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X-ray spectrometer having 12 000 resolving power at 8 keV energy

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Abstract

An x-ray spectrometer employing a thin (50 μm) silicon transmission crystal was used to record high-resolution Cu $K\alpha$ spectra from a laboratory x-ray source. The diffraction was from the (331) planes that were at an angle of 13.26° to the crystal surface. The components of the spectral lines resulting from single-vacancy (1s) and double-vacancy (1s and 3d) transitions were observed. After accounting for the natural lifetime widths from reference double-crystal spectra and the spatial resolution of the image plate detector, the intrinsic broadening of the transmission crystal was measured to be as small as 0.67 eV and the resolving power 12 000, the highest resolving power achieved by a compact (0.5 m long) spectrometer employing a single transmission crystal operating in the hard x-ray region. By recording spectra with variable source-to-crystal distances and comparing to the calculated widths from various geometrical broadening mechanisms, the primary contributions to the intrinsic crystal broadening were found to be the source height at small distances and the crystal apertured height at large distances. By reducing these two effects, using a smaller source size and vignetting the crystal height, the intrinsic crystal broadening is then limited by the crystal thickness and the rocking curve width and would be 0.4 eV at 8 keV energy (20 000 resolving power).

I. INTRODUCTION

The resolving power achieved by an x-ray spectrometer, operating in the Bragg reflection geometry at lower energies (typically <10 keV) or the Laue transmission geometry at higher energies, is limited by the angular width of the diffraction profile (the rocking curve width) and also by various geometrical factors such as source dimensions, crystal and slit aperture dimensions, detector spatial resolution, and crystal thickness. In the case of Bragg reflection, the broadening contributions have been greatly reduced by using double-crystal diffraction, small apertures, and highly accurate scanning mechanisms. In addition, extremely small instrumental broadening ($\Delta E/E < 10^{-6}$) has been demonstrated by recording the characteristic K spectral lines from electron-bombarded Cu anode sources.^{1,2} The two intense Cu $K\alpha_1$ and $K\alpha_2$ transitions near 8.04 keV from single (1s) vacancies are blended with much weaker spectral features from double vacancies (1s and 3d). The line widths of a few electron volts result from the distribution of the double-vacancy transition energies

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around the single-vacancy transitions and from the natural lifetime widths of the transitions. These high-resolution Cu K α spectra, having negligible instrumental broadening, serve as standard reference spectra for comparison to spectra recorded by a spectrometer having significant instrumental broadening. After accounting for the standard reference line widths and removing the detector broadening, the spectrometer's residual broadening is attributed to the intrinsic crystal broadening which is a figure of merit for high-resolution diffraction crystals. This process was used to determine the intrinsic crystal broadening of a cylindrically bent transmission crystal in the Cauchois geometry.

The motivation for this work is the development of spectrometers operating in the hard x-ray region (>8 keV) having sufficiently high resolving power ($>10\,000$) for the application of spectroscopic diagnostic techniques based on the measurement of the spectral line widths from hot, dense plasmas produced at high-intensity laser and other facilities. For example, ³⁻⁶ the plasma temperature can be determined from the line ratios of transitions from a distribution of charge states, the ion temperature and turbulent motions from the Doppler-broadened spectral line widths, and in dense plasmas the electron density from Stark broadening.

A schematic of the spectrometer setup is shown in Fig. 1. The crystal is a commercial (100) silicon wafer having 76 mm diameter and polished to a thickness of 50 μm . The (110) planes are perpendicular to the wafer surface and to the small reference flat on the edge of the wafer. The wafer was bent on an aluminum frame having 500 mm radius. By making the reference flat parallel to the plane of the bending circle (the Rowland circle in Fig. 1), the (110) planes were oriented perpendicular to the plane of the bending circle. In this study, the (331) diffraction planes were chosen; these are also perpendicular to the plane of the bending circle and at an angle $\alpha = 13.26^\circ$ to the (110) planes. The Cu K α transitions near 8.04 keV are diffracted from the (331) planes having lattice spacing $2d = 0.2487$ nm and with average Bragg angle $\theta = 38.3^\circ$.

The alignment of the crystal to the x-ray source and the tuning of the energy range are easily accomplished as discussed in Ref. 7. An alignment laser beam is pointed to the center of the x-ray source as shown in Fig. 1. The crystal is positioned on a rotation stage so that the center of the crystal is on the rotational axis of the stage and the alignment beam is incident on the center of the back face of the crystal. The crystal is oriented on the rotation stage so that the alignment beam is back reflected from the crystal surface to the laser. The crystal surface is then perpendicular to the alignment beam and the (110) planes at the crystal center are parallel to the alignment beam, and this defines the zero rotation angle of the crystal. The crystal is rotated by the Bragg angle θ , and the alignment beam reflected from the back surface of the crystal (at angle 2θ to the alignment beam) defines the path of the diffracted x rays to the detector as indicated in Fig. 1. A slit aperture is placed midway between the crystal and the detector and is centered on the reflected laser beam so that it passes the diffracted x rays. The purpose of this slit is to reduce background exposure on the detector resulting from fluorescence and scattering in the crystal, and it does not function as a limiting aperture and does not affect the spectral resolution. The crystal is rotated by an additional angle α (to angle $\theta + \alpha$ from the alignment beam) so that photons of energy $E = hc/(2d \sin \theta)$ are diffracted from the (331) planes at the center of the crystal.

A typical spectrum recorded by diffraction from the Si (331) planes is shown on a log scale in Fig. 2. Identified are the intense $K\alpha_1$ and $K\alpha_2$ transitions and also the $K\alpha_{3,4}$ feature which results from transitions having $1s$ and $2p$ vacancies.¹

The x-ray source was an electron-bombarded Cu anode with 30 kV electron beam energy. The detector that was employed was a Fuji¹³ TR image plate that was positioned on the Rowland circle whose diameter is equal to the crystal's 500 mm bending radius. While the spectral lines to first order are focused on the Rowland circle, the spectral lines are broadened by the detector spatial resolution, the blending and lifetime broadening of the spectral lines, and a number of crystal geometrical effects as discussed in Refs. 8 and 9. After removing the detector broadening (by measurement of the detector spatial resolution) and the blending and lifetime broadening of the spectral lines (using the line width measurements from Ref. 2), the residual broadening is the convolution of the crystal geometrical effects and represents the intrinsic crystal broadening.

II. DETECTOR SPATIAL RESOLUTION

The spatial resolution of the TR image plate detector was measured using x-ray exposures through a resolution test gauge. The gauge consisted of line pair per mm (LPMM) features fabricated in a 20 μm thick Pb foil. Each LPMM feature had four bars and five adjacent open spaces of the same lateral width, and the bar/space frequency ranged from 5 LPMM to 20 LPMM. An additional opening had no bars (0 LPMM). The overall size of the gauge was 25 mm long and 12 mm wide, and the gauge was packaged in a thin plastic case for handling and stability. The gauge was placed in contact with the TR image plate and was positioned 1.0 m from the x-ray source.

The resolution gauge was exposed to narrow bandpass x-ray fluence at the National Institute of Standards and Technology (NIST) absolute x-ray calibration facility. The x-ray source had an electron-bombarded W anode, and the upper limit of the bandpass was established by the peak kilovoltage provided by the computer-controlled power supply, selected to be 15 kV for these resolution tests. The lower bandpass limit was established by standard attenuation filters and was 7.5 keV. The photon-number weighted centroid energy of the bandpass was 11.8 keV. The x-ray fluence on different exposures was reproducible to an accuracy of better than 1%.

The exposed TR image plate was scanned by a ScanX¹³ scanner with 17 μm steps. As shown in Fig. 3, this scanner pulls the image plate into the scan region, and the laser scans the image plate while moving perpendicular to the image plate feed direction. The exposed TR image plate was fed into the scanner using one of the two selected orientations, with the LPMM bar pattern parallel to the laser scan direction (as illustrated in Fig. 3) or with the bar pattern perpendicular to the laser scan direction. Typical scans in the two orientations are shown in Fig. 4 where the curves are integrated and averaged over the width of the bar pattern. The orientations of the bar pattern are (a) parallel to the laser scan direction and (b) perpendicular to the laser scan direction. It is immediately apparent that the high-frequency bars are better resolved when the bar pattern is parallel to the laser scan direction.

Shown in Fig. 5 are the contrast transfer functions (CTFs) for the two cases of the bar pattern (1) parallel to the laser scan direction (three exposures) and (2) perpendicular to the laser scan direction (two exposures). The CTF is defined as $(C_{\max} - C_{\min}) / (C_{\max} + C_{\min})$, where $C_{\min}$ and $C_{\max}$ are the minimum and maximum count values (summed and averaged along the lateral length of the bars and open spaces) in each LPMM feature. The CTF curves derived from different exposures that were scanned with the same orientation are in good agreement.

As discussed in Ref. 10, which presents the spatial resolution of the Fuji SR and MS image plates scanned by a Logos scanner,¹³ the CTF is related to the modulation transfer function (MTF), and the line spread function (LSF) is the inverse Fourier transform of the MTF. The data points in Fig. 6 are the LSF (normalized to unity) derived from the averages of the CTF curves for each scan orientation (three curves with the gauge bar pattern oriented parallel to the laser scan direction and two curves with the gauge bar pattern perpendicular to the laser scan direction). The curves in Fig. 6 are fits of Voigt profiles to the LSF data by the least squares technique where the widths of the Gaussian and Lorentzian components were variables. The optimum full width at half maximum (FWHM) values of the Gaussian and Lorentzian components are (1) 39.9 μm and 4.3 μm , respectively, for the case of the bar pattern parallel to the laser scan direction and (2) 48.9 μm and 12.2 μm for the case of bar pattern perpendicular to the laser scan direction with $\pm 1 \mu\text{m}$ fitting uncertainties for the Gaussian components and for the Lorentzian components. This quantifies the superior spatial resolution of the TR image plate when the bar pattern is parallel to the laser scan direction. We note that a similar difference in the spatial resolution of the Fuji SR image plate with scan orientation was reported in Ref. 11.

III. INTRINSIC CRYSTAL BROADENING

The spectra diffracted by the silicon crystal's (331) planes were scanned by the ScanX with the spectral lines parallel to the laser scan direction as illustrated in Fig. 3, the orientation giving the superior spatial resolution. The intrinsic crystal broadening was derived by removing the TR image plate resolution, measured during the spatial resolution tests, and removing the broadening resulting from blending and lifetime as quoted in Ref. 2.

The data points in Fig. 7(a) represent the spectrum recorded using 150 cm source-to-crystal distance. The identifications $K\alpha_{11}$ and $K\alpha_{21}$ refer to the intense single-vacancy ($1s$) transitions usually designated $K\alpha_1$ and $K\alpha_2$. The identifications $K\alpha_{12}$ and $K\alpha_{22}$ refer to the much weaker features on the low-energy wings that result from double-vacancy ($1s$ and $3d$) transitions. In Refs. 1 and 2, four Voigt profiles were fitted to these spectral features; the transition energies and the Lorentzian FWHM values from Ref. 2 are listed in Table I (the corresponding values from Ref. 1 are similar).

Figure 7(a) shows the fits of four Voigt profiles to the spectrum data points recorded in this work. The differences between the Voigt fit and the data points shown in Fig. 7(b) are well within the $\pm 1\sigma$ curves, where σ is the Poisson counting uncertainty C/C and C is the spectrum count value. The fits were performed using the transition energies and Lorentzian FWHM values from Ref. 2 (listed in Table I) with additional Lorentzian and Gaussian

broadening resulting from the spatial resolution of the TR image plate and the intrinsic crystal broadening. It was found that the additional Lorentzian width was 0.09 eV, the product of the TR Lorentzian FWHM $4.3 \mu\text{m}$ and the 20.4 eV/mm dispersion provided by the Si (331) planes. The additional Gaussian width decreased with the source-to-crystal distance and was equal to 2.00 eV at the smallest distance (25.6 cm) and 1.05 eV at the largest distance (150 cm). Subtracting in quadrature the TR Gaussian FWHM 0.81 eV, the product of the TR Gaussian FWHM $39.9 \mu\text{m}$ and the 20.4 eV/mm dispersion, the resulting Gaussian FWHM values decreased from 1.83 eV at 25.6 cm distance to 0.67 eV at 150 cm distance, and these values represent the intrinsic crystal broadening. The highest resolving power, equal to 8.04 keV/0.67 eV, occurred at the largest source-to-crystal distance and was 12 000.

The intrinsic crystal broadening values, determined at five source-to-crystal distances, are shown by the data points in Fig. 8. The numbered curves are the calculated broadenings resulting from the various geometrical effects discussed in Ref. 9: (1) total width, (2) 2.5 mm source height in the direction perpendicular to the horizontal dispersion plane, (3) 13 mm crystal apertured height, (4) $50 \mu\text{m}$ crystal thickness, (5) $13 \mu\text{rad}$ rocking curve width calculated using the multilamellar mode of the XOP code,¹² (6) 1% possible error in the 500 mm crystal bending radius and 5 mm possible error in positioning the image plate on the Rowland circle, and (7) aberrations resulting from the 1 mm source width in the dispersion plane multiplied by a factor of 100 for display. The dominant broadening mechanisms result from the source height and the apertured crystal height. These broadenings could be reduced by using a smaller source and aperturing the crystal to a smaller height, and then the intrinsic crystal broadening would be limited by the crystal thickness and the rocking curve width and would be 0.4 eV (20 000 resolving power) at the largest source-to-crystal distance.

IV. DISCUSSION

By using the NIST Cu K α spectrum recorded by a double-crystal spectrometer with negligible instrumental broadening as a standard reference spectrum,² the instrumental broadening of a single transmission crystal in the Cauchois geometry was evaluated. After removing the Lorentzian line widths derived from the NIST reference spectrum and the measured detector resolution from the single-crystal spectrum, the intrinsic broadening by the single crystal was measured. The intrinsic broadening by the Si (331) planes of a thin ($50 \mu\text{m}$) silicon wafer was found to be Gaussian with FWHM values that varied from 2.00 eV at the smallest (25.6 cm) source-to-crystal distance to 0.67 eV at the largest distance (150 cm) corresponding to 12 000 resolving power. Based on comparisons with the calculated broadening effects, the dominant broadening mechanisms were identified to be source height and crystal apertured height. By using a smaller source and smaller crystal height, the broadening is limited by the crystal thickness and rocking curve width and would be 0.4 eV (20 000 resolving power), an order of magnitude improvement over the present generation of compact hard x-ray spectrometers at large laser facilities.⁴⁻⁶ This work points the way for the development of hard x-ray transmission crystal spectrometers having greatly improved resolution for the measurement of the line shapes and plasma properties in laser-produced and other hot, dense plasmas.

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13. Certain commercial equipment, instruments, or materials are identified in this paper 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 is necessarily the best available for the purpose.

FIG. 1. Schematic of the spectrometer's dispersion plane showing the bent crystal, Rowland circle, and alignment laser.

FIG. 2.
Spectrum of the Cu $K\alpha$ lines diffracted by the Si (331) planes recorded with 50 cm source-to-crystal distance.

FIG. 3.

Orientation of the image plate scan with the gauge bar pattern or spectral lines parallel to the laser scan direction, the orientation giving the superior spatial resolution.

FIG. 4.

Scan of the gauge bar pattern for the two image plate scan orientations having the gauge bar pattern (a) parallel to the laser scan direction and (b) perpendicular to the laser scan direction.

FIG. 5.

The contrast transfer function for the two cases of the gauge bar pattern oriented (1) parallel to the laser scan direction and (2) perpendicular to the laser scan direction.

FIG. 6.
Fits of Voigt profiles to the line spread functions for the two cases of the gauge bar pattern
(1) parallel to the laser scan direction and (2) perpendicular to the laser scan direction.

FIG. 7.

(a) Fits of four Voigt profiles (curves) to the experimental Cu K α spectrum (data points) recorded with 150 cm source-to-crystal distance. (b) Difference between the fitted Voigt profiles and the experimental spectrum. The curves labeled $\pm 1\sigma$ in (b) represent $\pm C/C$ where C is the spectrum count value in (a).

FIG. 8.

Comparison of the measured intrinsic crystal broadening (data points) with the calculated widths where the curves are (1) total width, (2) source height, (3) crystal apertured height, (4) crystal thickness, (5) rocking curve width, (6) possible error in the crystal bending radius and error in positioning the image plate on the Rowland circle, and (7) aberrations resulting from the source width multiplied by a factor of 100. The error bars on the crystal broadening data points represent the uncertainties resulting from the determination of the TR image plate broadening and the least-squares fitting of the Voigt profiles to the experimental spectrum.

TABLE I.

Transition energies and Lorentzian widths from Ref. 2.

	Energy (eV)	Energy $\pm 1\sigma$ (eV)	FWHM (eV)	FWHM $\pm 1\sigma$ (eV)
K α_{11}	8047.8254	0.0003	2.275	0.001
K α_{12}	8045.2956	0.0047	2.915	0.009
K α_{21}	8028.0503	0.0027	2.529	0.005
K α_{22}	8026.5386	0.0092	3.274	0.008