Hard X-ray Spectrographs with Resolution Beyond 100 µeV

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Spectrographs take snapshots of photon spectra with array detectors by dispersing photons of different energies into distinct directions and spacial locations. Spectrographs require optics with a large angular dispersion rate as the key component. In visible light optics diffraction gratings are used for this purpose. In the hard x-ray regime, achieving large dispersion rates is a challenge. Here we show that multi-crystal, multi-Bragg-reflection arrangements feature cumulative angular dispersion rates almost two orders of magnitude larger than those attainable with a single Bragg reflection. As a result, the multi-crystal arrangements become potential dispersing elements of hard x-ray spectrographs. The hard x-ray spectrograph principles are demonstrated by imaging a spectrum of photons with a record high resolution of ∆E ≃ 90 µeV in hard x-ray regime, using multi-crystal optics as dispersing element. The spectrographs can boost research using inelastic ultra-high-resolution x-ray spectroscopies with synchrotrons and seeded XFELs.

PACS numbers: 42.25.-p, 41.50.+h, 07.85.Nc, 78.70.Ck, 07.85.Fv

A dream x-ray spectrometer is actually a spectrograph that images x-ray spectra in one shot, and with an ultimate spectral resolution. State of the art single shot x-ray spectrometers are imaging spectra with array detectors, using Bragg’s law dispersion (BD). BD links the angle of incidence θ to the energy E of photons Bragg reflected from the crystal atomic planes. However, the spectral resolution of the BD-spectrometers is always limited by the Bragg reflection (Darwin) bandwidth.

Angular dispersion (AD) is one way how to overcome the Darwin width limitation and substantially improved spectral resolution of x-ray optics. AD is a variation of the photon angle of reflection θ′, for a fixed incidence angle θ, with the photon energy E. AD takes place in Bragg diffraction, albeit only if the diffracting atomic planes are at a nonzero angle η ≠ 0 (asymmetry angle) to the entrance crystal surface, see Fig. 1(b).

Unlike BD, AD links θ′ to E for a fixed θ. AD is independent of the Darwin width, and can be therefore used to resolve much narrower spectral features. Using angular-dispersive monochromators, x-rays were already monochromatized to bandwidths (0.45 meV) almost two orders of magnitude smaller than the width of the Bragg reflections (27 meV) involved. New concepts are required, however, to realize single shot angular-dispersive spectrographs.

We show here that multi-Bragg-reflection arrangements feature, in theory and in experiment, cumulative angular dispersion rates almost two orders of magnitude greater than those attainable in a single Bragg reflection. An angular-dispersive x-ray spectrograph of a Czerny-Turner-type is introduced with the enhanced angular-dispersive optics of a “diffraction grating”. A record high spectral resolution of ∆E ≃ 90 µrad is demonstrated in the hard x-ray regime.

Czerny-Turner grating spectrographs are nowadays standard in infrared, visible, and ultraviolet spectroscopies. In its classical arrangement, the spectrographs comprise, first, a collimating mirror M_c, which collects photons from a radiation source S and collimates the photon beam - see Fig. 1(a); second, an angular-dispersive element DE such as a diffraction grating or a prism, which disperses photons of different energies into different directions; third, a curved mirror M_f which focuses photons of different energies onto different locations x(E), and, last but not least, a spatially sensitive detector Det placed in the focal plane to record the whole photon spectrum. To achieve high resolution, the most important factor is the magnitude of the AD rate D = δθ′/δE, which measures the variation of the reflection angle θ′ with photon energy E upon reflection from the dispersing element. For the given mirror focal length F (M_f → Det), the AD rate D determines the variation of the source image position x(E) on the detector with respect to photon energy: δx(E) = D F δE. The smallest spectral interval ∆E which can be resolved is therefore

$$\Delta E = \frac{1}{D} \frac{\Delta x}{F}, \quad (1)$$

where Δx is the largest of either the source S image size on the detector for a particular monochromatic component or detector spatial resolution.

Nowadays, diffraction grating manufacturing technology has advanced to the extent that grating spectrographs are being successfully used with much shorter wavelengths, in particular in soft x-ray regime (≈ 1 keV) attaining a resolving power of E/∆E ≃ 10^4. Extension into the hard x-ray regime is, however, not trivial, because of the lack of hard x-ray optics elements with sufficiently large dispersion rate.

A hard x-ray equivalent of the diffraction grating is a Bragg diffracting crystal with diffracting atomic planes at an asymmetry angle η ≠ 0 to the entrance crystal surface - Fig. 1(b). The AD rates in Bragg diffraction are typically small D ≃ 8 µrad/meV, being the main
optics
dispersing
detector
mirror CDFDW
focusing
ations in
\( \theta \)
vention that the counterclockwise sense of angular vari-
wise, unlike that shown in Fig. 1(b), then
deflection of the ray upon the Bragg reflection is clock-
where
has to be used with sign minus.
rays are dispersed, \( \frac{d\theta}{E} \)\( \frac{d\theta}{D} \), using Bragg’s law \( 2K \sin \theta = H \),
and assuming \( |\theta’ - \theta| \ll 1 \), we obtain
\[
\frac{d\theta}{E} = -b \frac{d\theta}{dE} + D, \quad D = \frac{2\sin \theta \sin \eta}{E \sin(\theta - \eta)}
\]
Here \( b = -\sin(\theta + \eta)/\sin(\theta - \eta) \) is the asymmetry ra-
tio. If the incident beam is collimated, \( d\theta/dE = 0 \), then
\( d\theta'/dE = D \), where \( D \) is the intrinsic AD rate in
a Bragg reflection [5, 6]. If, however, the incident x-
rays are dispersed, \( d\theta/dE \neq 0 \), then the dispersion rate
\( d\theta'/dE = D_{\text{out}} \) becomes
\[
D_{\text{out}} = bD_{\text{in}} + D,
\]
where \( D_{\text{in}} = -d\theta/dE \). The minus sign follows the con-
vention that the counterclockwise sense of angular vari-
ations in \( \theta \) and \( \theta' \) is positive. Similarly, if the sense of
deflection of the ray upon the Bragg reflection is clock-
wise, unlike that shown in Fig. 1(b), then \( D \) in Eq. 3
has to be used with sign minus.

Equation 4 demonstrates that the AD rate \( D_{\text{in}} \) can be
indeed significantly enhanced by two successive asym-
metric Bragg reflections, if its asymmetry ratio is large:
\( |b| \gg 1 \). The enhancement can be even larger if several
\( (1, 2, ..., n) \) successive reflections are used:
\[
D_{\text{out}} = b_n D_{\text{in}} + D_n = b_n(b_{n-1} \cdots (b_1D_1 + D_2) + D_3) \cdots D_{n-1} + D_n.
\]

In the first experiment presented below, we demon-
strate this effect on an example of a four-crystal an-
gular dispersive CDFDW optics, with schematic shown in
Fig. 1(c). In the second proof of principle experiment
presented in this paper, we apply such optics as the “dis-
pergrating” of a prototype hard x-ray spectrograph
to image a spectrum of the CDFDW with record high
spectral resolution. The experiments were performed at
30ID beamline of the APS.

Details on the CDFDW optics used in this paper are
provided in [9]. CDFDW is a modification of the CDW
optics originally designed to achieve the very high
monochromatization of x-rays. The first element - C (col-
limator) - is a Si asymmetrically cut crystal, with the 220
Bragg reflection, \( \theta_c = 20.7^\circ, \eta_c = 19.0^\circ, b_c = -1/21.5 \),
accepting x-rays with photon energy \( E = 9.1315 \text{ keV} \)
in a wide angular range \( \sim 110 \mu\text{rad} \), and colimating
it to a beam with a \( |b_c| \) smaller divergence, and negli-
gible \( D_c = 0.040 \mu\text{rad}/\text{meV} \). The next two Si crystals
- \( D_1 \) and \( D_2 \), are designed to produce maximal intrin-
sic Bragg dispersion rate \( D \), using the 008 reflection
with \( \theta_{d_1} \approx 90^\circ, \eta_{d_1} = 88.0^\circ, b_{d_1} \approx -1 \) \( (i = 1, 2) \),
and \( D_{d_1} = -D_{d_2} = 6.27 \mu\text{rad}/\text{meV} \). Note, that the
scheme in Fig. 1(c) is shown in a generic configuration
with \( \theta_{d_1} \neq 90^\circ \). The fourth crystal - W, is equiv-
alent to the C-crystal, however applied in an inverse
configuration with \( \eta_w = -\eta_c, b_w = 1/b_c = -21.5, \)
and \( D_w = 0.86 \). It is used to enhance the AD rate

![FIG. 1: Scheme of the Czerny-Turner type spectograph with a diffraction grating (a), or a crystal in asymmetric x-ray Bragg diffraction (b) as dispersing element - DE. Other components include radiation source S, collimating and focusing mirrors \( M_s \) and \( M_r \), and position sensitive detector Det. (c) Multi-crystal multi-reflection CDFDW optics - is an example of a hard x-ray “diffraction grating” (DE element) with enhanced dispersion rate, suitable for hard x-ray spectrographs.](image-url)
of the D-crystals. Indeed, applying Eq. (5) we estimate the cumulative dispersion rate of the optics: $D = b_w(D_{d_2} - D_{d_1}) + D_w = 2b_wD_{d_1} \approx -270 \, \mu\text{rad}/\text{meV}$, enhanced by a factor $2|b_w| \approx 43$ compared to the dispersion rate achieved in a single Bragg reflection. The same enhancement factor was derived in [15] using DuMond diagram analysis.

Figure 2 shows a schematic of the first experiment and results of measurements of the AD rate of the CDFDW optics. A tunable monochromator with a $\approx 170 \, \mu\text{eV}$ bandwidth shown schematically in Fig. 2(a), and described in detail in the next paragraph, is used to measure transmission spectra through the CDFDW optics under study, presented in Fig. 2(b). An auxiliary element denoted as +W in Fig. 2(b), is used to extract from all x-ray photons emanating from the CDFDW optics a small part with $\approx 20 \, \mu\text{rad}$ divergence by the 220 symmetric Bragg reflection (angular acceptance 20 $\mu\text{rad}$) from a Si channel-cut crystal. For each angular position $\Theta_{+W}$ of the +W crystal, a spectrum of x-rays transmitted through the CDFDW optics and through the +W angular analyzer is measured, as shown in Fig. 2(c). Figure 2(d) presents a 2D plot of the spectra. The peak of the spectral distribution changes with the emission angle defined by $\Theta_{+W}$. Figure 2(e) shows that the dependence is linear, with the tangent $D_{\text{CDFDW}} = 314 \, \mu\text{rad}/\text{meV}$ representing the measured dispersion rate of the CDFDW optics. The number is even higher than the previously estimated one, which we attribute to the difference between the nominal and real asymmetry angles. The result confirms the theoretical prediction, expressed by Eqs. (4)-(5), that the angular dispersion rate can be substantially enhanced in multi-crystal arrangements.

A few details regarding the monochromator in Fig. 2(a) are in order: It is the same CDFDW optics, however, enhanced with the +W channel-cut that substantially decrease the CDFDW bandwidth. The energy tuning of the CDFDW+W monochromator is performed by synchronous change of the angular orientation of the D-crystals, as indicated by $\Theta_{d_i}$ in Fig. 2(a), and explained in more detail in [9]. Each spectral dependence in Figure 2(c) has a width of $\Delta E \approx 240 \, \mu\text{eV}$. This number represents the width of the convolution of the spectral distributions of the CDFDW+W monochromator - Fig. 2(a), and the analyzer - Fig. 2(b). Assuming they are equivalent, the spectral width of a single CDFDW+W optics is estimated as $\Delta E/\sqrt{2} \approx 170 \, \mu\text{eV}$. An envelope of spectral dependences in Fig. 2(c) reveals a total width of $\Delta E_{\text{CDFDW}} \approx 460 \, \mu\text{eV}$, and represents the spectral width of the CDFDW optics. A similar number.
The experimental scheme in Fig. 4(a) is complemented by the +W channel cut which, as we know from the results presented in Fig. 2, selects x-ray emanating in a certain direction from the CDFDW optics, and, as a result, within a reduced bandwidth, whose central photon energy is defined by the angle \( \Theta_{x,w} \). Figure 4(b) shows the spatial distribution of x-rays measured at different \( \Theta_{x,w} \) values. The peak positions, plotted in Fig. 4(b), change linearly with \( \Theta_{x,w} \) at a rate \( D_\perp = 1.13 \, \mu \text{m}/\mu \text{rad} \). Together with the results presented in Fig. 2(e), this proves that the spectral distribution of x-rays from the CDFDW optics is imaged by mirror \( M_F \) on the spatial scale, with a conversion factor \( D_\perp D_{\text{CDFDW}} \approx 355 \, \mu \text{m}/\text{meV} \). Using this number, we obtain that the total widths of the spatial distributions in Figs. 3(b) and 3(b) are \( \Delta_{\text{CDFDW}} \approx 450 \, \mu \text{eV} \), representing the spectral width of the CDFDW optics. The spatial widths of single lines vary from 32 \( \mu \text{m} \) (in green) to 50 \( \mu \text{m} \) (in yellow), corresponding to spectral widths \( \approx 90 \, \mu \text{eV} \) and \( \approx 140 \, \mu \text{eV} \) respectively. The resolution of the CDFDW spectrograph is at least \( \approx 90 \, \mu \text{eV} \), or better. The fact that this value changes across the CDFDW spectrum, as well as the fact that the CDFDW spectrum has a double-peak structure, imply that the CDFDW optics we have built is not yet perfect. This is consistent with the results of [9], where a somewhat broader line was measured as expected from theory. However, now using the CDFDW in the spectrograph setup, we can measure and analyze the CDFDW spectrum directly, without the need of another CDFDW optics as an analyzer. Using Eq. [1] with \( F = 1.38 \, \text{m} \), \( \Delta x = 32 \, \mu \text{m} \), and the theoretically estimated \( D_\perp = 270 \, \mu \text{rad}/\text{meV} \), we obtain \( \Delta E = 86 \, \mu \text{eV} \) in agreement with the measured energy resolution.

In conclusion, a principle is proposed and demonstrated how to enhance by more than an order of magnitude the angular dispersion rate of x-rays in Bragg diffraction, namely by successive asymmetric Bragg reflections. This effect opens an opportunity of realizing dispersing elements in the hard x-ray regime with an angular dispersion rate sufficiently large for x-ray spectrographs. The hard x-ray spectrograph principle is demonstrated with the multi-crystal multi-reflection CDFDW optics as dispersing element, by imaging an x-ray spectrum of 9.1315 keV photons in a 450 \( \mu \text{eV} \) window with a record small 90 \( \mu \text{eV} \) resolution, thus achieving spectral resolution power beyond \( 10^8 \) in hard x-ray regime. The main future effort should be directed not only to further improving the spectral resolution, but primarily into making the spectral window broader, to enhance the spectrographs’ throughput. Hard x-ray spectrographs can advance significantly research using high-resolution x-ray spectroscopies, in particular different branches of inelastic x-ray scattering, using synchrotrons and seeded XFELs.

We are grateful to L. Young for supporting this project at the APS, to S. Collins and G. Materlik at the DLS.

FIG. 3: Image of the CDFDW spectral function on the spatial x-scale (b) measured in the prototype spectrograph setup (a).
D. Shu, T. Roberts, K. Goetze, J. Kirchman, P. Jemian, M. Upton, and Y. Ding are acknowledged for technical support. R. Lindberg and X. Yang are acknowledged for reading the manuscript and valuable suggestions. Work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

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