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A. Polarization Dependent Transmission and Reflection
B. Polarization Dependent Photoeffect

by

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Investigations of Aluminum Films with Synchrotron Radiation of Wavelengths 500 to 1000 Å

A. Polarization Dependent Transmission and Reflection*

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ABSTRACT

Measurements of the transmittance and reflectance of Al films as a function of wavelength, polarization, angle of incidence, and film thickness have been carried out using synchrotron radiation together with a normal-incidence monochromator for the vacuum ultraviolet spectral region. A pronounced difference in the measurements for "s" and "p" polarized light was found near the Al plasma wavelength (λ_p = 835 Å). We measured a dip in thin film transmittance for "p" light at λ_p which is due to the excitation of plasmons. The "p" reflectance exhibits a Brewster minimum below λ_p. For "s" light there is no structure for very thin films. With films thicker than about 500 Å interference effects appear, especially in reflection, for both "p" and "s" light at wavelengths less than λ_p where Al becomes transparent. The transmittance and reflectance of Al films of various thicknesses, both with and without substrate materials and oxide layers, have been computed assuming optical constants based upon previous experiments. The calculations explain the structure exhibited in the experimental curves. Our measurements show that the vacuum ultraviolet synchrotron radiation is highly polarized even after passing through our monochromator.

I. Introduction

The interaction between the electromagnetic field and an electron gas can be described by a complex dielectric constant.[1] At the plasma frequency where the dielectric constant vanishes we expect a singular behavior giving rise to special effects. First discrete plasmon peaks in electron energy loss spectra [2] and associated radiation from thin films [3-5] were observed. The presence of a maximum at the plasma frequency for reflection of "p" light (electric vector parallel to the plane of incidence) due to the radiative decay of optically excited plasmons in thin metal films was later predicted by Ferrell and Stern [6]. This prediction was soon expanded to include structure in transmission and absorption [7-10]. The predicted dip in transmittance at the plasma wavelength was first found experimentally for Ag [7,10]. More recently transmission experiments have been performed which exhibit plasma resonance absorption in Al[11] and K[12]. In addition a peak in scattered "p" light at λ_p has been observed in Ag[13,14] and K[15].

In the present work we have studied Al which is particularly interesting since it behaves, to a good approximation, like a free-electron metal with little damping in the neighborhood of the plasma frequency.[16,17] Synchrotron radiation with its unique continuum and polarization properties is particularly suited for the study of Al near its plasma wavelength λ_p (835 Å), since λ_p lies in the technically troublesome extreme vacuum ultraviolet below the LiF transmission limit of 1050 Å.

In section II we discuss the predictions of electrodynamics for ideal thin films. The formulation is used in section IV to explain our measurements and in the following paper (B. Polarization Dependent Photoeffect). Section III presents a description of our experimental apparatus and some comments on using synchrotron radiation. Section IV contains the measured and calculated transmittance and reflectance of thin Al films.

II. Theory

Consider an ideal thin film of known complex scalar dielectric constant, $\tilde{\varepsilon}(\omega) = \varepsilon_1(\omega) - i\,\varepsilon_2(\omega)$, and thickness d.[18] The complex transmission and reflection coefficients, $\tilde{t}$ and $\tilde{r}$ respectively, may be obtained by application of Maxwell's equations. The film transmittance T and reflectance R are given by $|\tilde{t}|^2$ and $|\tilde{r}|^2$. For "p" light incident upon a film we get:

$$\tilde{t}_p = \frac{4\tilde{a}\tilde{b}\,e^{-\rho d}}{(\tilde{a}+\tilde{b})^2-(\tilde{a}-\tilde{b})^2 e^{-2\tilde{\rho} d}} \tag{1}$$

and

$$\tilde{r}_p = \frac{(\tilde{a}^2-\tilde{b}^2)(1-e^{-2\tilde{\rho} d})}{(\tilde{a}+\tilde{b})^2-(\tilde{a}-\tilde{b})^2 e^{-2\tilde{\rho} d}} \tag{2}$$

where $\tilde{a} = \tilde{\varepsilon}\cdot\cos\phi$, $\tilde{b} = (\tilde{\varepsilon} - \sin^2\phi)^{1/2}$, $\tilde{\rho} = i\omega\,\tilde{b}/c$, ω is the frequency and c the velocity of light, and ϕ is the angle of incidence. It is difficult to see immediately from these equations that they exhibit structure near the plasma frequency ω_p (or plasma wavelength $\lambda_p = 2\pi c/\omega_p$) which may be defined as that frequency where the real part ε_1 of the dielectric constant is zero with the imaginary part $\varepsilon_2 << 1$. In the neighborhood of the plasma frequency for angles of incidence which are "not too small", i. e.

$$|\tilde{\varepsilon}| \ll \sin^2\phi \quad \text{so that} \quad \tilde{b} \approx i\sin\phi \tag{3}$$

we have

$$\tilde{t}_p = \frac{2\tilde{\varepsilon}\cosh^{-1}(\beta\sin\phi)}{2\tilde{\varepsilon} - i\tan\phi\tanh(\beta\sin\phi)} \tag{4}$$

and

$$\tilde{r}_p = \frac{+i\tan\phi\tanh(\beta\sin\phi)}{2\tilde{\varepsilon} - i\tan\phi\tanh(\beta\sin\phi)} \tag{5}$$

where $\beta = \omega d/c$. A free electron model yields in the neighborhood of ω_p:

$$\tilde{\varepsilon}(\omega) = \left(1 - \frac{\omega_p^2}{\omega^2}\right) - i\varepsilon_2 \approx 2(\omega-\omega_p)/\omega_p - i\varepsilon_2 \qquad (6)$$

with ε_2 approximately constant. The transmittance and reflectance become in this model

$$T_p \approx \frac{\left[4(\omega-\omega_p)^2 + \omega_p^2\varepsilon_2^2\right]\left[\cosh(\beta\sin\phi)\right]^{-2}}{4(\omega-\omega_p)^2 + \frac{1}{4}\omega_p^2\left[\tan\phi\,\tanh(\beta\sin\phi) + 2\varepsilon_2\right]^2} \qquad (7)$$

and

$$R_p \approx \frac{\frac{1}{4}\omega_p^2\left[\tan\phi\,\tanh(\beta\sin\phi)\right]^2}{4(\omega-\omega_p)^2 + \frac{1}{4}\omega_p^2\left[\tan\phi\,\tanh(\beta\sin\phi) + 2\varepsilon_2\right]^2} \qquad (8)$$

For films which are "not too thick", i. e. $\beta \sin\phi \ll 1$, Eqs. (7) and (8) exhibit a Lorentzian minimum and a maximum respectively at the plasma frequency as has been pointed out.[6-10] The structure is a manifestation of the excitation of plasma oscillations by photons. It should be emphasized that the validity of formulas (4) - (8) is restricted by condition (3). If we put, for example $\phi = 30^\circ$, $|\tilde{\varepsilon}|$ for Al remains less than $\sin^2 30^\circ$ for wavelengths between 730 Å and 930 Å so that the formulas (4) - (8) are valid in a wavelength region narrower than 200 Å wide. For angles much smaller than 30° approximation (3) is invalid for Al.

The structure of R_p obtained from Eq. (2) without approximations deviates from the ideal resonance form of (8). For small angles a pronounced minimum which is not included in (8) occurs just below λ_p. It is the Brewster minimum for reflection of "p" light. Consider the numerator of (2). For films of arbitrary thickness a minimum in R_p will occur when the first factor of the numerator is a minimum. If

$$\left(\tilde{\varepsilon}\cos\phi\right)^2 - \left(\tilde{\varepsilon} - \sin^2\phi\right) = 0 \qquad (9)$$

R_p would be zero. With $\varepsilon_2 = 0$ Eq. (9) yields

$$\sqrt{\varepsilon_1} = n = \tan\phi \qquad (10)$$

which is the familiar Brewster condition. In the free-electron model without absorption the wavelength of the "Brewster minimum" λ_B is given by

$$\lambda_B = \lambda_p\left(1 - \tan^2\phi\right)^{1/2} \qquad (11)$$

This equation is valid for $\phi \leq 45^\circ$. The Brewster minimum is shifted to lower wavelengths with increasing angle of incidence.

The plasma resonance maximum in R_p for thin films is thus accompanied by a Brewster minimum below λ_p (see Fig. 3). The transmittance and reflectance of thin films for "s" light (electric vector perpendicular to the plane of incidence) show no structure near λ_p. There is neither a plasma excitation nor a Brewster minimum.

We digress here to point out the unusual properties of a metal near the plasma frequency. The condition for total reflection from bulk material (with $\varepsilon_2 = 0$)

$$\sqrt{\varepsilon_1} = n = \sin\phi \tag{12}$$

gives in a free-electron model total reflection for

$$\lambda \geq \lambda_T = \lambda_p \cos\phi \tag{13}$$

while for $\lambda < \lambda_T$ the electron gas becomes transparent as shown long ago by Wood's experiments [19]. From Eqs. (11) and (13) we see that $\lambda_B \leq \lambda_T \leq \lambda_p$ and that for small angles of incidence λ_B and λ_T are nearly the same.

Interference effects can produce structure in the transmittance and reflectance of thin films as a function of wavelength, in addition to that resulting from plasma resonances, the Brewster minimum, and the transition from total reflection to transparency. Interference maxima of m^{th} order in the reflectivity of films without absorption occur at wavelengths λ_m given by

$$\sqrt{\varepsilon_1}\, d \cos\phi' = \left(m + \frac{1}{2}\right)\lambda_m \; ; \quad m = 1, 2, \ldots\ldots \tag{14}$$

where ϕ' is the angle of refraction in the medium. Equation (14) is well known as the condition for constructive two-beam interference and follows directly from the exponential term of (2). Equation (14), together with Snell's law and the ε_1 of a free-electron metal predicts interference maxima (minima) in the reflectance (transmittance) at wavelengths λ_m ($\lambda_m < \lambda_p$ where the metal becomes transparent) given by:

$$\lambda_m = 2d\cos\phi\left[(2d/\lambda_p)^2 + \left(m+\tfrac{1}{2}\right)^2\right]^{-1/2} \qquad (15)$$

Until now we have discussed only the ideal case of the optical behavior of a free-electron gas near its plasma frequency. Film substrates, surface oxide layers, and impurities will cause the transmittance and reflectance of a real metallic film to be different from those predicted above. We would expect the absolute values to be modified much more than the spectral dependence if there is no marked structure in the optical constants of the added layers near the Al plasma frequency. That this is at least approximately true has been shown by computer calculations of the transmittance and reflectance of films on substrates and with surface layers. For this purpose a computer program was developed to calculate the optical properties of an arbitrary multilayer [20] when the optical constants and thickness of the different layers are known.

In the calculations the two parameter Drude model was used for Al. The plasma wavelength and damping constant were assumed to be 835 Å and 1.35×10^{15} sec^{-1} respectively. This model gives good agreement with the measured optical constants of Ditchburn and Freeman [21] in the neighborhood of λ_p. We have used Freeman's Al_2O_3 optical constants [22] and for our microscope glass substrates we have used $n = 1.17$ and $k = 0.61$ as average values in the region of 600 to 1000 Å [23].

III. Apparatus

a. Monochromator

Since Tomboulian and Hartman [24] investigated synchrotron radiation and found agreement with the theory of Schwinger [25] the advantages of using synchrotron radiation as a source for spectroscopy in the vacuum ultraviolet (vuv) have been recognized. Even though the theoretical treatment of synchrotron radiation already appears in textbooks the experimental study and utilization of synchrotron radiation has been limited.[26,27] The unique polarization properties of synchrotron radiation in the vuv have not been exploited at all.

In the present work a grating monochromator with near-normal incidence has been utilized in conjunction with the synchrotron radiation from the 6 GeV Deutsches Elektronen-Synchrotron (DESY). The monochromator has been described by Skibowski and Steinmann [28,29]. An eccentric mounting allows the grating to be rotated in order to vary the wavelength incident on the fixed exit slit. The monochromator yields a broad spectrum with a maximum near 700 Å. Measurements of gas absorption edges have shown the wavelength calibration to be accurate and reproducible to ±1 Å. The resolution has not been accurately checked in the vuv but is better than 3 Å at 800 Å. This resolution is adequate for the experiments described here. It is limited by the width, 0.5 mm, of the exit slit used. With a smaller slit it appears that the inaccurately known size and motion of the electron beam in the accelerator may limit the resolution. Since our gold coated Bausch & Lomb grating (blazed for 600 Å) reflects light with reasonable intensity only above 400 Å, we have only a first order spectrum from 400 to 800 Å. The second order peak at 1400 Å of our maximum near 700 Å has approximately 10% of the first order intensity. (Technical note: Our Au grating has darkled both in the bulk and on the surface from exposure to the x-ray component of the synchrotron radiation, but no noticeable degradation in monochromator performance has resulted.)

Calculations using the theory of synchrotron radiation, our experimental geometry, and DESY operating conditions of 10 mA at 6 GeV indicate that we have $6\text{x}10^{10}$ photons $\sec^{-1}\text{Å}^{-1}$ incident on our grating at a wavelength of 700 Å. The calculated degree of polarization $(I_p - I_s)/(I_p + I_s)$ of the light incident on our grating is shown in Fig. 1. I_p and I_s are the intensities of the "p" and "s" components of the light. The light is polarized with the electric vector in the synchrotron plane. Since our grating is used at near-normal incidence (for wavelengths under 1500 Å the angle of incidence is always less than 10^o), we assume that the degree of polarization is nearly unchanged by our monochromator. The measurements reported in this and the following paper show that the light coming from our monochromator is, indeed, highly polarized.

The monochromator grating chamber is directly coupled to the synchrotron vacuum system and has a pressure of about 10^{-7} torr.

b. Reflectometer

A reflectometer located behind the monochromator exit slit allows measurement of film transmittance or reflectance as a function of angle of incidence and polarization without opening the vacuum system. In transmission measurements the angle of incidence can be varied from 0° in steps of 7.5°, while in reflection it can be increased from 15° in steps of 7.5°. The upper angle attainable depends upon the geometry of the sample being investigated, but is typically about 45° in transmission measurements and 75° in reflection measurements. With the plane of incidence of the sample in the synchrotron plane we have almost pure "p" polarized light. By a 90° rotation of the sample and detector about the axis of the incident light we obtain "s" light. The reflectometer chamber can be isolated from the monochromator vacuum system for sample changes. It reaches a pressure on the order of 10^{-6} torr.

We have used an open electron multiplier (Bendix M 306 with W cathode) as detector. This multiplier is insensitive to stray light of the visible and near uv. A motor drive moves our grating to scan a wavelength spectrum. (Health physics dictates that experiments must be remotely controlled.) A potential proportional to the instantaneous wavelength is applied to the X axis of an X-Y plotter and the wavelength is simultaneously displayed on a digital voltmeter. The signal of our detector is divided by a reference signal and displayed on the plotter Y axis. The reference signal which is proportional to the instantaneous electron current of the synchrotron is necessary because the accelerator current varies erratically and is rarely stable to better than 5%.

(This behavior is not troublesome in elementary-particle physics experiments, so no determined efforts towards current stability seem likely. Storage rings should be free of this difficulty.) The reference signal is provided by a photomultiplier sensitive to the visible portion of synchrotron radiation.[30]

c. Film Preparation

The Al films were evaporated from W filmaments in a vacuum of 10^{-5} to 10^{-6} torr at a deposition rate of about 30 Å/sec. Film thicknesses were monitored during deposition with a quartz crystal oscillator and later measured with a Tolansky interferometer. The errors in quoted thicknesses are less than ±40 Å. In order to prepare unbacked films, Al was evaporated onto $(NaPO_3)_6$ covered glass. The Al films were then carefully floated free with water and caught on fine Cu grids. This technique yielded nearly hole free films with thicknesses as low as 200 Å.

IV. Results

a. Transmission

Equation (7) indicates there should be a plasma resonance minimum in the transmittance of "p" light at the plasma wavelength. Measurements of the transmittance of an unbacked 280 Å thick Al foil are shown in Fig. 2. Our results obtained with synchrotron radiation confirm those reported earlier by Ejiri and Sasaki [11]. They used a line source and the partial polarization produced by a Seya-Namioka monochromator. We have calculated the transmittance of a 250 Å thick Al foil with 40 Å of oxide on each surface and found a curve which nearly reproduced (except for a minor scale change) the measured curves of Fig. 2. The fit obtained with our assumed Drude parameters for Al is good. With thicker films the plasma minimum becomes less pronounced. It is still detectable with films as thick as 500 Å. With a 100 Å thick Al film deposited upon a Zapon backing we have measured an angle and polarization dependence of the transmittance similar to that measured with the unbacked 280 Å foil.

The results shown in Fig. 2 were obtained by the procedure described in IIIb. They are not corrected for the fact that the monochromator radiation is not totally polarized, but the high degree of polarization is clearly demonstrated. Estimations based upon our measurements show that the polarization of the radiation at the exit slit is certainly near the calculated polarization of the synchrotron radiation (Fig. 1). Direct measurements of polarization at the exit slit are planned.

In both "s" and "p" light we observe interference effects (similar to those reported earlier [21,31]) for films thicker than about 500 Å at wavelengths less than λ_p. The position and the separation of maxima and minima are predicted approximately by Eq. (15). They are naturally the same for "s" and "p" light. With increasing angle of incidence the maxima are shifted to smaller wavelength and the oscillatory interference structure gradually disappears with the onset of total reflection.

For a given angle of incidence the "p" interferences are damped more than the "s" interferences. Calculations of the transmittance also exhibit this effect.

b. Reflection

Equations (8) and (11) and the discussion accompanying them indicate that the reflectance of "p" light at oblique incidence may exhibit interesting structure as a function of angle and wavelength near λ_p. A computer calculation of the reflectance of an unbacked 100 Å Al film (unattainable in practice) for various angles of incidence yields the curves shown in Fig. 3. In calculations for thicker films the structure is less pronounced. Figure 4 shows the measured reflectance of a 100 Å thick Al film evaporated on a glass microscope slide. For comparison Fig. 5 is the calculated reflectance of a film composed of 40 Å Al_2O_3--100 Å Al---semi-infinite glass. In both the measured curves and those calculated for a composite film the Brewster minimum is very pronounced but the plasma peak seen in the calculations of the unrealistic unbacked unoxidized Al film has disappeared. The reflectance minimum is enhanced and shifted more in the direction of λ_p compared with the unbacked and unoxidized foil. Curves similar to Figs. 4 and 5 have been found for film thicknesses up to about 500 Å.

As in the case of transmission measurements interference effects appear with films thicker than about 500 Å at wavelengths less than the plasma wavelength. The interference effects are much more pronounced in reflection than transmission and can be seen in films with a glass substrate as well as with unbacked films. Figure 6 shows the reflectance versus wavelength spectrum of a 3040 Å Al film for $\phi = 15^{\circ}$. The spacing between maxima is given with reasonable accuracy by Eq. (15). For a thickness 3040 Å, for instance, we calculate an average spacing of about 45 Å, for a 1000 Å thickness 100 Å spacing both of which are in good agreement with the measurements. For the angle dependence and damping of "p" and "s" interferences we found the features reported above in transmission. Computer calculations indicate that thin oxide layers amplify the interference effects considerably and shift the position of the maxima but not their separation.

Our transmission and reflection measurements were reproducible to better than five percent. Systematic errors in our measurements can be somewhat larger. A significant error may enter through the inhomogeneity of both the synchrotron radiation intensity and the sensitivity of our detector photocathode. This makes absolute and accurate measurements, particularly of reflectance, difficult. For more exact comparison of the theoretical and measured curves we should take into account the possible influence of higher order and stray light and that our light is less than 100% polarized. The agreement of the curves calculated assuming the light was 100% polarized and the measured curves show conclusively, however, that the light emerging from our monochromator is highly polarized.

c. Summary

The experiments on thin Al films described here have not been carried out with the intent of making precise absolute reflection and transmission measurements. Rather we were interested in the relative behavior of these quantities under variation of the wavelength, polarization, and angle of incidence, as well as for various film thicknesses. Measuring these quantities allows the study of the excitation of plasmons by photons, the Brewster minimum, and interference effects. The properties found in the experiments are closely related to the singular behavior of the complex dielectric constant of a free-electron metal in the neighborhood of the plasma frequency.

A second aim of this research program was to test the feasibility of the application of synchrotron radiation and its unique properties, particularly its intense continuum and its high degree of polarization, to solid state physics studies.

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Footnotes

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FIGURE CAPTIONS

Fig. 1 Polarization of the light incident on the monochromator grating

Fig. 2 Measured transmittance of an unbacked 280 Å thick Al foil

Fig. 3 Calculated reflectance of a 100 Å thick unbacked Al film for "p" light

Fig. 4 Measured reflectance of an oxidized 100 Å thick Al film on a glass substrate for "p" light

Fig. 5 Calculated reflectance of a composite film 40 Å Al_2O_3-100 Å Al-glass for "p" light

Fig. 6 Measured reflectance of a 3040 Å thick unbacked Al foil for "s" light, angle of incidence 15°

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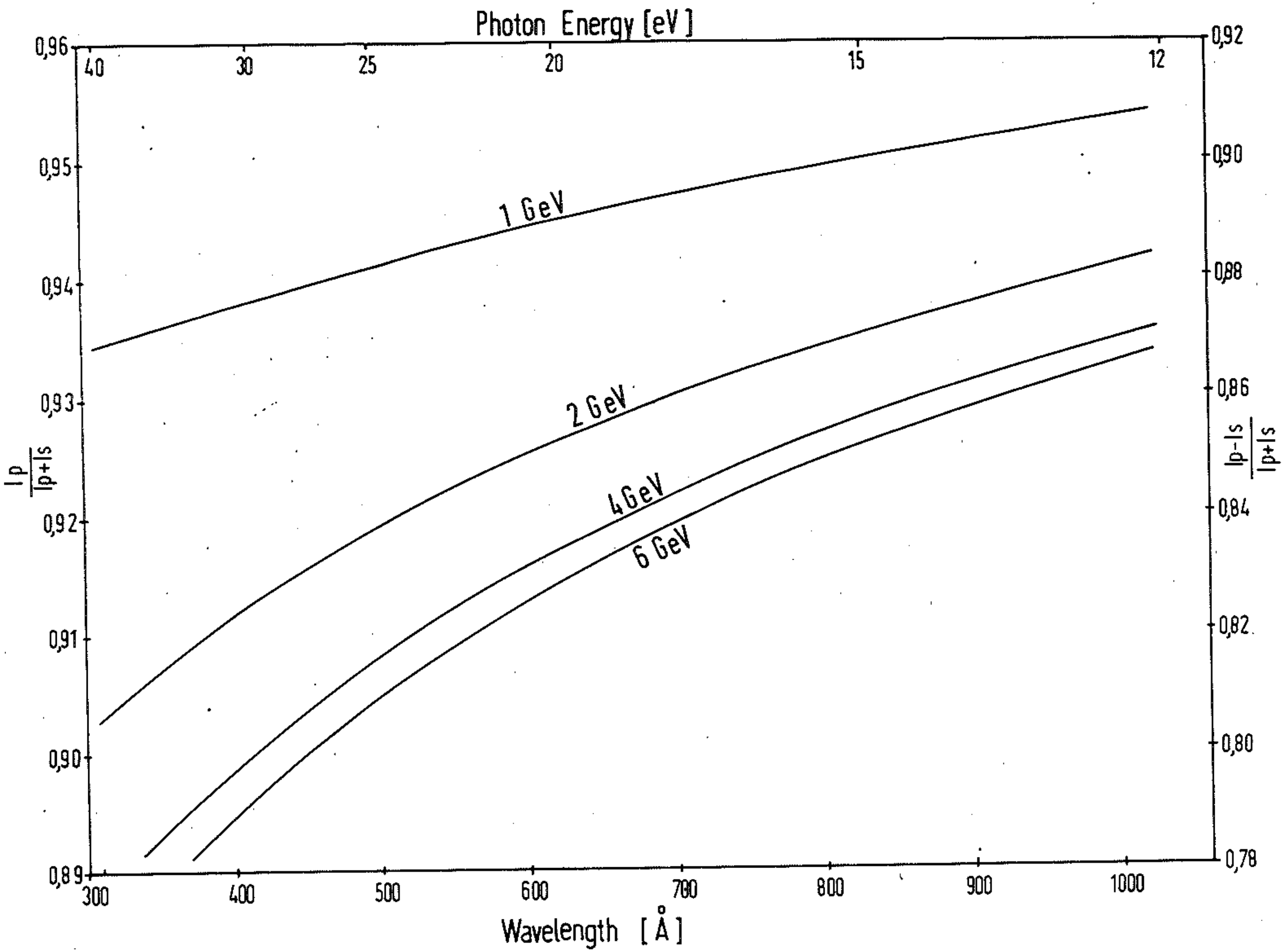

Photon Energy [eV]
40
30
25
20
15
12
0,96
0,95
0,94
0,93
0,92
0,91
0,90
0,89
$\frac{Ip}{Ip+Is}$
0,92
0,90
0,88
0,86
0,84
0,82
0,80
0,78
$\frac{Ip-Is}{Ip+Is}$
1 GeV
2 GeV
4 GeV
6 GeV
300
400
500
600
700
800
900
1000
Wavelength [Å]

Fig. A1

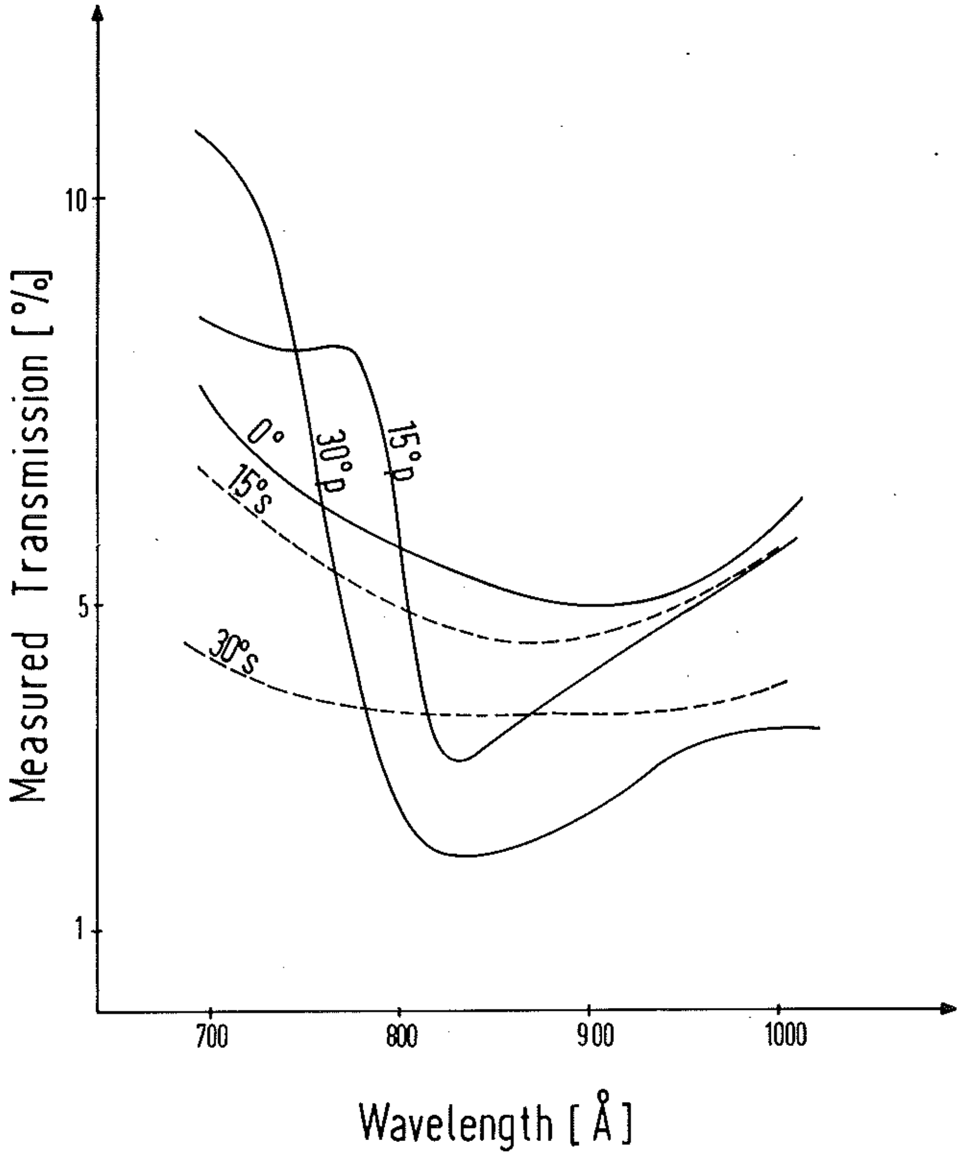
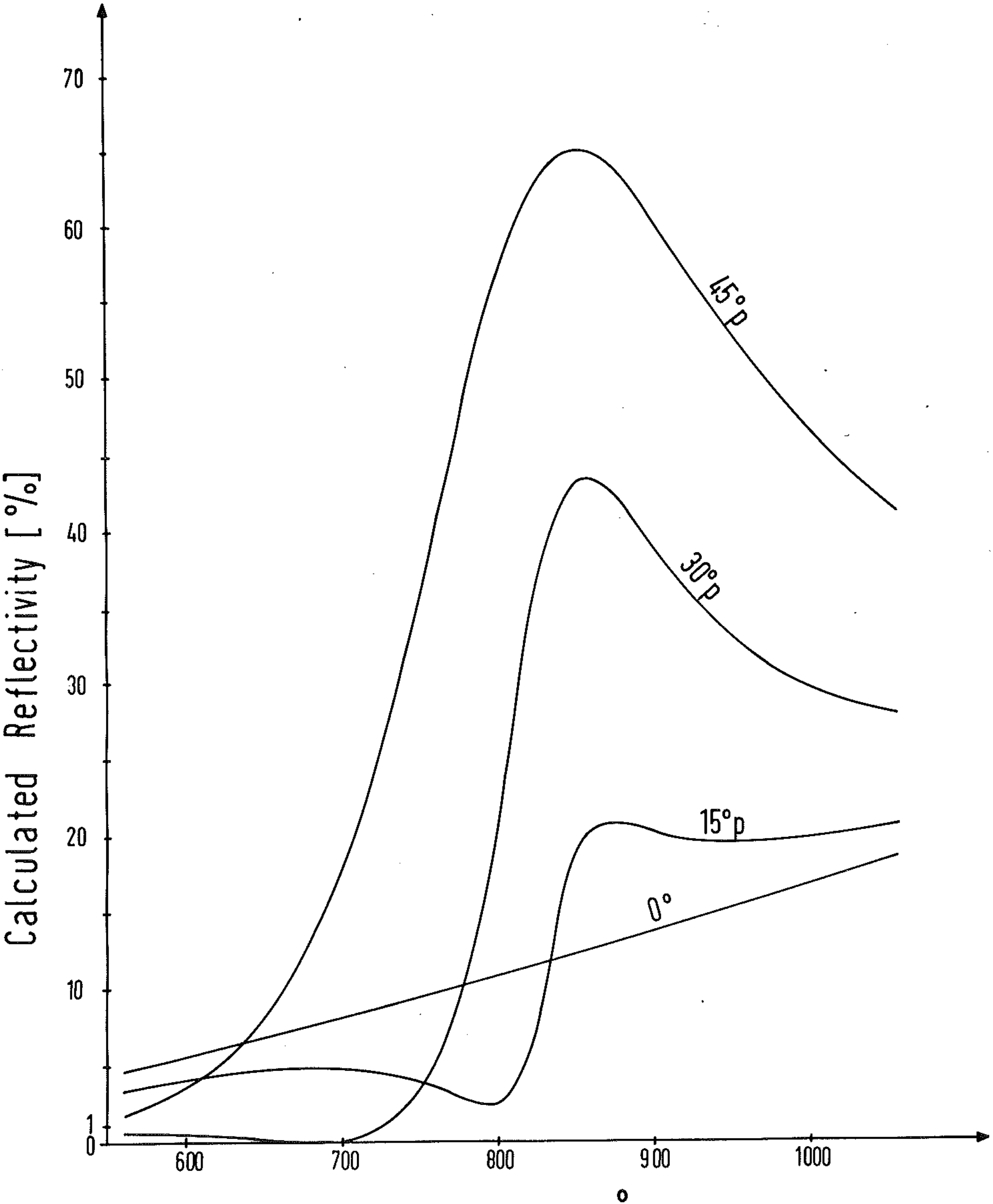

10
5
1
Measured Transmission [%]
0°
30°p
15°p
15°s
30°s
700
800
900
1000
Wavelength [Å]

Fig.A2



Calculated Reflectivity [%]
70
60
50
40
30
20
10
1
0
600
700
800
900
1000
Wavelength [Å]
45° p
30° p
15° p
0°

Fig. A3

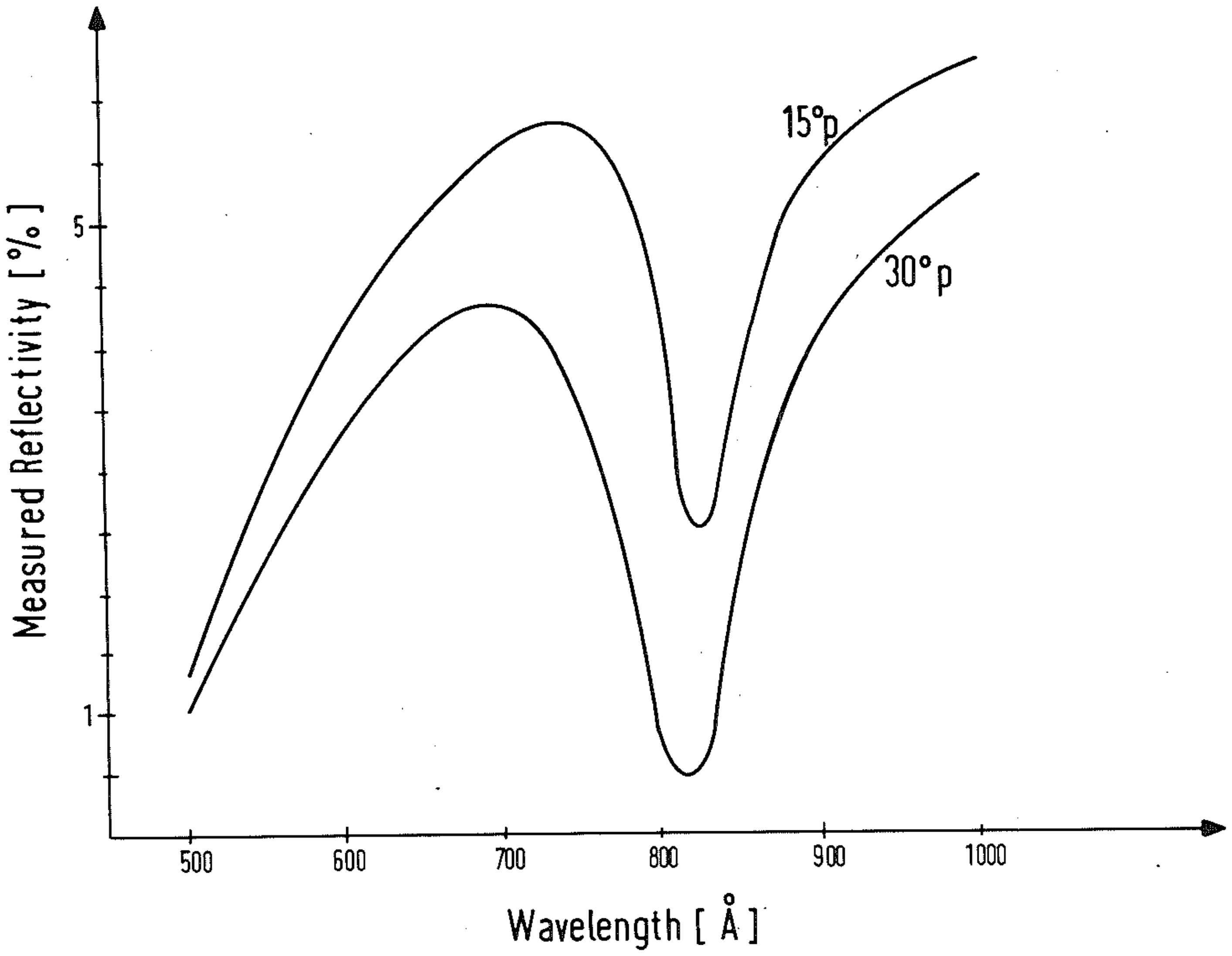

Measured Reflectivity [%]
5
1
15°p
30° p
500
600
700
800
900
1000
Wavelength [Å]

Fig. A4

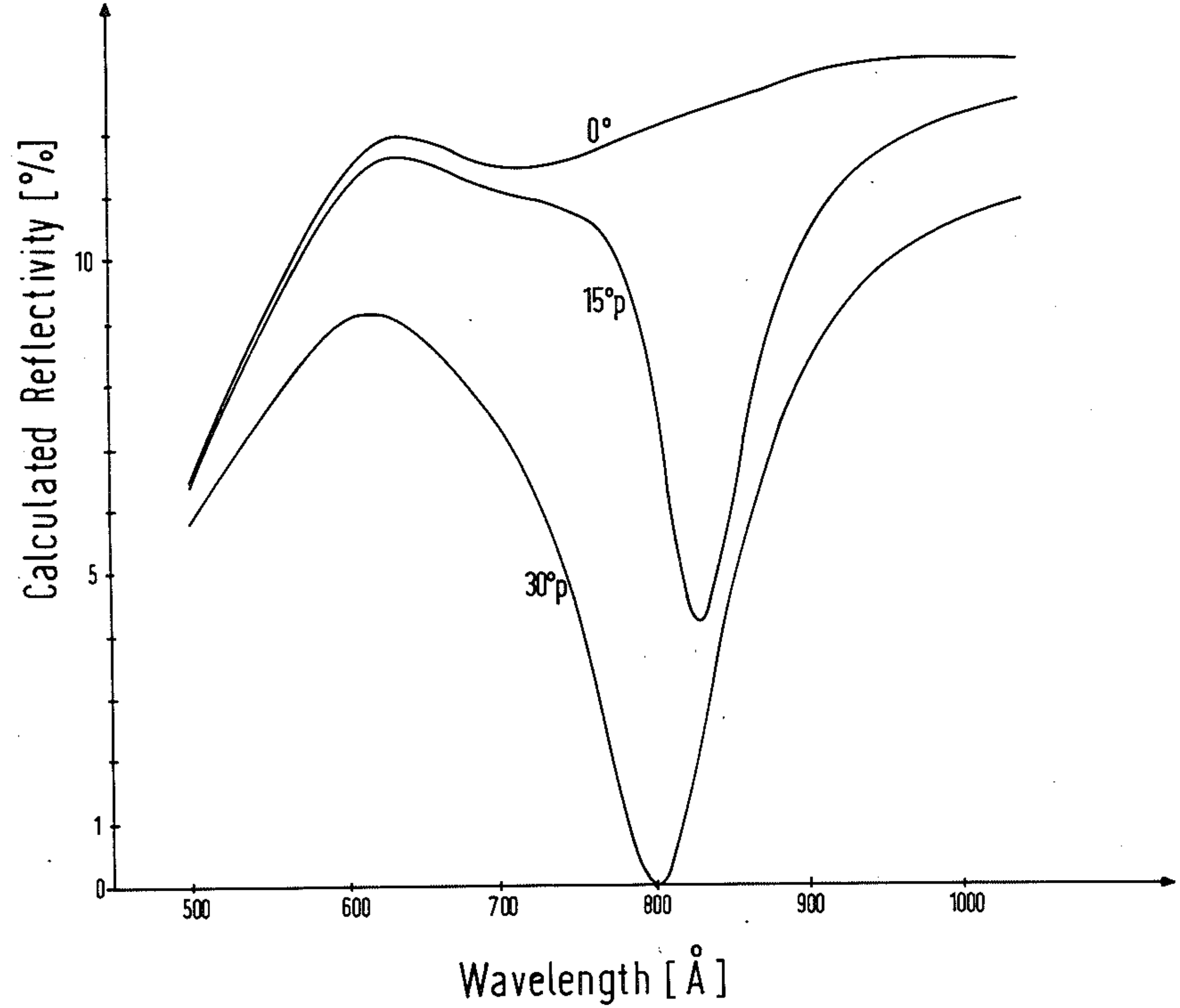
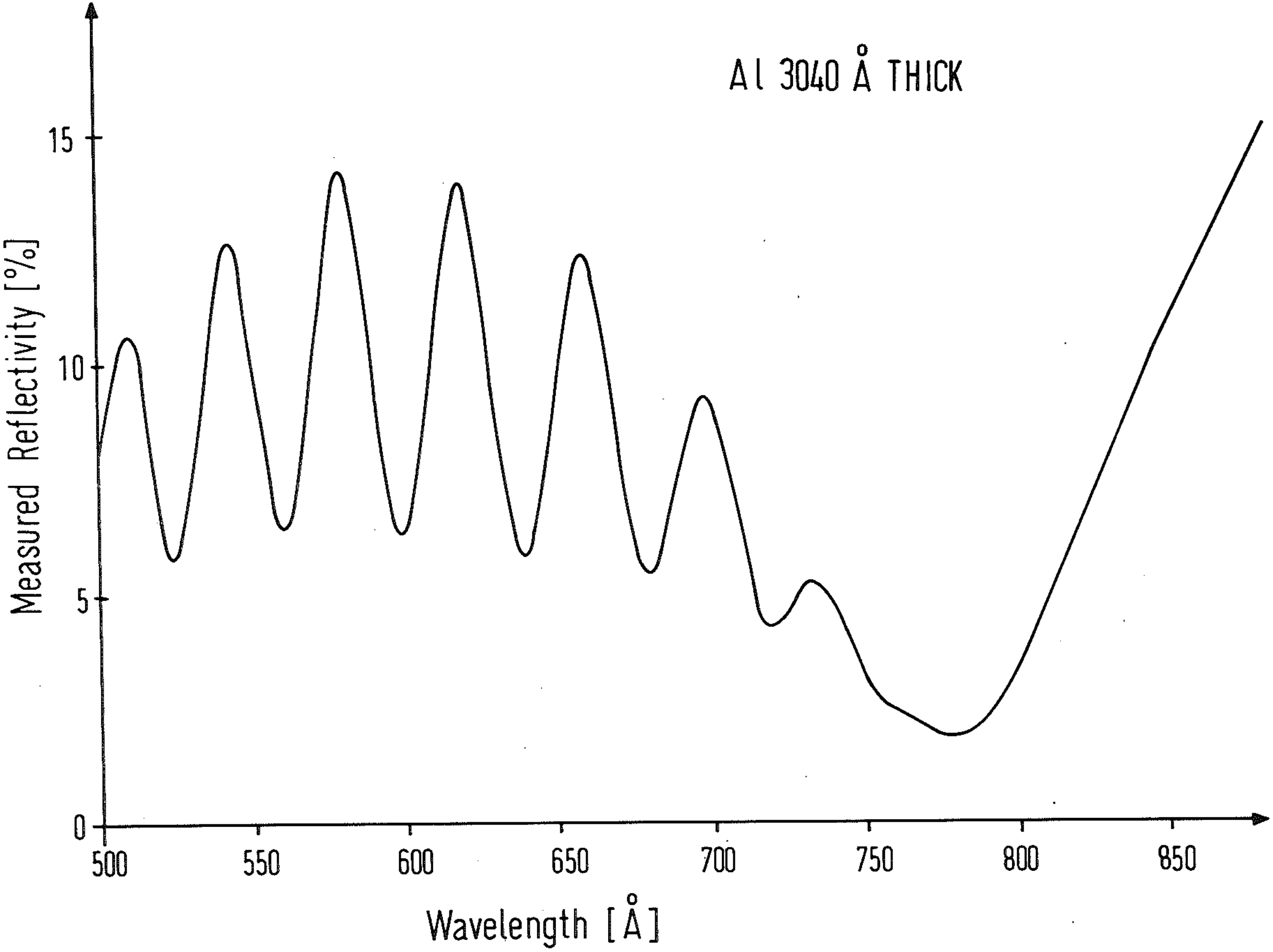

Calculated Reflectivity [%]
10
5
1
0
500
600
700
800
900
1000
Wavelength [Å]
0°
15°p
30°p

Fig. A5



Al 3040 Å THICK
Measured Reflectivity [%]
15
10
5
0
500
550
600
650
700
750
800
850
Wavelength [Å]

Fig. A6

Investigations of Aluminum Films with Synchrotron Radiation of Wavelengths 500 to 1000 Å

B. Polarization Dependent Photoeffect*

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ABSTRACT

Measurements of the photocurrent from thin Al cathodes in a windowless electron multiplier as a function of wavelength, polarization, angle of incidence, and film thickness have been carried out in the extreme vacuum ultraviolet. A normal incidence monochromator utilizing synchrotron radiation provided highly polarized light. A marked difference is found between the photocurrent measured during irradiation of thin films with "s" and "p" polarized light at wavelengths near the Al plasma wavelength (835 Å). For films thicker than about 500 Å pronounced interference effects are found in both "p" and "s" light at wavelengths less than the plasma wavelength. The observations can be explained by assuming the photocurrent is related to the photon density (electromagnetic energy density) in the photocathode. Calculations of the energy density in films irradiated with light give the structure found in the measured photocurrent. The measurements indicate our monochromator yields light with a degree of polarization consistent with the calculated polarization of the synchrotron radiation incident upon our grating (about 85%).

I. Introduction

The influence of polarization on the photoeffect has been recognized for a long time. In thin alkali metal foils there is a maximum in the photocurrent at the plasma wavelength λ_p in the near ultraviolet for foils irradiated with "p" light at an oblique angle of incidence [1,2]. For incident "s" light there is no structure near λ_p. Steinmann and Skibowski [3] recently reported a similar effect for Al near its plasma wavelength 835 Å. In this work we have utilized thin Al photocathodes in an open electron multiplier to investigate more extensively the polarization dependent photoeffect with the unique polarization properties of synchrotron radiation in the extreme vacuum ultraviolet.

In section II we point out the connections between electrodynamic quantities and the photoeffect used to explain our measurements. Section III contains the results of our measurements as well as comparisons with the predictions of calculations. Little mention is made of experimental techniques since they are nearly identical to those discussed in the previous paper (A). Throughout this paper we use the notation of A.

II. Theory

The volume photoeffect [4-6] is believed to be predominant in the extreme vacuum ultraviolet. We thus write the number of electrons N' excited per unit volume and time at a depth $z > 0$ under the surface of the photocathode and contributing to the photocurrent

$$N'(\omega,\phi,\alpha,z) = \sigma(\omega,z)\cdot\left|\tilde{E}(\omega,\phi,\alpha,z)\right|^2 \qquad (1)$$

where $|\tilde{E}(\omega, \phi, \alpha, z)|^2$ indicates the energy density (photon density) in the film which is a function of the light frequency ω incident on the cathode, the angle of incidence ϕ, the polarization α (α = p or s for "p" or "s" light respectively), as well as the depth z. The factor $\sigma(\omega, z)$ is related to the cross section for photoexcitation and the scattering of the electrons on their way to the surface. It contains matrix elements depending upon the detailed electronic properties of the metal. (For a rigorous treatment see Fan [4].) N' is linearly related to the absorption per unit volume which is proportional to $\varepsilon_2|\tilde{E}|^2$. With isotropic absorption, excitation, and scattering of the electrons $\sigma(\omega, z)$ will be independent of polarization and angle of incidence. Integration of Eq. (1) over the irradiated volume of the cathode gives the total number N of photoelectrons per unit time

$$N(\omega, \phi, \alpha) = S \cdot \sec\phi \cdot \int_0^\infty N'(\omega, \phi, \alpha, z)\, dz \qquad (2)$$

where S is the fixed cross section of the incident beam. It is believed that photoelectrons have mean free paths ℓ of magnitude 10 - 100 Å [7,8]. $N'(\omega, \phi, \alpha, z)$ is appreciably different from zero only for $z \leq \ell$. If the wavelength $\lambda \gg \ell$ the energy density will not vary noticeably in a depth ℓ from the value at the surface so that we can approximate $|\tilde{E}(\omega, \phi, \alpha, z)|^2$ by $|\tilde{E}(\omega, \phi, \alpha, 0)|^2$ the energy density just under the surface. With these assumptions we obtain from Eq. (2):

$$N(\omega, \phi, \alpha) \approx S \cdot \sec\phi \cdot |\tilde{E}(\omega, \phi, \alpha, 0)|^2 \cdot \int_0^\ell \sigma(\omega, z)\, dz \qquad (3)$$

In Al interband transitions lie far from the plasma frequency.[9,10] Thus $\varepsilon_2(\omega)$ and also $\sigma(\omega, z)$ will be smoothly varying functions of ω, so that the spectral dependence of the photoyield may be essentially that of the energy density. The reduced photocurrent

$$i_r \equiv \frac{N(\omega,\phi,\alpha)}{N(\omega,0,\alpha)} \approx \frac{|\tilde{E}(\omega,\phi,\alpha,0)|^2}{|\tilde{E}(\omega,0,\alpha,0)|^2} \cdot \sec\phi \tag{4}$$

is independent of the poorly known (but interesting) $\sigma(\omega, z)$ and the electronic mean free path which are removed by normalization to 0° angle of incidence.

A straightforward calculation of the energy density just under the surface using the electromagnetic boundary conditions yields for "p" light

$$|\tilde{E}(\omega,\phi,p,0)|^2 = |\tilde{E}_{inc}|^2 \left\{ \cos^2\phi \cdot |1-\tilde{r}_p|^2 + \sin^2\phi \cdot |1+\tilde{r}_p|^2 \cdot |\tilde{\varepsilon}|^{-2} \right\} \tag{5}$$

and for "s" light

$$|\tilde{E}(\omega,\phi,s,0)|^2 = |\tilde{E}_{inc}|^2 \cdot |1+\tilde{r}_s|^2 \tag{6}$$

where $\tilde{E}_{inc}$ is the electric vector of the incident light, $\tilde{r}_p$ and $\tilde{r}_s$ are the complex angle and frequency dependent reflection coefficients for "p" and "s" light, and $\tilde{\varepsilon}(\omega)$ is the complex dielectric constant.[11,12] Equations (5) and (6) are quite general. They are valid not only for a semi-infinite medium or a thin film, but also for an arbitrary multilayer. They include as a special case the equations given for a semi-infinite medium by Juenker et al. [13] in their analysis of the vectorial photoeffect of molybdenum. The cosine squared term in (5) corresponds to the component of the electric field parallel to the surface and the sine squared term to the normal component. It follows that the absorption of "p" light just below the surface (which is proportional to $\varepsilon_2|\tilde{E}|^2$) has two terms. The first is proportional to ε_2 and the second to $\mathrm{Im}(1/\tilde{\varepsilon})$ which is known as the energy loss function and which peaks at the plasma frequency. Both terms are modulated by factors including the complex reflection coefficients. If the thickness d of the film is very small compared to the wavelength of the incident radiation we can neglect the modulating factors.

In the special case of a free electron gas in the neighborhood of the plasma frequency for angles of incidence and thicknesses such that $|\tilde{\varepsilon}| \ll \sin^2\phi$ and $2\pi d/\lambda \ll 1$

$$|\tilde{E}(\omega,\phi,p,0)|^2 = |\tilde{E}_{inc}|^2 \sin^2\phi \, \frac{\omega_p^2}{4(\omega-\omega_p)^2 + \frac{1}{4}\omega_p^2(\beta\sin\phi\tan\phi + 2\varepsilon_2)^2} \qquad (7)$$

where we have utilized Eq. (5) and also Eq. (5) of A. (We drop the first term of (5) since it is small compared to the second.) Thus according to Eqs. (5) and (7) a resonance maximum of the photocurrent at the plasma frequency is expected. This is due to the maximum in the absorption occurring in conjunction with the excitation of plasmons for "p" light under oblique incidence. In "s" light we have no normal component and thus no maximum at the plasma frequency. Figure 1 shows the result of a computer calculation of the electromagnetic energy density for "p" light just under the surface of a 100 Å thick Al film. Here the exact Eq. (5) was used together with the Drude-model parameters given in A II. The direct calculation of the absorption $(Ab)_p = 1 - (T_p + R_p)$ of a thin film with the same approximations used in deriving Eq. (7) yields the same resonance behavior as that given in Eq. (7). See also Eqs. A(7) and A (8). Equations (5) and (6) show that the energy density at the surface is closely related to the reflectance. According to the discussion in A it should thus be possible to find in addition to the plasma effect just described structure in the photoyield near the plasma wavelength which is due to the Brewster minimum for "p" light and also interference effects. Some influence of total reflection on the photoyield is also expected.

Aluminum films which have been exposed to air (as ours were) have an oxide layer some tens of Ångstroms thick [14]. Since photoelectrons may have mean free paths of the order of 10 - 100 Å, most of the photoelectrons will originate in the oxide rather than Al. Thus the analysis above cannot be expected to predict accurately the behavior of our measurements. The energy density in the oxide layer determined by the optical constants of the oxide and those of the Al film under it has to be taken into account. Therefore, rigorous calculations of the energy density in a multilayer [12] have been carried out assuming optical constants and thicknesses of the different layers. Some results of computer calculations for realistic situations will be presented together with experimental results.

III. Measurements of the Photocurrent

We have investigated the polarization dependent photoeffect for wavelengths between 500 and 1000 Å and film thicknesses from 100 to 4000 Å.

The films were prepared as described in A. A square microscope cover glass 18 x 18 mm in area and 0.2 mm thick served as the substrate. The cover glass was installed as the cathode of an open photomultiplier (Bendix M 306) mounted in the reflectometer described in A. The angle of incidence could be varied between 0° and 45° in steps of 7.5°. Rotating the detector 90° about the axis of the incident radiation varied the radiation from "s" to "p" just as in A.

The results of a measurement on a 100 Å thick Al cathode deposited on glass are shown in Fig. 2. The photocurrent is displayed as a function of wavelength and angle of incidence for "p" polarized light. The curves measured with "s" light show no structure and vary less then 5% from the curve obtained at 0°. The structure of Fig. 2 between 700 and 900 Å is due to Al. The fall in photocurrent at less than 700 and greater than 900 Å is due to the combined characteristics of our monochromator and the synchrotron radiation together with the quantum efficiency of our Al cathode. A similar angle and polarization dependence has been measured for Al films up to 300 Å thickness. We cannot expect to find the ideal resonance at the plasma wavelength predicted by Eq. (7) in experiments since we have no unbacked Al photocathode, but rather an Al film deposited on a glass substrate and oxide coated. If we calculate the energy density for the composite film 40 Å Al_2O_3--100 Å Al--semi-infinite glass with the optical constants used in A we find at the surface (i. e. just inside the oxide layer) the behavior shown in Fig. 3 for incident "p" light. In "s" light we find no marked difference in the photocurrent upon varying the angle of incidence. A comparison of Figs.2 and 3 shows the qualitative agreement between the measured photocurrent and that predicted from the energy density calculation. The minimum just below the plasma wavelength is related to the Brewster minimum found in reflection through Eq. (5). (see also A.) With increasing angle of incidence the structure gradually disappears due to the onset of total reflection. The maximum at the plasma wavelength is very weak, but the ideal resonance form may be observable under ultra-high vacuum conditions.

Figure 4 shows both the experimental and theoretical photocurrent curves for a 100 Å thick Al film on glass normalized to the 0°-curve as in Eq. (4). The difference between the experimental and theoretical curves is less than 10 per cent. Our calculated curve was made for 100% polarized light. In reality the radiation falling upon the grating of our monochromator is about 85 per cent polarized. A reduction in the polarization brings the theoretical and experimental curves into better agreement.

With films of thickness greater than about 600 Å the photocurrent versus wavelength curves are different from those of thinner foils. Figures 5 and 6 show the photocurrent as a function of wavelength and angle for a 970 Å Al film in "s" and "p" polarization respectively. For wavelengths less than 800 Å periodic maxima appear with a spacing of about 100 Å, while above 850 Å the photocurrent has no structure. Cairns and Samson [15] have observed similar effects with unpolarized light. Our observations can be simply explained as optical interference by realizing that according to Eqs. (5) and (6) interference effects in the reflectance are associated with those in the energy density at the surface. The periodic maxima occur at wavelengths less than the plasma wavelength where the metal becomes transparent. Maxima and minima for the "s" and "p" curves lie at the same position. With increasing angle of incidence they are shifted to smaller wavelengths and finally vanish when total reflection begins. At a given angle of incidence the interference effects are more pronounced in "s" than "p" light. A special difference between the "s" and "p" measurements appears in the 15° curve near 800 Å. For "s" polarization the 15° curve lies above the 0° curve, while for "p" polarization it lies below the 0° curve. In addition, the 15° "p" curve shows a saddle at 790 Å which does not appear in the "s" polarization measurement. This pronounced difference is a manifestation of the Brewster minimum in reflection of "p" light. In films of increasing thickness the interference maxima become closer together. For a 3040 Å film we have measured a spacing of about 40 Å in agreement with Eq. A (15). The same sample was also used in an optical reflection measurement of the type described in A. Identical spacing was found in the photoemission and reflectance interferences. The polarization and angular dependence of measurements on 3000 to 4000 Å thick films were the same as those found with the 970 Å films. The ratio of the photocurrent on a maximum to that at a minimum becomes smaller with larger film thicknesses. Calculations of the energy density at the surface of an oxide-Al film-glass multilayer give not only the polarization dependent properties of the photoemission but also the observed damping of the interference effects with increasing angle of incidence or thickness.

We have performed a similar experiment with In. Interferences can be seen in the In window between 750 Å (the N_{45} edge) and1100 Å (the plasma wavelength). Figure 7 shows the photoemission measured with In films 1200 and 2010 Å thick.

The reduced photocurrent was determined with an accuracy of better than five per cent. Since the incident intensity and the amplification factor of our multiplier were only approximately known we could not quote an exact photoefficiency. It seems to be 0.1 to 0.2 electrons per photon in our spectral region as it is with the W cathode. (The radiation at the exit slit of width 0.5 mm corresponding to 3.5 Å was so intense that we obtained with Al cathodes an anode current of the order 10^{-9}A at 800 Å with only 1100 V applied to the dynode strip of the Bendix multiplier while the accelerator operated with 5 mA at 6 GeV.)

IV. Summary

Measurements of the photocurrent of thin Al films in the vuv under variation of wavelength, polarization, and angle of incidence, as well as of thickness give results which can be closely related to the electromagnetic energy density in the film. The spectral and polarization dependence of the photocurrent reflects the dielectric behavior typical for a free electron gas near its plasma wavelength.

In addition the measurements, together with those of the previous paper (A), give conclusive evidence that the synchrotron radiation in the vacuum ultraviolet is polarized as predicted by electrodynamics and that our near-normal incidence monochromator preserves this polarization.

The experiments described show it may be possible to utilize open multipliers with thin metal cathodes as simple polarization analysers in a limited spectral region.

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Footnotes

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FIGURE CAPTIONS

Fig. 1 Calculated energy density under the surface of a 100 Å thick Al film.

Fig. 2 Measured photocurrent of a glass backed 100 Å thick Al film.

Fig. 3 Calculated energy density multiplied by sec ϕ at the surface of an oxidized Al film deposited on glass (40 Å Al_2O_3--100 Å Al--glass).

Fig. 4 Experimental and theoretical reduced photocurrents:

——————— experimental

- - - - - - - - - theoretical assuming 100% light polarization.

Fig. 5 Measured photocurrent of a 970 Å thick Al film in "s" light.

Fig. 6 Measured photocurrent of a 970 Å thick Al film in "p" light.

Fig. 7 Interference effects in the photocurrent of In films. (The photocurrent scale is different for each thickness.)

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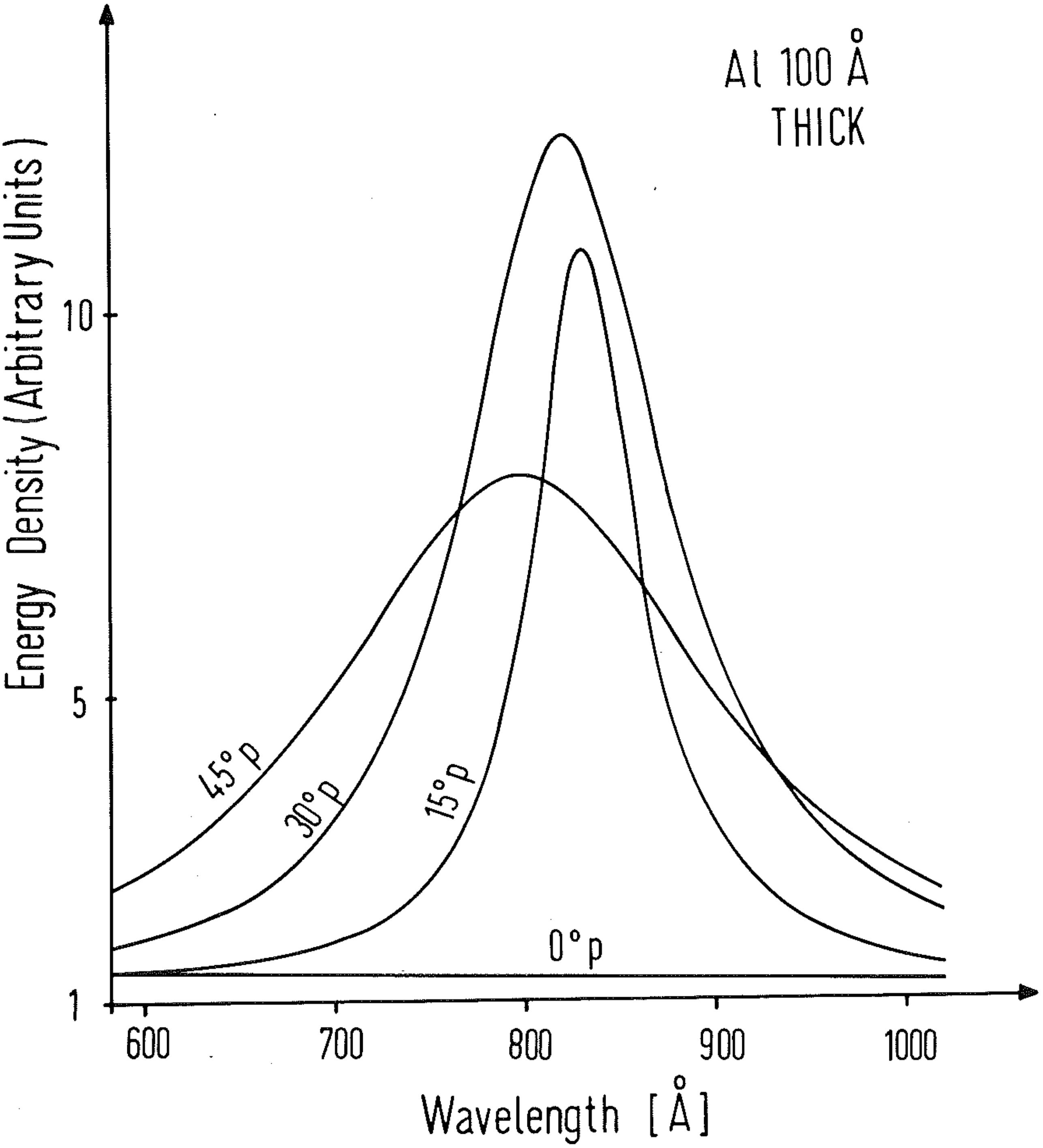

Al 100 Å
THICK
Energy Density (Arbitrary Units)
10
5
1
600
700
800
900
1000
Wavelength [Å]
45° p
30° p
15° p
0° p

Fig. B1

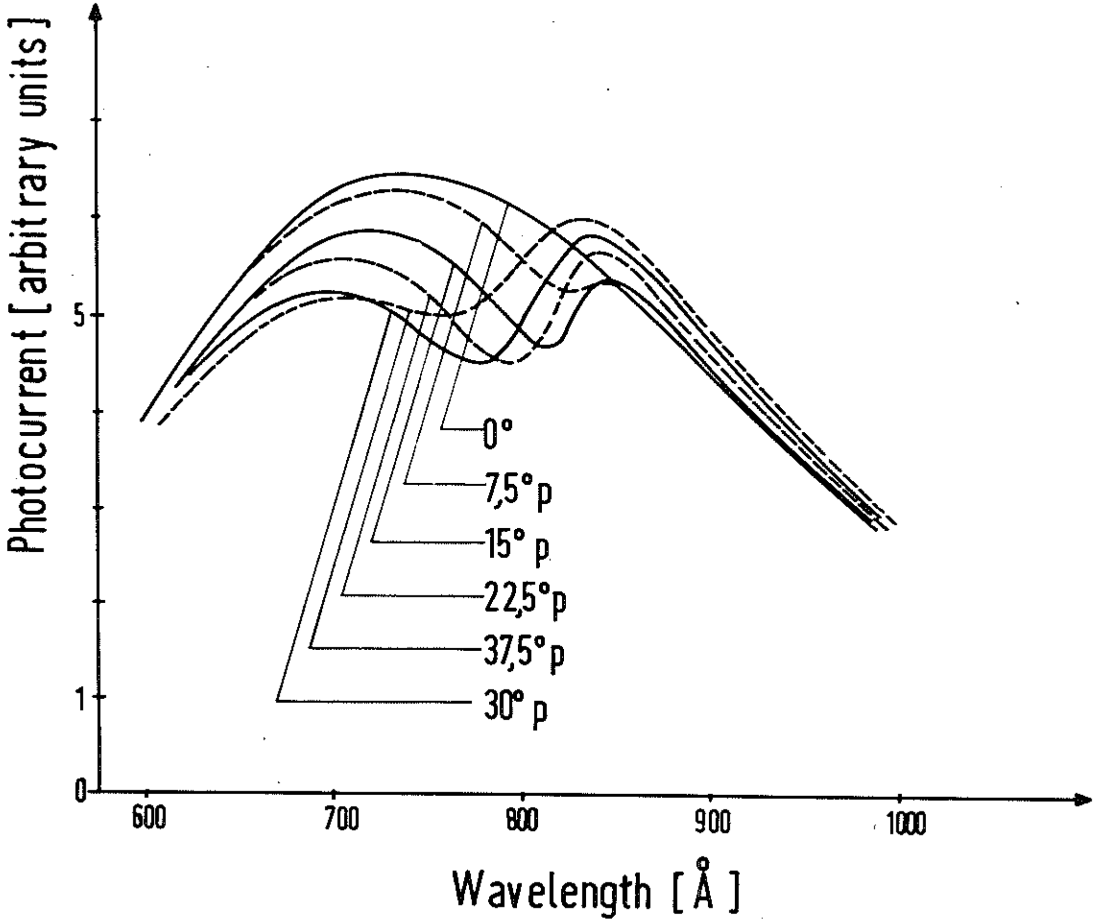
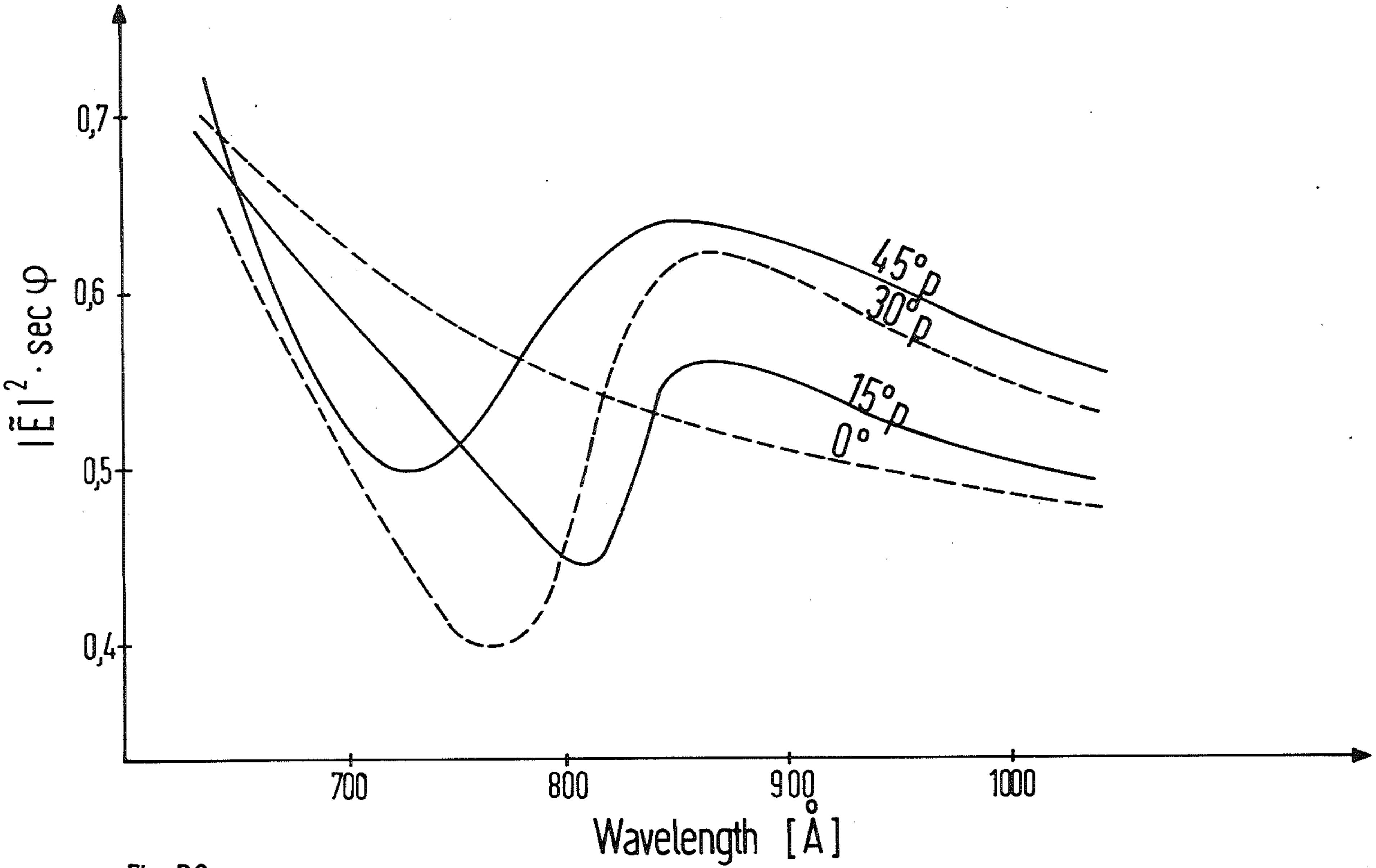

Photocurrent [arbitrary units]
5
1
0
600
700
800
900
1000
Wavelength [Å]
0°
7,5° p
15° p
22,5° p
37,5° p
30° p

Fig. B 2



$|\tilde{E}|^2 \cdot \sec \varphi$
0,7
0,6
0,5
0,4
700
800
900
1000
Wavelength [Å]
45° p
30° p
15° p
0°

Fig. B3

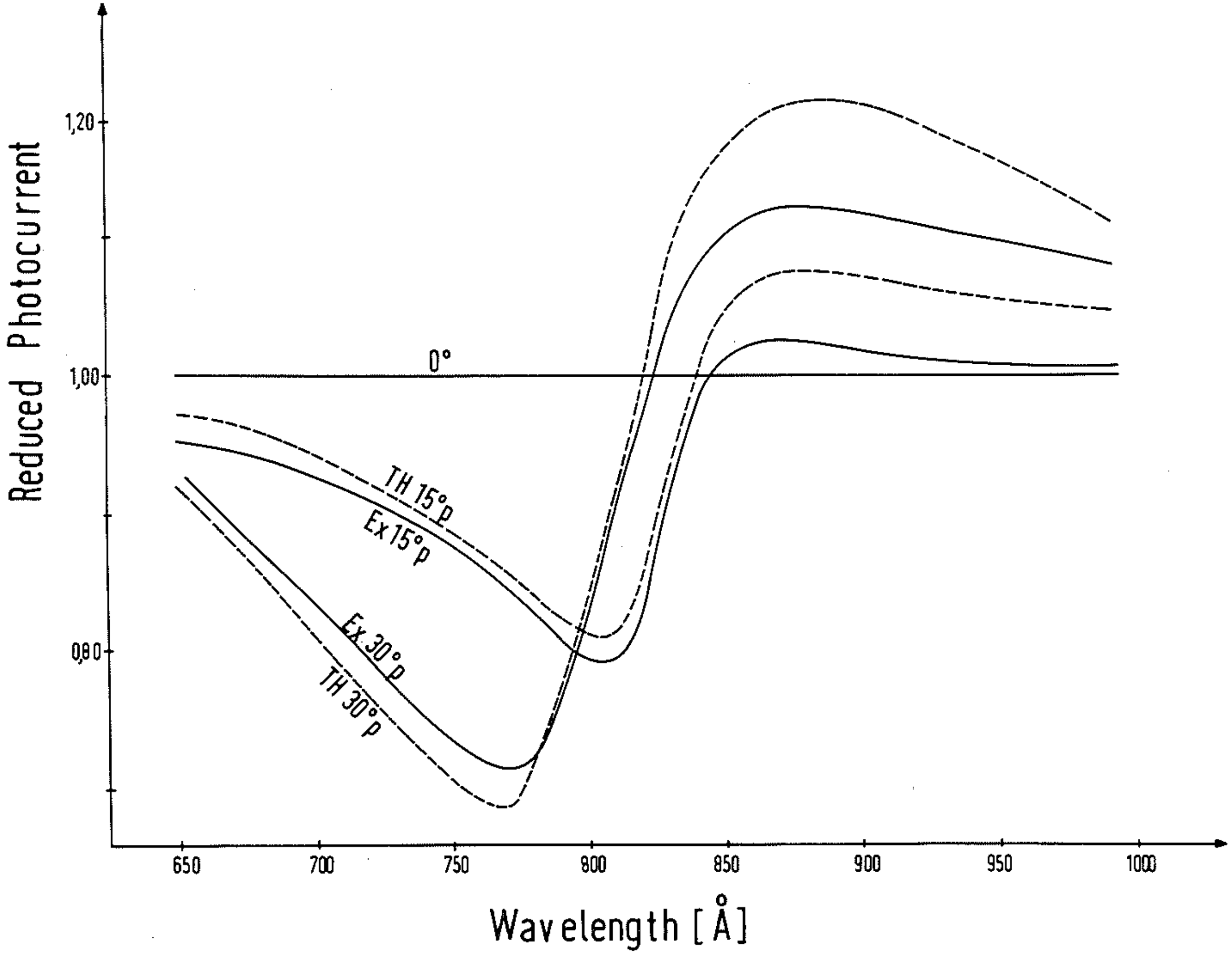

Reduced Photocurrent
1,20
1,00
0,80
0°
TH 15°p
Ex 15°p
Ex 30°p
TH 30°p
650
700
750
800
850
900
950
1000
Wavelength [Å]

Fig.B4

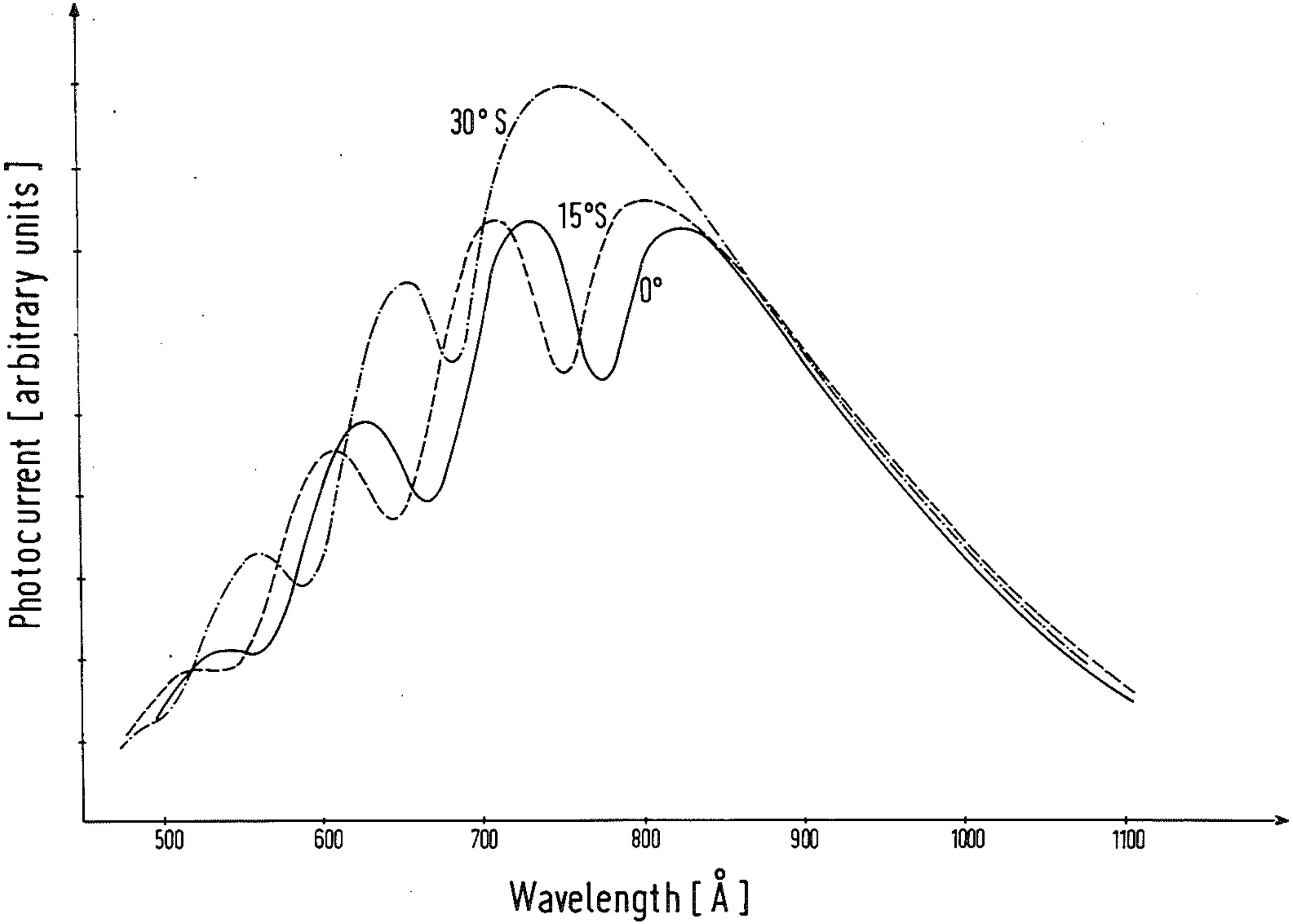

30° S
15°S
0°
Photocurrent [arbitrary units]
500
600
700
800
900
1000
1100
Wavelength [Å]

Fig.B5

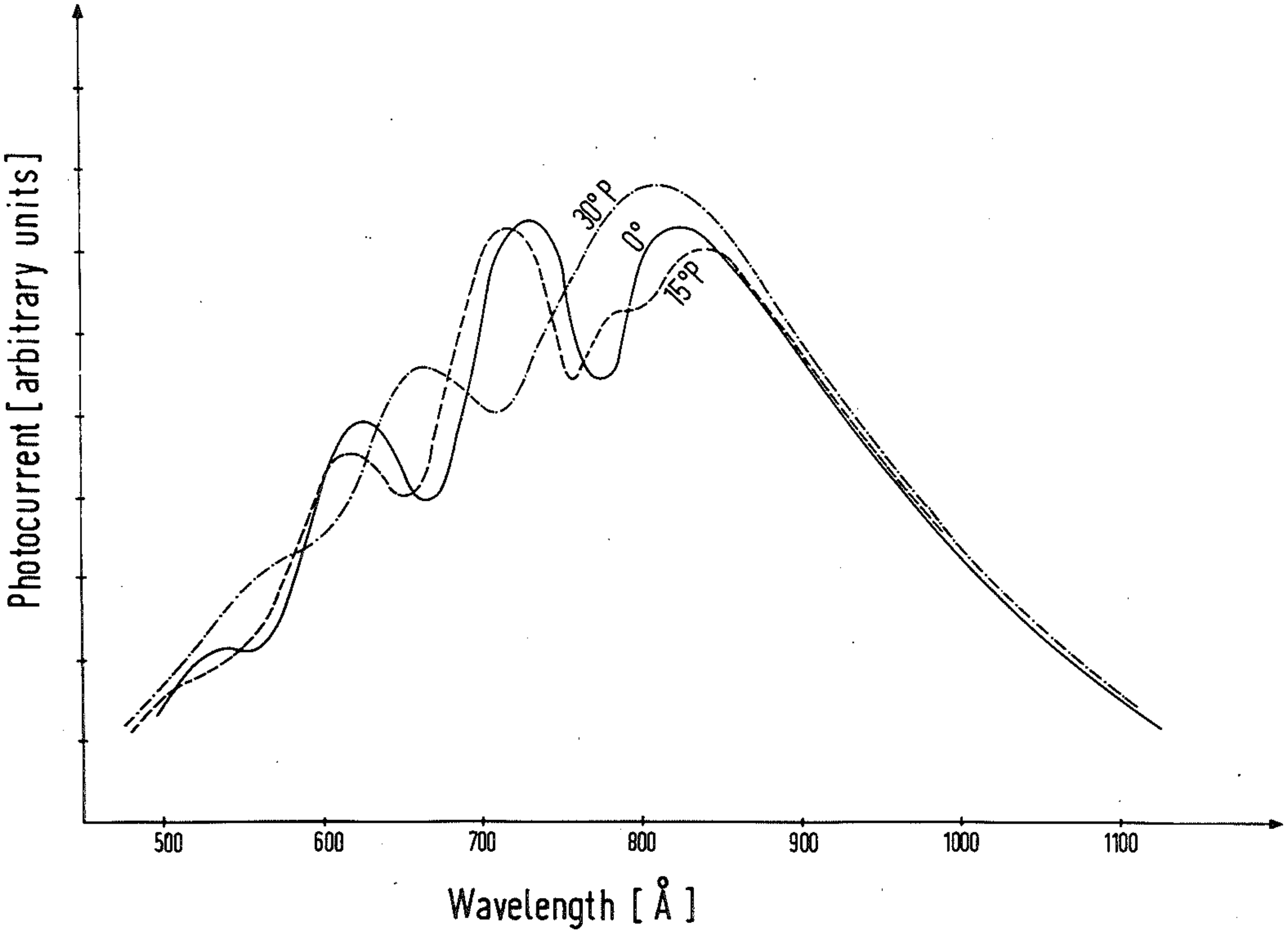
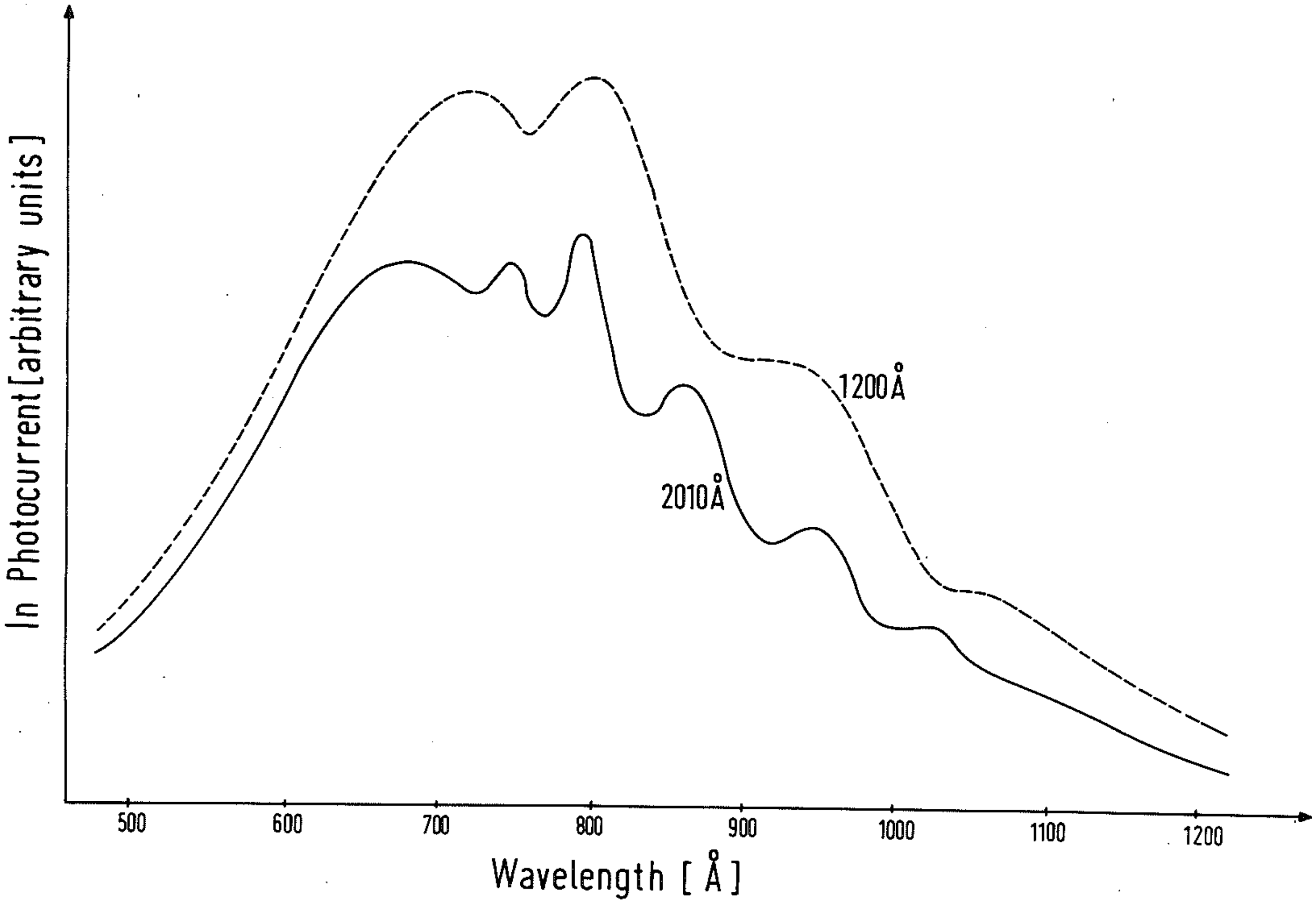

Photocurrent [arbitrary units]
Wavelength [Å]
500
600
700
800
900
1000
1100
30° P
0°
15°P

Fig. B6



ln Photocurrent [arbitrary units]
1200 Å
2010 Å
500
600
700
800
900
1000
1100
1200
Wavelength [Å]

Fig. B7