X-RAY OPTICAL CONSIDERATIONS ON PARABOLIC GRADED MULTILAYERS ON THE DIFFRACTED BEAM SIDE IN X-RAY DIFFRACTION

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ABSTRACT

Parabolically curved graded multilayer mirrors, so-called Göbel mirrors, have opened up new applications as x-ray optical devices. They are applied as condensing reflectors to convert divergent x-rays from laboratory x-ray sources into a parallel beam. The use of such graded multilayers on the diffracted beam side of x-ray scattering experiments is an interesting alternative to the state of the art analyzer crystals. As medium resolution analyzers in triple-axis diffractometry, graded multilayers allow to distinguish between different real structure effects in epitaxial sample systems. For the application on the diffracted beam side in parallel beam optics, the resolution properties of the parabolically curved graded multilayer mirror have to be examined. Therefore, a new method has been developed to prove the focussing properties of adjacent area elements on the mirror. As medium resolution analyzers with high intensity throughput, parabolically curved graded multilayers are ideally suited to cover the range of resolution limited by soller slits on the low-resolution side and by crystals on the high-resolution side. The performance of mirrors applied as analyzers is demonstrated on epitaxial GaN layers and compared with the performance of an analyzer crystal.

1. INTRODUCTION

Quality and structural perfection of epitaxial films are mostly investigated by x-ray diffraction techniques. The real-structure effects in epitaxial samples (crystal curvature, layer thickness, lateral granularity, mosaicity, variations in d spacing) result in angular shifts and angular broadening of lattice Bragg-peaks [1-3]. Generally, the instrumental resolution shows up as an additional broadening of peaks for non-perfect crystals. Without going too much into detail, the instrumental resolution function needs to be deconvoluted from the diffraction pattern from such samples. Essentially, the instrumental resolution is determined by the spread of wavelength $\Delta \lambda$, the angular divergence of the primary beam and the angular acceptance of the analyzer. For a nearly parallel and monochromized ($\Delta \lambda/\lambda = 2.2 \times 10^{-5}$ for $\lambda = 0.154056\text{nm}$ CuK$\alpha_1$) primary beam, the instrumental broadening is dominated by the angular acceptance of the analyzer. To decrease the angular acceptance on the receiving side, one can use narrow slits [4] or soller slits with a minimum acceptance of approx. 0.07°. To increase the resolution for real structure effect separation further, one uses an analyzer crystal with low acceptance [5-9], e.g. 0.0033° (Ge 022). For samples with poor reflectance, the intensity losses by an analyzer crystal may be too high. Therefore, the counting time needs to be increased. The high resolution of an analyzer crystal can lead to another drawback: sometimes, the goniometer alignment is not quite correct. Due to the small reflection width of an analyzer crystal, lattice reflections are hard to discover. For samples that allow to dispense with highest resolution to determine the real structure, analyzer mirrors combine sufficient resolution with a high gain of intensity.
This document was presented at the Denver X-ray Conference (DXC) on Applications of X-ray Analysis.

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2. MIRROR TEST METHOD

Fig. 1 displays a schematic view of the mirror test method. A parallel, monochromized x-ray primary beam ($\lambda=0.154056\text{nm}$) incides onto the parabolically curved, graded multilayer mirror. The purpose of the slit with variable position $x$ in front of the multilayer mirror is to cut out a part $l_0$ of the primary beam, usually $l_0 \approx 20\text{\mu m}$. The graded multilayer mirror is adjusted to maximum reflection, i.e. $\alpha=\Theta_{\text{Bragg}}$. For the mirrors investigated, $0.9^\circ<\Theta_{\text{Bragg}}<1.5^\circ$. As a result, the illuminated length $l_j$ of mirror element $j$ varies within $770\text{\mu m} \ldots 1270\text{\mu m}$ for $j=1 \ldots N$. The shift of the slit position along $x$ results in a shift of the illuminated area on the mirror. For every mirror element $j$, the angle of incidence $\alpha$ is changed within $\Theta_{\text{Bragg}}-0.216^\circ < \alpha \leq \Theta_{\text{Bragg}}+0.216^\circ$, and the focussing properties of the mirror elements are recorded by the detector. Hereby, the angle between detector and multilayer surface remains fixed. A second slit in front of the detector with a slit width of $30\text{\mu m}$ is aligned to the focus of the parabola. Its purpose is to separate the specular reflected intensity and to prevent the diffuse scattering from reaching the detector. The test provides position sensitive information on the reflectivity and focussing properties of adjacent mirror elements. This method is close to experimental conditions, where analyzer mirrors are fixed to the 2$\Theta$ circle of the goniometer. The situation in reciprocal space is shown in Fig. 2. $q_x$ and $q_z$ denote the vertical and horizontal wavevector transfers with respect to the multilayer mirror surface. The Bragg reflection of the analyzer mirror is given as a shaded ellipse on the $q_z$-axis. Its width in $q_z$ direction is given by the material combination of the multilayer system and the number of multilayers as well as their vertical perfection. Its width in $q_x$ direction is given by the lateral perfection of the multilayer [10]. Applying an $\alpha$-scan, the Bragg reflection is crossed in $q_z$ and $q_x$ direction in reciprocal space in the manner shown in Fig. 2. Therefore, the resolution of the analyzer mirror is determined by the material combination of the multilayer and its vertical and lateral perfection.

Fig. 3 shows the simulation of the reflectivity pattern of adjacent mirror elements. The left-hand side of Fig. 3 gives the profile of a parallel, monochromized beam. In the form of a trapezoid, the beam profile is the best approximation for many experimental situations (see Fig. 4). The profile is recorded versus $2\Theta$. Here, $2\Theta$ refers to the angle of the 2$\Theta$ circle, i.e. detector circle, of the goniometer (see setup, Fig. 7). The primary beam profile can be measured by attaching a narrow slit to the detector and applying a $2\Theta$-scan, i.e. detector-scan,
over the primary beam. Here, we want to emphasize particularly that the primary beam profiles recorded versus 2θ in Figs. 3 - 6 do not stand for any angular divergence of the primary beam. Experimentally, it is an easy to realize way of primary beam profile measurement, and the plot versus the 2θ scale is quantitatively consistent with the (Θ_{Bragg}-α) scale of the reflectivity patterns in Figs. 3 - 6. The plot of the reflected intensity of the mirror versus (Θ_{Bragg}-α) and versus mirror element j gives the horizontal intensity stripe that shows up in the middle of Fig. 3. For (Θ_{Bragg}-α)=0 - the angle of incidence fulfills the Bragg-condition α-Θ_{Bragg} - every mirror element j shows maximum reflectivity. For α deviating from Θ_{Bragg}, the reflected intensity decreases rapidly as can be concluded from the α-scan shown in Fig. 2. The acceptance profile of the analyzer mirror is given on the right-hand side of Fig. 3. Its FWHM gives the angular acceptance Δα of the mirror. Δα is crucial for x-ray diffraction. It determines the resolution of the analyzer. The acceptance profile can be obtained by adding up the reflected intensities of all mirror elements, or by applying a α-scan across a parallel, monochromatized beam.

Fig. 4 exhibits the experimental data obtained from a W/BaC parabolically curved, graded multilayer mirror. The mirror presented in this chapter may not be confused with mirrors A and B in chapter 3. The focal length f₀ was 90mm. The Bragg-angle Θ_{Bragg} of the multilayer ranged from 1.466° to 1.476°. The primary beam from the 1.5kW Cu x-ray tube was parallelized by a collimator mirror and coupled into a Ge 022 (+−−−−−−) channel-cut monochromator (see setup, Fig. 7). For CuKα1, the monochromator had a divergence in the scattering plane of 0.0033°. At this stage of experiment, the angular beam width, i.e. FWHM, was 0.1°, which corresponds to a width of 0.48mm. The mirrors under investigation were dimensioned to match a beam width of 1mm. Therefore, the primary beam profile was further broadened by a single Ge 011 reflection. After broadening, the angular width was 0.24°, which corresponds to a width of 1.15mm. A variable precision-slit (Fig. 1) was used to cut out 20μm of the primary beam. With a mean Bragg-angle of 1.471°, a mean length of ⟨l⟩=780μm was illuminated on the mirror. The reflectivity pattern in Fig. 4 is similar to that displayed in Fig. 3. However, there are deviations from ideal conditions. The primary beam profile is no ideal trapezoid. The intensity streak on the top right-hand side of the reflectivity pattern is caused by deviations from the parabola of the multilayer mirror. Nevertheless, the maximum intensity gathers at α=Θ_{Bragg} for the mirror elements j=1...45, as predicted by the simulation in Fig. 3. The acceptance profile shown on the right-hand side has an angular acceptance Δα of 0.025°. Obviously, the resolution of a parabolically curved, graded multilayer applied as analyzer cannot compete with the resolution of an analyzer crystal, but without doubt, it is an improvement in resolution compared to soller slits. The main advantage of mirrors used as analyzers, intensity gain provided by x-ray condensation, will show up in chapter 3.

Fig. 5 gives the same situation as Fig. 3: The reflection properties of adjacent mirror elements are proved step by step with the primary beam. In contrast to Fig. 3, the beam profile is not broadened. It is smaller than the width the mirrors under investigation are dimensioned for. The simulation of the reflectivity pattern shows a small distortion of the intensity streak. As a result, the angular acceptance width Δα of the acceptance profile broadens a little bit. The primary beam profile has a trapezoidal shape. The acceptance of the mirror elements is determined by the perfection of the graded multilayer, as mentioned previously. Besides deviations from the parabola and mismatches of the mirror alignment, the intensity gradient on both sides of the trapezoid plateau can lead to the distortion of the reflectivity profile in Fig. 5. As a result, the acceptance profile of the analyzer mirror broadens.

Fig. 6 displays the measurement. The beam broadener Ge 011 crystal was taken away. The primary beam had an angular width at FWHM of 0.1° corresponding to a width of 0.48mm. The measurement in Fig. 6 confirms the simulation in Fig. 5. Deviations from the
parabola curvature can be ruled out. This was confirmed by an optical determination of the curvature. The angular acceptance $\Delta \alpha$ of the acceptance profile increases to 0.042°. In contrast to Fig. 4, the maximum number $N$ of mirror elements was 36. This is attributed to the smaller illumination length of x-rays on the multilayer mirror caused by a smaller projection of the narrow primary beam profile. To avoid changes of the focussing properties of analyzer mirrors, the primary beam width and the accepted beam width (here: 1mm) of the analyzer mirror need to be matched with each other. This guaranties the full illumination of the mirror.

Fig. 3. Simulation of the reflectivity pattern of adjacent mirror elements. The angular width of the acceptance profile gives the resolution of the multilayer mirror applied as analyzer.

Fig. 4. The experimental determination of the reflectivity pattern of a W/B$_4$C multilayer mirror. The angular acceptance is 0.025°.
Fig. 5. Simulation of the reflectivity pattern of adjacent mirror elements. Here, the primary beam profile is not broadened.

Fig. 6. The experimental determination of the reflectivity pattern of a W/B₄C multilayer mirror. The angular acceptance increases to 0.042°.

3. APPLICATION OF GÖBEL MIRRORS AS ANALYZERS

The performances of two different multilayer mirrors (denoted A and B) with different angular acceptances are compared to that of a Ge 022 (+ − +) analyzer crystal. The sample used to compare the analyzers is a GaN 001 layer grown on c-plane sapphire, Al₂O₃ 001, by
metal organic chemical vapor deposition (MOCVD). The samples were provided by Walter-Schottky Institut, Munich.

3.1. Experimental Setup

The schematic setup of the diffractometer is given in Fig. 7. On the left-hand side, the x-ray source provided a 8mm x 0.04mm line focus at a 6° take-off angle. The line focus was positioned in the focus of the collimator mirror. The collimator mirror converted the divergent x-rays from the line focus into a parallel beam [11, 12]. CuKα1 and CuKα2 are preselected by the mirror. This is discussed in detail in ref. [13, 14]. Parallel-beam coupling into a Ge 022 monochromator by means of a parabolically curved, graded multilayer mirror revealed an intensity gain based on the exploitation of a larger exit angle range of the x-ray tube. On the diffracted beam side, the analyzer mirrors (A and B) were mounted on the 2θ circle of the goniometer. Together with the scintillation counter and a slit aligned to the focus of the mirror, the mirrors replaced the Ge 022 analyzer crystal. The slit

![Fig. 7. The "medium-resolution diffractometer" using a parabolically curved, graded multilayer mirror as analyzer on the diffracted beam side. The incident-beam side mirror acts as collimator for the Ge 022 monochromator.](image)

![Fig. 8. The acceptance profiles of the analyzer mirrors A and B. The profile of the analyzer crystal is not shown. The acceptance angles are displayed in the insets.](image)
preceeding the detector had a width of 30\,\mu m. Fig. 8 displays the acceptance profiles of the mirrors A and B. Mirror A consisted of 50 layer pairs of Ni/C. Its focal length \( f_0 \) was 150\,mm. The acceptance angle \( \Delta \alpha \) of mirror A was found to be 0.013°. Analyzer mirror B was fabricated from 50 layer pairs of W/B\textsubscript{4}C with a focal length \( f_0 \) of 90\,mm. Its acceptance angle \( \Delta \alpha \) was 0.032°. The profile of the analyzer crystal Ge 022 is not shown in Fig. 8. Its FWHM was 0.0033°. The acceptance profiles are normalized to the maximum intensity. For both mirrors, the acceptance profile has a Gaussian shape. The better resolution, i.e. the smaller angular acceptance, of analyzer mirror A in contrast to analyzer mirror B is attributed to the smaller width of the Ni/C multilayer Bragg-reflection. Additionally, the vertical and lateral perfection of the Ni/C multilayer structure is improved.

### 3.2. Comparative measurements on GaN

Fig. 9 shows \( \Omega/2\Theta \)-scans at the GaN 002 reflection using the analyzer mirrors A and B and the crystal analyzer Ge 022. The measured intensity is normalized by the maximum intensity and plotted on a linear scale versus \( 2\Theta-2\Theta_{002} \). \( 2\Theta_{002} \) is 34.58°. The FWHM using a crystal analyzer is found to be 0.013°. Close to that result is the FWHM of the GaN 002 reflection obtained by analyzer mirror A. In contrast to the maximum intensity of 6500\,cps obtained with the analyzer crystal at the 002 reflection, the maximum intensity using mirror A was 22000\,cps.

![Graph](image)

**GaN 002**

**Fig. 9.** \( \Omega/2\Theta \)-scans at the GaN 002 reflection using analyzer mirrors A and B and the crystal analyzer Ge 022.

The \( \Omega \)-scans at the 002 reflection of GaN are given in Fig. 10. \( \Omega_{002} \) is 17.29°. Here, the profiles of the \( \Omega \)-scans using mirrors A and B and the crystal analyzer match each other. The FWHM values of the profiles are within 0.115° ... 0.123°. In Figs. 11 and 12, the scattering profiles of the weak asymmetric GaN 105 reflection are measured. \( 2\Theta_{105} \) is 105.09° and \( \Omega_{105} \) is 31.9°. Both analyzer mirrors A and B and the crystal analyzer provide nearly the same scattering profile. For the mirrors, the FWHMs of the GaN 105 reflection for the \( \Omega/2\Theta \)-scans...
are 0.067°. With analyzer crystal, the FWHM is 0.062°. As can be seen from the statistical fluctuations of the scattering curve measured with a crystal, the intensity of the asymmetric reflection was comparably weak. Using mirrors, the intensity throughput to the detector was sufficiently high to provide a regularly shaped scattering profile. For the Ω-scans at the GaN 105 reflection, the scattering profiles obtained with mirrors and crystal are similar to each other. Again, the data fluctuations of the scattering profile are higher for the measurement using the crystal. The FWHMs of the Ω-scans at the 105 reflection are 0.065°.

![Graph](image1.png)

**Fig. 10.** Ω-scans at the GaN 002 reflection using mirrors A and B and the crystal analyzer. The diffraction patterns match each other.

![Graph](image2.png)

**Fig. 11.** Ω/2Θ-scans at the weak asymmetric GaN 105 reflection. For the analyzer crystal, the low intensity throughput leads to deviations from the symmetric shaped diffraction pattern.
4. CONCLUSION

A new test method has been developed for parabolically curved, graded multilayer mirrors that is close to experimental conditions. The test provides position sensitive information on the reflectivity and focussing properties of adjacent mirror elements. The method is sensitive to deviations from the parabola. Its main advantage is its sensitivity to the lateral as well as vertical perfection of the multilayer structure of the mirror. In addition to that, it determines the resolution of mirrors applied as analyzers.

The angular position, diffraction shape and angular broadening of Bragg peaks is determined by the real-structure of the samples. To get reliable results for the real-structure of non-perfect crystals, the instrumental broadening has to be known. Additionally, it has to be small compared to the real-structure induced broadening. This guarantees that the shape and broadening of x-ray scattering profiles is dominated by the sample structure, not by the instrumental resolution. In Figs. 9 - 12 it is demonstrated that parabolically curved, graded multilayers are well suited to improve x-ray diffraction experiments from samples that allow to dispense with highest resolution.

Mirrors combine sufficient resolution with a high intensity throughput. For instance, at the Ga 002 reflection mirror A (50 layer pairs of Ni/C) provided a maximum intensity of 22000cps compared to 6500cps with a Ge 022 crystal analyzer. The intensity gain is advantageous for the measurement of weak lattice reflections from samples. For real structure determination, the angular acceptance of 0.013° applying mirror A was sufficient.

Göbel mirrors are an innovative tool for "medium-resolution diffraction". Dependent on material combination and perfection, resolution as well as intensity throughput of analyzer mirrors can be changed for various requirements. The new approach to use mirrors as analyzers is still in the beginning. For the near future of x-ray diffractometry, the main application will be to cover the range of resolution limited by soller slits on the low resolution side and by crystals on the high resolution side.
ACKNOWLEDGEMENT

We gratefully acknowledge Dr. O. Ambacher from Walter Schottky Institution (Garching, Germany) for the preparation of the MOCVD grown GaN layers. We are indebted to Prof. Dr. U. Pietsch from the Institute of Solid State Physics, University of Potsdam (Potsdam, Germany) for valuable discussions.

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