Elements of Modern X-ray Physics

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About this course

“To explain the physics underlying the production and exploitation of X-rays with emphasis on application in condensed matter and materials physics”

1. Sources of X-rays

2. X-rays and their interaction with matter: scattering

3. Case studies

4. X-ray imaging
1. Introduction: contact region, the near and far fields

2. Absorption contrast imaging
   - Radiography and tomography
   - Microscopy

3. Phase contrast imaging
   - Free-space propagation
   - Grating interferometry

4. Coherent diffraction imaging
   - Coherent beams and speckle patterns
   - Phase retrieval via oversampling

5. Holography
Why X-ray imaging?
Advantages for real space imaging

1. Imaging contrast:

Refractive index $n=1-\delta+i\beta$ \quad $\beta<\delta<<1$
charge, magnetic, orbital….

2. Penetrative power:

$$\sigma_{abs} \propto \left(\frac{\hbar \omega}{Z}\right)^{-3} Z^4$$

3. Spatial resolution:

$$\lambda_{x-rays} \ll \lambda_{light}$$

4. Temporal resolution:

$$< 100 \text{ fs @ XFELs}$$

5. Coherence:

Holography, Lensless imaging
From the near to the far field
Fresnel and Fraunhofer diffraction

P and O illuminated by an incident plane wave radiate outgoing circular waves detected at D.

In the far-field limit (Fraunhofer diffraction) the phase difference is $Q \cdot r = -k' \cdot r$.

In the near field (Fresnel diffraction) must allow for shortening of path length difference by $\Delta = OF' - OF = R - R \cos \psi \approx \frac{a^2}{2R}$.

Expect far-field limit to apply when $\lambda \ll \Delta$.

Fraunhofer region: $R \gg \frac{a^2}{\lambda}$
Fresnel region: $R \approx \frac{a^2}{\lambda}$
Contact region: $R \ll \frac{a^2}{\lambda}$
The contact, Fresnel and Fraunhofer regions

5 micron disks
red: absorption only
blue: phase only

Image courtesy of Timm Weitkamp
Relationship between scattering and refraction

Resonant scattering

\( f(Q, \hbar \omega) = f^0(Q) + f'(\hbar \omega) + i f''(\hbar \omega) \)

Rayleigh scattering

Visible light

Thomson scattering

X-rays

Refractive index

\( n = 1 - \delta + i \beta \)

\( \delta = (f^0(0) + f') \frac{2 \pi \rho_a r_0}{k^2} \)

\( \beta = -f'' \left( \frac{2 \pi \rho_a r_0}{k^2} \right) \)

Scattering and refraction: different ways of understanding the same phenomena
Relationship scattering, refraction and absorption

\[ n = 1 \quad n = 1 - \delta + i \beta \]

\[ \delta = \left( \frac{2\pi \rho_a (f^0(0) + f'' r_0)}{k^2} \right) \]

\[ \beta = \left( \frac{2\pi \rho_a f'' r_0}{k^2} \right) \]

Absorption coefficient \( \mu \) defined by \( I = I_0 e^{-\mu z} \) and absorption cross-section \( \sigma_a = \mu / \rho_a \)

\[ f'' = -\left( \frac{k^2}{2\pi \rho_a r_0} \right) \]

\[ \frac{\mu}{2k} = -\left( \frac{k}{4\pi r_0} \right) \sigma_a \]

Absorption is proportional to the imaginary part of the forward scattering amplitude (Optical Theorem)
X-ray absorption edges

Absorption cross-section scales as

$$\sigma_{abs} \propto (\hbar \omega)^{-3} Z^4$$

$$\varepsilon_0 = 12.398 \text{ keV} \approx 1 \text{ Å}$$

$$H_1 = \left( \frac{e^2}{2mc^2} \right) A^2 + \left( \frac{e}{mc} \right) A \cdot p$$

$$H_1$$  $$H_2$$
Absorption contrast imaging
Computer axial tomography (CAT)

Brain courtesy of Mikael Haggstrom
The Radon transform $R(\theta, x')$

The line integral of the absorption coefficient can be determined from the ratio $I_0/I$.

The Radon transform is the line integral evaluated at a specific viewing angle $\theta$ as a function of the coordinate $x'$ perpendicular to the direction of viewing.
Fourier slice theorem

Image reconstruction from the Radon transform

\[ p(x) = \int f(x, y) \, dy \]

\[ P(q_x) = \int p(x) \, e^{i q_x x} \, dx \]

\[ F(q_x, q_y = 0) = \int p(x) \, e^{i q_x x} \, dx = P(q_x) \]

\[ F(q_x, q_y) = \iiint f(x, y) \, e^{i(q_x x + q_y y)} \, dx \, dy \]
Example of image reconstruction from the Radon transform

- (a) $\theta = 0$
- (b) $\theta = 90^\circ$
- (c) Model $f(x,y)$
- (d) Sinogram
- (e) Reconstructed $f(x,y)$
CAT
Three dimensional image reconstruction

Commercial, fully automated, desktop systems with resolution < 10 microns available from various companies
For X-rays, $n < 1$ which implies total external reflection for angles below a critical angle of order 0.1 degrees.

Due to absorption, refractive lenses operate efficiently above 10 keV.
X-ray lenses
The ideal shape

Applying Fermat’s Theorem to optical path lengths

\[ AF = P'P + PF \]

leads to

\[ x^2 + (2\delta - \delta^2)y^2 - 2f\delta y = 0 \]

c.f. equation of an ellipse

\[ x^2 + (a/b)^2 y^2 - 2(a^2/b)y = 0 \]

Hence

\[ b = \frac{f}{2 - \delta} \approx \frac{f}{2} \]

and

\[ a = f\sqrt{\frac{\delta}{2 - \delta}} \approx f\sqrt{\frac{\delta}{2}} \]

Therefore

\[ \Delta x = 1.22\left(\frac{\lambda f}{2a}\right) = 1.22\left(\frac{\lambda}{\sqrt{2\delta}}\right) \]
X-ray Fresnel zone plates

Kinoform

Performance of lens is insensitive to relative change in the optical path length by an integer number of wavelengths

\[ \Lambda = (N + 1)\lambda_0 = N\lambda = N \left(1 + \delta \right) \lambda_0 \]

For X-rays \( \Lambda \sim 10\text{-}100\mu\text{m} \).

[Evans-Lutterodt et al., 2003]
X-ray Fresnel zone plates

Binary approximation

Zone radius:

\[ r_m^2 + f^2 = \left(f + \frac{m\lambda}{2}\right)^2 \]

\[ r_m \approx \sqrt{m\lambda f} \]

Outermost zone width:

\[ \Delta r_M = \sqrt{\lambda f} \left(\sqrt{M} - \sqrt{M-1}\right) \]

\[ \Delta r_M \approx \frac{\sqrt{\lambda f}}{2\sqrt{M}} \]
X-ray Fresnel zone plates

Spatial resolution

The resolution of a FZP depends on the width of the outermost zone.

State-of-the-art is around 15 nm

\[ \Delta x = 1.22 \frac{\lambda f}{D} \]

\[ f = 4M \frac{(\Delta r_M)^2}{\lambda} \]

\[ D = 2r_M = 2 \sqrt{M \lambda f} = 2 \sqrt{M \lambda f} = 4M \Delta r_M \]

Image courtesy of Christian David
X-ray Absorption Microscopy
FZP based scanning and full-field transmission microscopes

Magnification

$$M = \frac{d_{O-D}}{d_{S-O}}$$

>1,000
X-ray Absorption Microscopy
Imaging of biological materials

The water window
X-ray Absorption Microscopy

Cell division in a unicellular yeast

Image courtesy of Carolyn Larabell
X-ray Magnetic Circular Dichroism (XMCD)

Photon polarization dependent absorption

Dipole selection rules
\[ \Delta l = \pm 1 \]
\[ \Delta S = 0 \]

Conservation of angular momentum and Pauli exclusion principle produce different absorptions of left and right circular polarized light.
Magnetic Spectro-Microscopy

X-ray Magnetic Linear Dichroism: **Antiferromagnets**

![Graph showing normalized electron yield versus photon energy for LaFeO$_3$.](image)

\[ I \sim \cos^2 \theta \]

X-ray Magnetic Circular Dichroism: **Ferromagnets**

![Graph showing absorption coefficients versus photon energy for L$_2$ and L$_3$ transitions for Fe.](image)

\[ I \sim \cos \theta \]

*Image courtesy of Jo Stohr*
X-ray Magnetic Circular Dichroism (XMCD)

Sum rules for transitions to 3d states

\[ m_{\text{orb}}[\mu_B/\text{atom}] = -\frac{4q(10 - n_{3d})}{r} \]
\[ m_{\text{spin}}[\mu_B/\text{atom}] \approx -\frac{(6p - 4q)(10 - n_{3d})}{r} \]

\[ p = \int_{L_{\text{III}}} (\mu^+ - \mu^-) d\varepsilon \]
\[ q = \int_{L_{\text{III}} + L_{\Pi}} (\mu^+ - \mu^-) d\varepsilon \]
\[ r = \int_{L_{\Pi} + L_{\Pi}} (\mu^+ + \mu^-) d\varepsilon \]

Iron thin films, Chen et al. PRL (1995)
X-rays can pick materials apart: layer-by-layer

Image courtesy of Jo Stohr
X-Ray Microscopy Methods - toward Nanometer Resolution

Present resolution in the 20 - 40 nm range

Image courtesy of Jo Stohr
X-ray magnetic microscopy

STXM

Image courtesy of Jo Stohr
Alignment of Ferromagnetic by Antiferromagnetic Domains
Nolting et al., Nature 405, 767 (2000)

a) LaFeO$_3$ layer

b) Co layer

Normalized Intensity (a.u.)

Normalized Intensity (a.u.)

Photons Energy (eV)

2 µm
Phase contrast imaging

\[ n = 1 - \delta + i\beta \]
Phase contrast imaging

The phase of the wave is: $\phi(r) = \mathbf{k}' \cdot \mathbf{r}$

While the direction of propagation is:

$$\hat{\mathbf{n}} = \mathbf{k}'/|\mathbf{k}'| = (\lambda/2\pi)\nabla \phi(r)$$

Angular deviation perpendicular to beam:

$$\alpha_x = \frac{\lambda}{2\pi} \frac{\partial \phi(x, y)}{\partial x} \quad \text{and} \quad \alpha_y = \frac{\lambda}{2\pi} \frac{\partial \phi(x, y)}{\partial y}$$

(a) Homogeneous

$$\alpha = \frac{\lambda(1 + \delta) - \delta \lambda}{\Delta x} = \delta \frac{\lambda}{\Delta x} = \delta \tan \omega$$

(b) Inhomogeneous

$$\alpha = \frac{\lambda \Delta x \frac{\partial \delta(x)}{\partial x}}{\Delta x} = \lambda \frac{\partial \delta(x)}{\partial x}$$
Phase contrast imaging

Depends on being able to determine the deflection angle $\alpha$ accurately.

Essentially three methods:

1. Free-space propagation
2. Interferometric techniques
3. Analyser crystals
Phase contrast imaging
Free-space propagation

Beam displacement encodes information on the phase gradient perpendicular to the optical axis
Phase contrast imaging
Example of free-space propagation
Etched silicon (100) wafer

Images courtesy of Martin Bech and Torben Jensen
Phase contrast imaging
Free space propagation

Blood cell infected with malaria parasite

Images courtesy of Martin Bech, Martin Dierolf and Torben Jensen
Phase contrast imaging
Grating interferometry

David et al. (2002), Momose et al. (2003), Weitkamp et al. (2006), Pfeiffer et al. (2006)
Phase contrast imaging
Grating interferometry

Wavevector:
\[ \mathbf{k} = (k_x, k_z) \]

For \( k_x \ll k \)

we have
\[ k_z \approx k - k_x^2/(2k) \]

Propagated wave
\[ e^{i [k z - k_x^2 z/(2k)]} \]

Transverse to optical axis, expect periodic wavefield with \( k_x = 2\pi/p \)

\[ \frac{z}{2k} \left( \frac{2\pi}{p} \right)^2 = 2\pi m \]

Talbot length
\[ d_T = 2p_1^2/\lambda \]
Phase contrast imaging
Grating interferometry

Resolution determined by pixel size, currently around 10 microns
Phase contrast imaging
Grating interferometry

Absorption contrast

Water
Sugar

Differential phase contrast

“Dark field” (scattering)

Fringe Visibility

\[ V = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}} \]

Images courtesy of Franz Pfeiffer and Martin Bech
Phase contrast imaging
Grating interferometry

Absorption contrast
“Dark field” (scattering)
Differential phase contrast

Images courtesy of Franz Pfeiffer and Martin Bech
Phase contrast imaging
Grating interferometry

“Pictures of the Future”
Siemens Magazine for Research and innovation

Images courtesy of Franz Pfeiffer
The Phase Problem

Measure intensity in a scattering experiment

\[ I = |F|^2, \quad F = \sum_j f_j e^{iQ \cdot r_j} \]

and it appears that all information on phase is lost.
Transverse Coherence Length

Sources have a finite size and produce divergent waves

Monochromatic waves are in phase at P, and again after a distance $2L_T$

\[
\therefore 2L_T \Delta \theta = \lambda \quad \text{with} \quad \Delta \theta = \frac{D}{R}
\]

\[
L_T = \frac{1}{2} \frac{\lambda}{(D/R)} = \frac{\lambda}{2} \left( \frac{R}{D} \right)
\]
Longitudinal Coherence Length

Sources are not perfectly monochromatic

\[ 2L_L = N\lambda \]

\[ N\lambda = (N + 1)(\lambda - \Delta\lambda) \]

\[ \Rightarrow N \approx \frac{\lambda}{\Delta\lambda} \]

\[ L_L = \frac{1}{2} \frac{\lambda^2}{\Delta\lambda} \]

Can also define coherence time \( t_0 = L_L / c \)
Photon Degeneracy $\Delta_c$

Photon degeneracy:

The number of photons per eigenstate of the photon field. This is equivalent to the number of photons in a coherence volume.

The photon degeneracy can be calculated from the brilliance:

Thus we have

$$\Delta_c \propto B \times t_0 \times \Delta \Omega_r \times A_S \times \frac{\Delta \nu}{\nu}$$

$B$: brilliance; $t_0$: coherence time $\approx 1/\Delta \nu$; $A_S$: source area

$\Delta \Omega_r$: solid angle subtended by transverse coherence area $\approx \frac{(L_T)^2}{R^2} \approx \frac{1}{R^2} \frac{\lambda^2 R^2}{D^2} = \frac{\lambda^2}{A_S}$

$$\frac{\Delta \nu}{\nu}:$$ bandwidth

Thus

$$\Delta_c \propto \frac{B \lambda^3}{c}$$
Photon Degeneracy $\Delta_c$

Comparison of different sources

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Jens Als-Nielsen and Des McMorrow

P. Guertler, HASYLAB
Imaging using coherent X-rays
Speckle patterns

SAXS thought experiment

single spherical particle
Imaging using coherent X-rays

Time average scattering and instantaneous speckle

SAXS from dilute suspension of 500 nm silica spheres

Coherent beam illumination, average of 200 exposures

Data courtesy of Anders Madsen
X-ray photon-correlation spectroscopy (XPCS)
Direct measurement of antiferromagnetic domain fluctuations

Chromium:
SDW below $T_N = 311$ K
CDW induced at $T_N$
X-ray photon-correlation spectroscopy (XPCS)
Direct measurement of antiferromagnetic domain fluctuations

Time dependent speckle reveals information on the domain dynamics
X-ray photon-correlation spectroscopy (XPCS)
Direct measurement of antiferromagnetic domain fluctuations

\[ g_2(t) = \frac{\langle I(\tau)I(\tau + t) \rangle}{\langle I(\tau) \rangle^2} = 1 + A |F(Q, t)|^2 \]

\[ |F(Q, t)| = a \exp[-(t/\tau_F)\beta] + (1 - a) \exp[-(t/\tau_S)\beta] \]
X-ray photon-correlation spectroscopy (XPCS)
Direct measurement of antiferromagnetic domain fluctuations

Temperature dependent domain wall dynamics
Coherent diffraction imaging
Phase retrieval via oversampling

Three critical factors allow the phase problem to be overcome by illuminating a finite size object of size $L^3$ with a coherent beam (Sayre, 1980):
• Diffraction pattern is extended in reciprocal space over volume $\sim 1/L^3$
• Can sample the diffraction pattern with resolution better than $1/L$
• Phases can be recovered by iterative numerical procedure (Fienup, 1982)
Coherent diffraction imaging
Phase retrieval via oversampling

Example of real space constraint:

$$I(Q) = \int_{-\infty}^{\infty} P(r) e^{iQr} dr$$

$$P(r) = \int_{-\infty}^{\infty} f^*(r') f(r + r') dr'$$

auto-correlation function is a measure of the sample size and can be obtained by the FT of the intensity
Coherent Diffraction Imaging
Phase retrieval via oversampling

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Jens Als-Nielsen and Des McMorrow
Coherent diffraction imaging
Reconstruction of three dimensional electron density

First demonstrated by Miao et al., Nature (1999) for a non-crystalline sample
Coherent diffraction imaging
Reconstruction of three dimensional electron density of a crystal

Real space resolution limited by inverse of maximum $Q$ sampled.

Currently around 30 nm

Data courtesy of Ian Robinson
**X-ray Holography**


- Image produced by interference between object and reference beams
- X-ray resonant magnetic scattering at Co $L_3$ edge
- Fourier transform holography – record interference pattern in far-field limit

Image courtesy of Jo Stohr
X-ray holography
Fourier transform holography

In the far-field limit the intensity can be simply calculated as:

\[ |A(Q)_T|^2 = |A(Q)_R + A(Q)o|^2 \]
\[ = |A(Q)_R|^2 + |A(Q)o|^2 + A(Q)_RA(Q)_o^* + A(Q)_oA(Q)_R^* \]

If reference and object beams are spatially offset, then cross-correlation and auto-correlations terms are offset when the diffraction pattern is Fourier transformed.

From the convolution theorem, the FT of the cross-correlation term is the convolution of the FT of \( A_R(Q) \) and \( A_O(Q) \). In the case that the reference hole is small it may be represented as a delta function, and the convolution integral is then simply the FT of \( A_R(Q) \), or in other words the real space image of the structure.
X-ray holography
X-ray holography
Fidelity of reconstructed image
Where are we heading……

Utilizing the highly brilliant coherent beams from XFELs it is envisaged (hoped!) that in the near future we will be able to:

• Obtain atomic-scale, real-space images of single molecules (particles)

• Have a time resolution of < 100 fs

• Study chemical/biological processes as they happen

...and how far have we come

Femtosecond diffractive imaging with a soft XFEL

- FLASH facility at DESY, Hamburg
- Single pulse 32 nm, 25 fs, $4 \times 10^{13} \text{ Wcm}^{-2}$
- Coherent diffraction imaging
- Real space image reconstructed
- Sample destroyed!