Applications of New, High Intensity X-Ray Optics - Normal and thin film diffraction using a parabolic, multilayer mirror

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Abstract

Corundum (300) peak intensity from a NIST SRM 1976 plate improved dramatically for narrow samples when using a parabolic, multilayer mirror compared to standard Bragg-Brentano parafocusing optics, but was poorer than Bragg-Brentano optics for wide samples. Peak intensity from the mirror was 5.2 times higher than the for Bragg-Brentano case for a 1.15 mm wide sample. Conversely, the Bragg-Brentano case was superior for large samples, presumably because the greatly decreased irradiated volume due to the narrow parallel beam more than compensated for the increased X-ray flux density from the mirror.

Likewise, compared to both Bragg-Brentano and parallel beam optics, the parabolic, multilayer mirror produced superior peak intensities in grazing incidence X-ray diffraction (GIXRD) studies for small grazing angles on both a 600 Å Pt thin film and a 350 Å TiN thin film. Parallel beam optics with conventional incident beam slits were superior for grazing angles above 3 degrees. The amount of intensity improvement for mirror optics over parallel beam optics with a divergent incident beam increased as the grazing angle decreased, reaching values of 2.6 times at 0.3 degrees for the Pt film and 1.6 times at 0.5 degrees for the TiN film.

Introduction

Several new technologies have recently been proposed with the goal of increasing X-ray intensity on laboratory diffractometers equipped with conventional sealed X-ray tubes. The two principle ones are parabolic, multilayer mirrors and capillary optics utilizing a bundle of polycapillaries. Both approaches attempt to increase beam intensity by capturing a larger solid angle from the anode and directing it toward the sample.

Synthetic multilayer lattices have long been known to have high reflectivity for X-rays and a narrow energy bandpass. These characteristics made them attractive for a variety of X-ray and neutron applications (Schoenborn et al, 1974; Thompson et al, 1989, Barbee, 1986). In fact, Nagel (1982) suggested the possibility of making parabolic monochromators with graded-thickness multilayers to produce a parallel X-ray beam. A similar device was patented by Keem (1985). However, it is only fairly recently that thin film deposition techniques have evolved to the point where it is possible to produce such a device.

We performed two experiments to investigate some of the capabilities of this new tool for XRD analysis. First, we compared the performance of the parabolic, multilayer mirror to Bragg-Brentano parafocusing optics for conventional diffraction analysis as a function of sample size. Finally, we compared mirror, Bragg-Brentano and parallel beam optics with a divergent incident beam in GIXRD studies on thin films.
Experimental

All data was collected with a Scintag X1 Theta:Theta diffractometer. Three hardware configurations were used in these studies: 1) parabolic, multilayer mirror with parallel beam attachment, 2) conventional incident beam slits with parallel beam attachment, and 3) Bragg-Brentano parafocusing optics. These configurations are summarized in table 1. The multilayer mirror was supplied by Seifert Optik and consisted of 50 multilayers on a silicon substrate. The geometric form was parabolic with the multilayer repeat distance varying between 27.4 and 37.1 Å. It's physical size was 80 x 20 x 10 mm and it's peak reflectivity was 75%.

Table 1: Hardware configurations used in this study.

<table>
<thead>
<tr>
<th></th>
<th>Parabolic Mirror</th>
<th>Parallel Beam</th>
<th>Bragg-Brentano</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Diffractometer:</strong></td>
<td>Scintag X1 Theta:Theta</td>
<td>Scintag X1 Theta:Theta</td>
<td>Scintag X1 Theta:Theta</td>
</tr>
<tr>
<td><strong>X-Ray Anode:</strong></td>
<td>Copper</td>
<td>Copper</td>
<td>Copper</td>
</tr>
<tr>
<td><strong>Anode Size:</strong></td>
<td>1 x 10 mm (line focus)</td>
<td>1 x 10 mm (line focus)</td>
<td>1 x 10 mm (line focus)</td>
</tr>
<tr>
<td><strong>Tube Current:</strong></td>
<td>40 mA</td>
<td>40 mA</td>
<td>40 mA</td>
</tr>
<tr>
<td><strong>Tube Voltage:</strong></td>
<td>45 kV</td>
<td>45 kV</td>
<td>45 kV</td>
</tr>
<tr>
<td><strong>Tube Optics:</strong></td>
<td>Multilayer Mirror</td>
<td>0.2 deg. divergence slit</td>
<td>1.4 deg. divergence slit</td>
</tr>
<tr>
<td></td>
<td>2 mm beam width</td>
<td>0.7 deg. scatter slit</td>
<td>(0.2 deg. for GIXRD)</td>
</tr>
<tr>
<td></td>
<td>&lt;0.02 deg. divergence</td>
<td>1.1 deg. Soller slit</td>
<td>(0.7 deg. for GIXRD)</td>
</tr>
<tr>
<td><strong>Anode-to-Sample:</strong></td>
<td>250 mm</td>
<td>250 mm</td>
<td>250 mm</td>
</tr>
<tr>
<td><strong>Detector:</strong></td>
<td>Scintillation</td>
<td>Scintillation</td>
<td>Peltier-cooled Si(Li)</td>
</tr>
<tr>
<td><strong>Detector Area:</strong></td>
<td>506 mm²</td>
<td>506 mm²</td>
<td>24 mm²</td>
</tr>
<tr>
<td><strong>Detector Optics:</strong></td>
<td>Parallel-Beam Optic</td>
<td>Parallel-Beam Optic</td>
<td>1.2 mm receiving slit</td>
</tr>
<tr>
<td></td>
<td>0.4 mm x 150 mm long</td>
<td>0.4 mm x 150 mm long</td>
<td>3 mm scatter slit</td>
</tr>
<tr>
<td></td>
<td>8 x 13 mm opening</td>
<td>8 x 13 mm opening</td>
<td>1.1 deg. Soller slit</td>
</tr>
<tr>
<td><strong>Sample-to-</strong></td>
<td>185 mm</td>
<td>185 mm</td>
<td>250 mm</td>
</tr>
<tr>
<td><strong>Detector:</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Small, uniform, polycrystalline samples were “created” by covering a NIST SRM 1976 sintered corundum plate with a layer of 0.25mm lead foil. The edges of the plate were left uncovered so that the corundum sample would be tangential to the goniometer focusing circle. We cut openings of various widths in the lead foil and then covered all but one of the openings with another layer of lead foil to insure that diffraction from only one “sample” was measured at a time. We collected data on the corundum (300) near 68 degrees two-theta because the lead foil thickness effectively screened the corundum plate at lower two-theta values. This effect limited the minimum useable width to about 1mm.

Data was collected by step scan from 67-69 degrees two-theta with a 0.02 degree step size and a dwell time of 25 seconds. Two measurements were made with each size sample to minimize the error in aligning the opening with the X-ray beam. Linear background was subtracted from each spectrum. Peak heights were defined as the maximum intensity after background subtraction.
Two thin films were measured at grazing incidence: a 600 Å Pt thin film on a Si (100) substrate and a 350 Å TiN thin film also on Si (100). Both films showed significant texture in normal scans; the Pt film in the (111) direction and the TiN in the (100) direction.

GIXRD data was collected by step scan on the Pt (111) peak from 33-51 degrees two-theta with a step size of 0.04 degrees and a dwell time of 25 seconds. TiN data was collected for the TiN (200) peak from 33-45 degrees two-theta with a 0.04 degree step size and 15 second dwell time. Grazing angles varied from 0.3 degrees to 5 degrees. The spectra were processed as discussed above.

Results

Figure 1 shows the ratio of corundum (300) peak intensity for the parabolic, multilayer mirror to that of conventional Bragg-Brentano parafocusing optics. The mirror-based configuration showed increased intensity compared to Bragg-Brentano optics as the sample width decreased. However, as the sample width increased toward values expected for bulk samples, the Bragg-Brentano configuration actually performed slightly better. These results are consistent with the narrow, brilliant beam expected from the parabolic, multilayer mirror. As the sample width decreased, the higher X-ray flux density from the mirror compared to Bragg-Brentano optics is the dominant effect. On the other hand, the irradiated volume is the dominant effect at larger sample widths which favors Bragg-Brentano optics because of its wide, divergent X-ray beam. This effect makes it unlikely that parabolic, multilayer mirrors will become the standard optics for general-purpose diffractometers.

![Figure 1](image_url)

**Figure 1:** Comparison of Corundum (300) peak height ratios between mirror optics and Bragg-Brentano optics as a function of sample width.

Figure 2 shows grazing incidence data collected for an approximately 600 angstrom platinum thin film on a silicon (100) substrate for the three different hardware configurations described earlier. As you can see, both the parabolic, multilayer mirror and parallel beam cases show significant (about a factor of 7) improvement over the Bragg-Brentano case. This is because the sample in grazing angle geometry is not in the focusing conditions required for Bragg-Brentano optics; namely that the incident and diffracted beam
angles are equal. The performance of mirror optics is better seen by normalizing intensity from the mirror to intensity from the parallel beam case. This is shown in figure 3. This curve is very similar to figure 1.

![Graph showing comparison of Pt(111) peak heights measured by grazing angle diffraction using different optical configurations.](image1)

**Figure 2:** Comparison of Pt(111) peak heights measured by grazing angle diffraction using different optical configurations. Film thickness is approximately 600 angstroms.

![Graph showing Pt(111) peak height ratios as a function of grazing angle.](image2)

**Figure 3:** Pt (111) peak height ratios as a function of grazing angle for an approximately 600 angstrom thin film. Ratios compare parabolic, multilayer mirror optics to parallel beam optics using fixed divergence slits on the incident beam.
At low grazing angles, the brilliance of the X-ray beam is most important because only a fraction of the incident beam is actually illuminating the sample. However, at higher grazing angles where the size of the beam is dominant, there is no advantage to the mirror and it may actually be worse than the parallel beam case.

Figure 4 shows grazing angle diffraction data for a 350 angstrom TiN film on a silicon (100) substrate. Intensities are much weaker in this data, but the trend is about the same as in the previous experiment. Both the mirror and parallel beam cases produce significantly more intensity than the Bragg-Brentano case. The ratio of mirror to parallel beam intensities (figure 5) show the same trend as before, but the maximum improvement seen is only about 1.7 times the parallel beam case.

The mirror’s gain over the parallel beam case is less for the TiN thin film. It doesn’t appear that this can be due to a difference in texture because both films have significant texture. However, we do know that the reflectivity of the parabolic, multilayer mirror is not constant across the length of the mirror. At lower grazing angles when the incident beam is wide, these areas of differing reflectivity result in stripes of higher and lower X-ray illumination and can be seen as bright and dark stripes on a fluorescent screen. These two films were measured at different times with different mirror alignments so it is quite possible that the alignment of these bright and dark stripes was somewhat different.

![Graph](image)

**Figure 4:** Comparison of TiN(200) peak heights measured by grazing angle diffraction using different optical configurations. Film thickness is approximately 350 angstroms.
Figure 5: TiN (200) peak height ratios as a function of grazing angle for an approximately 350 angstrom thin film. Ratios compare parabolic, multilayer mirror optics to parallel beam optics using fixed divergence slits on the incident beam.

Conclusions

Compared to conventional Bragg-Brentano parafoseing optics, a parabolic, multilayer mirror was found to produce significantly increased peak intensities from small samples. The amount of increase varied inversely with the width of the sample and was as great as 5.2 times the Bragg-Brentano intensity for a 1.2 mm wide sample. Conversely, Bragg-Brentano optics outperformed the mirror for large sample sizes; presumably because of the mirror's very narrow incident beam.

The parabolic, multilayer mirror was also shown to be effective in grazing angle analysis of thin films. Studies carried out on 600 Å Pt and 350 TiN Å thin films showed that the mirror became more effective compared to parallel beam optics as the grazing angle decreased. Intensity improvements of 2.6 times the parallel beam case for the Pt film and 1.7 times the parallel beam case for the TiN film were measured. The differences in the amount of improvement compared to the parallel beam case were probably due to variations in reflectivity across the mirror surface.

Acknowledgment

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References