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0. Abstract

We present angular resolved scattering (ARS) distributions of various glass mirrors and of SiC mirrors at glancing incidence. The measurements have been performed at the XUV reflectometer station at the DORIS2 storage ring (HASYLAB) between 25 and 1.2 nm wavelength. The data are compared with different scattering theories by using least squares fitting procedures. Of all the available theories the Rayleigh-Rice vector perturbation theory is based on the most realistic model and gives by far the best agreement between experiment and theory. Indeed, even near the critical angle, where anomalous scattering arises, excellent agreement was obtained. Also, convincing evidence for scattering from dielectric fluctuations inside some of the glass samples was found. The rms-roughness, autocorrelation length (measure of the mean lateral separation of the surface irregularities), and the type of correlation function could be determined. Furthermore, we show that ARS-measurements are sensitive to detect irregularities with a mean lateral separation from 50 to 1500 nm. Therefore ARS-measurements in the soft x-ray range could become an important tool to supplement other methods characterizing optical surfaces. Practically all other familiar methods are restricted in spatial frequency corresponding to a lateral resolution above a few microns.

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1. Introduction

In many optical problems, such as monochromator- and beamline design, it is desirable to predict the angular and spectral dependence of the light scattered from the optical elements. Since no exact treatment of the scattering problem is feasible and since all theories are approximations in one or another way, an experimental test in the XUV spectral region of the different theories is very necessary. Furthermore a precise knowledge of the surface finish parameters, which have to be used in a particular scattering theory, is needed. A number of investigators have determined the statistical properties of surfaces by means of stylus, interferometric, and microdensitometer analysis methods[1-5], in particular the rms-roughness and the autocovariance function, and some have used their results to calculate angular resolved scattering (ARS) distributions [2,6]. Other authors have only measured the ARS-distributions but not the surface statistics. In the visible and near UV spectral region, light scattering is easily measurable and can be predicted from surface roughness measurements with reasonable accuracy. It has been shown by Bennett et. al. [3], that total integrated scattering (TIS) only depends on the value of the rms-roughness height δ_r as long as this is small compared to wavelength λ of the incident light. The ARS-distribution however depends not only on the value of δ_r but also on the autocorrelation length τ which is a measure of the mean lateral distance between the surface microirregularities. Excellent agreement between rms-roughness values obtained from TIS-measurements and those determined directly using profilometer- or interferometric methods was obtained in the visible spectral range [2,3]. Agreement was somewhat less between measured ARS-distributions and angular dependent light scattering calculated from directly measured surface roughness using a vector perturbation scattering theory [2,6]. Most of the available theories, as e.g. Beckmanns simple scalar theory or the more subtle Rayleigh-Rice vector perturbation theory, are based on the condition ($\delta_r \ll \lambda$). This could become a severe problem in the XUV spectral range, since for common optical surfaces their δ_r-values of typically 1 - 2 nm are in the order of the wavelength and the theoretical condition is violated. We have measured the ARS-distribution and the specular reflectivity versus angle of incidence $R(\theta_i)$ of various glass mirrors, metal-coated mirrors, and SiC-mirrors at different wavelengths. Different theories were fitted to the experimental data to obtain values for δ_r and the autocorrelationlength τ_a, in order to get a deeper insight into the applicability of available scattering theories.

2. Theory

When a plane harmonic wave strikes a rough surface polarization currents are generated. Some of the energy contained in these currents is radiated off as diffuse straylight. The interaction between light and surface roughness involves a change in electromagnetic momentum along the surface and therefore allows the surface currents to radiate in non specular directions. A rough surface can be considered to be composed of a Fourier superposition of sinusoidal waves having different amplitudes, spatial frequencies, and phases. Each Fourier component must satisfy its corresponding "grating" equation

$$\frac{2\pi}{\lambda} \cdot (sin\theta_s - sin\theta_i) = \triangle K \quad , \tag{1}$$

where $\triangle K$ denotes the wave vector transfer between the incident and scattered light, and θ_i and θ_s are the angles of incidence and of diffraction respectively. Thus, scattering from a slightly rough surface can be visualized as a distribution of the plus and minus first-order diffraction peaks produced by each Fourier component of the rough surface [8]. The range of $\triangle K$-values of the Fourier spectrum detectable in a straylight experiment can be deduced from eq.1. Consider θ_i and λ fixed and vary $\triangle K$. As $\triangle K$ becomes very small θ_s approaches θ_i and the diffracted orders will come close to the specular direction. The lower limit of detectable values of $\triangle K$ is given by the diameter of the probing light beam. Conversely, as $\triangle K$ increases, the diffraction orders move away from the specular direction until they vanish into the surface.

Angular resolved scattering is usually expressed in terms of the differential cross section per unit area : $\frac{1}{P_0} \cdot (\frac{dP}{d\Omega})$ [9]. Here $\frac{dP}{d\Omega}$ denotes the power scattered per unit solid angle $d\Omega$ into a direction specified by the angles (θ_s, ϕ) and P_0 is the total power incident on the surface. There exist two basic types of theories dealing with the interaction of light with surface irregularities : scalar and vector theory. Either theory can provide a basis for estimating statistical parameters from the measurement of the ARS-distribution. In order to get closed expressions for $\frac{1}{P_0}\cdot\langle\frac{dP}{d\Omega}\rangle$ an assumption about surface statistics is needed.

Within this paper the surface height is considered to be a random variable which is completely characterized by a two dimensional probability density function $P(z_1, z_2)$ giving the probability that the surface height $\zeta_r(x, y)$ at two laterally separated points 1 and 2 assumes values $z_1 \le \zeta_1 < z_1 + dz_1$ and $z_2 \le \zeta_2 < z_2 + dz_2$. Generally, this function is assumed to be Gaussian and isotropic, since several authors have reported about experimental evidence for a Gaussian height distribution [1,4,5]. The height distribution is then completely determined by its first moments : the variance $\langle\zeta_r^2\rangle$ and the autocovariance function $C_r(\tau) = \langle\zeta_r(\vec{\rho})\zeta_r(\vec{\rho}+\vec{\tau})\rangle$ [1]. The square root of the variance $\langle\zeta_r^2\rangle$ yields the rms-roughness while the autocovariance function describes the lateral surface statistics. $C_r(\tau)$ reaches its maximum at $\tau = 0$ ($C_r(0) = \langle\zeta_r^2\rangle$) and usually decreases monotonically with increasing separation parameter τ. Surface points which are infinitely separated are expected to be fully uncorrelated, i.e. $\lim_{\tau\to\infty} C_r(\tau) = 0$. The smallest value of τ which satisfies the equation $C_r(\tau_a) = a \cdot C_r(0)$ is called the autocorrelation length and provides a measure of the lateral separation of the surface microirregularities. The value of a ($a < 1$) depends on the type of the autocovariance function, of particular interest is 1/e. It has been confirmed many times, that a rough surface can be considered to be generated by two or more statistically independent roughness amplitudes $\zeta_r(x, y) = \zeta_{r1}(x, y) + \zeta_{r2}(x, y)$, e.g. short range roughness and long range waviness. In this case the autocovariance function of the surface is simply the superpositon of the single autocovariance functions of each roughness-amplitude.

[1] $\langle ... \rangle$ denotes the ensemble average and r stands for roughness

This is the assumption with which we analyze our data in this paper.

Beckmanns scalar scattering theory is based on the Helmholtz integral and neglects the vector character of light [10]. The central point is, that a solution of the electrical field to the Helmholtz wave equation at some point in space surrounded by a closed surface may be obtained, if the field values are known at all points on the surface. The Helmholtz integral may also be applied to non closed surfaces - in our application to a rough surface - which demands the field values to be known on the rough surface itself. Since these field values are generally unknown, certain approximations are made. The field values at each point of the surface are expressed in terms of the Fresnel reflection coefficients for a tangent plane at this point (Kirchhoff-method). This approximation requires the radii of curvature of the surface to be much larger than the wavelength λ of the incident light. Further, Beckmann assumes an average reflectivity $\bar{R}(\theta_i) = R(\bar{\theta}_i)$ neglecting the variation of the local reflectivity at each point of the surface to obtain a manageable Helmholtz integral.

Historically, a perturbation technique was first used by Rayleigh for scattering of accoustical waves. Meanwhile there have been several variations of first order perturbation techniques concerning the scattering of electromagnetic waves from rough surfaces [11,12,13,14]. The surface roughness is treated as a perturbation parameter having a weak influence on the situation of the smooth surface. Thus the diffusely scattered fields are a small correction to the zeroth-order fields (perfect smooth surface), which may be expressed by the Fresnel coefficients. These assumptions are generally suitable if $\delta_r \ll \lambda$, no further restrictions are necessary.

Kröger and Kretschmann have developed a perturbation approach with respect to the roughness amplitude which is based on a model consisting of a smooth surface and roughness induced surface currents [14]. This equivalent current model has an obvious physical meaning. It is assumed that the roughness and the slope of the surface are sufficiently small, so that the polarization currents of the rough surface have the same direction as the currents of the smooth surface. The difference between the polarization currents with and without roughness is taken as the source of the scattered fields. Basically, the different perturbation calculations lead to the same results. In this work we used a perturbation approach given by J.M. Elson [11] to calculate $\frac{1}{P_0} \cdot \langle \frac{dP}{d\Omega} \rangle$ for surface scattering from a plane bounded semi-infinite medium :

$$\frac{1}{P_0} \cdot \left\langle \frac{dP}{d\Omega} \right\rangle = \left(\frac{2\pi}{\lambda} \right)^4 \cdot \frac{|1-\tilde{\epsilon}|^2}{(2\pi)^2} \cdot \left[\frac{cos^2\theta_s}{cos\theta_i} \cdot (|s_r|^2 + |p_r|^2) \right] \cdot g_r(\triangle \vec{K}) \qquad (2)$$

Eq.2 is composed of three parts. The first term contains the factor $(\frac{2\pi}{\lambda})^4$,which is characteristic of dipole radiation, and gives the spectral dependence of scattering. The term in square brackets is the so-called optical factor, containing the polarizability and the angular radiation characteristics of the dipole currents. The factors s_r and p_r are the scattering coefficients [11] for s- and p-polarized scattered light respectively, they are related to the Fresnel coefficients. The surface statistics is contained in the third factor, $g_r(\triangle \vec{K})$. This surface factor gives a measure of the relative strength of the roughness Fourier component $\triangle \vec{K}$ involved in the scattering mechanism and is called the power-spectral-density function. The autocovariance function and the power-spectral-density function are Fourier transforms of each other. If the surface is generated by several independent roughness-amplitudes, then $g_r(\triangle \vec{K})$ has to be replaced by the corresponding sum of power-spectral-density functions.

Elson has also developed a vector scattering theory, for a medium with an isotropic fluctuation of the dielectric function. The dielectric perturbation is assumed to fluctuate at random in the plane parallel to the surface : $\tilde{\epsilon}(\vec{\rho}, z) = \tilde{\epsilon}_1 + \triangle\tilde{\epsilon}(\vec{\rho}, z)$, where

$\tilde{\epsilon}_1$ is the constant dielectric function of the host material and $\triangle\tilde{\epsilon}(\vec{\rho}, z)$ is a random variable, which fluctuates around zero. Further Elson assumes an exponential decay of the fluctuations with depth into the surface in order to simplify the model : $\triangle\tilde{\epsilon}(\vec{\rho}, z) = \triangle\tilde{\epsilon}(\vec{\rho}) \cdot e^{\alpha \cdot z}$, $z \leq 0$. The decay constant α determines the size of the scattering volume. The main physical difference between scattering from surface roughness and dielectric fluctuations is, that in the latter case the polarization currents are distributed throughout the sample rather than concentrated at the surface. The radiation emerging from these currents must travel through the scattering medium and pass through the boundary into the medium of observation. To obtain solutions of the scattered fields to Maxwells equations, Elson applied a perturbation approach using $\triangle\tilde{\epsilon}(\vec{\rho})$ as the perturbation parameter. This approximation is applicable as long as $\zeta_d / |\tilde{\epsilon}_1| \ll 1$, where $\zeta_d^2 = \langle |\triangle\tilde{\epsilon}(\vec{\rho})|^2 \rangle$ is the variance of the dielectric fluctuation, which is analogous to the rms-roughness parameter. The expression of $\frac{1}{P_0} \cdot \langle \frac{dP}{d\Omega} \rangle$ for scattering from dielectric fluctuations is similar to that of rough surface scattering. The statistics of the fluctuations are contained in the factor $g_d(\triangle \vec{K})$ which is the two dimensional Fourier transform of the autocovariance function of the dielectric fluctuations : $C_d(\tau) = \langle \triangle\tilde{\epsilon}(\vec{\rho}) \cdot \triangle\tilde{\epsilon}(\vec{\rho} + \vec{\tau}) \rangle$ where $C_d(0) = \zeta_d^2$. Elson has also considered cross-correlation effects between surface roughness and dielectric fluctuations in detail, but in this work the two sources of light scattering are assumed to be independent, in order to avoid overinterpreting of our experimental data. The equations by Elson are applicable for arbitrary angles of incidence θ_i, angles of scattering (θ_s, ϕ), dielectric function $\tilde{\epsilon}$, and state of polarization of incident and scattered light. For more details the reader is referred to reference [11].

3. Experiment

The experiments have been performed at the synchrotron radiation laboratory HASYLAB. Our experimental setup consists of a plane grating monochromator and an UHV-reflectometer. The monochromator supplies the reflectometer with radiation of photon energy between 50 and 1200 eV. Its principles and characteristics have been described in reference [15]. A toroidal mirror behind the exit slit of the monochromator refocusses the beam; near the sample, the beam size is $0.85 \times 2.2mm^2$ (FWHM, vert. $\times$ horiz.) and its divergence is $2.1 \times 4.8mrad^2$. The reflectometer provides computer-controlled independent rotation and translation of the sample and detector. Details of the reflectometer are given in reference [16]. The measurements were performed with a semiconductor diode (Hamamatsu G1127) in connection with a Keithley 617 electrometer, which provides a dynamic range of seven orders of magnitude. The capability of the diode has been investigated and is reported elsewhere [17]. The aperture of the detector is $1.0 \times 2.5mm^2$ (vert. $\times$ horiz.); the distance between sample and detector is 150 mm. Since the sample can be removed from the direct beam, normalization of the spectra can be done by moving the detector into the direct beam. Corrections for changes in the incoming photon flux are made by monitoring the total electron yield from the toroidal mirror in front of the reflectometer.

4. Results

We present results of ARS-measurements performed on zerodur-glass-, quartz-glass-, and SiC-mirrors. As an example fig.1-3 show results of our angular resolved straylight measurements of zerodur-glass. In all cases the incident beam was s-polarized. The data are normalized to the incident power, so that the intensity in the specular direction equals the specular reflectivity of the sample. Thus, the spectra show the intensity scattered into the detector divided by the incident intensity versus the polar angle of the detector, which was rotated inside of the plane of incidence. The accuracy of the data is estimated to be of about 5 % of the normalized detector signal. We fitted Elson's equations for $\frac{1}{P_0} \cdot \langle\frac{dP}{d\Omega}\rangle$ to the measured spectra using the method of least-squares. The expression for $\frac{1}{P_0} \cdot \langle\frac{dP}{d\Omega}\rangle$ as a function of the scattering angles (θ_s, ϕ) has its maximum in the specular direction and decreases very rapidly with angles away from the specular direction - perpendicular to the plane of incidence the steepness of this function is extremely pronounced. In calculating the theoretical quantity of the intensity scattered into the detector, the expression for $\frac{1}{P_0} \cdot \langle\frac{dP}{d\Omega}\rangle$ was numerically integrated with respect to the detector aperture within the fit-procedure, in order to account for the analytical behavior of the scattering distribution. The necessity of this convolution was confirmed by test calculations based on the geometrical parameters of our experimental setup. It was found that differences between convoluted and unconvoluted expressions of up to half an order of magnitude can occur, depending on the parameters δ_r, τ_a, and λ. The rms-roughness and the corresponding autocorrelation length served as free fit-parameters. Values of the dielectric function $\tilde{\epsilon}$ of the material under investigation were determined from reflectivity versus θ_i measurements and served as fixed parameters. The influence of the surface roughness on the specular reflectivity was taken into account by the factor $exp(-(4\pi \cdot \delta_r \cdot cos\theta_i/\lambda)^2)$ [10]. Fig.1 shows a plot of the measured ARS-distributions of a zerodur-glass mirror for three different angles of incidence $\theta_i = 75°, 80°, 83°$ at $\lambda = 10.3$ nm wavelength. The circles denote the experimental data and the closed lines represent the results of the fits. The crosses label the first and the last point of each branch used in the fit, thus points belonging to the specular peak were omitted. In each fit shown in Fig.1 two independent roughness-amplitudes are assumed, each of which has an exponential autocovariance function. The fit results are summarized in table1. We found out, that it is necessary to assume at least two roughness-amplitudes, in order to obtain reasonable agreement between experiment and theory. Other types of correlation functions were also tested, in particular a Gaussian and a Lorentzian form, but the exponential type gives by far the best results. Strong disagreement between experiment and theory occurs when using a Gaussian function, whereas the application of a Lorentzian gives results very close to those of the exponential function. Also the combination of an exponential with a Gaussian function yields quite unreasonable results. Fig.2 shows ARS - spectra measured on the same sample as in Fig.1 together with the fitted curves. The wavelength is $\lambda = 6.4$ nm. As in the case of Fig.1, a reasonable fit could only be achieved by using a superpositon of two exponential autocovariance functions. As the wavelength becomes shorter, the shape of the spectra changes significantly as can be seen in fig.3a-b. The curve in fig.3a, measured at $\lambda = 2.6$ nm wavelength and $\theta_i = 84°$ angle of incidence, reveals a pronounced bump on the right side of the specular peak. The position of the centre of this bump is at the critical angle of reflection $\theta_c = 86.5°$ of the material. The spectrum taken at $\theta_i = 85°$ (see fig.3b) also shows a bump in the vicinity of the critical angle θ_c, but somewhat less pronounced as in the case of fig.3a.

The 2nd spectrum in fig.3b, which was measured at an angle of incidence equal to the critical angle ($\theta_i = \theta_c$), has no anomalous features. Its angular shape is similar to those of the spectra taken at longer wavelengths (see fig.1-2).

In evaluating the spectra at $\theta_i = \theta_c = 86.5°$ the same fit criteria were applied as in the cases of fig.1-2, assuming light scattering from a rough surface, which is generated by two independent roughness amplitudes. When trying to evaluate the data of fig.3a in the same way, systematic deviations between experiment and theory (– · – · – line) occur, especially at scattering angles in the vicinity of the critical angle θ_c. The value of the reduced χ^2_{red}-function, which is a measure of the fit quality, in this case is 2.57. We tried to explain these deviations by assuming dielectric light scattering from the interior of the sample. It is not completely unexpected that with very small roughnesses other sources of scattering could come into play. Therefore Elson's equations for dielectric scattering were adopted within the fit procedure assuming statistical independence between rough surface and dielectric scattering. The dielectric perturbation is considered by a single fluctuation amplitude. The fit result is represented by the solid line in fig.3a. Here the χ^2_{red}-value is 0.35, indicating a significant improvement of the fit. The data measured at $\theta_i = 85°$ were evaluated in the same way, resulting in values for δ_r, τ_r, ζ_d, and τ_d [2] in agreement with those from the data taken at $\theta_i = 84°$. Furthermore, the surface roughness parameters obtained from the different spectra shown in fig.3a-b agree. The ARS-distributions of the quartz-glass and the SiC-mirrors were also evaluated as described. In all cases quite encouraging agreement between Elson's Vector perturbation theory and the experimental data was obtained. As in the case of the zerodur-glass sample, it is necessary to consider two roughness amplitudes, each of which has an exponential autocovariance function. In no case the

[2] Here r, d refer to roughness and dielectric scattering respectively

application of a Gaussian function leads to reasonable results. The two latter samples also reveal pronounced anomalous scattering when λ becomes short and it is also necessary there to assume dielectric scattering in order to explain the straylight spectra. We also tried to use Beckmann's scalar light scattering theory in evaluating the ARS - measurements in connection with different types autocovariance functions. In all cases a strong disagreement between theory and experiment occurs, especially when λ becomes short and anomalous scattering arizes. The reason of this disagreement may be the following : since Beckmann involves the tangent plane approximation, the surface profile has to be a very gently varying function along the zero-mean-plane, in order to meet the theoretical condition. But in our measurements, we found evidence for exponential autocorrelations functions. Surfaces, which are described by exponential autocorrelation functions, reveal profiles very jagged, rather than having gentle undulations [18]. A detailed comparison between data derived from the vector- and the scalar scattering theory will be published in a separate paper.

5. Summary

In conclusion, we have demonstrated that statistical parameters characterizing a moderately rough surface can be determined by means of ARS-measurements in the XUV - spectral region. Excellent agreement between experiment and Elson's vector perturbation theory for roughness scattering was obtained. We found, that rough surfaces have to be described by at least two statistically independent roughness amplitudes. The most appropriate type of autocorrelation function was found to be an exponential; the assumption of a Gaussian seems to be inadequate. Values of the rms-roughnesses and of the autocorrelation length determined at different angles of incidence and wavelengths are in good agreement for each individual sample. Furthermore, we found evidence for light scattering from dielectric fluctuations in applying Elson's dielectric scattering theory to our experimental data. Thus ARS-measurements in the XUV - spectral region seem to be a very useful method in characterizing optical surfaces, since it is sensitive to roughness features with lateral separations in the submicron region.

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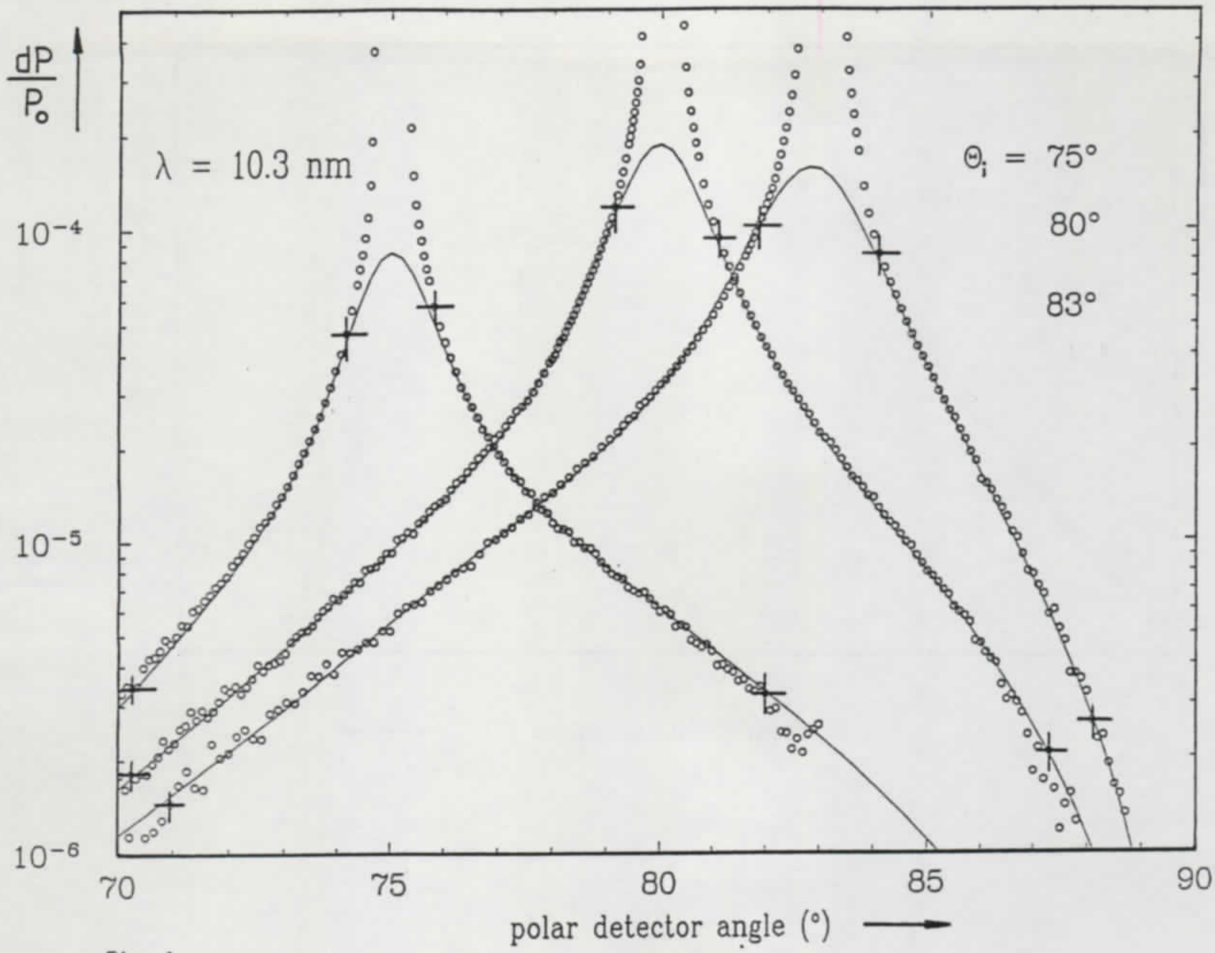
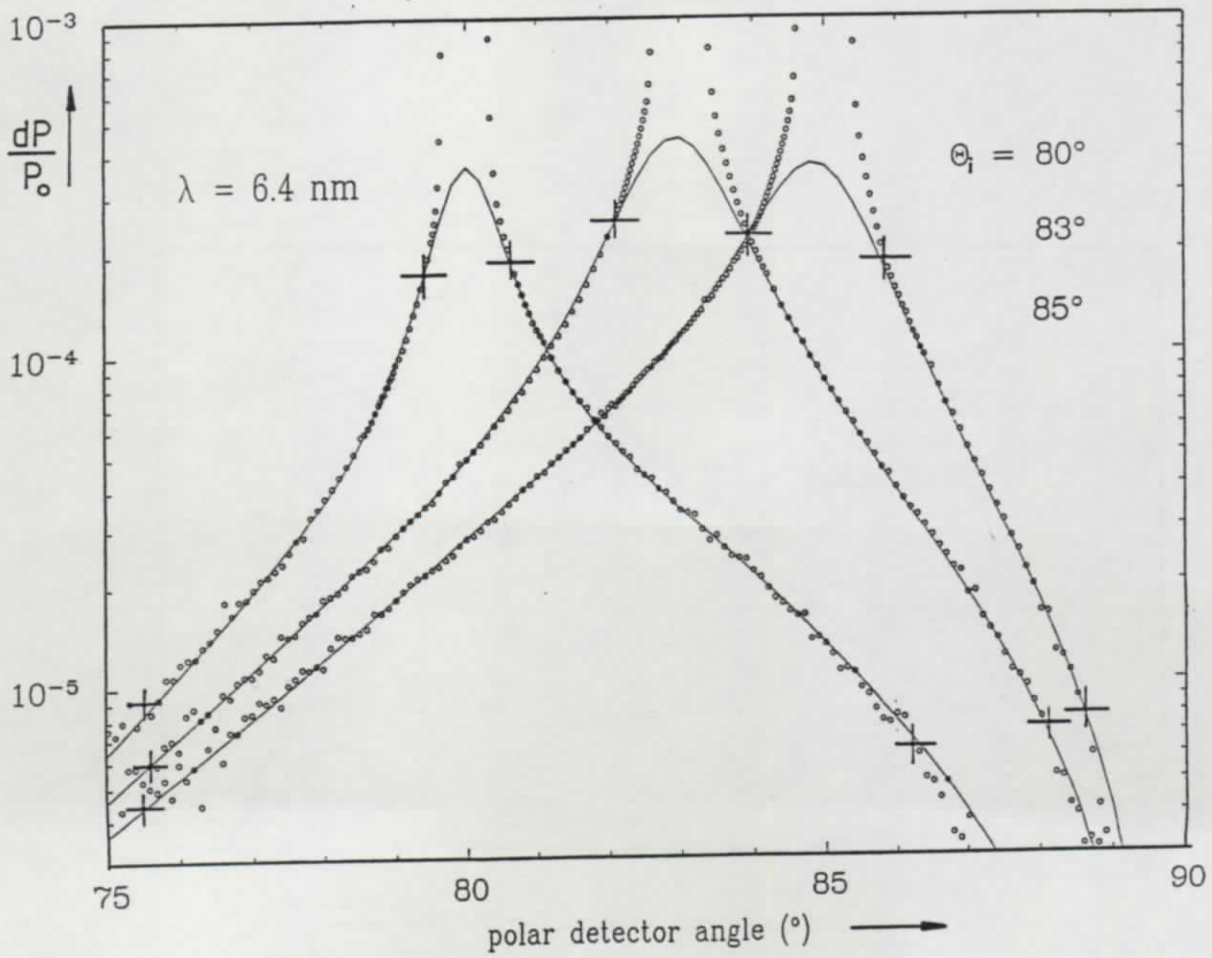


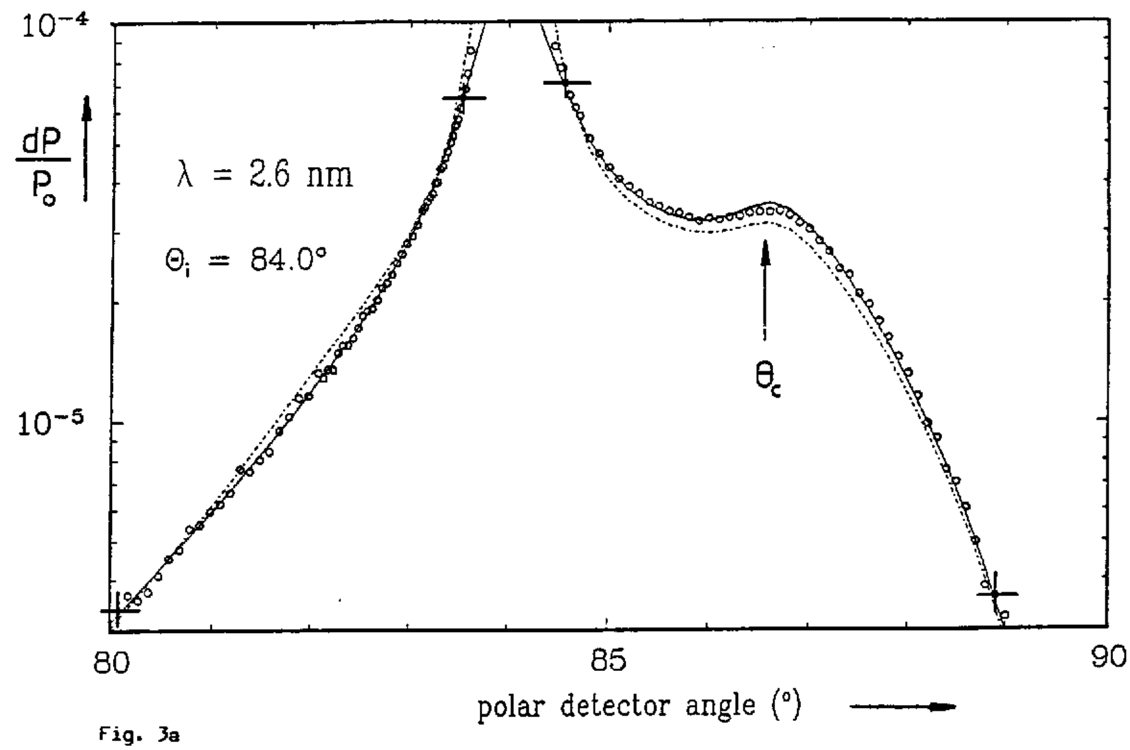
dP/P₀
λ = 10.3 nm
Θᵢ = 75°
80°
83°
10^{-4}
10^{-5}
10^{-6}
70
75
80
85
90
polar detector angle (°)

Fig. 1



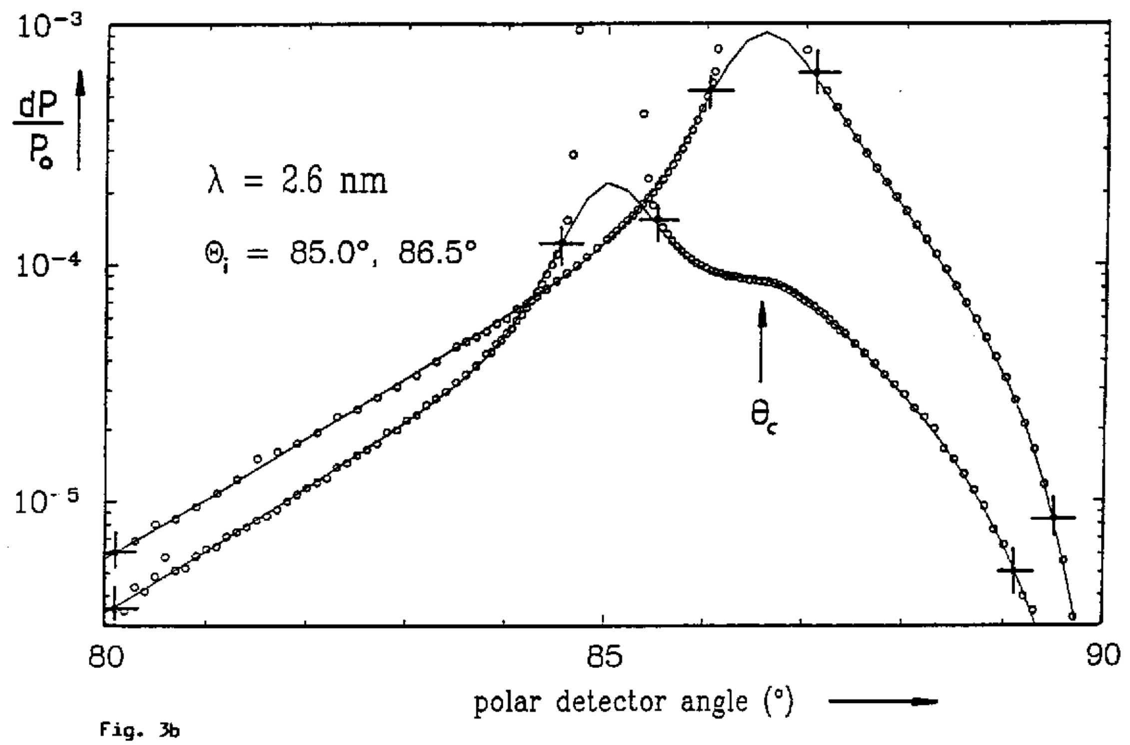
10^{-3}
dP/P₀
λ = 6.4 nm
Θᵢ = 80°
83°
85°
10^{-4}
10^{-5}
75
80
85
90
polar detector angle (°)

Fig. 2



10^{-4}
$\frac{dP}{P_o}$
λ = 2.6 nm
Θ_i = 84.0°
θ_c
10^{-5}
80
85
90
polar detector angle (°)

Fig. 3a



10^{-3}
$\frac{dP}{P_o}$
λ = 2.6 nm
Θ_i = 85.0°, 86.5°
10^{-4}
θ_c
10^{-5}
80
85
90
polar detector angle (°)

Fig. 3b