

#### OPTICAL COMPONENTS FOR HARD X-RAY SYNCHROTRON RADIATION SOURCES



**DENNIS MILLS** Deputy Associate Laboratory Director Advanced Photon Source

National School for Neutron and X-ray Scattering July 2022

#### **Outline of Presentation**

- 1. Historical Perspectives
- 2. Why Do We Need Optics?
- 3. X-ray Mirrors (Reflective Optics)
- 4. X-ray Lenses (Refractive Optics)
- 5. Single Crystal, Multilayers & Zone Plates (Diffractive Optics)
- 6. High Heat Load Optics (monochromators)

I will not be discussion gratings as they are used in the soft x-ray region of the spectrum and the focus of this talk will be hard x-ray optics.



#### **X-RAY PHYSICS AND X-RAY OPTICS**

- The first time the term "X-ray Optics" was used (that I could find in the literature) was in Compton's 1927 Nobel Prize lecture: "X-rays as a Branch of Optics"
  - One of the most fascinating aspects of recent physics research has been the gradual extension of familiar laws of optics to the very high frequencies of X-rays, until at the present there is hardly a phenomenon in the realm of light whose parallel is not found in the realm of X-rays. Reflection, refraction, diffuse scattering, polarization, diffraction, emission and absorption spectra, photoelectric effect, all of the essential characteristics of light have been found also to be characteristic of X-rays....
  - It has not always been recognized that X-rays is a branch of optics. As a result of the early studies of Röntgen and his followers it was concluded that X-rays could not be reflected or refracted, that they were not polarized on transversing crystals, and that they showed no signs of diffraction on passing through narrow slits. In fact, about the only property which they were found to possess in common with light was that of propagation in straight lines.
  - Many will recall also the heated debate between Barkla and Bragg, as late as 1910, one defending the idea that X-rays are waves like light, the other that they consist of streams of little bullets called "neutrons".
  - Thus, by a study of X-rays as a branch of optics we have found in X-rays all of the well-known wave characteristics of light, but we have found also that we must consider these rays as moving in directed quanta. It is these changes in the laws of optics when extended to the realm of X-rays that have been in large measure responsible for the recent revision of our ideas regarding the nature of the atom and of radiation.



### **IMAGING WITH X-RAYS**

- This year is the 75<sup>th</sup> anniversary (actually 74<sup>th</sup>, but close enough) of Paul Kirkpatrick and Albert Baez's famous 1948 paper on the *"Formation of Optical Images by X-Rays".*
  - Several conceivable methods for the formation of optical images by x-rays are considered...
  - Point images of points and therefore extended images of extended objects may be produced by causing the radiation to reflect from two concave mirrors in series....



Paul Kirkpatrick 1894 - 1992

- Born on South Dakota to a family of homesteaders
- Ph.D from Berkeley specializing in X-ray research
- Spend time at U of Hawaii and Cornell before becoming a professor at Stanford
- Pioneer in using x-rays for imaging/microscopy
- Published "An Approach to X-ray Microscopy" in Nature, 1950
- Early practitioner of holography



Albert Baez 1912 - 2007

- With Kirkpatrick as his advisor, he wrote his thesis, "Principles of X-Ray Optics and the Development of a Single Stage X-Ray Microscope" in 1950.
- 1952 he outlined the theoretical advantages and fabrication methods of zone plates for EUV/Soft x-rays
- A lifelong pacifist, he opposed nuclear weapons buildup of the 1950s and, later, the Vietnam War.
- His daughter, Joan Baez, was a singer/song writer who was induced into the Rock and Roll Hall of Fame in 2017



## **X-RAY OPTICS**

- Control the energy (E) and bandwidth ( $\Delta E$ ) of the beam.
  - ∆E = 1-2 keV @ 10 keV; ∆E/E = 10<sup>-1</sup> (wide bandpass to increased flux for time-resolved studies – lectures latter this week)
  - ∆E = 1-2 eV @ 10 keV; ∆E/E = 10<sup>-4</sup> (typical diffraction exp.)
  - ΔE = a few milli-eV @ 10 keV; ΔE/E = 10<sup>-7</sup> (inelastic scattering lecture on Monday)
- Control the size/divergence of the beam (often related).
  - Micro- or nano-beams (spot sizes microns to 10's of nanometers)
  - Highly collimated beams
- Control the polarization of the beam.
  - Linear
  - Circular (magnetic x-ray scattering or spectroscopy)

5





# **REFLECTIVE OPTICS: X-RAY MIRRORS**



#### **INDEX OF REFRACTION FOR X-RAYS:** N < 1

This expression for the (real part) index of refraction:

n = 
$$[1 - (n_e(e^2/mc^2) \lambda^2/\pi)]^{1/2} \approx 1 - (n_e r_e/2\pi)\lambda^2$$

is usually written as:

n = 1 -  $\delta$ , where  $\delta = (n_e r_e/2\pi)\lambda^2$ .

varies as the density and the square of the wavelength.

and  $r_e = (e^2/mc^2)$  is the classical radius of the electron (2.82 x 10<sup>-13</sup> cm),  $n_e$  is the electron density, and  $\lambda$  is the wavelength of the x-ray.

• When you plug in the numbers for the real part of the index of refraction you find:

 $\delta = 10^{-5}$  to  $10^{-6}$ 

- So you have:
  - an index of refraction less than one
  - differing from unity by only a few ppm

This simple treatment did not include any absorption. A more detailed calculation would result in an expression:

$$n = 1 - \delta - i\beta$$

Where  $\beta = \lambda \mu / 4\pi$ , with  $\mu$  the linear absorption coefficient (I = I<sub>o</sub>e<sup>- $\mu$ t</sup>).

#### **CRITICAL ANGLE FOR TOTAL EXTERNAL REFLECTION**

Let an x-ray (in vacuum, where n<sub>1</sub> = 1) impinge on a material with index of refraction n<sub>2</sub>. From Snell's Law (when φ<sub>2</sub> = 90°), we have:

 $n_1 sin(\phi_c) = n_2 sin(90^\circ);$ 

$$\cos(\theta_c) = n_2 \cos(0) \quad (\theta = 90^\circ - \phi)$$

 $\cos(\theta_c) = n_2$ 

 Expanding the cosine of a small angle and substituting for n<sub>2</sub> in the above equation gives:

$$1 - \frac{1}{2}(\theta_c)^2 = 1 - \delta$$
  
 $\theta_c = (2\delta)^{1/2}$ 

 $\theta_c$  is the so-called **critical angle**, the angle at which there is **total external reflection** and the material behaves like a mirror.



Recall that the typical values for δ at 1 Å is 10<sup>-5</sup> to 10<sup>-6</sup> and so the critical angle is going to be about 10<sup>-3</sup> or a few milliradians



## X-RAY REFLECTIVITY

 $\theta_{c}$  is the so-called **critical angle**, the angle at which there is total external reflection and the material behaves like a mirror.

 $\theta_{c} = (2\delta)^{1/2}$ 

- The amplitude of the reflected wave can be determined through the Fresnel equations. Sparing you the details, it can be shown that:
  - **Below**  $\theta_{c}$ , there is unit reflectivity (when  $\beta$ , the absorption. equals 0)
  - Above  $\theta_c$ , the reflectivity falls rapidly



Below  $\theta_c$ , there is unit reflectivity (if  $\beta = 0$ )



9

#### ENERGY CUTOFF FOR A FIXED ANGLE-OF-INCIDENCE MIRROR

- Often mirrors are used as first optical components. This means a polychromatic incident beam strikes the mirror at some fixed angle.
- The relationship for the critical angle and wavelength can be re-written, for a fixed angle of incidence θ, in terms of a critical wavelength, λ<sub>c</sub>, where wavelengths above λ<sub>c</sub> are reflected and those below λ<sub>c</sub> are not. Since E = hc/λ, I can re-write this and get a relationship for a fixed incident angle. θ, and determine the maximum, or cut-off energy, E<sub>cut off</sub>, that will be totally reflected by the mirror.

$$\theta_{c} = (2\delta)^{1/2} = \lambda (n_{e}r_{e}/\pi)^{1/2}$$

 $E_{\text{cut off}} = hc/\lambda_c = (hc /\theta) (n_e r_e/\pi)^{1/2}$ 



Critical or cut-off energy,  $\mathsf{E}_{\mathsf{cut\,off}}$  for fixed angle  $\theta$ 



#### **GRAZING INCIDENCE X-RAY MIRRORS**

- Because the incidence angle are small (a few milliradians) to capture the full extent of the beam (say 1 mm or so), x-ray mirrors tend to be very long (sometimes over a meter).
- Low-pass filters
  - mirrors can be used to effectively suppress high energies
  - mirrors are designed so that the cutoff energy, E<sub>cut off</sub>, can be varied by having several different coatings deposited on the mirror substrate
- Mirrors can effectively remove a considerable amount of the heat from the raw (incident) beam and reduce the thermal loading on downstream optics.



Courtesy Chandra mission website: http://chandra.harvard.edu



Chandra's mirrors are positioned so they're almost parallel to the entering X-rays. The mirrors look like open cylinders, or barrels. The X-rays skip across the mirrors much like stones skip across the surface of a pond.



Most mirrors are made from silicon coated with one or multiple stripes of high-Z material after polishing.

11





 Two orthogonal singly focusing mirrors, from which incident Xrays reflect successively, was first proposed in 1948 by Kirkpatrick and Baez (KB).

# REFRACTIVE OPTICS: X-RAY LENSES



#### **COMPOUND REFRACTIVE LENSES**







 $1/F = \delta (1/R_1 + 1/R_2 + etc.)$ 

• For a single lens:



Compound Refractive Lens





If we have N surfaces, all with radius r:

 $F = R/2N\delta$ 

Using the same numbers as before but with 50 lenses, i.e.:

R = 1 mm  $\delta \approx 10^{-5}$  N = 50

- Then the focal length, F, would be at 1 m.
- These lenses focus at rather larger distances and are well adapted to the scale of synchrotron radiation beamlines.
  Argonne A



These are chromatic, i.e. the focal length is dependent on x-ray wavelength since  $\delta$  is a function of  $\lambda$ .

#### FOCUSING IN ONE DIMENSIONS WITH **REFRACTIVE LENSES**

- Planar technologies
  - Leverage planar technologies from micro-electronics industry
  - Fabricate compound lens systems in a small space
  - Small radius means moderate focal spots with a single lens or nano-focusing with a moderate number of lenses



Sawtooth lens - The amount of lens material projected on the lateral plane is a (nearly) parabolic profile Vary opening angle to keep focal length fixed as energy is changed or to vary the focal length





Parabolic lenses etched 400 µm deep into Si wafer made at CNM and tested at APS. The gray shaded area is one lens. At left is focusing performance at 87 keV.



#### FOCUSING IN TWO DIMENSIONS WITH REFRACTIVE LENSES

- 2-D lenses typically "embossed" and typically made from Be, Al or Ni
- Spherical lenses are easy to make but suffer from spherical aberrations.
- Paraboloids eliminate spherical aberrations.





b

# DIFFRACTIVE OPTICS: SINGLE CRYSTALS, MULTILAYERS & ZONE PLATES



#### **DIFFRACTION FROM PERFECT CRYSTALS**

 The theory that describes diffraction from perfect crystals is called <u>dynamical diffraction theory</u> (as compared with kinematical theory, which describes diffraction from imperfect or mosaic crystals) first proposed in 1914 by C. G. Darwin in two seminal papers.



Mosaic crystal model

Perfect crystal model

In the case of a strong reflection from a perfect crystal of a monochromatic x-ray beam, the penetration of the x-rays into the crystal is not limited by the (photoelectric) absorption, but the beam is attenuated due to the reflecting power of the atomic planes. (This type of attenuation is called "extinction".) " *if the crystal is perfect all the radiation that can be reflected is so, long before the depth at which the rays at a different angle are appreciably absorbed.*"





http://www.eoht.info/p age/C.G.+Darwin

**Aside:** C. G. Darwin was the first to calculate the index of refraction for x-rays. Charles G. Darwin was the grandson of the "more famous" Charles Darwin of evolution fame.



#### TWO CONSEQUENCES OF LIMITED PENETRATION IN DIFFRACTION FROM PERFECT CRYSTALS

- The limited penetration due to extinction (reflection by the atomic planes) means at the Bragg condition, the x-ray beam is limited in the number of atomic planes it "experiences".
- Consequence #1:
  - There is a finite angular width over which the diffraction occurs. This is is often called the *Darwin width*,  $\omega_D$
  - Depends on the strength of the reflection, F(hkl), and square of the wavelength,  $\lambda^2.$
- Consequence #2:
  - The reflectivity over this narrow angular width is nearly unity, even in crystals with a finite absorption.

Using modern notation, Darwin width,  $\omega_D$ , can be written as:

 $ω_{\rm D} = 2r_{\rm e}F(hkl)\lambda^2/\pi Vsin(2\theta)$ 

F(hkl) = structure factor and V = volume of unit cell







#### PERFECT CRYSTAL MONOCHROMATORS

- Simply use Bragg's Law to select a particular wavelength (or energy)  $\lambda = 2d \sin(\theta).$
- If we differentiate Bragg's Law ( $\Delta \lambda = \cos(\theta) \ \Delta \theta$ ), divide this by the original equation and recall that  $\Delta \lambda / \lambda = \Delta E/E$  for small deltas, then we can determine the energy resolution of the monochromator.



 $\Delta\lambda / \lambda = \Delta E / E = \cot(\theta) \Delta\theta$ 

- At 8 keV (1.5Å) for Si(111)  $\omega_D \approx 40$  microradians. Recall that, for an APS undulator, the opening angle is about 5-10 microradians.
- In this case the energy resolution of the mono is determined by the crystal. Plugging in the values you get ΔE/E = 10<sup>-4</sup>. So for at 8 keV x-ray the bandwidth (or ΔE) would be about 0.8 eV.



Silicon is used for monohromtor crystals as as it can be easily obtained and has good thermomechanical properties for cooling.



Synthetic diamonds are also a good choice but much harder to find with the required quality



#### **DOUBLE CRYSTAL MONOCHROMATORS**

- The most common arrangement for a monochromator is the double-crystal monochromator (DCM). It:
  - is **non-dispersive**, that is all rays that diffract from the first crystal simultaneously diffract from the second crystal (if same crystals with same hkl's are used)
  - keeps the beam parallel to the incident beam as the energy is changed (by changing the Bragg angle,  $\theta$ ).
- There is little loss in the throughput using two crystals because the reflectivity is near unity over the Darwin width.



- Monochromators need to be cooled to maintain the desired properties.
  - Silicon monos are often liquid N<sub>2</sub> cooled to enhance thermal properties (higher conductivity and coefficient of thermal expansion goes through a zero at about 120° K).



polychromatic beam going into the slide



#### HARD X-RAY MULTILAYER OPTICS

- A "periodic multilayer" coating is a film stack comprising a number of identical repetitions of two or more optically dissimilar component layers.
  - Wide energy band-pass
  - Focusing :  $\theta_B$  multilayer >>  $\theta_c$  mirror so multilayer length << mirror length
  - Increased numerical aperture
  - Soft X-ray monochromators (when Si atomic spacing is too small)
- The energy is selected using Bragg's Law.
- The energy bandwidth is determined by the number of layers N; ∆E/E ≈ 1/N.





http://xray0.princeton.edu/~phil/Facility/G uides/XrayDataCollection.html

Diffracted beam from a W/B<sub>4</sub>C (d = 27.7 A) on a Si substrate provides an X-ray spectrum with 1% energy bandwidth. The energy of the X-ray spectrum can be tuned by changing the Bragg angle of the multilayer.

"100 ps time-resolved solution scattering utilizing a wide-bandwidth X-ray beam from multilayer optics", Ichiyanagi et al, JSR **16**, 2009.

22



#### FRESNEL ZONE PLATES

- Zone plates are diffraction gratings, that is, structures composed of alternating concentric zones of two materials with different (complex) refractive indices.
- The focusing capability is based on constructive interference of the wavefront modified by passage through the zone plate. The wave that emerges from the zone plate is the superposition of spherical waves, one from each of the zones.



Cape Meares Lighthouse (Oregon); first-order Fresnel lens

In general, the size of the **focal spot from the zone plate**,  $\Delta x$ , is determined by the **width of the outermost ring**,  $\Delta r_{out}$ , and is given by:

$$\Delta x = 1.22 \Delta r_{out}$$

 The challenge in making zone plates for hard x-rays is making ∆r<sub>out</sub> small while maintaining a high thickness for efficiency) i.e. a high aspect ratio.



Zone plates are chromatic, i.e. the focal length is dependent on x-ray wavelength.





#### VARIOUS OPTICAL COMPONENTS ARE OFTEN FOUND IN A SINGLE BEAMLINE



Design of an ultrahigh-resolution crystal monochromator and analyzer (from sub-10 meV to submeV) for inelastic x-ray scattering.





#### **HIGH HEAT LOAD X-RAY OPTICS**



#### THERMAL LOADING ON OPTICAL COMPONENTS

 Along with the enormous increase in x-ray beam brilliance from insertion devices comes unprecedented powers and power densities that must be effectively handled so that thermal distortions in optical components are minimized and the full beam brilliance can be delivered to the sample.

<u>Process</u>	Approx. Heat Flux (W/mm <sup>2</sup> )
Interior of rocket nozzle	10
Commercial plasma jet	20
Fusion reactor components	0.05 to 80
Meteor entry into atmosphere	100 to 500
APS Undulator @ 30m (on-axis 2.4 m 100 mA)	10 to 160



In order to maintain the beam intensity and collimation (i.e., brightness) through the optics, special attention must be paid to the issue of thermal management for those first optical components.



#### **PROPERTIES OF SI, GE, AND C(DIAMOND)**

Thermal gradients,  $\Delta T$ , and coefficient of thermal expansion,  $\alpha$ , contribute to crystal distortions:

 $\alpha \Delta T = \Delta d/d$ 

We therefore need to look for materials that have a very low coefficient of thermal expansion,  $\alpha$ , and/or have a very high thermal conductivity, k, so that the material cannot support large  $\Delta T$ 's.



#### FIGURE OF MERIT (FOM) OF TYPICAL X-RAY MONOCHROMATOR MATERIALS

#### **FOM of Typical Monochromator Materials**

Material and Temperature	Thermal Conductivity (k)	Coef. Thermal Expansion ( $\alpha$ )	Figure of Merit FOM (k/α )
Si (300K)	1.2 W/cm-K	2.3 x 10 <sup>-6</sup> /K	0.5
Si (78K)	14 W/cm-K	-0.5 x 10 <sup>-6</sup> /K	28
Dia. (300K)	20 W/cm-K	0.8 x 10 <sup>-6</sup> /K	25

These properties motivate us to use cryogenically cooled silicon or room temperature diamond as high heat load monochromators.



#### LN<sub>2</sub> COOLED Si MONOCHROMATORS



The historical development of cryogenically cooled monochromators for third-generation synchrotron radiation sources, Bilderback, Freund, Knapp, and Mills, J. Synch. Rad. 7, 2000.





#### **SUMMARY**

- X-ray optics is an active area of research at both universities and national laboratories.
- High-brightness sources provide new opportunities but ever higher demands on the quality of optics to ensure beam coherence is preserved through the optics.
- Metrology is key to making good optics "You Can't Improve What You Can't Measure"





High-reflectivity high-resolution X-ray crystal optics with diamonds

Yuri V. Shvyd'ko1\*, Stanislav Stoupin<sup>1</sup>, Alessandro Cunsolo<sup>1,2</sup>, Ayman H. Said<sup>1</sup> and Xianrong Huang<sup>2</sup>

SCIENTIFIC **REPORTS OPEN** Interlaced zone plate optics for hard X-ray imaging in the 10 nm range Received: 18 August 2016 Istvan Mohacsi<sup>1,2</sup>, Ismo Vartiainen<sup>1,3</sup>, Benedikt Rösner<sup>1</sup>, Manuel Guizar-Sicairos<sup>1</sup>, Accepted: 26 January 2017 Vitaliy A. Guzenko<sup>1</sup>, Ian McNulty<sup>4</sup>, Robert Winarski<sup>4</sup>, Martin V. Holt<sup>4</sup> & Christian David<sup>1</sup>

Department of Physics & Astronomy, Purdue University, West Lafavette Indiana 47907, USA



Correct interpretation of diffraction properties of guartz crystals for X-ray optics applications. Corrigendum

Xian-Rong Huang,<sup>a</sup>\* Thomas Gog,<sup>a</sup> Jungho Kim,<sup>a</sup> Elina Kasman,<sup>a</sup> Ayman H. Said,<sup>a</sup> Diego M. Casa,<sup>a</sup> Michael Wieczorek,<sup>a</sup> Marcelo G. Hönnicke<sup>b</sup> and Lahsen Assoufid<sup>a</sup>

Received 27 February 2018 Accepted 14 March 2018

30

"Advanced Photon Source, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439, USA, and <sup>b</sup>Instituto de Ciencias da Vida e da Natureza, Universidade Federal da Integração Latino-Americana, 2044 Foz do Iguaçu, Parana, 85867-970, Brazil. \*Correspondence e-mail: xiahuang@aps.anl.gov

# QUESTIONS



# DETECTORS



## **DETECTOR PROPERTIES**

- We would like detectors to have *all* of the following characteristics (but they never do):
  - high efficiency (count every photon or make every photon count)
  - high count rate capabilities
    - single photon counting detectors that can "process" one x-ray at a time very fast or
    - integrating detectors that accumulate (integrate) over a fixed period of time, and the detector output is proportional to the total number of x-rays detected)
  - good spatial resolution for 1D (linear) or 2D(area) detectors - a few microns would be nice but that is very hard
  - energy resolution (signal proportional to energy of the incident x-ray)



## **ENERGY RESOLVING DETECTORS - SSDs**

- Solid state detectors (SSDs) are basically large reversebiased pin diodes that collect the electrons (holes) that are made as the x-ray is absorbed in a biased diode.
- They have quite good energy resolution (△E/E of 3-5%) since the energy required to generate and electron-hole pair is on the order of 3 eV in silicon and germanium.
- To keep the leakage currents as low as possible, SSDs are typically cooled to near liquid nitrogen temperature.







## **ENERGY RESOLVING DETECTORS – TES**

- Transition edge superconducting (TES) detectors
  - Currently under development, they work at less than a degree above absolute zero but have energy resolutions 3-15 times better than SSDs (but are very slow).
  - To get around the low count rates assemble arrays of individual detectors.
  - The sensor array shown at the bottom is a Mo/Cu bilayer on a SiN suspended membrane (developed at APS) that operates at  $T_c$ ~100 mK.





The TES is biased on the superconducting transition to maximize  $\alpha$ , a measure of the sensitivity of the device.





## **AREA DETECTORS - CCDs**

- The original area detector was x-ray sensitive film.
- Now a days, all area detectors are electronic.
- Charge Coupled Detectors (CCDs)
  - These are basically the same devices that are in your phone:
    - a larger number of pixels (4096 x 4096)
    - smaller pixel size (10 microns)
    - higher resolution readout (16 bits)
  - To get a large area, can be tiled or coupled to a tapered fiber optic with a scintillator



PHOSPHOR X-RAY PHOTON VISIBLE LIGHT PHOTONS FIBEROPTIC TAPER (3.6:1 TAPER RATIO) B ELECTRONS IN CCD PIXEL VISIBLE LIGHT PHOTONS



# **AREA DETECTORS - (PADS)**

- In a pixel array detector (PAD) each pixel is a stand alone detector (usually a diode) that has its own electronics.
- Electronics can be tailored to the specific needs of the experiment via application specific integrated circuits (ASICs)
  - single channel analyzers
  - autocorrellators
  - local background subtraction
  - lock-in techniques
- High framing rates possible with built in memory, but total number of frames limited



## THE PROBLEM

Over the next decade, the 5 Light Sources are projected to generate ~ 1 exabyte of data/year and will require 10s of petaflop/s to an exaflop/s of peak computing power



#### 1 exabyte/year = 1.5 million Netflix movies every day

We don't need to just watch these movies, we need to look at every frame of every movie, analyze it in near real-time, make decisions about what to do next based on that analysis and archive the data.

#### 1 exaflop/s = 500,000 servers

This will require up to 1 exaflop of peak compute power, fast networks (multiple Tbs<sup>-1</sup>), archival storage, and a robust software infrastructure to support near real-time analysis.



Slide courtesy of Nicholas Schwarz (APS)