Methoden moderner Röntgenphysik II: Streuung und Abbildung

Lecture 12	Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2017 G. Grübel, A. Philippi-Kobs, O. Seeck, T. Schneider, L. Frenzel, M. Martins, W. Wurth		
Location	Lecture hall AP, Physics, Jungiusstraße		
Date	Tuesday	12:30 - 14:00	(starting 4.4.)
	Thursday	8:30 - 10:00	(until 13.7.)

Resonant magnetic small angle X-ray scattering (mSAXS) of magnetic domain patterns

1.) Ferromagnetism in a nutshell

- forms of magnetic phenomena
- contributions to free energy
- focus on systems with perpendicular magnetic anisotropy (Co/Pt multilayers)
- magnetic domains and domain walls

2.) Interaction of **polarized** photons with matter

- Recap: Charge and **Spin** X-ray Scattering by a single electron
- Recap classical concept of Resonant Absorption & Scattering (forced oscillator)
- Resonant Absorption and Scattering (QM concept, Fermi's Golden rule)
- Interactions of photons with ferromagnetic materials ➔ XMCD effect

(- XMLinearD and X-ray Natural Dichroism)

3.) Resonant magnetic SAXS of magnetic domain patterns

Part II

Magnetism – Magnetic Thin Films

by André Philippi-Kobs (AP)

[23.5.] Magnetic small angle scattering of magnetic domain patterns

- Introduction of magnetism in thin films
- Resonant scattering & X-ray magnetic circular dichroism (XMCD),

[30.5.] Imaging of magnetic domains

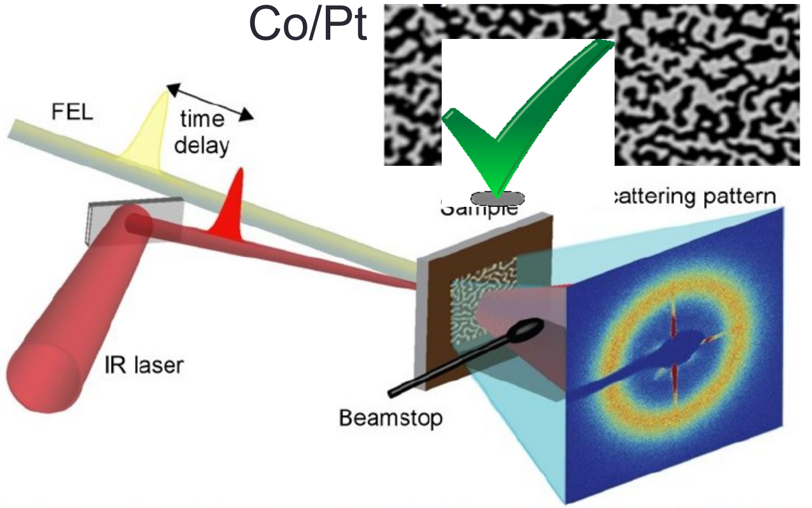
- Fourier transform holography (FTH)
- Scanning transmission X-ray microscopy (STXM)
- Coherent diffraction imaging (CDI), Ptychography

[1.6.] Femtomagnetism

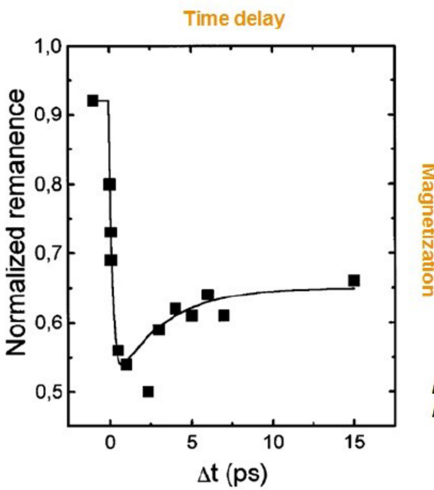
- Introduction of ultrafast magnetization dynamics
- Pump-probe experiments of nano-scale magnetic domain patterns

[13.6.] Related aspects

- Determination of coherence via magnetic domain patterns
- Magnetic XRD of antiferromagnets and chiral systems
- Further electronic inhomogeneities probed by X-rays (charge density wave; Abrikosov vortices in superconductors)



B. Pfau et al., Nature Communications, Vol. 3, 11; DOI:doi:10.1038/ncomms2108 (2012)
L.Müller et al., Rev. Sci. Instrum. 84, 013906 (2013)



Time-resolved MOKE (Ni 120 fs)

E. Beaurepaire et al., PRL 76 (1996) 4250

Interaction of **polarized** photons with matter

- Recap: Interaction of X-rays with matter (**consider also light's polarization ε**)

$$n(\omega,\boldsymbol{\varepsilon}) = 1 - \delta(\omega,\boldsymbol{\varepsilon}) + i\,\beta(\omega,\boldsymbol{\varepsilon})$$ Refractive index (classical refraction theory)

$$f(\mathbf{q},\omega,\boldsymbol{\varepsilon}) = f^0(\mathbf{q}) + \underbrace{f'(\omega,\boldsymbol{\varepsilon}) - if''(\omega,\boldsymbol{\varepsilon})}$$ Atomic scattering factor (scattering theory)

Atomic form factor
~ *Z* for forward scattering (or soft X-rays)

Anomalous scattering factors
(electrons are bound in a solid
➔ "resonances" at atomic transitions)

- Equivalence between scattering and refraction picture (lecture 4)

$$1 - n(\omega,\boldsymbol{\varepsilon}) = \frac{r_0\lambda^2}{2\pi}\rho f(\omega,\boldsymbol{\varepsilon})$$

Atomic density

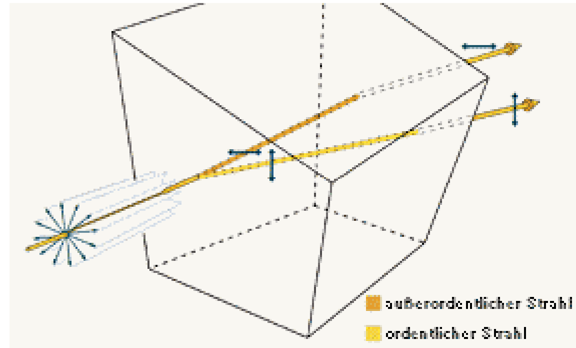
$$\delta(\omega,\boldsymbol{\varepsilon}) = \frac{r_0\lambda^2}{2\pi}\rho\,(Z + f'(\omega,\boldsymbol{\varepsilon}))$$

$$\beta(\omega,\boldsymbol{\varepsilon}) = \frac{r_0\lambda^2}{2\pi}\rho\, f''(\omega,\boldsymbol{\varepsilon})$$

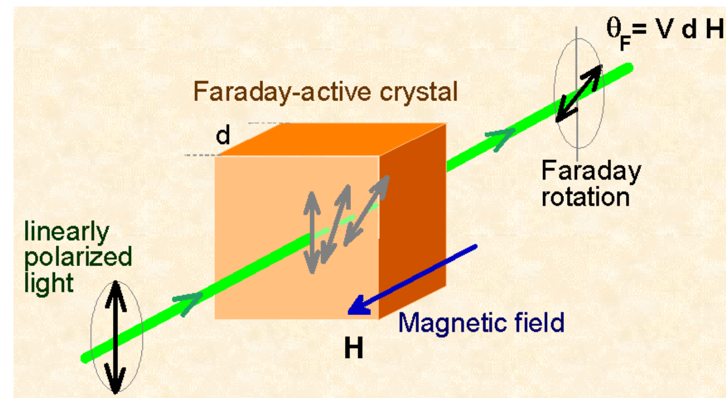
Interaction of **polarized** photons with matter

> Polarization **ε** dependent effects in transmission geometry

- The dependence of δ on **ε** is called birefringence (Doppelbrechung)



- The change of polarization **ε** is called optical rotation (Faraday effect in case of magnetic materials)



- The dependence of β on **ε** is called Dicroism (Zweifarbigkeit)
  - X-ray Natural (charge) linear dicroism (XNLD)
  - X-ray Natural (charge) circular dicroism (XNCD)
  - X-ray magnetic linear dicroism (XMLD)
  - X-ray magnetic circular dicroism (XMCD)

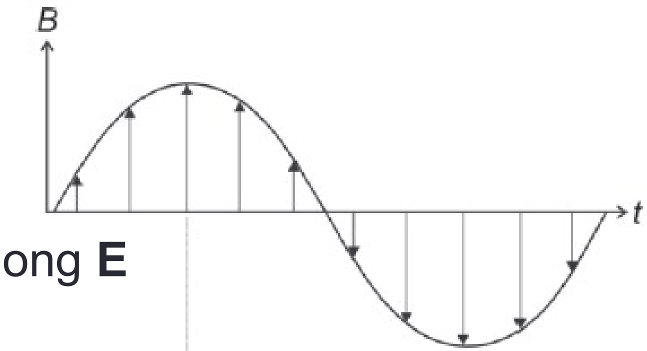
Interaction of **polarized** photons with matter

> Scattering of X-rays by a single electron (also **consider spin of electron**)

Incoming plane wave

$$E(r,t) = \epsilon\, E_0\, \mathrm{e}^{-\mathrm{i}(\omega t - \boldsymbol{k}\cdot\boldsymbol{r})}$$

$$B(r,t) = \frac{1}{c}(k_0 \times \epsilon) E_0\, \mathrm{e}^{-\mathrm{i}(\omega t - \boldsymbol{k}\cdot\boldsymbol{r})}$$

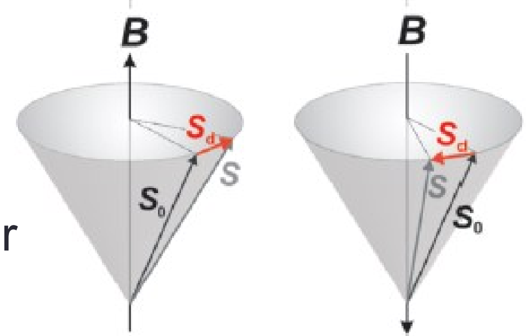


Electric dipole moment (charge movement) oscillates along **E**

$$p(t) = -\frac{e^2}{m_e \omega^2}\, E_0\, \mathrm{e}^{-\mathrm{i}\omega t}$$

Spin of electron precesses around magnetic field according to

$$\frac{d s_d(t)}{dt} = -\frac{e}{m_e}\, \vec{s}_0 \times \vec{B}(t)$$



g-factor

With definition of magnetic moment $m = -2\mu_B s_d$

Magnet dipole moment (spin movement) oscillates in the direction perpendicular to **B** and **s** (initial spin direction)

$$m(t) = \mathrm{i}\,\frac{e^2 \hbar \mu_0}{\omega m_e^2}\, s \times B_0\, \mathrm{e}^{-\mathrm{i}\omega t}$$

Interaction of **polarized** photons with matter

> Scattering by a single electron (also **consider Spin of electron**)

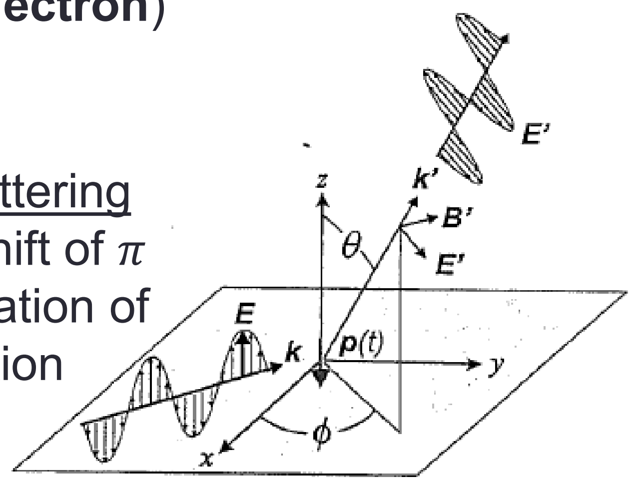
Electric fields radiated by

- electric dipole (Jackson text book):

$$\boldsymbol{E}'(t) = \frac{\omega^2}{4\pi\epsilon_0 c^2}\frac{\mathrm{e}^{\mathrm{i}k'r}}{r}\left[\boldsymbol{k}_0' \times \boldsymbol{p}(t)\right] \times \boldsymbol{k}_0'$$

$$\boldsymbol{E}'(t) = - \frac{1}{4\pi\epsilon_0}\frac{e^2}{m_e c^2}\frac{\mathrm{e}^{\mathrm{i}k'r}}{r}\underbrace{\left[\boldsymbol{k}_0' \times \boldsymbol{E}(t)\right] \times \boldsymbol{k}_0'}_{\vec{\varepsilon} \text{ for } \vec{k}_0 \parallel \vec{k}_0'}$$

Electric dipole scattering



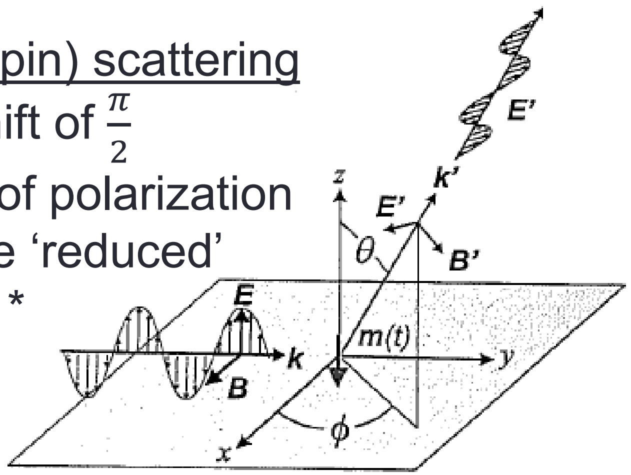
Charge scattering
- Phase shift of π
- Conservation of polarization

- magnetic dipole (Jackson text book):

$$\boldsymbol{E}'(t) = -\frac{\omega^2}{4\pi c}\frac{\mathrm{e}^{\mathrm{i}k'r}}{r}\left[\boldsymbol{k}_0' \times \boldsymbol{m}(t)\right]$$

$$\boldsymbol{E}'(t) = \mathrm{i}\frac{1}{4\pi\epsilon_0}\frac{e^2}{m_e c^2}\left(\frac{\hbar\omega}{m_e c^2}\right)^{*}\frac{\mathrm{e}^{\mathrm{i}k'r}}{r}\left[\boldsymbol{s} \times (\boldsymbol{k}_0 \times \boldsymbol{E}(t))\right] \times \boldsymbol{k}_0'$$

Magnetic dipole scattering



Magnetic (Spin) scattering
- Phase shift of $\frac{\pi}{2}$
- Rotation of polarization
- Amplitude 'reduced' by factor *

Interaction of **polarized** photons with matter

- Scattering by a single electron (**also consider Spin of electron**)

Polarization dependent scattering lengths: $$f(\epsilon,\epsilon') = -\frac{r e^{-\mathrm{i}k'r}}{E}\, E'\cdot\epsilon'$$

$$f_\mathrm{e}(\epsilon,\epsilon') = \frac{1}{4\pi\epsilon_0}\frac{e^2}{m_\mathrm{e}c^2}\,\epsilon\cdot\epsilon' = r_0\,\underbrace{\epsilon\cdot\epsilon'}\qquad f_\mathrm{s}(\epsilon,\epsilon') = -\mathrm{i}\,r_0\,\frac{\hbar\omega}{m_\mathrm{e}c^2}\,s\cdot(k_0\times\epsilon)\times(k_0'\times\epsilon')$$

Remember: Polariazion factor P = sinθ (lecture 2)

Differential scattering cross-section: $$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = |f(\epsilon,\epsilon')|^2$$

$$\frac{d\sigma}{d\Omega} = r_0^2 \sin^2\theta \text{ for } f_e$$

Total cross-section: $$\sigma_e = \int |f(\vec\epsilon,\vec\epsilon')|^2 d\Omega = r_0^2\int_0^{2\pi} \sin^2\theta \sin\theta\, d\theta \int_0^{\pi} d\varphi$$

$$\sigma_\mathrm{e} = \frac{8\pi}{3}\,r_0^2 = 0.665\times 10^{-28}\,\mathrm{m}^2 = 0.665\,\mathrm{barn}$$

$$\sigma_\mathrm{s} = \frac{8\pi}{3}\,\frac{1}{4}\left(\frac{\hbar\omega}{m_\mathrm{e}c^2}\right)^2 r_0^2 = \frac{\sigma_\mathrm{e}}{4}\left(\frac{\hbar\omega}{m_\mathrm{e}c^2}\right)^2$$

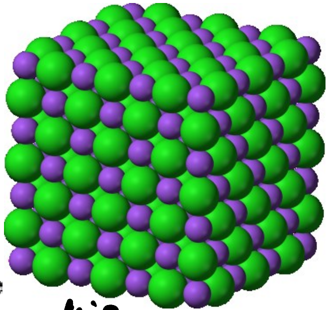
$E = 10\ \mathrm{keV} \rightarrow \frac{\sigma_s}{\sigma_e} = 0.0004$ Only weak spin-scattering signal

DESY

Interaction of **polarized** photons with matter

> Scattering by a single electron (**also consider Spin of electron**)

Example: magnetic XRD of antiferromagnetic NiO



NiO Ni

Gitterperiode
a,a,a
→ $\left(\frac{2\pi}{a}, \frac{2\pi}{a}, \frac{2\pi}{a}\right)$
hkl (111)
Spin-Struktur
(2a,2a,2a)
hkl (1/2,1/2,1/2)

Volume 39A, number 2 PHYSICS LETTERS 24 April 1972

OBSERVATION OF MAGNETIC SUPERLATTICE PEAKS BY X-RAY DIFFRACTION ON AN ANTIFERROMAGNETIC NiO CRYSTAL

F. De BERGEVIN and M. BRUNEL
Laboratoire de rayons-X, Cédex 166, 38–Grenoble-Gare, France

Received 14 February 1972

. We have searched and measured in the zone $\{hhh\}$ the first two superlattice magnetic reflections $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ and $(\frac{3}{2}\frac{3}{2}\frac{3}{2})$ and the first ordinary reflection (111). If an equal amount of all possible magnetic domains or twins is supposed to form the crystal, the formula (1) applied to these reflections gives a ratio R between magnetic and ordinary (111) intensities, approximately equal to 4×10^{-8}. Such a small value obliges to take an unusual care of obtaining a maximum intensity and a minimum background.

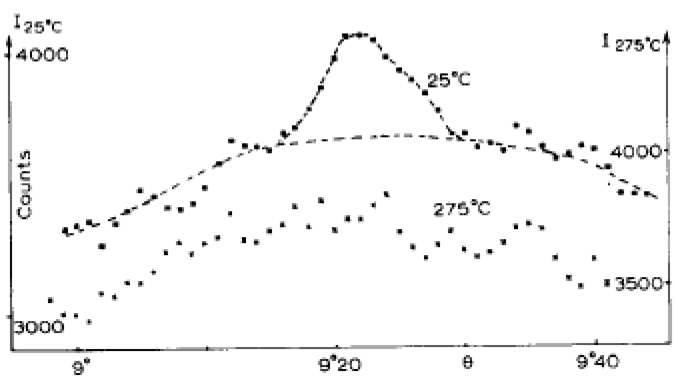


Fig. 1. Intensity $I_t(\theta)$ near the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ position at $t = 25°$ C and 275° C in counts/225 min. The hump which cover the interval could be due to some impurity.

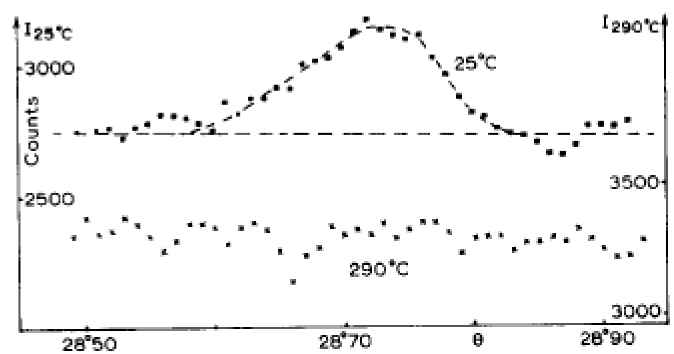


Fig. 2. Intensity $I_t(\theta)$ near the $(\frac{3}{2}\frac{3}{2}\frac{3}{2})$ position at $t = 25°$C and 290°C in counts/225 min.

Interaction of **polarized** photons with matter

> Recap from lecture 8: Absorption and Resonant Scattering (classical concept)

Picture: Electrons are bound to atoms

➔ Forced oscillator model with resonances ω_s and damping Γ to describe equation of motion of electrons

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

atomic scattering length f_s (in units of $-r_0$) for bound electron

note: $f_s \rightarrow 1$ $(\omega \gg \omega_s)$

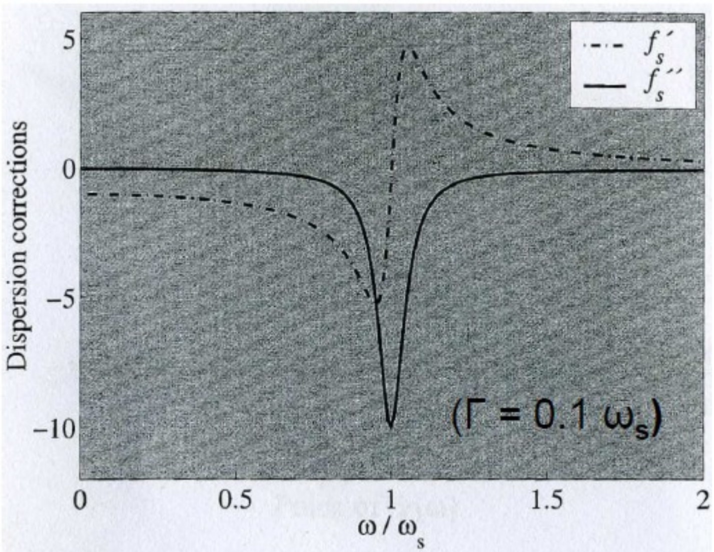
total cross-section: $\sigma_T = (8\pi/3)\, r_o^2$ (free electron)

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

(scattering cross-section)

Interaction of **polarized** photons with matter

> Recap from lecture 8: Absorption and Resonant Scattering (classical concept)



with:

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

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

$f'' = -(k/4\pi)\,\sigma_a\,(E)$ (optical theorem $2k\beta = \mu = \rho\sigma_a$)

$$\boxed{\sigma_{a,s}(\omega) = 4\,\pi\, r_0\, c\,\frac{\omega_s^2\,\Gamma}{(\omega^2 - \omega_s^2)^2 + (\omega\Gamma)^2}}$$

Measure absorption cross-section in experiment

Use Kramers-Kronig relations to obtain *f'*

$$f'(\omega) = \frac{1}{\pi}\, P \int_{-\infty}^{+\infty} \frac{f''(\omega')}{(\omega' - \omega)}\, d\omega'$$

$$f''(\omega) = -\frac{1}{\pi}\, P \int_{-\infty}^{+\infty} \frac{f'(\omega')}{(\omega' - \omega)}\, d\omega'$$

Interaction of **polarized** photons with matter

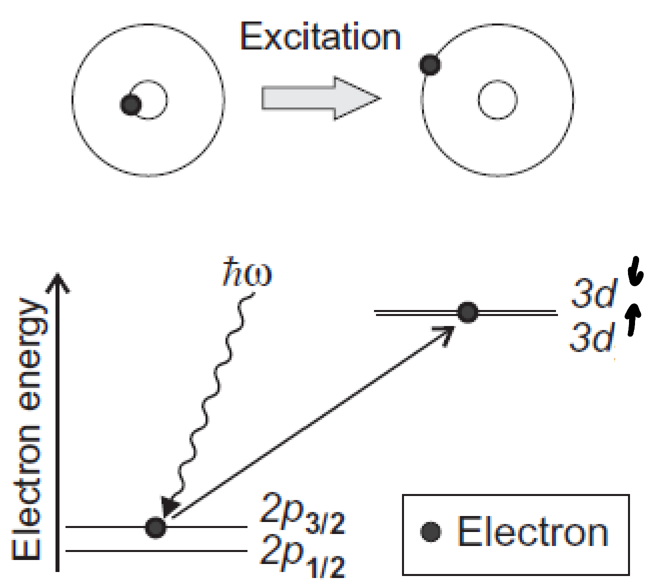
> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

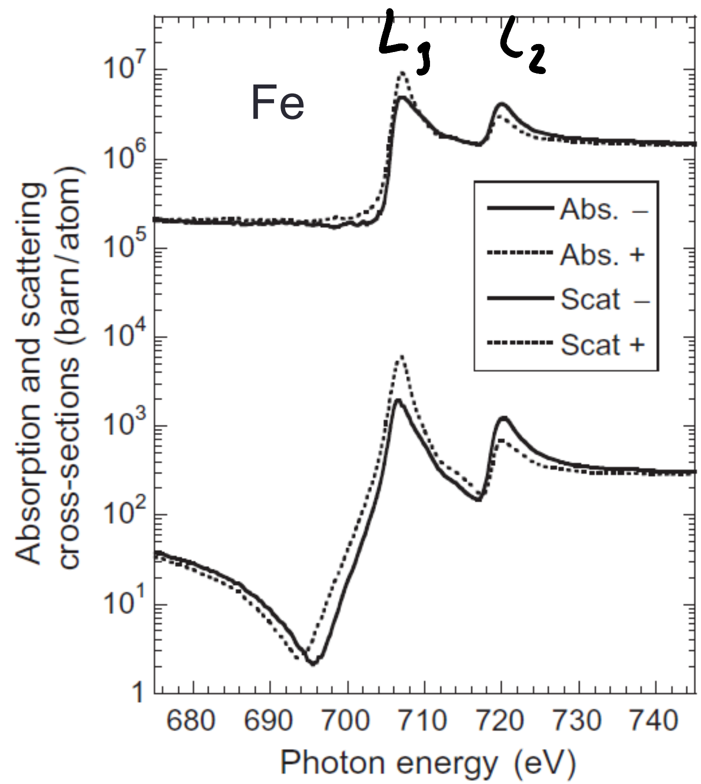
Motivation: Understand polarization dependent 2p ➔ 3d transition in ferromagnets, i.e., XMCD effect

$L_3: 2p_{3/2} \to 3d$

$L_2: 2p_{1/2} \to 3d$

($2p^1 \to 3d^1$ electron transition)





Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

- Time-dependent pertubation theory (up to second order) = „Fermi's Golden rule"

$$T_{if} = \frac{2\pi}{\hbar} \left| \underline{\langle f | \mathcal{H}_{\text{int}} | i \rangle} + \sum_n \frac{\langle f | \mathcal{H}_{\text{int}} | n \rangle \langle n | \mathcal{H}_{\text{int}} | i \rangle}{\varepsilon_i - \varepsilon_n} \right|^2 \delta(\varepsilon_i - \varepsilon_f) \rho(\varepsilon_f)$$

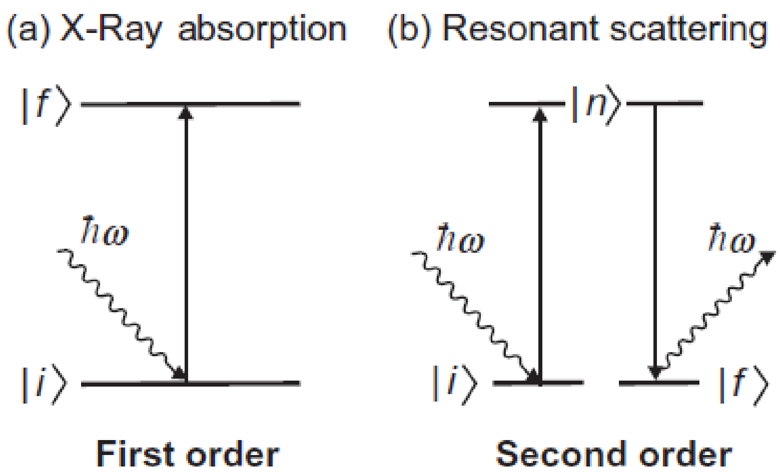
T_{if}: transition rate from state i to f; $[T_{if}] = s^{-1}$; i and f are initial and final states of the combined electron and photon system

$\rho(\varepsilon_f)$: density of final states

ε_n: energy of all possible intermediate states n

- Total cross-section given by $\sigma = \frac{T_{if}}{\Phi_0}$

Incident photon flux



Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

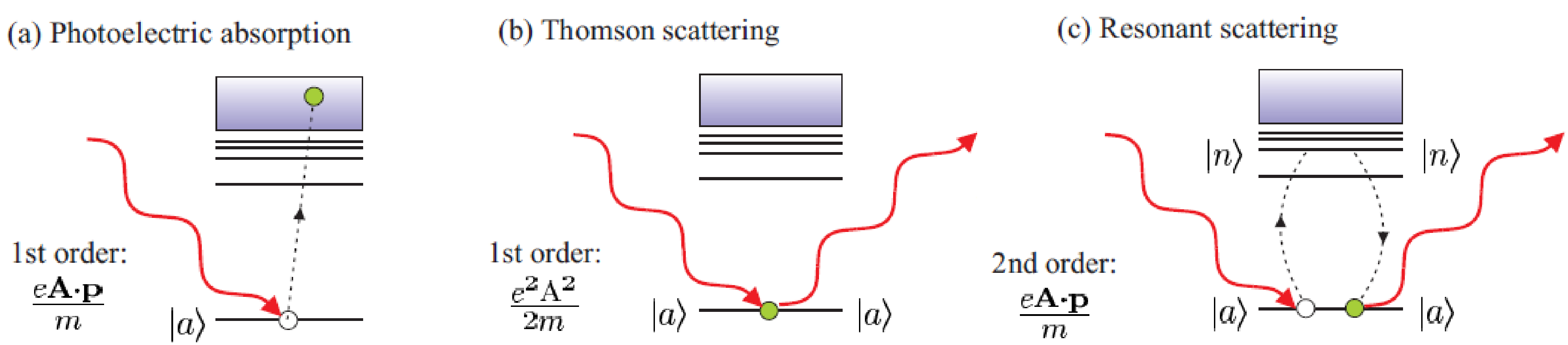
- Time-dependent pertubation theory (up to second order)

$$T_{if} = \frac{2\pi}{\hbar} \left| \langle f|\mathcal{H}_{\text{int}}|i\rangle + \sum_n \frac{\langle f|\mathcal{H}_{\text{int}}|n\rangle\langle n|\mathcal{H}_{\text{int}}|i\rangle}{\varepsilon_i - \varepsilon_n} \right|^2 \delta(\varepsilon_i - \varepsilon_f)\rho(\varepsilon_f)$$

- Interaction Hamiltonian
(derivation again via force on "atom in electric and magnetic field")

$$\mathcal{H}_{\text{e}}^{\text{int}} = \frac{e}{m_e}\mathbf{p}\cdot\mathbf{A} + \cancel{\frac{e^2A^2}{2m_e}}$$

p: momentum of electrons
A: vector potential



see begin of lecture

Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

- Time-dependent pertubation theory (up to second order)

$$T_{if} = \frac{2\pi}{\hbar} \left| \underbrace{\langle f|\mathcal{H}_{\text{int}}|i\rangle}_{\mathcal{M}} + \sum_n \frac{\langle f|\mathcal{H}_{\text{int}}|n\rangle\langle n|\mathcal{H}_{\text{int}}|i\rangle}{\varepsilon_i - \varepsilon_n} \right|^2 \delta(\varepsilon_i - \varepsilon_f)\rho(\varepsilon_f)$$

- Interaction Hamiltonian
  (derivation again via force on "atom in electric and magnetic field")

$$\mathcal{H}_{\text{e}}^{\text{int}} = \frac{e}{m_e}\mathbf{p}\cdot\mathbf{A}$$

p: momentum of electrons
A: vector potential

- Consider only the much stronger impact of electric field component of EM wave on electrons (begin of lecture: Charge scattering >> Spin scattering)

$$\vec{E} = -\frac{\partial \vec{A}}{\partial t}$$

with plane wave

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

$|a\rangle, |b\rangle$ atomic states

$$\mathcal{M} = \langle b|\, \vec{p}\cdot\vec{\varepsilon}\, e^{i\vec{k}\cdot\vec{r}}\, |a\rangle$$

Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

Matrix elements for atomic transitions between states a and b

$$\mathcal{M} = \langle b | \boldsymbol{p} \cdot \boldsymbol{\epsilon} \, e^{i\boldsymbol{k}\cdot\boldsymbol{r}} | a \rangle$$

Dipole-Approximation : ellimination of $\vec{k}$-dependence of $\mathcal{M}$

Expansion of $e^{i\vec{k}\cdot\vec{r}} = 1 + i\,\vec{k}\cdot\vec{r} - \frac{(\vec{k}\cdot\vec{r})^2}{2} + \dots$

size of e^--Radius : $|\vec{r}| = 0.1\,\text{Å}$ for p-core shells

soft X-ray $E_\gamma \leq 1\,\text{keV} \rightarrow \lambda \geq 1\,\text{nm}$

$|\vec{k}| = \frac{2\pi}{\lambda} \leq 5 \cdot 10^9\,\text{m}^{-1}$

$|\vec{k}||\vec{r}| \leq 0.05 \ll 1 \rightarrow e^{i\vec{k}\cdot\vec{r}} \approx 1$

$\rightarrow \mathcal{M} = \langle b | \vec{p} \cdot \vec{\varepsilon} | a \rangle$ dipole approximation

Interaction of **polarized** photons with matter

- Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

Matrix elements for atomic transitions between states a and b

$$\mathcal{M} \simeq \langle b|\, \boldsymbol{p} \cdot \boldsymbol{\epsilon} |a\rangle$$

$\vec{p} \rightarrow \vec{r}$ ("length operator")

Reformulation of Matrix-elements

via commutation relation $\vec{p} = \frac{mi}{\hbar} [\mathcal{H}_0, \vec{r}]$

$$\mathcal{M} = \langle b | \vec{p} \cdot \vec{\varepsilon} | a \rangle = \frac{mi}{\hbar} \langle b | [\mathcal{H}, \vec{r}] \vec{\varepsilon} | a \rangle$$

$$= \frac{mi}{\hbar} \left(\langle b | \mathcal{H} \cdot \vec{r} \, \vec{\varepsilon} | a \rangle - \langle b | \vec{r} \, \mathcal{H} \vec{\varepsilon} | a \rangle \right)$$

$$= \frac{mi}{\hbar} \left(E_b \langle b | \vec{r} \cdot \vec{\varepsilon} | a \rangle - E_a \langle b | \vec{r} \cdot \vec{\varepsilon} | a \rangle \right)$$

$$= \frac{mi}{\hbar} (E_b - E_a) \langle b | \vec{r} \cdot \vec{\varepsilon} | a \rangle = mi\omega \langle b | \vec{r} \cdot \vec{\varepsilon} | a \rangle$$

$[\vec{\varepsilon}, \vec{r}] = 0$

$[\mathcal{H}, \vec{r}] = 0$

Absorption cross-section in dipole approximation

$$\sigma^{\mathrm{abs}} = 4\pi^2 \frac{e^2}{4\pi\epsilon_0 \hbar c} \hbar\omega \, |\langle b| \, \boldsymbol{\epsilon} \cdot \boldsymbol{r} |a\rangle|^2 \, \delta[\hbar\omega - (E_b - E_a)] \, \rho(E_b)$$

Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

Polarization dependent Dipole Operator $P = \boldsymbol{\epsilon} \cdot \boldsymbol{r}$

Electron position vector or length operator $\mathbf{r} = x\mathbf{e}_x + y\mathbf{e}_y + z\mathbf{e}_z$

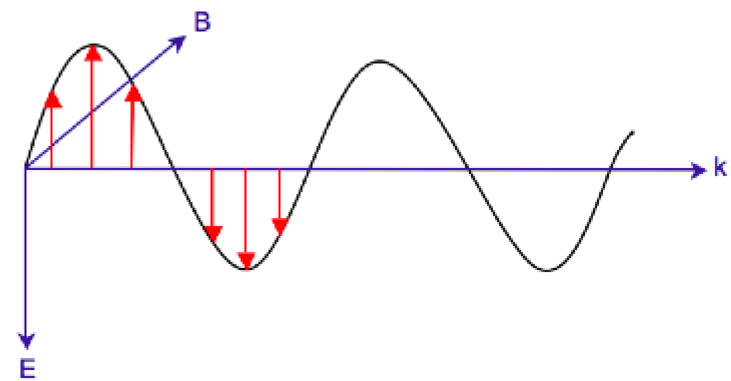
Linear polarized light $\epsilon_x^0 = \epsilon_x = e_x \quad \epsilon_y^0 = \epsilon_y = e_y \quad \epsilon_z^0 = \epsilon_z = e_z$

$$P_z^0 = \vec{\varepsilon}_z \cdot \vec{r} = z = r\cos\theta$$

$$= r\sqrt{\frac{4\pi}{3}}\, Y_{1,0}$$

$Y_{l,m}$: "Kugelflächenfkt"
spherical harmonics

Linearly polarized



Interaction of **polarized** photons with matter

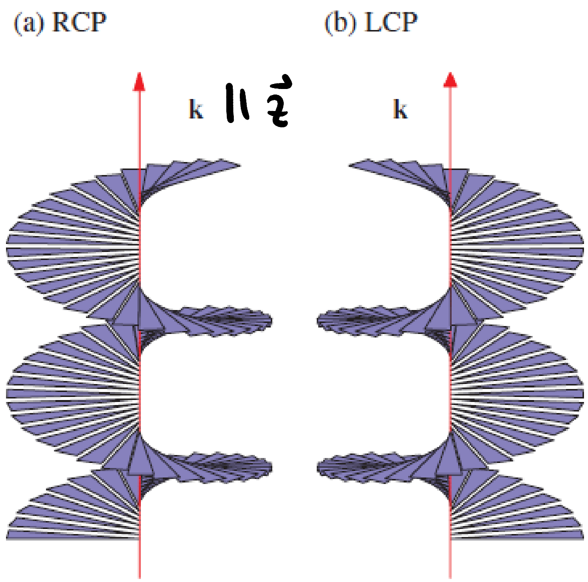
- Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

Polarization dependent Dipole Operator $P = \boldsymbol{\epsilon} \cdot \boldsymbol{r}$

Circular polarized light $(\vec{k} \parallel \vec{z})$

$$\vec{\mathcal{E}}_z = \pm\frac{1}{\sqrt{2}}\, \vec{\mathcal{E}}_x \pm i\, \vec{\mathcal{E}}_y \qquad (i = e^{i\pi/2})$$

$\vec{k}$-direction



Definition of Helicity
(photon angular momentum or spin $\mathbf{L}_{ph,z} \parallel \mathbf{z}$):

$$|\mathbf{L}_{ph,z}| = \pm\, q\, h$$

"+": q = +1 right circularly polarized light (RCP)
"-": q = -1 left circularly polarized light (LCP)
("0": q = 0 lin. pol. Light)

DESY

Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

→ Polarization dependent Dipole Operator for circularly polarized light:

$$P_z^{\pm} = \vec{\varepsilon}_z^{\,\pm} \cdot \vec{r} = \mp \frac{1}{\sqrt{2}} (x \pm iy) = \mp r \sin\theta \, e^{\pm i\phi}$$
$$= r \sqrt{\frac{4\pi}{3}} \, Y_{\overset{l}{1},\overset{m_l}{\pm 1}}$$

Racah's spherical tensor operators are defined as [181],

$$C_m^{(l)} = \sqrt{\frac{4\pi}{2l+1}} \, Y_{l,m}(\theta,\phi) \, , \qquad \left(C_m^{(l)}\right)^* = (-1)^m C_{-m}^{(l)} \, .$$

Dipol operator : $P_z^0 = r \, C_0^{(1)}$ lin pol.

$P_z^{\pm} = r \, C_{(\pm 1)}^{(1)}$ RCP(+) , LCP(−)

Interaction of **polarized** photons with matter

> Absorption (qm concept, Fermi's Golden rule)

➔ Transition-Matrix-Elements with atomic wave functions (non-relativistic approx.)

$$|a\rangle = |R_{n,l}(r);\, l, m_l;\, s, m_s\rangle$$

$$|b\rangle = |R_{n',l'}(r);\, l', m_l';\, s', m_s'\rangle$$

$$\langle b|P_z^q|a\rangle = \underbrace{\delta(m_s, m_s')}_{\text{spin}}\ \underbrace{\langle R_{n',l'}(r)|r|R_{n,l}(r)\rangle}_{\text{radial}} \cdot \underbrace{\sum_{m_l, m_l', q} \langle l', m_l'|C_q^{(1)}|l, m_l\rangle}_{\text{angular}}$$

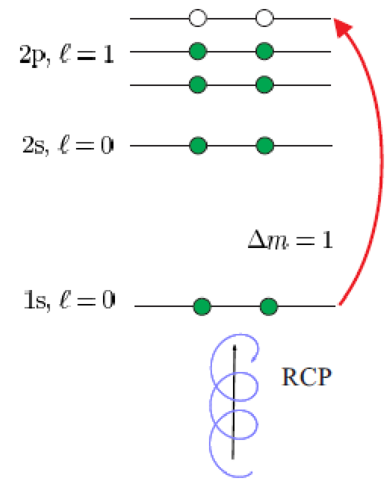
spin: electron spin direction is conserved

radial: transition strength : $2p \to 3d$ only considered in lecture
↳ the same strength for all transitions $2p \to 3d$!

Interaction of **polarized** photons with matter

> Absorption (qm concept, Fermi's Golden rule)

(a) Simplified energy level diagram



Non-vanishing matrix elements (use in today's excercise)

Table 1 Nonvanishing angular momentum dipole matrix elements $\langle L, M | C_q^{(1)} | l, m\rangle$. The matrix elements are real, so that $\langle L, M | C_q^{(1)} | l, m\rangle^* = \langle L, M | C_q^{(1)} | l, m\rangle = (-1)^q \langle l, m | C_{-q}^{(1)} | L, M\rangle$. Nonlisted matrix elements are zero.[a]

Lin. pol

$$\langle l+1, m | C_0^{(1)} | l, m\rangle = \sqrt{\frac{(l+1)^2 - m^2}{(2l+3)(2l+1)}}$$

$$\langle l-1, m | C_0^{(1)} | l, m\rangle = \sqrt{\frac{l^2 - m^2}{(2l-1)(2l+1)}}$$

RCP

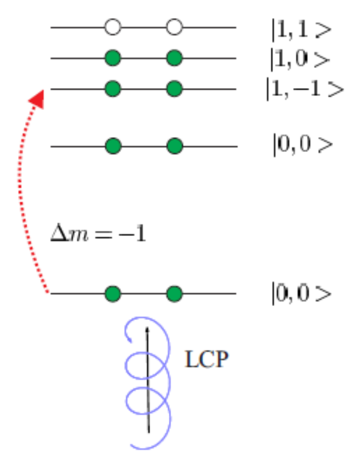
$$\langle l+1, m+1 | C_1^{(1)} | l, m\rangle = \sqrt{\frac{(l+m+2)(l+m+1)}{2(2l+3)(2l+1)}}$$

$$\langle l-1, m+1 | C_1^{(1)} | l, m\rangle = -\sqrt{\frac{(l-m)(l-m-1)}{2(2l-1)(2l+1)}}$$

LCP

$$\langle l+1, m-1 | C_{-1}^{(1)} | l, m\rangle = \sqrt{\frac{(l-m+2)(l-m+1)}{2(2l+3)(2l+1)}}$$

$$\langle l-1, m-1 | C_{-1}^{(1)} | l, m\rangle = -\sqrt{\frac{(l+m)(l+m-1)}{2(2l-1)(2l+1)}}$$



Interaction of **polarized** photons with matter

> Absorption (qm concept, Fermi's Golden rule)

Dipole selection rules (for states of the form $|n, l, m_l, s, m_s\rangle$)

$$\Delta l = l' - l = \pm 1$$

$$\Delta m_l = m_l' - m_l = q = 0, \pm 1$$

Helizität

$$\Delta s = s' - s = 0$$

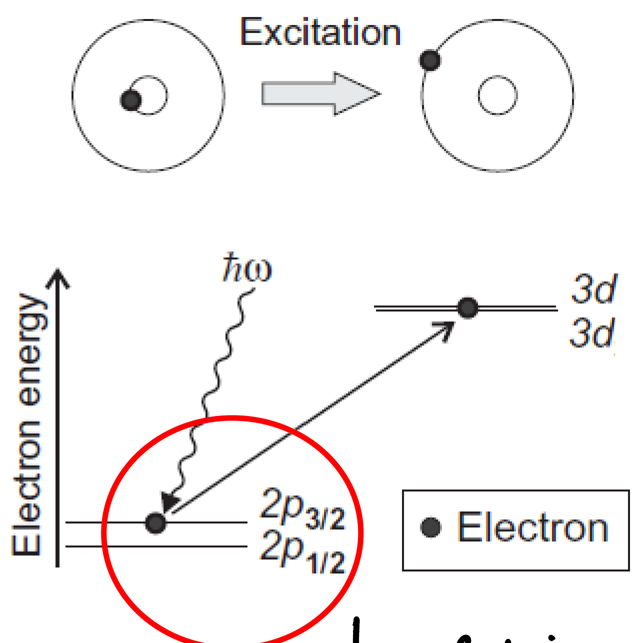
$$\Delta m_s = m_s' - m_s = 0$$

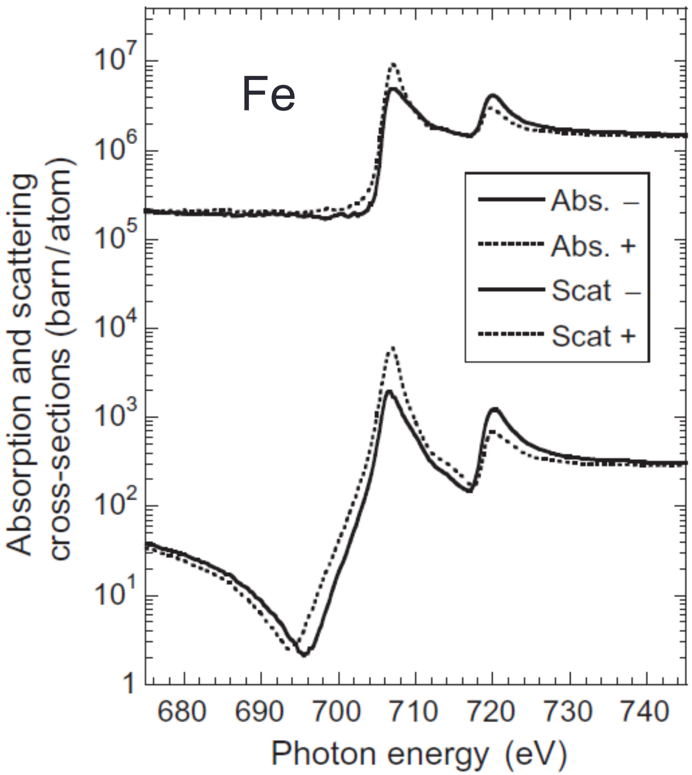
Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

<u>Motivation</u>: Understand polarization dependent 2p ➔ 3d transition in ferromagnets, i.e., XMCD effect

($2p^1 \rightarrow 3d^1$ electron transition)





SO1 → $|n, l, s, j, m_j\rangle$

Interaction of **polarized** photons with matter

- Absorption (qm concept, Fermi's Golden rule)

Atomic core shell states are split due to spin-orbit split interaction (use in today's lecture)
➔ Clebsch-Gordon coefficients *C*

$$|l,s,j,m_j\rangle = \sum_{m_l,m_s} C_{m_l,m_s;j,m_j}\,|l,s,m_l,m_s\rangle$$

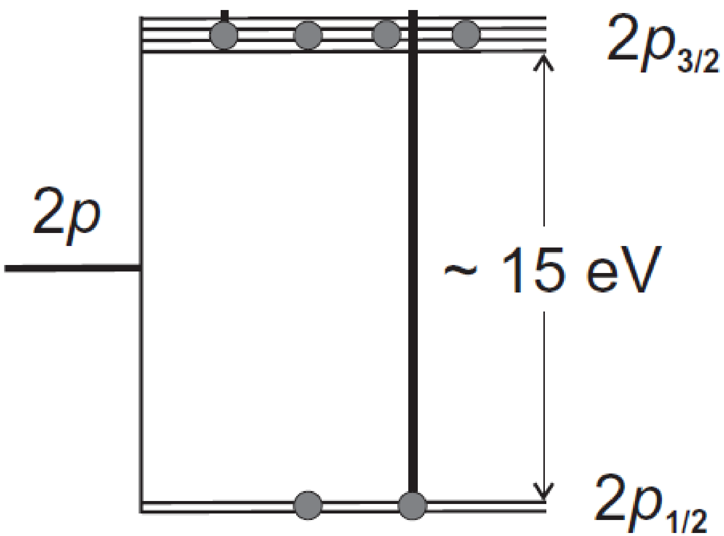


Table 2

$\|l,s,j,m_j\rangle$ basis		$\|l,m_l,s,m_s\rangle$ basis
j	m_j	$Y_{l,m_l}\chi^{\pm}$
$\frac{1}{2}$	$+\frac{1}{2}$	$\frac{1}{\sqrt{3}}(-Y_{1,0}\,\alpha + \sqrt{2}\,Y_{1,+1}\,\beta)$
	$-\frac{1}{2}$	$\frac{1}{\sqrt{3}}(-\sqrt{2}\,Y_{1,-1}\,\alpha + Y_{1,0}\,\beta)$
$\frac{3}{2}$	$+\frac{3}{2}$	$Y_{1,+1}\,\alpha$
	$+\frac{1}{2}$	$\frac{1}{\sqrt{3}}(\sqrt{2}\,Y_{1,0}\,\alpha + Y_{1,+1}\,\beta)$
	$-\frac{1}{2}$	$\frac{1}{\sqrt{3}}(Y_{1,-1}\,\alpha + \sqrt{2}\,Y_{1,0}\,\beta)$
	$-\frac{3}{2}$	$Y_{1,-1}\,\beta$

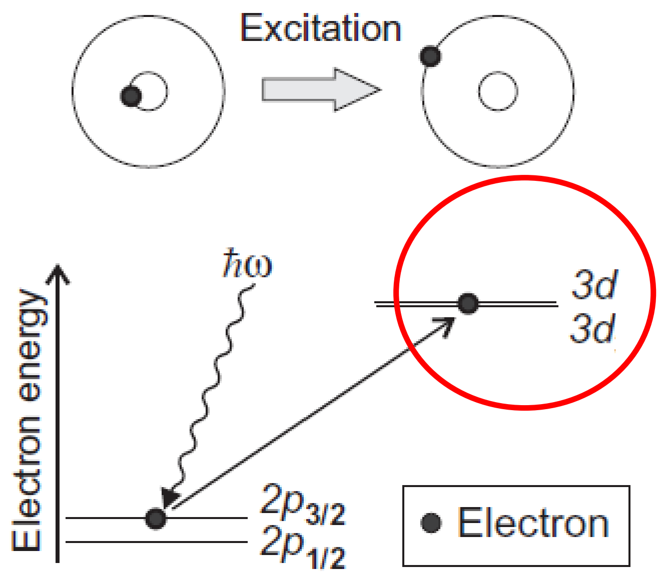
$\alpha \hat{=} \uparrow$
$\beta \hat{=} \downarrow$

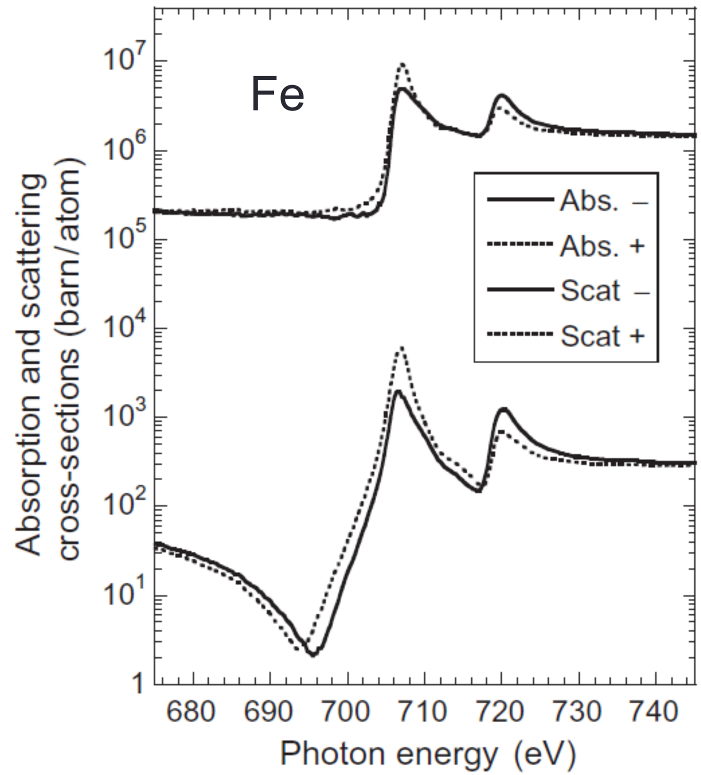
Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

Motivation: Understand polarization dependent 2p ➔ 3d transition in ferromagnets, i.e., XMCD effect

($2p^1 \rightarrow 3d^1$ electron transition)



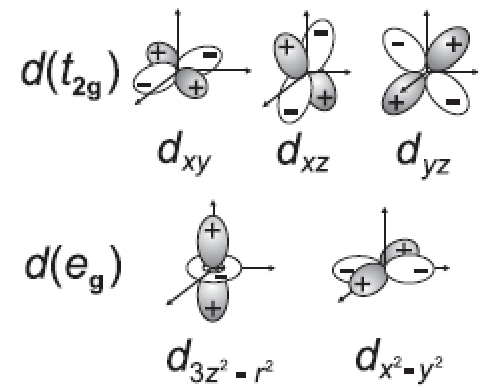


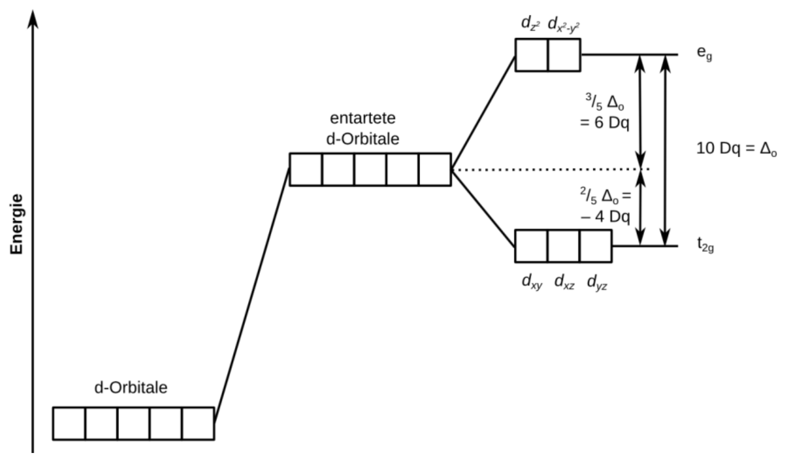
d-states: SOI negligible small but crystal field splitting

Interaction of **polarized** photons with matter

> Absorption (qm concept, Fermi's Golden rule)

Itinerant *d*-states are split due to crystal field
(can be neglected to a good approximation as splitting is small
➔ use atomic wave functions without SOC; today's lecture)





crystal field split d-states

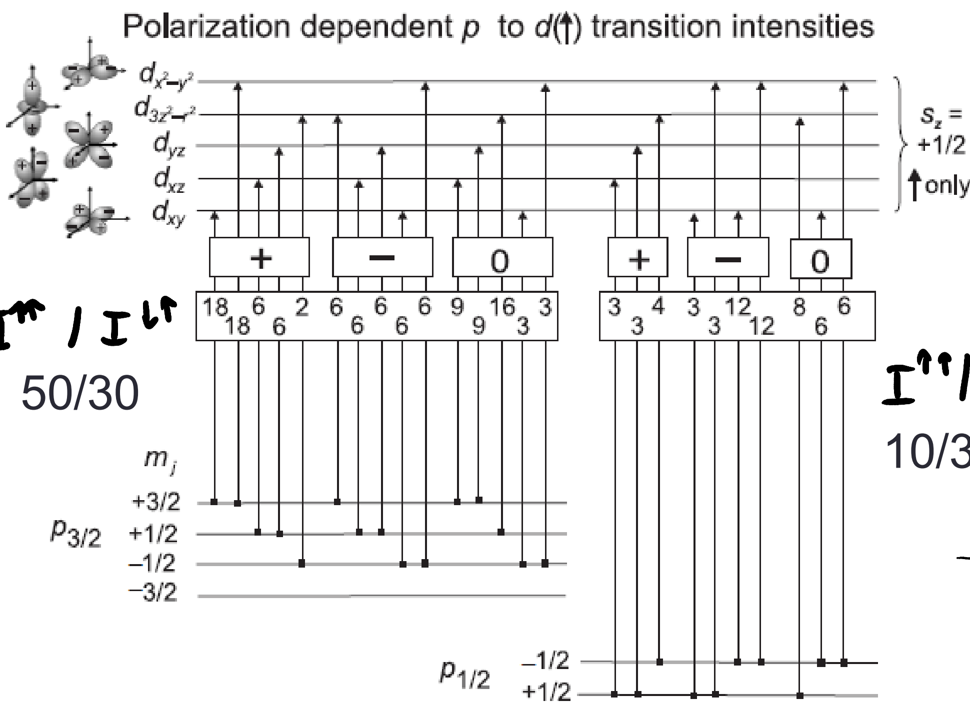
$|l, m_l\rangle$ basis

$$
\begin{array}{llll}
d_{xy} & = \sqrt{\frac{15}{4\pi}\frac{xy}{r^2}} & = & \frac{i}{\sqrt{2}}(Y_{2,-2} - Y_{2,+2}) \\
d_{xz} & = \sqrt{\frac{15}{4\pi}\frac{xz}{r^2}} & = & \frac{1}{\sqrt{2}}(Y_{2,-1} - Y_{2,+1}) \\
d_{yz} & = \sqrt{\frac{15}{4\pi}\frac{yz}{r^2}} & = & \frac{i}{\sqrt{2}}(Y_{2,-1} + Y_{2,+1}) \\
d_{x^2-y^2} & = \sqrt{\frac{15}{16\pi}\frac{(x^2-y^2)}{r^2}} & = & \frac{1}{\sqrt{2}}(Y_{2,-2} + Y_{2,+2}) \\
d_{3z^2-r^2} & = \sqrt{\frac{5}{16\pi}\frac{(3z^2-r^2)}{r^2}} & = & Y_{2,0}
\end{array}
$$

Interaction of **polarized** photons with matter

> X-ray magnetic circular dicroism (XMCD) effect

Crystal-field-split-d-states



$I^{\uparrow\uparrow} / I^{\downarrow\uparrow}$

50/30

- Strong ferromagnet: one subband is completely filled
- Spin in conserved during transition

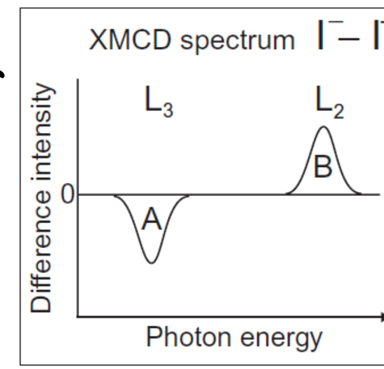
➔ Calculate transition matrix elements for **Spin-Up** electrons & helicity q = ± 1 (RCP and LCP)

$I^{\uparrow\uparrow} / I^{\downarrow\uparrow}$

10/30

➔ XMCD: m_s q

$\Delta I = I^{\uparrow\downarrow} - I^{\uparrow\uparrow} \neq 0$



$$\frac{I_{L_3\,\text{total}}}{I_{L_2\,\text{total}}} = \frac{80}{40} = 2:1$$

$$\Delta I_{L_3} = A R^2 \sum_{n,m_j} |\langle d_n, \chi^+ | C^{(1)}_{-1} | p_{3/2}, m_j \rangle|^2 - |\langle d_n, \chi^+ | C^{(1)}_{+1} | p_{3/2}, m_j \rangle|^2 \quad \propto 20$$

$$\Delta I_{L_2} = A R^2 \sum_{n,m_j} |\langle d_n, \chi^+ | C^{(1)}_{-1} | p_{1/2}, m_j \rangle|^2 - |\langle d_n, \chi^+ | C^{(1)}_{+1} | p_{1/2}, m_j \rangle|^2 \quad \propto 20$$

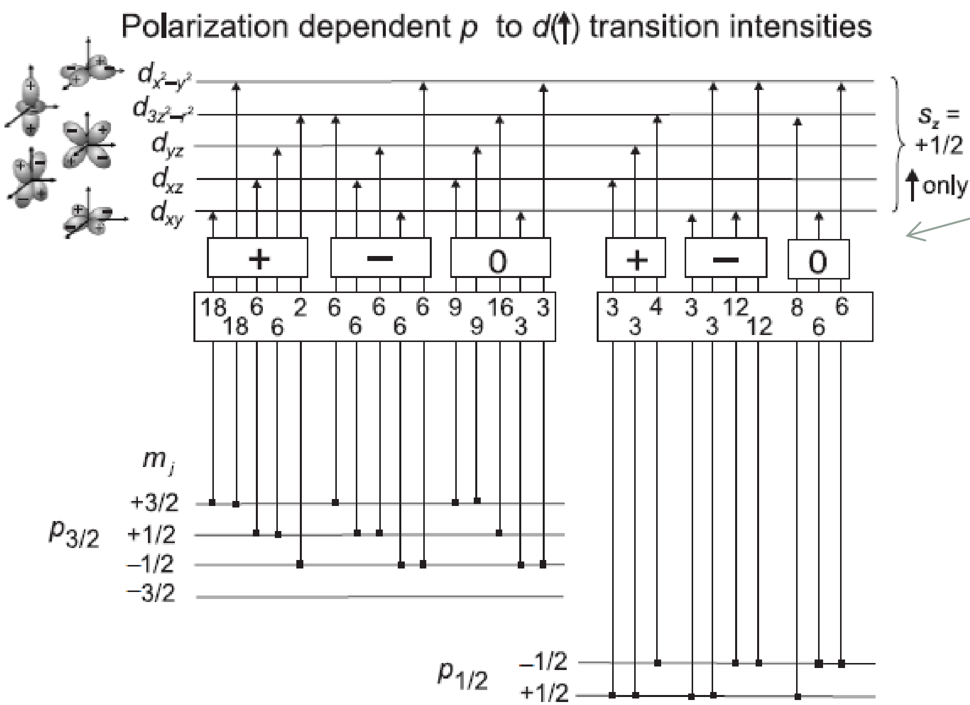
$\Big\}\ \Delta I_{L_3} = -\Delta L_{L_2}$

DESY

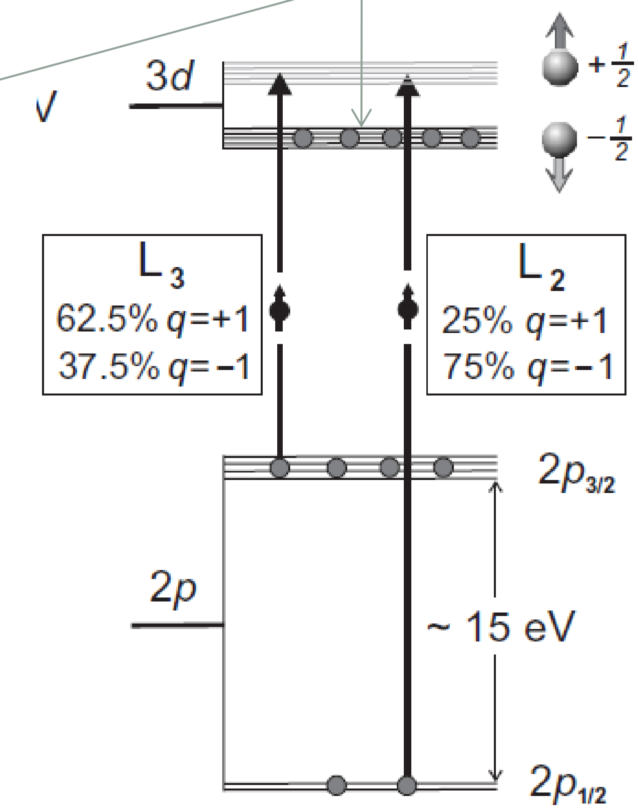
Interaction of **polarized** photons with matter

> <u>X-ray magnetic circular dicroism (XMCD) effect</u>

Strong ferromagnet:
One subband is
Completely filled



Crystal-field-split-d-states



Atomic-exchange-split d-states (w/o SOC)

Same results for $I_{L_{3,total}} : I_{L_{2,total}} = 2:1,$

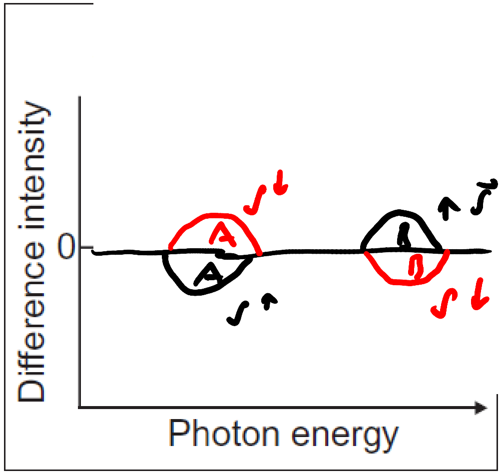
$$\Delta I_{L_{3,total}} : \Delta I_{L_{2,total}} = 1:-1$$

when using atomic d-states (w/o SOC); today's lecture

Interaction of **polarized** photons with matter

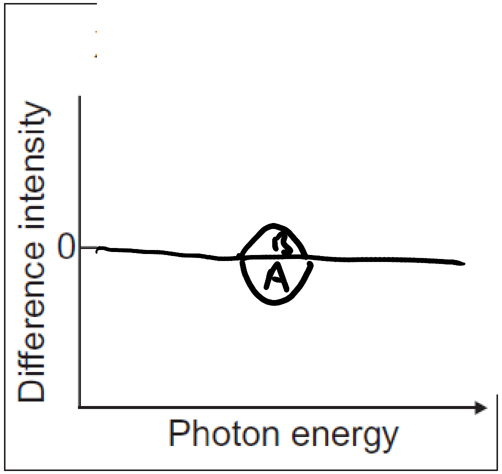
> X-ray magnetic circular dicroism (XMCD) effect

What is happening in a paramagnet?



→ No XMCD

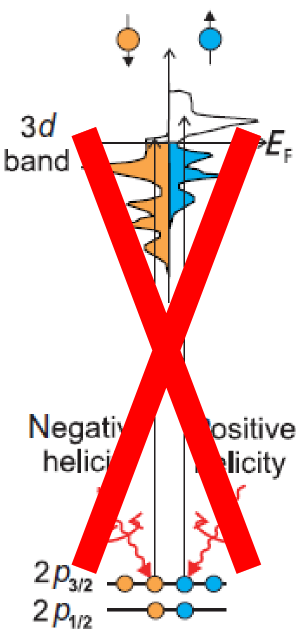
What is happening w/o Spin-Orbit-Coupling for the p-states?

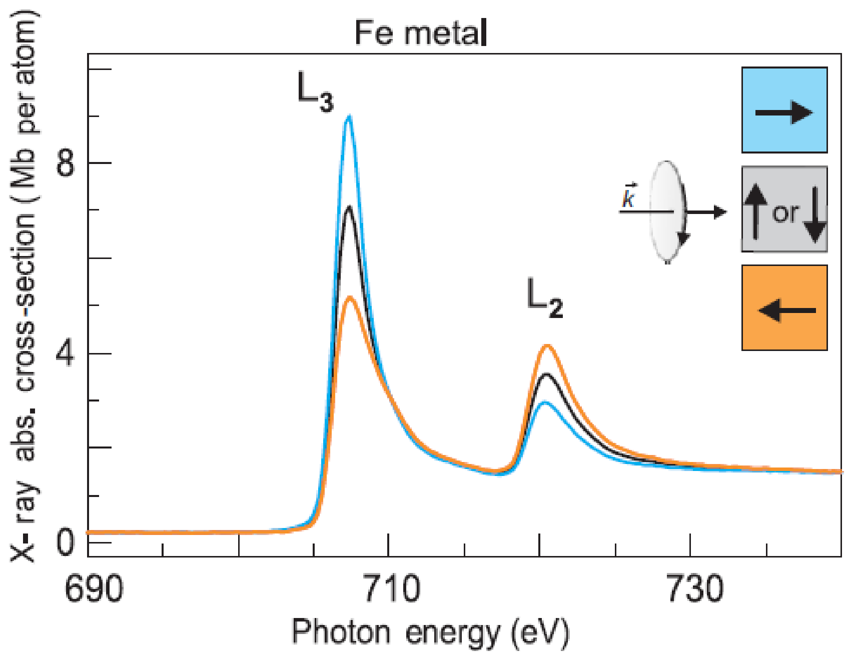


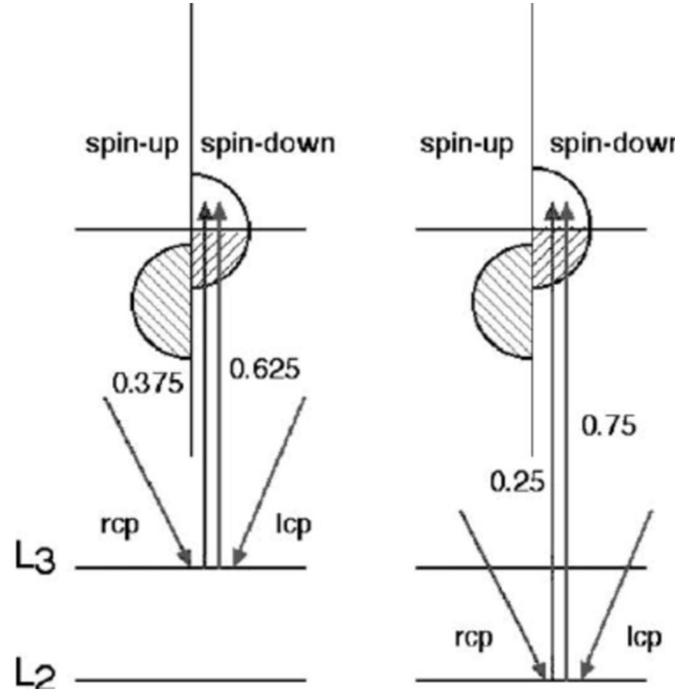
→ No XMCD

Interaction of **polarized** photons with matter

> X-ray magnetic circular dicroism (XMCD) effect







(sketch in textbooks can be misleading!)

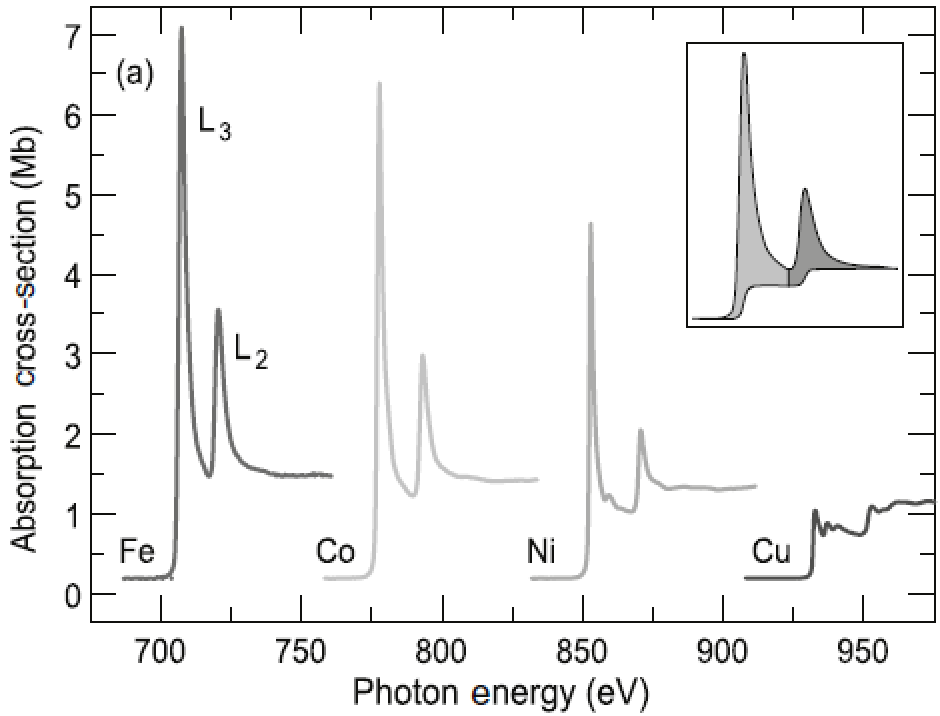
$$\Delta I_{XMCD} \propto \vec{M} \cdot \vec{L}_{ph} \propto \cos\Theta$$

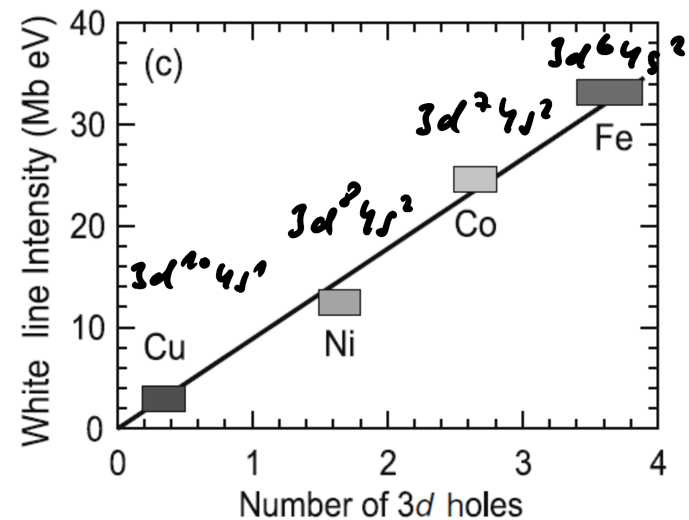
$$\Theta \sphericalangle (\vec{M}, \vec{L}_{ph})$$

Interaction of **polarized** photons with matter

> (Orientation averaged) Sum rules $$\langle I \rangle = \frac{1}{3}\left(I_\alpha^{-1} + I_\alpha^{0} + I_\alpha^{+1}\right) \qquad (\alpha = z)$$

Density of d-states at E_F $$\sigma^{\mathrm{abs}} = 4\pi^2 \frac{e^2}{4\pi\epsilon_0 \hbar c} \hbar\omega \, |\langle b|\, \epsilon \cdot r \,|a\rangle|^2 \, \delta[\hbar\omega - (E_b - E_a)] \, \rho(E_b)$$





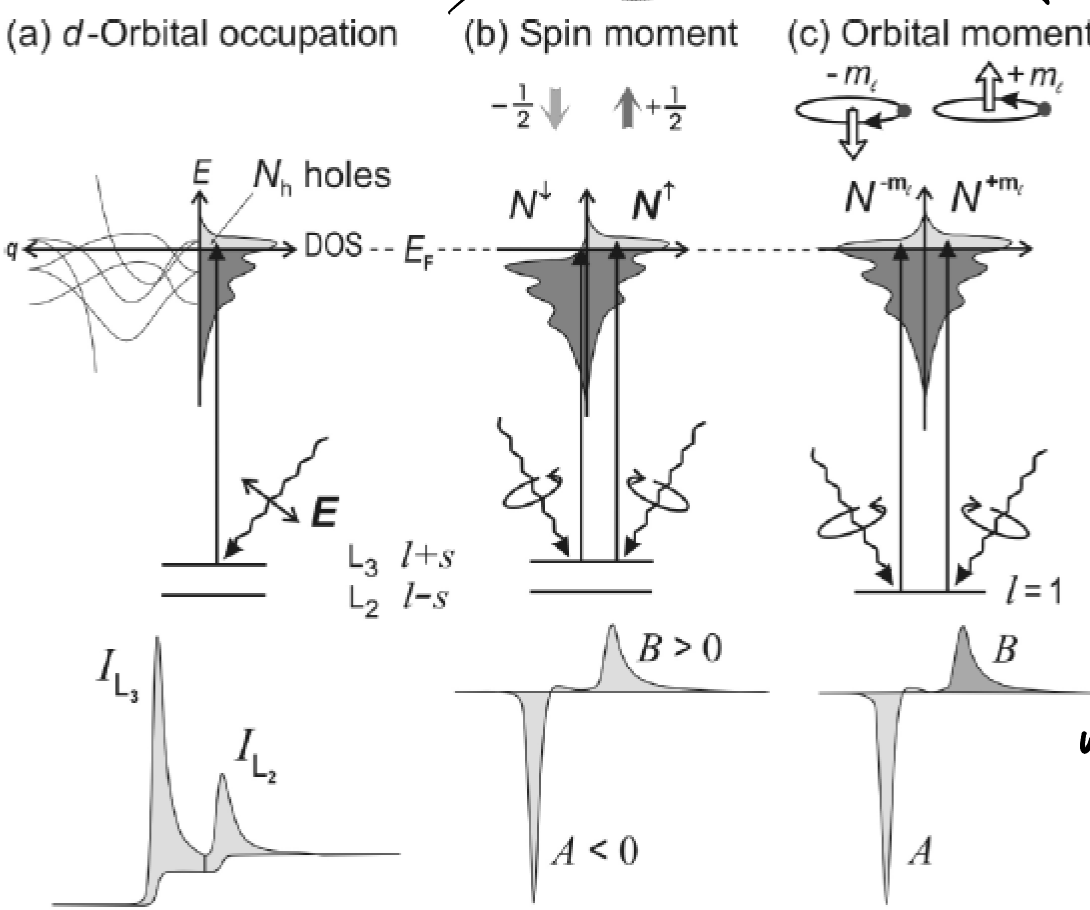
$$D_d(E_F) = \frac{\langle I_{L_3} + I_{L_2} \rangle}{C}$$

$p \to s$ transitions have to be considered as well but as $D_s(E_F) \ll D_d(E_F)$ $p \to d$ channels dominate!

Interaction of **polarized** photons with matter

- (Orientation averaged) Sum rules

XMCD



→ as SOC (~50 meV) and $\vec{L}$ exist for d-e⁻!
$m_l \ll m_s$ (moments!)

angular moment
$m_l = -2\mu_B \frac{\langle A+B \rangle}{3C}$

see last slide

$m_s = \mu_B \frac{\langle -A+2B \rangle}{C}$

Interaction of **polarized** photons with matter

> <u>History of XMCD</u>

1846 - **M. Faraday**: polarisation of visible light changes when trasmitted by a magnetic material

1975 - **Erskine and Stern** - first theoretical formulation of XMCD effect
excitation from a core state to a valence state for the $M_{2,3}$ edge of Ni.

1987 - G. Schütz et al. - first experimental demonstration of the XMCD at the K-edge of Fe

Application also for thin films; example

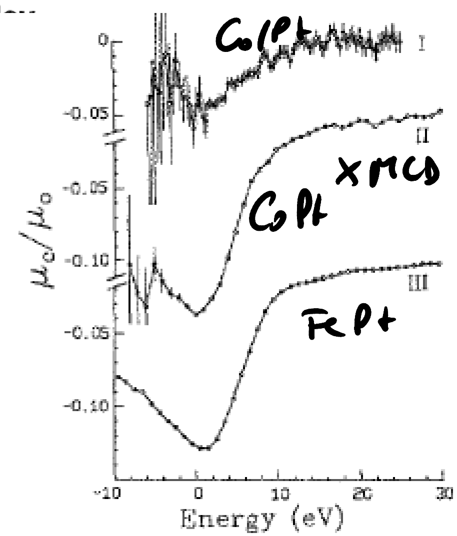
Spin-dependent x-ray absorption in Co/Pt multilayers and $Co_{50}Pt_{50}$

G. Schütz, R. Wienke, and W. Wilhelm
Fak. f. Physik, TU München, D-8046 Garching, Federal Republic of Germany
W. B. Zeper
Philips Research Laboratories, P.O. Box 80.000, 5600 JA Eindhoven, The Netherlands
H. Ebert
Siemens AG, ZFE ME TPH 11, Postfach 3220, D-8520 Erlangen, Federal Republic of Germany
K. Spörl
Institut für Angew. Physik, University of Regensburg, Federal Republic of Germany

The spin dependence of $L_{2,3}$ absorption in 5*d* atoms oriented in a ferromagnetic matrix contains information on the spin density of the empty *d*-projected states of the absorbing atom. Spin-dependent absorption spectroscopy using circularly polarized synchrotron radiation was applied to study the polarization of the Pt atoms in the binary alloy $Co_{50}Pt_{50}$ and Pt/Co layered structures, which are promising candidates for magneto-optical recording. The spin-dependent absorption signals for vapor-deposited 250(4 Å Co + 18 Å Pt) and 250(6 Å Co + 18 Å Pt) multilayers indicate a ferromagnetic coupling on Pt and Co atoms with a significant Pt polarization. This is reduced on average by about 60% with respect to the Pt polarization in the $Co_{50}Pt_{50}$ alloy. The experimental results are discussed on the basis of spin-polarized band-structure calculations.

J. Appl. Phys. 67 (9), 1 May 1990

DORIS II at HASYLAB, DESY, Hamburg.



$E_\gamma^{L_3 Pt} = 11.5$ keV

$|m_s|_{Pt} = 0.35$ μ_B/atom for alloy

$|m_s|_{Pt} = 0.08$ μ_B/atom Co/Pt interface