Neutron and x-ray spectroscopy

B. Keimer

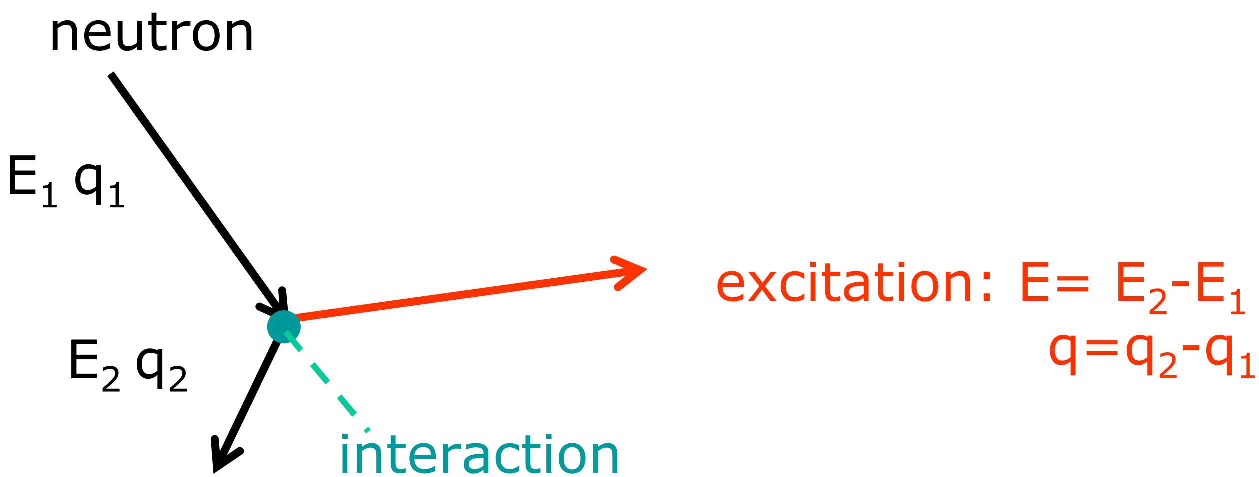
Max-Planck-Institute for Solid State Research

outline

1. self-contained introduction
   - neutron scattering and spectroscopy
   - x-ray scattering and spectroscopy
2. application to correlated-electron materials
   - bulk
   - interfaces

Max-Planck-Institut für Festkörperforschung

Neutron scattering



strong (nuclear) interaction

elastic	lattice structure
inelastic	lattice dynamics

magnetic (dipole-dipole) interaction

elastic	magnetic structure
inelastic	magnetic excitations

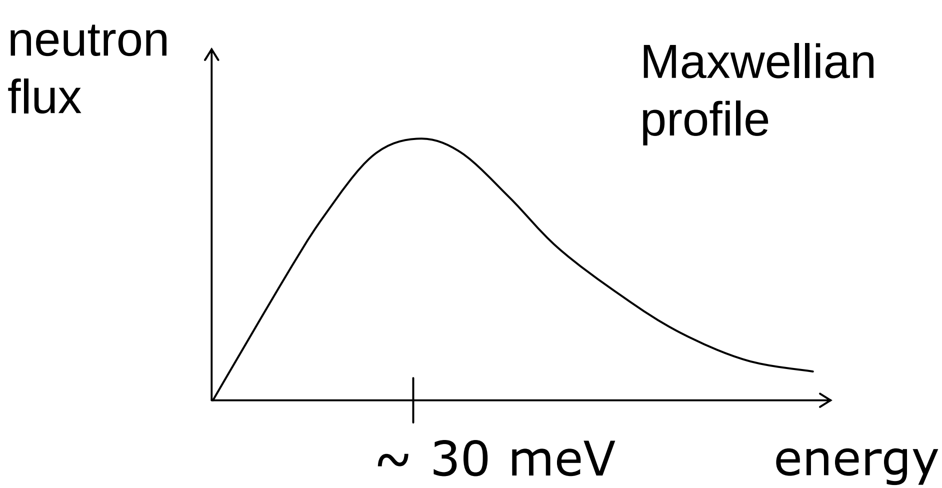
Neutron sources

example research reactor

$$^{235}U + n \rightarrow A + B + 2.3n$$

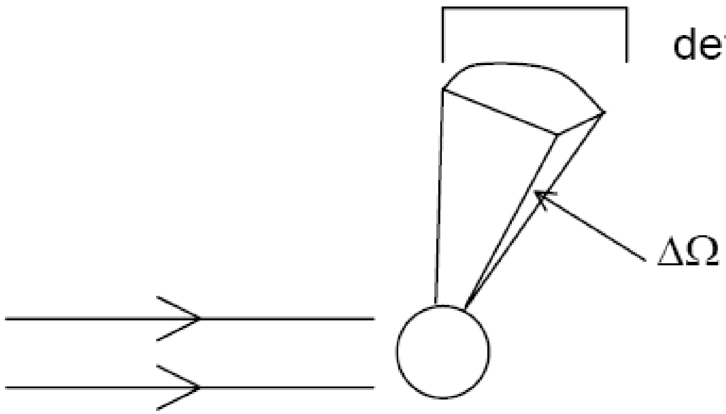
FRM-II
Garching, Germany

spectrum



Elastic neutron scattering

basic quantity: differential cross-section $\frac{d\sigma}{d\Omega}$



incident neutron beam, flux $\phi = \dfrac{\text{\# of neutrons}}{\text{area} \cdot \text{time}}$

$\frac{d\sigma}{d\Omega} =$ # of neutrons scattered into solid angle element $d\Omega$ per unit time,

normalized to incident flux.

dimensions: $\left[\dfrac{d\sigma}{d\Omega}\right] = \dfrac{1}{[\Delta\Omega]\,[t]\,[\phi]} =$ area

dimensionless

Elastic neutron scattering

calculation of $\frac{d\sigma}{d\Omega}$ through Fermi's Golden Rule:

transition rate (# of transitions per unit time): $W = \frac{2\pi}{\hbar} \left| \left\langle \vec{k}_f \middle| V \middle| \vec{k}_i \right\rangle \right|^2 \underbrace{\rho_f(E)}_{\text{Density of final states}}$

$$\left.\begin{aligned} |k_i\rangle &= \frac{1}{\sqrt{L^3}} e^{i\vec{k}_i \cdot \vec{r}} \\ |k_f\rangle &= \frac{1}{\sqrt{L^3}} e^{i\vec{k}_f \cdot \vec{r}} \end{aligned}\right\} \text{ plane waves}$$

incident neutron flux: $\frac{\text{velocity}}{L^3} = \frac{\hbar k_i}{m_n L^3}$

$k_i = k_f$ for elastic scattering

$$\rho_f(E) = \underbrace{\left(\frac{L}{2\pi}\right)^3}_{\text{density of states in } k\text{-space}} \frac{d\vec{k}_f}{dE}$$

$$d\vec{k}_f = k_f^{\,2} dk_f d\Omega$$

$$\rho_f(E) = \left(\frac{L}{2\pi}\right)^3 k_f^{\,2} \frac{dk_f}{dE} d\Omega = \left(\frac{L}{2\pi}\right)^3 \frac{m_n k_f}{\hbar^2} d\Omega \qquad \text{with } \frac{dE}{dk_f} = \frac{\hbar^2 k_f}{m_n}$$

$$\Rightarrow \frac{d\sigma}{d\Omega} = \frac{W}{\text{incident flux}} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left| \int V e^{i\left(\vec{k}_i - \vec{k}_f\right)\cdot\vec{r}} d\vec{r} \right|^2$$

$$= \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left| \int V(\vec{r}) e^{-i\vec{Q}\cdot\vec{r}} d\vec{r} \right|^2$$

"Born approximation"

Elastic nuclear neutron scattering

For short range strong force, use approximate interaction potential

$$V(\vec{r}) = \frac{2\pi\hbar^2}{m_n} b\, \delta(\vec{r} - \vec{R})$$

position of nucleus

"scattering length"

scattering length b ~ size of nucleus ~ 10^{-15} m

for single nucleus: $\frac{d\sigma}{d\Omega} = |b|^2$

total cross section: $\sigma = \int \frac{d\sigma}{d\Omega} d\Omega = 4\pi b^2$

lattice of nuclei: $V(\vec{r}) = \frac{2\pi\hbar^2}{m_n} \sum_{\vec{R}} b_{\vec{R}}\, \delta(\vec{r} - \vec{R})$

$b_{\vec{R}}$: scattering length of nucleus at lattice site $\vec{R}$

$$\frac{d\sigma}{d\Omega} = \left| \int d\vec{r} \sum_{R} b_R\, \delta(\vec{r} - \vec{R}) e^{i\vec{Q}\cdot\vec{r}} \right|^2 = \left| \sum_{R} b_R\, e^{i\vec{Q}\cdot\vec{R}} \right|^2 = b^2 \frac{N(2\pi)^3}{v_o} \sum_{\vec{K}} \delta(\vec{Q} - \vec{K})$$

Bragg peaks
at reciprocal lattice vectors K

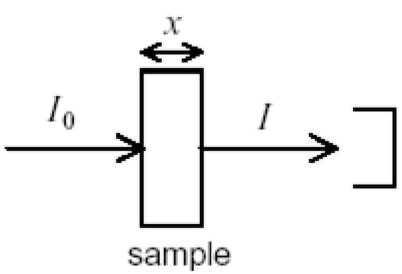
for unit cell with several atoms, basis vector $\vec{d}$

$$\frac{d\sigma}{d\Omega} = N \frac{(2\pi)^3}{v_0} \sum_{\vec{K}} \delta(\vec{Q} - \vec{K}) \left| F_N(\vec{K}) \right|^2$$

$$F_N(\vec{K}) = \sum_{\vec{d}} e^{i\vec{Q}\cdot\vec{d}} b_{\vec{d}}$$

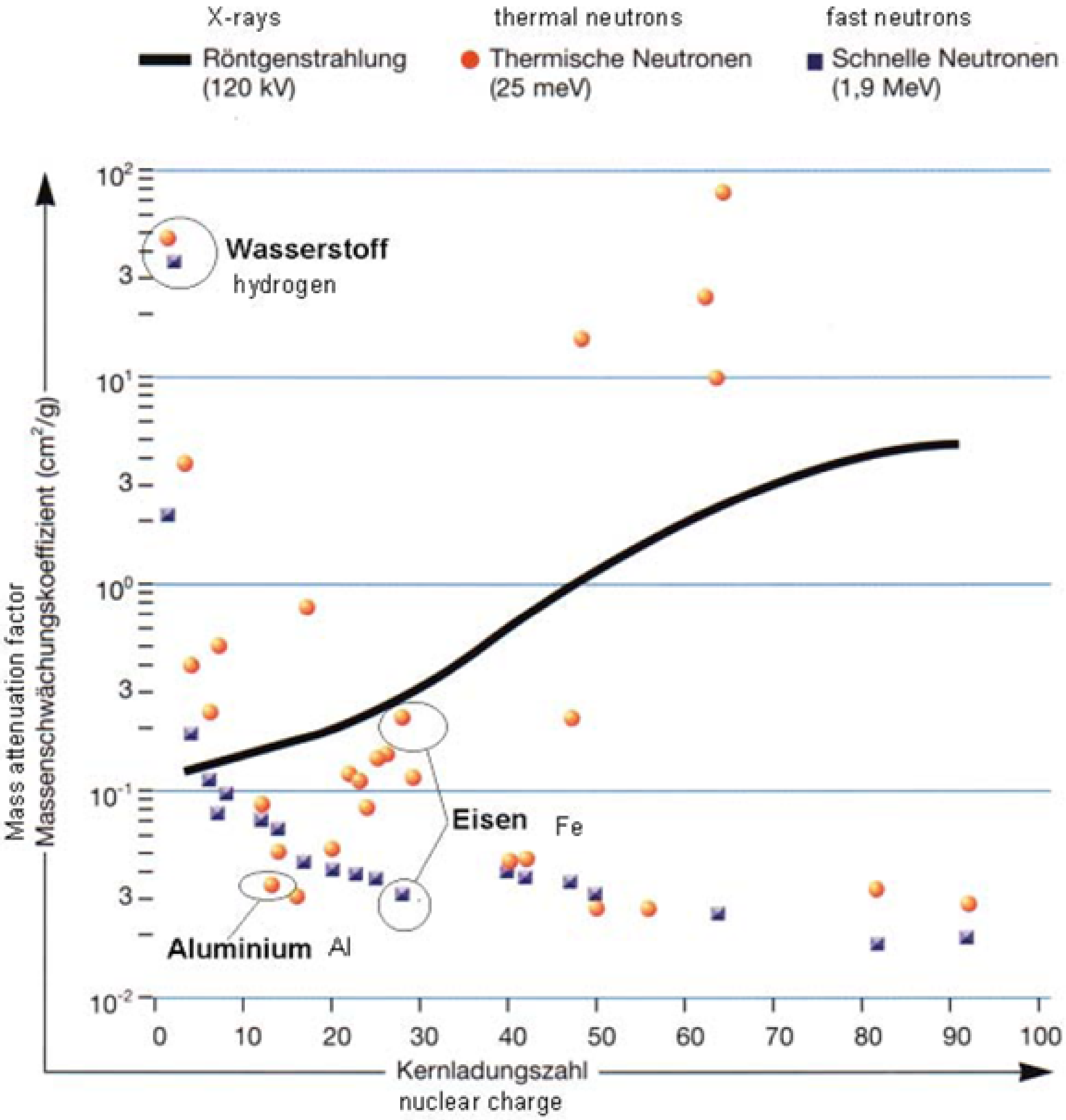
"nuclear structure factor"

Neutron radiography

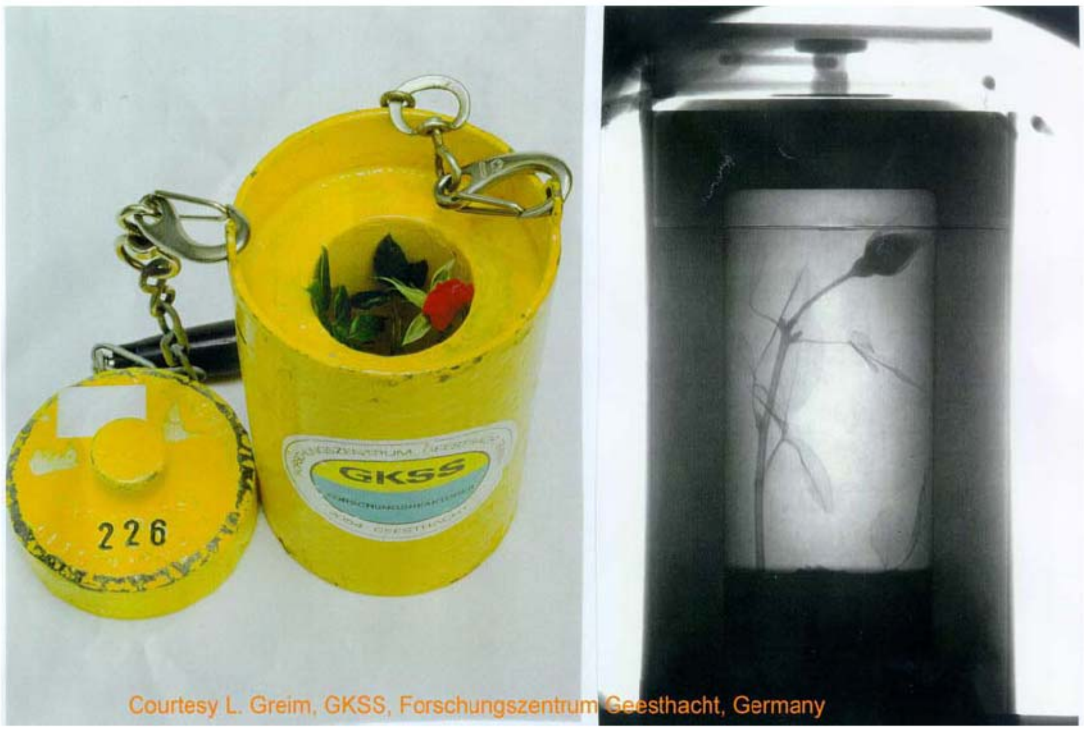


$$\frac{I}{I_0} = e^{-(\mu/\rho_a)x}$$

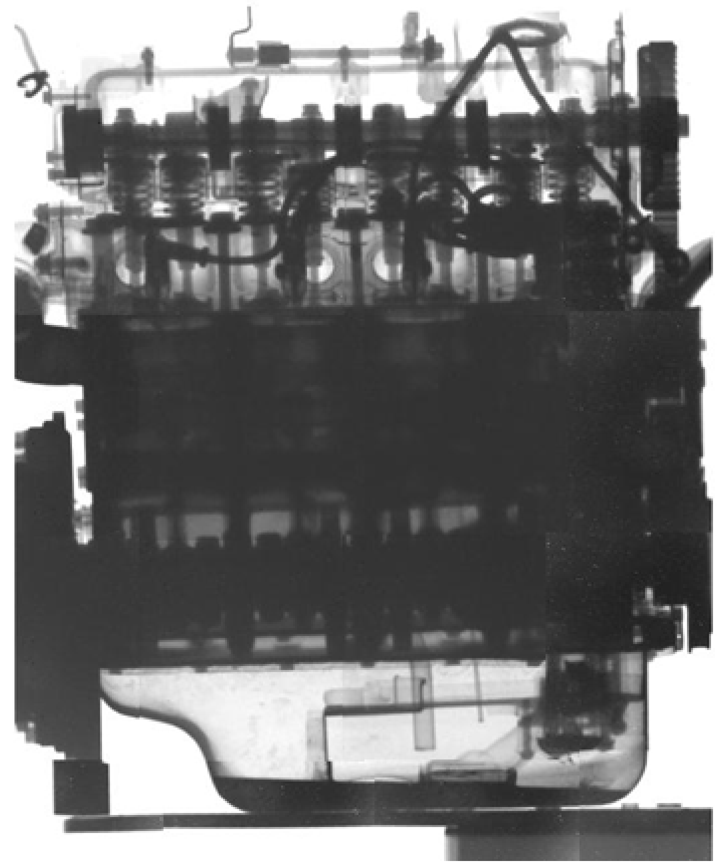
mass attenuation coefficient



Neutron radiography

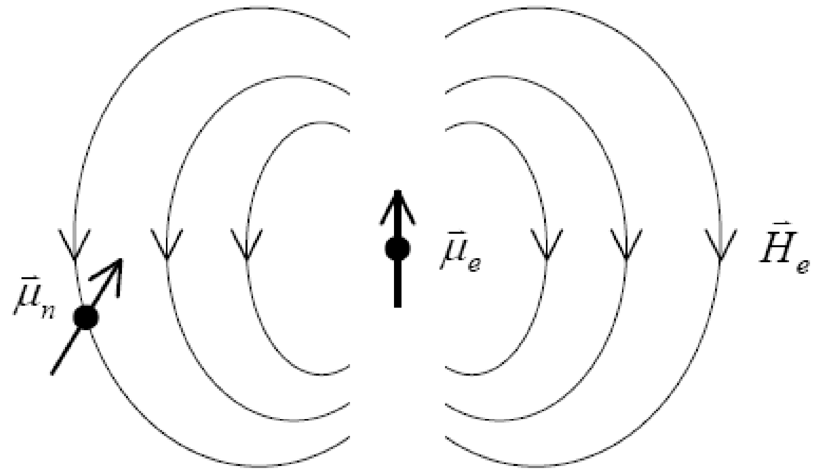


Courtesy L. Greim, GKSS, Forschungszentrum Geesthacht, Germany



Max-Planck-Institut
für Festkörperforschung

Elastic magnetic neutron scattering



$$\vec{\mu}_e = -2\mu_B \vec{s}_e \text{ with } \mu_B = \frac{e\hbar}{2m_e}$$

$$\vec{\mu}_n = -g_n \mu_N \vec{s}_n \equiv -\gamma \mu_N \vec{\sigma} \text{ with } \mu_N = \frac{e\hbar}{2m_n} \text{ and } \gamma = \frac{g_n}{2} = 1.913$$

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left|\left\langle \vec{k}_f m_f \middle| H_{\text{int}} \middle| \vec{k}_i m_i \right\rangle\right|^2 \text{ with } H_{\text{int}} = -\vec{\mu}_n \cdot \vec{H}_e$$

Elastic magnetic neutron scattering

$$\vec{A}_e = \frac{\mu_0}{4\pi}\frac{\vec{\mu}_e \times \vec{r}}{|\vec{r}^{\,3}|} = \frac{\mu_0}{4\pi}\vec{\mu}_e \times \vec{\nabla}\frac{1}{|\vec{r}|}$$

$$\vec{H}_e = \vec{\nabla} \times \vec{A}_e = \frac{\mu_0}{4\pi}\vec{\nabla} \times \left(\vec{\mu}_e \times \nabla \frac{1}{|\vec{r}|}\right)$$

collect all prefactors:

$$\left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left(2\gamma\mu_N\mu_B\right)^2 \left(\frac{\mu_0}{4\pi}\right)^2 (4\pi)^2 = (\gamma r_0)^2$$

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left(2\gamma\mu_N\mu_B\right)^2 \left|\left\langle \vec{k}_f, m_f \left| \vec{\sigma}_n \cdot \vec{\nabla} \times \left(\vec{s}_e \times \nabla \frac{1}{|\vec{r}|}\right)\right| \vec{k}_i, m_i \right\rangle\right|^2$$

$$\int \frac{d\vec{p}}{|\vec{p}|^2} e^{i\vec{p}\cdot\vec{r}} = 2\pi\int_0^\infty d|\vec{p}| \int_{-1}^{1} e^{i|\vec{p}||\vec{r}|\cos\Theta} d(\cos\Theta) = 2\pi\int_0^\infty d|\vec{p}| \frac{\sin|\vec{p}|\,|\vec{r}|}{|\vec{p}|\,|\vec{r}|} = \frac{2\pi^2}{|\vec{r}|}$$

$\vec{p}$ auxiliary variable

$$\nabla \times \left(\vec{s}_e \times \nabla \frac{1}{|\vec{r}|}\right) = \frac{1}{2\pi^2}\int \frac{d\vec{p}}{|\vec{p}|^2} \vec{\nabla} \times \left(\vec{s}_e \times \vec{\nabla}\right) e^{i\vec{p}\cdot\vec{r}}$$

$$= \frac{1}{2\pi^2}\int \hat{p} \times \left(\vec{s}_e \times \hat{p}\right) e^{i\vec{p}\cdot\vec{r}} d\vec{p}$$

$$\left\langle \vec{k}_f \left| \nabla \times \left(s_e \times \nabla \frac{1}{|\vec{r}|}\right)\right| \vec{k}_i \right\rangle = \frac{1}{2\pi^2}\int d\vec{r} e^{-i\vec{Q}\cdot\vec{r}} \int d\vec{p}\;\; \hat{p} \times \left(\vec{s}_e \times \hat{p}\right) e^{i\vec{p}\cdot\vec{r}}$$

$$= 4\pi \underbrace{\hat{Q} \times \left(\vec{s}_e \times \hat{Q}\right)}_{\equiv \vec{s}_{e\perp}}$$

Elastic magnetic neutron scattering

one electron

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \left| \langle m_f | \vec{\sigma} \cdot \vec{s}_{e\perp} | m_i \rangle \right|^2$$

$r_0 (= 2.8 \times 10^{-5} \mathring{A}$ "classical electron radius"

$$\langle m_f | \vec{\sigma} \cdot \vec{s}_{e\perp} | m_i \rangle = s_{e\perp} \langle m_f | \sigma_z | m_i \rangle = \begin{cases} s_{e\perp} & \text{if } m_f = m_i \\ 0 & \text{otherwise} \end{cases}$$

non-spin-flip

$\sigma_z \rightarrow \sigma_x, \sigma_y$ spin-flip (not possible for nuclear scattering)

average for unpolarized beam

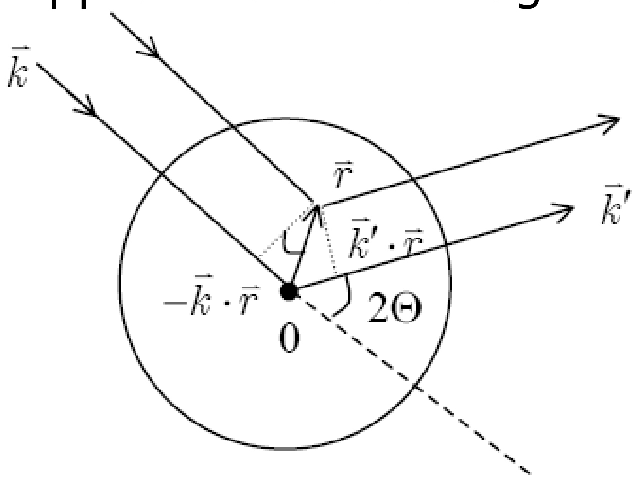
$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \left| \vec{s}_{e\perp} \right|^2$$

$\vec{s}_{e\perp} = 4\pi \hat{Q} \times (\vec{s}_e \times \hat{Q})$ projection of the electron spin perpendicular to $\vec{Q}$

Elastic magnetic neutron scattering

one atom

approximated as magnetized sphere, magnetization density *M(r)*



elastic scattering: $\left|\vec{k}\right| = \left|\vec{k}'\right|$

$\left|\vec{r}\right| \ll \left|\vec{R}\right|$

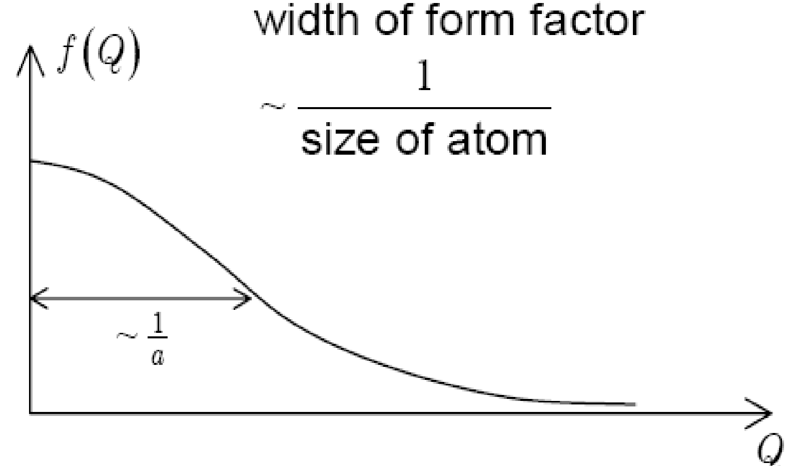
phase difference between wave scattered at 0 and at $\vec{r}$: $\left(\vec{k} - \vec{k}'\right) \cdot \vec{r} \equiv \vec{Q} \cdot r$

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \left[1 - \left(\hat{\eta} \cdot \hat{Q}\right)^2\right] \left|f\left(\vec{Q}\right)\right|^2$$

$$f\left(\vec{Q}\right) = \frac{1}{2\mu_B} \int \mathcal{M}\left(\vec{r}\right) e^{-i\vec{Q}\cdot\vec{r}}$$ magnetic form factor

$$\vec{\mathcal{M}}\left(\vec{r}\right) = \mathcal{M}\left(\vec{r}\right)\hat{\eta}$$ magnetic dipole moment density

width of form factor $\sim \dfrac{1}{\text{size of atom}}$



Elastic magnetic neutron scattering

generalization for collinear magnets

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 \left[1-\left(\hat{\eta}\cdot\hat{Q}\right)^2\right] \left|\sum_{\vec{R}} (\pm) f_{\vec{R}}\left(\vec{Q}\right) e^{i\vec{Q}\cdot\vec{R}}\right|^2$$

$$= (\gamma r_0)^2 \left[1-\left(\hat{\eta}\cdot\hat{Q}\right)^2\right] N \frac{(2\pi)^3}{V_0} \sum_{\vec{K}_M} \left|F_M\left(\vec{K}_M\right)\right|^2 \delta\left(\vec{Q}-\vec{K}_M\right)$$

Bragg peaks

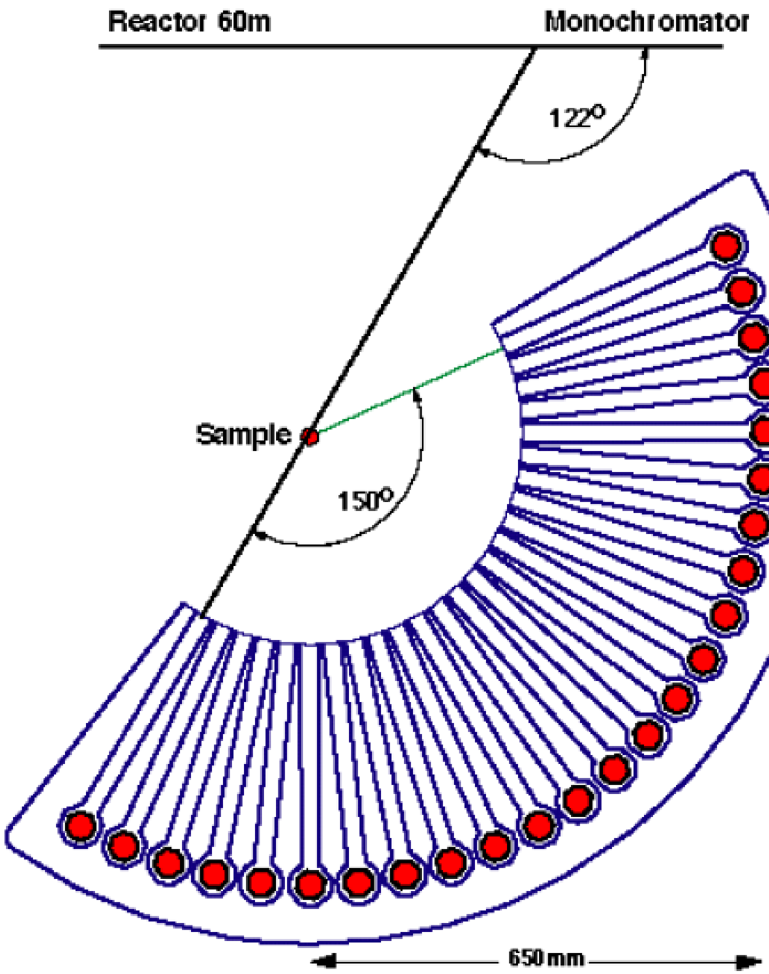
polarization factor

$$F_M\left(\vec{Q}\right) = \sum_{\vec{d}} (\pm) f_{\vec{d}}\left(\vec{Q}\right) e^{i\vec{Q}\cdot\vec{d}}$$ magnetic structure factor

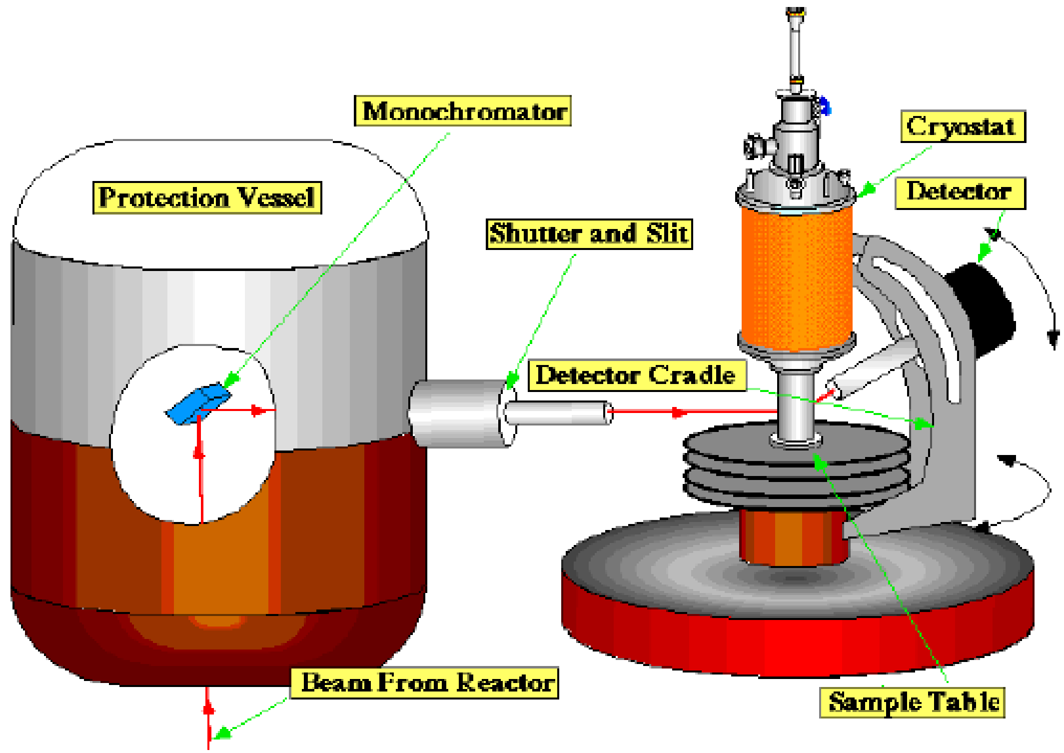
$\vec{K}_M$ magnetic reciprocal lattice vectors

Neutron diffractometer

powder

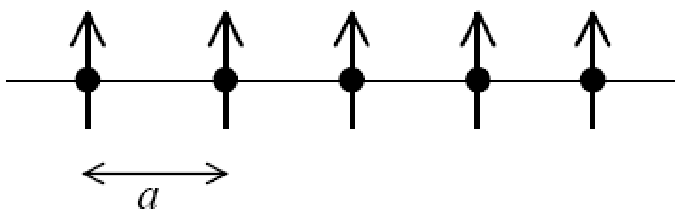


single crystal

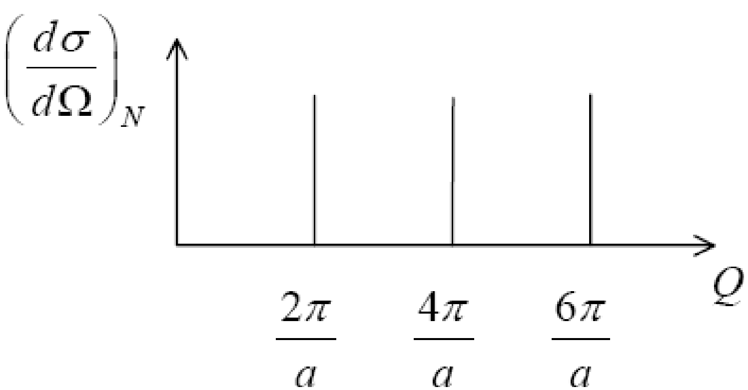


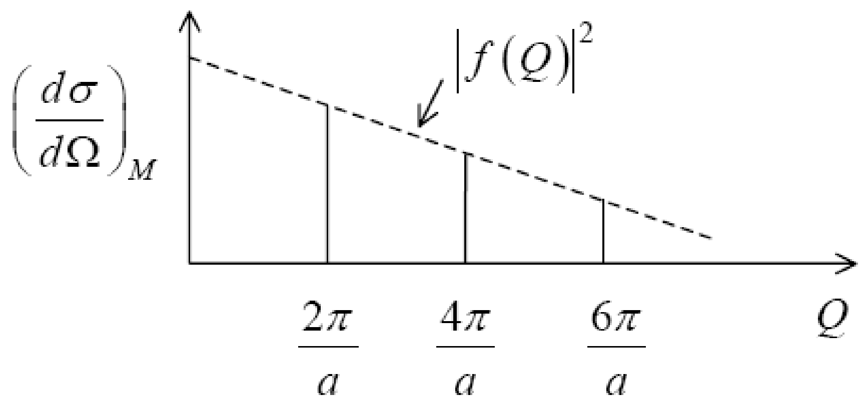
Max-Planck-Institut für Festkörperforschung

Example one-dimensional ferromagnet



nuclear and magnetic unit cells identical $\Rightarrow K_M = K_N = \frac{2\pi}{a} n$, n integer.





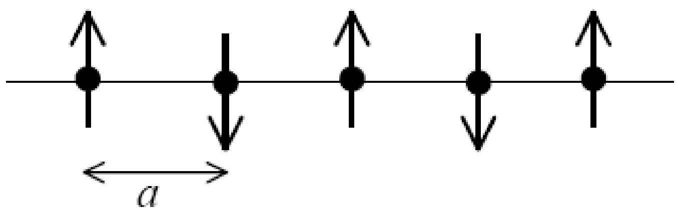
use **interference** between nuclear and magnetic scattering to create spin-polarized neutrons

$$\frac{d\sigma}{d\Omega} \sim |b|^2 + |\hat{\eta}|^2 + b\hat{\eta}$$

(up to prefactors)

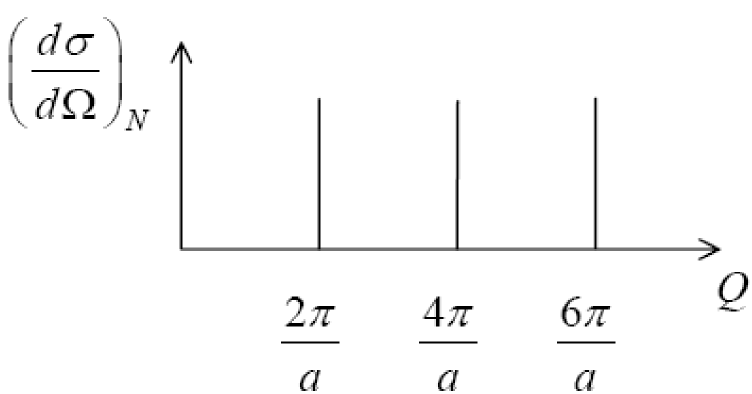
Max-Planck-Institut für Festkörperforschung

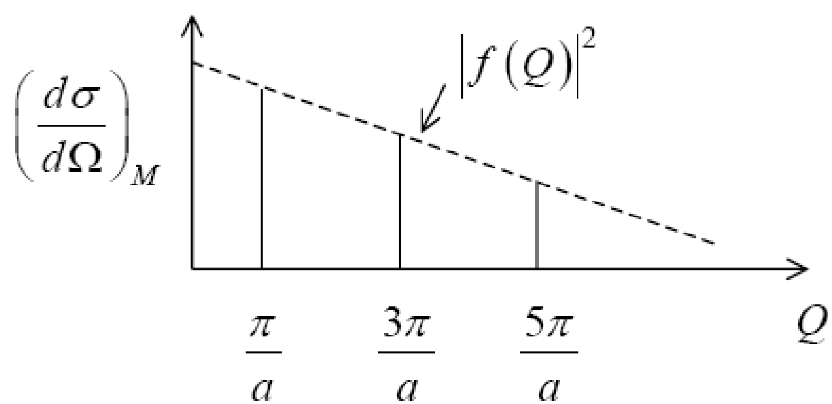
Example one-dimensional antiferromagnet



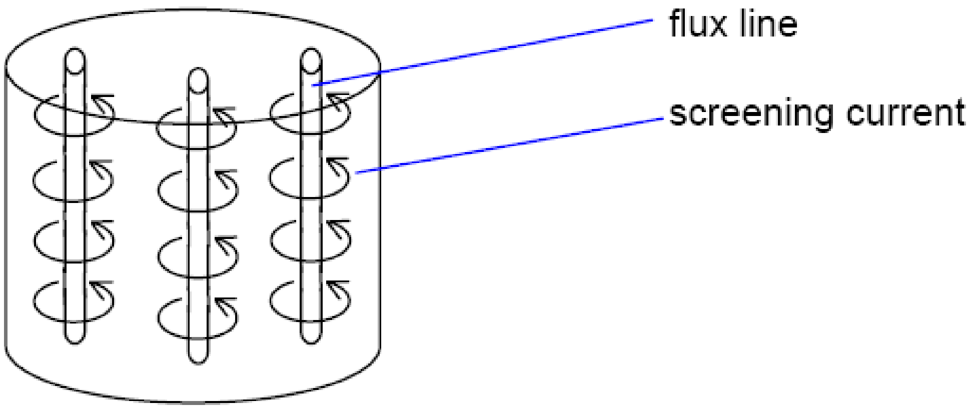
magnetic unit cell twice as large as nuclear unit cell $\Rightarrow\; K_M = \dfrac{\pi}{a} n \neq K_N = \dfrac{2\pi}{a} n$

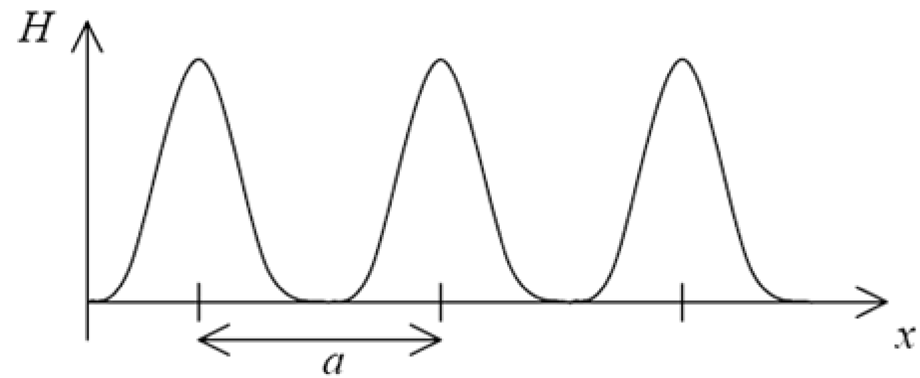
$$|F_M|^2 = |f(Q)|^2 \; |+1 - e^{iQa}|^2 = 4|f(Q)|^2 \sin^2 \frac{Qa}{2} = \begin{cases} 4|f(Q)|^2 & \text{for } n \text{ odd} \\ 0 & \text{for } n \text{ even} \end{cases}$$





Example vortex lattice in type-II superconductor



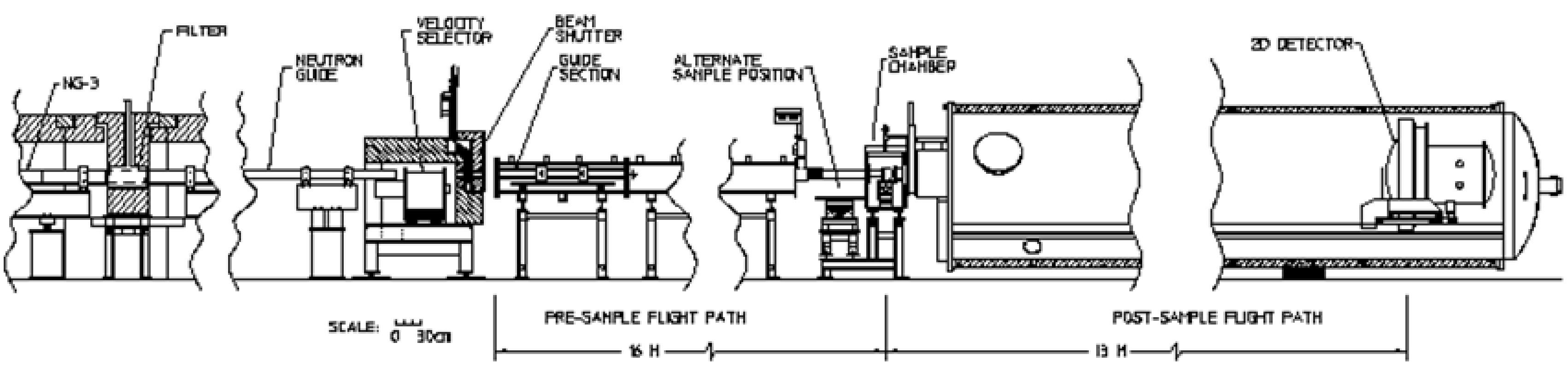


$$a \sim \sqrt{\frac{\Phi_0}{H}} \qquad \Phi_0 = \frac{h}{2e}$$

$a \sim 500\text{Å}$ for $H = 1T$

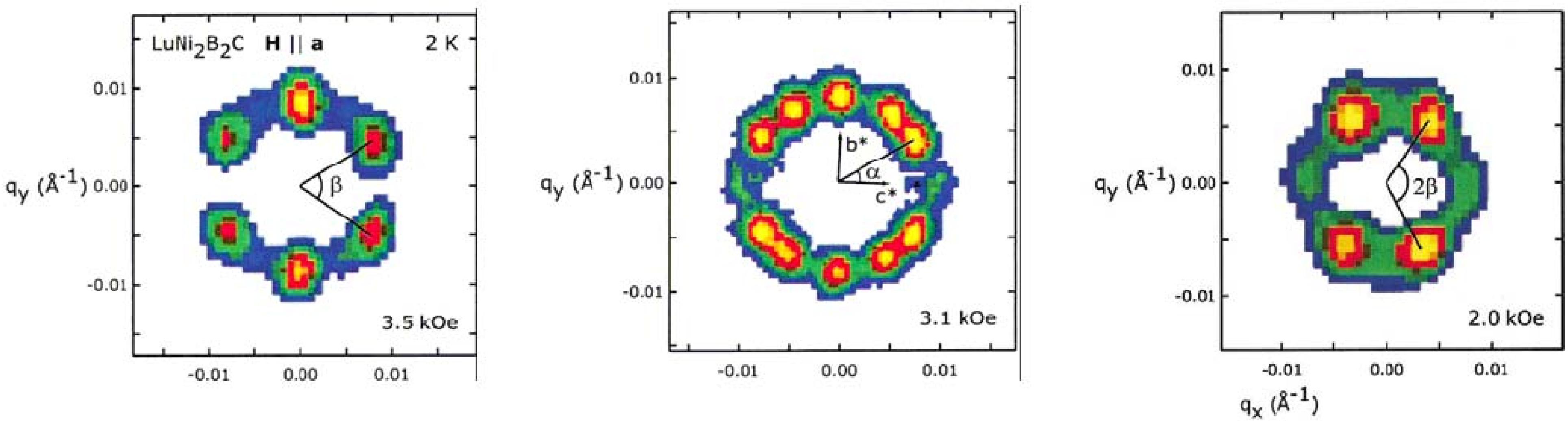
small-angle neutron scattering

CHRNS 30 METER SANS INSTRUMENT



Max-Planck-Institut für Festkörperforschung

Example vortex lattice in type-II superconductor



structural phase transition in vortex lattice

H ~ H_{c1}: electrodynamic interaction between flux lines

H ~ H_{c2}: vortex cores overlap

structure depends on superconducting coherence length

Max-Planck-Institut für Festkörperforschung

Inelastic neutron scattering

elastic cross section $\frac{d\sigma}{d\Omega} = \frac{\text{\# of neutrons scattered into } d\Omega}{(\text{unit time})\bullet(\text{incident flux})}$

inelastic cross section $\frac{d\sigma}{dEd\Omega} = \frac{\text{\# of neutrons scattered into } d\Omega}{(\text{unit time})\bullet(\text{incident flux})\bullet(\text{energy})}$

inelastic nuclear neutron scattering

$$\frac{d^2\sigma}{d\Omega\, dE} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'} b_{j'} b_j \int_{-\infty}^{\infty} \sum_{\lambda_i} p_{\lambda_i} \left\langle \lambda_i \left| e^{-i\mathbf{QR}_{j'}(0)} e^{i\mathbf{QR}_j(t)} \right| \lambda_i \right\rangle e^{-i\omega t} dt$$

$|\lambda_i\rangle \; |\lambda_f\rangle$ initial, final state of sample

$\hbar\omega = \frac{\hbar^2 k_i^2}{2m_n} - \frac{\hbar^2 k_f^2}{2m_n} = E_{\lambda_i} - E_{\lambda_f}$ energy of excitation created by neutron in sample

$p_{\lambda_i} = \exp(-E_{\lambda_i}\beta)/Z \quad Z = \sum_{\lambda_i} \exp(-E_{\lambda_i}\beta)$ partition function

Inelastic nuclear neutron scattering

$$\frac{d^2\sigma}{d\Omega\, dE} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\frac{\sigma_{coh}}{4\pi}\sum_{jj'}\int_{-\infty}^{\infty}\left\langle e^{-i\mathbf{Q}\mathbf{R}_{j'}(0)}e^{i\mathbf{Q}\mathbf{R}_{j}(t)}\right\rangle \exp\left(-i\omega t\right) dt$$

$\langle e^{\cdots}e^{\cdots}\rangle = \sum p_{\lambda_i}\langle\lambda_i| e^{\cdots}e^{\cdots}|\lambda_i\rangle$ thermal average $\sigma_{coh} = 4\pi(\overline{b})^2$

$|\lambda\rangle$ characterized by population n_s of phonons of energy $\hbar\omega_s(\vec{k})$ in branch s

Debye-Waller factor due to thermal lattice vibrations

$$\frac{d^2\sigma}{d\Omega\, dE} = \frac{\sigma_{coh}}{4\pi}\frac{k_f}{k_i}\frac{(2\pi)^3}{v_0}\frac{1}{2M}e^{-2W}\sum_s\sum_\eta\frac{(\mathbf{Q}\cdot\mathbf{e}_s)^2}{\omega_s}\times$$
$$\{\langle n_s+1\rangle\,\delta(\omega-\omega_s)\,\delta(\mathbf{Q}-\mathbf{q}-\mathbf{K}) + \langle n_s\rangle\,\delta(\omega+\omega_s)\,\delta(\mathbf{Q}+\mathbf{q}-\mathbf{K})\}$$

phonon creation
neutron energy loss

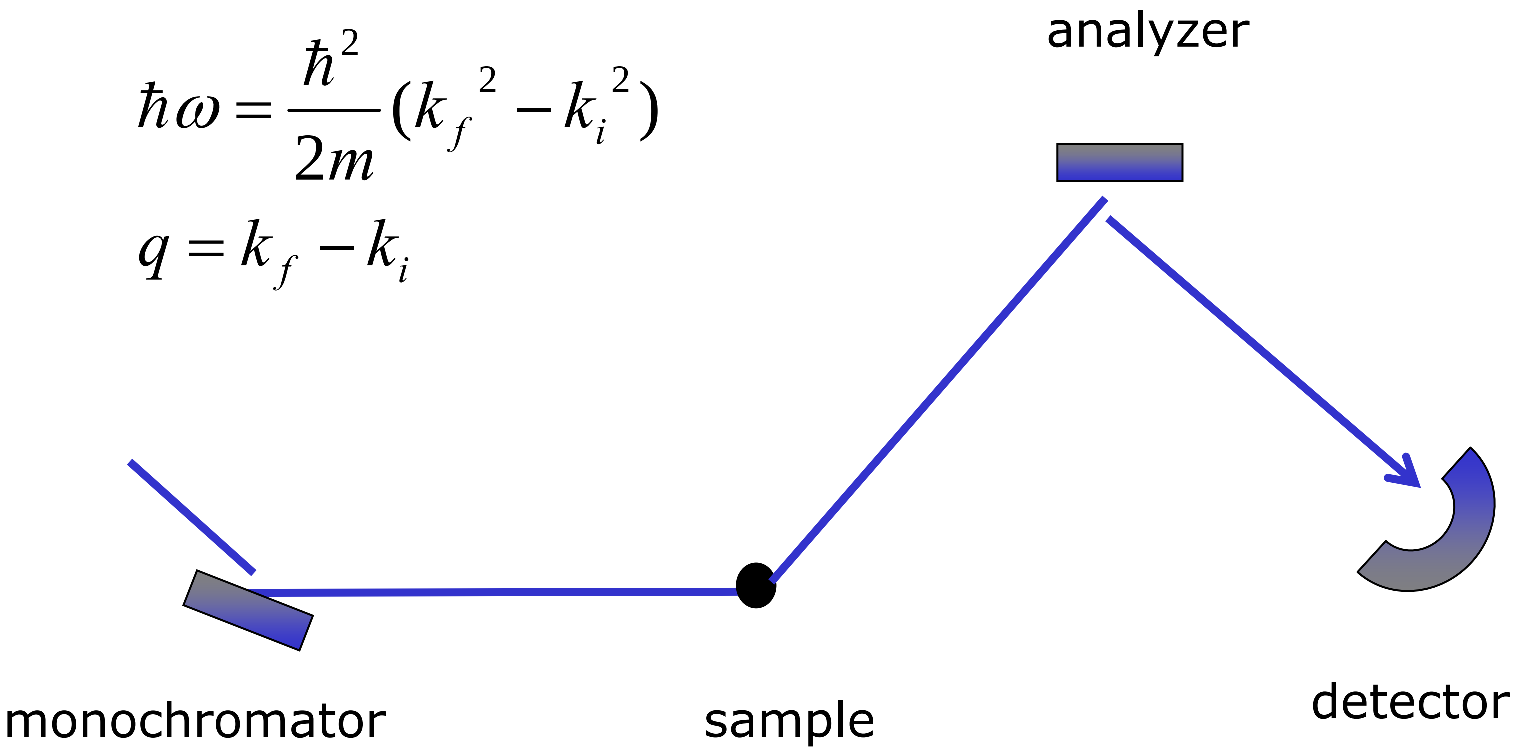
phonon annihilaion
neutron energy gain

für Festkörperforschung

Triple-axis spectrometer

$$\hbar\omega = \frac{\hbar^2}{2m}(k_f{}^2 - k_i{}^2)$$

$$q = k_f - k_i$$



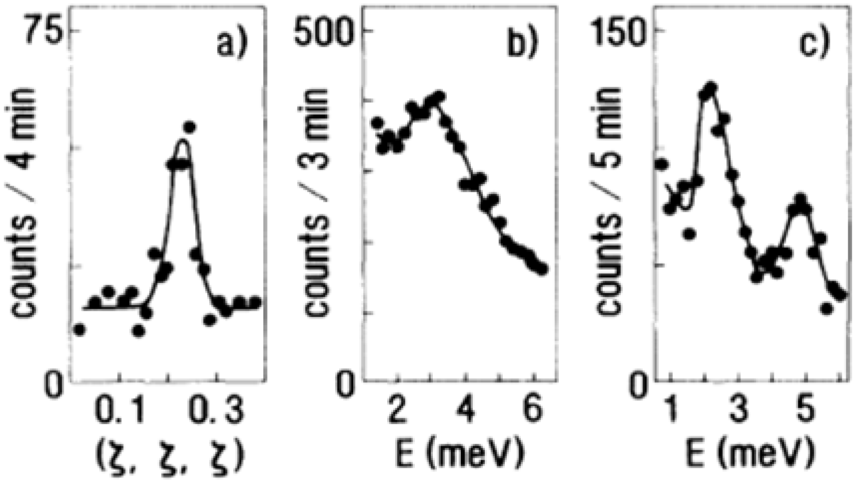
Triple-axis spectrometer

TRISP at FRM-II

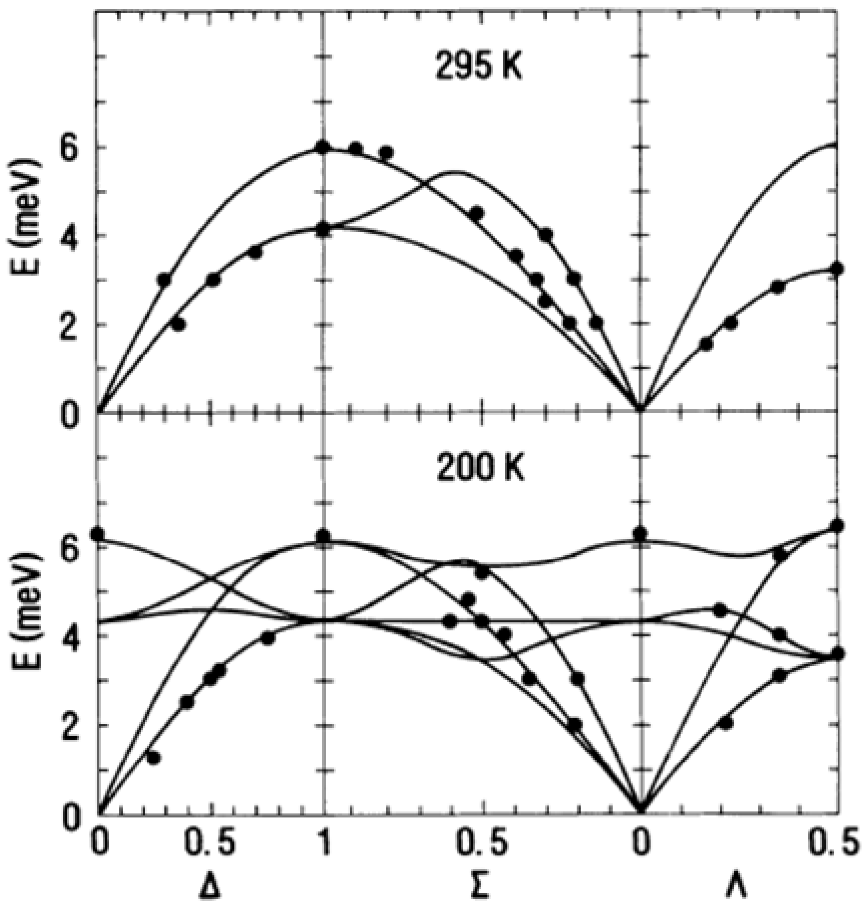
Example C_{60}

lattice structure

typical inelastic nuclear scattering scans with a triple-axis spectrometer



Example C_{60}



fcc lattice at room temperature

molecules rotate freely

molecules "lock in" at low temperatures

unit cell becomes larger

new optical phonon modes appear

Inelastic magnetic neutron scattering

$$\frac{d^2\sigma}{d\Omega\, dE} = (\gamma r_0)^2 \frac{k_f}{k_i} N \left|F(\mathbf{Q})\right|^2 e^{-2W} \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(\mathbf{Q},\omega)$$

polarization factor

$$S^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar} \int \sum_l e^{i\mathbf{Q}\mathbf{r}_l} \left\langle S_0^\alpha(0) S_l^\beta(t) \right\rangle e^{-i\omega t} dt$$

spin-spin correlation function

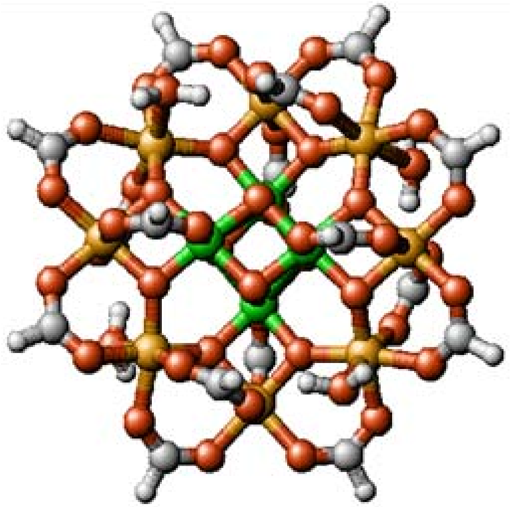
Heisenberg antiferromagnet, magnon creation

$$\frac{d^2\sigma}{d\Omega\, dE} = (\gamma r_0)^2 \frac{k_f}{k_i} \left|F(\mathbf{Q})\right|^2 e^{-2W} \frac{(2\pi)^3}{4N v_0} \{1 + (\hat{Q}\hat{\eta})^2\} \times \sum_{a=0,1} \sum_{q,\, K_m} \langle n_{q,a} + 1 \rangle\, \delta(\omega_{q,a} - \omega)\, \delta(\mathbf{Q} - \mathbf{q} - \mathbf{K_m})$$

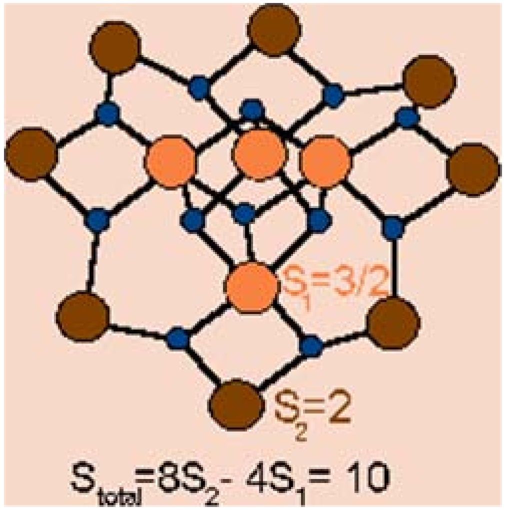
itinerant electrons → next lecture

Example molecular magnetism

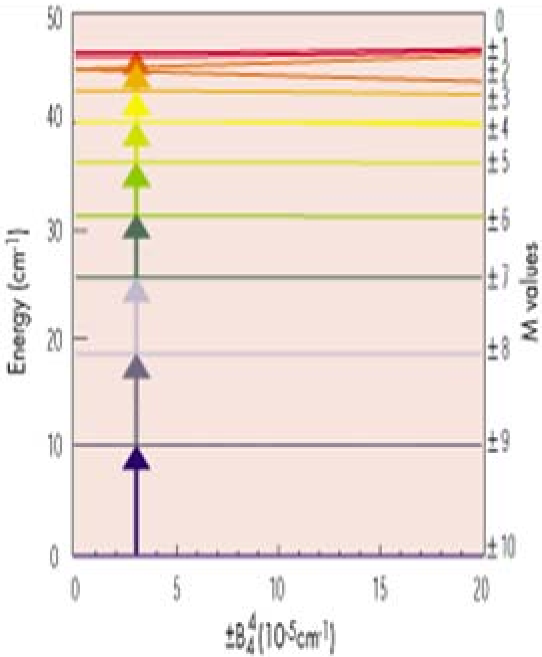
Mn_{12} acetate molecule



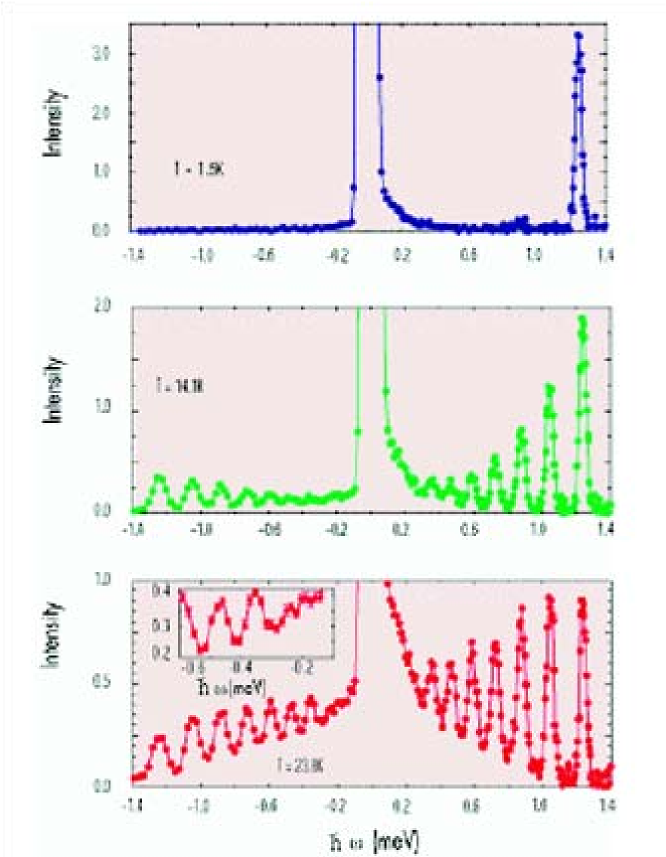
Mn atoms



energy levels

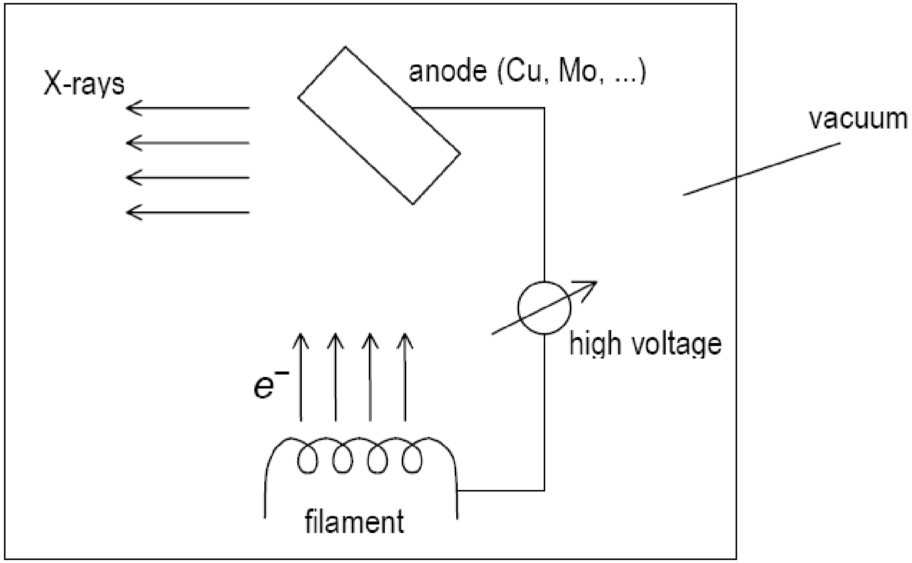


inelastic magnetic neutron scattering intensity

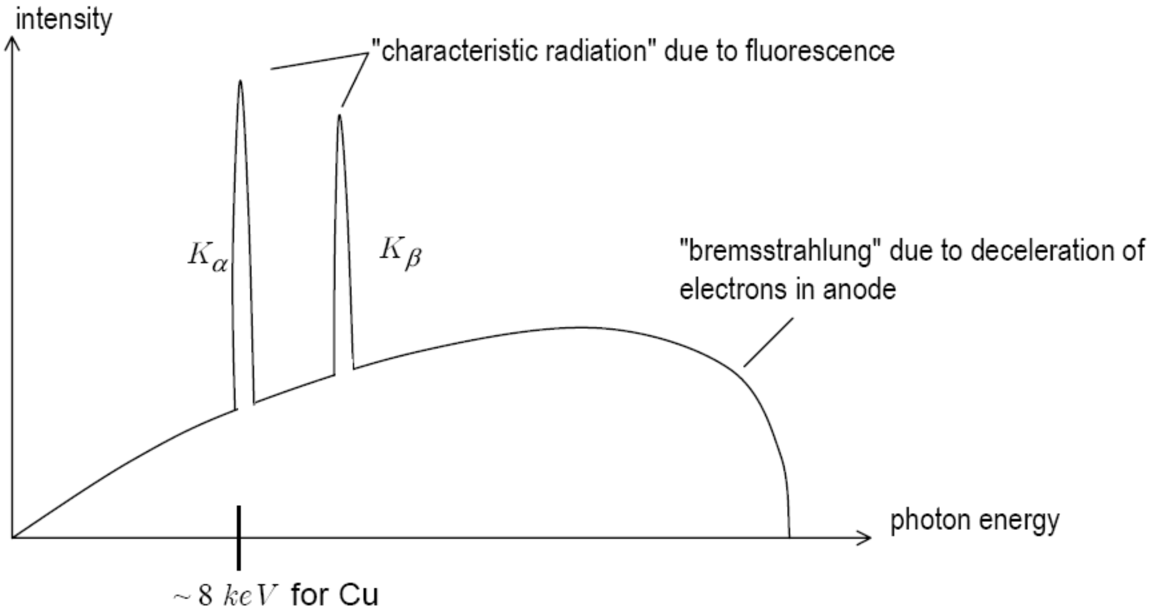


X-ray sources: tube

setup

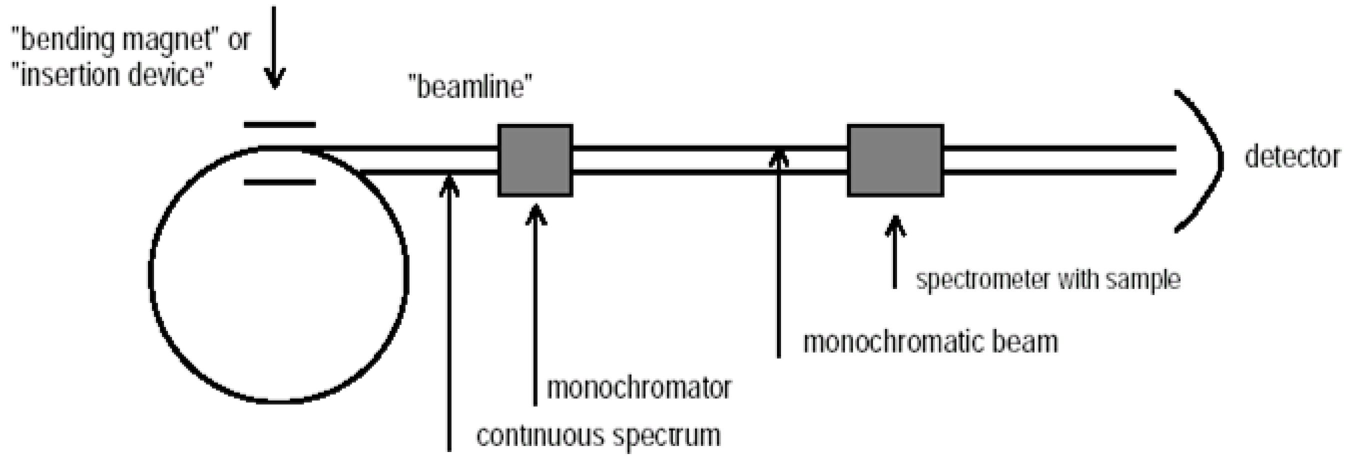


spectrum



X-ray sources: synchrotron

synchrotron

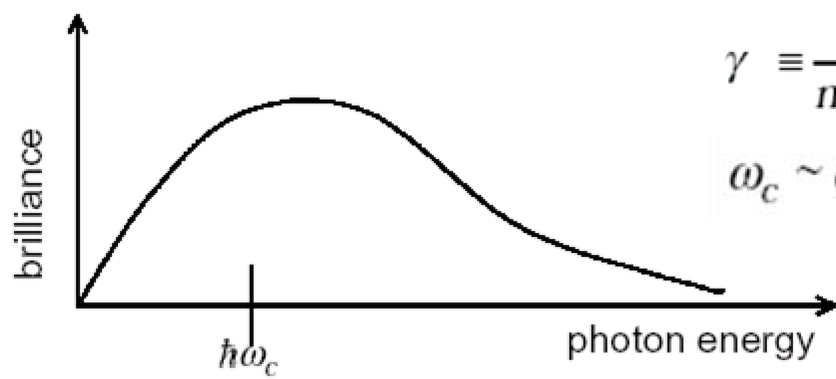


$$\varepsilon_e \sim 5GeV \qquad m_e c^2 = 0.5MeV$$

$$\gamma \equiv \frac{\varepsilon_e}{m_e c^2} \sim 10^4$$

$$\omega_c \sim \omega_o \gamma^3 \sim 20\ keV$$

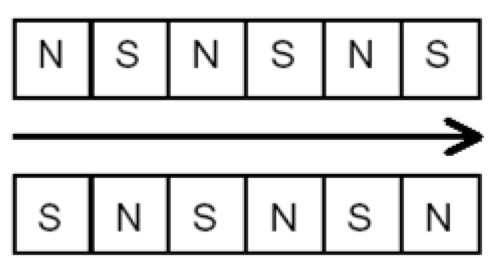
primary spectrum



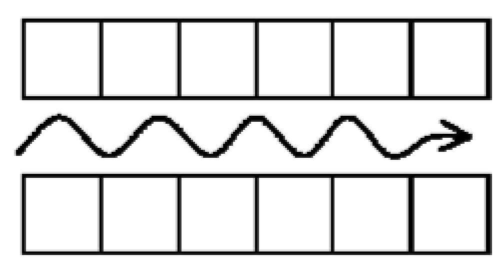
insertion devices:

wiggler, undulator

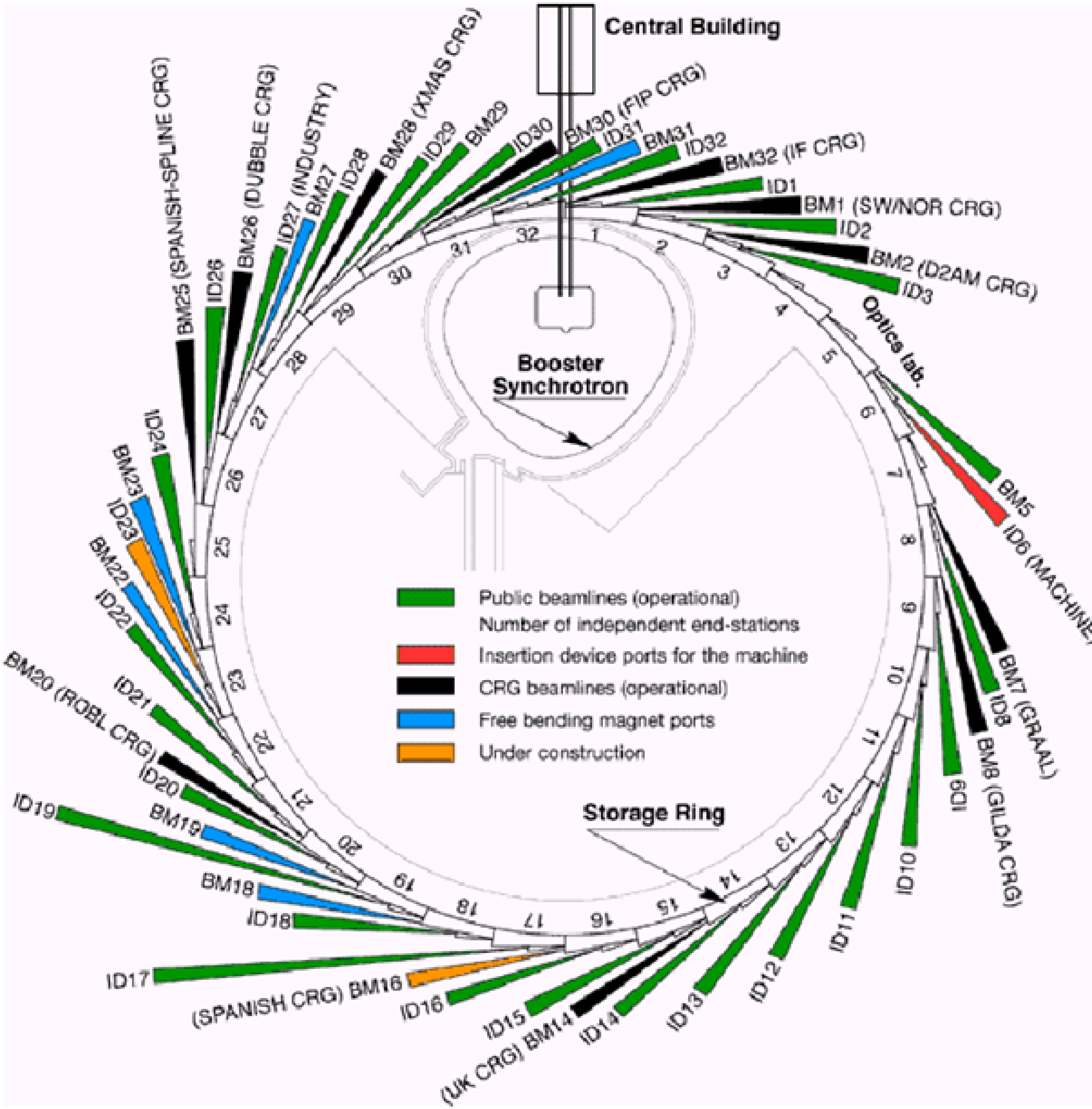
side view:



top view:



X-ray sources: synchrotron

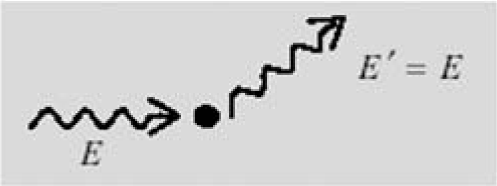


ESRF
Grenoble, France

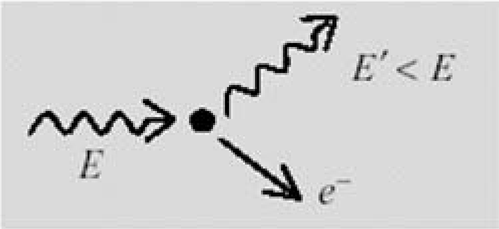
Max-Planck-Institut für Festkörperforschung

Interaction of x-rays with matter

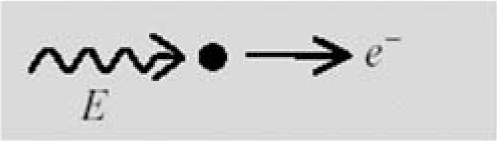
– elastic scattering (Thompson or Rayleigh scattering)



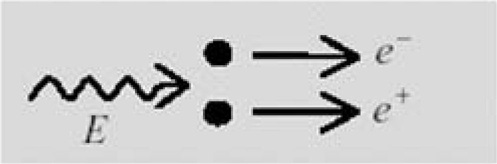
– inelastic scattering (Compton scattering)



– photoelectric absorption

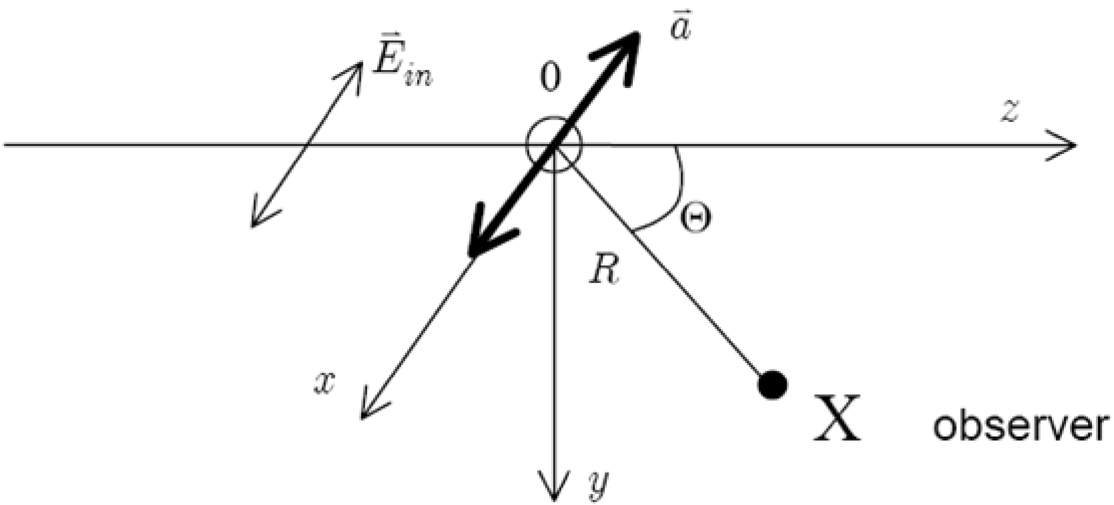


– pair creation



für Festkörperforschung

Thompson scattering – one electron



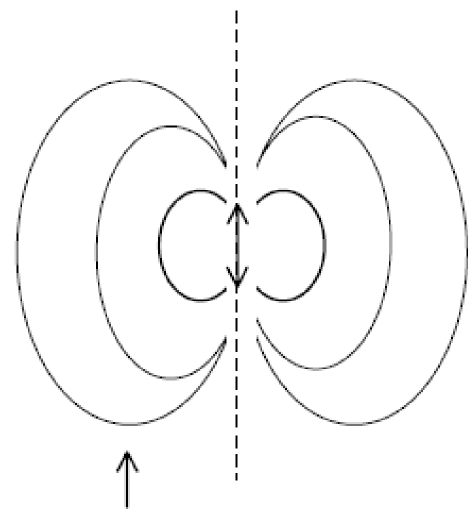
Electron is accelerated by incoming E-field.

$$a(t) = \frac{F_x}{m} = \frac{e}{m} E_{in}(t)$$

re-radiates E-field ("antenna")

$$E_{rad}(R,t) = \frac{-e}{4\pi\varepsilon_0 c^2 R} \; a_x(t') \qquad t' = t - \frac{R}{c}$$

energy density $\propto E^2$
angle-integrated energy density
independent of R
(energy conservation)



contour plot of
radiated intensity

Thompson scattering – one electron

apparent acceleration seen by observer:

$$a_x(t') = -\frac{e}{m} E_o e^{-i\omega t'} \cos\Theta$$

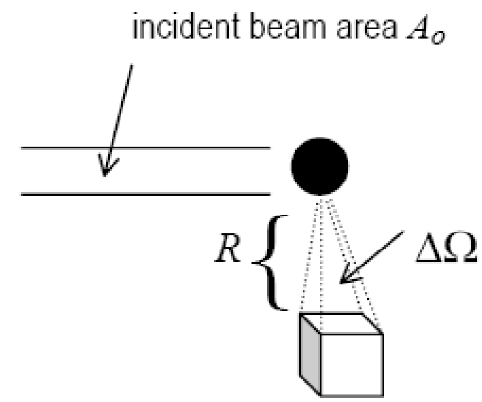
$$= -\frac{e}{m} E_{in}(t) e^{i\frac{\omega R}{c}} \cos\Theta$$

$$\frac{E_{rad}(R,t)}{E_{in}(0,t)} = \underbrace{\frac{e^2}{4\pi \in_o mc^2}}_{\text{dimension of length}} \frac{e^{ikR}}{R} \cos\Theta$$

$\equiv r_0$ ("classical electron radius")

$= 2.8\times10^{-5}\,\mathring{A}$

$$\frac{I_{det}}{I_{in}} = \frac{|E_{rad}|^2 R^2 \Delta\Omega}{|E_{in}|^2 A_o}$$

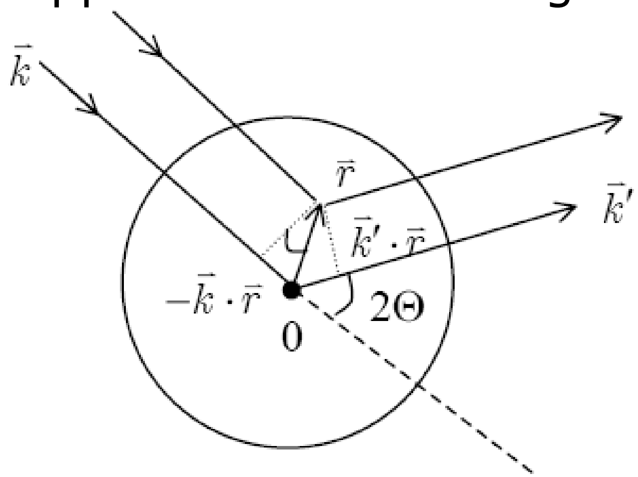


differential cross section: one electron

$$\frac{d\sigma}{d\Omega} = \frac{\#\text{ of photons detected}}{\Delta\Omega\text{ (incident flux)}} = \frac{|E_{rad}|^2 R^2}{|E_{in}|^2} = r_o{}^2 \cos^2\Theta$$

Thompson scattering – one atom

approximated as charged sphere, charge density *ρ(r)*



elastic scattering: $|\vec{k}| = |\vec{k}'|$

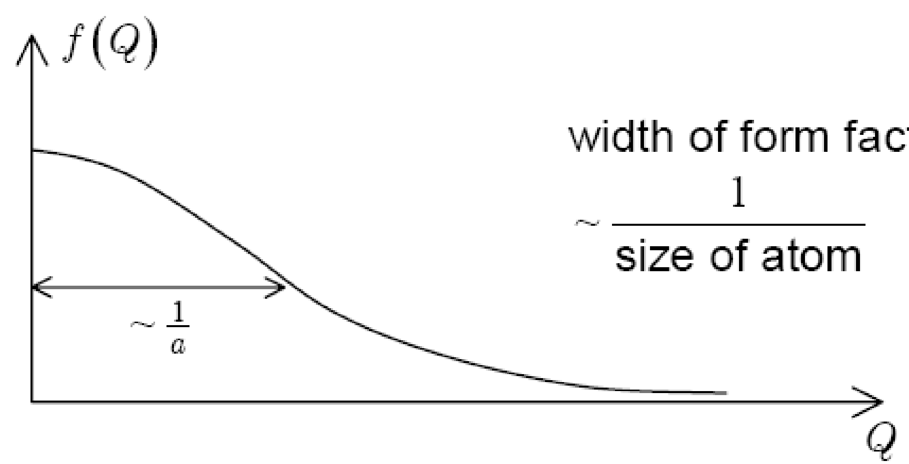
$|\vec{r}| \ll |\vec{R}|$

phase difference between wave scattered at 0 and at $\vec{r}$: $\left(\vec{k} - \vec{k}'\right)\cdot\vec{r} \equiv \vec{Q}\cdot r$

atomic form factor

$$\frac{d\sigma}{d\Omega} \sim \left| r_o \underbrace{\int \rho(\vec{r})\, e^{i\vec{Q}\cdot\vec{r}}\, d\vec{r}}_{\equiv f(\vec{Q})} \right|^2$$

× (polarization factor)



width of form factor

$$\sim \frac{1}{\text{size of atom}}$$

Thompson scattering – crystal lattice

$$\frac{d\sigma}{d\Omega} = r_0^2 \left\langle \left| \sum_n e^{i\vec{Q}\cdot\vec{R}_n} f(\vec{Q}) \right|^2 \right\rangle = r_0^2 \left| f(\vec{Q}) \right|^2 \sum_{mn} e^{i\vec{Q}(\vec{R}_{0m}-\vec{R}_{0n})} \left\langle e^{i\vec{Q}(\vec{u}_m-\vec{u}_n)} \right\rangle$$

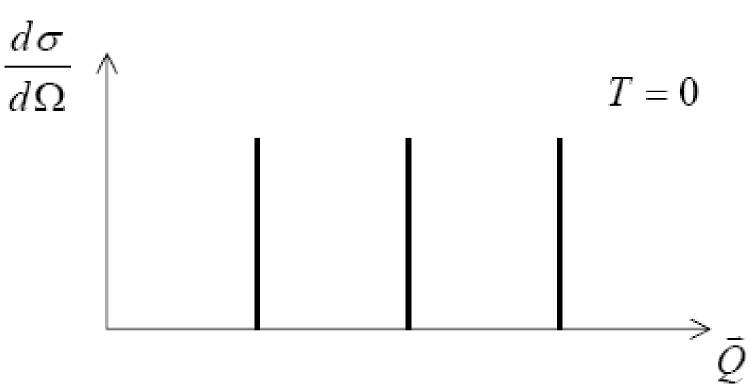
equilibrium positions   thermal vibrations

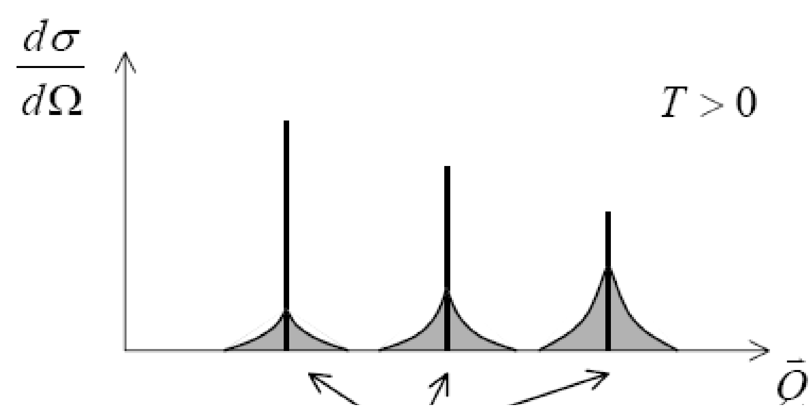
expansion of $\langle \ldots \rangle$

$$= r_0^2 \left| f(\vec{Q}) \right|^2 e^{-2W} \frac{(2\pi)^3}{v_0} \sum_{\vec{K}} \delta(\vec{Q}-\vec{K}) \quad + \ldots$$

Debye-Waller factor

Bragg reflections at reciprocal lattice vectors *K*





thermal diffuse scattering

ür Festkörperforschung

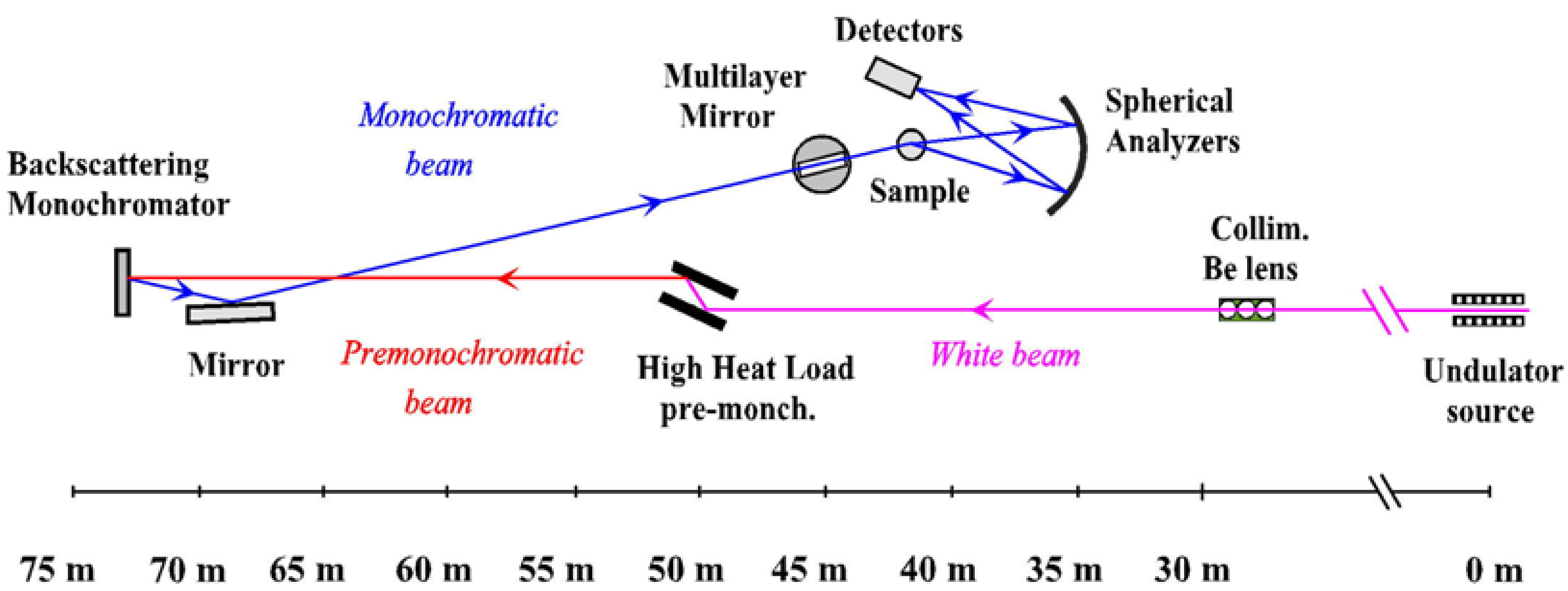
Inelastic x-ray scattering

photon energy ~ 10 keV

phonon energy ~ 10 meV

→ resolution $\Delta E/E < 10^{-7}$ required

triple-axis spectrometer



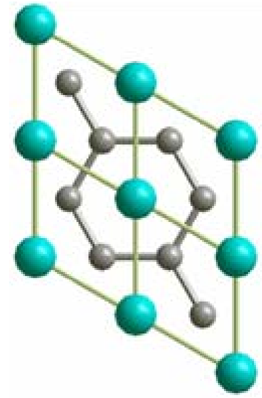
Inelastic x-ray scattering

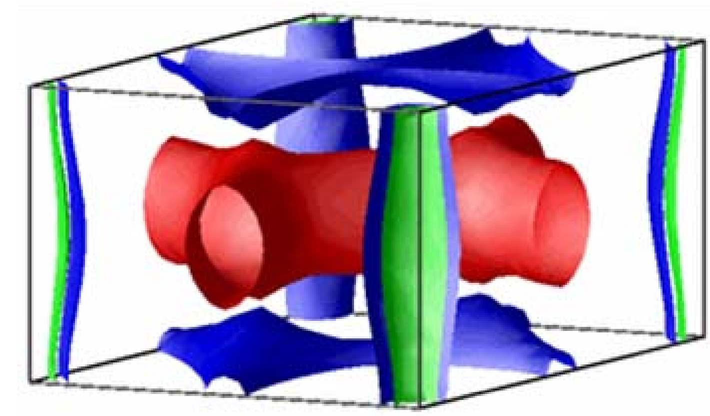
ID-16
ESRF Grenoble, France

Example MgB_2

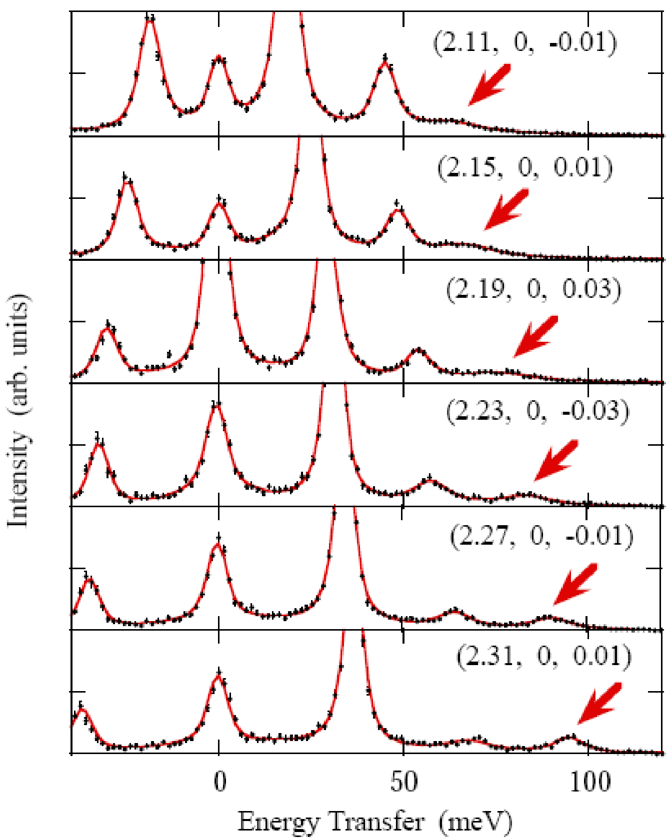
B vibration

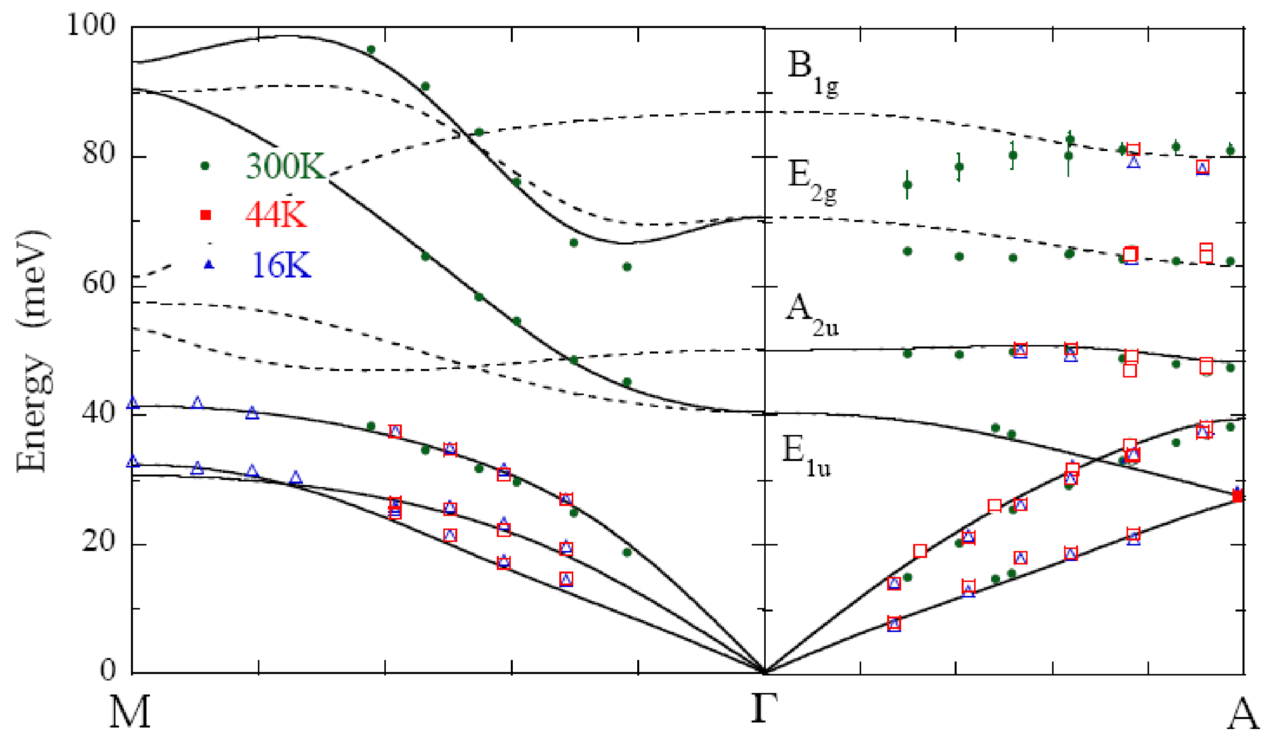
- modulates Fermi surface
- drives superconductivity (T_c = 39 K)





IXS data





X-ray absorption

interaction Hamiltonian

$$H = H_e + H_{rad} = \frac{\vec{p}^2}{2m} + H_{rad} \qquad \vec{p} \rightarrow \vec{p} - e\vec{A} = \frac{\hbar}{i}\vec{\nabla} - e\vec{A}$$

$$H = \frac{\vec{p}^2}{2m} + H_{rad} + \quad \frac{e}{m}\vec{A}\cdot\vec{p} + \frac{e^2\vec{A}^2}{2m} \qquad \vec{A} = \hat{\varepsilon}\sqrt{\frac{\hbar}{2\epsilon_0 V\omega}}\left(a_k^+ e^{-i\vec{k}\cdot\vec{r}} + a_k e^{i\vec{k}\cdot\vec{r}}\right)$$

annihilation operator

photon creation

absorption
photon annihilated

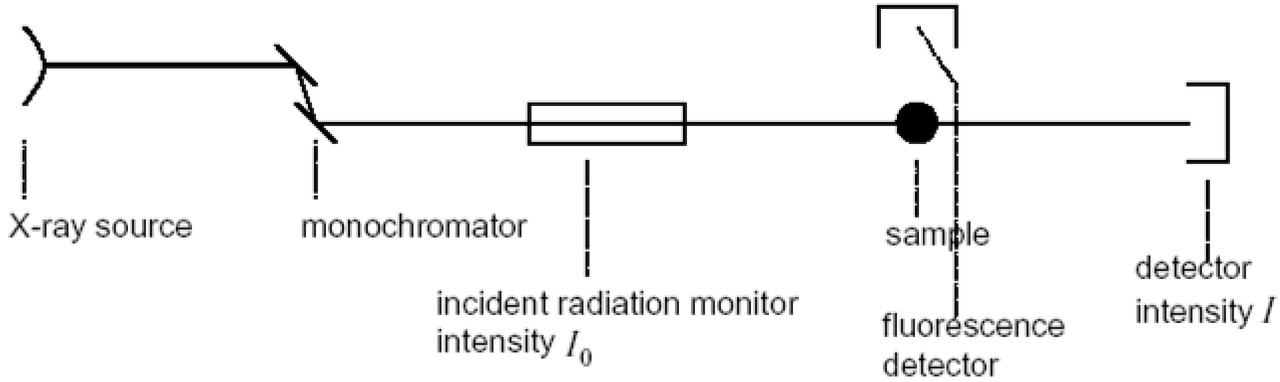
scattering
photon number conserved

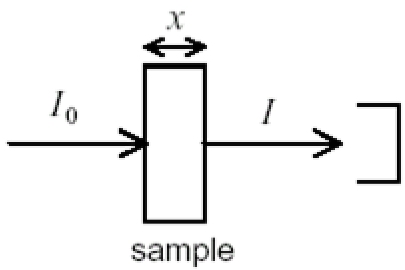
absorption cross section

$$\sigma_a = \frac{W}{\Phi_0} \qquad W = \frac{2\pi}{\hbar}\left|M_{fi}\right|^2 \rho(E_f) \qquad M_{fi} = \langle f | H_{\text{int}} | i \rangle$$

für Festkörperforschung

X-ray absorption

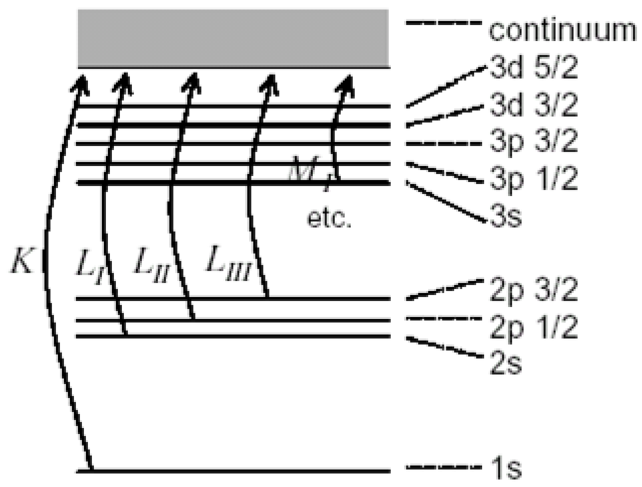


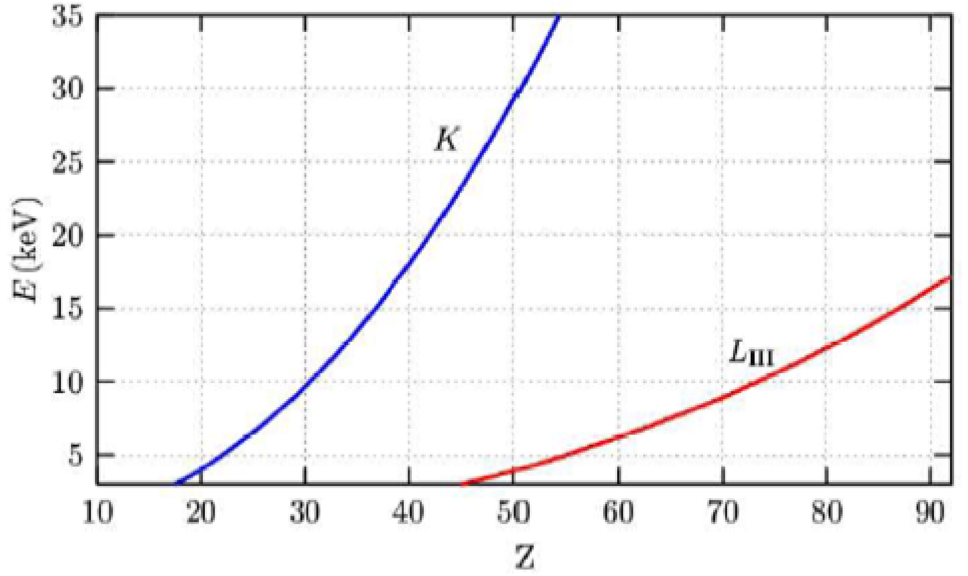


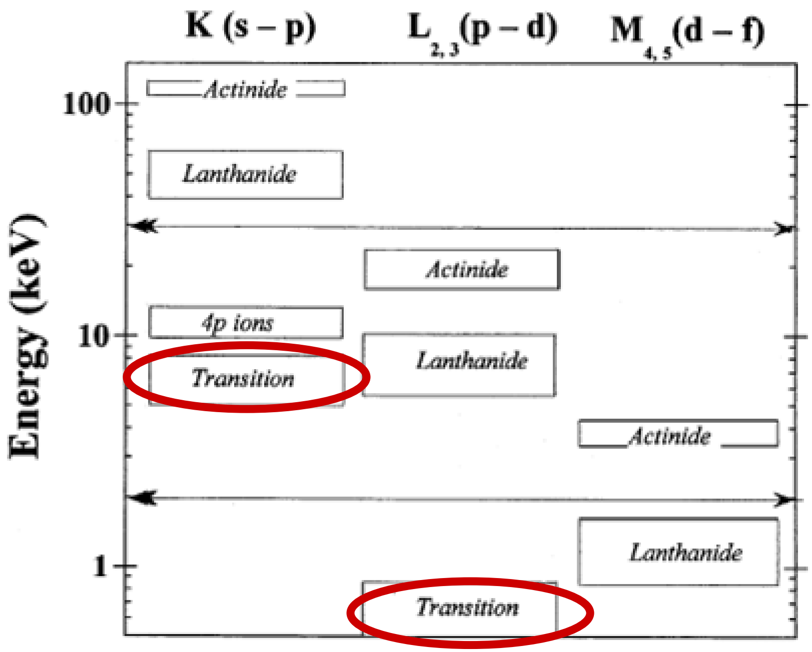
$$\frac{I}{I_0} = e^{-(\mu/\rho_a)x}$$

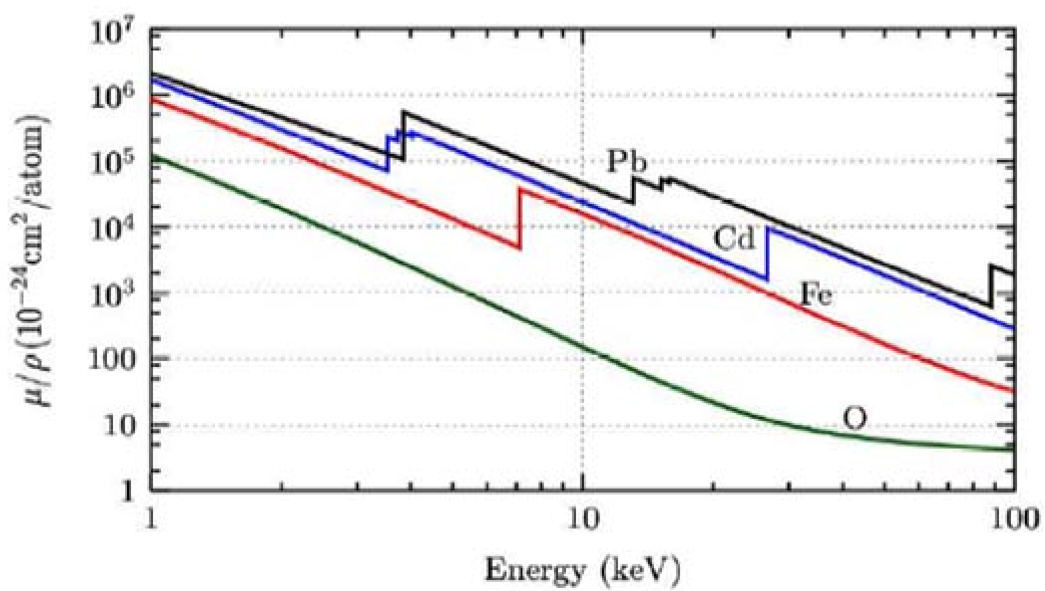
mass absorption coefficient

X-ray absorption edges









Example K-edge

absorption cross section

$$\sigma_a = \frac{W}{\Phi_0} \qquad W = \frac{2\pi}{\hbar}\left|M_{fi}\right|^2 \rho(E_f) \qquad M_{fi} = \langle f | H_{\text{int}} | i \rangle$$

transitions into continuum

$$|i\rangle = |i\rangle_{photon} |i\rangle_{electron} = |1\rangle \Psi_{100}(\vec{r})$$

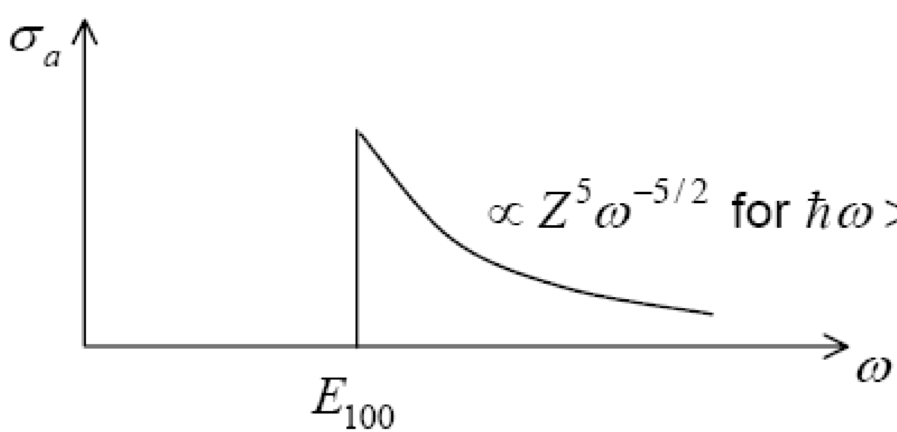
with $\Psi_{100}(\vec{r}) = \frac{1}{\sqrt{\pi}}\left(\frac{Z}{a_B}\right)^{3/2} e^{-Zr/a_B}$

$$|f\rangle = |f\rangle_{photon} |f\rangle_{electron} = |0\rangle \frac{1}{\sqrt{V}} e^{i\vec{q}\cdot\vec{r}}$$

with $\frac{\hbar}{i}\vec{\nabla} e^{i\vec{q}\cdot\vec{r}} = \hbar\vec{q}$ (photoelectron momentum)

$$M_{fi} = \frac{e\hbar}{mV}\sqrt{\frac{\hbar}{2\epsilon_0\omega}} \int d\vec{r}\, e^{-i\vec{q}\cdot\vec{r}} \, (\hat{\varepsilon}\cdot\vec{q}) e^{i\vec{k}\cdot\vec{r}} \Psi_{100}(\vec{r})$$

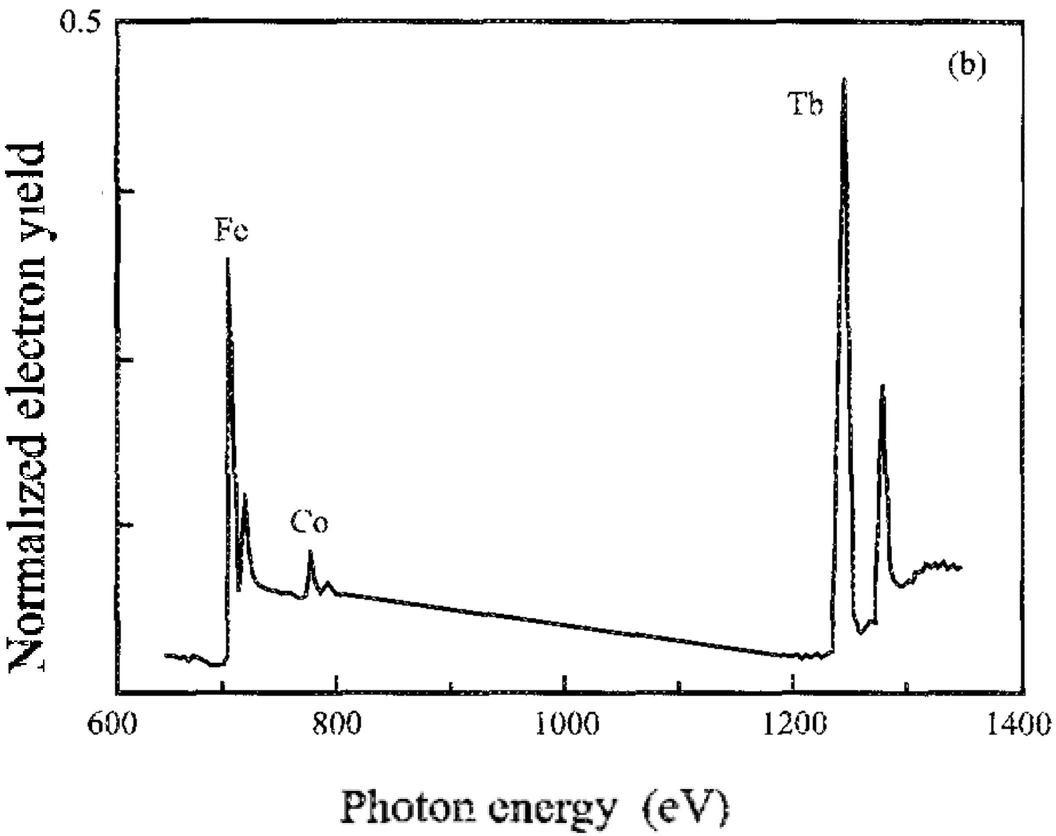
$$= \frac{e\hbar}{mV}\sqrt{\frac{\hbar}{2\epsilon_0\omega}} (\hat{\varepsilon}\cdot\vec{q}) \underbrace{\frac{Z^{5/2}}{\left[\left(\frac{Qa_B}{2}\right)^2 + Z^2\right]^2}}_{\text{Fourier transform of } \Psi_{100} \text{ with } \vec{Q} \equiv \vec{k} - \vec{q}}$$



Example Fe L-edge

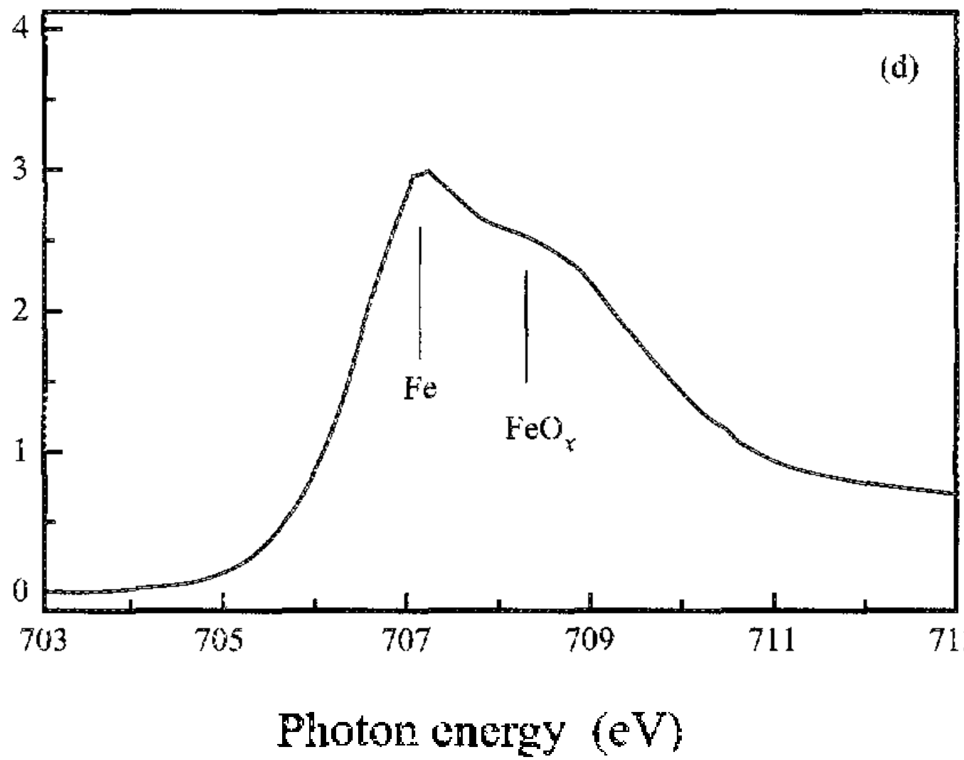
chemical analysis

example TbFeCo alloy



valence state

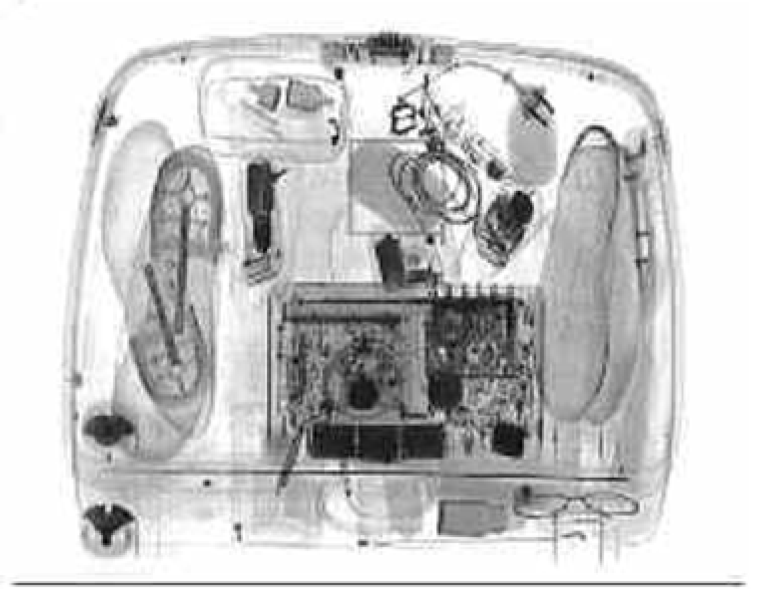
example Fe thin film



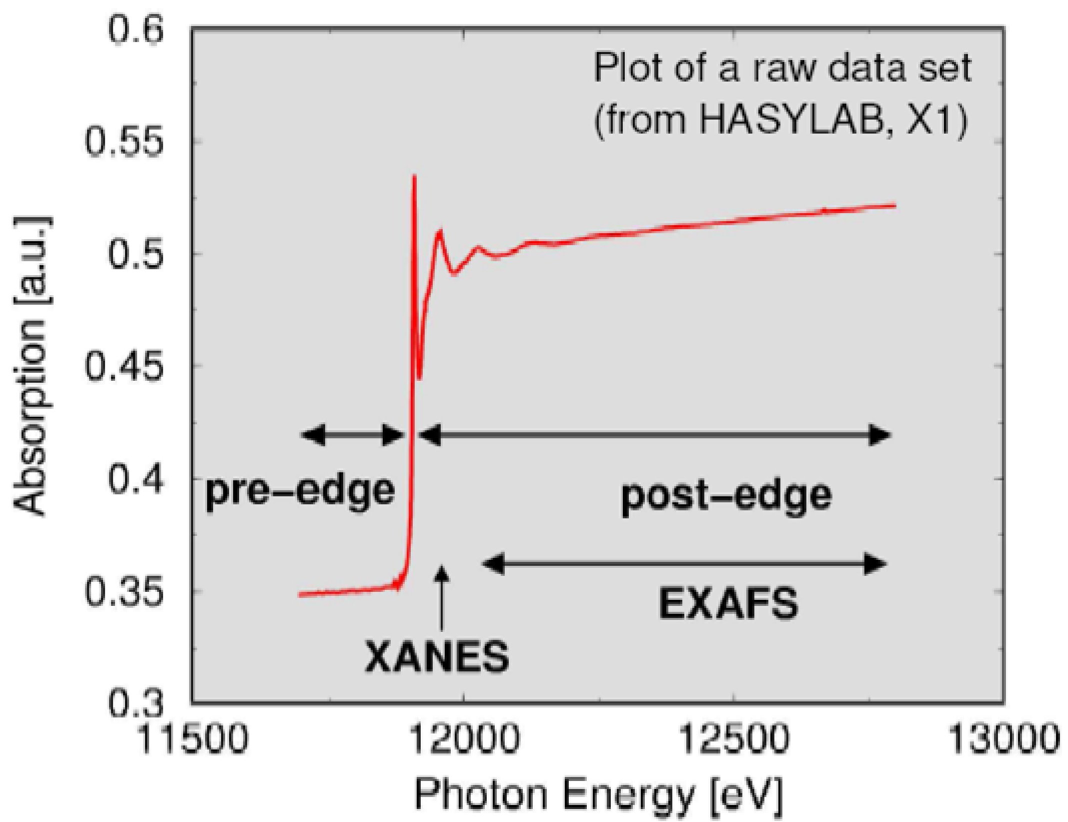
X-ray radiography

dual-energy
x-ray radiography

discriminate between
carbohydrates and metals



X-ray absorption fine structure



Example K-edge

$$|i\rangle = |i\rangle_{photon} |i\rangle_{electron} = |1\rangle \Psi_{100}(\vec{r})$$

with $\Psi_{100}(\vec{r}) = \frac{1}{\sqrt{\pi}} \left(\frac{Z}{a_B} \right)^{3/2} e^{-Zr/a_B}$

transition into unoccupied excited state

$$|f\rangle = |f\rangle_{photon} |f\rangle_{electron} = |0\rangle \Psi_{nlm}(\vec{r})$$

$$\frac{\hbar}{i} \vec{\nabla} \Psi_{100}(\vec{r}) = \frac{\hbar}{i} \frac{\hat{r}}{\sqrt{\pi}} \left(\frac{Z}{a_B} \right)^{5/2} e^{-Zr/a_B}$$

$$M_{fi} = \frac{e\hbar}{im} \sqrt{\frac{\hbar}{2\epsilon_0 V \omega}} \underbrace{\langle 0|a_k|1\rangle}_{=1} \frac{Z}{a_B} \int d\vec{r}\, \Psi_{nlm}^{*}(\vec{r})\, (\hat{\epsilon} \cdot \vec{r})\, e^{i\vec{k}\cdot\vec{r}} \Psi_{100}(\vec{r})$$

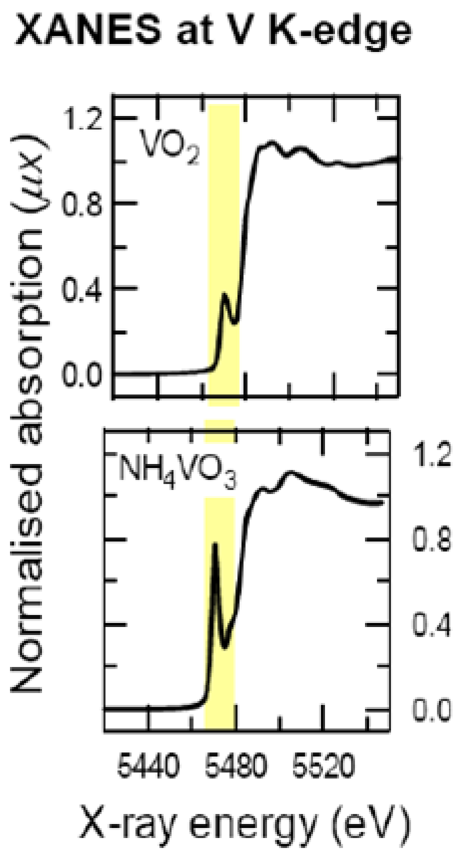
electric dipole matrix element

selection rules

$\Delta l = \pm 1$

$\Delta m = 0, \pm 1$

Example vanadium K-edge



nearly octahedral coordination

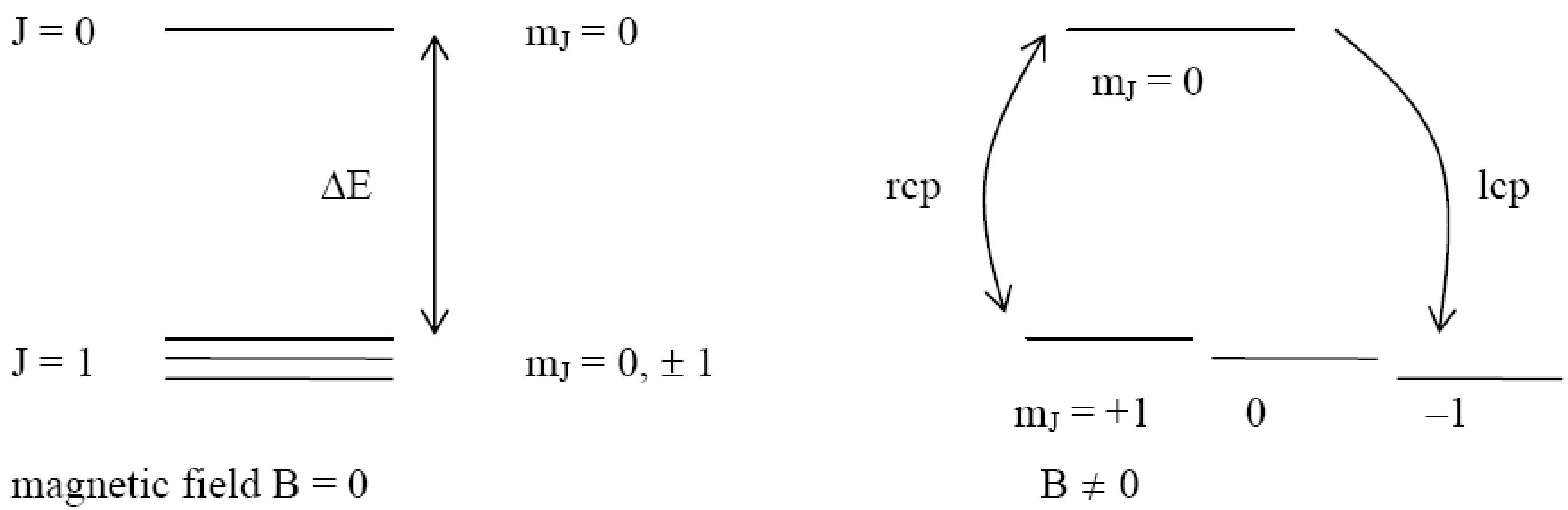
Centrosymmetry, no pd mixing,
only quadrupole transitions

nearly tetrahedral coordination

Strong pd-mixing, dipole transitions
contribute to pre-edge structure

Magnetic circular dichroism

example single atom

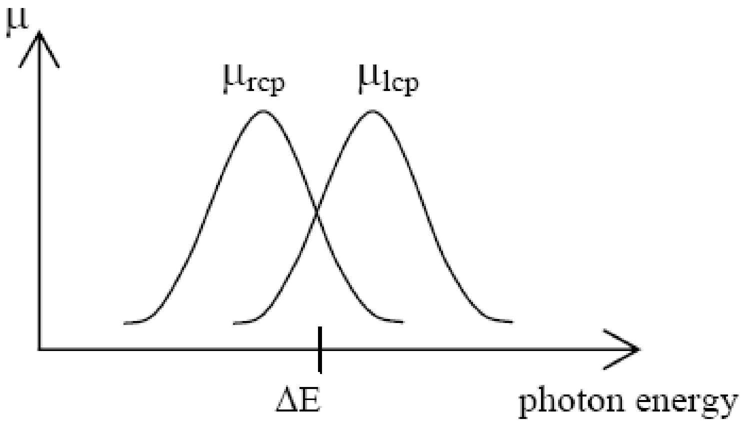


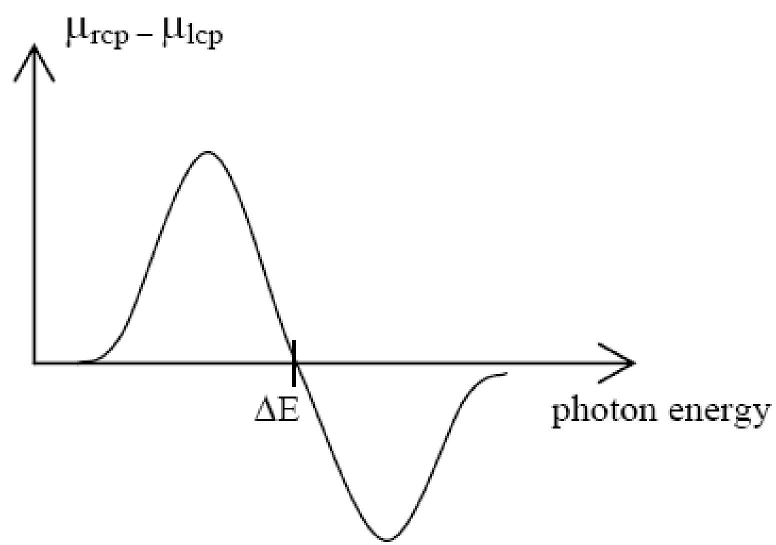
electric dipole selection rules

$m_J = +1 \to 0$ transitions induced by right circularly polarized light

$m_J = -1 \to 0$ transitions induced by left circularly polarized light

Magnetic circular dichroism





Magnetic circular dichroism

classical calculation for bound electron

analogous to Thompson scattering

$$\ddot{x} + \Gamma \dot{x} + \omega_0{}^2 x = -\frac{eE_0}{m} e^{-i\omega t}$$

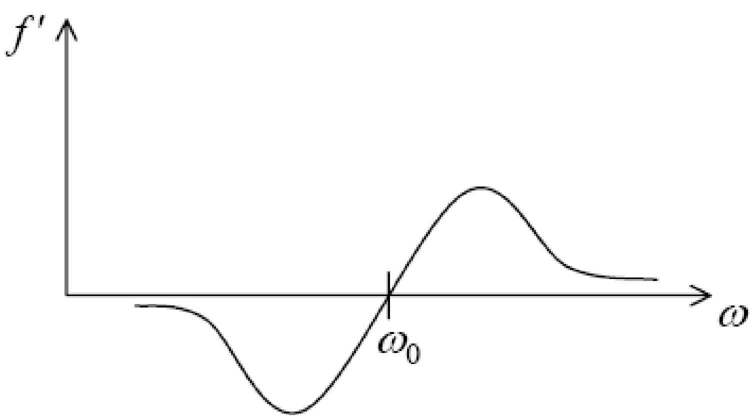
$$E_{rad} = \frac{e}{4\pi\epsilon_0 R c^2}\, \ddot{x}\left(t - \frac{R}{c}\right)$$

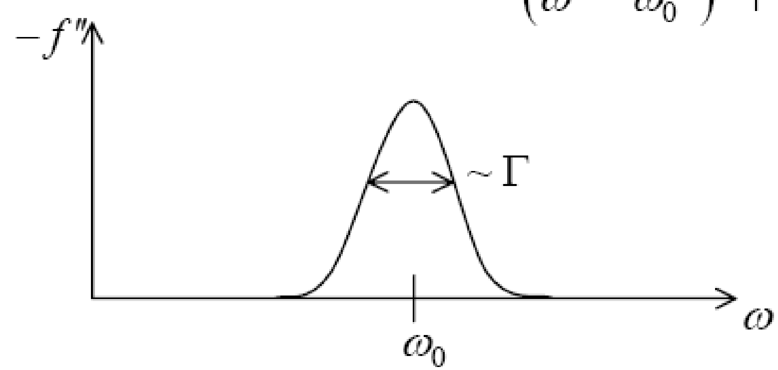
$$= \frac{e}{4\pi\epsilon_0 R c^2}\, \frac{\omega^2}{\omega_0{}^2 - \omega^2 - i\omega\Gamma}\, \frac{eE_0}{m}\, e^{-i\omega t}\, e^{ikR}$$

$$\frac{E_{rad}}{E_0} = -r_0 \frac{\omega^2}{\omega^2 - \omega_0{}^2 + i\omega\Gamma} \frac{e^{ikR}}{R} \equiv f(\omega)\, r_0 \frac{e^{ikR}}{R}$$

$$f' = \frac{\omega^2\left(\omega^2 - \omega_0{}^2\right)}{\left(\omega^2 - \omega_0{}^2\right)^2 + \omega^2\Gamma^2}$$

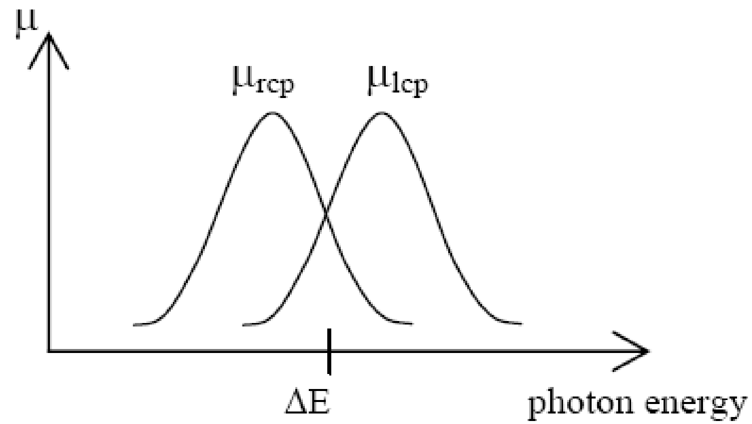
$$f'' = \frac{-\omega^3\Gamma}{\left(\omega^2 - \omega_0{}^2\right)^2 + \omega^2\Gamma^2}$$

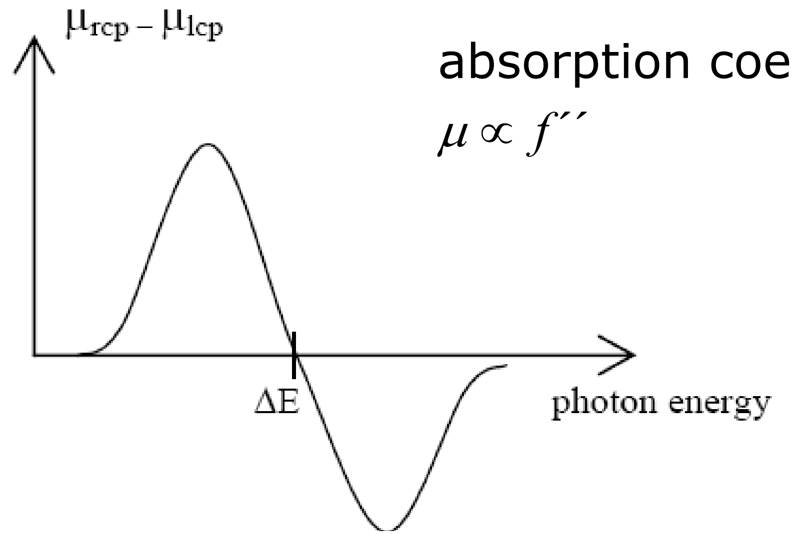




Magnetic circular dichroism

circular dichroism

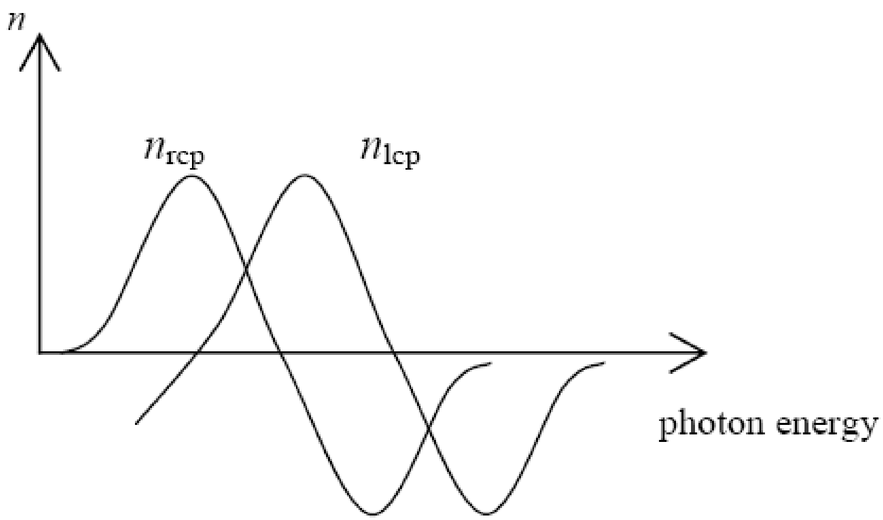


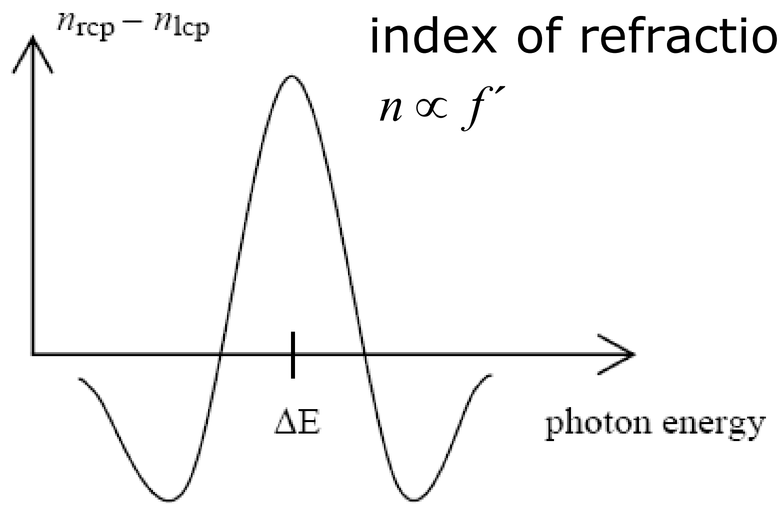


absorption coefficient

$\mu \propto f''$

circular birefringence





index of refraction

$n \propto f'$

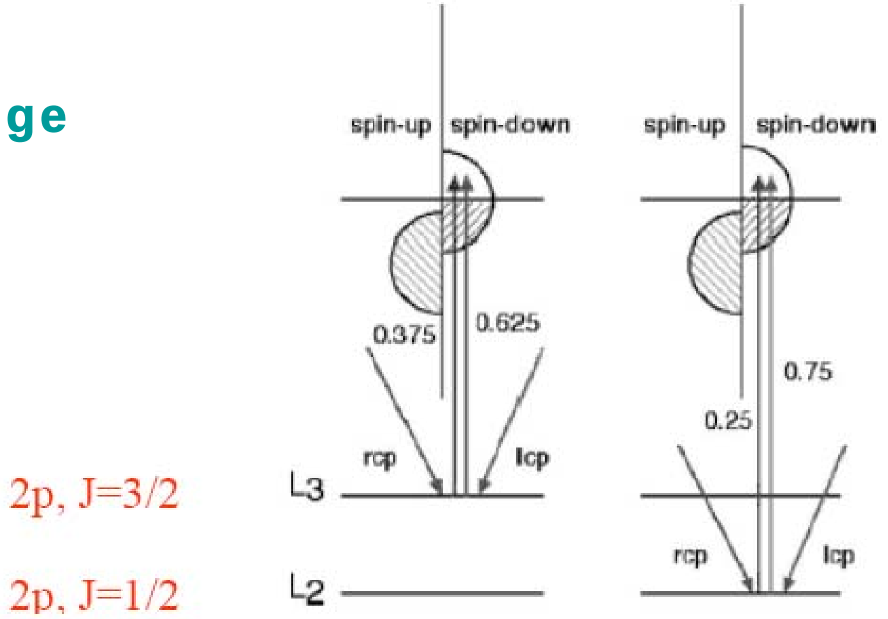
rotation of plane of polarization by $\varphi = \frac{\pi}{\lambda}\left(n_{rcp} - n_{lcp}\right)x$ after distance x

Kerr effect

Faraday effect

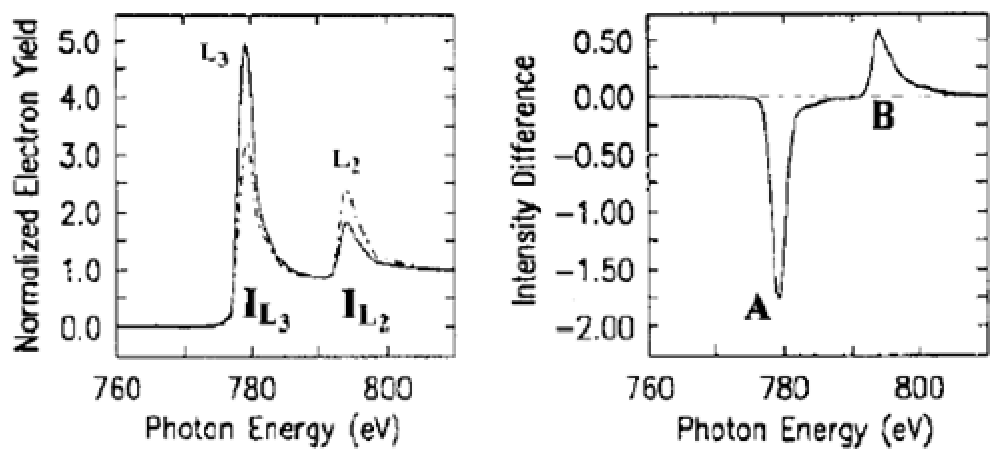
X-ray magnetic circular dichroism (XMCD)

L-absorption edge
of ferromagnet

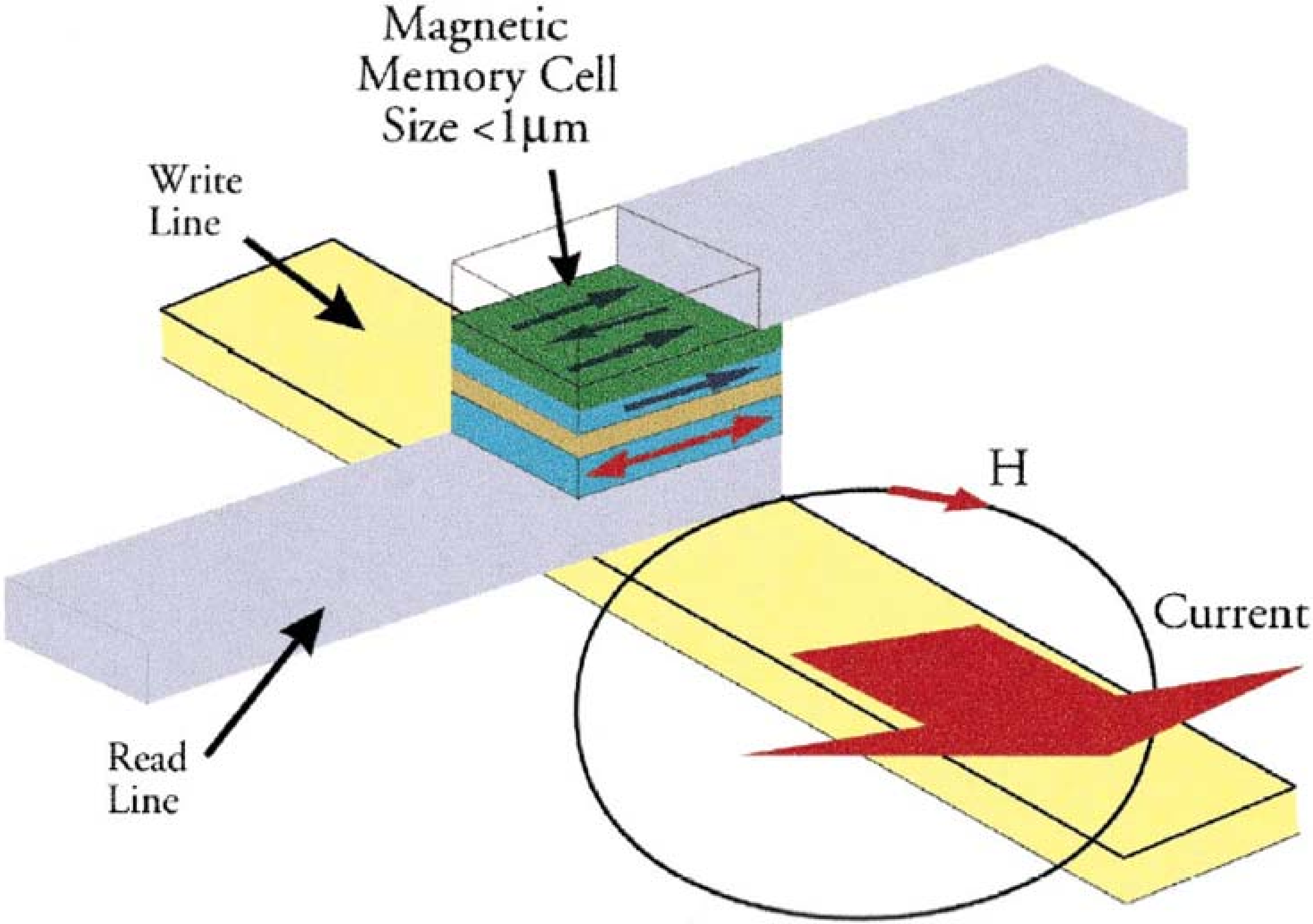


cobalt L-edge

Co $L_{2,3}$ XMCD Spectra

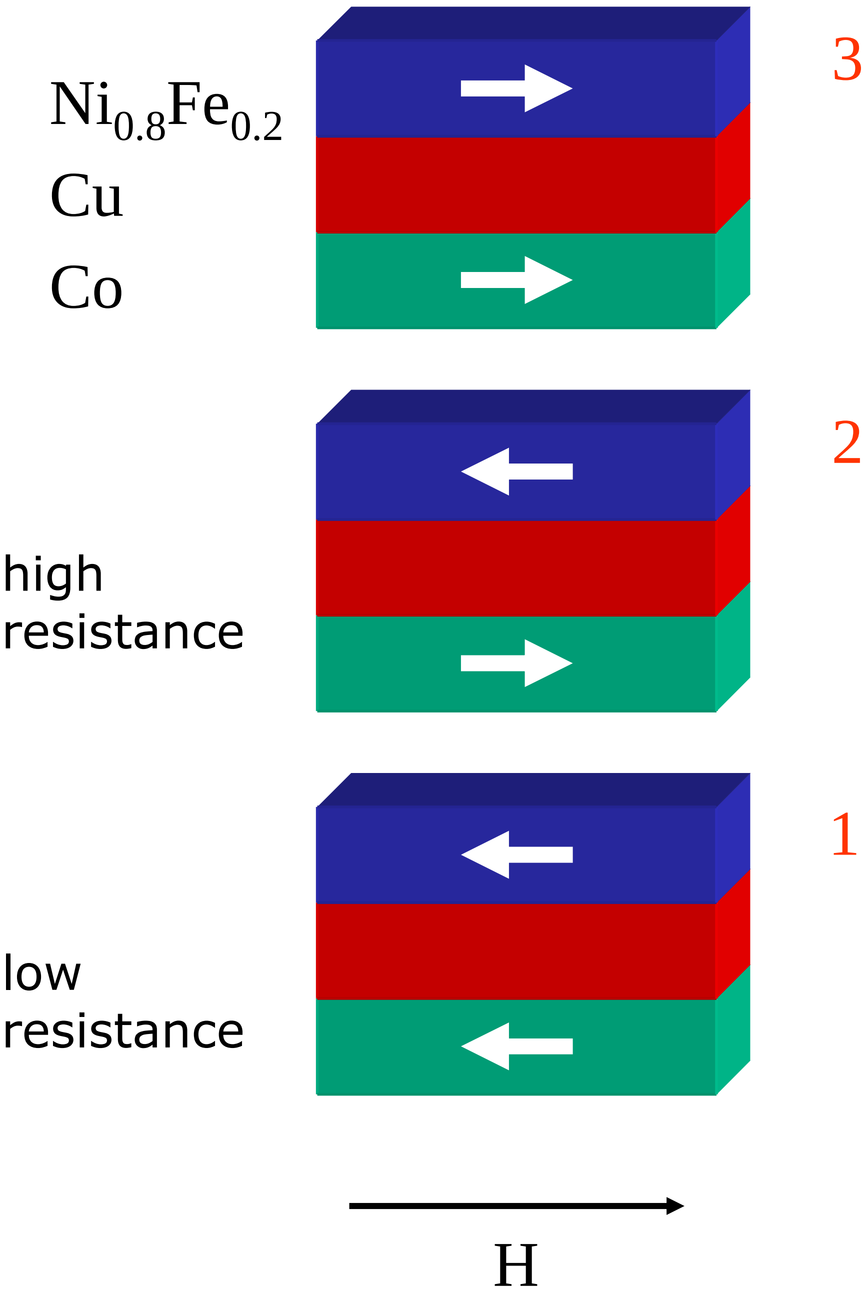


Example GMR device

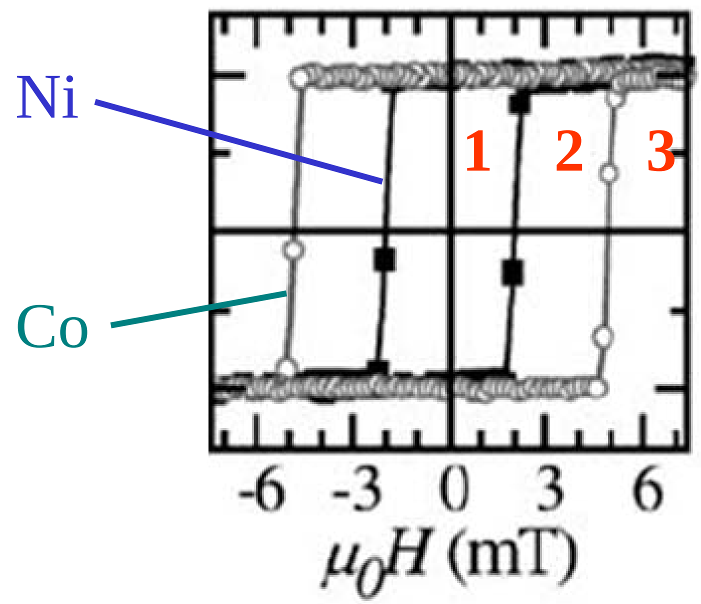


Kortright et al., JMMM 207, 7 (1999)

Example GMR device

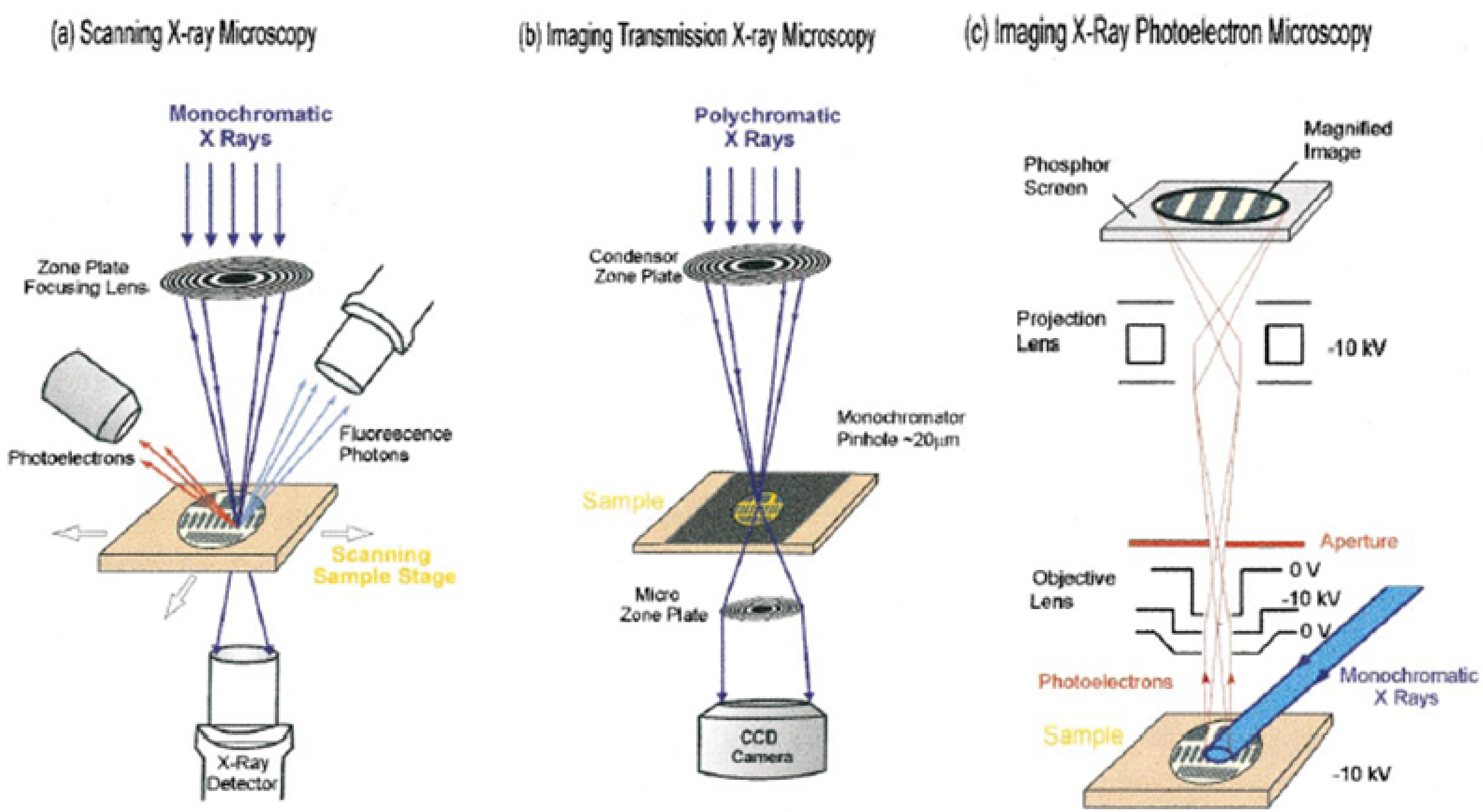


XMCD amplitude

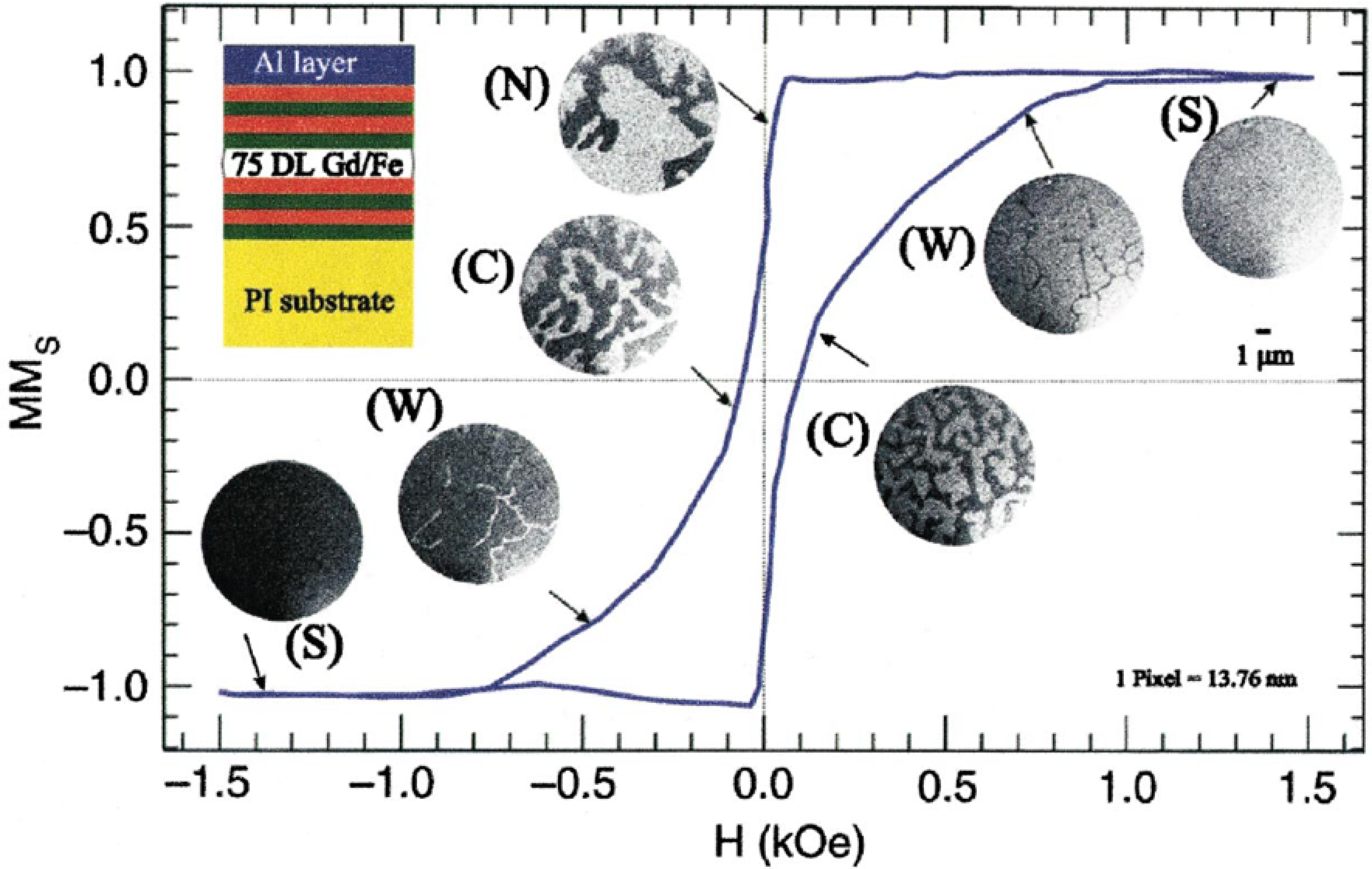


Bonfim et al., PRL 86, 3646 (2001)

XMCD microscopy

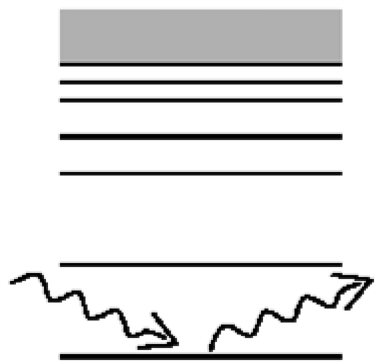


XMCD microscopy

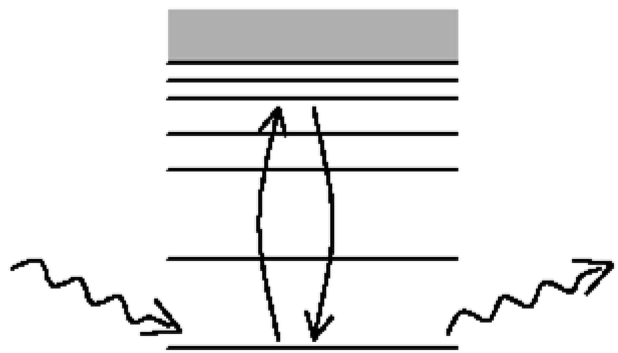


Resonant x-ray scattering

Thompson scattering



resonant (or "anomalous") scattering



form factor

$$f(Q,\omega)=\underbrace{f_0(Q)}_{\text{contribution of electrons with characteristic frequencies different from } \omega_0}+f'(Q,\omega)+i\,f''(Q,\omega)$$