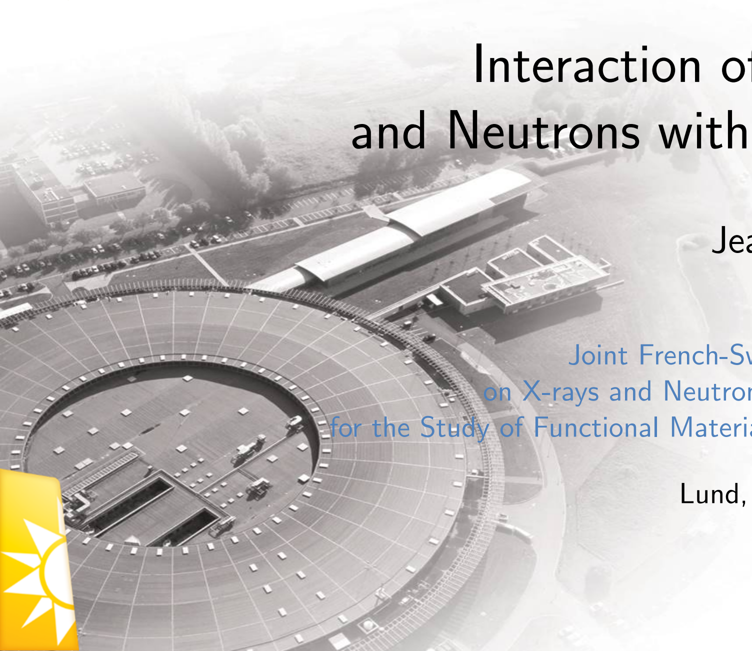


Interaction of X-rays and Neutrons with Matter

Jean Daillant

Joint French-Swedish School on X-rays and Neutrons Techniques for the Study of Functional Materials for Energy

Lund, 13 May 2019

- Provide you with a simple conceptual framework to understand synchrotron and neutron experiments .
- Give an overview of the methods which will be discussed during the week and introduce vocabulary.
- Give orders of magnitude.

Introduction
Production of X-rays and Neutrons
- Synchrotron Radiation
- Free Electron Lasers
- Neutron Sources

Interactions of X-rays and Neutrons with Matter
- Photons and Neutrons
- Fundamentals of Scattering

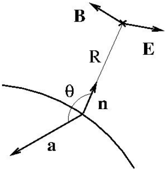
Elastic Scattering
- Elastic Scattering in the Born Approximation
- Scattering Lengths
- Reflection from Surfaces
- Refractive Index

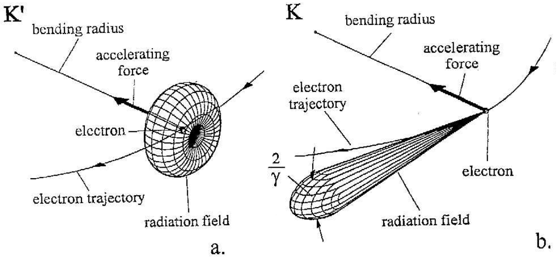
Inelastic Scattering, Spectroscopy
- Compton Scattering
- Absorption
- Photoelectric Effect - Photoemission
- Fluorescence
- Absorption Spectroscopy
- Resonant processes
- Neutron spin-echo

Production of X-rays and Neutrons

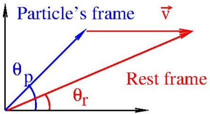
Radiation by a moving charge: $\mathbf{E} = \frac{q}{4\pi\epsilon_0 c^2 R}\mathbf{n}\times\mathbf{n}\times\mathbf{a}$

$$E = \frac{q\sin\theta}{4\pi\epsilon_0 c^2 R}$$



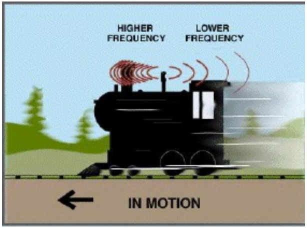


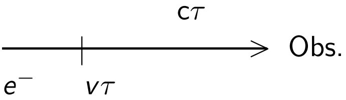
$$\gamma = \frac{1}{\sqrt{1 - v^2/c^2}}$$



- Emission in a cone $\approx 1/\gamma$, with $\gamma = E/m_{e^-}c^2$; $m_0c^2 = 511$ keV; for E = 2.75 GeV, $\gamma = 5382$, $1/\gamma = 0.186$ mrad $= 0.01$deg
- Polarization

Relativistic effects



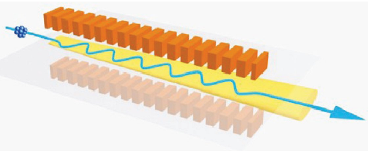


$$\beta = v/c$$

- Doppler effect
- $\lambda = (1 - \frac{v}{c})\lambda_0 = \frac{1-\beta^2}{1+\beta}\lambda_0 \approx \frac{\lambda_0}{2\gamma^2}$
  as $\beta \approx 1$.
- X-rays!

Undulators

- Magnetic field: $B_z = B_0 \cos(2\pi x/\lambda_0)$
- Lorentz force: $\gamma m_0 (dv_y/dt) \approx e v_0 B_0 \cos(2\pi x/\lambda_0)$
- Trajectory: $y = -K\lambda_0/(2\pi\gamma)\cos(2\pi x/\lambda_0)$ with $K = EB_0\lambda_0/(2\pi m_0 c)$ the undulator strength.

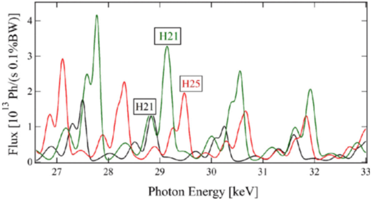


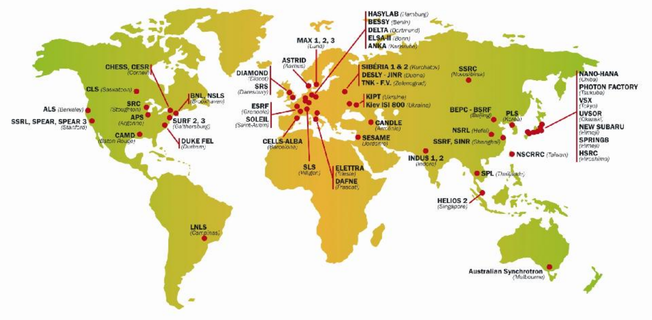
With E=2.75Gev, B_0=1T, λ_0=20mm, K=1.9, the maximum deviation of the e^- beam is 1.1μm.

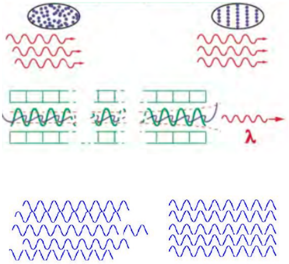
Over λ_0, extra distance $\delta L = \int_0^{\lambda_0}\left(\sqrt{1+(dy/dx)^2}-1\right)dx = K^2\lambda_0/(4\gamma^2)$

Time needed by the e^- to cover a period is larger than time needed by the photon by: $\delta t = (\lambda_0 + \delta L)/v_0 - \lambda_0/c = \lambda_0/(2\gamma^2 c)(1+K^2/2)$.

→ Harmonics: $\dfrac{\lambda_0}{2n\gamma^2}\left(1+\dfrac{K^2}{2}\right)$

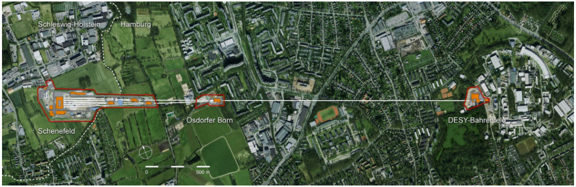


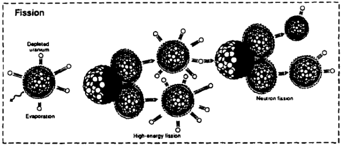

HASYLAB (Hamburg)
BESSY
DELTA (Dortmund)
ELSA-II (Bonn)
ANKA
MAX 1, 2, 3
(Lund)
ASTRID
CHESS, CESR
(Cornell)
DIAMOND
SRS
(Daresbury)
CLS (Saskatoon)
BNL, NSLS
SRC
(Stoughton)
ALS (Berkeley)
APS
(Argonne)
SSRL, SPEAR, SPEAR 3
(Stanford)
SURF 2, 3
(Gaithersburg)
CAMD
(Baton Rouge)
DUKE FEL
(Durham)
ESRF
(Grenoble)
SOLEIL
(Saint-Aubin)
CELLS-ALBA
(Barcelona)
SLS
(Villigen)
ELETTRA
(Trieste)
DAFNE
(Frascati)
LNLS
(Campinas)
SIBÉRIA 1 & 2 (Kurchatov)
DESLY - JINR (Dubna)
TNK - F.V. (Zelenograd)
KIPT (Ukraine)
Kiev ISI 800 (Ukraine)
CANDLE
SESAME
SSRC
(Novosibirsk)
BEPC - BSRF
(Beijing)
PLS
(Korea)
NSRL (Hefei)
SSRF, SINR (Shanghai)
INDUS 1, 2
(Indore)
NSCRRC (Taiwan)
SPL
HELIOS 2
(Singapore)
NANO-HANA
(Chiba)
PHOTON FACTORY
(Tsukuba)
VSX
(Tokyo)
UVSOR
(Okazaki)
NEW SUBARU
(Himeji)
SPRING8
(Himeji)
HSRC
(Hiroshima)
Australian Synchrotron
(Melbourne)

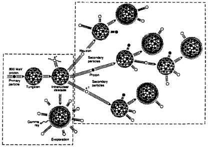


$I \propto N_{e^-}^2$

$+$ LCLS, SACLA, Pohang...

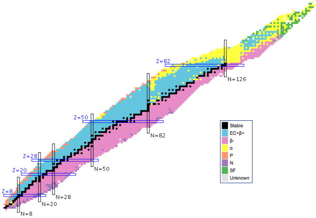






Fission:

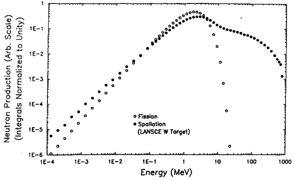
- Capture of a neutron by a "fissionable" nucleus; exothermal chain reaction.
- A few ($\propto 1$) neutron per event; mainly evaporation from fission fragments.



National Nuclear Data Center, BNL

Spallation:

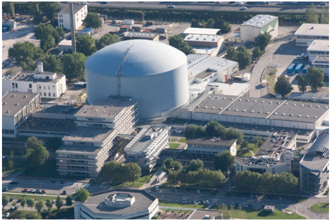
- High energy p^+ (1 GeV) hits a heavy metal target (e.g. Hg).
- $\propto 10$ neutrons per p^+; intra- and inter-molecular cascade followed by evaporation.
- Naturally pulsed sources.



G.J. Russell, ICANSXI, Tsukuba, 1990

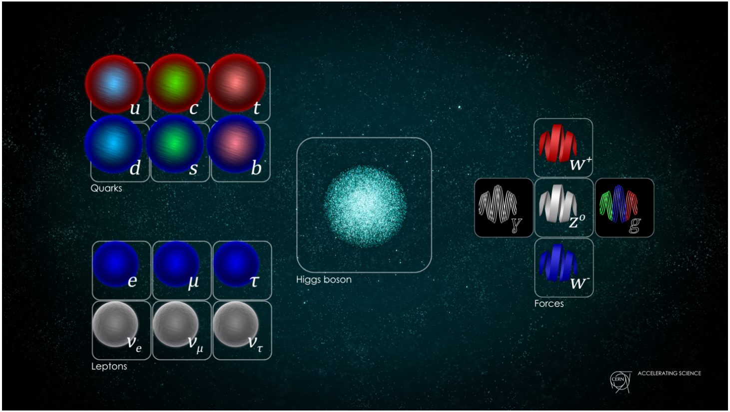
Neutron Facilities

https://www.iucr.org/resources/commissions/neutron-scattering/where-neutrons



X-ray and Neutron Interactions with Matter

Elementary particles in the standard model



Neutron:

2 down quarks, 1 up quark
Mass 1,675$\times 10^{-27}$kg $\approx$ 940 MeV/c^2
No charge
Spin 1/2

Photon:

Quantum of electromagnetic field and carrier of electromagnetic force
Zero mass, travels at the speed of light
Polarization

Photons:

Maxwell equations in a vacuum:

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial \mathbf{t}}$$

$$\nabla \times \mathbf{B} = \frac{1}{c^2}\frac{\partial \mathbf{E}}{\partial \mathbf{t}}$$

→ Propagation equation:

$$\nabla\times\nabla\times\mathbf{E}+\frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial \mathbf{t^2}} = -\nabla^2\mathbf{E}+\frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial \mathbf{t^2}} = 0$$

Solution: Plane waves
$A_0 \exp i(\omega t - k_0 x)$ $k_O = \omega/c$
Wavelength $\lambda = 2\pi/k_0$
Momentum p = h /λ = E/C
Energy $E = hc/\lambda = \hbar\omega = h\nu$

Neutrons:

Schrödinger equation:

$$i\hbar\frac{\partial|\psi\rangle}{\partial t} = \mathcal{H}|\psi\rangle; \quad \mathcal{H} = \frac{\mathbf{P}^2}{2m}; \quad \mathbf{P} = \frac{\hbar}{i}\nabla$$

Stationary solution: $-\frac{\hbar^2}{2m}\nabla^2\psi = E\psi$.
Solution $A_0 \exp i(kx - \omega t)$
$E = \hbar\omega = h\nu; = \frac{\hbar^2 k^2}{2m}$
$\lambda = h/p$ (de Broglie); $p = \hbar k\sqrt{2mE}$

"Thermal neutrons"
300K; $k_B T = 4.1 \times 10^{-21} J = 0.0256$ eV
$\lambda = h/\sqrt{2mE} \approx 1.78Å$;
$v = h/m\lambda \approx 2.22 km.s^{-1}$.

→ Time-of-flight ≡ E

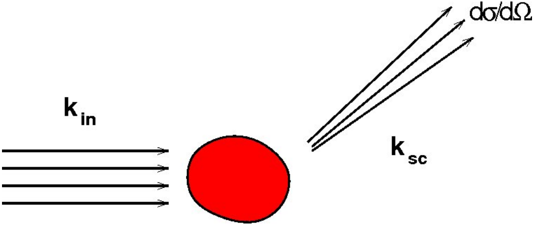
Exchange of energy?

- No: "Elastic scattering". Information on distribution of matter only.
- Yes: "Inelastic scattering": also information on the energy distribution in the sample (whatever it is).

Scattering ↔ Spectroscopy

Coherent or incoherent process?

- Yes: Possibility of interferences. Structural information can be recovered.
- No: No possibility of interferences, structural information is lost.

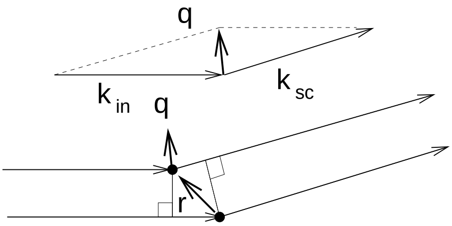


The differential scattering cross-section $d\sigma/d\Omega$ is the intensity scattered per unit solid angle in the direction $\mathbf{k}_{\mathrm{sc}}$ per unit incident flux in the direction $\mathbf{k}_{\mathrm{in}}$.

$$I = \Phi_0 \int_\Omega \frac{d\sigma}{d\Omega} d\Omega$$

Dimension of an area.

Elastic Scattering



Plane wave: $A \exp i(\omega t - \mathbf{k}.\mathbf{r})$
Phase shift: $(\mathbf{k}_{\rm sc} - \mathbf{k}_{\rm in}).\mathbf{r}$

$$\frac{d\sigma}{d\Omega} = \left| \sum_j b_j e^{i\mathbf{q}.\mathbf{r}_j} \right|^2 = \sum_j \sum_k b_j b_k e^{i\mathbf{q}.(\mathbf{r_j} - \mathbf{r_k})} = b^2 \left| \int d\mathbf{r} \rho(\mathbf{r}) e^{i\mathbf{q}.\mathbf{r}} \right|^2$$

ρ: density of scatterers

b_j: scattering lengths

b: scattering length density

For x-rays: $b = r_e = (e^2/4\pi\epsilon_0 m_e c^2) = 2.810^{-15} m$, classical radius of the electron
For neutrons, b $\sim$ fm (10^{-15}m) depends on atom, isotope, can be > 0 or < 0 $\rightarrow$ isotopic substitution (D_2O, H_2O)

Thomson Scattering Length (X-rays)

Low atomic numbers, $\omega \gg$ atomic frequencies $\rightarrow$ free e^-

$$m_e d\mathbf{v}/dt = -e\mathbf{E}e^{i\omega t}$$

$\mathbf{v} = i\omega \mathbf{x}$
For a $e^{i\omega t}$ time dependence of the electric field
$\mathbf{v} = (ie/m_e\omega)\mathbf{E}e^{i\omega t}$
$\rightarrow$ oscillating dipole $\mathbf{p} = -e\mathbf{x} = -\frac{e^2}{m_e\omega^2}\mathbf{E}e^{i\omega t}$

$$\mathbf{E}_{sc} = \frac{-p\omega^2 e^{-ik_0 r}}{4\pi\epsilon_0 c^2 r}\sin\theta\ \widehat{\mathbf{e}}_{\mathrm{sc}}$$

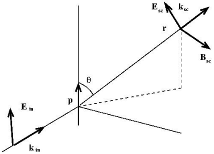
Intensity scattered in a unit solid angle $r^2|E|^2$.

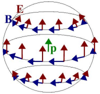
$$\boxed{b = \frac{e^2}{4\pi\epsilon_0 mc^2}(\widehat{\mathbf{e}}_{\mathrm{in}}.\widehat{\mathbf{e}}_{\mathrm{sc}})}$$

$r_e = e^2/4\pi\epsilon_0 mc^2 = 2.818 \times 10^{-15} m$ classical electron radius (Thomson radius).

Note : with $\omega = 2\pi c/\lambda$, $p = -(\epsilon_0\lambda^2 r_e/\pi)E$.

Free electrons





$m_{p^+} \approx 1836 \times m_{e^-}$

Fermi pseudo-potential (Strong interaction)

$$V(r) = (2\pi\hbar^2/m)b\delta(r)$$

b depends on isotope and spin → isotopic substitution

$$b = b_c + \frac{1}{2} b_N \mathbf{I}.\sigma,$$

describes the interaction between the neutron spin and the nuclear magnetic moment. Eigenvalues of $\mathbf{I}.\sigma$ are I for $J = I + 1/2$ and $-(I+1)$ for $J = I - 1/2$. With b^+ and b^- the corresponding scattering lengths,

$$\begin{cases} b^+ = b_0 + \frac{1}{2} b_n I \\ b^- = b_0 - \frac{1}{2} b_n (I+1) \end{cases}$$

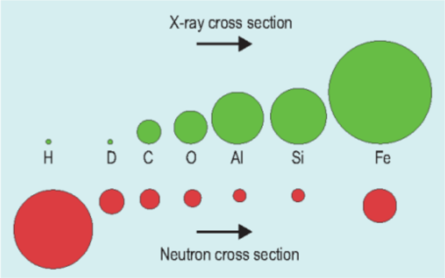


Fig. 2. Neutron and x-ray scattering cross-sections compared. Note that neutrons penetrate through Al much better than x rays do, yet are strongly scattered by hydrogen.

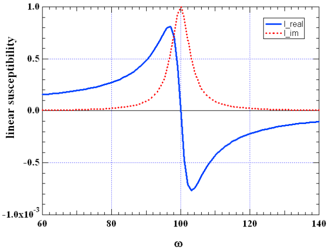
NIST

Anomalous scattering of x-rays

Classical description in an harmonic potential including damping:
$md^2\mathbf{x}/dt^2 = -m\omega_0^2\mathbf{x} - 2m\gamma d\mathbf{x}/dt - e\mathbf{E}. \rightarrow x(t) = -\frac{e}{m}\frac{Ee^{-i\omega t}}{\omega_0^2 - \omega^2 - 2i\gamma\omega}.$
Following the same procedure as before:

$$\boxed{b = \frac{e^2}{4\pi\epsilon_0 mc^2}\frac{\omega^2}{\omega_0^2 - \omega^2 - 2i\gamma\omega}(\widehat{\mathbf{e}}_{\rm in}.\widehat{\mathbf{e}}_{\rm sc}),} \quad (1)$$

Similar to the full quantum calculation.



- FWHM $= 2\gamma$
- lifetime $1/\gamma$ ($\Delta E.\Delta\tau \simeq \hbar$)
- For a typical natural width $\approx 1eV \rightarrow$ fs (10^{-15}s)

Example: Ion Solvation via Neutron Scattering

With different species in a solution:

$$\begin{aligned}\frac{d\sigma}{d\Omega} &= \Sigma_\alpha \Sigma_\beta b_\alpha b_\beta \langle \Sigma_{\mathbf{r}_{i(\alpha)}} \Sigma_{\mathbf{r}_{j(\beta)}} \exp i\mathbf{k}(\mathbf{r}_{j(\beta)} - \mathbf{r}_{i(\alpha)}) \rangle \\ &= N\left[\Sigma_\alpha c_\alpha b_\alpha^2 + \Sigma_\alpha \Sigma_\beta c_\alpha c_\beta b_\alpha b_\beta (S_{\alpha\beta}(k) - 1)\right],\end{aligned}$$

with α and β different chemical species of concentration c_α and c_β and the first sum runs over their positions.

$S_{\alpha\beta}$ is the partial strcuture factor of α and β. It is related to the partial distribution function $g_{\alpha\beta}(r)$ via Fourier transform.

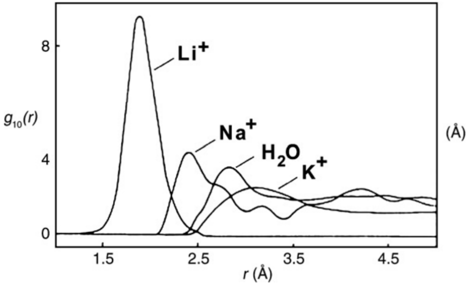
$$g_{\alpha\beta}(r) = 1 + \frac{V}{2\pi^2 N r}\int dk (S_{\alpha\beta} - 1) k \sin(kr),$$

with $4\pi\rho_\beta g_{\alpha\beta}(r) r^2 dr$ being the probability of finding a β particle in a spherical shell of radius r and thickness dr, knowing that there is an α particle at origin.

→ **The full collection of $S_{\alpha\beta}(k)$ contains in principle all information about the structure of the solution.**

Ion Solvation via Neutron Scattering (II)

The way to separate out the different $S_{\alpha\beta}(k)$ is to use isotopic substitution.

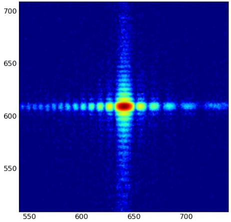


N.T. Skipper and G.W. Neilson, J. Phys. Cond. Matt. **1** 4141-4154 (1989).

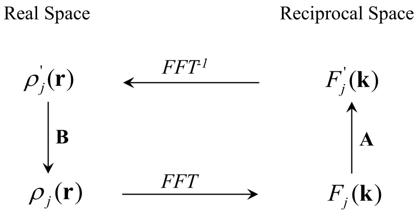
Weaker binding when surface charge decreases.

$$\frac{d\sigma}{d\Omega} = \sum_j \sum_k b_j b_k e^{i\mathbf{q}.(\mathbf{r_j}-\mathbf{r_k})} \rightarrow \text{Absolute phase is lost.}$$

It can be recovered provided using appropriate constraints and iterative reconstruction algorithms provided the sample is coherently illuminated.

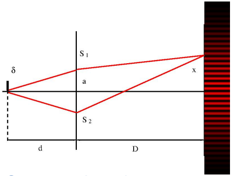


Soleil, Cristal beamline



J. Fienup *Op; Lett.* **3** 27 (1978)

Young's slits: path difference ax/D.
Fringes become invisible if $a \approx \lambda d/\delta$
$\rightarrow$ transverse coherence length.
$\lambda = 1\mathring{A}$, $\delta = 10\mu$m, d = 5m $\rightarrow 25\mu m$.



No correlation between an atom's position and its isotope and/or spin state.
Random distribution of isotopes and spin states. $b_i = \langle b \rangle + \delta b_i$, where $\langle \delta b \rangle = 0$.

$$\frac{d\sigma}{d\Omega} = \left| \sum_j b_j e^{i\mathbf{q}.\mathbf{r}_j} \right|^2 = \sum_i \sum_j (\langle b \rangle + \delta b_i)(\langle b \rangle + \delta b_j) e^{i\mathbf{q}.(\mathbf{r}_i - \mathbf{r}_j)}$$

$$= \langle b \rangle^2 \left| \sum_j e^{i\mathbf{q}.\mathbf{r}_j} \right|^2 + N(\langle b^2 \rangle - \langle b \rangle^2)$$

$\langle \delta b_i \rangle = \langle \delta b_j \rangle \langle \delta b_i \rangle_{i \neq j} = 0$
$\langle \delta b_i \delta b_i \rangle = \langle b^2 \rangle - \langle b \rangle^2$ as $b = \langle b \rangle + \delta b_i$.

The coherent scattering length is the average and the incoherent scattering length the variance.
Strong incoherent scattering with H.

Magnetic scattering (of neutrons)

Dipolar interaction of the neutron spin with the magnetic field created by the unpaired electrons of the magnetic atoms. This field contains two terms, the spin part and the orbital part.

Class	Interaction	δb (fm)
I	Strong interaction	10.0
	Atomic magnetic dipole moment*	10.0
II	Spin–orbit (Schwinger)	0.1
	Foldy	0.1
	Neutron electric polarizability	0.05
	Intrinsic electrostatic	0.01
	Nuclear magnetic dipole moment*	0.005
III	Neutron electric dipole moment*	$\lesssim 10^{-8}$
	Neutron electric charge*	$\lesssim 10^{-10}$
	Weak interaction	$\sim 10^{-34}$

$$V(\boldsymbol{r}) = -\boldsymbol{\mu}_n.\boldsymbol{B} = -g_n \mu_N \boldsymbol{\sigma}.\boldsymbol{B}$$

$\boldsymbol{\mu}$ magnetic moment of the electron; $\mu_N = e\hbar/(2m_p)$ nuclear magnetic moment; $g_n = 3,8260855$ Landé factor ; $\boldsymbol{B}$ magnetic field produced by the atom.

$$f = \frac{2m}{\hbar}\boldsymbol{\mu}.\boldsymbol{M}_\perp,$$

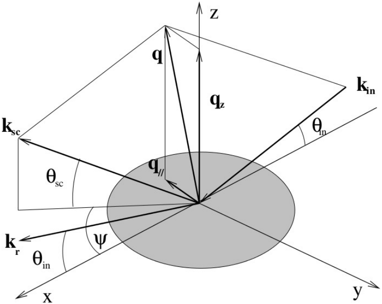
$\boldsymbol{M}_\perp$ transverse part of the atomic magnetization (projection of $\boldsymbol{M}$ in the plane $\perp$ to $\boldsymbol{q}$.

Note: e^- also has a 1/2 spin which can interact with the magnetic part of electromagnetic waves. One has $b_{mag} = -ir_e\left(\hbar\omega/m_{e^-}c^2\right)\left[\left(\widehat{\mathbf{e}}^*_{\rm sc}.\overline{\overline{\mathbf{T}}}_S.\widehat{\mathbf{e}}_{\rm in}\right).\mathbf{S} + \left(\widehat{\mathbf{e}}^*_{\rm sc}.\overline{\overline{\mathbf{T}}}_L.\widehat{\mathbf{e}}_{\rm in}\right).\mathbf{L}\right]$, where tensors $\overline{\overline{\mathbf{T}}}_S$, $\overline{\overline{\mathbf{T}}}_L$ depend on scattering geometry. For 10keV photons $\hbar\omega/m_{e^-}c^2 = 10/511 \approx 0.02$ As only unpaired electrons contribute, magnetic scattering is $\approx 10^7$ of Thomson scattering.

$$\frac{d\sigma}{d\Omega} = b^2\rho_{\rm sub}^2 \int dzdz' \int d\mathbf{r}d\mathbf{r}' e^{i\mathbf{q}_{\parallel}\cdot(\mathbf{r}_{\parallel}-\mathbf{r}'_{\parallel})} e^{iq_z z} e^{iq_z z'}$$

$$\frac{d\sigma}{d\Omega} = \frac{4\pi^2 A b^2 \rho_{\rm sub}^2 \delta(\mathbf{q}_{\parallel})}{q_z^2}$$

with $\int d\mathbf{r}_{\parallel} e^{i\mathbf{q}_{\parallel}\mathbf{r}_{\parallel}} = 4\pi^2\delta(\mathbf{q}_{\parallel})$,
$\mathbf{q}_{\parallel}$ wave-vector transfer component in the surface plane, A illuminated area



Integrating over $\delta\Omega = d\theta_{\rm sc}d\psi = (2/k_0q_z)d\mathbf{q}_{\parallel}$, and normalizing to the incident flux ($I_0/A\sin\theta_{in}$)

$\rightarrow$ reflection coefficient:

$$R = \frac{I}{I_0} = \frac{16\pi^2 b^2\rho_{\rm sub}^2}{q_z^4} = \frac{q_c^4}{16q_z^4}$$

Brewster angle 45°

Averaging over all possible orientations $\rightarrow 1/Q^4$ Porod's law of Small Angle Scattering.

X-rays → Maxwell Equations:

$$\begin{aligned}\nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial \mathbf{t}}\\ \nabla \times \mathbf{B} &= \mu_0 \mathbf{j} + \mu_0 \frac{\partial \mathbf{D}}{\partial \mathbf{t}},\\ \mathbf{D} &= \epsilon_0 n^2 \mathbf{E} = \epsilon_0 \mathbf{E} + \mathbf{P}\end{aligned}$$

Polarization of the medium $\mathbf{P} = \rho_{el} \mathbf{p}$

$$\boxed{n = 1 - \frac{\lambda^2 r_e}{2\pi} \rho_{el} \approx 1 - 10^{-6}}$$

Neutrons → Schrödinger equation:

$$\left(-\frac{\hbar^2}{2m} \nabla^2 + \frac{2\pi\hbar^2}{m} \sum_i \rho_i b_i \right) \psi(r) = \mathcal{E} \psi(r)$$

b_i scattering length density of nucleus i of density ρ_i.

$$\boxed{n = 1 - \frac{\lambda^2}{2\pi} \sum_i \rho_i b_i}$$

→ Optical description

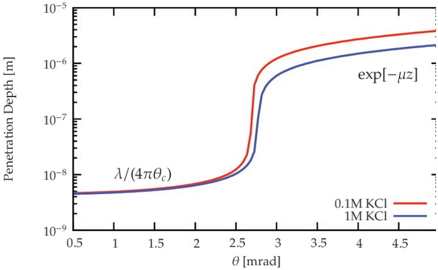
Total External Reflection

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

$\cos\theta_1 = n\cos\theta_2$ **grazing angles of incidence!**

$\theta_2 = 0$ for $\theta_{\rm in} \le \theta_c = \sqrt{2\delta} \approx 10^{-3}$

Total external reflection.



Wave $\exp i(\omega t - k_z z)$; $k_z = n\sin\theta$.

Penetration depth $1/(2\mathcal{I}mk_z)$ with

$$\mathcal{I}m(k_z) = \frac{1}{\sqrt{2}}\, k_0 \sqrt{[(\theta^2 - 2\delta_i)^2 + 4\beta^2]^{1/2} - (\theta^2 - 2\delta)}.$$

Inelastic Scattering Spectroscopy

Exchange of energy between x-rays or neutrons and matter
X-rays:

- Compton scattering.
- Absorption ($\rightarrow$ (N)EXAFS).
- Photoelectric effect. Photoemission.
- Fluorescence.
- Anomalous scattering / Resonant scattering.
- Inelastic scattering / Raman scattering.

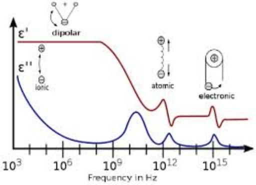
Neutrons:

- Inelastic scattering of neutrons.

Orders of Magnitude

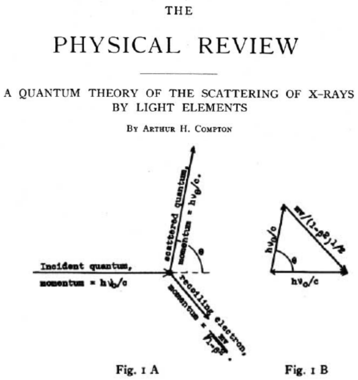
	Energy	Wavelength	Frequency	
Molecular rotations	1-50 meV	25μm - 1mm (400-10 cm^{-1})	0.1 - 10 THz	Far IR
Molecular vibrations	50-500 meV	1-30 μm (4000-100 cm^{-1}	10 - 100 THz	IR, Raman
Phonons	10-100 meV	0.01 - 0.1 μm	10 - 100 THz	IXS,INS
Magnetic excitations (magnons)	100meV-1eV	0.1-1. μm	10^{14} Hz	RIXS
Chemical shifts	$\sim$1eV			Photoemission
Band gap	1-10 eV	10 - 100nm		ARPES, RIXS
Electronic molecular transitions	10 -100 eV	$\sim$ 100 nm	10^{16}-10^{17} Hz	UV-vis
Atomic core levels	1-100 keV	0.1 $\AA$ - 1nm	10^{17}-10^{19} Hz	X-rays (soft, hard)

INS: $E \sim 25meV$; $\delta E/E \sim 10^{-1} - 10^{-2}$; IXS: $E \sim 10keV$; $\delta E/E \sim 10^{-7} - 10^{-8}$



- THz, IR : dipole fluctuations, bond vibrations, hydrogen bonds.
- UV: electronic transitions.
- Hard x-rays: scattering by almost free electrons.

Electron-photon collision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PHYSICAL REVIEW

A QUANTUM THEORY OF THE SCATTERING OF X-RAYS BY LIGHT ELEMENTS

By Arthur H. Compton

Fig. 1 A Fig. 1 B

Conservation of energy and momentum:

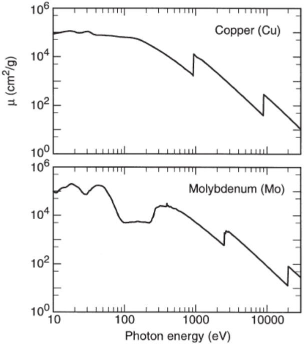
$$\mathbf{p_1} = \mathbf{p_2} + \mathbf{p_e}$$
$$p_e^2 = p_1^2 + p_2^2 - 2p_1.p_2\cos\theta$$
$$p_1 c + m_e c^2 = p_2 c + \sqrt{m_e^2 c^4 + p_e^2 c^2}$$

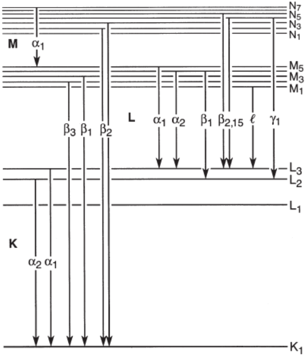
$$\boxed{\Delta\lambda = \frac{h}{m_e c}(1 - \cos\theta)}$$

→ Electronic properties in condensed matter

→ Imaging

Absorption of a photon by an atom

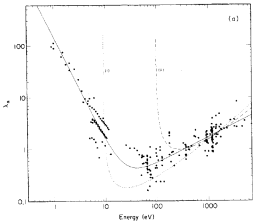




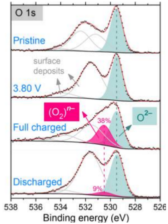
- Energy transfered to an e^- which is excited to an empty upper state.
- Absorption varies as E^{-3} and Z^4.
- Leaves the atom in an excited state.
- Most frequently, the e^- will be expelled from the atom
  → Photoelectric effect
  → Fluorescence or non-radiative de-excitation (Auger).

Photoelectric effect, Einstein 1905

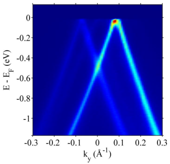
- Photon of energy hν transfers energy and momentum to the system (atom, solid).
- For core levels of atoms, "chemical shifts" $\rightarrow$ chemical environment. KE = hν -ΔE - eϕ, "work function".
- In solids, energy and momentum transfered to electrons. Conservation of $\|$ component of momentum $\rightarrow$ band structure if both kinetic energy and angle are measured (Angle Resolved Photoemission Spectroscopy, ARPES).
- Photoelectrons lose energy in matter (collisions with the electrons of the other atoms $\rightarrow$ secondary electrons are produced. The probability of not suffering an inelastic collision after travelling a distance x in matter (solid, gases) is $\exp(-x/\lambda)$ where λ is the inelastic mean free path $\rightarrow$ Varying the photon energy allows depth profiling.



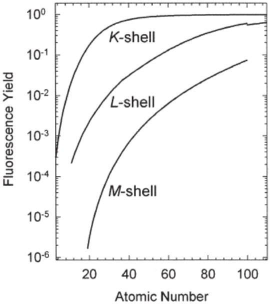
Inelastic Mean Free Path (IMFP).
M. P. Seah and W. A. Dench, Surface and Interface Analysis, VOL. **1**, 2 (1979).

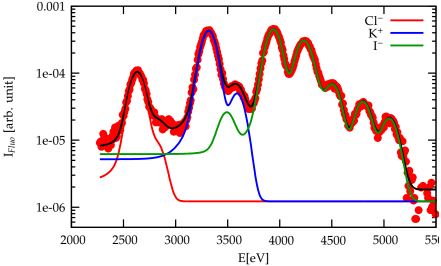


G. Assat et al. ACS Energy Lett. 3, 2721 (2018)



M. Sprinkle et al., Phys. Rev. Lett. 103 226803 (2009)





Fluorescence is characteristic of the atom

Lifetime of excited states $\sim$ fs.

In 1fs, the light travels $10^{-15} \times 3.10^{8} = 300nm \gg \lambda \rightarrow$ incoherent process.

Transition probability from an initial state $|i\rangle$ to a final state $|f\rangle$ per unit time (second order):

$$w = \frac{2\pi}{\hbar} \left| \langle f|\mathcal{H}_{int}|i\rangle + \sum_n \frac{\langle f|\mathcal{H}_{int}|n\rangle\langle n|\mathcal{H}_{int}|i\rangle}{E_i - E_n} \right|^2 \delta(E_f - E_i)$$

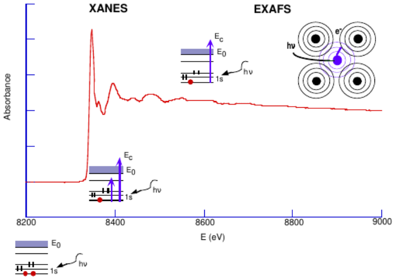
$|i\rangle = |a, \boldsymbol{k}\lambda\rangle$, $|f\rangle = |b, \mathbf{k'}\lambda'\rangle$, $E_i = E_a + \hbar\omega_k\rangle$, $E_i = E_a + \hbar\omega_k\rangle$
$\mathcal{H}_{int}$ interaction Hamiltonian, $\langle f|W|i\rangle$ Transition matrix element.
First term: Thomson scattering, non-resonant magnetic scattering.
Second term: Anomalous scattering, resonant magnetic scattering.

$$\mathcal{H}_{int} = \sum_{i=1}^{N} \frac{e}{m} \boldsymbol{A}(\boldsymbol{r}_i).\boldsymbol{p} + \frac{e^2}{2m} \boldsymbol{A}^2(\boldsymbol{r}_i) + ...$$

$\boldsymbol{A}$ vector potential.

$$\boldsymbol{A} \propto a \exp(i\mathbf{k}.\mathbf{r}) + a^\dagger \exp(-i\mathbf{k}.\mathbf{r})$$

Creation and annihilation of photons.



$$\frac{d^2\sigma}{d\Omega dE} = w\rho(E_f)/I_0$$

$\rho(E_f)$ density of final states.

$CsNi[Cr(CN)_6]$, Ni K edge
V. Briois et al., Actualité Chimique **3**, 31 (2000)

Empty states

- Pre-edge: first empty levels.

NEXAFS: Near Edge X-ray Absorption Fine Structure ($\equiv$ XANES: X-ray Absorption Near Edge Structure)

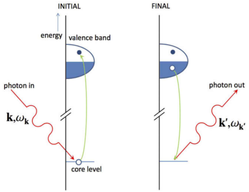
- Local environment and electronic structure

EXAFS: Extended X-Ray Absorption Fine Structure ($h\nu - E \gtrsim 50eV$)

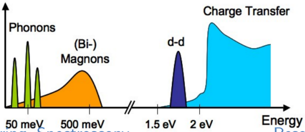
- The wave corresponding to the photoelectron is diffracted by neighboring atoms $\rightarrow$ local structure.
- $k = \sqrt{2m/\hbar^2 \times (E - E_0)}$
- $\chi(k) = \sum_j \frac{N_j f_j(k) \exp(-2k^2\sigma_j^2)}{kR_j^2} \sin(2kR_j + \delta_j(k))$

$$w = \frac{2\pi}{\hbar} \left| \langle f|\mathcal{H}_{int}|i\rangle + \sum_n \frac{\langle f|\mathcal{H}_{int}|n\rangle\langle n|\mathcal{H}_{int}|i\rangle}{E_i - E_n} \right|^2 \delta(E_f - E_i)$$

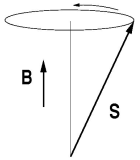
- Resonant process if E_n energy level of the system.
- Conservation of energy and momentum
  → excitations can be investigated.
- X-rays with p = E/c carry much more momentum than visible photons or neutrons with $p = \sqrt{2mE}$
  → wide range of momentum transfer possible.
- Element and orbital specific.
- Bulk sensitive.
- Good energy resolution as it is not affected by the core level short lifetime.



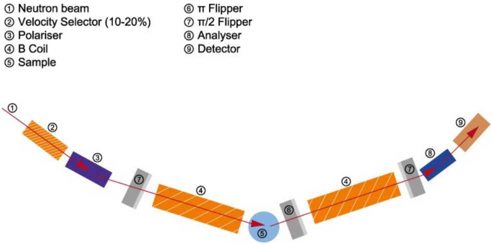
E. Pavarini, E. Koch, J. van den Brink, and G. Sawatzky (eds.) Quantum Materials: Experiments and Theory Modeling and Simulation Vol. 6, Forschungszentrum Jülich, 2016



Energy of neutrons can be measured by analyzer crystals in "Triple axis" instruments as x-rays or time-of-flight and spin-echo.



www.nmi3.eu



- Polarized neutrons. Spins precess around magnetic fields with the Larmor frequency: $\frac{d\mathbf{s}}{dt} = \gamma \mathbf{s} \times \mathbf{B}$.
- $\omega_L = \gamma B = (g_n \mu_N / \hbar) B = -3.826 (e/2m_p) B = 183.3 \times 10^6 B(T)$.
- For elastic scattering, precession after scattering cancels precession before scattering.
- If the neutron changes energy, the precession phases will be different.
- $\phi = \omega_L t = \gamma B d/v$; $\delta\phi = \omega_L t \approx \gamma B d \delta v / v^2 \approx \gamma B d \hbar\omega / (m v^3)$.
- 100s of ns $\rightarrow$ investigation of slow dynamics, e.g. in soft matter. Equivalently, highest energy resolution (neV).

Thank you!

Questions? jean.daillant@synchrotron-soleil.fr