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Multilayer Interference Mirrors for the XUV Range
Around 100 eV Photon Energy

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Multilayer Interference Mirrors for the XUV Range

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For the first time successful results demonstrating interference effects in the reflectance of multilayer films at high photon energies where n=1 and k is large were obtained. Films with 3.5 to 4.5 periods of Au/C and Cu/C were investigated for angles of incidence $\alpha=15^\circ$ to $\alpha=60^\circ$ in the wavelength interval 80 to 300 Å. The highest near normal incidence reflectance found were R=2.7 % for Au/C and R=1.2 % for Cu/C at $\lambda \gtrsim 190$ Å and $\alpha=15^\circ$. These values were by a factor of 7 higher than those for a single layered opaque Au film (respectively a factor of 6 for Cu) evaporated under the same conditions. Further improvements appear to be possible. Calculations were performed to guide the investigations and to help the interpretation of the results.

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1. Introduction

Multilayer coatings on optical components are widely used in the spectral region where nonabsorbing materials are available. Recently Spiller (1) proposed to extend the use of such coatings into the vacuum ultraviolet region by preparing interference coatings which consist of a sequence of alternating films of two materials with high and low absorption coefficients. The basic design idea is to place the strong absorber in thin layers at the nodes of the desired wavefield. Thus a fairly large penetration depth of the wavefield can be achieved with a participation of many layers in the reflection process. The goal is to boost up the notoriously weak reflectivity of mirrors at near normal incidence in the vacuum ultraviolet and at the same time to obtain wavelength selective reflectors. The effect used is conceptually equivalent to the Borrmann Effect observed in x-ray physcis.

In a series of calculations Spiller (1) has demonstrated that fairly large reflectivities could be expected. He was able to demonstrate agreement between calculations and experiments with a 10 layer coating of absorbing gold and transparent MgF_2 in the wavelength range above 3000 Å (2) and with several layers of Al and MgF_2 around 2000 Å (3).

No demonstration of this effect, however, has been given, up to now, with two absorbing materials in the spectral region of extreme vacuum ultraviolet where the production of such coatings would mean a great technical progress.

In this paper we shall show for the first time realization of interference mirrors which have maxima of reflection in the photon energy range 65 to 160 eV (190 Å to 90 Å in wavelength). The films were made of carbon as the material with low absorption and Au (respectively Cu) as the highly absorbing component. The fact that metals and carbon usually do not diffuse readily into each other is an essential point (4). The investigations were performed by means of a reflectometer which allowed for measurements in s-polarization with angles of incidence between 15 and 90 degrees. Monochromatized synchrotron light (5) in the energy range 40 to 230 eV was available from the DESY synchrotron. Calculations using a multilayer scheme are described in section 2. They were used to guide the preparation of the films and to interprete the results. The experimental procedure is described in section 3 and the results are presented in section 4 and discussed on the basis of model calculations in section 5.

2. Design calculations

Because of the special arrangement of the reflectometer used with respect to the direction of polarization of synchrotron radiation all measurements were made with s-polarization and all calculations refer only to this type of polarization.

The amplitude reflectance r of a thin film on a substrate is given by (6):

$$r = \frac{r_1 + r_2 e^{2i\, d_1 k_1}}{1 + r_1 r_2 e^{2i\, d_1 k_1}}$$

r_1, r_2 are the amplitude reflectance coefficients of the interfaces vacuum/film and film/substrate respectively. The film has a thickness d_1 and the normal component of the light vector in the film is denotes by k_1. This equation can easily be expanded for a two layer system with a second film of thickness d_2 and the normal component of the light vector k_2 by replacing r_2 by:

$$r_2 = \frac{r_2' + r_3 e^{2i\, d_2 k_2}}{1 + r_2' r_3 e^{2i\, d_2 k_2}}$$

where r_2', r_3 refer to the interfaces film 1 / film 2 or film 2 / substrate respectively. In this way the scheme can be generalized to as many layers as desired (6).

All calculations were performed according to this algebraic scheme on a digital desk computer.

For Au, Cu, and C the wavelength dependent optical constants of Hagemann et al. (7) were used while the value $\tilde{\epsilon} = 0.9 + 0.1\, i$ for glass was used throughout since we are not aware of any actual data in our wavelength region.

The calculations showed that it is advantageous to have an odd number of layers with highly absorbing ones on top and at the bottom. For a periodic structure of alternating, absorbing and transparent layers the ratio of the thickness of both films and the length of the period can easily be optimized. As quoted by Spiller (1) a nonperiodic structure will improve the performance. Therefore the thickness of each layer was varied in steps of 2 Å to obtain maximum reflectivity. This procedure resulted in an decreasing thickness of the absorbing layer from bottom to top. The gain of reflectance was small but we hoped to obtain thus a system which would not suffer too much from the thickness errors which occur on evaporating the films. Table 1 gives the parameters of two nonperiodic mirrors with gold and carbon optimized for $\alpha=15^{\circ}$ and $\lambda=200$ Å, 150 Å respectively and a periodic mirror with copper and carbon optimized for $\alpha=15^{\circ}$ and $\lambda=170$ Å.

3. Experimental procedure

As a substrate for the mirrors we used microscope object slides. Gold and copper were evaporated from a tungsten boat, carbon with an electron gun. The base pressure in the system was 5×10^{-7} Torr. During deposition of Au and Cu the pressure rose only little while for carbon it reached 1×10^{-5} Torr. The thickness was controlled by an oscillating quartz monitor (Balzers). The deposition of Au and Cu was initiated and stopped by a shutter. When evaporating carbon from an electron gun the quartz monitor drifted because of excessive heating. Therefore no shutter was used and the thickness was obtained by reading the monitor in its steady state before and after deposition of the film. Repeated evaporations were usually necessary in order to obtain the desired thickness. Table 1 gives an impression of the practicability of the method. Long term instabilities of the quartz monitor may introduce addtional errors. After preparation the mirrors were stored in atmosphere between 3 and 5 days before measurement.

The measurements were performed with synchrotron radiation monochromatized by a fixed exit slit grazing incidence monochromator [5] which covers the range from 20 to 280 eV. The reflectometer is installed behind the exit slit of the monochromator. It allows independent rotation of the sample and the detector around an axis normal to the plane of incidence. The angle of incidence can be varied from 15 degrees to grazing incidence. For measuring the intensity of light incident on the sample which serves as a reference the sample can be removed. A Johnston MM-1 photoelectron multiplier with a special cathode coated with KCl served as the detector.

4. Results

The reflectivity of the multilayer films is measured as a function of wavelength for different angles of incidence α. Figure 1 gives the result for the mirror Au/C with 4.5 periods and 106 Å average periodicity. At all angles of incidence pronounced interference structures show up with one dominating peak. Its position is determined by the condition that the nodes of the standing wave inside the film coincide with the Au layers. At normal incidence the wavelength λ_{max} of this peak is given by twice the optical period (which due to the difference of the index of refraction from one will differ from the geometrical periodicity). With increasing angle of incidence the peak shifts to shorter wavelengths. With all the samples the measured values of λ_{max} at $\alpha = 15^o$ differ at most by 10 % from those aimed at when producing the mirrors. At the long-wavelength side of the main peak a side peak shows up which obviously must originate also from an interference effect since it shifts along with the main peak.

Figure 2 gives the results for another mirror with a smaller periodicity (Au/C, 81 Å). The interference peaks come to lie at shorter wavelengths and shift with angle of incidence in the same systematic way. The side-peak is more pronounced comared to the sample Au/C 106 Å and shows a stronger increase with larger α. For $\alpha = 15^o$ one of the Au $N_{6,7}$ structures is superimposed onto the main peak.

In order to test also other combinations of materials a mirror was produced with 3.5 periods of Cu and C (see Table 1). Figure 3 shows the reflectivity spectrum for $\alpha = 30^o$. The result is compared with the reflectivity of a single opaque (thickness 1000 Å) Cu film at the same angle of incidence. In addition, a calculated reflectivity based on optical constants is plotted. Compared to the measured reflectivity of the opaque Cu film the gain of the interference-mirror is a factor of 7 at the peak. The results for other angles α are listed in Table 2. Figure 4 shows the corresponding result for the Au/C 106 Å film. The maximum gain with respect to a 1000 Å thick opaque Au film was 7. The values for other angles and the other film are given in Table 2. The gain in reflectivity is considerable for all films and angles.

Especially with Au the reflectivity as calculated from the optical constants deviate by a large factor from those measured. The optical constants of Au were obtained in this region from absorption data by means of a Kramers-Kronig analysis. The result was scaled at 300 Å to the reflectivity measurements of Canfield et al. (8). Thus for the shorter wavelength region these reflectivities are only an extrapolation. It is well known that measured reflectivities are notoriously low when compared with values of optical constants which originate from absorption measurements. Surface roughness is usually quoted to be one of the reasons. Thick Au films are supposed to be especially rough (8). On the other hand the alternative Au and C evaporations might also yield some roughness. We therefore felt that the comparison on the basis of the experimentally measured values is quite reasonable in order to obtain an idea of the gain in reflectivity from these interference mirrors.

5. Model calculations and discussion

Calculations of the performance of the interference mirrors were carried out using the method described in section 2 in order to obtain a feeling for the quality of the films obtained. The thicknesses of the films as obtained from the oscillating quartz monitor multiplied with a common factor f were used. The correction factor f was chosen so that the interference peak at $\alpha = 15^{o}$ coincides in the experimental curve and in the calculated curve, f is also given in Table 2.

In Figs. 1 and 2 the calculated curves are compared with the measured curves. The shape of the peaks and their shift to shorter wavelengths with increasing α are quite well reproduced by the calculation. At shorter wavelengths the position of the main peak is quite sensitive to the optical constants of the glass substrate since the absorption coefficients of Au and C are rapidly decreasing. As a consequence the transparency of the total film increases in spite of the increasing geometrical path length of the light. Model calculations in which the $\hat{\varepsilon}$ of glass was varied in the expected way showed that the positions of the peaks at large angles α then come to lie even closer to the experimental values.

The side-peak at the longer wavelengths shows up also in the calculated curves. There it is, however, less pronounced relative to the main peak. In a simple explanation this peak can be characterized as a wave which has nodes at the substrate and at the vacuum interface but inside the multilayer film it has one node less than the main peak. At present it is quite unclear why this peak is not as pronounced in the calculation as it is in the experiment.

The absolute reflectivities of the calculated curves is 5 to 10 times larger than measured (see Table 2). Important reasons for this difference could be undetected errors in the thicknesses of the individual films, roughness and partial diffusion at the carbon metal interfaces.

A more realistic way to estimate the possible gain in reflectance from interference films produced with an improved preparation technique which would avoid the just mentioned sources of errors is probably the following: Compare the theoretical gain in reflectivity from an opaque one layer film to a multi-layer film with the experimental gain from an opaque film to a multi-layer film. While according to Table 2 the theoretical gain is 10 - 30 the experimental gain is on the average only half as large. From this point of view it appears to be quite reasonable to expect at least factor of two improvement in the reflectivity of interference films prepared under better control than ours. In addition an increase in the number of layers should yield an even larger gain in absolute reflectivity. Such highly reflecting coatings could find useful applications in spectrographs and astrophysical instrumentation for increasing the efficiency and suppressing unwanted radiation. Further, the fairly large reflectivities at near normal incidence could open the possibility to set up a microscope with mirror elements in the wavelength range around 100 Å or below. Also applications of these mirrors for the attempt to build lasers in the soft x-ray region can be envisaged. While the possible reflectivities are still low compared to ordinary laser mirrors, a resonator equipped with interference mirrors could well serve to define the direction of lasing.

Acknowledgement

We wish to thank E. Spiller for fruitful discussions.

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Table 1: Sequences of interference layers (in Å units) as envisaged and as obtained from reading a quartz monitor. The Au/C mirrors were optimized in reflectivity by making them slightly aperiodic.

		glass	Au	C	Au	C	Au	C	Au	C	Au
Au/C 106 Å	envisaged	∞	50	64	42	70	42	72	40	72	36
	achieved	∞	50	64	42	69	42	72	40	71	36
Au/C 81 Å	envisaged	∞	39	47	35	47	35	47	35	47	33
	achieved	∞	39	47	35	49	35	46	35	47	33

		glass	Cu	C	Cu	C	Cu	C	Cu
Cu/C 103 Å	envisaged	∞	35	65	35	65	35	65	35
	achieved	∞	37	60	32	70	27	60	32

Table 2: Peak values of the reflectivity and wavelength position of interference maxima of multi-layer films and reflectivities of opaque single metal films at the same wavelengths. The calculations are based on the optical constants of Ref. 7. α = angle of incidence, λ_{max} = wavelength (in Å units) of the interference peak; f = common correction factor for measured film thickness (see text); R_I^{ex}, R_I^{th} measured resp. calculated reflectivity of multi-layer mirrors at peak wavelength; R_M^{ex}, R_M^{th} measured resp. calculated reflectivities of an opaque metal film. All reflectivities in %.

	α	λ_{max}	f	R_I^{ex}	R_I^{th}	R_I^{th}/R_I^{ex}	R_M^{ex}	R_M^{th}	R_I^{th}/R_M^{th}	R_I^{ex}/R_M^{ex}
Au/C 106 Å	15	186	.95	2.7	12.5	5	0.4	1.3	10	7
	30	167		3.1	15.0	5	0.4	1.4	11	8
	45	134		4.8	23.3	5	0.6	1.8	13	8
	60	96		4.8	32.2	7	0.5	1.3	25	9
Au/C 81 Å	15	146	.99	1.2	11.3	9	0.1	0.6	19	12
	30	133		1.5	15.0	10	0.1	0.7	21	15
	45	109		1.5	16.5	11	0.1	0.6	28	15
	60	78		0.6	11.6	19	0.1	0.3	39	6
Cu/C 103 Å	15	188	1.12	1.2	3.2	2.7	0.2	0.3	11	6
	30	167		1.3	4.0	3.1	0.2	0.4	10	7
	45	139		2.0	7.2	3.6	0.3	0.9	6	7
	60	97		4.6	11.0	2.4	0.5	1.7	7	9

Figure Captions

Fig. 1 Reflectivity R of a multi-layer interference mirror with 4.5 periods of Au/C on glass as a function of photon wavelength λ. The average periodicity is 106 Å. The angle of incidence α is varied in steps from 15^o to 60^o. Solid line: experimental values, dashed line: theoretical values based on optical constants of Ref. 7 (for the glass substrate $\tilde{\varepsilon} = 0.9 + 0.1\,i$ was used independent of wavelength). The theoretical curve is reduced by a factor of 0.2.

Fig. 2 Reflectivity R of a multi-layer interference mirror with 4.5 periods of Au/C and 81 Å average periodicity (see Fig. 1)

Fig. 3 Comparison of the measured reflectivity R of an interference mirror (solid line) with 3.5 periods of Cu/C at $\alpha = 30^o$ with the reflectivity of a single opaque (1000 Å) Cu film (dashed line) as a function of wavelength λ. The interference film has an average periodicity of 103 Å. The dashed-dotted line gives the theoretical reflectivity of Cu based on optical constants of Ref. 7.

Fig. 4 Comparison of the measured reflectivity R of the Au/C 106 Å, 4.5 periods interference film at $\alpha = 15^o$ with the measured and calculated reflectivities of an opaque Au film (designations see Fig. 3).

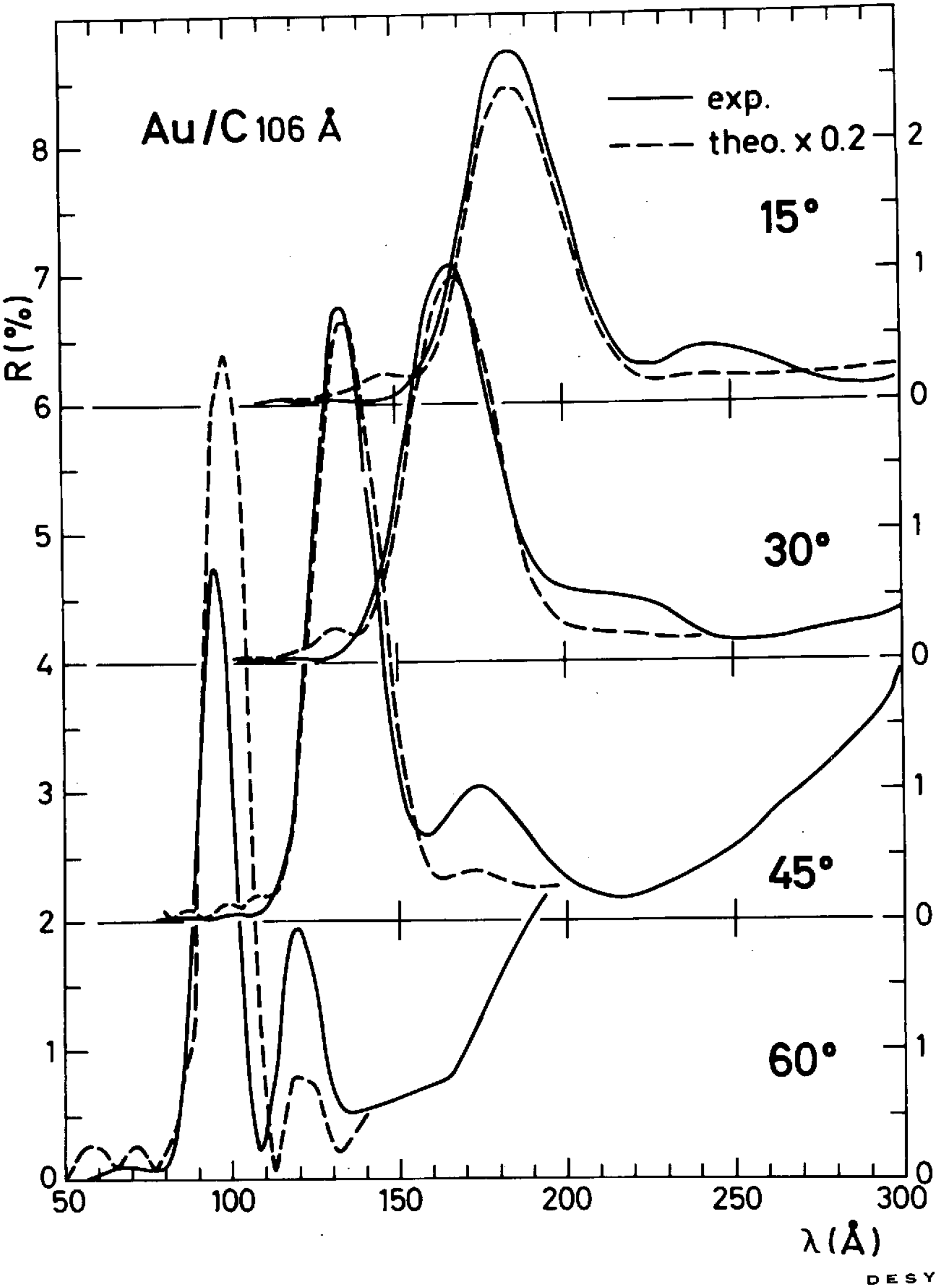

Fig. 1

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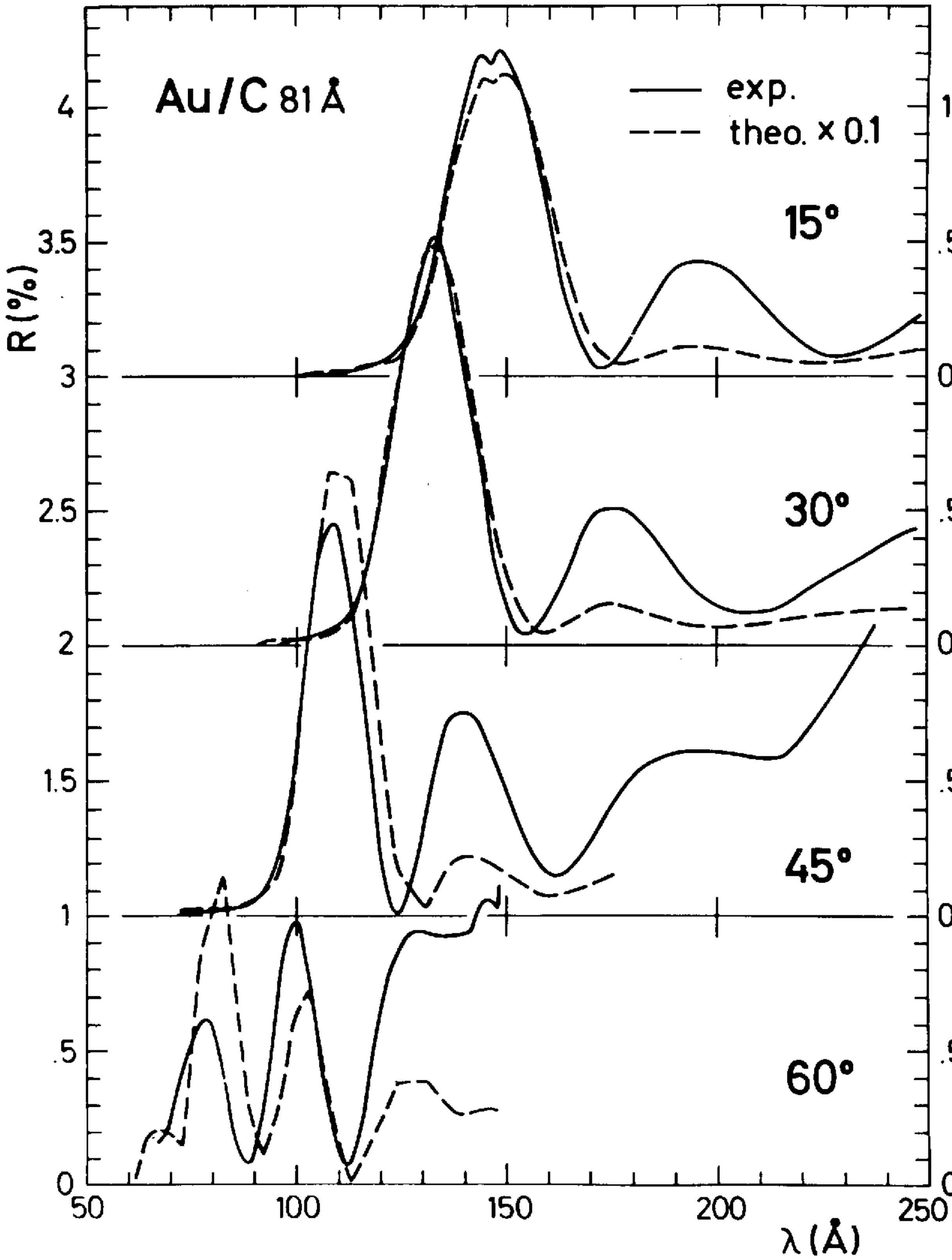

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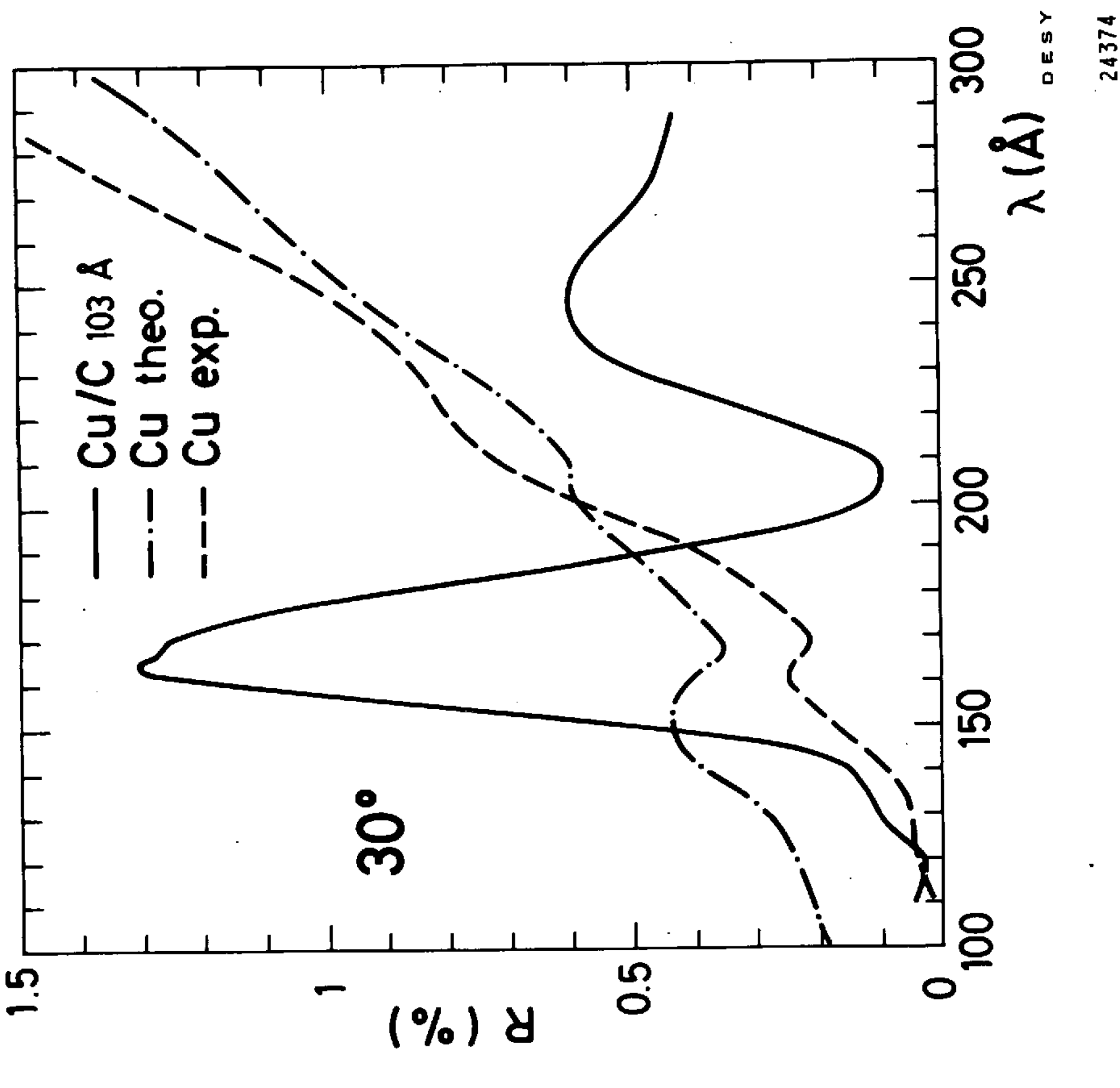
Fig. 2



Cu/C 103 Å
Cu theo.
Cu exp.
30°
R (%)
λ (Å)
0
0.5
1
1.5
100
150
200
250
300
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24374

Fig. 3

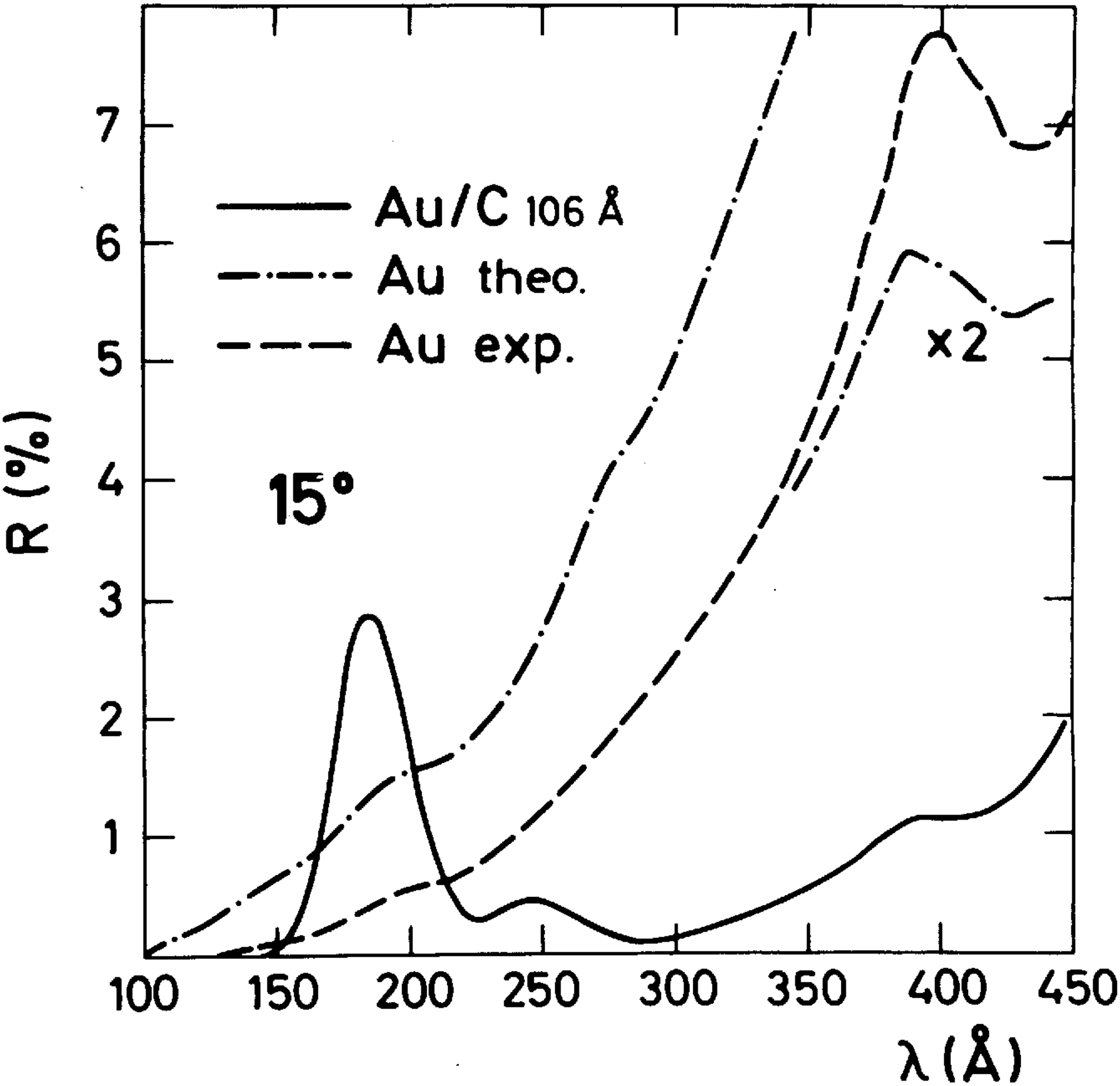

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Fig. 4