"X-rays and their interaction with matter"

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OUTLINE

"X-rays and their interaction with matter"

1 – Synchrotron radiation properties
2 – Waves and photons
3 – X-ray Scattering
4 – Photon Absorption
5 – Optical properties

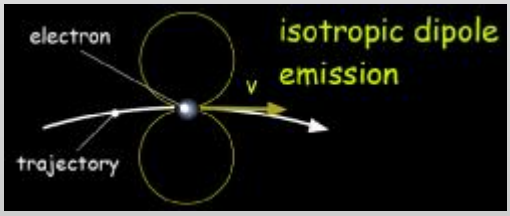
WHAT THE SYNCHROTRON RADIATION?

Electromagnetic radiation emitted when charge particles moving at ultra-relativistic energies are forced to change direction under the action of a magnetic field.

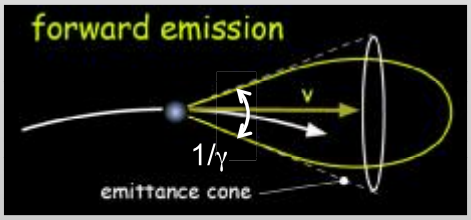
$E>m_0c^2\sim0.511$ MeV $\gamma= E/m_0c^2$ $\gamma\sim1957\ E[GeV]$

Classical emission $v_e<< c$
Lorenz Force $F=q\ \mathbf{v}\times\mathbf{B}$

Relativistic emission when $v_e\sim c$
Forward direction emittance cone $1/\gamma\sim1$mrad



Ex: Radio waves



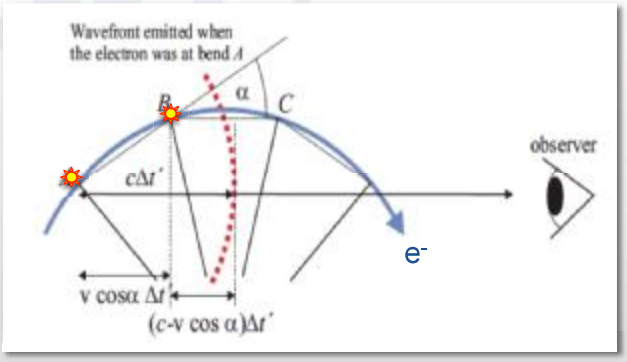
Synchrotron emission

The radiation emitted by an accelerated charge particle appear shifted in frequency from an observer sit in the tangent of trajectory.

Time interval Δt between the two flash emitted in A and B at the observer position:

$$\Delta t = (1-\beta_e\cos\alpha)\Delta t'$$

$\Delta t'$= electron time interval between two flashes in A and B
v = electron velocity
c = light speed
β_e=v/c



… and because α is small:

$$\Delta t \cong \left[\frac{1+(\alpha\gamma)^2}{2\gamma^2}\right]\Delta t'$$

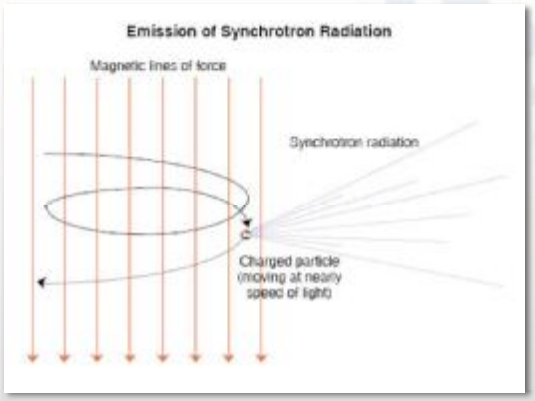
Maximum Doppler effect when $\alpha=0$
$\Delta t \sim 1/(2\gamma^2)\ \Delta t' \sim 10^{-8}\ \Delta t'$

When $\alpha=1/\gamma$ Doppler effect decrease of ½

Most of the radiation is then contained in a solid angle $1/\gamma$

SYNCHROTRON RADIATION IN ASTRONOMY

Most of the radiation of our galaxy is composed by synchrotron radiation.

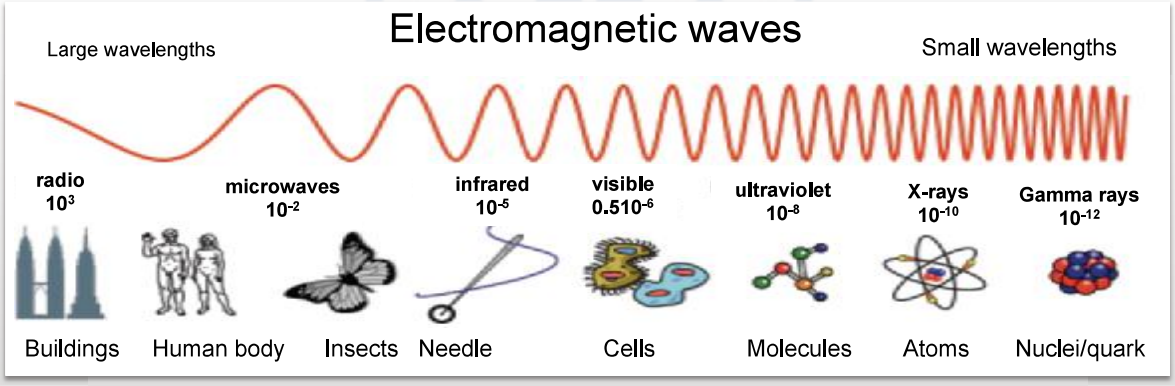


Synchrotron light was first detected in a jet emitted by M87 in 1956 by Geoffrey R. Burbidge.

Gas emission (red) and synchrotron light (blu) produced by high energy electrons in the magnetic field of a neutron star (Crab nebula)

ELECTROMAGNETIC SPECTRUM

Small wavelength probe small objects



Radio waves

Radar

Microscopes

Synchrotrons

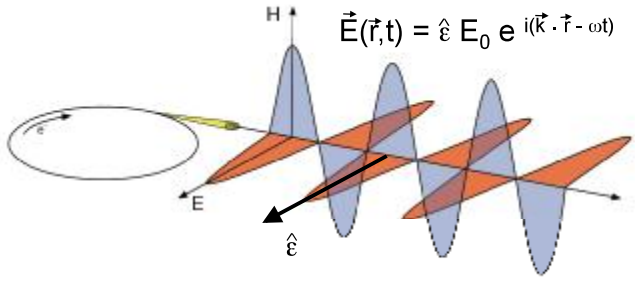
Large accellerators

ELECTROMAGNETIC WAVES

A Light for Science

The synchrotron radiation delivered by insertion devices is a polarized electromagnetic wave with polarization vector ε parallel to the electric field E and lying in the synchrotron orbit plane.

Transverse EM waves



$\vec{E}(\vec{r},t) = \hat{\varepsilon}\, E_0\, e^{i(\vec{k}\cdot\vec{r} - \omega t)}$

λ wavelength

$k=2\pi/\lambda$ wavenumber

$\nu=\omega/2\pi$ frequency

$\hat{\varepsilon}$ polarization

Energy

$\mathcal{E}$ [keV] $= \hbar\omega = hc/\lambda = 12.398 / \lambda$ [Å]

λ [Å] $= hc/\mathcal{E} = 12.398/E$[keV]

ex. 1 Å = 12.398 keV

Spectral intensity

$I_0(\omega) = \langle E_0^2 \rangle = N(\omega)\, \hbar\omega$

SYNCHROTRON RADIATION PROPERTIES

A Light for Science

High brillance
- Easy focusing, fast detection

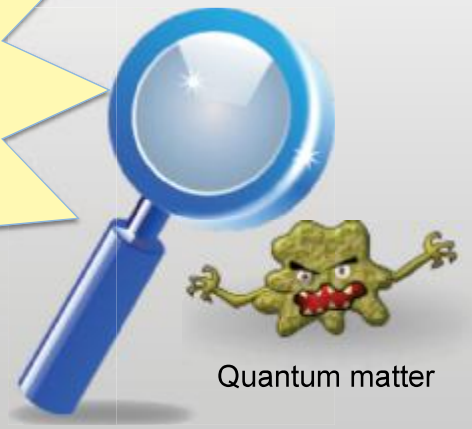
Short Wavelengths
- Comparable with atomic distances

Energy resolution and tuning
- Core-hole electron transitions energies

Polarization
- Magnetic and electronic symmetries

Time structure
- Pump probe experiments

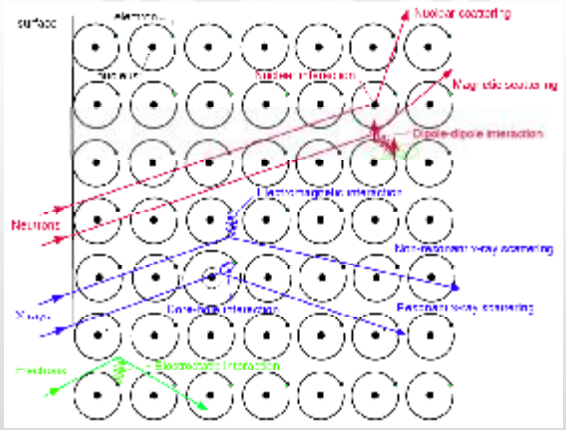
Coherence
- Photon correlation spectroscopies



Quantum matter

QUANTUM PROBES

Neutron and x-ray magnetic scattering are a powerfull probe to study the electronic interactions at atomic scale in bulk and nanostructured materials



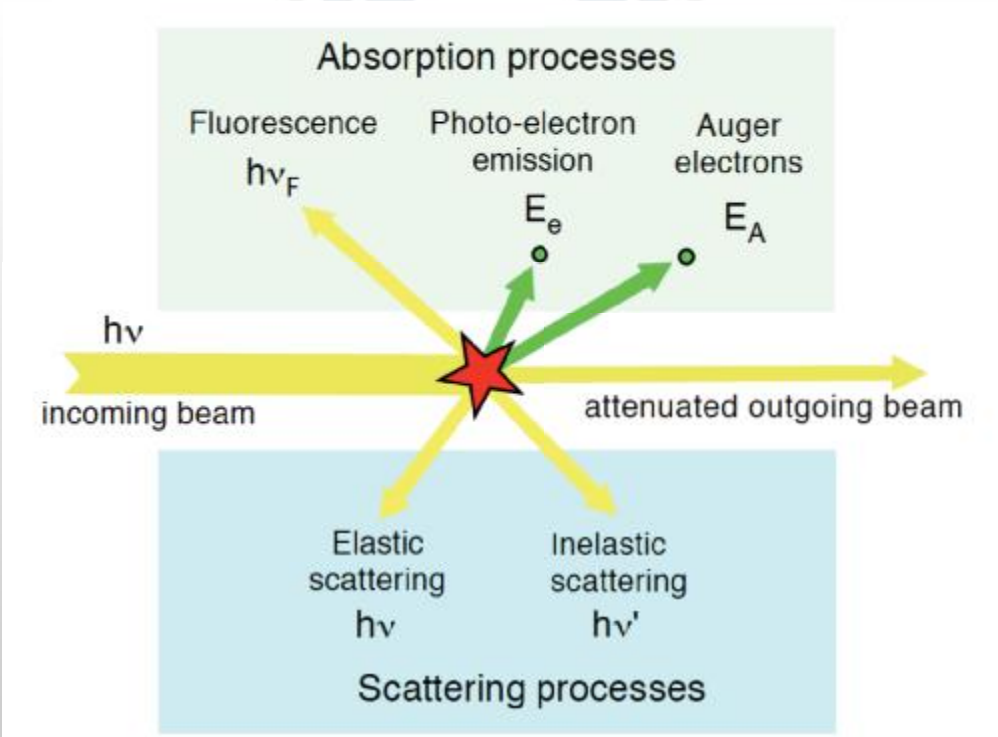
Neutron scattering (Cold-Thermal)

- Bulk sensitivity (low absorp., ~10 cm)
- Amplitudes: Nuclear/Magnetic ~ 1
- High E-resolution
- **Unpolarized source**
- Soft interaction neutron-sample
- Well established sample environment

X-ray scattering (3-30 keV)

- Surface sensitivity (high absorp., ~10 μm)
- Amplitudes: Charge/Magnetic ~ 10^5
- High Q-resolution
- **Polarized source**
- **Easy Focusing**
- Hard probe (T-heating, sample damage)…

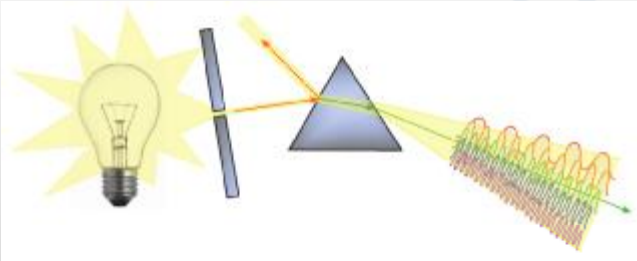
X-RAYS / MATTER INTERACTIONS



CLASSICAL AND QUANTUM ASPECTS

Electromagnetic waves

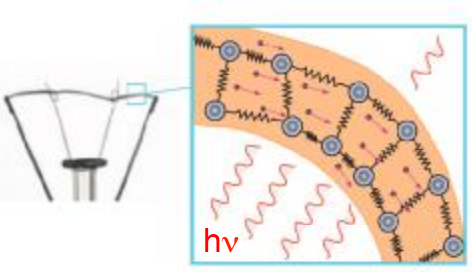
Far from the emission sources
Large length scales and forces strengths
Classical electromagnetism



- Maxwell equations
- Optics laws:
  - Reflection
  - Refraction
  - Diffraction

Photons

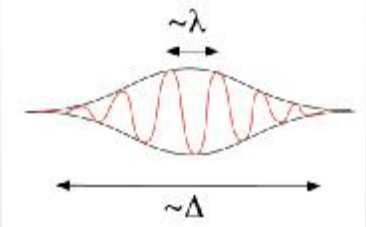
Near the emission sources
Atomic length scales
Quantum mechanics



- Plank laws
- Quantum electrodynamics
- Absorption/emission processes
- Elastic and inelastic scattering

THE DUALISM: PARTICLES OR WAVES?

Matter wave



Heisenberg uncertainly principle:
Cannot define both Δ and λ to an arbitrary accuracy

Oscillations ➔ wave
Envelope ➔ particle

Classical: Δ>>λ

Quantum: Δ<<λ

Decreasing Δ to define better the position, but we lose information on λ

Kinematics

Energy: $E = h\nu = \hbar\omega$
Momentum: $p = h/\lambda = \hbar k$

where

h= Planck's constant
v=frequency
k=wavevector

Assumption of quantum mechanics:

(i) Particle is represented mathematically by a wavefunction, $\psi(\underline{r})$

(ii) $|\psi(\underline{r})|^2 dV$= Probability of finding a particle in a (infinitesimal) volume dV

Flux of particles

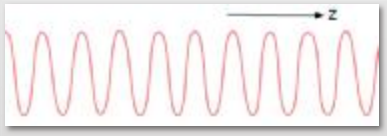
I = No. of particles incident normally on unit area per second =
= particle density x velocity
= $|\psi|^2$ x v = $|\psi|^2$ $\hbar k/m$ $(m^{-2}\ s^{-1})$

Infinite plane wave:

$\psi(\underline{z}) = e^{ikz} = \cos(kz) + i \sin(kz)$

$|\psi|^2 = \psi\psi^* = e^{ikz}e^{-ikz} = 1$

1 particle per unit volume everywhere!

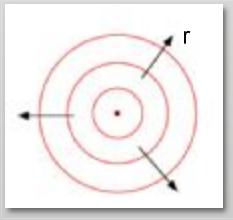


Spherical wave:

$\psi(\underline{r}) = b/r\ e^{ikr}$

$|\psi(\underline{r})|^2 = b^2/r^2$

Density of particles falls as $1/r^2$



Definition: Total cross section

$$\sigma = \frac{\text{Total no. particles scattered in all the directions per second}}{\text{Incident flux } (I_0)}$$

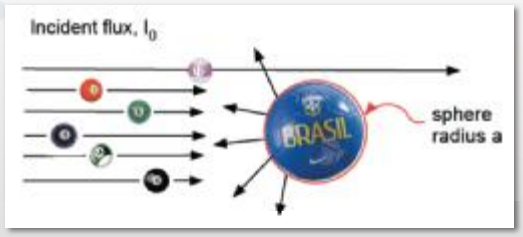
Classical case:

no. particles scattered per second = $I_0 \times \pi a^2$

$\sigma = \pi a^2$

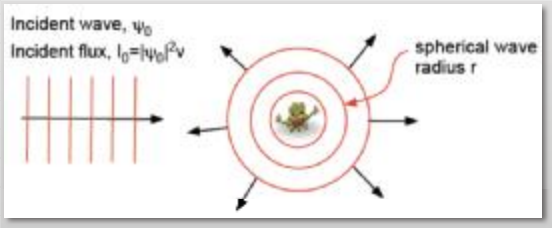
Particles per unit area per sec.

Cross-sectional area of sphere



Quantum case:

Incident wave	$\psi_0 = e^{ikz}$
Incident flux	$I_0=\|\psi_0\|^2 v = v$
Scattered wave	$\psi_{sc} = b/r\ e^{ikr}$
Scattered flux	$I_{sc}=\|\psi_{sc}\|^2 v = b^2v/r^2$



no. particles scattered per second = $I_{sc} \times 4\pi r^2$

$\sigma = 4\pi b^2$

Effective area viewed by scattering particles!
b is the scattering amplitude

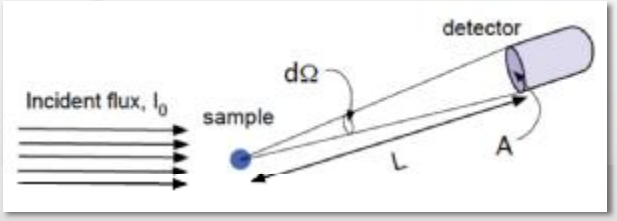
Definition: differential cross section

$$\frac{d\sigma}{d\Omega} = \frac{\text{no. particles scattered into solid angle } d\Omega \text{ per sec.}}{I_0 \times d\Omega}$$

Solid angle subtended by the detector:
$\Delta\Omega = A/L^2$

No. particles detected per sec.:
$I_0 \, \Delta\Omega \, d\sigma/d\Omega = |\psi_{sc}|^2 \, v \times A$

Flux ↙ ↘ Detector area



$$\frac{d\sigma}{d\Omega} = \frac{|\psi_{sc}|^2}{|\psi_0|^2} L^2 = b^2 = \frac{d\sigma}{4\pi}$$

(barns/steradian) 1 barn=10^{-28} m^2

Notice that ψ_{sc} is a spherical wave $|\psi_{sc}|^2 = b^2/L^2$

X-RAYS SCATTERING

X-RAYS SCATTERING BY A FREE ELECTRON

- The electric field E_{in} of the incident x-rays forces the motion of the electron which radiates a spherical wave E_{rad}.
- Electron at rest: Electric Force **F=Eq**

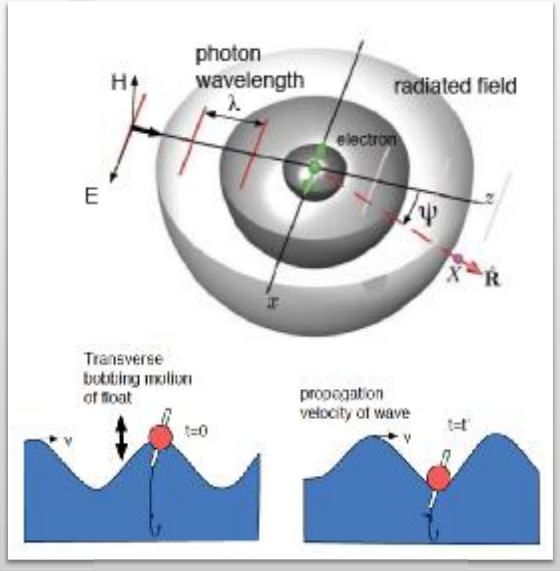
Radiated spherical field E_{rad} :
- proportional to the electron acceleration
- anti-phase with respect E_{in}
- decreases with cos(ψ)

$$\frac{E_{rad}(R,t)}{E_{in}} = -\left(\frac{e^2}{4\pi\epsilon_0 mc^2}\right)\frac{e^{ikR}}{R}\cos\psi$$

Thomson scattering length:

$$r_0 = \left(\frac{e^2}{4\pi\epsilon_0 mc^2}\right) = 2.82\times10^{-5}\ \text{Å}$$

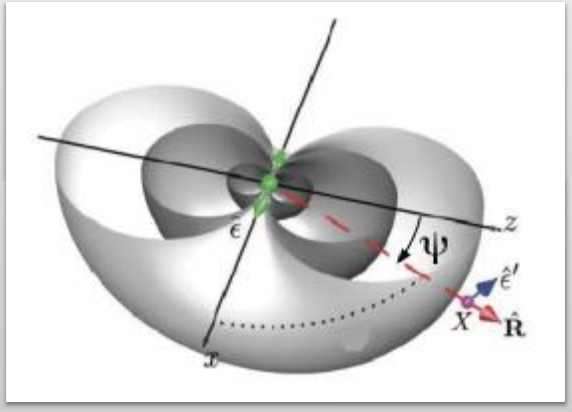
Notice that $b=r_0$

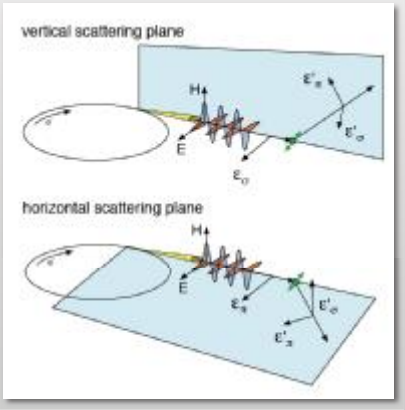


POLARIZATION DEPENDENCE OF THOMSON SCATTERING

The differential cross section for the Thomson scattering depends from the incident and scattered photon polarizations

$$\left(\frac{d\sigma}{d\Omega}\right) = r_0^2\left|\hat{\varepsilon}\cdot\hat{\varepsilon}'\right|^2 \qquad P = \left|\hat{\varepsilon}\cdot\hat{\varepsilon}'\right|^2 = \begin{cases} 1 & \text{synchrotron: vertical scattering plane} \\ \cos^2\psi & \text{synchrotron: horizontal scattering plane} \\ \frac{1}{2}\left(1+\cos^2\psi\right) & \text{unpolarized source} \end{cases}$$





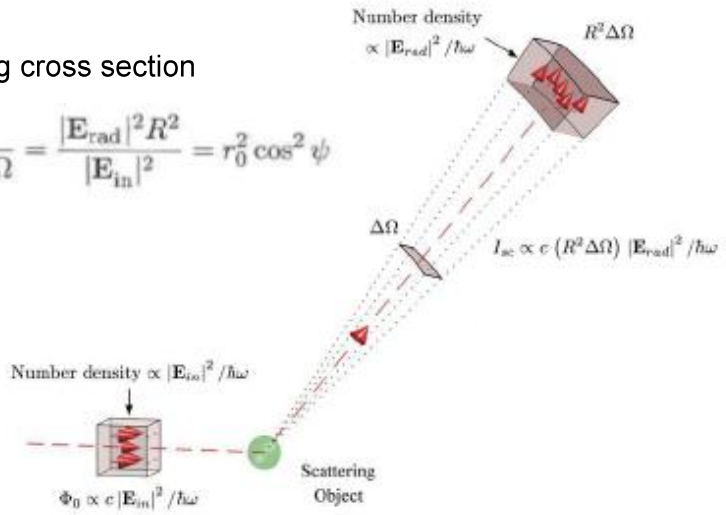
THOMSON SCATTERING CROSS SECTION

Def.: Thomson differential cross section

$$\left(\frac{d\sigma}{d\Omega}\right) = \frac{(\text{Number of X-rays scattered per second into } \Delta\Omega)}{(\text{Incident flux})(\Delta\Omega)}$$

Thomson scattering cross section

$$\left(\frac{d\sigma}{d\Omega}\right) = \frac{I_{sc}}{(I_0/A_0)\Delta\Omega} = \frac{|\mathbf{E}_{rad}|^2 R^2}{|\mathbf{E}_{in}|^2} = r_0^2 \cos^2\psi$$

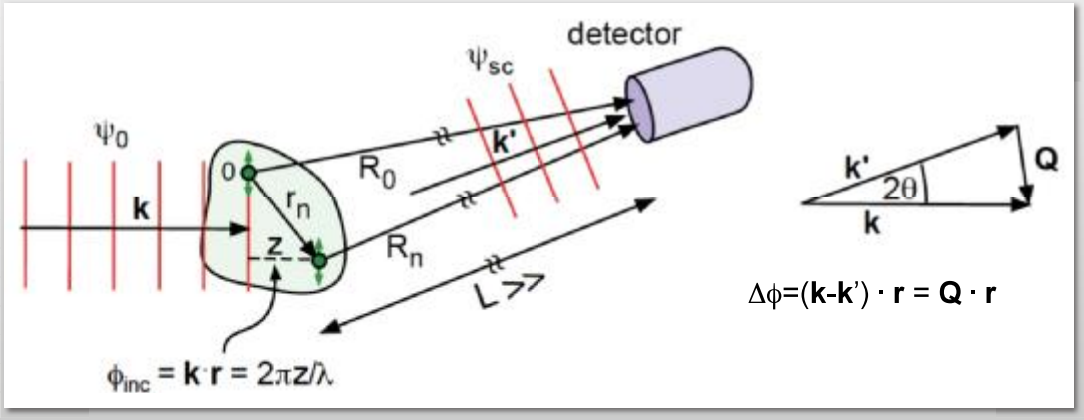


ELASTIC SCATTERING BY TWO ELECTRONS

- Interference between scattered X-rays observed in the direction **k'** and at large distances (far field limit), with $|\mathbf{k}| = |\mathbf{k'}| = 2\pi/\lambda$.
- The incident wave **k** arrives at the second electron at $\mathbf{r}_n$ with a phase shift ϕ_{inc}
- The phase difference between the two scattered X-rays is $\Delta\phi = (\mathbf{k}-\mathbf{k'}) \cdot \mathbf{r} = \mathbf{Q} \cdot \mathbf{r}$

$$\frac{d\sigma}{d\Omega} = \frac{|\psi_{sc}|^2}{|\psi_0|^2} L^2 = 2r_0^2\,[1+\cos(\mathbf{Q}\cdot\mathbf{r}_n)]$$

Elastic scattering cross section



$\Delta\phi = (\mathbf{k}-\mathbf{k'}) \cdot \mathbf{r} = \mathbf{Q} \cdot \mathbf{r}$

RANDOM DISTRIBUTION OF TWO ELECTRONS

The scattering intensity depends from the relative orientation of scattering vector Q and the vector r_n.

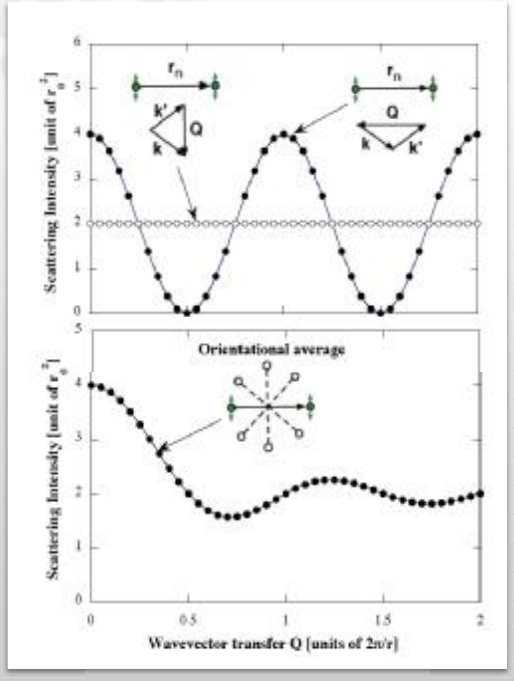
Scattering intensity (two electrons)

$$\frac{d\sigma}{d\Omega} = 2r_0^2(1+\cos(\mathbf{Q}\cdot\mathbf{r}))$$

Scattering intensity (two electrons): orientational average

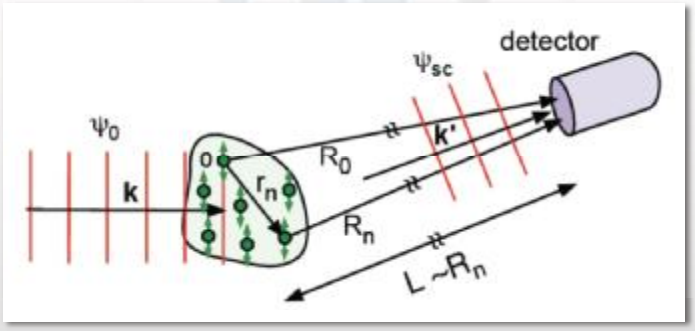
$$\langle\frac{d\sigma}{d\Omega}\rangle_{\mathbf{or.av.}} = 2r_0^2(1+\langle e^{(i\mathbf{Q}\cdot\mathbf{r})}\rangle_{\mathbf{or.av.}})$$

$$\langle e^{(i\mathbf{Q}\cdot\mathbf{r})}\rangle_{\mathbf{or.av.}} = \frac{\sin(Qr)}{Qr}$$



SCATTERING BY MANY ELECTRONS

In general if we have a random distribution of electrons, the scattering function is obtained sum coherently all the individual terms



$$\psi^{sc} = \sum_n \psi_n^{sc} = \sum_n -r_0\frac{e^{i\mathbf{k}\cdot\mathbf{R}_n}}{R_n} \approx \frac{1}{L}\sum_n -r_0 e^{i\mathbf{k}\cdot\mathbf{R}_n} = \frac{1}{L}\sum_n -r_0 e^{i\mathbf{k}\cdot(\mathbf{R}_0+\mathbf{r}_n)} = \frac{1}{L}\sum_n -r_0 e^{i\mathbf{k}\cdot\mathbf{r}_n}$$

$$\frac{d\sigma}{d\Omega} = r_0^2\left|\sum_n e^{i\mathbf{Q}\cdot\mathbf{r}_n}\right|^2$$

Electron form factor

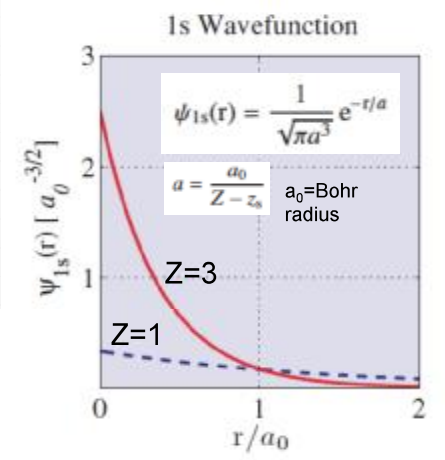
Obs.: We neglect polarization factor ($\varepsilon\cdot\varepsilon'$)

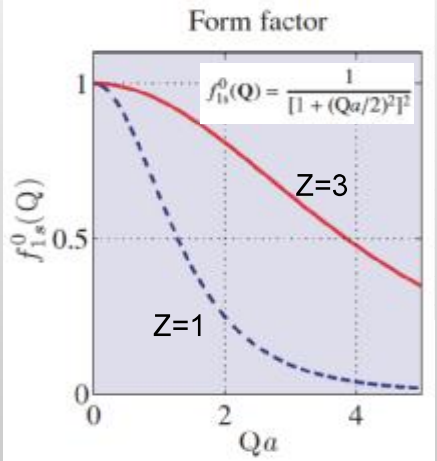
SCATTERING BY ONE ATOM: THE FORM FACTOR

The form factor is related to the Fourier transform of charge density distribution:

$$f^0(\mathbf{Q}) = -r_0 \int \rho(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r} = \begin{cases} Z & \text{for } \mathbf{Q} \to 0 \\ 0 & \text{for } \mathbf{Q} \to \infty \end{cases}$$

The Q dependence is due to the fact that the Thomson scattering is produced by all atomic electrons, which have a spatial extent of the same order of magnitude as the X-ray wavelength.

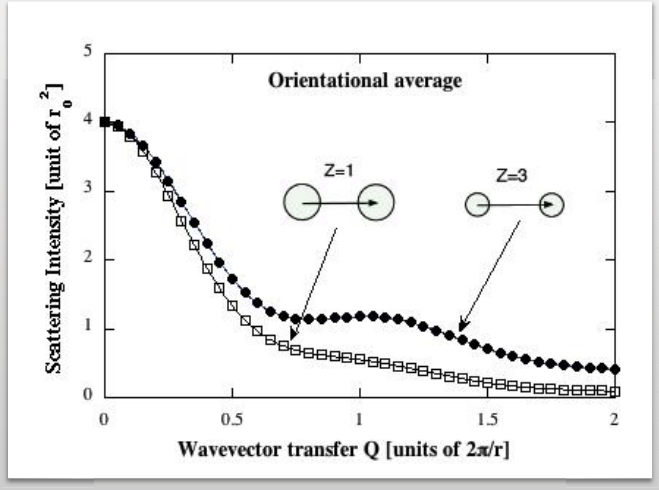


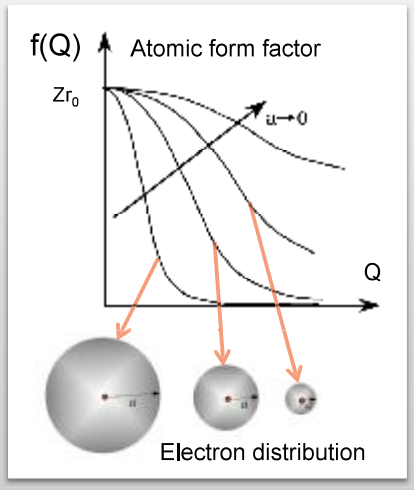


SCATTERING BY TWO ATOM

The scattering of two atoms which have a spatial extent of electron distribution of the same order of magnitude as the X-ray wavelength.

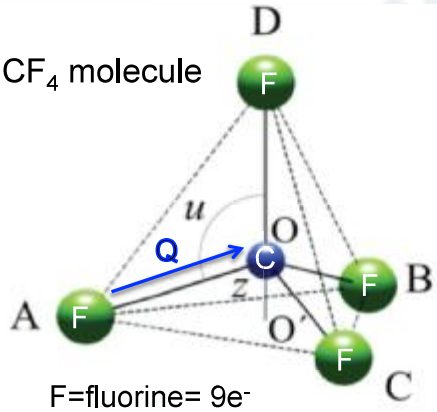
$$\langle I(\mathbf{Q}) \rangle_{\text{orient. av.}} = f_1^2 + f_2^2 + 2 f_1 f_2 \langle e^{i\mathbf{Q}\cdot\mathbf{r}} \rangle_{\text{orient. av.}}$$



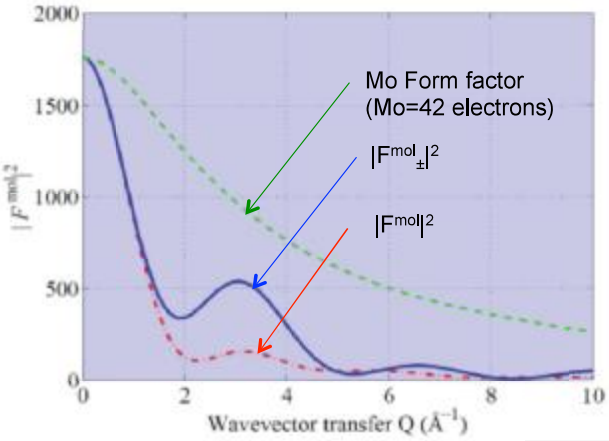


SCATTERING FROM A MOLECULE

CF_4 molecule



F=fluorine= 9e⁻
C=carbon= 6e⁻



Molecular structure factors $F^{\mathrm{mol}}(\mathbf{Q}) = \sum_j f_j(\mathbf{Q})\, e^{i\mathbf{Q}\cdot\mathbf{r}_j}$

Q // C-F bond $F^{\mathrm{mol}}_{\pm}(\mathbf{Q}) = f^{\mathrm{C}}(\mathrm{Q}) + f^{\mathrm{F}}(\mathrm{Q})\left[3e^{\mp iQR/3} + e^{\pm iQR}\right]$

Orientational average $\left|F^{\mathrm{mol}}\right|^2 = \left|f^{\mathrm{C}}\right|^2 + 4\left|f^{\mathrm{F}}\right|^2 + 8 f^{\mathrm{C}} f^{\mathrm{F}} \frac{\sin(QR)}{QR} + 12\left|f^{\mathrm{F}}\right|^2 \frac{\sin(Q\sqrt{8/3}R)}{Q\sqrt{8/3}R}$

SCATTERING FROM SOLIDS

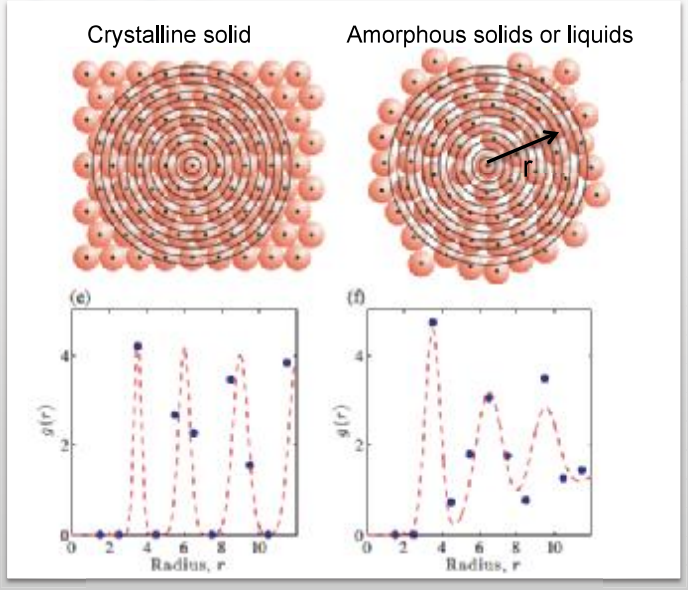
Cristalline solids exhibit long range structural order and sit on a regular array. Amorphous materials present a degree of randomness in the position of the atoms and the structural order, if present at all, can only be described in a statistical sense.

Radial density
2D N(r)/2πrdr
3D N(r)/4πr²dr

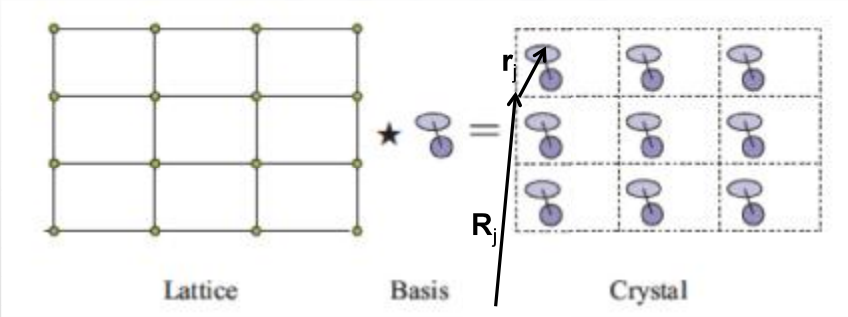
Radial distribution function
$g(r)=\rho(r)/\rho_{at}$

$g(r)\rightarrow 1$ $\rho(r)\rightarrow\rho_{at}$

The radial distribution function can be obtained from the Fourier transform of the observed intensity as a function of wavevector transform Q.



A two-dimensional crystal structure is built from the convolution of a lattice and a basis.



The Structure factor could be written as a product between the lattice sum and the unit cell structure factor.

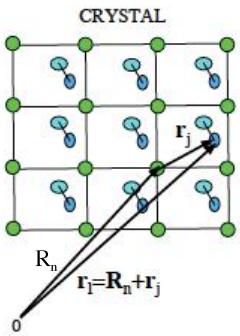
$$F^{crystal}(\mathbf{Q}) = \sum_{\mathbf{R}_n+\mathbf{r}_j}^{\text{All atoms}} f_j(\mathbf{Q}) e^{i\mathbf{Q}\cdot(\mathbf{R}_n+\mathbf{r}_j)} = \overbrace{\sum_n e^{i\mathbf{Q}\cdot\mathbf{R}_n}}^{\text{Lattice}} \overbrace{\sum_j f_j(\mathbf{Q}) e^{i\mathbf{Q}\cdot\mathbf{r}_j}}^{\text{Unit cell}}$$

The Fourier transform of the crystal (the crystal structure factor) is equal to the product of the FT of lattice and the basis

$$F^{crystal}(\mathbf{Q}) = -r_0 \sum_{\mathbf{R}_n+\mathbf{r}_j}^{all\ atoms} f_j(\mathbf{Q}) e^{i\mathbf{Q}\cdot(\mathbf{R}_n+\mathbf{r}_j)}$$

$$= -r_0 \sum_n^{lattice} e^{i\mathbf{Q}\cdot\mathbf{R}_n} \sum_j^{unit\ cell} f_j(\mathbf{Q}) e^{i\mathbf{Q}\cdot\mathbf{r}_j}$$

FT Lattice FT Basis



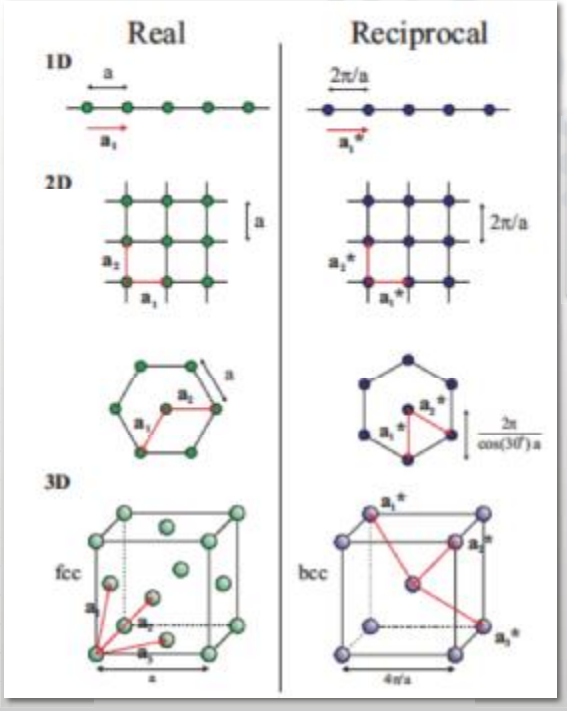
$$S_N(\mathbf{Q}) = \sum_n^{lattice} e^{i\mathbf{Q}\cdot\mathbf{R}_n}$$

The lattice sum is non vanishing only when:

$$\mathbf{Q}\cdot\mathbf{R}_n = 2\pi \times \text{integer}$$

$$\mathbf{G}_{hkl}\cdot\mathbf{R}_n = 2\pi \times integer = 2\pi(hn_1 + kn_2 + ln_3)$$ Laue conditions

THE RECIPROCAL SPACE



Definition of reciprocal space

$$\mathbf{a}_i \cdot \mathbf{a}_j^* = 2\pi\,\delta_{ij} \qquad \mathbf{G} = h\,\mathbf{a}_1^* + k\,\mathbf{a}_2^* + l\,\mathbf{a}_3^*$$

$$\mathbf{a}_1^* = \frac{2\pi}{v_c}\,\mathbf{a}_2 \times \mathbf{a}_3 \qquad \mathbf{a}_2^* = \frac{2\pi}{v_c}\,\mathbf{a}_3 \times \mathbf{a}_1 \qquad \mathbf{a}_3^* = \frac{2\pi}{v_c}\,\mathbf{a}_1 \times \mathbf{a}_2$$

Laue condition for x-ray diffraction

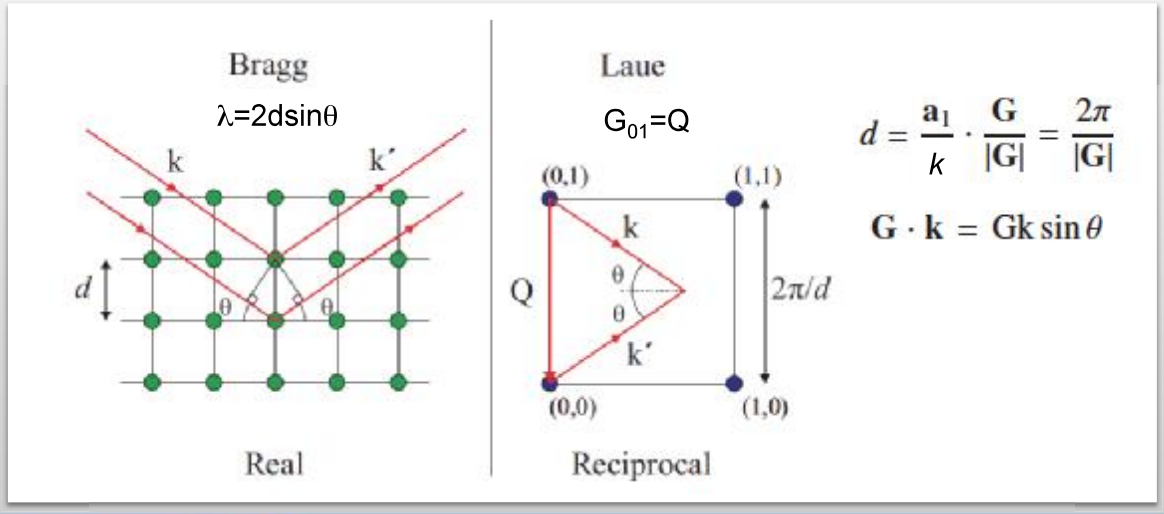
$$\mathbf{Q} = \mathbf{G}$$

$$\mathbf{G} \cdot \mathbf{R_n} = 2\pi(hn_1 + kn_2 + ln_3)$$

EQUIVALENCE OF BRAGG AND LAUE CONDITIONS

Bragg condition for x-ray elastic diffraction:
the path length difference between the incident k and reflected k' waves is an integer multiple of their wavelength λ.

The Bragg and Laue conditions are equivalent:
- G_{hkl} is perpendicular to the planes with Miller indices (hkl).
- $|G_{hkl}|$ is equal to the inverse of lattice spacing d_{hkl} of (hkl) planes



$$d = \frac{\mathbf{a}_1}{k} \cdot \frac{\mathbf{G}}{|\mathbf{G}|} = \frac{2\pi}{|\mathbf{G}|}$$

$$\mathbf{G} \cdot \mathbf{k} = Gk\sin\theta$$

THE STRUCTURE FACTOR

- *The Structure factor F(hkl) describes the interference between the resultatant waves diffused from each atom in the unit cell for any given reciprocal lattice vector Q=G_{hkl} i.e.:*

$$\left(\frac{d\sigma}{d\Omega}\right)_{Bragg} = N\frac{(2\pi)^3}{v_0}\sum_{hkl}\delta(\mathbf{Q}-\mathbf{G}_{hkl})\,|F(hkl)|^2$$

N° unit cell — *unit cell volume* — ***Laue's condition***

$$F(hkl) = \sum_s f_s\, e^{i\mathbf{G}_{hkl}\cdot\mathbf{d}_s - W_s}$$

Structure factor: information about the atom distribution inside the unit cell

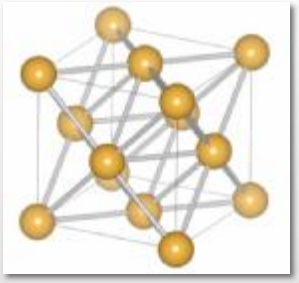
$$f_s = \int_{atom} \rho_s(\mathbf{r}')e^{-i\mathbf{Q}\cdot\mathbf{r}'}d\mathbf{r}'$$

f_s = atomic scattering amplitude for the atoms s related to the Fourier transform of the atomic electron density

UNIT CELL STRUCTURE FACTOR

Example: FCC lattice
Composed by four atoms at (000), (½½0), (0½½) and (½0½) unit cell positions

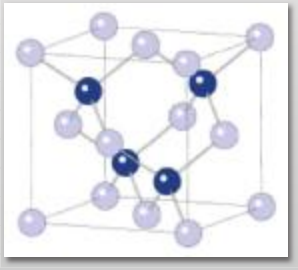
$$\begin{aligned} F_{hkl}^{fcc} &= f(\mathbf{G})\sum_j e^{i\mathbf{G}\cdot\mathbf{r}_j} \\ &= f(\mathbf{G})(1+e^{i\pi(h+k)}+e^{i\pi(k+l)}+e^{i\pi(l+h)}) \\ &= f(\mathbf{G})\times\begin{cases}4 & \text{if } h,k,l \text{ are all even or all odd}\\ 0 & \text{otherwise}\end{cases}\end{aligned}$$



Example: AsGa (Zinc-blende)
convolution of a FCC lattice and a "motif" of two atoms at (000) and (¼¼¼)

$$\begin{aligned} F_{hkl}^{GaAs} &= (1+e^{i\pi(h+k)}+e^{i\pi(k+l)}+e^{i\pi(l+h)}) \\ &\quad\times(f^{Ga}(\mathbf{G})+f^{As}(\mathbf{G})e^{i2\pi(h/4+k/4+l/4)}) \end{aligned}$$

$$F_{200}^{GaAs} = 4(f^{Ga}(2,0,0)-f^{As}(2,0,0))$$

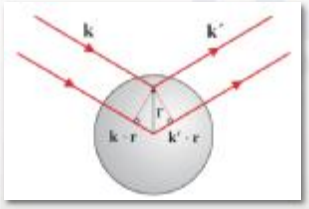


COHERENT ELASTIC SCATTERING PROCESSES

Scattering processes conserve the number of photons
If the photon energy is conserved, the scattering is elastic

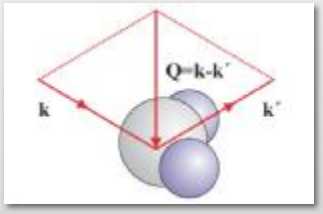
Structure factors

one atom



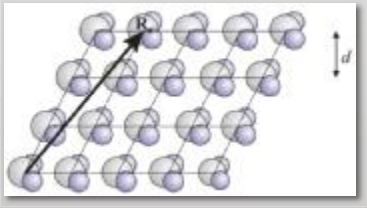
$$-r_0 f^0(\mathbf{Q}) = -r_0 \int \rho(\mathbf{r})\, e^{i\mathbf{Q}\cdot\mathbf{r}}\, d\mathbf{r}$$

a molecule



$$F^{\text{mol}}(\mathbf{Q}) = \sum_j f_j(\mathbf{Q})\, e^{i\mathbf{Q}\cdot\mathbf{r}_j}$$

a crystal



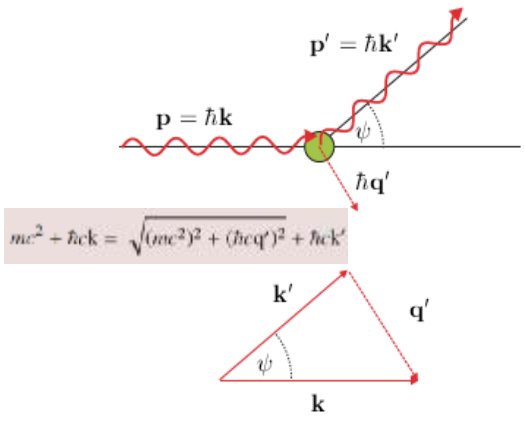
$$F^{\text{crystal}}(\mathbf{Q}) = \overbrace{\sum_j f_j(\mathbf{Q}) e^{i\mathbf{Q}\cdot\mathbf{r}_j}}^{\text{Unit cell structure factor}} \overbrace{\sum_n e^{i\mathbf{Q}\cdot\mathbf{R}_n}}^{\text{Lattice sum}}$$

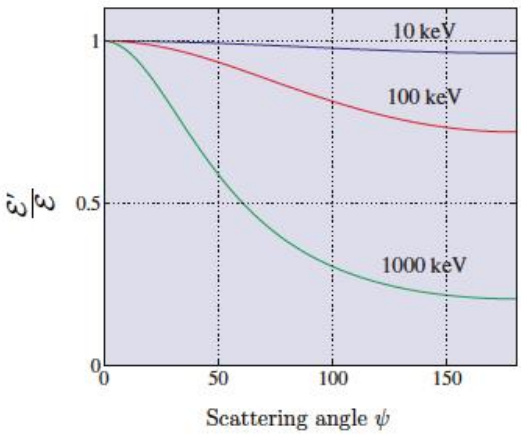
INELASTIC SCATTERING BY FREE ELECTRONS

Compton scattering:
Inelastic collision between a photon and an electron at the rest in which part of the the photon energy is transferred to the electron.
This scattering is **incoherent!**

$$\frac{k}{k'} = 1 + \lambda_C k(1 - \cos\psi) = \frac{\mathcal{E}}{\mathcal{E}'} = \frac{\lambda'}{\lambda}$$

$$\lambda_C = \frac{\hbar}{mc} = 3.86 \times 10^{-3}\ \text{Å}$$





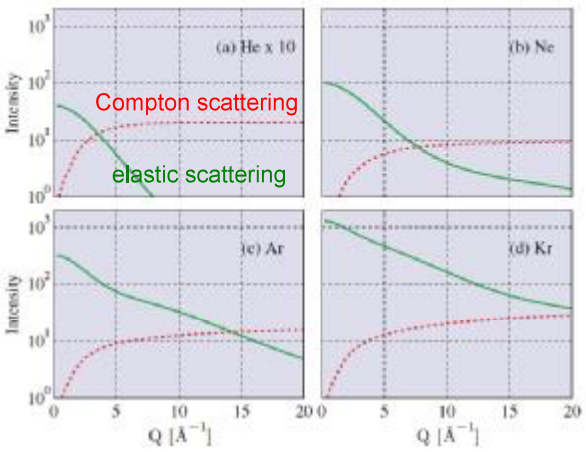
INELASTIC SCATTERING BY FREE ELECTRONS

The inelastic scattering dominates at high Q vectors and for low Z elements

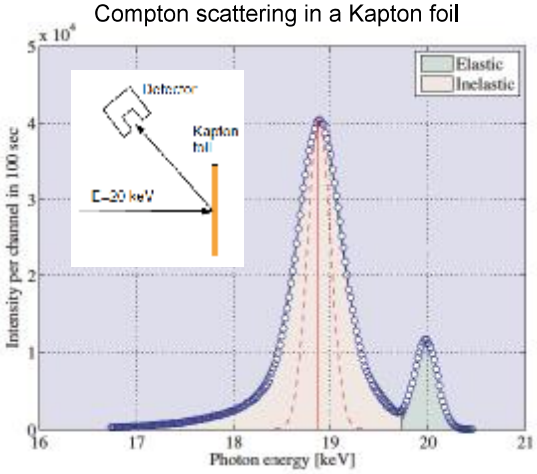
Thomson scattering intensity approach Z^2 when Q➔0

Compton scattering approaches Z when Q➔∞

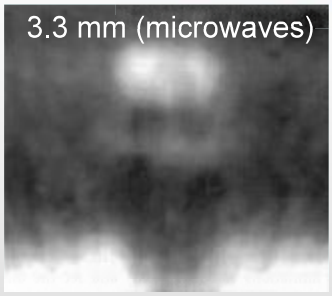
Elastic and inelastic scattering in noble gas

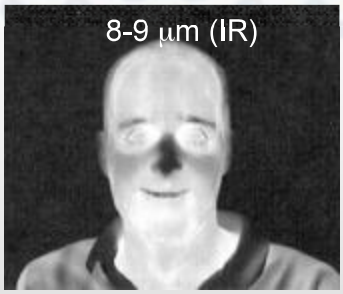


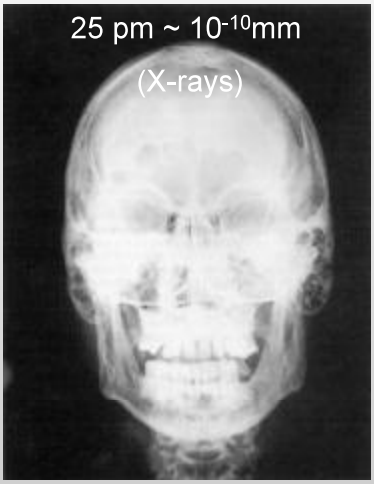
Compton scattering in a Kapton foil



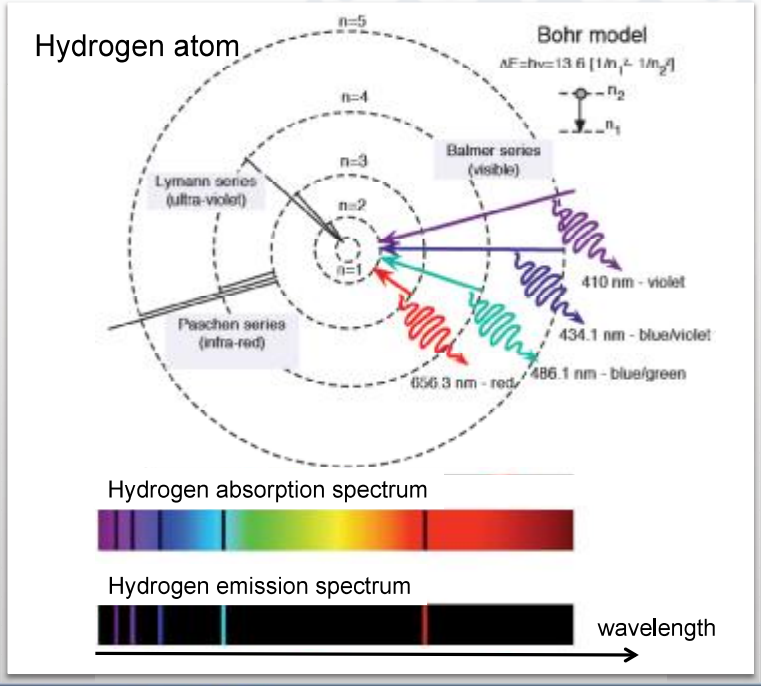
PHOTON ABSORPTION

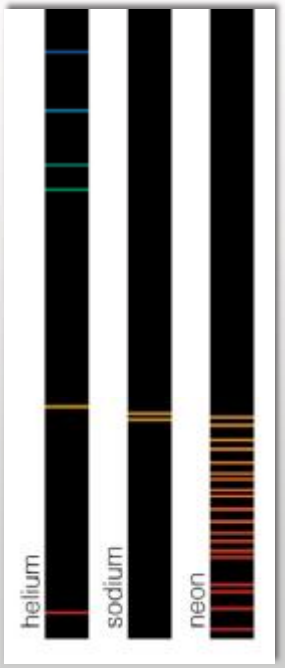






Photons are adsorbed or re-emitted by atomic electrons at discrete energies.
Determination of elemental composition by its electromagnetic response.

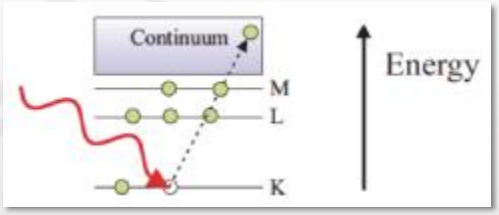




Absorption and emission processes are tools for basic analysis of the electronic structure of atom, molecules and solids over different energy scales.

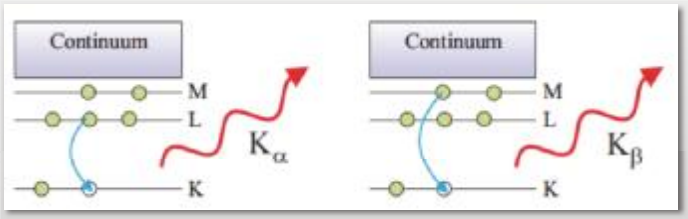
Photo-electric absorption

Photon absorbed and electron emitted in the continuum



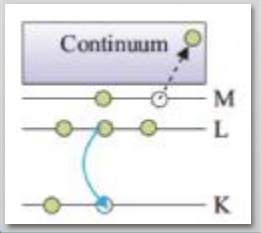
Fluorescent emission

An electron from the outer shell fill the hole and emit a photon



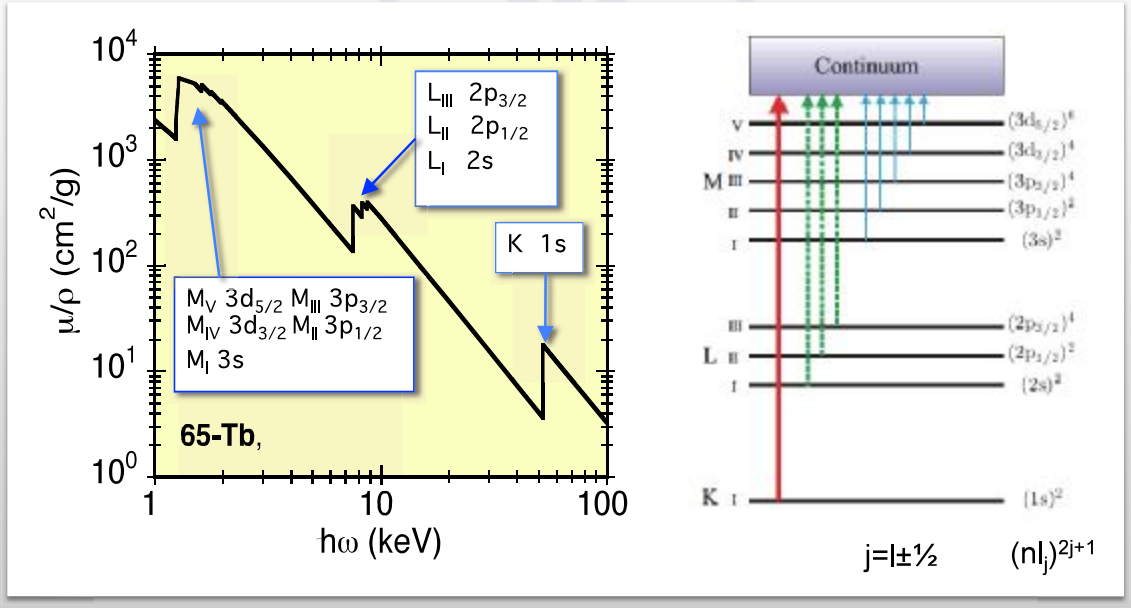
Auger electron emission

The atom relax into the ground state by emitting an electron



X-RAY ABSORPTION EDGES

X-rays energies are able to extract atomic electrons from the atomic core!
The element-specific energies of the discontinuous jumps in the x-rays absorption spectra are called absorption edges.



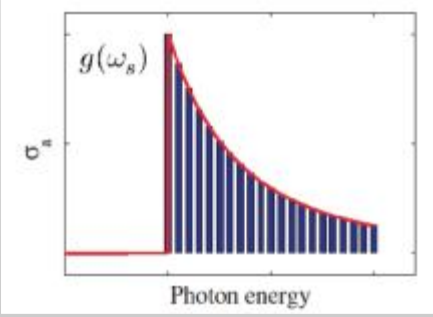
BOUND ELECTRONS AND DISPERSION CORRECTIONS

Because the electrons are bound in atoms with discrete energies, a more elaborate model than that of a cloud of free electrons must be invoked.

The scattering amplitude includes two energy dependent term f'(ω) and f"(ω) which are called "dispersion corrections".

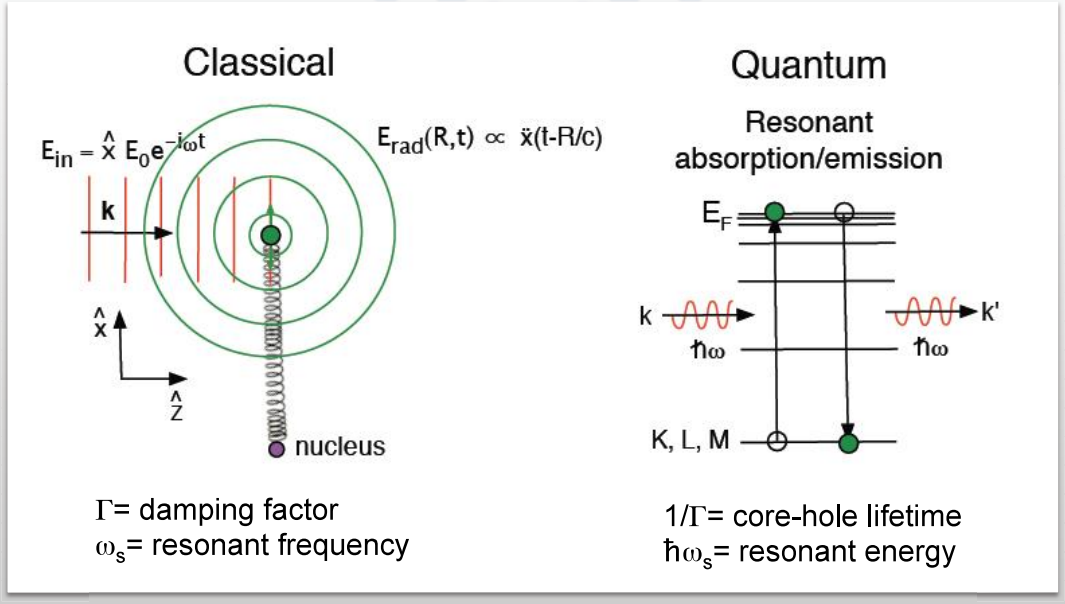
$$f(\mathbf{Q},\omega) = f^0(\mathbf{Q}) + f'(\omega) + i\,f''(\omega)$$

The dispersion corrections are derived by treating atomic electrons as harmonic oscillators. The absorption cross section σ_a is a superposition of oscillators with relative weights, so-called oscillator strengths, $g(\omega_s)$, proportional to $\sigma_a(\omega = \omega_s)$.



RESONANT SCATTERING: CLASSICAL OSCILLATOR MODEL

We suppose the electron be subject to the electric field E_{in} of an incident X-ray beam and to a damping term proportional to the electron velocity $\Gamma\dot{x}$ which represents dissipation of energy.



Γ= damping factor
ω_s= resonant frequency

$1/\Gamma$= core-hole lifetime
$\hbar\omega_s$= resonant energy

BOUND ELECTRONS: THE CLASSICAL FORCED OSCILLATOR MODEL

The amplitude of the forced oscillations:

$$x_0 = -\left(\frac{e\,E_0}{m}\right)\frac{1}{(\omega_s^2 - \omega^2 - i\,\omega\Gamma)}$$

Γ= damping factor
ω_s= resonant frequency

The radiated field E_{rad} is proportional to the acceleration of the electron $\ddot{x}(t-R/c)$ at the detector position R and at retarded time t'=t-R/c:

$$\ddot{x}(t - R/c) = -\omega^2 x_0 e^{-i\,\omega t} e^{i\,(\omega/c)R}$$

$$\frac{E_{rad}(R,t)}{E_{in}} = -r_0 \frac{\omega^2}{(\omega^2 - \omega_s^2 + i\,\omega\Gamma)}\left(\frac{e^{ikR}}{R}\right)$$

$$f_s \approx -r_0\left(1 + \frac{\omega_s^2}{\omega^2 - \omega_s^2 + i\omega\Gamma}\right) = f_0 + f'(\omega) + if''(\omega)$$

Total scattering length

Thomson term (Q dependence)

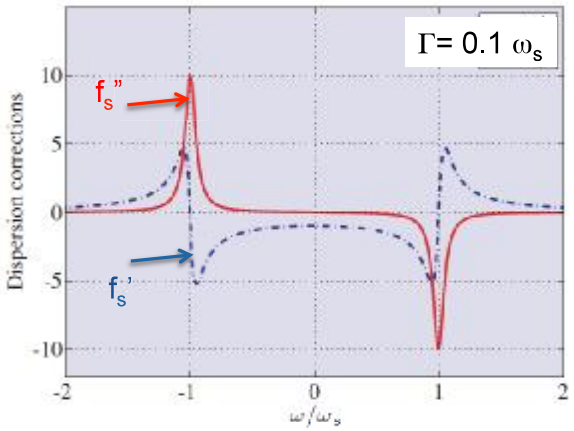
Frequency-dependent refraction index n(ω)

Absorption correction (dissipation term)

Real and imaginary part of dispersion correction to the scattering factor

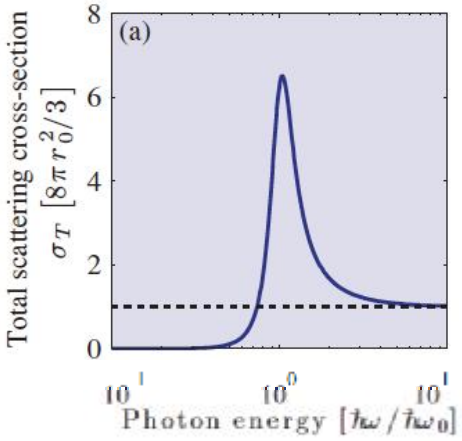
$$f_s' = \frac{\omega_s^2(\omega^2 - \omega_s^2)}{(\omega^2 - \omega_s^2)^2 + (\omega\Gamma)^2}$$

$$f_s'' = -\frac{\omega_s^2\omega\Gamma}{(\omega^2 - \omega_s^2)^2 + (\omega\Gamma)^2}$$



Total scattering cross section for a bound electron

$$\sigma_T = \left(\frac{8\pi}{3}\right)\frac{\omega^4}{(\omega^2 - \omega_s^2)^2 + (\omega\Gamma)^2}\, r_0^2$$



Scattering and refraction are alternative ways to view the same physical phenomenon.

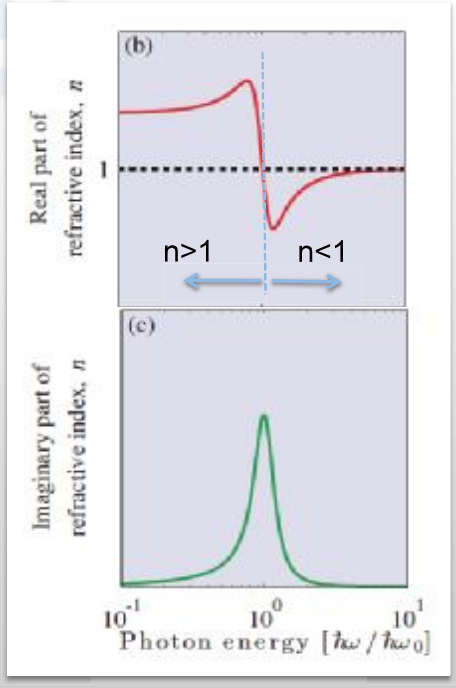
The existence of resonant scattering terms arising from the dispersion corrections can therefore be expected to lead to a frequency dependence of the refractive index n(ω).

$$n^2 = 1 + \left(\frac{e^2\rho}{\epsilon_0 m}\right)\frac{1}{(\omega_s^2 - \omega^2 - i\,\omega\Gamma)}$$

For $\omega << \omega_s$ => n>1 visible light
For $\omega >> \omega_s$ => n<1 x-rays

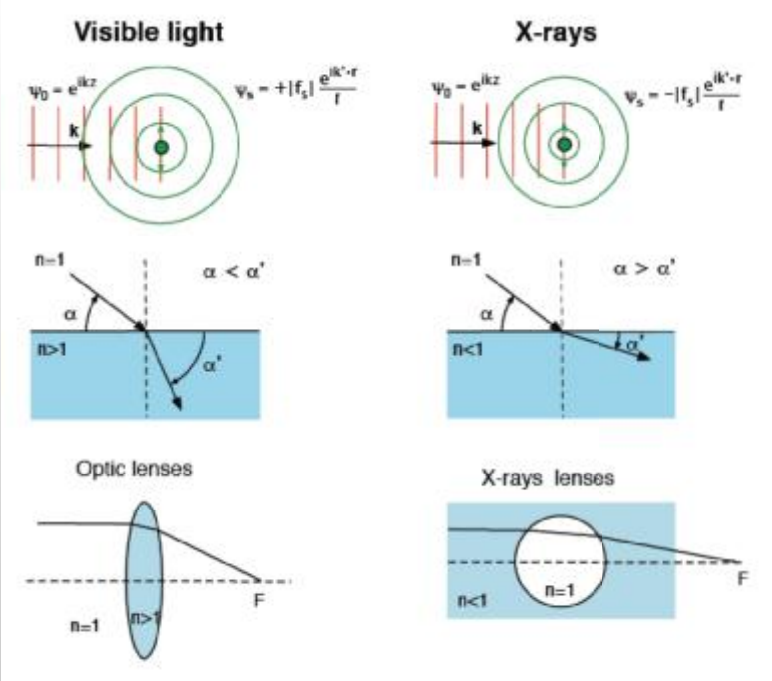
Notice that if $\omega >> \omega_s >> \Gamma$

$$n \approx 1 - \frac{1}{2}\frac{e^2\rho}{\epsilon_0 m\omega^2} = 1 - \frac{2\pi\rho r_0}{k^2}$$



REFRACTION: VISIBLE LIGHT VS X-RAYS

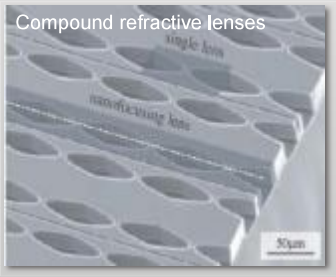
Snell law: $n_1\cos\alpha=n_2\cos\alpha'$



Refraction index for X-rays:

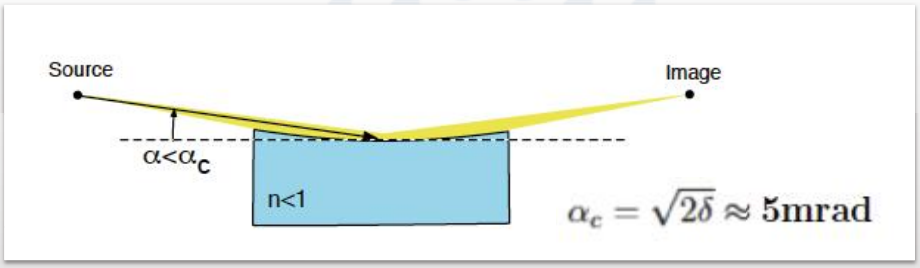
$n=1-\delta+i\beta$

$\delta(air)\sim10^{-8}$
$\delta(solids)\sim10^{-5}$
$\beta\sim10^{-8}<<\delta$



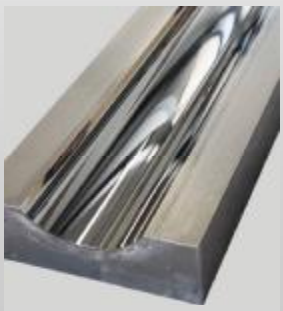
REFLECTION OF X-RAYS

The critical angle for the total reflection:



High quality mirrors are required for x-rays focusing and a large radius tangential focusing

Ex: silicon mirror with toroidal shape
Distance from source p=76m,
Distance mirror object q=26m
θ=2.7 mrad
$\rho_{sagital}$=27 cm ρ_{tang}=27 km

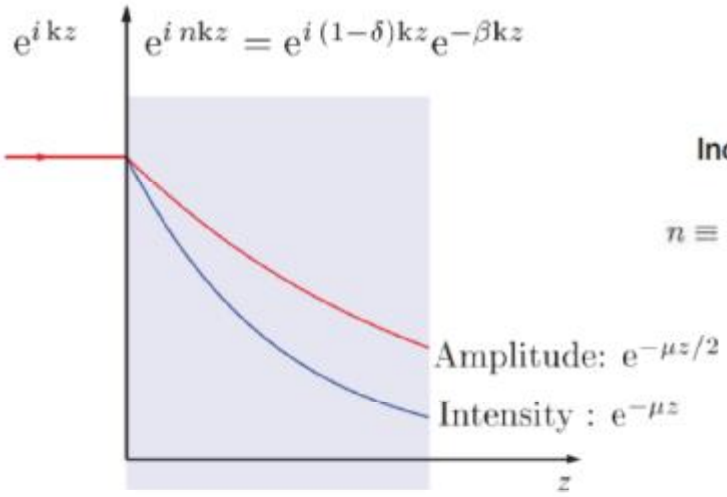


REFRACTIVE INDEX AND ABSORPTION

The attenuation of x-rays in a medium depends from the imaginary part β of refraction index:

e^{ikz} $e^{inkz} = e^{i(1-\delta)kz} e^{-\beta kz}$

$$\beta = \frac{\mu}{2k}$$



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

Including dispersion corrections:

$$n \equiv 1 - \frac{2\pi \rho_a r_0}{k^2} \{f^0(0) + f' + i f''\}$$

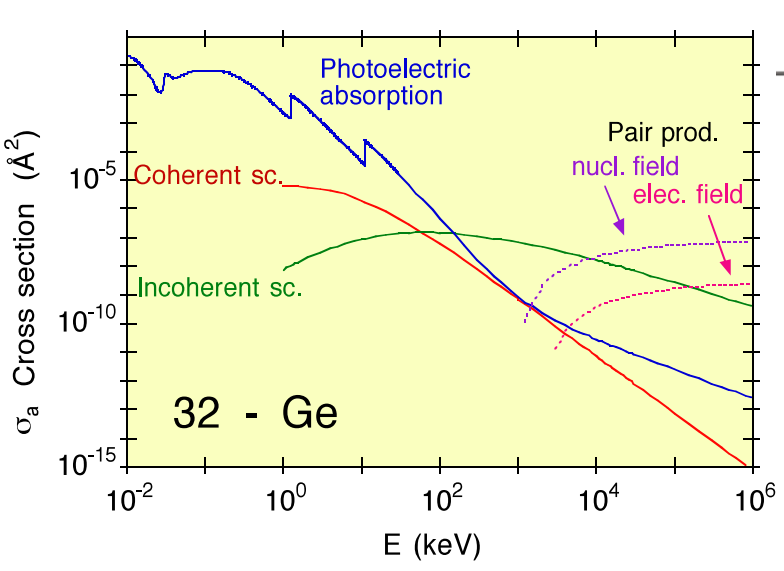
and:

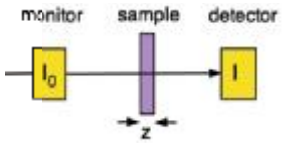
$$f'' = -\left(\frac{k}{4\pi r_0}\right) \sigma_a$$

Optical theorem

ABSORPTION CROSS SECTION

The absorption cross section can be measured by a transmission method.



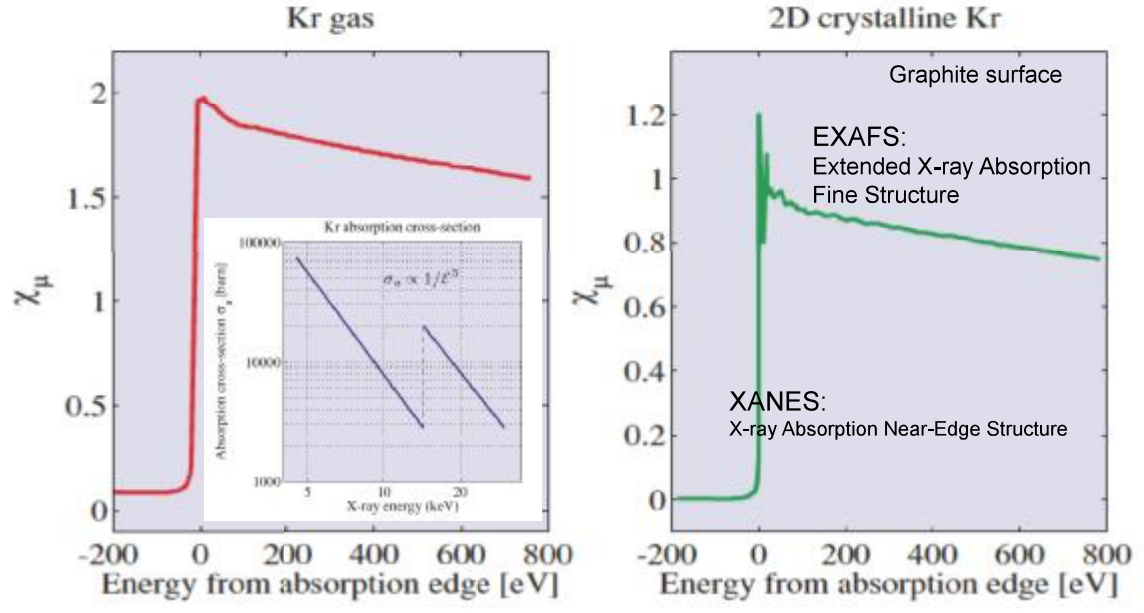


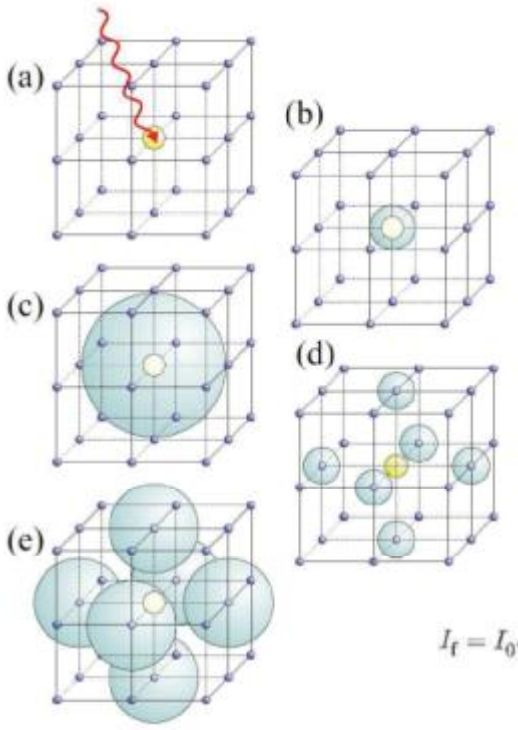
$$T = \frac{I}{I_0} = e^{-\mu z}$$

Absorption coefficient

$$\mu = \left(\frac{\rho_m N_A}{M}\right) \sigma_a$$

The transitions from core electron levels to empty states produce in the absorption spectrum simple threshold structures in the case of isolated atoms or modulated structures well above the threshold when the atom is embedded in a crystalline environment.





$$\chi(q(\mathcal{E})) = \frac{\mu_\chi(\mathcal{E}) - \mu_0(\mathcal{E})}{\mu_0(\mathcal{E})}$$

$$\frac{\hbar^2 q^2}{2m} = \mathcal{E} - \hbar\omega_K$$

In transmission:

$$T = \frac{I_1}{I_0} = e^{-\mu(\mathcal{E})d}$$

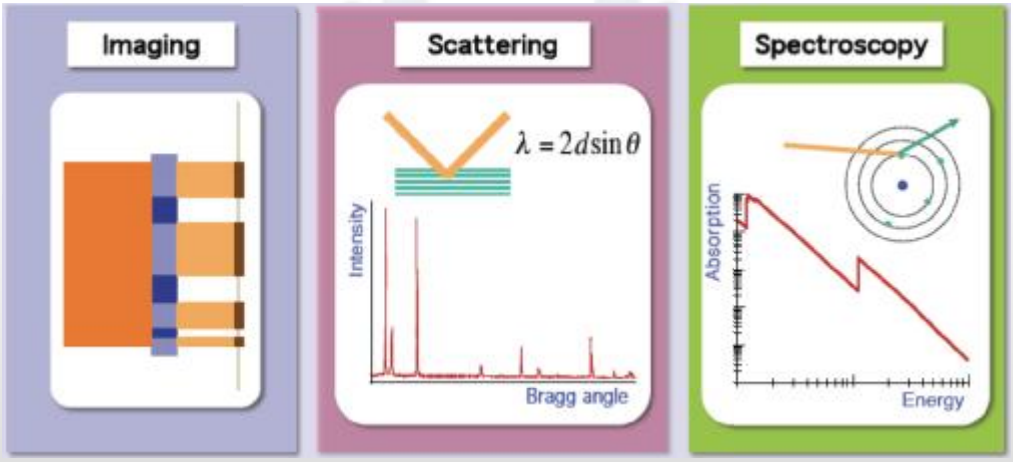
$$\mu(\mathcal{E}) = \mu_A(\mathcal{E}) + \mu_\chi(\mathcal{E})$$
$$= \mu_A(\mathcal{E}) + \mu_0(\mathcal{E})\,[1 + \chi(q)]$$

In fluorescence:

$$I_f = I_0 \epsilon \left(\frac{\Delta\Omega}{4\pi}\right) \frac{\mu_\chi(\mathcal{E})}{\mu(\mathcal{E}) + \mu(\mathcal{E}_f)} \left[1 - e^{-(\mu(\mathcal{E}) + \mu(\mathcal{E}_f))d}\right]$$

X-RAYS METHODS

A Light for Science



Spatial informations

Structural informations

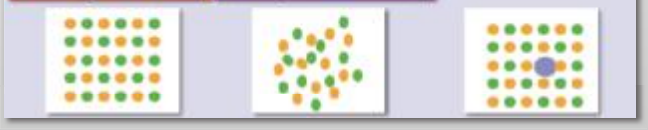
Absorption contrast

Phase contrast

Long-range order (crystals)

Short-range order (amorphous solids)

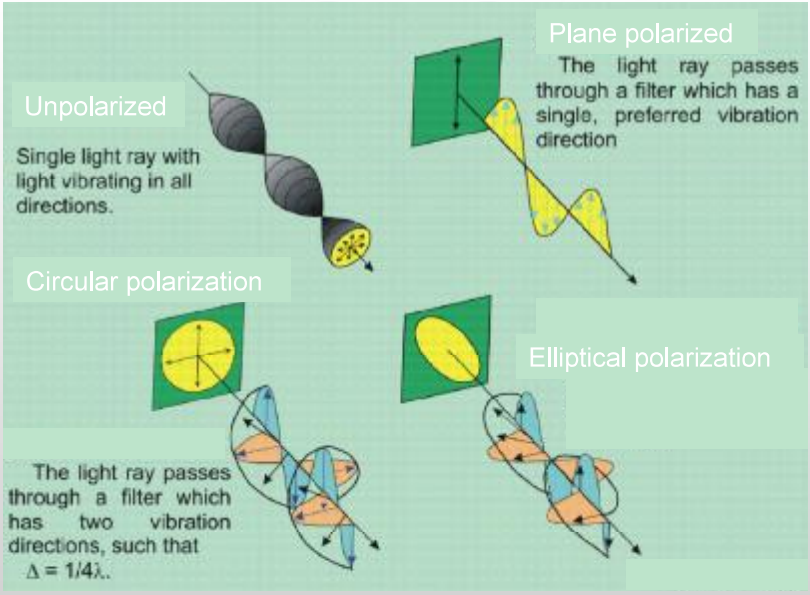
Local properties



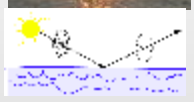
LIGHT POLARIZATIONS

A Light for Science

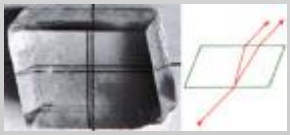
Direct Sun light



Reflected Sun light



Birifrangence in calcite



Linear dichroism
Produced by the preferential absorption of one of the two orthogonal photon polarization

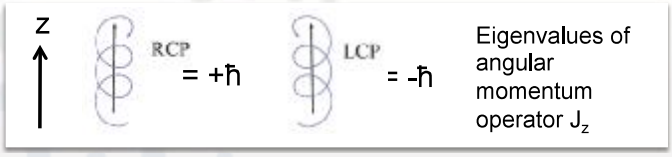
Linearly oriented polimers
When the electric field is parallel to the preferential molecular axis, it is absorbed

Circular dichroism
Produced by the preferential absorption of one of the two circular photon polarization

Combination of λ/4 and linear polarized filters have the different effect on circular polarization.
Circular dichroism is found also in chiral molecules which select only one circular polarization (ex. sugar)

Quantum description of a a circular polarised photon beam:

RCP and LCP eigenstates of J_z

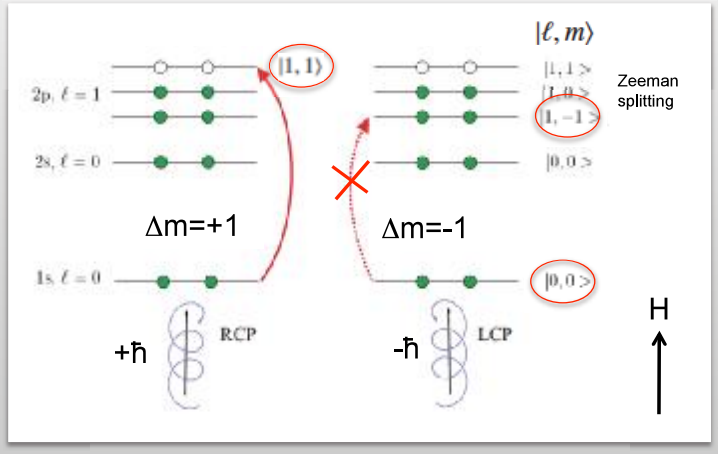


The sum rule for the conservation of angular momentum in electronic transition produces a difference in the absorption of RCP and LCP photons.

Ex.: Dipole electric transitions in Oxygen (8 electrons)

selection rule: Δl±1
(odd function for coordinate exch.)

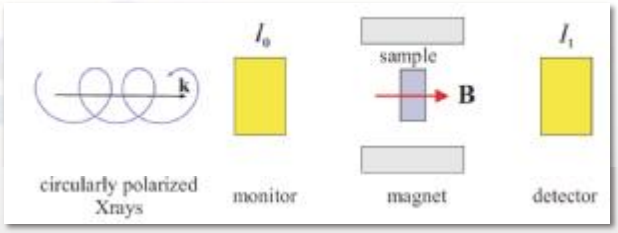
Transition allowed:
Δm=+1 for RCP
Δm=-1 for LCP



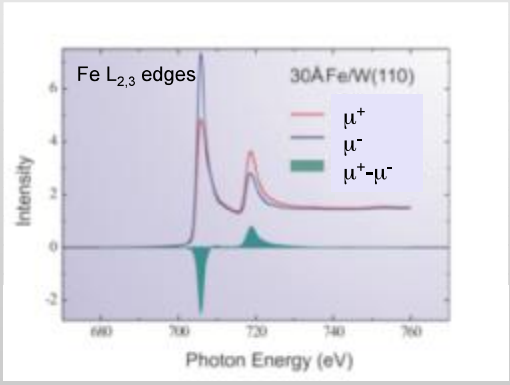
Absorption coefficients

$$\mu^+(E) = \left(\frac{1}{d}\right) \ln\left(\frac{I_0^+(E)}{I_1^+(E)}\right)$$

$$\mu^-(E) = \left(\frac{1}{d}\right) \ln\left(\frac{I_0^-(E)}{I_1^-(E)}\right)$$



The difference between $\Delta\mu=\mu^+-\mu^-$ is enhanced across the absorption edge because the electronic transitions to final states are strongly magnetically polarized



Thole et al., Phys. Rev .Lett. 68 (1992) 1943; Carra et al., Phys. Rev .Lett. 70 (1993) 694

Orbital and spin magnetic moment determination in 3d electron systems

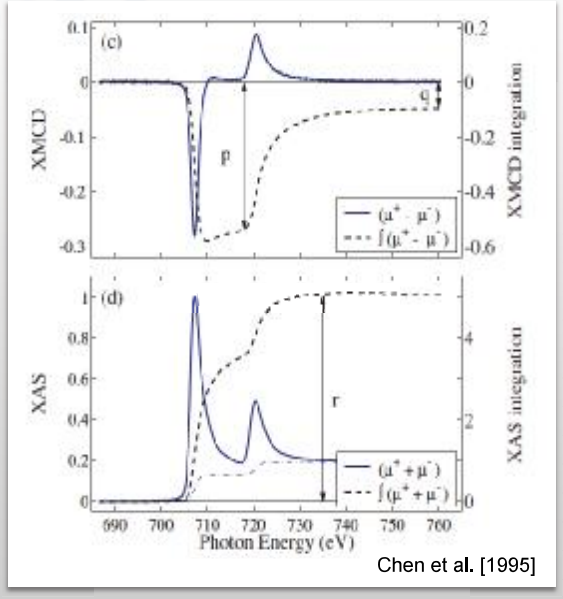
$$m_{\text{orb}}[\mu_B/\text{atom}] = -\frac{4q(10-n_{3d})}{r}$$

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

$$p = \int_{L_{III}} (\mu^+ - \mu^-)\, d\mathcal{E}$$

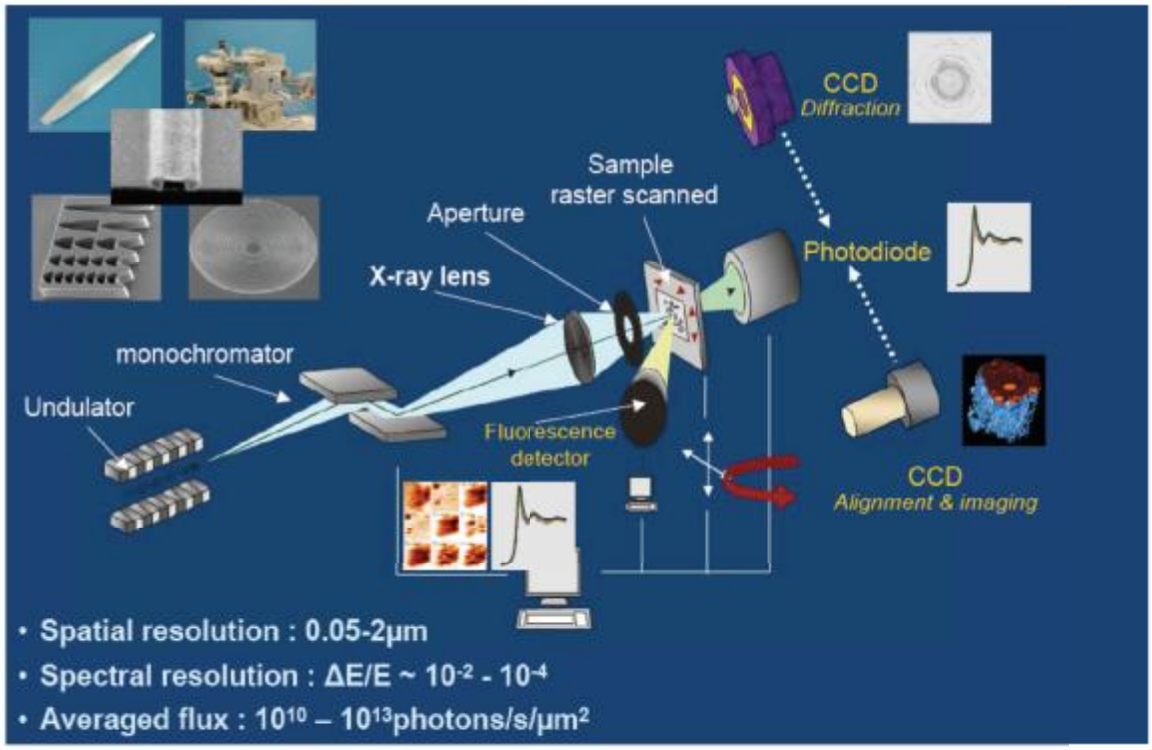
$$q = \int_{L_{III}+L_{II}} (\mu^+ - \mu^-)\, d\mathcal{E}$$

$$r = \int_{L_{III}+L_{II}} (\mu^+ + \mu^-)\, d\mathcal{E}$$



Chen et al. [1995]

X-RAYS A POWERFUL NANOPROBE



- Spatial resolution : 0.05-2µm
- Spectral resolution : $\Delta E/E \sim 10^{-2} - 10^{-4}$
- Averaged flux : $10^{10} – 10^{13}$photons/s/µm^2