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INVESTIGATIONS ON THE CONSISTENCY OF OPTICAL CONSTANTS IN THE XUV DETERMINED BY DIFFERENT METHODS

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In the photon energy range from 40 to 1000 eV, we have measured reflectances and transmittances of polyimide foils as well as reflectances and total photoelectron yields of Cu and Pt films. We compare sets of optical constants derived by the different methods applied to the same sample. The degree of agreement gives an indication of the present status of determining optical properties in the XUV. This is relevant for the planning of advanced instrumentation.

INTRODUCTION

Published optical constants, reflectivities, and transmission coefficients in the photon energy range from 6 eV to some keV show numerous inconsistencies. Materials commonly used for mirror coatings are not exempted. In principle, the discrepancies may arise from differences in the samples or from invalid assumptions in evaluating the measurements. For practical applications like multilayer tayloring, for example, it is mandatory to have more reliable data, at least data with reliable error margins.

We therefore attack the problem by comparing results, which have been obtained by different methods applied to the same sample; i.e., we measure the reflectance and the relative total photoelectron yield as a function of the angle of incidence θ, and for free-standing thin films the transmittance as a function of the photon energy hν. By a careful evaluation of the discrepancies between optical constants obtained from two of these data sets, systematic errors in the measurements and evaluation procedures can be revealed.

With polyimide foils we were able to perform both transmittance and reflectance measurements on the same samples. In the XUV, this normally fails due to two contradictory requirements, both of which were fulfilled in our case. The measurement of the reflectance requires samples of about 10 mm length with surfaces of optical quality, while sufficient transmittance is available only from samples that are thinner than 1 µm.

Reflectance versus angle of incidence spectra are evaluated by least-squares fits to the Fresnel formulas. Interface roughness is taken into account by a factor $\exp[-(4\pi\cdot\sigma\cdot\cos\theta/\lambda)^2]$, where σ, θ, and λ denote rms roughness, angle of incidence, and vacuum wavelength, respectively. In the case of polyimide foils two independent roughness amplitudes have been used for the front and rear side of each foil [1,2].

The photoelectric yield can theoretically be described in terms of the dielectric function ε of the material and an average electron escape depth L. As shown by Pepper [3], for a thin film its thickness and the dielectric properties of the substrate must be taken into account. We used Pepper's equations in a least squares fit procedure.

Transmittance versus photon energy data taken at normal incidence are submitted to a Kramers-Kronig transformation. Since in our experiments the reflectance is very small, it is neglected in the evaluation.

I. EXPERIMENT

The experiments were performed at the synchrotron radiation laboratory HASYLAB. As shown in Fig. 1, our setup consists of a plane grating monochromator combined with a UHV reflectometer.

The monochromator BUMBLE BEE supplies the experiment with radiation of photon energies between 15 and 1500 eV. Its principles and characteristics have been described in ref. [4]. By proper choice of the operating modes, the spectral purity of the monochromatized beam can be optimized. Further improvement is achieved by the use of transmission filters (Al, Ag, In, Cu). In most parts of the energy range, where the present data were obtained, the purity was clearly above 99 %.

A toroidal mirror behind the exit slit refocuses the beam; near the sample, the beam size is 0.85×2.2 mm^2 (FWHM, vertical × horizontal) and its divergence is 2.1×4.8 mrad^2. For the experiments on the polyimide foils, the vertical parameters were reduced to 0.24 mm and 1.1 mrad.

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The reflectometer provides computer-controlled independent rotations and translations of the sample and detector by means of special feedthroughs [5]. Their rotational increment is 0.005° and the uncertainty of angles is less than 0.03°. An electron gun for in situ preparation of thin films is installed in the lower part of the chamber. The whole setup is bakable and allows pressures down to $3 \cdot 10^{-8}$ Pa.

The measurement of the transmitted or reflected photon fluxes is performed with semiconductor diodes (Hamamatsu G1127). Their suitability has been investigated and is reported elsewhere [6,7]. Since the sample can be totally removed from the beam, the necessary normalization can be done by just moving the detector into the direct beam. The corrections for changes of the incoming photon flux are made by monitoring the total electron yield from the toroidal Kanigen mirror in front of the chamber.

The polyimide foils were manufactured by polymerization of liquid PIQ 13 that was obtained from Hitachi Chemicals Europe. By mounting them on steel rings, we achieve a sample flatness such that no deflection of the beam was detectable when scanning along the sample.

The Cu and Pt samples were prepared in situ in an UHV chamber by evaporating films onto substrates of borosilicate glass or "white crown" glass. During the evaroration processes, the pressure rose to values between 10^{-4} and 10^{-3} Pa. The evaporation rates were 5 – 9 nm/min.

II. RESULTS AND DISCUSSION

A. TRANSMITTANCE AND REFLECTANCE OF THE POLYIMIDE FOILS

The experiments on polyimide foils offer the possibility to compare optical constants obtained by reflectance versus angle measurements with those derived from transmittance measurements on the same sample.

Our reflectance measurements usually cover more than three decades on the ordinate; an example is given in Fig. 2. The experimental data obtained for a photon energy of 236 eV are very well reproduced by the fitted curve. The necessity of including different roughnesses for the two foil surfaces can be expected from the manufacturing process and, in fact, we get significantly different values from the fit procedure. They are about 1.0 and 2.0 nm for the upper and lower surface. Via the pronounced interferences the thickness of the foils is obtained. The value for this specimen is 291 nm. The remaining discrepancies in the fit are plotted in an expanded scale. The most significant differences occur in the vicinity of the critical angle at about 85°. They arise from the very small absorption of the polyimide combined with the fact that our present data evaluation procedure does not include a convolution with the divergence of the incident radiation.

The transmittance measurements were performed for photon energies between 50 and 1500 eV. With the film thickness as determined by the reflectance measurements and neglecting the very small normal-incidence reflectance, the absorption coefficient is easily obtained. For the Kramers-Kronig transformation, we extrapolate our data with values from Arakawa et al. [8] that were obtained from a different polyimide (Kapton). In the overlapping region between 50 and 65 eV, there is a systematic difference of about 10 %. Therefore the transition to the values of Ref. 8 at 65 eV leads to both the step in Im ε and the peak in Re ε in the spectra presented in Fig. 3. The high-energy extrapolation assumes a $(h\nu)^{-3}$ scaling of the absorption coefficient.

The results for Re ε show a systematic difference between the values obtained by reflectance measurements and those from the Kramers-Kronig analysis. The deviation amounts to 10 % at the lower photon energies; it decreases with increasing energy and almost vanishes above 300 eV. The differences may be explained under the assumption that the absorption coefficients of Kapton exceed those of our PIQ polyimide over the whole energy range from 3 to 65 eV. By the nature of the KK analysis, the effect of this error decreases with increasing energy. However, we want to prove this assumption before including it in our evaluation.

The two sets of values for Im ε agree very well between 70 and 170 eV. The increasing discrepancy between 200 eV and the carbon 1s absorption edge may result from inappropriate evaluation of the reflectance data due to the very low absorption as explained before. The degree of agreement is well again at 330 and 400 eV but decreasing in the vicinity of the oxygen 1s edge. Our analysis of the reflectance measurements at even higher energies shows that the truncation of our measurements above angles of 87° due to the finite size of our samples results in unreliable values of Im ε. We therefore exclude them from the presentation here.

B. REFLECTANCE AND YIELD OF CU AND PT FILMS

As we have shown in the case of thin Al films, the dielectric function and the thickness of a conducting film deposited on an insulator can be obtained by means of total yield versus θ measurements [9]. These data compared favorably with results determined by reflectance measurements on the same sample. Since the optical constants of the "white crown" glass substrate are required for the evaluation of film measurements, they were determined in the energy range 40 - 1000 eV from angular-dependent reflectance measurements [10]. For more details of the evaluation procedure of the yield method, the reader is referred to Ref. 9.

We present results of Cu and Pt films (Figs. 4 and 5). These investigations have been performed in order to see whether the yield method is also applicable to materials other than Al with the same quality of results. Figure 4 shows the dielectric function ε of our Cu films. There the closed symbols refer to the reflectance versus θ measurements and the open symbols to the yield versus θ measurements. The circle and the diamond symbols are chosen to distinguish between two different films of 22.5 ± 0.4 and 31.7 ± 0.6 nm thickness. For comparison with our results, values of ε calculated from atomic scattering factors from Henke et al. [11] are also included in the figure (dash-dotted line).

Two regions with inconsistencies appear in the ε spectra of our Pt films (Fig. 5). First, the pronounced absorption minimum in Im ε vs hν at about 120 eV, which is attributable to the delayed onset of the 4d - nf transitions, is about 40 % smaller than that in the data from Ref. 11. Photoabsorption cross-section data, which were compiled by Henke and co-workers to establish atomic scattering factors in this energy range, originate from thin-film absorption measurements [12]. These values can significantly depend on impurities within the sample and surface contaminants from the preparation process under non-UHVconditions. Further, in the energy region from 300 to 600 eV, the reflectance method results in Im ε values which systematically exceed the corresponding values of the yield method. An analogous systematical deviation was found in the 1 - Re ε spectra. In that region, we obtained fits of the yield versus θ spectra with a much higher quality than in the case of the reflectance spectra. Nonetheless, we do not want to prematurely decide which data set should be discarded before clarifying the origin of the discrepancies. Further studies are definitely necessary. In particular, the influence of surface roughness on the electron yield needs to be investigated, which has been ignored up to now. Furthermore, stray light investigations have to be performed in order to check whether the exponential factor is a proper treatment of the reflectance loss due to roughness. The microscopic structure of the films must also be checked.

The differences between the data sets of Pt appear to be rather unsatisfactory but they present the state of the art. Such unclarified problems have also been found with materials other than with Pt.

Further investigations along these lines could follow several paths. The experiments could be extended to other important materials in order to establish the quality of optical data. In cases where the data are well behaved as with polyimide films and Cu, we hope to be able to attach significant error bars to the ε values. In cases like Pt where discrepancies between the two methods occur in certain energy regions, we hope to obtain a clarification of their origins.

ACKNOWLEDGEMENTS

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FIGUR CAPTIONS

Fig. 1 side view of the reflectometer beamline at HASYLAB

Fig. 2 reflectance vs angle of incidence of 291 nm polyimide at 236 eV
circles experimental spectrum
line theoretical spectrum from fit
bars difference theory - experiment in units of reflectance/100 (right ordinate)

Fig. 3 optical constants of polyimide
a) decrement from unity of Re ε
b) Im ε
circles from reflectance vs angle of incidence measurements
line from Kramers-Kronig analysis including data from Arakawa et al. [8] (cf. text)

Fig. 4 optical constants of Cu
a) decrement from unity of Re ε
b) Im ε
closed symbols from reflectance measurements
open symbols from total yield measurements
circles film thickness 22.5 ± 0.4 nm
diamonds film thickness 31.7 ± 0.6 nm
-·-· calculated from atomic scattering factors from Henke et al. [11]

Fig. 5 optical constants of Pt
a) decrement from unity of Re ε
b) Im ε
closed symbols from reflectance measurements
open symbols from total yield measurements
circles film thickness 37.7 ± 0.6 nm
diamonds film thickness 30.8 ± 0.4 nm
-·-· calculated from atomic scattering factors from Henke et al. [11]

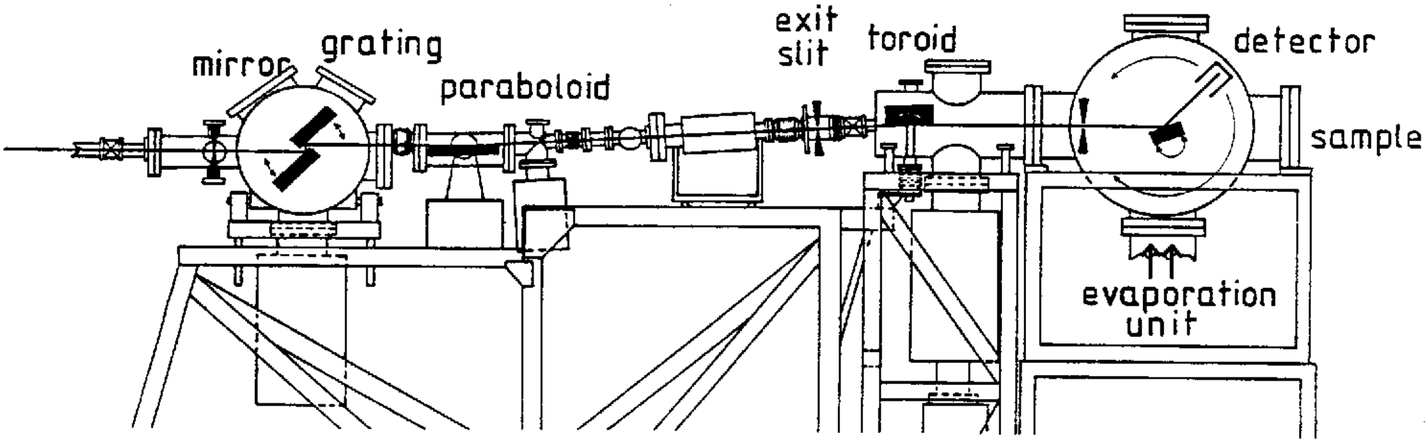
mirror
grating
paraboloid
exit slit
toroid
detector
sample
evaporation unit

Fig. 1

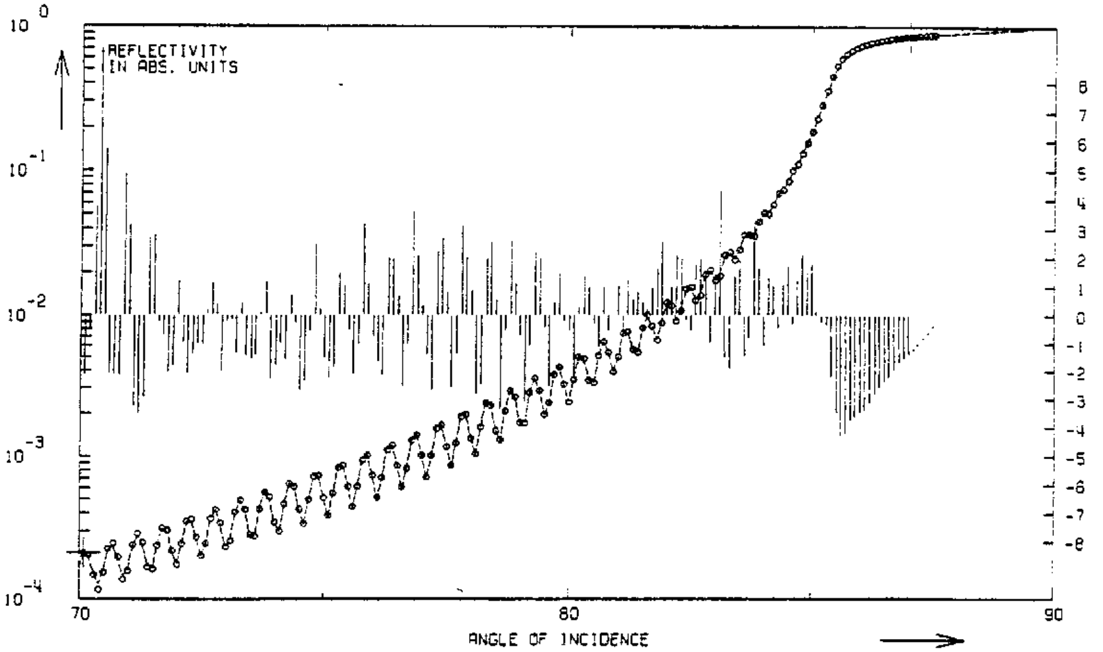
REFLECTIVITY
IN ABS. UNITS
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
8
7
6
5
4
3
2
1
0
-1
-2
-3
-4
-5
-6
-7
-8
70
80
90
ANGLE OF INCIDENCE

Fig. 2

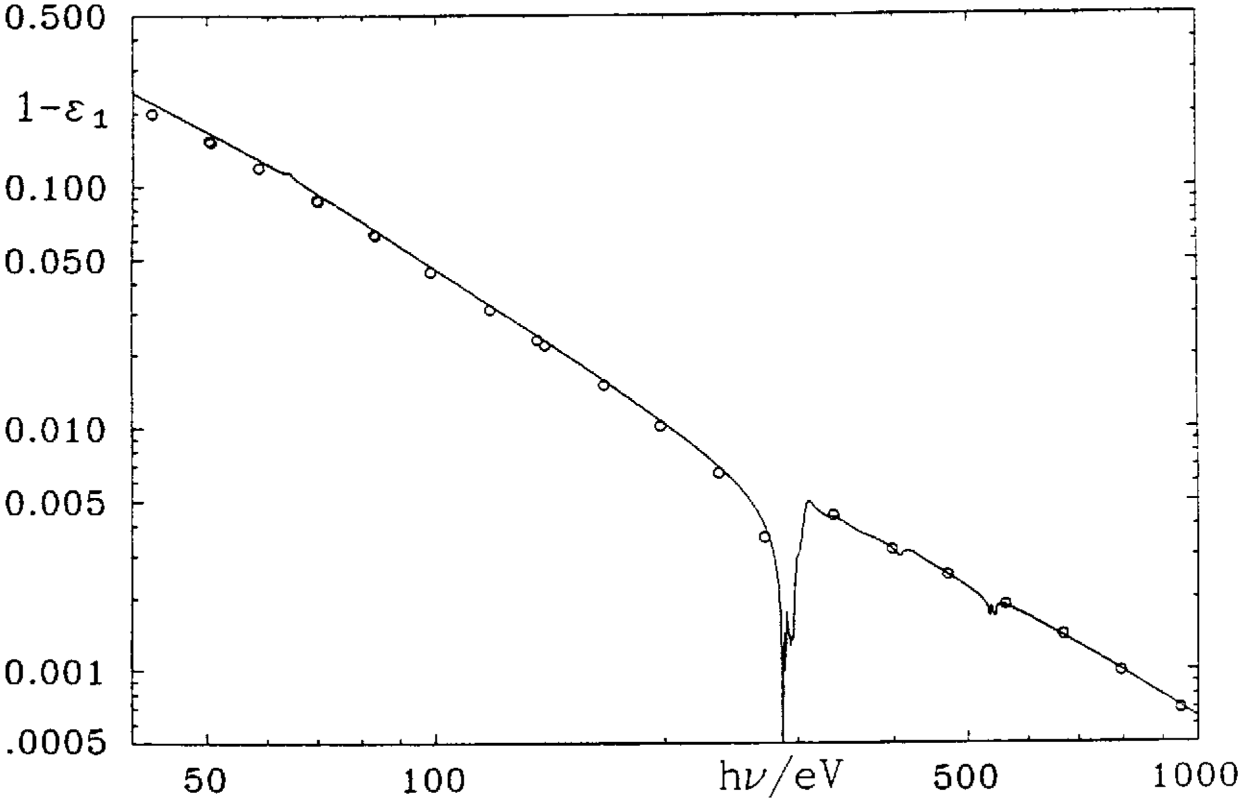
0.500
1-ε₁
0.100
0.050
0.010
0.005
0.001
.0005
50
100
hν/eV
500
1000

Fig. 3 a

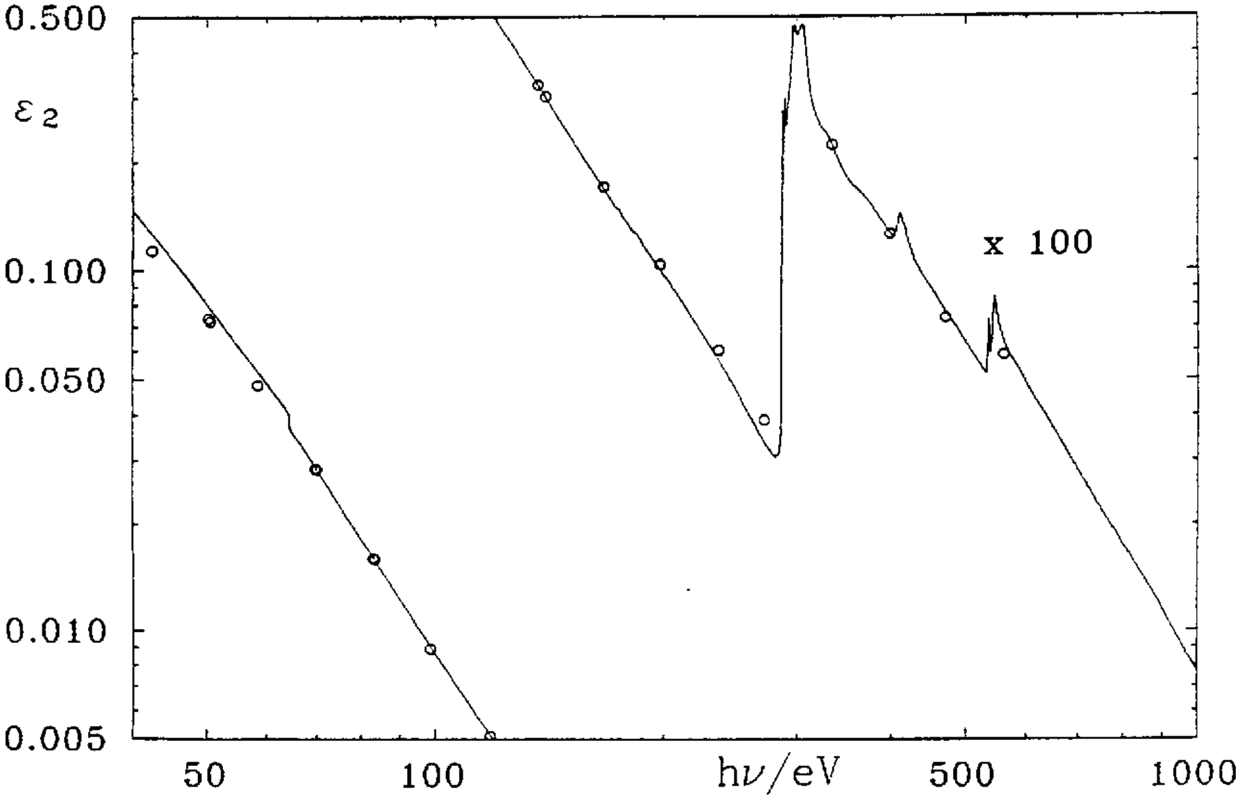
0.500
ε₂
0.100
0.050
0.010
0.005
x 100
50
100
hν/eV
500
1000

Fig. 3 b

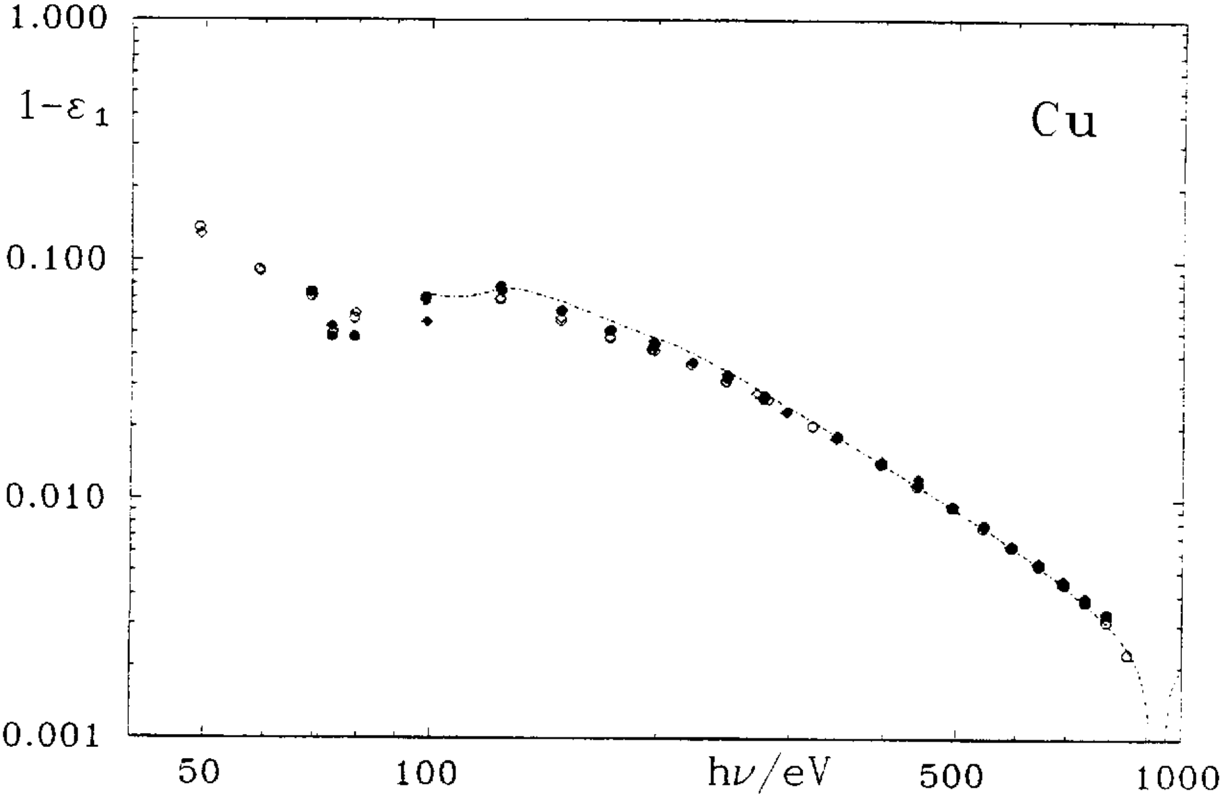
1.000
1-ε_1
Cu
0.100
0.010
0.001
50
100
$h\nu$/eV
500
1000

Fig. 4 a

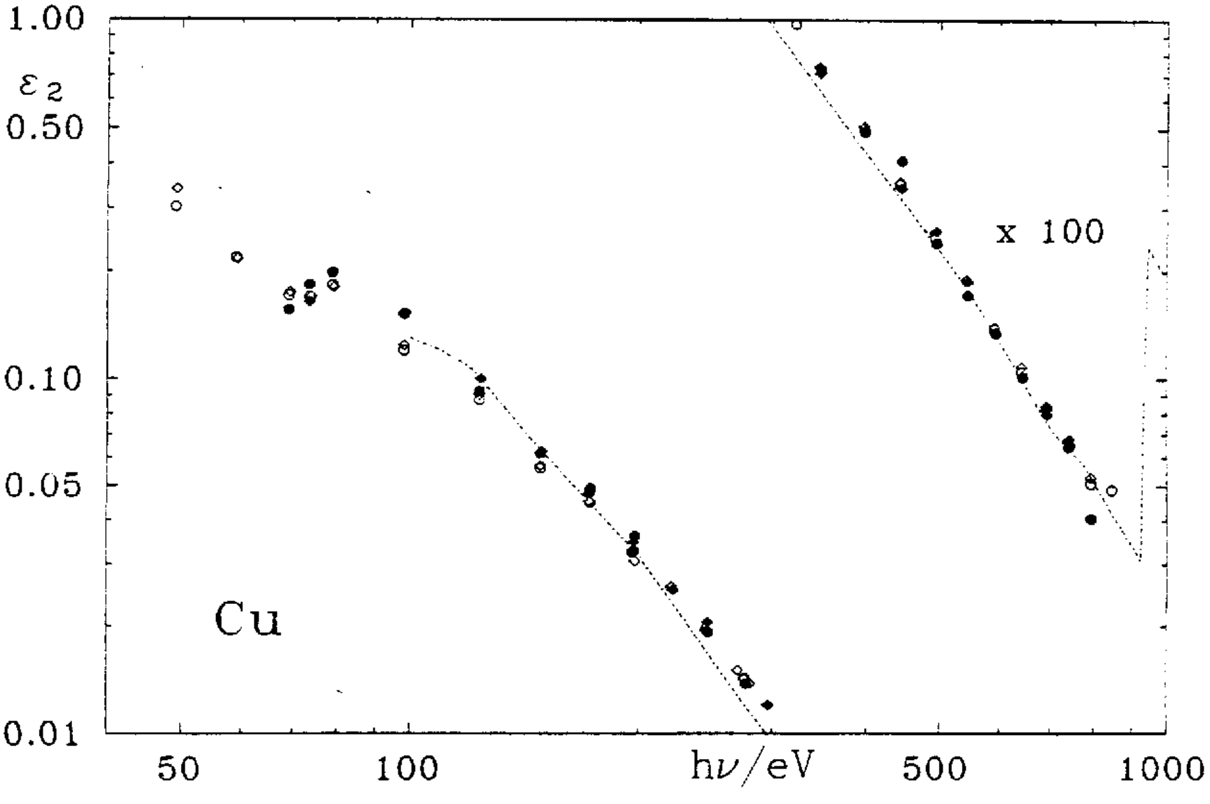
1.00
ε_2
0.50
x 100
0.10
0.05
Cu
0.01
50
100
$h\nu$/eV
500
1000

Fig. 4 b

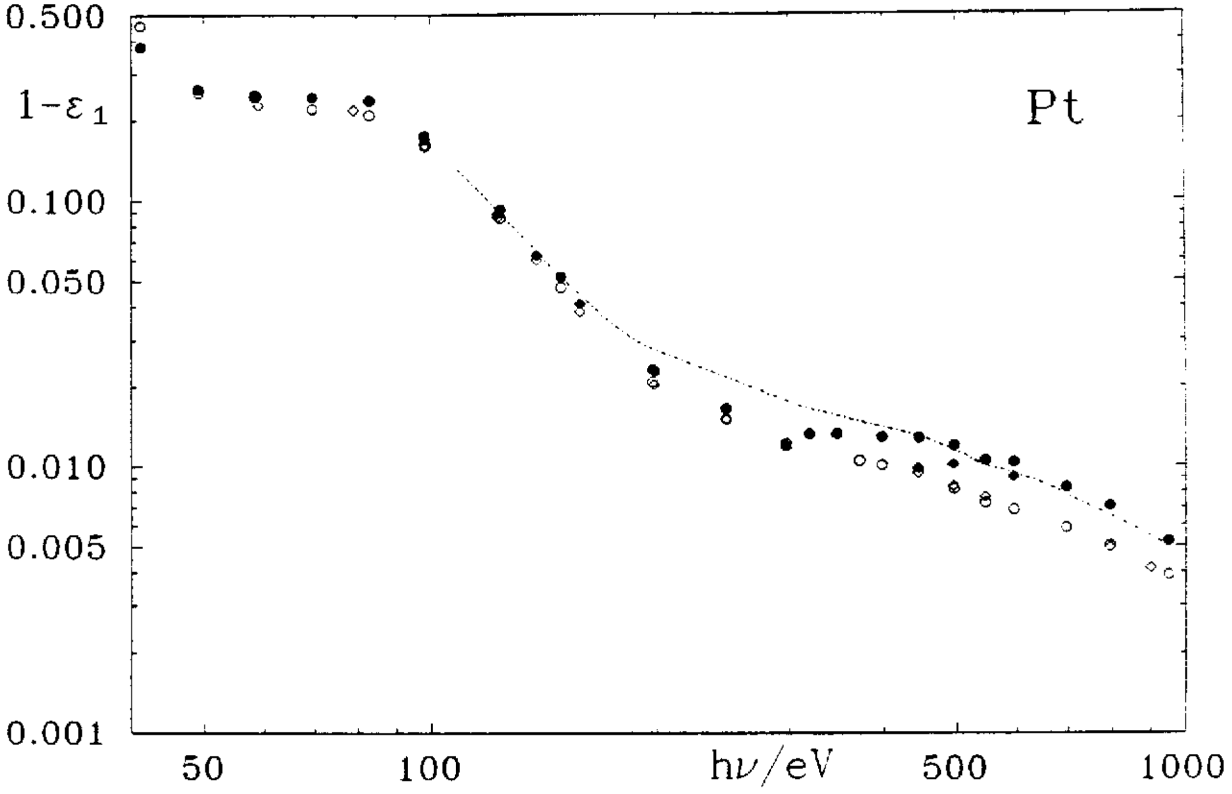
0.500
1-ε₁
0.100
0.050
0.010
0.005
0.001
Pt
50
100
hν/eV
500
1000

Fig. 5 a

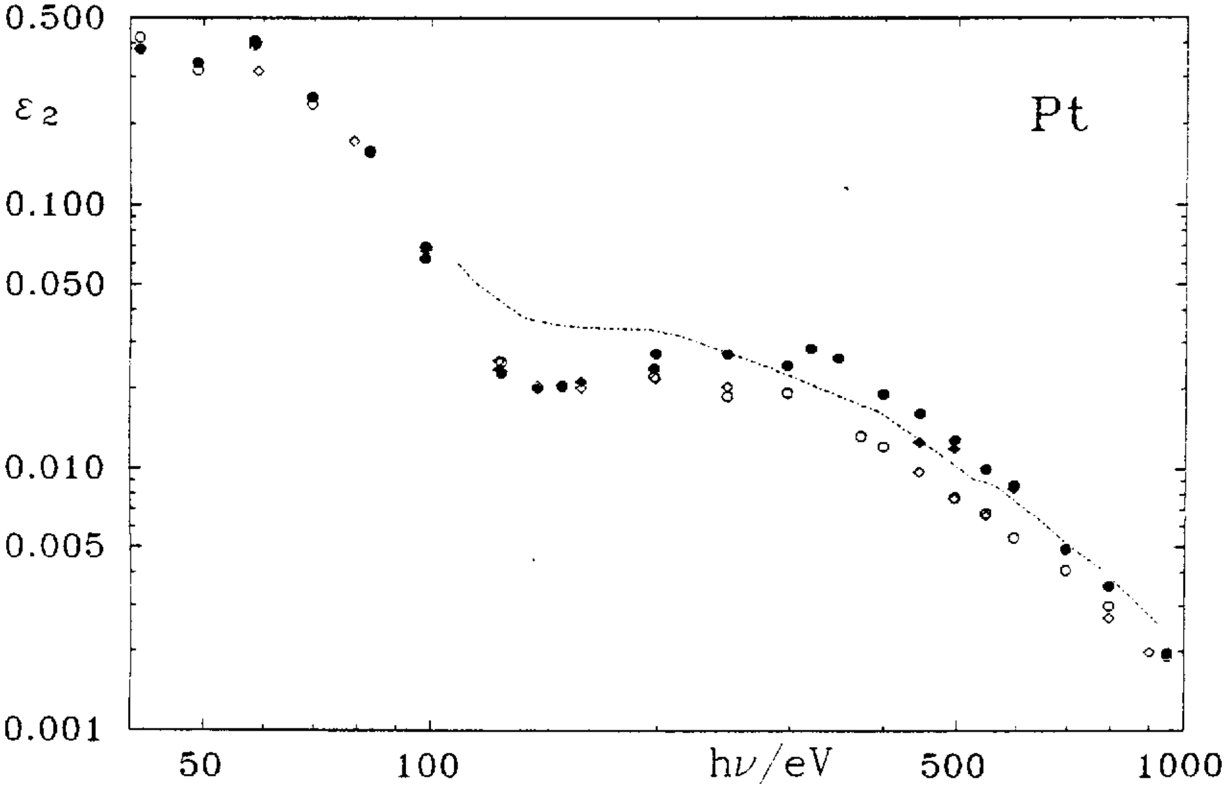
0.500
ε₂
0.100
0.050
0.010
0.005
0.001
Pt
50
100
hν/eV
500
1000

Fig. 5 b