Ultra-thin curved transmission crystals for high resolving power (up to $E/\Delta E = 6300$) 

x-ray spectroscopy in the 6–13 keV energy range

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Ultra-thin curved transmission crystals operating in the Cauchois spectrometer geometry were evaluated for the purpose of achieving high spectral resolution in the 6–13 keV x-ray energy range. The crystals were silicon (111) and sapphire R-cut wafers, each 18 μm thick, and a silicon (100) wafer of 50-μm thickness. The W Lα spectral line at 8.398 keV from a laboratory source was used to evaluate the resolution. The highest crystal resolving power, $E/\Delta E = 6300$, was achieved by diffraction from the (33-1) planes of the Si(100) wafer that was cylindrically bent to a radius of curvature of 254 mm, where the (33-1) planes have an asymmetric angle of 13.26° from the normal of the crystal surface facing the x-ray source. This work demonstrates the ability to measure highly resolved line shapes of the K transitions of the elements Fe through Kr and the L transitions of the elements Gd through Th using a relatively compact spectrometer optical system and readily available thin commercial wafers. The intended application is as a diagnostic of laser-produced plasmas where the presence of multiple charged states and broadenings from high temperature and density requires high-resolution methods that are robust in a noisy source environment.

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The measurement of properties of energetic x-ray sources such as produced in the laboratory by intense laser or electron irradiation often requires the resolution of the shapes of the emitted spectral lines. Similarly, the detailed analysis of material properties by the x-ray diffraction technique requires the resolution of subtle changes in the transition line shapes and intensities. For x-ray energies below about 10 keV, this is typically achieved using high-dispersion reflection crystals. Typically curved reflection crystal geometries return very narrow spectral bandwidths that require tuning by rotation of the crystal and detector (and shielding) while maintaining the theta-two-theta registration. This adds to the engineering budget and optical design tolerances. While trading some resolving power for bandwidth, the curved transmission crystal geometry has no moving parts, and since the entire spectrum converges at a specific distance between the crystal and detector, a simple shielding configuration can ensure a high signal-to-noise spectrum. When using a reflection or transmission crystal, high spectral resolution generally recommends using a small 2d crystal lattice spacing to increase the dispersion. Because high-Miller index crystal planes typically have a small integrated reflectivity (rocking curve width), there is a trade-off between dispersion and throughput, and the crystal diffraction planes must be chosen with the end resolution and the convenience of the optical system in mind.

In this Letter, we present the analysis of high-dispersion diffraction planes of thin single-crystal wafers for the purpose of achieving resolving power $E/\Delta E$ exceeding 5000 in the 6–13 keV energy range and in the Cauchois transmission geometry having instrument length and width <0.5 m. Such resolving power permits the measurement of source and material properties such as temperature, density, and opacity in an x-ray range where the photons can penetrate and escape from dense materials with minimal perturbation, and the relatively small instrument size permits convenient deployment at the energetic x-ray source.

The dispersion plane of the spectrometer is shown in Fig. 1. The x rays from the source are diffracted by the cylindrically bent crystal, converge through a slit aperture, and are incident on a detector that is placed on the Rowland circle (RC) having diameter equal to the crystal bending radius of curvature. The Cauchois spectrometer has the property that to first order, monoenergetic x-rays from an extended source are focused at one point on the RC, and different energies are dispersed along the RC. Previously, we have used this type of spectrometer to record the W L spectra from electron-bombarded tungsten anode sources with resolving power $E/\Delta E = 1800$ [1,2]. In this study, ultra-thin crystals are employed to minimize absorption at the lower energies of interest.

As shown in Fig. 1, the source-to-crystal distance $S$ and the offset angle $\alpha$ are chosen so that the central energy $E$ of the x-ray spectrum is incident at a desired place on the

![Fig. 1. Schematic of the x-ray spectrometer.](Image)
crystal with the Bragg angle \( \theta \) given by 
\[
\sin \theta = \frac{h c}{2 d E},
\]
where \( h \) is Planck’s constant, \( c \) is the speed of light, and \( 2d \) is the lattice spacing. While the diffraction planes are typically perpendicular to the crystal surface facing the source, higher dispersion can often be achieved using planes that are at an asymmetric angle \( \delta \) to the crystal surface. It is this case that is illustrated in Fig. 1, showing the crystal rotated by \( \delta \) (about the intersection point of the spectrometer axis and the crystal) so that the grazing angle on these planes is the Bragg angle \( \theta \). Pb apertures are placed between the source and crystal and between the crystal and detector so that only dispersed x rays reach the detector.

As shown in Fig. 1, the spectrometer axis is defined by the line from the center of the source and intersecting the RC at the center of curvature of the crystal. X-rays having energy \( E \) from the extended source are focused on the RC at a distance \( x = R_c (\theta - \delta) \) from the spectrometer axis. The dispersion is \( \Delta x / \Delta E = R_c \tan \theta / E [1] \), and high dispersion is achieved by using a large crystal-bending radius \( R_c \), within the limitations of the instrument footprint, and large Bragg angle \( \theta \) that results from small lattice spacing \( 2d \). In addition, the crystal should be sufficiently thin so that attenuation of the x-rays of central energy \( E \) is small and sufficiently thick so that the diffraction efficiency is optimal. Since the diffraction efficiency of a curved transmission crystal depends on the bending radius and can vary with the polising and other crystal properties, the accurate calculation of the optimal thickness can be estimated by using the crystals listed in Table 1, the W L lines were observed in the 8.3–12.1 keV range, and the thinner crystals can operate down to 6 keV.

The crystal wafers, diffraction lattice planes, wafer thicknesses, and radii of curvature (ROC) that were studied are listed in Table 1. The value of \( \delta \) was determined using Crystal Maker [3,4] visualization software (\( \delta = 0 \) for symmetric diffraction). In all cases, the crystals were conformed to aluminum cylindrical forms by a covering plate without glue. The first and third entries in the table indicate the use of different internal diffraction planes of the same wafer. The (110) planes are perpendicular to the (100) planes of the large surface of that wafer that faces the x-ray source; the (33-1) planes are rotated 13.26 degrees from the (110) about an axis that is parallel to the (100) planes. The R-cut sapphire (Al2O3) wafer was oriented such that the (1-12) planes were parallel to the large surface of the lamella, and the (110) planes are perpendicular to this surface; spectra were recorded from the (121) planes that are at an angle of 5.49° to the (110) planes. Finally, a Si(111) wafer was studied that had the (11-2) planes perpendicular to the large-area surface of the wafer; high-dispersion spectra were recorded in second order from these planes (22-4).

The free spectral range primarily depends on the length of the bent crystal that is illuminated by the x-rays and the source-to-crystal distance. When using the 50.8-mm diameter Si(100) wafer, the entire tungsten L spectrum covering the 7–13 keV range was recorded.

The highest resolving power was achieved using the Si (33-1) planes, and those results are described here. The (33-1) lattice planes were oriented to diffract the W \( \La_1 \) and \( \La_2 \) spectral lines having 8.398 and 8.335 keV energies [5], respectively, near the center of the crystal. The \( \La_1 \) transition was selected for the evaluation of the crystal’s resolving power because it is the most intense of the L transitions, is well isolated from the other intense \( \Lp_1 \) and \( \Lp_2 \) transitions, and is near the much weaker L\( \alpha \) transition, which provides an excellent energy scale without perturbing the \( \La_1 \) transition. In addition, the \( \La_1 \) transition originates from the radiative decay of the tightly bound 3d\( ^{5/2} \) level of W and is unperturbed by multiple vacancies, oxidation, and other material properties of the tungsten anode that might influence the higher-lying levels [1]. Finally, the natural lifetime width of the \( \La_1 \) transition is well known and can be accurately accounted for when determining the crystal’s resolving power [6].

The spectrum recorded from the Si(33-1) planes when the crystal was positioned at a distance of 1065 mm from the source is shown by the data points in Fig. 2(a), and the curve is the fit of the sum of two Voigt profiles by the least squares technique. The fit was performed by varying eight parameters: two transition energies, two line intensities, two full width at half-maximum (FWHM) values of the Gaussian components, and two FWHM values of the Lorentzian components. The differences between the data points and the fitted curves are shown in Fig. 2(b) and indicate the high quality of the fit to the data points.

The Gaussian and Lorentzian FWHM values for the Voigt profile fitted to the \( \La_1 \) line were 1.71 and 7.52 eV, respectively, with 0.01-eV one-sigma fitting uncertainties. These values were used to infer the intrinsic resolution of the (33-1) diffracting planes after removing the broadening by the natural lifetime of the transition and the detector spatial resolution. The natural width of the W \( \La_1 \) transition is 6.51 ± 0.2 eV [6] and will be subtracted from the Lorentzian FWHM value.

The spectra were recorded on a Fuji [4] type TR image plate that was scanned with 17.1-μm steps by a ScanX [4] scanner. The spatial resolution of the TR image plate was measured from the edge spread function (ESF) and the line spread function (LSF) resulting from a Pb foil that was opaque to the x-ray emission from the source operating at 55 kV. The foil edge was oriented perpendicular

<table>
<thead>
<tr>
<th>Wafer</th>
<th>Perpendicular Planes</th>
<th>Diffraction Planes</th>
<th>2d (nm)</th>
<th>( \delta ) (deg)</th>
<th>Thickness (μm)</th>
<th>ROC (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si(100)</td>
<td>(110)</td>
<td>(220)</td>
<td>0.3833</td>
<td>0.00</td>
<td>50.00</td>
<td>254</td>
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<tr>
<td>Al2O3(1-12)</td>
<td>(110)</td>
<td>(121)</td>
<td>0.3095</td>
<td>5.43</td>
<td>18.00</td>
<td>165</td>
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<tr>
<td>Si(100)</td>
<td>(110)</td>
<td>(33-1)</td>
<td>0.2487</td>
<td>13.26</td>
<td>50.00</td>
<td>254</td>
</tr>
<tr>
<td>Si(111)</td>
<td>(11-2)</td>
<td>(22-4)</td>
<td>0.2213</td>
<td>0.00</td>
<td>18.00</td>
<td>165</td>
</tr>
</tbody>
</table>

Table 1. Diffraction Planes that Were Studied in Detail
to the dispersion direction and along the spectral lines on the TR image plate, and its image was scanned in the same manner as the W spectral lines. The ESF and its derivative, the LSF, are shown by the points in Fig. 3(a). The Fourier transform of the LSF, i.e., the modulation transfer function (MTF), is shown by the points in Fig. 3(b). A Voigt profile was fitted to the LSF, and the FWHM values of the Gaussian and Lorentzian components are found to be 23.77 and 22.03 μm, respectively, with 0.01 μm fitting uncertainties.

Dividing by the (33-1) dispersion, 22.32 μm/eV having estimated 1% uncertainty resulting from possible positioning error of the image plate on the RC, the TR spatial resolution contributes 1.06/0.00060.01 and 0.99/0.00060.01 eV to the Gaussian and Lorentzian components of the Voigt profile fitted to the W Lα1 line. Linearly subtracting the 6.51 ± 0.2 eV natural broadening and the 0.99 ± 0.01 eV TR detector Lorentzian FWHM broadening from the 7.52 ± 0.01 eV FWHM value of the Lorentzian component of the Lα1 line, the result is 0.02 ± 0.2 eV. Subtracting in quadrature the TR detector 1.06 ± 0.01 eV Gaussian FWHM broadening from the 1.71 ± 0.01 eV FWHM value of the Gaussian component of the Lα1 line, the result is 1.34 ± 0.01 eV. Thus within the accuracy of the measurements undertaken in the present geometry, the intrinsic instrumental broadening profile due to the (33-1) lattice planes is Gaussian with 1.34 ± 0.01 eV FWHM, and the resolving power is $E/\Delta E = 8398 \text{ eV}/1.34 \text{ eV} = 6300$.

The intrinsic resolving power of the (33-1) crystal was also measured as a function of source-to-crystal distance, and the results are summarized in Fig. 4. The various Gaussian and Lorentzian FWHM values are shown by the data points (having one-sigma error bars) in Fig. 4(a), where straight lines are drawn between the data points, and the resolving power is shown in Fig. 4(b). For each source-to-crystal distance, the FWHM value of the Lorentzian component of the intrinsic resolution of the (33-1) planes, curve 3 in Fig. 4(a), is zero within the experimental accuracy. The FWHM value of the Gaussian component of the intrinsic resolution, curve 5, decreases with source-to-crystal distance, and the resulting resolving power shown in Fig. 4(b) increases with distance to the value 6300 at 1065 mm distance.

The resolving power increases with the source-to-crystal distance because the intrinsic Gaussian FWHM decreases with the source-to-crystal distance. The crystal properties that contribute to the intrinsic resolution of the bent crystal are crystal thickness, rocking curve width, and optical aberrations. The thickness and rocking-curve broadenings are independent of the source-to-crystal distance, and the aberration broadening is proportional to $\omega R_c / (R_c + S)$ where $\omega$ is the lateral width of the source in the dispersion plane, $R_c$ is the crystal bending radius, and $S$ is the source-to-crystal distance. Thus the decrease in the intrinsic Gaussian FWHM with source-to-crystal distance results from the decrease in the aberration broadening with distance. In effect, the angle subtended by the source as viewed from the crystal decreases as the source-to-crystal distance increases, and the effectively smaller source size reduces the detrimental aberration effects. In addition to the aberration broadening, additional broadening may result from imperfect bending of the crystal. In any case, for extremely high resolving power such as 6300, very small effects can influence the resolving power, and it is necessary to...
experimentally measure the resolving power to have confidence that a high-resolution spectrometer will perform as expected. While the (33-1) planes had the highest resolving power in the present study, the other diffraction planes listed in Table 1 may have the optimal resolving power in other applications and spectrometer geometries. The calculated Bragg angles of the four planes listed in Table 1 are shown in Fig. 5(a), and the dispersions are shown in Fig. 5(b) for the two cases of 254- and 500-mm crystal bending radii. As always, the dispersion increases with decreasing energy and with increasing bending radius, and the resolving power achieved in practice is influenced by the product of the dispersion and the detector spatial resolution. The selection of the optimal diffracting planes depends on the x-ray energy, the Bragg angle that influences the spectrometer width, the bending radius that influences the spectrometer width and length, and the detector spatial resolution compared to the dispersion. The instrument sensitivity and throughput decrease with larger source-to-crystal distance, larger crystal radius of curvature, higher Miller indices and dispersion, and these effects should also be considered for applications. Finally, because the higher resolving powers are achieved for larger source-to-crystal distances and since imperfect bending of the crystal and crystal preparation can affect the resolving power and the integrated reflectivity and throughput, the properties of the selected crystal diffraction planes must be experimentally characterized for each geometry and application.

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References and Notes

4. Certain commercial equipment, instruments, or materials are identified in this Letter in order to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by the U.S. government, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.