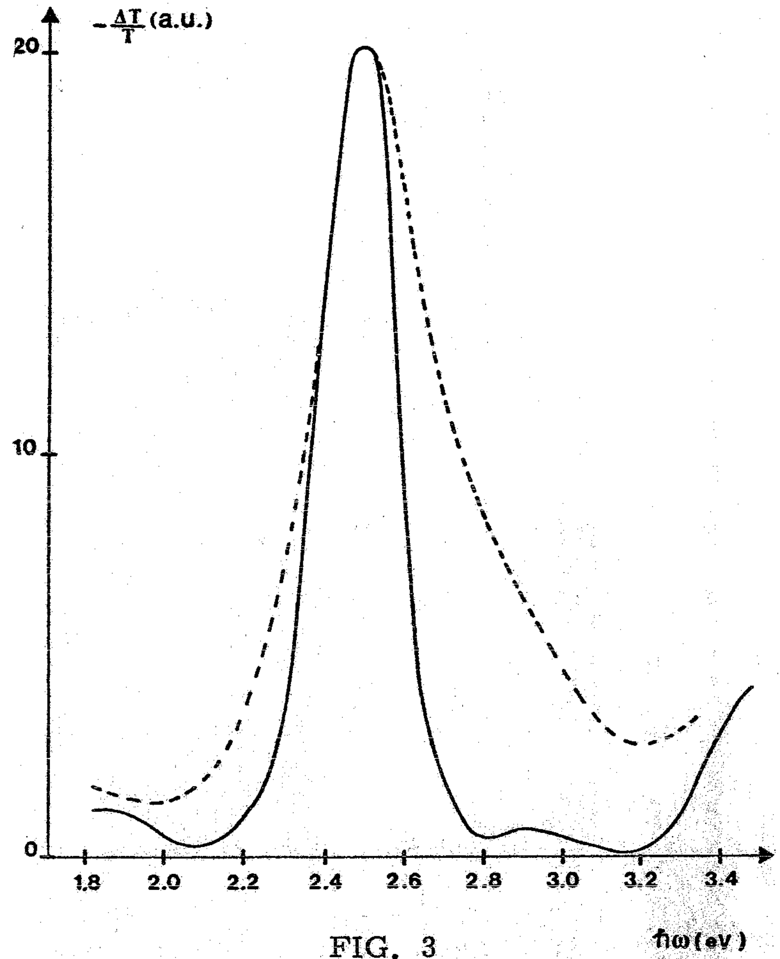
3. - EXPERIMENTAL RESULTS AND DISCUSSION

The spectrum of thermotransmission for a 220 Å thick self-sustained film of gold is shown in Fig. 2. The operating frequency was 75 Hz and the



$-\frac{\Delta T}{T} x 10^3$
20
15
10
5
0
1.8
2.0
2.2
2.4
2.6
2.8
3.0
3.2
3.4
ħω(eV)

FIG. 2



$-\frac{\Delta T}{T}$ (a.u.)
20
10
0
1.8
2.0
2.2
2.4
2.6
2.8
3.0
3.2
3.4
ħω(eV)

FIG. 3

power dissipation 0.25 W.

The strong peak at 2.50 eV is due to the L_3 - $L'_2(E_F)$ transition, as it is well known[16]. The structure on the high energy side, beginning at about 3.2 eV, comes from the $L'_2(E_F)$ -L_1 transition, as it is now well established[17].

The tiny bump at 2.9 eV has been attributed to a transition from a second lower lying d-level to the Fermi surface[15]. The structure at about 1.85 eV is new and it should probably be attributed to the X_5-$X'_4(E_F)$ transition[16].

The important point here,however, is not the origin of the different structures but the quality of the spectrum we have obtained. Indeed one of the major points was to check whether the sensitivity and reliability of this technique would be sufficient to obtain significative results in more difficult spectral ranges (V.U.V. and soft X-ray regions).

In Fig. 3 we compare our spectrum with the thermotransmission spectrum of quartz backed film (taken from Rosei et al.[15]). The spectra have been normalized to the same peak height to make the comparision easier.

It is easily seen that no spurious signals are present in our spectrum. This was a very important point to check. Indeed it was feared that the current pulses may cause vibrations in our sample which in turn may give origin to spurious signals in the photomultiplier.

We note also that,although the temperature of our film is estimated to be roughly the same as that of the sample of Rosei at al.[15] (slightly above room temperature), the width of the lineshape of the most prominent peak is almost a factor of two smaller. Also, the transition at 2.9 eV is completely resolved in our spectrum. Indeed it had been shown[18, 19] that metal films evaporated on glass or quartz substrates are rather badly strained. This is probably due to the difference in the thermal expansion coefficients of sample and substrate. It was shown also[20] that thermoreflectance measurements on a copper high quality bulk sample at room temperature gave smaller widths than measurements performed on a quartz backed film at liquid nitrogen temperature.

Our present results show that selfsupported samples are much less strained than quartz backed samples.

Other interesting and important features of our experiment are:

a) The peak value of $\frac{\Delta T}{T}$ is about $2x10^{-2}$ for a power input in the sample of only 250 mW. This is to be compared with a peak value of $\frac{\Delta T}{T}$ about an order of magnitude smaller obtained by Rosei et al.(15) using a substancially higher power input (about 1.5 W).

b) The system has a very fast thermal response. Fig. 4 reports the peak height of $\frac{\Delta T}{T}$ versus frequency. As expected, the thermal system behaves approximately as a low pass filter: the cutoff frequency is about 270 Hz and above it the signal diminuishes at 20 dB/decade.

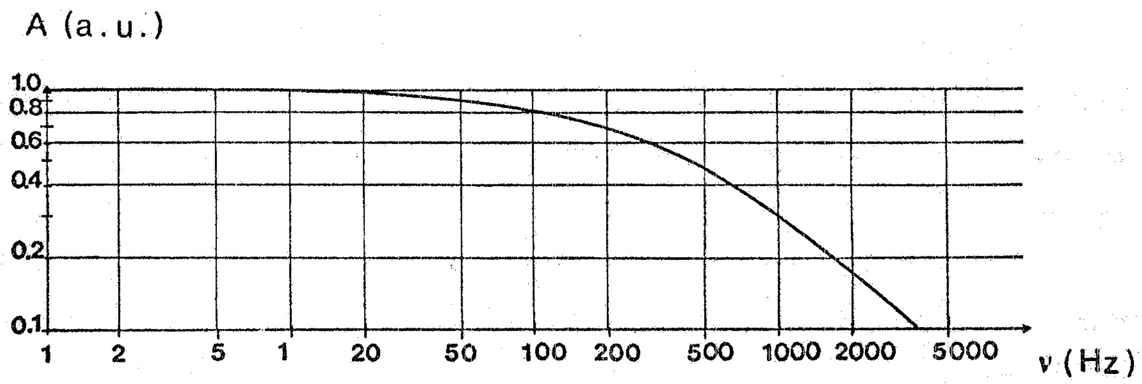


FIG. 4

Since the limit of power dissipation in the sample is substantially higher than 250 mW (several watts), the technique seems suitable for retrieving particularly small contributions in thermomodulation spectra. An attempt in this direction is at present pursued in our laboratory to see whether it may be possible to detect very weak structures discovered recently in the static optical absorbtion spectra(21) of Al.

A technique with such an enhanced sensitivity seems extremely suitable for measurements in the V.U.V. and in the soft X-ray range where even the most stable storage ring provides performances in optical modulated spectra which are somewhat limited by fluctuations.

4. - CONCLUSIONS

We have set up a thermotransmission experiment which is extremely promising for extending modulation measurements in the soft X-ray range. Very strong signals are obtained with a low power dissipation and the thermal system is very fast (response in the milliseconds range). As an extra bonus, the technique seems also very promising and competitive in the more convetional optical range.

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