Methoden moderner Röntgenphysik: Streuung und Abbildung

Lecture 2	Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2019 G. Grübel, L. Müller, O. Seeck, L. Frenzel, F. Lehmkühler, M. Martins, W. Wurth		
Location	Lecture hall AP, Physics, Jungiusstraße		
Date	Tuesdays	12:30 - 14:00	(starting 2.4.)
	Thursdays	8:30 - 10:00	(until 11.7.)

Methoden moderner Röntgenphysik: Streuung und Abbildung

Part I:

Basics of X-ray Physics

by Gerhard Grübel (GG)

[2.4.]	Organisation and Introduction
[4.4.]	X-ray Scattering Primer
[9.4.]	Sources of X-rays, Synchrotron Radiation
[11.4.]	Refraction and Reflection
[16.4.]	Kinematical Scattering Theory (I)
[18.4.]	Kinematical Scattering Theory (II), Applications
[23.4.]	Small Angle Scattering and Soft Matter
[25.4.]	Anomalous Scattering
[30.4.]	Introduction: Coherence I
[2.5.]	Coherence II; Applications of Coherent X-ray Beams

Literature

Basic concepts:

Elements of Modern X-Ray Physics
J. A. Nielsen and D. McMorrow, J. Wiley&Sons (2001)

X-Ray Diffraction
B.E. Warren, DOVER Publications Inc., New York

Principles of Optics
M. Born and E. Wolf, Cambridge University Press, 7th ed.

Soft X-rays and Extreme Ultraviolet Radiation
D. Attwood, Cambridge University Press (2000)
http://www.coe.berkeley.edu/AST/sxreuv/

Physik der Teilchenbeschleuniger und Synchrotronstrahlungsquellen
K. Wille, Teubner Studienbücher 1996

Lecture Notes

http://photon-science.desy.de/research/students__teaching/lectures__seminars/ss19

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Laboratory Sources, Accelerator Bases Sources

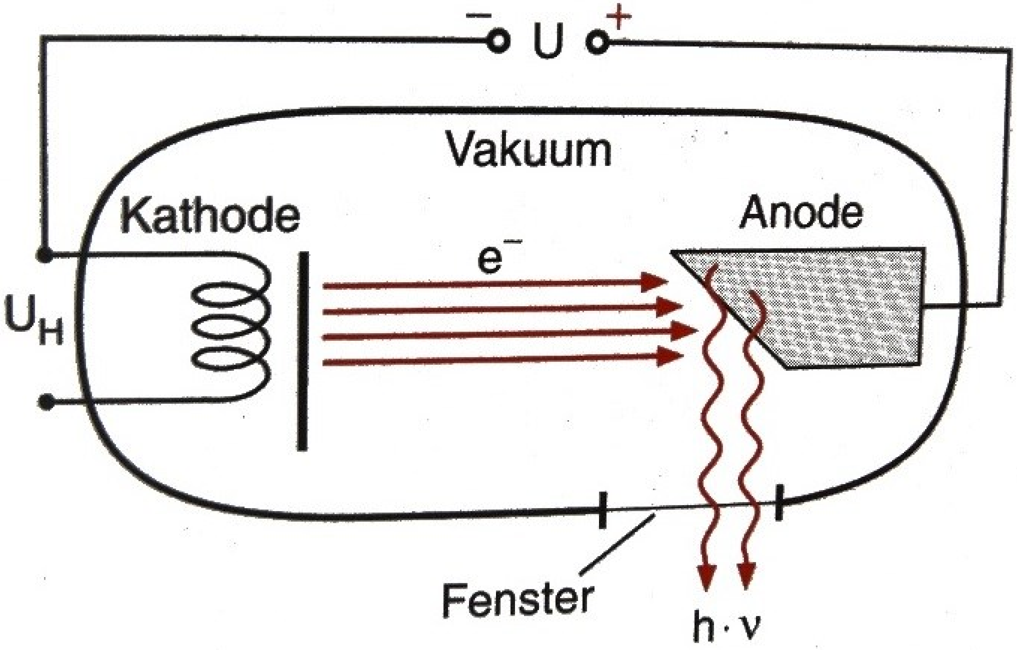
Reflection and Refraction from Interfaces
Snell's Law, Fresnel Equations

Kinematical Diffraction (I)
Diffraction from an Atom, a Molecule, from Liquids, Glasses, …

Kinematical Diffraction (II)
Diffraction from a Crystal, Reciprocal Lattice, Structure Factor, …

Würzburg, 8. November 1895

Wilhelm C. Röntgen (1845 - 1923)



Klassische Röntgenröhre

X-rays

≡

Electromagnetic Radiation

≡

Wavelength

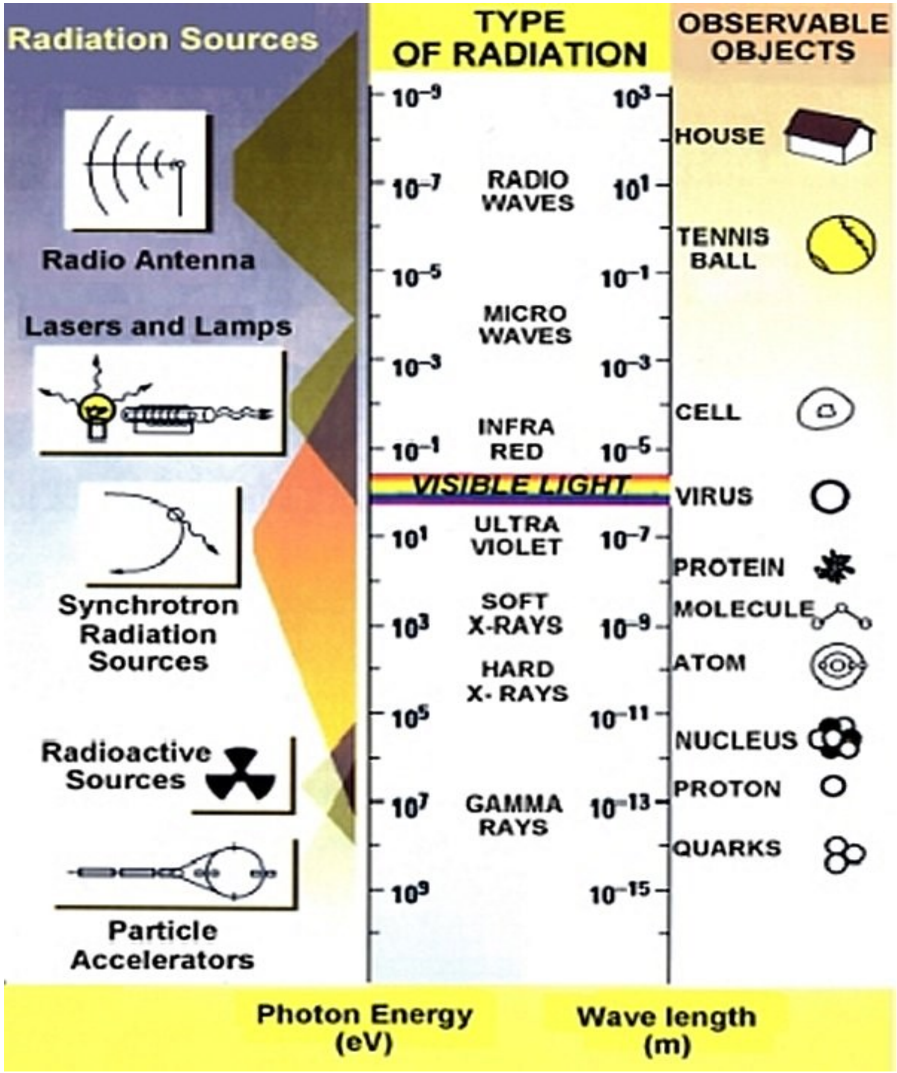
$$(\lambda[\text{Å}] = \frac{\mathbf{12.398}}{\boldsymbol{E}\ [\boldsymbol{keV}]})$$

≡

Object Size

≡

Angstroms for Condensed Matter Research



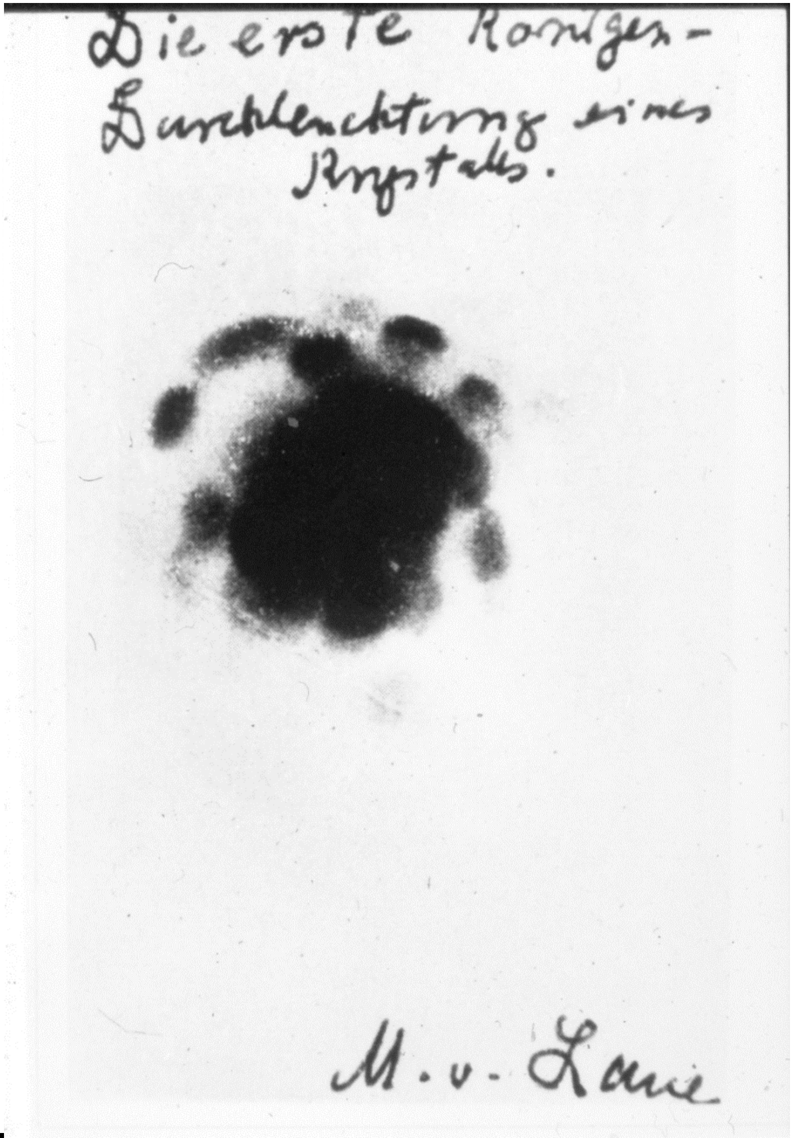
Nobel Prices

1901	W.C. Röntgen in **Physik** für die **Entdeckung der Röntgenstrahlen**
1914	M. von Laue in **Physik** für **Röntgenbeugung an Kristallen**
1915	W.H. Bragg und W.L. Bragg in **Physik** für Bestimmung der **Kristallstruktur mit Röntgenbeugung**
1917	C.G. Barkla in **Physik** für die **charakteristische Strahlung der Elemente**
1924	K.M.G. Siegbahn in **Physik** für **Röntgenspektroskopie**
1927	A.H. Compton in **Physik** für **Streuung von Röntgenstrahlen durch Elektronen**
1936	P. Debye in **Chemie** für **Beugung von Röntgenstrahlen und Elektronen in Gasen**
1946	H.J. Muller in **Medizin** für die Entdeckung von **Mutationen durch Röntgenstrahlung**
1954	L. Pauling in **Chemie** für Entwicklungen in der **Strukturchemie**
1956	A.F. Cournand, W. Forssmann und D.W. Richards in **Medizin** für die **Entwicklung des Herzkatheters unter Röntgenkontrolle**
1962	J. Watson, M. Wilkins und F. Crick in **Medizin** für die **Strukturaufklärung des DNA-Moleküls**
1962	M. Perutz und J. Kendrew in **Chemie** für die **Strukturaufklärung von Hämoglobin**
1964	D.C. Hodgkin in **Chemie** für die **Röntgenstrukturanalyse von Penicillin** und wichtigen biochemischen Substanzen
1976	W.N. Lipscomb in **Chemie** für **Röntgenstrukturuntersuchungen an Boranen**
1979	A.M. Cormack und G.N. Hounsfield in **Medizin** für **Computertomographie**
1981	K.M. Siegbahn in **Physik** für **hochaufgelöste Elektronenspektroskopie**
1985	H.A. Hauptman und J. Karle in **Chemie** für die Entwicklung direkter Methoden zur **Bestimmung von Röntgenstrukturen**
1988	J. Deisenhofer, R. Huber und H. Michel in **Chemie** für die **Bestimmung der dreidimensionalen Struktur von Proteinen für die Photosynthese**
1997	P.D. Boyer, J.E. Walker und J.C. Skou in **Chemie** für **Aufklärung der Funktion des Enzyms ATP**
2002	R. Giacconi in **Physik** für die **Entwicklung der Röntgenastronomie**
2003	R. MacKinnon in **Chemie** für **Röntgenstrukturbestimmung von Ionenkanälen in Zellmembranen**
2009	V. Ramakrishnan, T. A. Steitz, A. E. Yonath in **Chemie** für **Studies of the Structure and Function of the Ribosome**

v. Laue et. al.

Interferenzen am Kristallgitter

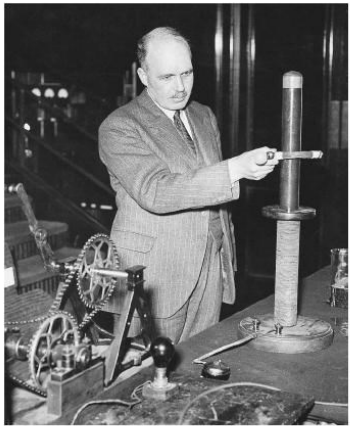
Laue, Friedrich und Knipping 1912

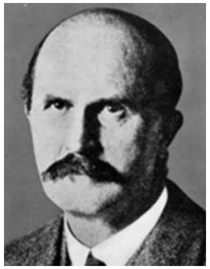


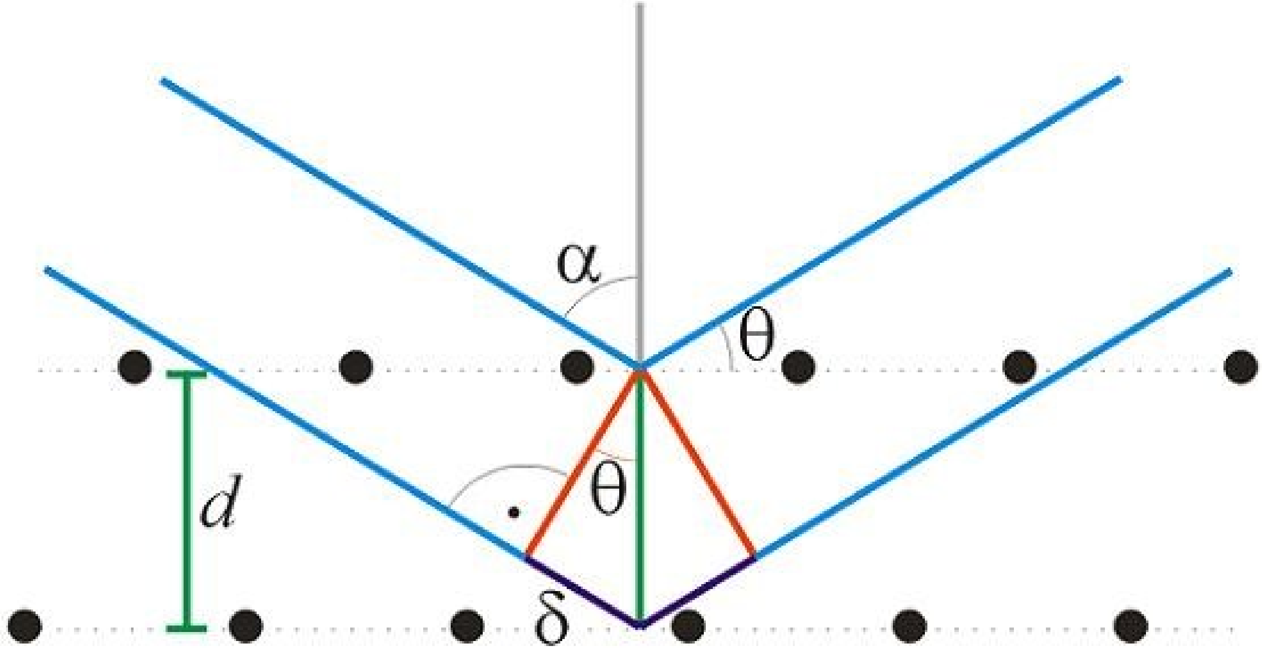
Die erste Röntgen-
Durchleuchtung eines
Krystalls.

M. v. Laue

Bragg's Law

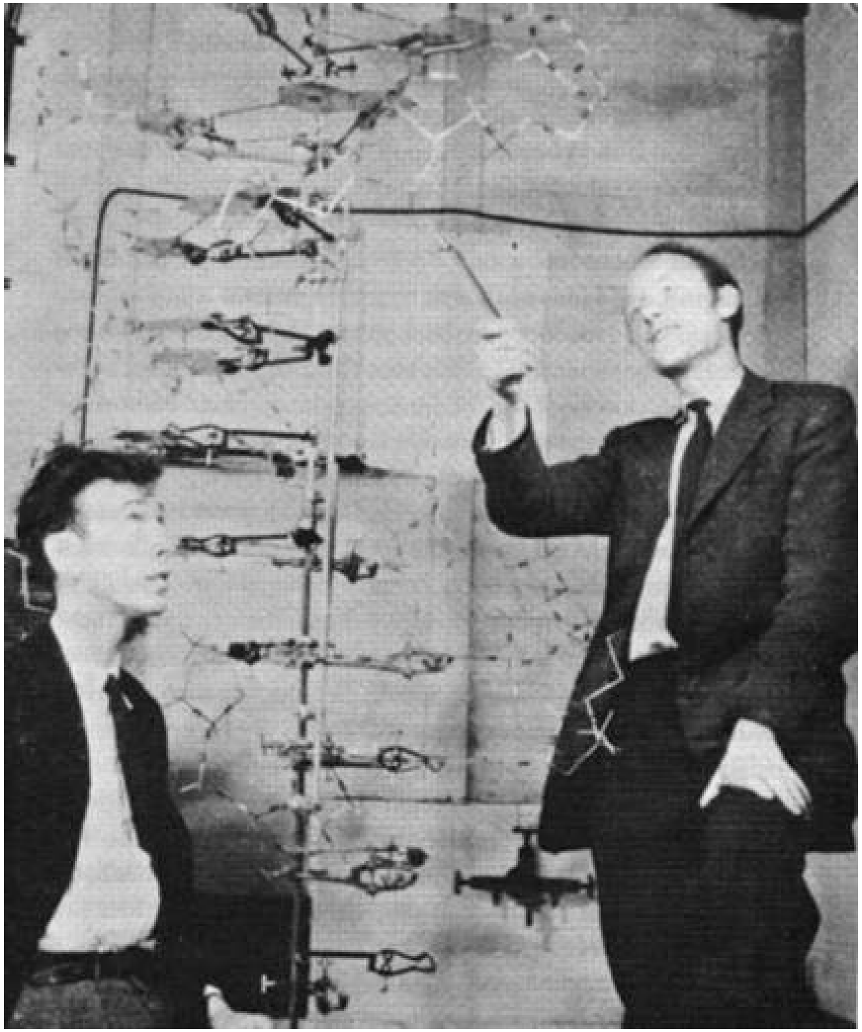




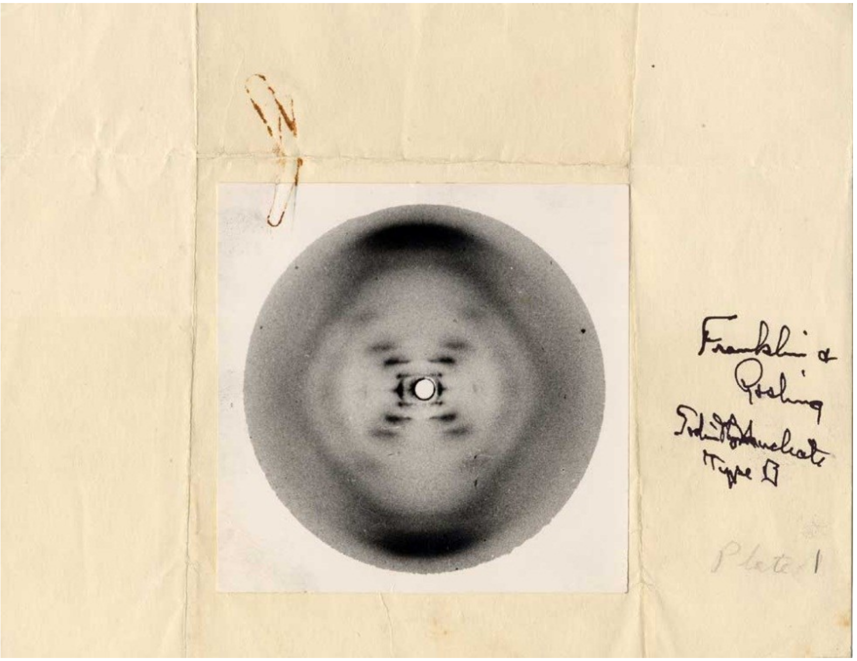


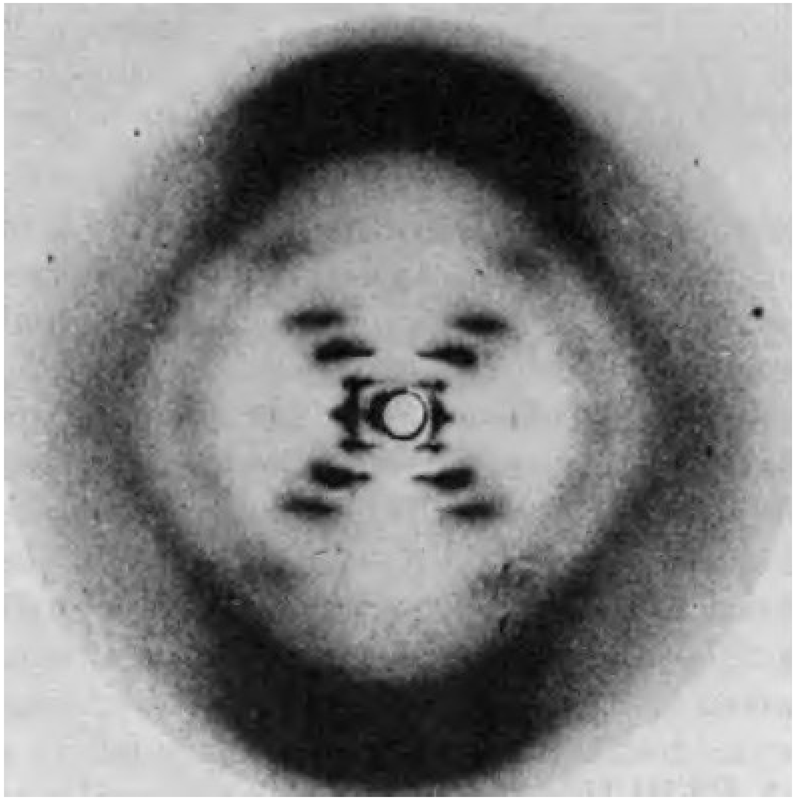
Scattering intensity only if: $n\lambda = 2d\sin(\Theta)$

Watson & Crick 1953

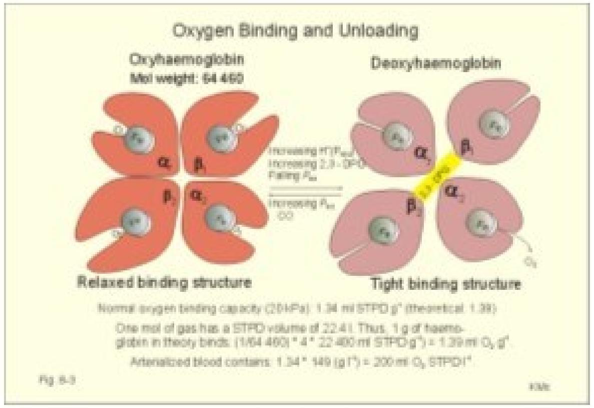


Rosalind Franklin's Famous X-ray that provided enough evidence to establish that DNA was a helix.





Hämoglobin und Myoglobin
Struktur mit Röntgenstrahlen



Normal oxygen binding capacity (20 kPa): 1.34 ml STPD g⁻¹ (theoretical: 1.39)

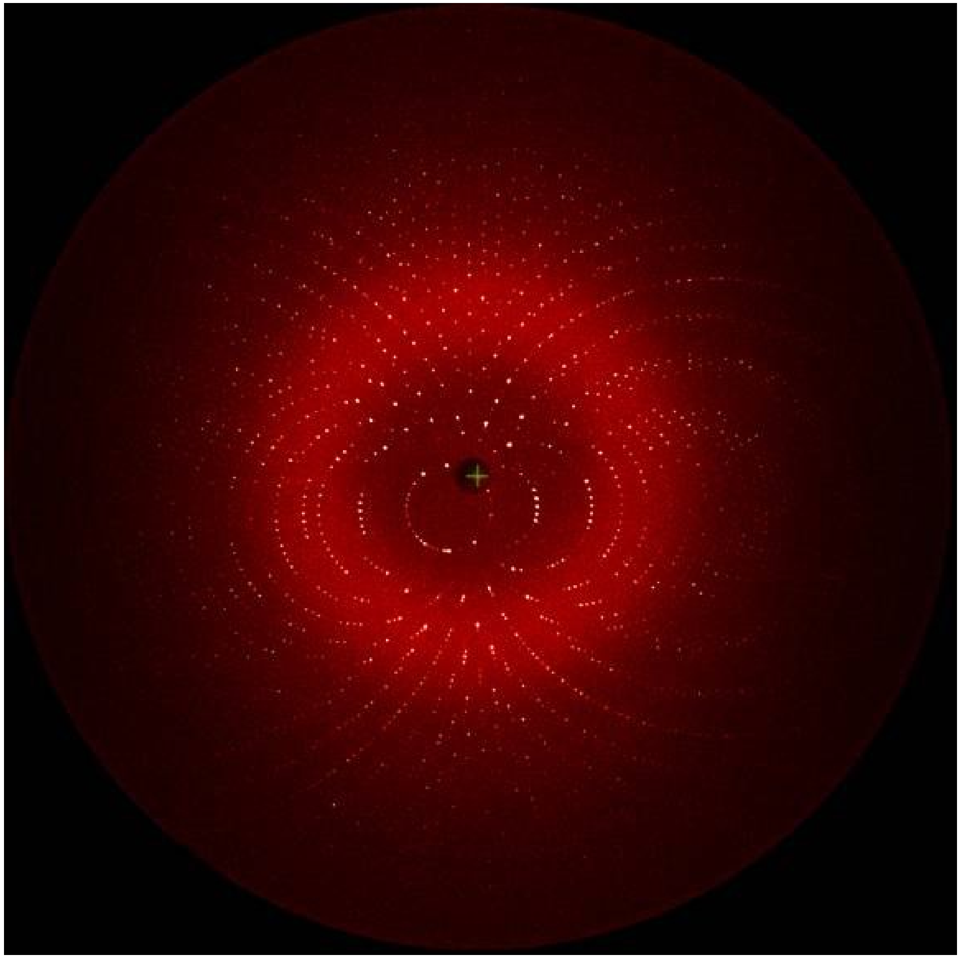
One mol of gas has a STPD volume of 22.4 l. Thus, 1 g of haemoglobin in theory binds: (1/64 460) * 4 * 22 400 ml STPD g⁻¹) = 1.39 ml O_2 g⁻¹

Arterialized blood contains: 1.34 * 149 (g l⁻¹) = 200 ml O_2 STPD l⁻¹

John Cowdery Kendrew
Max Perutz
Nobelpreis 1962

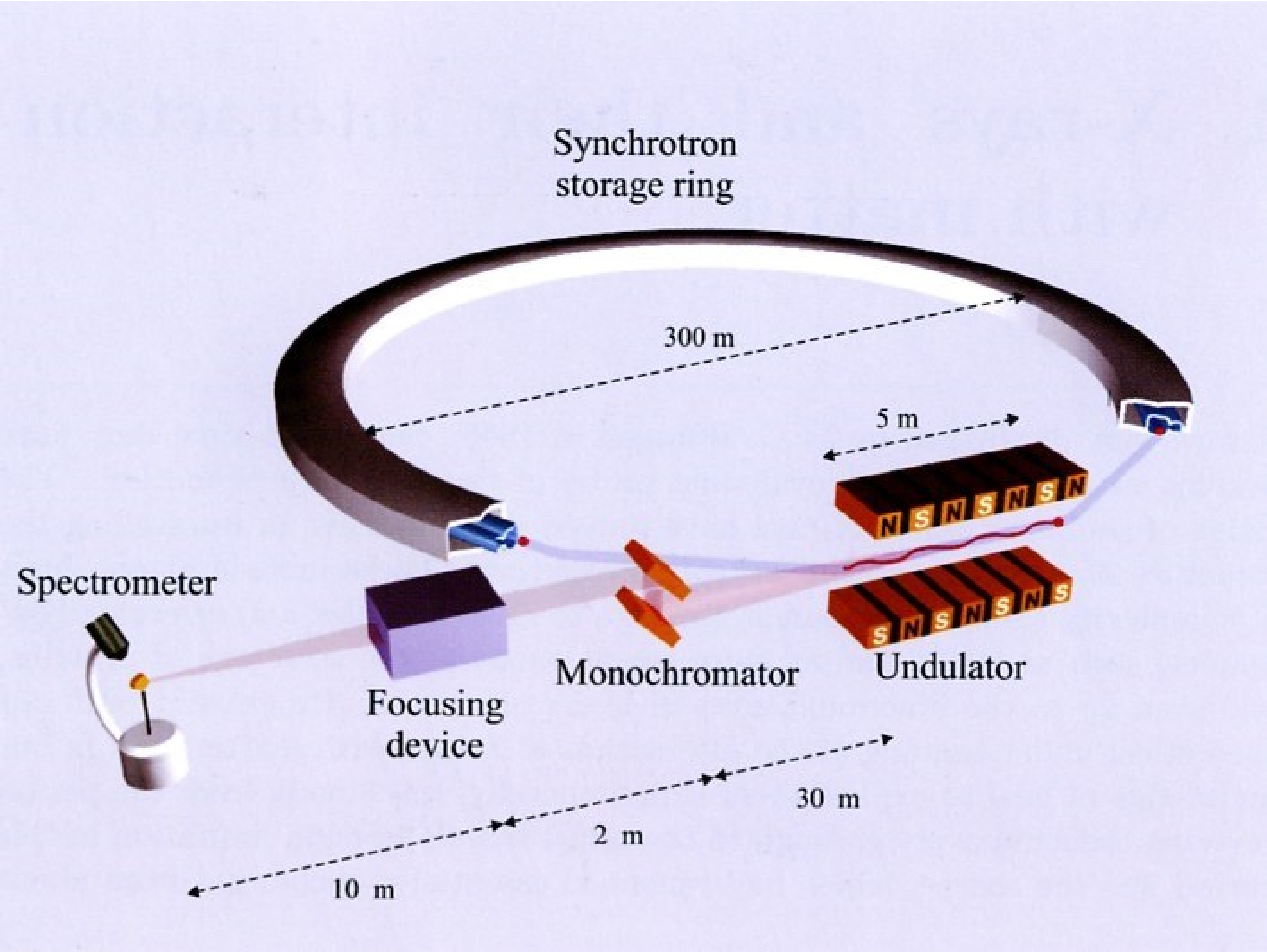
X-ray Scattering Research Today

Modern Protein Crystallography

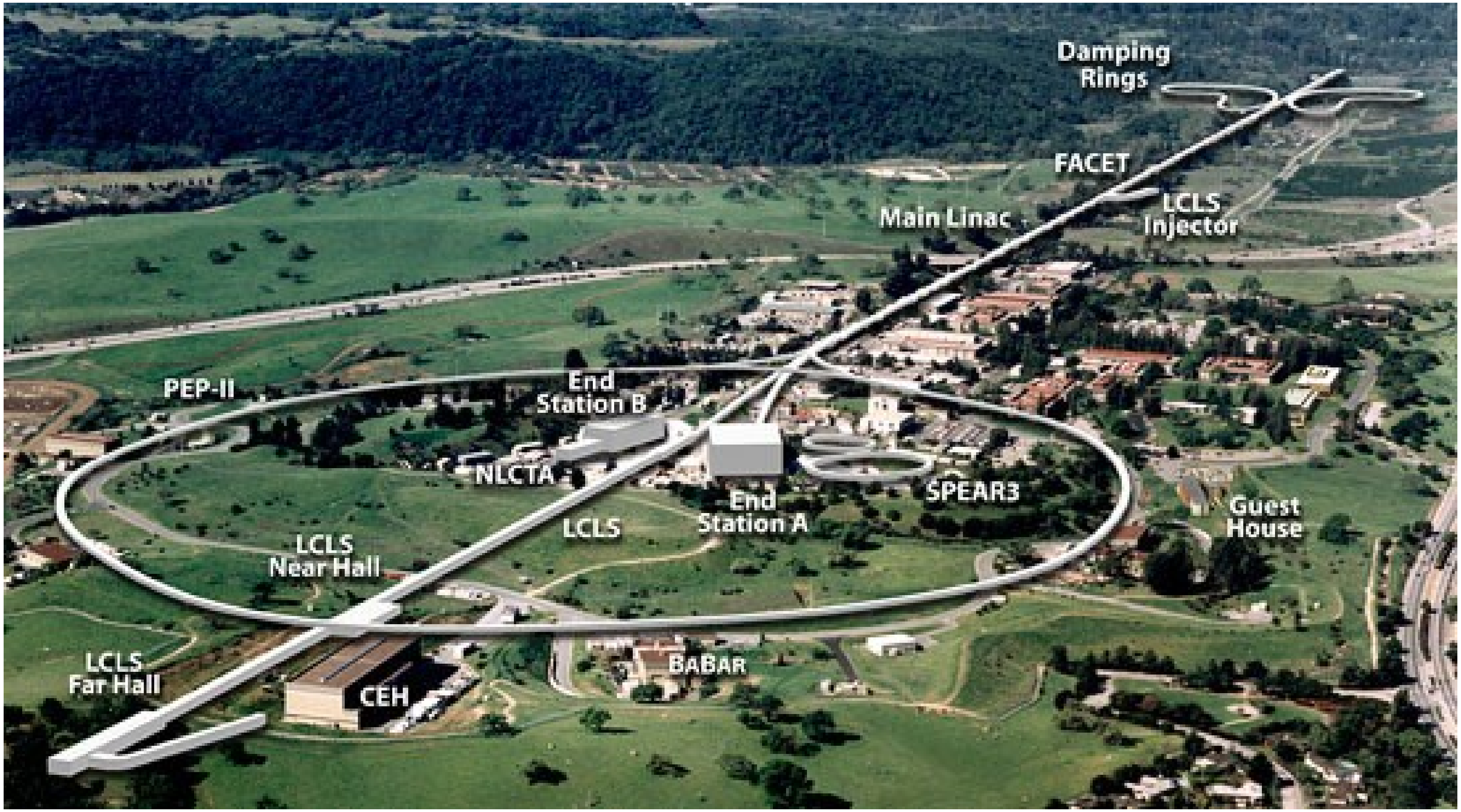


BioCARS 14-ID-B station of APS using an undulator with a gap of 25 mm from a crystal of the M37V mutant of CO-bound dimeric clam hemoglobin.

Made Possible by Storage-Rings



LCLS – Linac Coherent Light Source - SLAC



Serial Femtosceond Crystallography

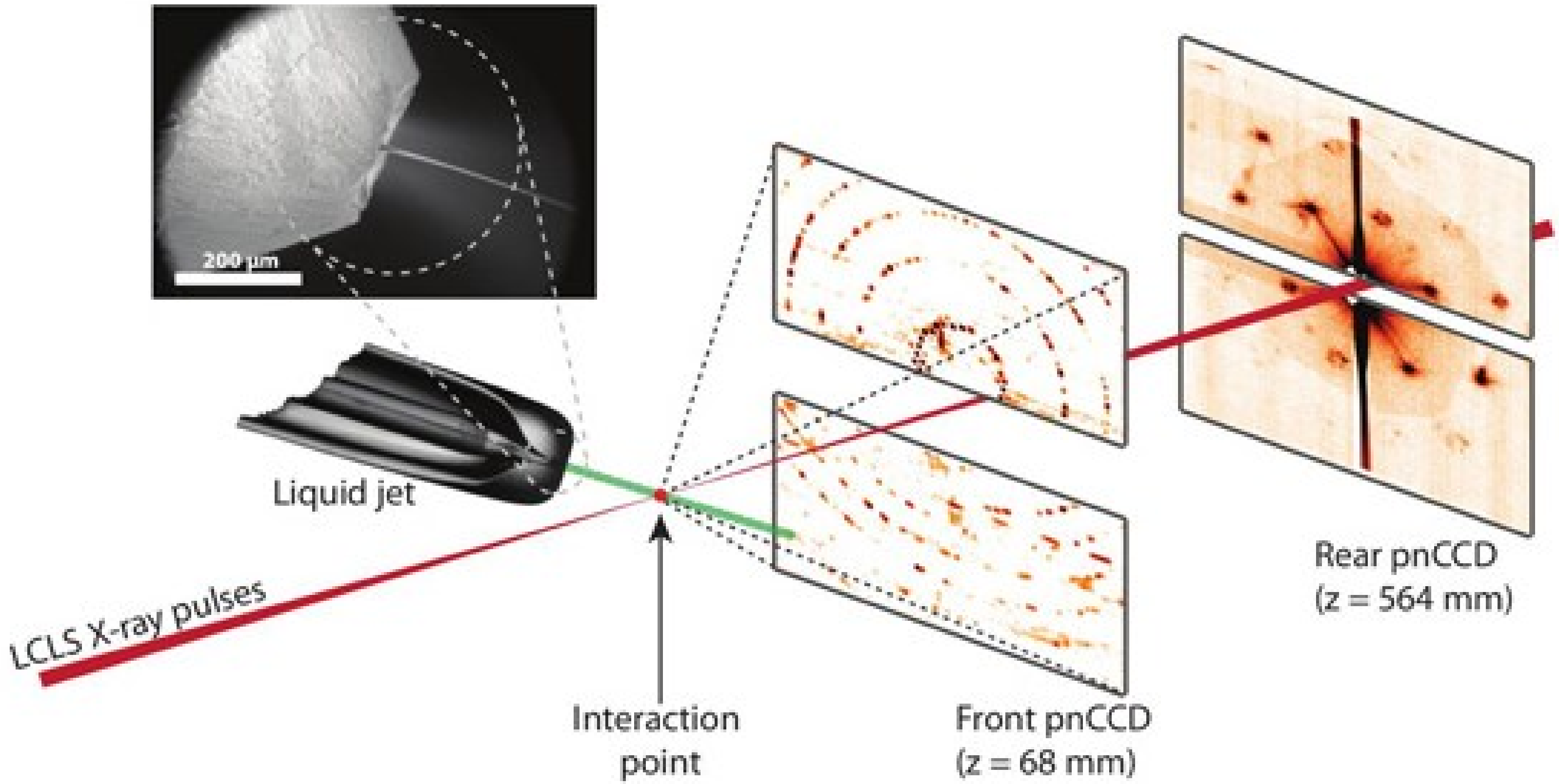


Figure 1
Experimental set-up for serial femtosecond crystallography. First published in Nature 470, 73 – 78 (2011).

Henry N. Chapman et al., NATURE 470, 73 (2011)

Serial Femtosceond Crystallography

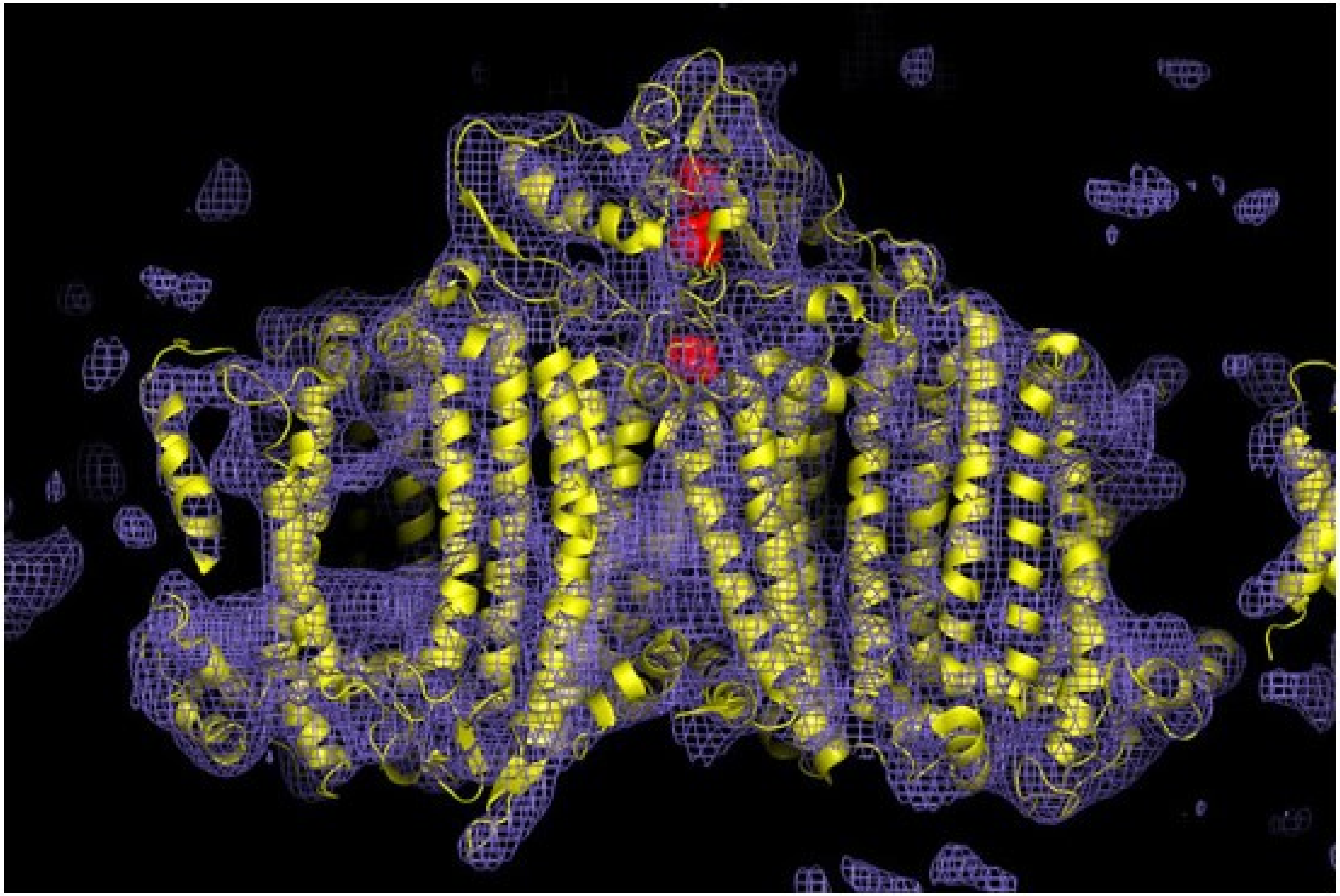


Figure 3

Electron density map of the photosystem I protein complex obtained from the LCLS diffraction data. First published in Nature 470, 73 – 78 (2011). Nanocrystals were grown by Petra Fromme of Arizona State University.

Extracted from 3 million diffraction patterns from photosystem I nanocrystals (200nm to 2 micron size) LCLS:30 Hz at 1.8 keV

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Kinematical Diffraction (II)
Diffraction from a Crystal, Reciprocal Lattice, Structure Factor, …

X-ray Scattering: A Primer

Scattering From a Single Electron

Scattering From a Single Atom

Scattering From a Crystal

Compton Scattering

Photoelectric Scattering

Photoelectric Absorption

Absorption and Reflection

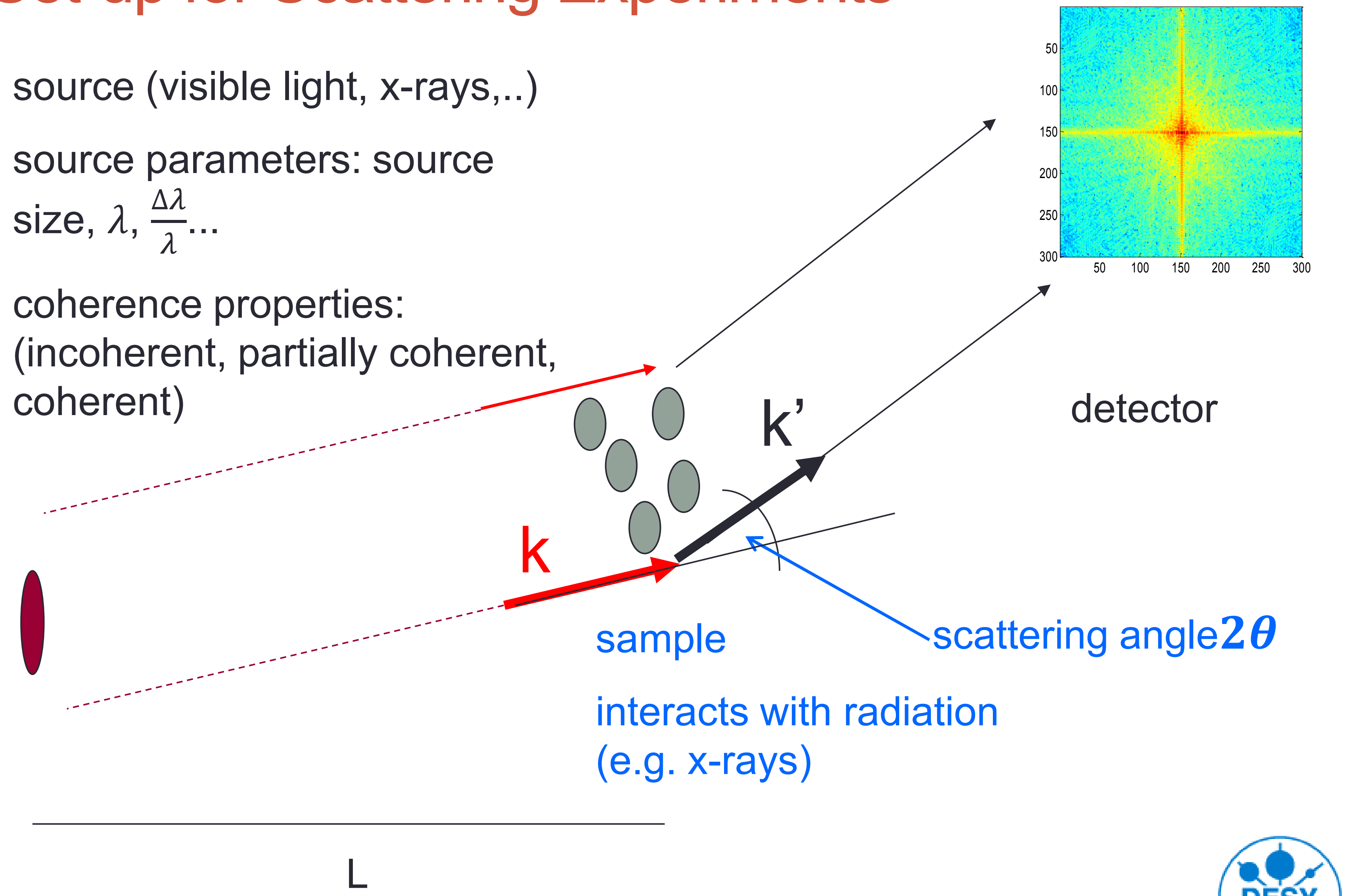
Coherence Properties

Set-up for Scattering Experiments

source (visible light, x-rays,..)

source parameters: source size, λ, $\frac{\Delta\lambda}{\lambda}$...

coherence properties: (incoherent, partially coherent, coherent)



k'

detector

k

sample

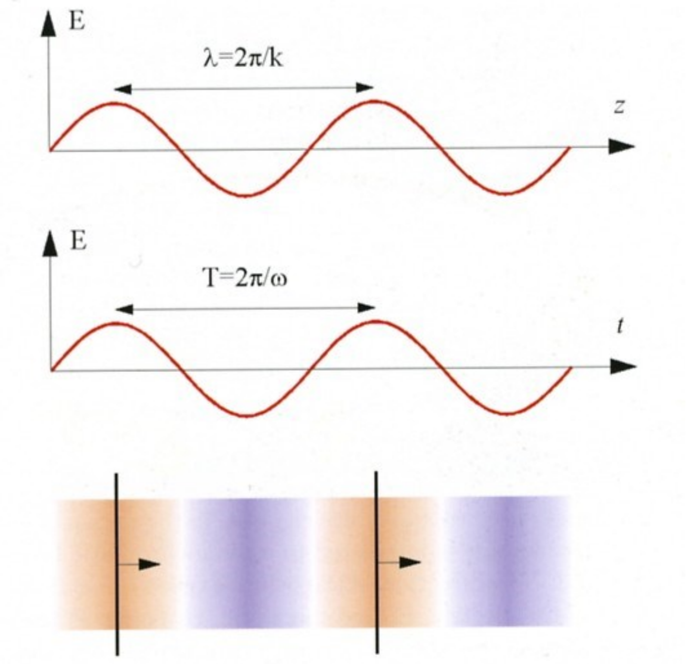
scattering angle 2θ

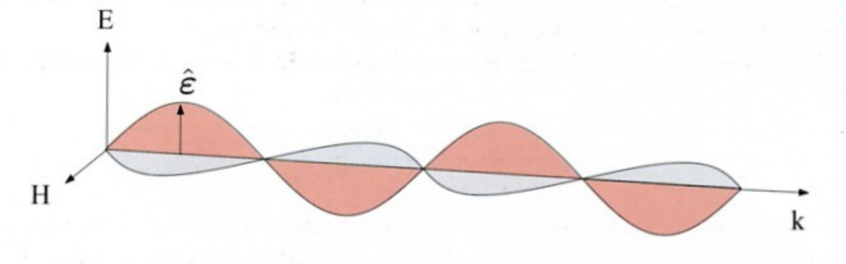
interacts with radiation (e.g. x-rays)

L

X-rays: Electromagnetic Waves and Photons

X-rays are electromagnetic waves with wavelengths in the region of Ångstroms (10^{-10} m). X-rays are transverse electromagnetic waves, where the electric and magnetic fields **E** and **H** are perpendicular to each other and to the propagation direction **k**.





with

ε: polarization vector

$$|\mathbf{k}| = \frac{2\pi}{\lambda}; \mathrm{E} = \mathrm{h}\nu = \hbar\omega = \frac{hc}{\lambda}$$

$$\lambda[\text{Å}] = \frac{\mathrm{hc}}{\mathrm{E}} = \frac{12.398}{\mathrm{E[keV]}}$$

Neglecting the H field one may write:

$$\mathbf{E}(\boldsymbol{r}, t) = \boldsymbol{\varepsilon}\, \mathrm{E}_0 \mathrm{e}^{i(\mathbf{kr}-\omega t)}$$

Scattering of X-rays

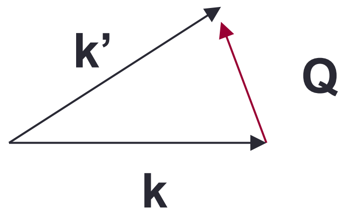
Consider a monochromatic plane (electromagnetic) wave with wave vector $\mathbf{k}$:

$$\mathbf{E}(\mathbf{r},t) = \boldsymbol{\varepsilon}\,\mathrm{E}_0\mathrm{e}^{i(\mathbf{kr}-\omega t)}$$

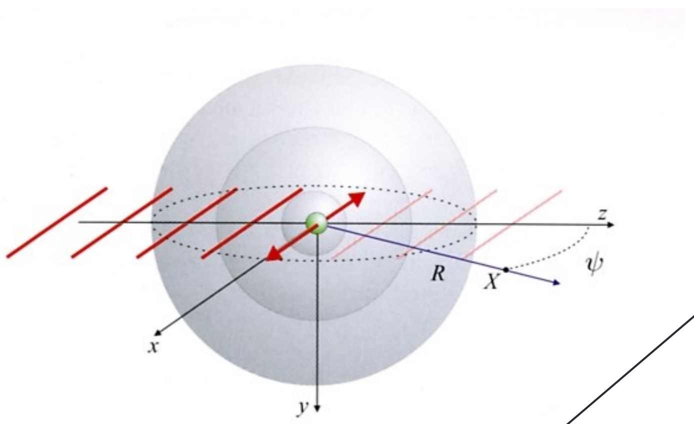
with $|\mathbf{k}| = \frac{2\pi}{\lambda}$

Elastic scattering:

$$\hbar\,\mathbf{k}' = \hbar\,\mathbf{k} + \hbar\mathbf{Q}$$



Scattering by a Single Electron:



$$\frac{\mathrm{E}_{\mathrm{rad}}(R,t)}{\mathrm{E}_{\mathrm{in}}} = -\frac{e^2}{4\pi\varepsilon_0 mc^2}\underbrace{\frac{\mathrm{e}^{ikR}}{R}}\cos\psi$$

spherical wave

Thomson scattering length r_0

$(= 2.82 \text{ x } 10^{-5}\text{Å})$

phase shift of π btw. incident and radiated field

Scattered intensity:

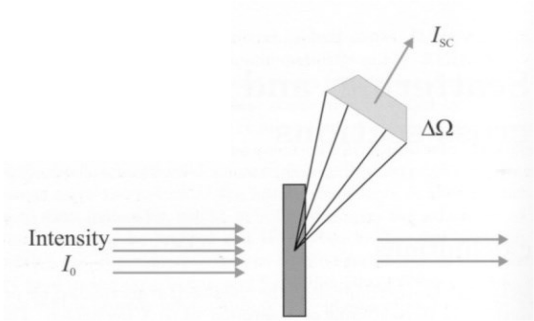
$$\frac{I_s}{I_0} = \frac{|E_{rad}|^2 R^2 \Delta\Omega}{|E_{in}|^2}$$

$$\frac{I_s}{I_0} = \left(\frac{d\sigma}{d\Omega}\right)\left(\frac{\Delta\Omega}{A_0}\right)$$

$\Delta\Omega$: solid angle seen by detector

$R^2\Delta\Omega$: cross sectional area scattered beam

A_0: incident beam size



with ($d\sigma$ / $d\Omega$) being the differential cross section (for Thomson scattering):
(# photons scattered/s into $\Delta\Omega$: $I_s/\Delta\Omega$ / incident flux: I_0/A_0)

$$\left(\frac{d\sigma}{d\Omega}\right) = r_0^2 P$$

$$P = \begin{cases} 1 & \text{vertical} \\ \cos^2\psi & \text{horizontal} \\ \frac{1}{2}(1+\cos^2\psi) & \text{unpolarized} \end{cases}$$

Note: $\boldsymbol{\sigma_{total} = \int \left(\frac{d\sigma}{d\Omega}\right) = \left(\frac{8\pi}{3}\right) r_0^2}$

Scattering by a Single Atom: scattering amplitude $A(Q) = -r_0 f(Q)$

phase factor

- **scattering amplitude by an ensemble of electrons** $-r_0 f^0(Q) = -r_0 \sum_{rj} e^{i\,Q\,r_j}$

(atomic) form factor

position of scatterers

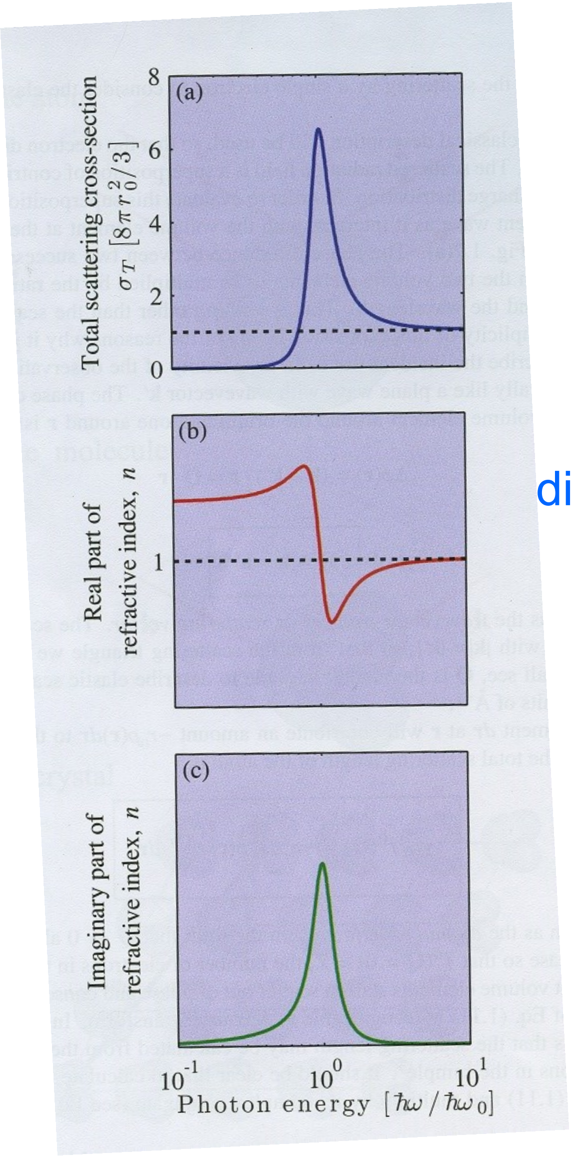
$$\{f^2(Q \rightarrow 0) = Z, \qquad f^2(Q \rightarrow \infty) = 0\}$$

form factor of an atom: $f(Q, \hbar\omega) = f^0(Q) + f'(\hbar\omega) + i\, f''(\hbar\omega)$

dispersion corrections: level structure absorption effects

scattering intensity: $I_s = A(Q)A(Q)^* = r_0^2 f(Q) f^*(Q) P$

Scattering by a Single Atom:

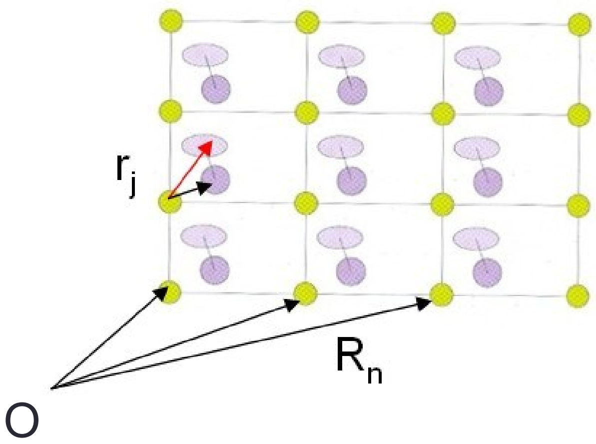


form factor of an atom:

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

dispersion corrections: level structure absorption effects

Scattering by a Crystal:



$$r_j' = R_n + r_j$$

lattice vector + atomic position in lattice

$$F^{\mathrm{crystal}}(Q) = \underbrace{\sum_{r_j} f_j(Q) e^{iQr_j}}_{\text{unit cell structure factor}} \underbrace{\sum_{R_n} e^{iQR_n}}_{\text{lattice sum}}$$

unit cell structure factor   lattice sum

$$I_s = r_0^2 F(Q) F^*(Q) P$$

lattice sum ≡ phase factor of order unity or N (number of unit cells) if:

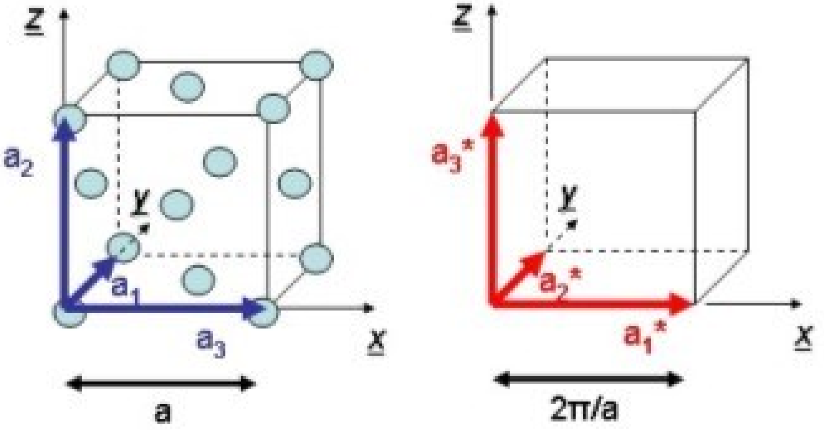
$$Q \bullet R_n = 2\pi \times \text{integer and } Q = G$$

Unit cell structure factor:

$$\sum_{r_j} f_j(Q)e^{iQr_j}$$

e.g. fcc lattice:

$$r_1 = 0$$
$$r_2 = \frac{1}{2}(a_1 + a_2)$$
$$r_3 = \frac{1}{2}(a_2 + a_3)$$
$$r_4 = \frac{1}{2}(a_3 + a_1)$$



$$a_1 = a\underline{x}\ ;\ a_2 = a\underline{y}\ ;\ a_3 = a\underline{z}\ ;\ v_c = a^3\ ;\ a_1^* = \left(\frac{2\pi}{a}\right)\underline{x}\ ; a_2^* = \left(\frac{2\pi}{a}\right)\underline{y}\ ; a_3^* = \left(\frac{2\pi}{a}\right)\underline{z}$$

$$F_{hkl}^{fcc} = f(Q)\sum e^{iQr_j}$$

with $Q = G = h\,a_1^* + k\,a_2^* + I\,a_3^*$

$$= f(Q)\{1 + e^{i\pi(h+k)} + e^{i\pi(k+l)} + e^{i\pi(l+h)}\} \qquad (£)$$

$$= f(Q)\ \times \begin{cases} 4 & \text{if } h, k, l \text{ are all even or odd} \\ 0 & \text{otherwise} \end{cases}$$

Compton Scattering

Consider photon with momentum initially at rest

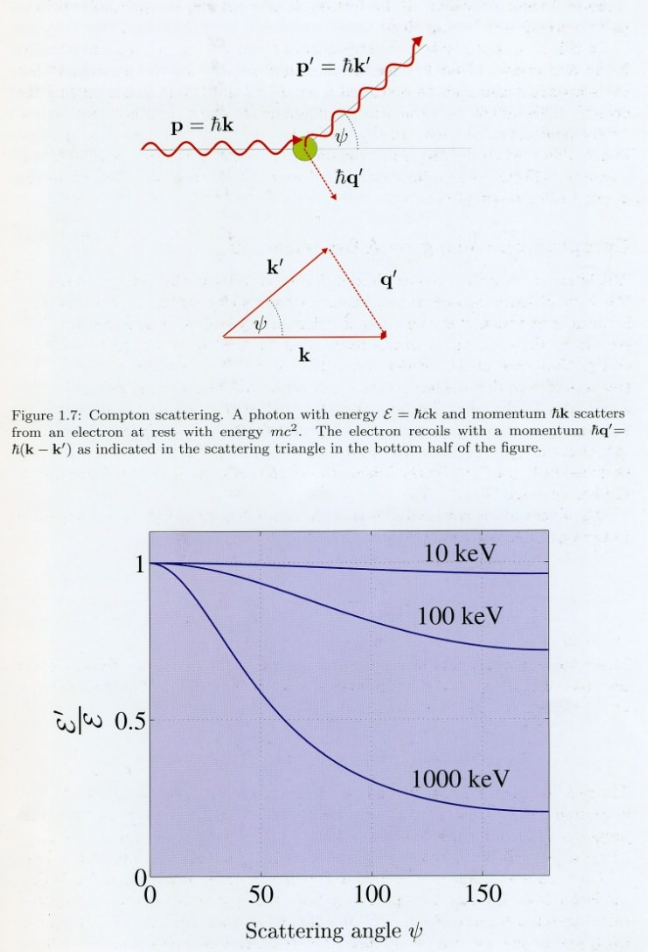


Figure 1.7: Compton scattering. A photon with energy $\mathcal{E} = \hbar ck$ and momentum $\hbar\mathbf{k}$ scatters from an electron at rest with energy mc^2. The electron recoils with a momentum $\hbar\mathbf{q}' = \hbar(\mathbf{k} - \mathbf{k}')$ as indicated in the scattering triangle in the bottom half of the figure.

$p = \hbar\, \mathbf{k}$ scattered by a electron,

Energy conservation:

$$m_0c^2 + \hbar ck = \sqrt{\{(m_0c^2)^2 + (\hbar cq')^2\}} + \hbar ck'$$

with $\lambda_c = \frac{\hbar c}{m_0c^2}$: Compton wavelength

$$q'^2 = (k - k')^2 + 2\frac{(k - k')}{\lambda_c} \qquad (1)$$

Momentum conservation: $p' = k - k'$

$$q' \bullet q' = q'^2 = (k - k')\bullet(k - k') = k^2 + k'^2 - 2kk' \cos\psi \qquad (2)$$

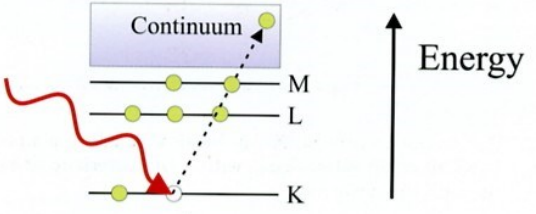
$(1) = (2)$

$$\frac{\mathbf{k}}{\mathbf{k}'} = \mathbf{1} + \boldsymbol{\lambda}_c\mathbf{k}(\mathbf{1} - \mathbf{cos}\,\boldsymbol{\psi}) = \frac{\boldsymbol{\varepsilon}}{\boldsymbol{\varepsilon}'} = \frac{\boldsymbol{\lambda}'}{\boldsymbol{\lambda}}$$

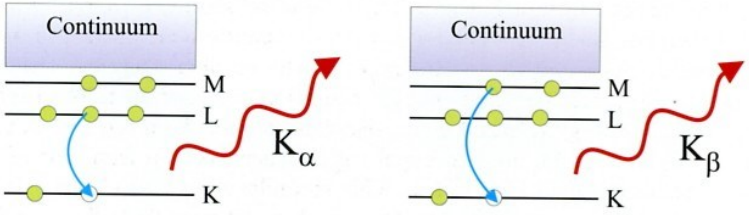
- ➔ origin of background
- ➔ determine electronic momentum distribution of materials

Photoelectric Absorption

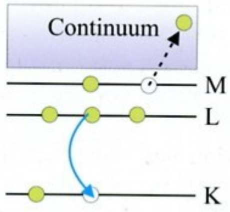
(a) Photoelectric absorption



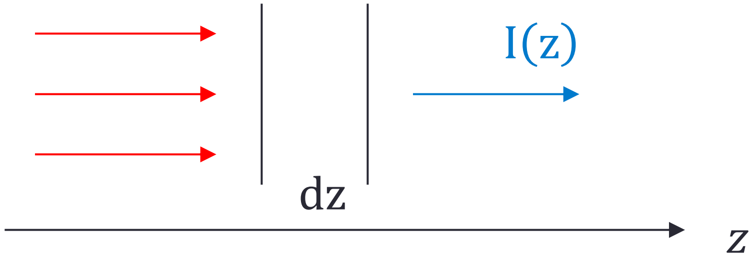
(b) Fluorescent X-ray emission



(c) Auger electron emission



$$-dI = I(z)\mu\, dz$$



$$I(z) = I_0 e^{-\mu z}$$

$$\mu = p_a \sigma_a = \left(\frac{p_m N_A}{A}\right) \sigma_a$$

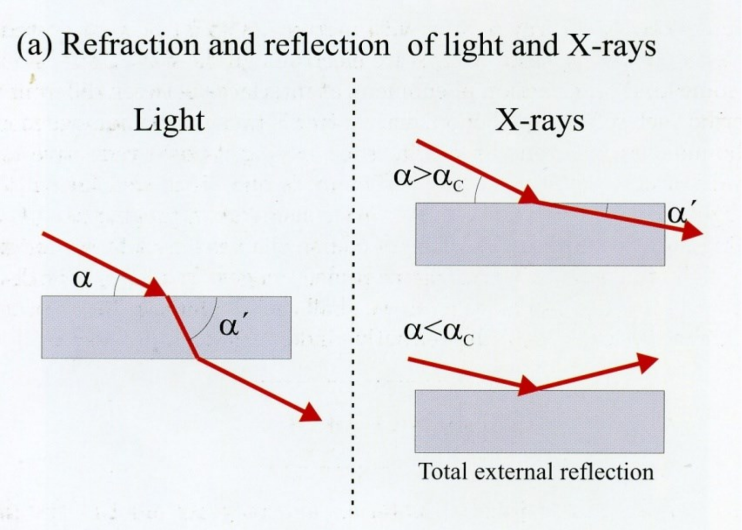
p_a	atomic number density
$\sigma_a = \sigma_a(E)$	absorption cross section
p_m	mass density
N_A	Avogadro’s number
A	atomic mass number

Refraction

$$\mathbf{n = 1 - \delta + i\beta} \qquad \mathbf{< 1}$$

$\uparrow$ $\uparrow$

10^{-5} absorption (<< δ)



Snell's law:

$$\cos\alpha = \mathrm{n}\cos\alpha'$$

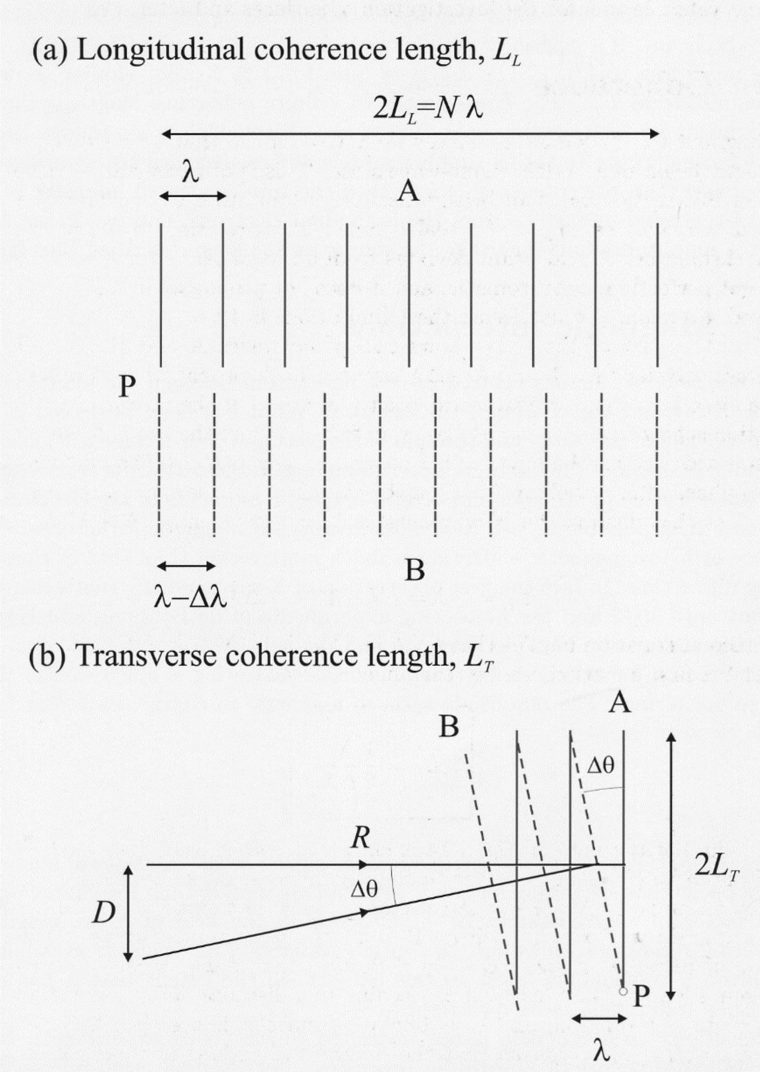
Note: total external reflection for x-rays $(\alpha' = 0)$

n < 1

$\alpha_c = \sqrt{2\delta}$

Note: cos z = $1 - z^2/2! + z^4/4! - z^6/6! \ldots$

Coherence



Longitudinal coherence:

Two waves are in phase at point P. How far can one proceed until the two waves have a phase difference of π:

$$\xi_l = \frac{1}{2}\frac{\lambda^2}{\Delta\lambda}$$

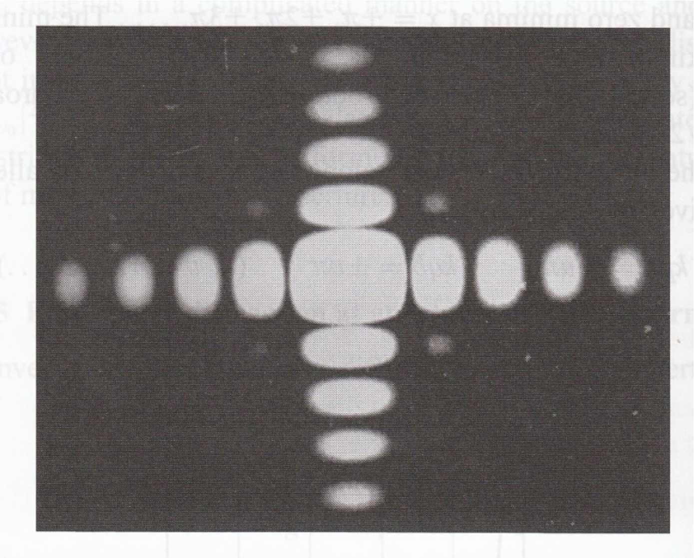
Transverse coherence:

Two waves are in phase at P. How far does one have to proceed along A to produce a phase difference of π :

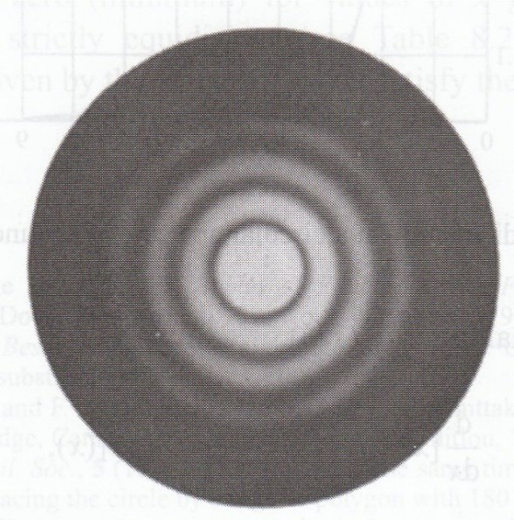
$$2\xi_t \, \Delta\theta = \lambda$$

$$\xi_t = \frac{\lambda}{2}\left(\frac{R}{D}\right)$$

Fraunhofer Diffraction

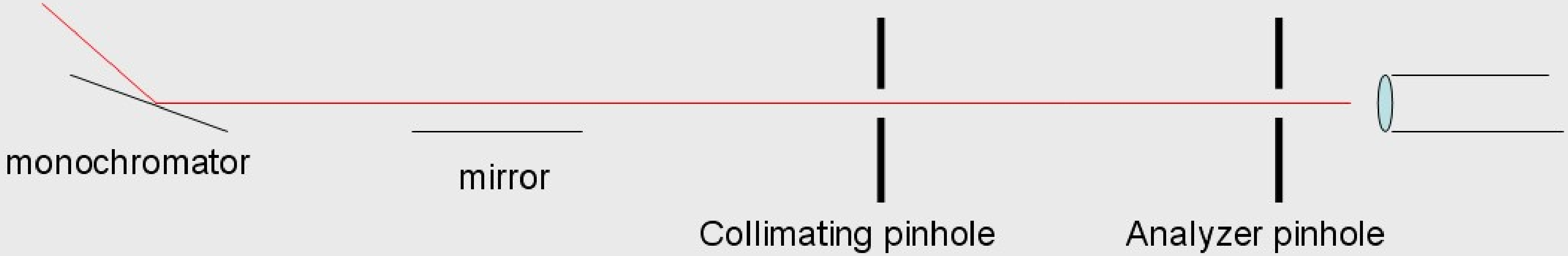


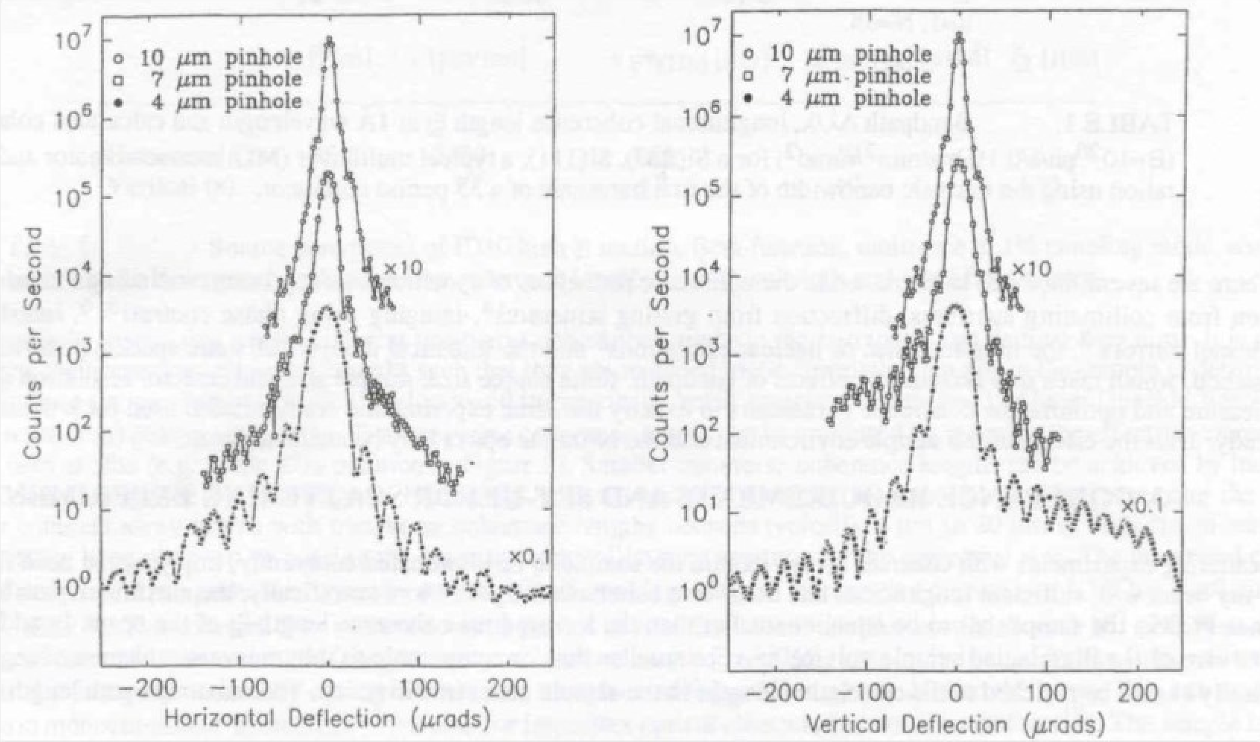
Fraunhofer diffraction of a rectangular aperture 8 x 7 mm^2, taken with mercury light λ=579nm
(from Born&Wolf, chap. 8)



Fraunhofer diffraction of a circular aperture, taken with mercury light λ=579nm
(from Born&Wolf, chap. 8)

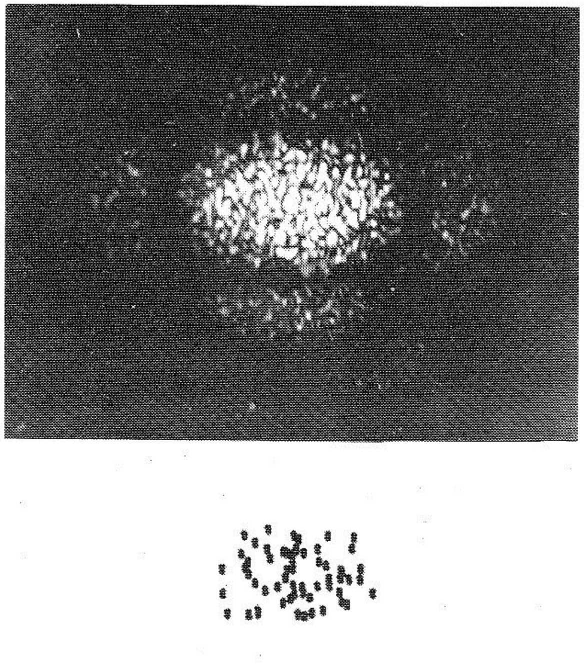
Fraunhofer Diffraction $(\lambda = 0.1nm)$



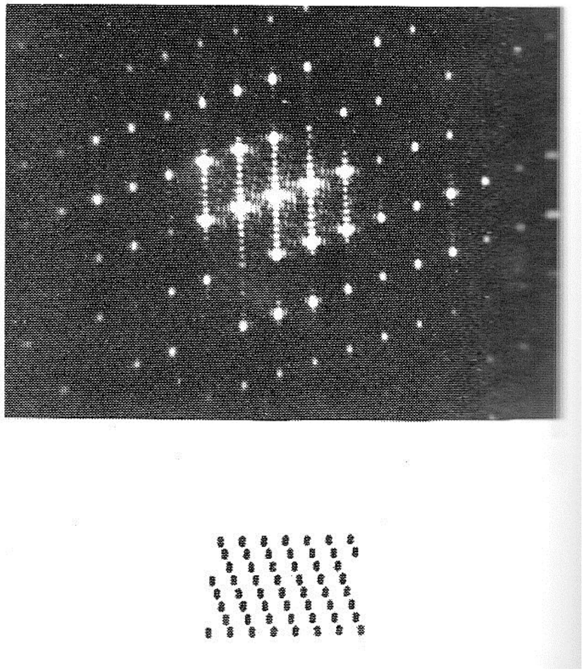


ID10 (ESRF)
E=11.8 keV;
λ=1.05Å

Speckle Pattern



random arrangement of apertures: speckle



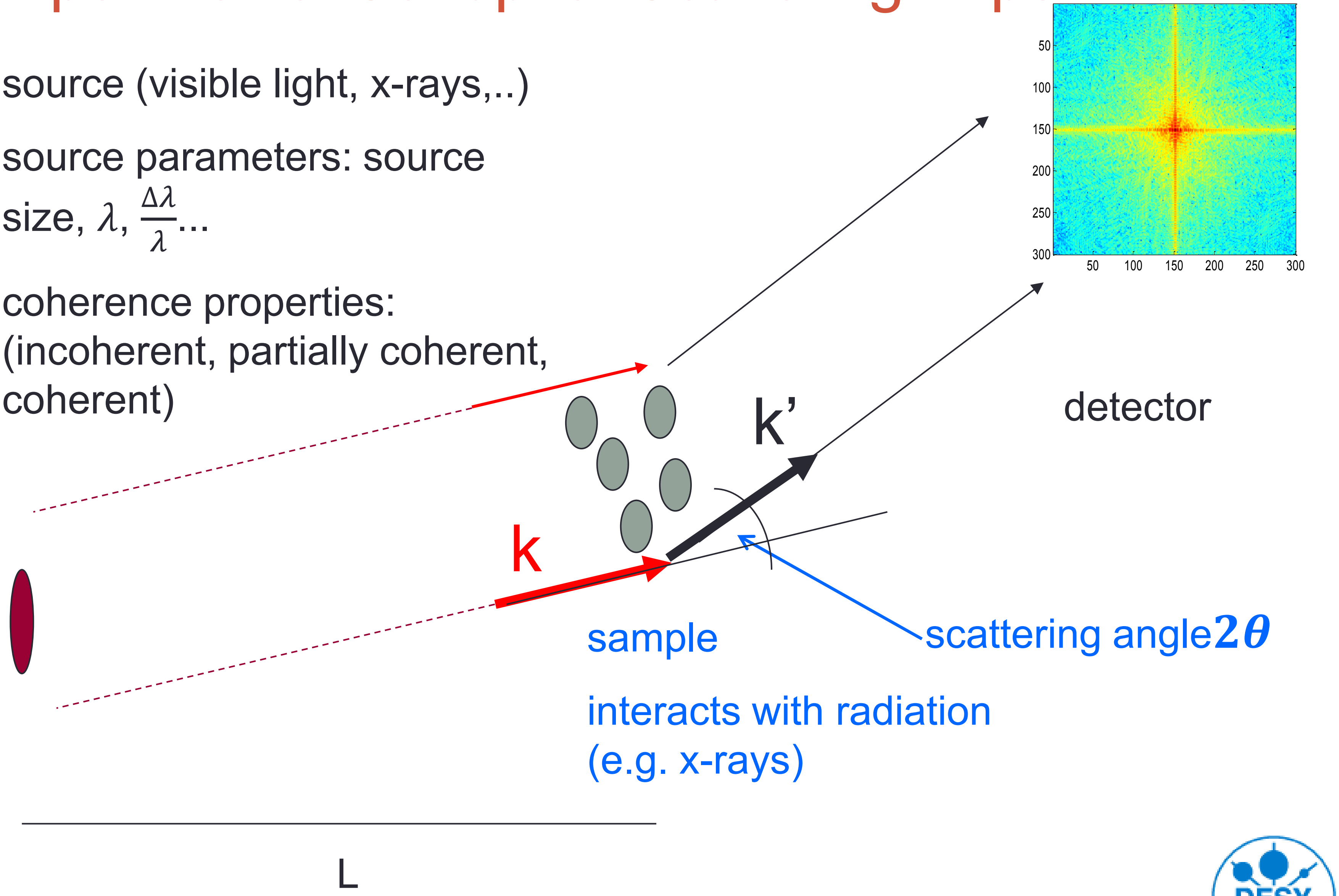
regular arrangement of apertures

Experimental Set-up for Scattering Experiments

source (visible light, x-rays,..)

source parameters: source size, λ, $\frac{\Delta\lambda}{\lambda}$...

coherence properties: (incoherent, partially coherent, coherent)



detector

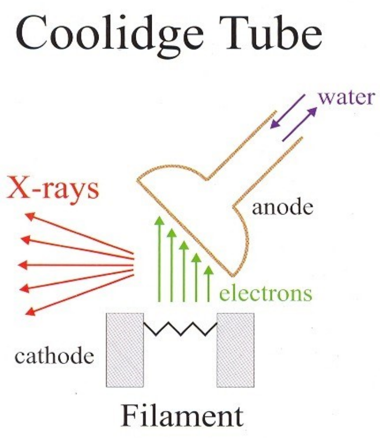
sample

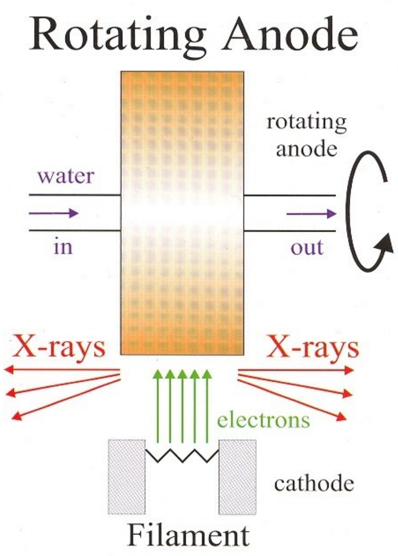
scattering angle $2\boldsymbol{\theta}$

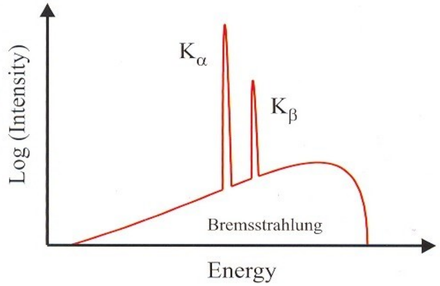
interacts with radiation (e.g. x-rays)

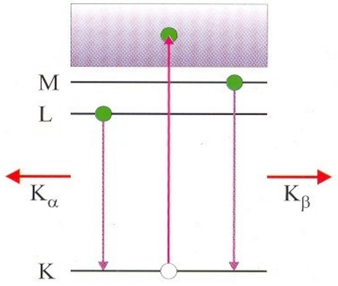
Source of X-Rays

1895	Discovered by W.C. Röntgen
1912	First diffraction experiment (v. Laue)
1912	Coolidge tube (W.D. Coolidge, GE)
1946	Radiation from electrons in a synchrotron, GE, Physical Review, 71,829 (1947)

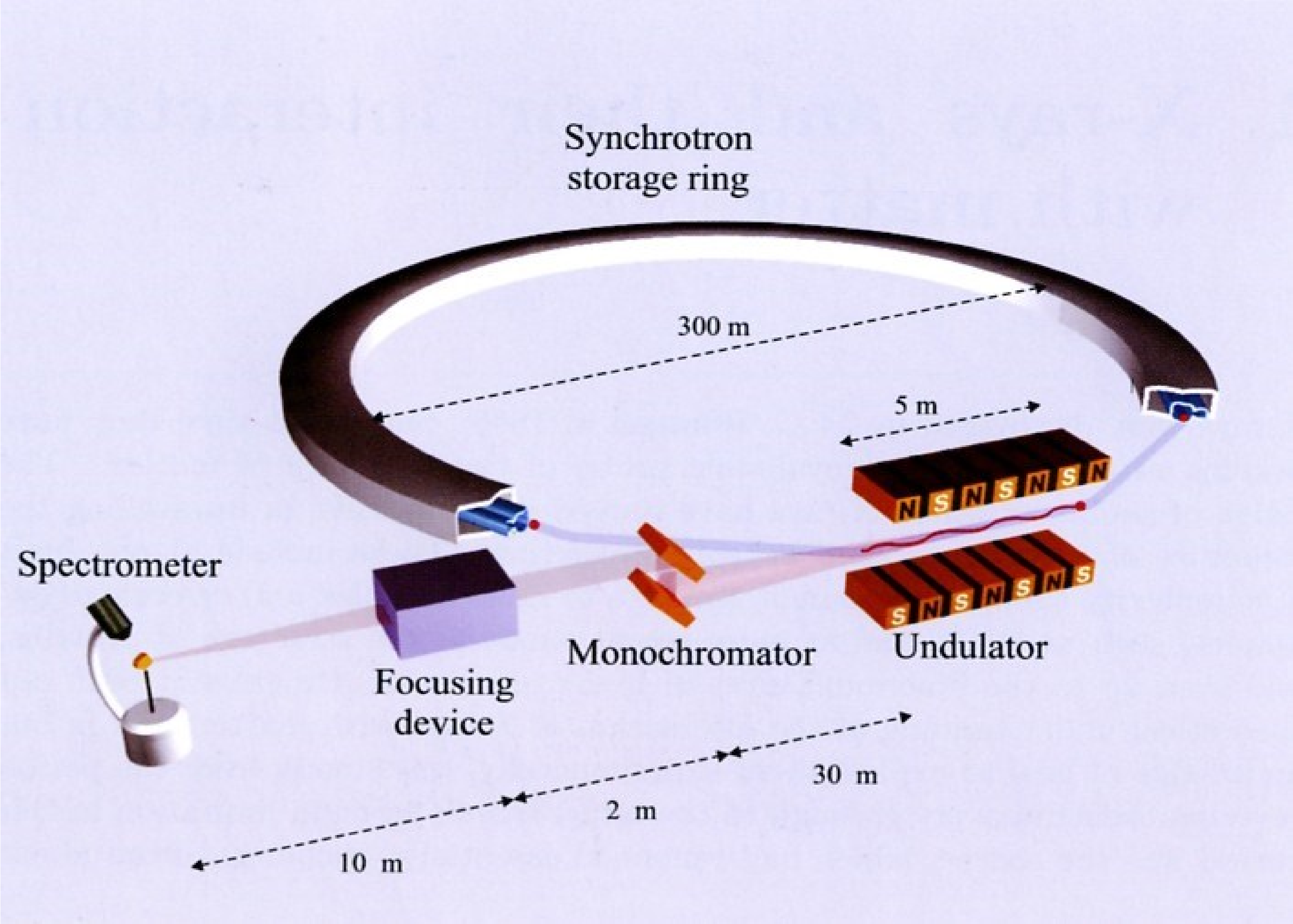








Synchrotron Radiation Storage Ring



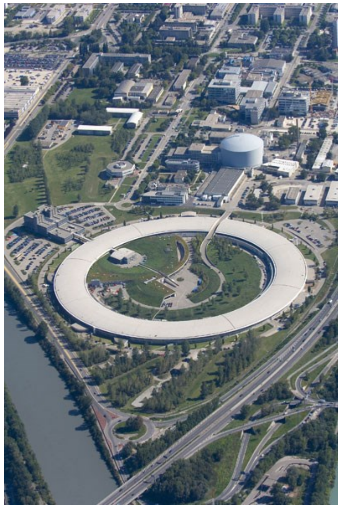
Photon Machines

The three largest and most powerful synchrotrons in the world

APS, USA

ESRF, Europe-France

Spring-8, Japan



The most recent third generation machine:

Petra III at DESY/Hamburg