V44 X-ray reflectrometry

Abstract

Studying interfaces and their properties with the help of X-ray reflectivity is an important topic of current research in solid-state physics. X-ray reflectivity offers the possibility of obtaining structural information about a layer on the surface of a sample that is only a few nanometer thick. The scattering image provides information about the thickness, the electron density and the roughness of a thin layer. Layers on solids as well as on liquid substrates can be studied. In this experiment, the density, roughness and layer thickness of a polysterol film on a silicon wafer are to be determined.

References

[1] Heinz Kiessig, *Interferenz von Röntgenstrahlen an dünnen Schichten*, Annalen der Physik **402** (1931), pp 769-788.

[2] Jens Als-Nielsen and Des McMorrow, *Elements of modern X-ray physics*, John Wiley & Sons, 2011

[3] L.G. Paratt, *Surface Studies of Solids by Total Reflection of X-Rays*, Phys. Rev. **95** (1954), pp 359-369.

[4] Metin Tolan, *X-Ray Scattering from Soft-Matter Thin Films* Material Science and Basic Research, Springer Verlag (1999).

[5] B.L. Henke, E.M. Gullikson and J.C. Davis, Atomic Data and Nuclear Data Tables **54** p. 181-342 (1993)

Preparation

In order to understand all aspects of this experiment, it is necessary to work on the following questions:

- How is X-ray radiation produced in an X-ray tube?
- What is a Göbel mirror and how does it work?
- How is the wave vector transfer defined in an X-ray reflectrometry experiment?
- What are Fresnel´s formulae and what do they describe? What is the special feature of Fresnel´s formulae in the case of X-rays?
- What are Kiessing oscillations and how do they occure?

- How does the Parratt algorithm work (qualitatively)?
- How must the Parratt algorithm be modified to apply to rough surfaces?

Experimental setup and Adjustment

The X-ray reflectivity measurements are carried out with the D8 laboratory diffractometer from Bruker-AXS. It is a so-called $\theta - \theta$ laboratory diffractometer designed for routine use in which the X-ray tube and the detector can be rotated around the sample. The X-rays are emitted by a copper anode tube operated with a current of 35 mA and a voltage of 40 kV.

Adjustment of the D8 Diffractometer

The program **XRD Commander** controls the diffractometer for adjusting the sample and for collecting data. The most used mask is the **adjust mask** seen on Figure 2. On the top there is a toolbar with the **Move-Drive, Init-Drive** and **Zi** buttons as well as a button to change the scales of the **Chart Area**. On the left side of the **adjust mask** are the **Motor Drives Controls** and **Generator Controls**. On the bottom are the **Scan Controlls**.

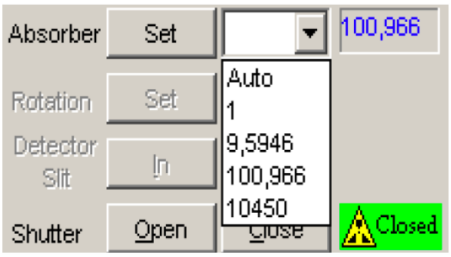

Figure 1: Absorber control mask menu.

In order to protect the detector from damage, the **Absorber** must first be set to **Auto** in the drop down menu (Figure 1). Don‘t forget to press the Set button after selecting **Auto**. After activating the absorber, the geometry of the set up has to be adjusted by positioning X-ray beam, sample and detector.

The position of the sample can be changed in **X, Y** and **Z** direction by using the **Motor Drive Controls**. Also the position of the detector relativ to the tube is changed by driving the 2 Θ motor. The motors are moved by typing the desired position into the **Requested** value field and setting the checkbox nextby.

> For example: The X-ray tube and the detector are moved to position 0
> The value 0 is first typed in the corresponding requirement field (2 Θ) field and the checkmark on the right of the field has to be set. In most situations, the program should set the corresponding checkmark by itself, so that it does not have to be set manually. After pressing the **Move-Drives** button, the motors move to the desired position.

Attention: Do not mix up the button **Init-Drives** whith the **Move-Drives** button. Please **NEVER** press the **Init-Drives** button, because this button moves the motor back to their reference position and they have to be adjusted again from the beginning.

In order to be able to measure reflectivity, the sample must be brought into the center of rotation of the diffractometer and the sample surface must be aligned parallel to the X-ray beam. To do this there are three **Scantypes** available: the **Rocking Curve**, the **Detector Scan**, and the **Z-Scan**. The measuring method can be selected via drop down menu **Scantype** on the bottom of the **adjust mask** (Figure 2). In addition, the scanning ranges and the scan speed must be adjusted to each scantype. After performing the adjustment scans, the data

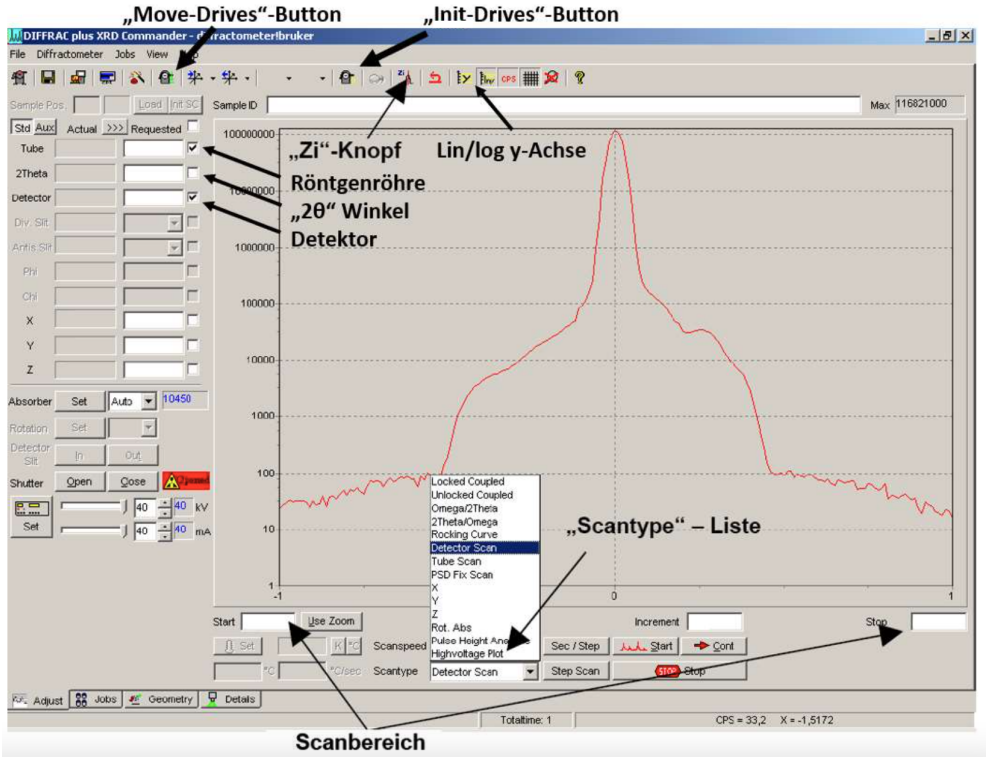

Figure 2: Adjust mask of **XRD Commander**.

should be saved in raw format, as they are still needed for later analysis.
At the beginning of the adjustment, the sample must be moved out of the beam by changing the Z-coordinate (Figure 3). Furthermore, both the tube and the detector must be moved to an angle of 0°.

Adjustment of the primary beam
To adjust the primary beam, a **detector scan** has to be run. The detector moves in a small angular range around the expected position of the X-ray beam. The resulting scan should be similar to a Gaussian distribution (Figure 5). The position of the maximum in 2θ should represent the new zero position of the detector. By pressing the **Zi button** (Figure 2) in the toolbar, the centre of gravity of the measured peak is determined. At the same time, the input window **ZI Determination** appears (right picture in Figure 3), via which the theoretical zero position is sent to the diffractometer. For this purpose, the value **0** is typed in the field **Enter theoretical position** and the button **save and send new Zi** is pressed.

Ajustment of the sample position
Following the alignment of the primary beam, the sample should be roughly positioned into the center of the Diffractometer by moving the sample table using the X, Y, and Z coordinates with stepper motors (left picture in Figure 3). The sample is optimally adjusted when it is

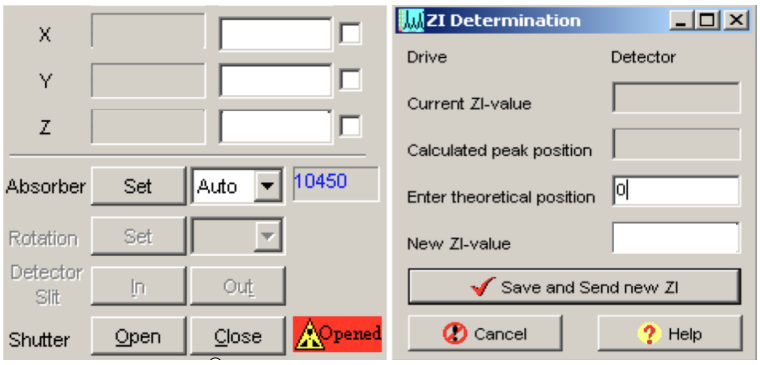

Figure 3: Left: Input mask for the adjustment of the sample in x, y, and z direction. Right: Input mask "ZI Determination" for the recalibration of the zero position.

parallel to the beam and shades half the intensity of the primary beam.
Start with the Z-coordinate using the **Z-scan** (Figure 5). The **Z-scan** shifts the sample in hight and is used to determine the shading of half the intensity of the primary beam. As long as the sample is below the beam, the full intensity I_{max} is measured. The intensity decreases more and more the further the sample is moved into the beam. The Z-value at which the intensity is $\frac{1}{2} I_{max}$ is to be estimated here. For a rough determination of half the shading, double-click on half the maximum intensity in the graph shown. By double-click, the Z coordinate automatically adopts the corresponding value.
If the sample is within the range of the beam along the X-coordinate, it must cover approx-

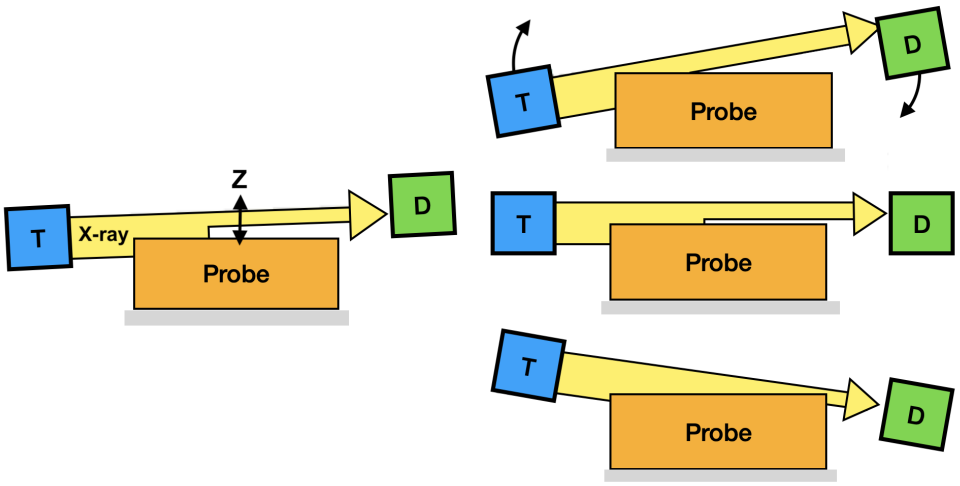

Figure 4: Left: The sample shades half the intensity of the primary beam after the use of a Z-scan. However, the beam may not be parallel to the sample surface. Right: The Rockingscan should help to correct the possible misalignment by determining the maximum intensity.

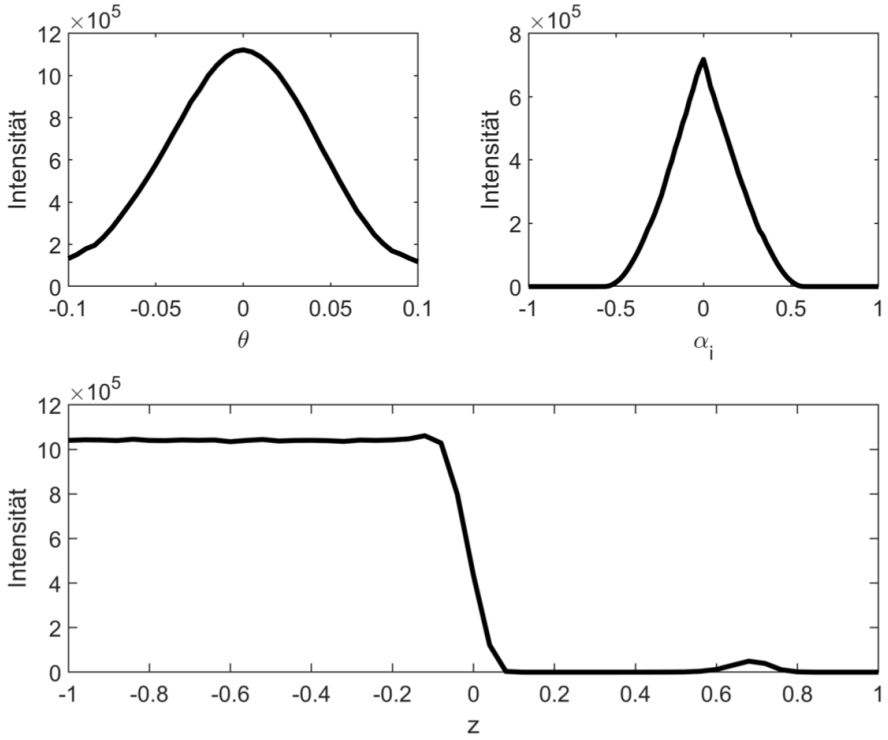

Figure 5: Example measurements of different **Scantypes**. Top left: Detector scan, Top right: Rockingscan at $2\Theta = 0$; Bottom: Z-scan.

imately half of the primary beam to be in the correct position. To ensure that the sample is correctly positioned along the X coordinate, an **X-Scan** is performed. This scan should result in a plateau of lowered intensity within which you are free to choose your position depending on which area of the sample you wish to examine.
If the sample is adjusted in the X and Z directions and the intensity is $\frac{1}{2}\, I_{max}$, a **Rockingscan** is performed (Figure 4) to adjust the Y-coordinate (in beam direction). The X-ray tube and the detector rotate araound the sample so that the angular sum $\alpha_i + \alpha_f = 2\,\theta$ remains constant. This scan corresponds to a rotation of the sample in the beam. On the one hand it provides information about the tilt of the sample relative to the X-ray beam. On the other hand it can be used to bring the sample in Y-direction to the center of rotation of the diffractometer. The measurement should result in a symmetrical triangle. In reality, the triangle can be asymmetrical and also the maximum value to the intensity is not reached at 0°, but at a different value. There is still a possibility that the rocking scan will produce a plateau. This indicates a misalignment of z in combination with a strong tilting of the sample. In the first case (asymmetry), the beam does not hit the sample exactly in the middle (i.e. the sample is not in the centre of rotation of the diffractometer). If the triangle is asymmetrical and e.g. the slope on the left side is flatter than on the right side, the Y-coordinate must be increased (and vice versa). In the second case, the angular orientation of the tube in relation to the

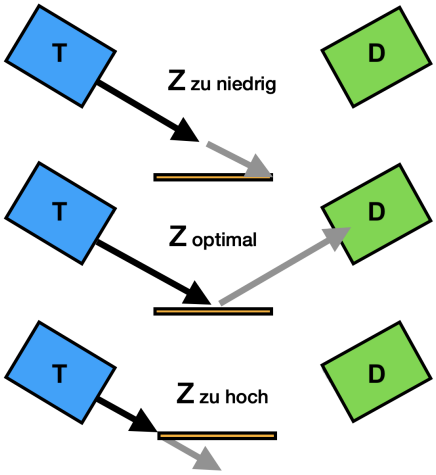

Figure 6: Z-Scan under a angle of $2\Theta = 0.3°$.

sample surface is still not correct. As soon as the triangle is symmetrical, a double-click on the maximum determines it position and transmit it to the motors. With **Move-Drives**, the motors can be moved to the correspondng positions. The X-ray tube and the detector are now in the position where the Rockingscan has detected the maximum intensity. This position is normally not exactly at $\Theta = 0$. Due to the parallel alignment of the X-ray beam, the sample is no longer centered in the beam. For this reason, a new Z-Scan must be performed to bring the sample back to half the shading.
A second Rockingscan is then performed at an angle of $2\,\theta = 0.3°$ to ensure even more precise adjustment of the sample in the beam. If a clear reflex can be seen, the angle of incidence and emergence can be set to 0.15° by pressing the **Zi** button, the value **0.15** is entered in the **Enter theoretical position** field.
Finally, the half shading can be determined more precisely from the third **Z-scan**, as it can be read from the maximum of the intensity curve. Accordingly, the intensity of the primary beam is completely reflected in a certain height range Δz. In the following, a Z-scan is now run at an angle (Figure 6). The scan range should be close to the zero point. The maximum is determined by double-clicking on the centre of gravity of the curve and moving to the corresponding Z position. This adjusts the sample.
For a more detailed adjustment, an additional Rockscanning is run at a higher angle of $2\,\theta = 0.5°$. Also in this case, by operating the **Zi** key and by entering the value **0.25** in the **Enter theoretical Position** input field, the angle of incidence and angle of reflection can be calibrated.
Table 1: Summary of the alignment measurements with all important values.

Type	Range	Step size	Measuring time per mesuring point [s]
Detector scan	-0.5 to 0.5	0.02	1
Z-Scan	-1 to 1	0.04	1
X-Scan	-20 to 20	1	1
Rockingscan $2\theta = 0$	-1 to 1	0.04	1
Z-Scan	-0.5 to 0.5	0.02	1
Rockingscan $2\theta = 0.3$	0 to 0.3	0.005	1
Z-Scan $2\theta = 0.3$	-0.5 to 0.5	0.02	1
Rockingscan $2\theta = 0.5$	0.2 to 0.5	0.005	1

The reflectivity of the polymer coated silicon wafer can now be measured. The angle of incidence α_i on the sample and the angle between sample surface and detector α_f are the same for such a scan. The scan type **Omega/2Theta** is used for this. A scan range from 0° to 2.5° can be selected for the measurement. For this range, the step width and the time per step must also be specified. For the step widths, 0.005° is suggested. The measuring time should be at least 5 s per step. To obtain the **true reflectivity** a **diffuse scan** which measures the diffusely scattered radiation, must be carried out. For this reason, a second measurement is started in which the detector angle is shifted by 0.2° relative to the angle of incidence. The same step size should be used. All data should be saved in *raw* format and converted using the program **File Exchange**. The files can then be saved on a USB stick (the participants **have brought with them**). This completes the measurement process of the polymer film on a silicon wafer and the data can be analysed.

The geometry factor

The geometry factor G (Figure 7) takes into account that only at a sufficiently large angle of incidence, the geometry angle α_g of the full beam hits the surface. With a diameter D of the sample surface and height d_0 of the beam the following applies for the geometry angle

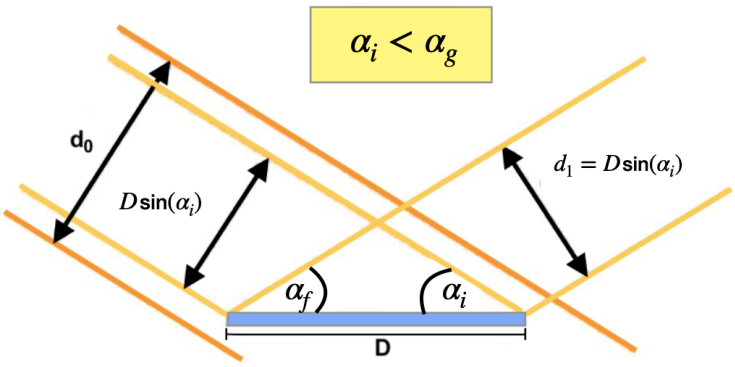

Figure 7: Beam path showing the geometry.

$$\begin{aligned} G &= \frac{D\,\sin(\alpha_i)}{d_0} && \text{if} \quad \alpha_i < \alpha_g \\ G &= 1 && \text{if} \quad \alpha_i > \alpha_g \end{aligned} \tag{1}$$

In this case the angle of incidence α_i are initially very small. Consequently the area swept by the beam is larger than the sample surface. Thus, not all of the irradiated intensity is reflected from the sample surface and can enter the detector. This effect results in a drop in reflectivity in the range of very small angles $\alpha_i < \alpha_g$. The geometry factor G takes this efect into account and is defined as a ratio of the beam width $D\ \sin(\alpha_i)$ that hits the sample surface to the total beam width d_0 [3].

Measurement and analysis

In oder to determine the dispersion and the roughness of the silicon wafer (D = 20 mm) as well as the layer thickness, dispersion, and roughness of the polysteroal layer, proceed as follows:

- Fit a Gaussian function to the detector scan and thereby determine the half width and maximum intensity in the detector scan.
- Plot the measured values of the reflectivity scan and the diffuse scan in a graph. Substract the diffuse scan from the reflectivity scan and present the result in a diagram. In addition, draw the Fresnel reflectvity of an ideally smooth silicon surface in the diagram.
- Calculate the geometry angle α_g of the examined sample from the data you recorded during the adjustment! Derive the correction factor G and correct the recorded data with it. Calculate the geometry angle from the beam width and the sample length and compare it with the measured one.
- Estimate the layer thickness via the recorded reflection curve.
- Calculate the dispersion of silicon and polysterol from the literature values and compare them with your results. Literature values[5]:
  Silicon: $r_e\rho = 20 \cdot 10^{10}\ cm$, $\delta = 7.6 \cdot 10^{-6}$, $\alpha_c = 0.174°$
  Polysterol: $r_e\rho = 9.5 \cdot 10^{10}\ cm$, $\delta = 3.5 \cdot 10^{-6}$, $\alpha_c = 0.153°$
- Calculate the position of the critical angle and compare it with your measurement.
- Determine the dispersion of the sample under investigation using the parrat algorithm. Use modified Fresnel coefficients to include the roughness. You can use a program of your choice for this (include program code as an attachment). Don´t forget error analysis.
  Note: Please note that the sign in front of the complex part in the refraction index ($n = 1 - \delta \pm i\beta$) depends on the definition of your wave equation.
- Compare your estimate of layer thickness with the value determined by the Parratt algorithm.