Methoden moderner Röntgenphysik II: Streuung und Abbildung

Lecture 23

Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2021

G. Grübel, O. Seeck, V. Markmann, F. Lehmkühler, A. Philippi-Kobs, M. Martins

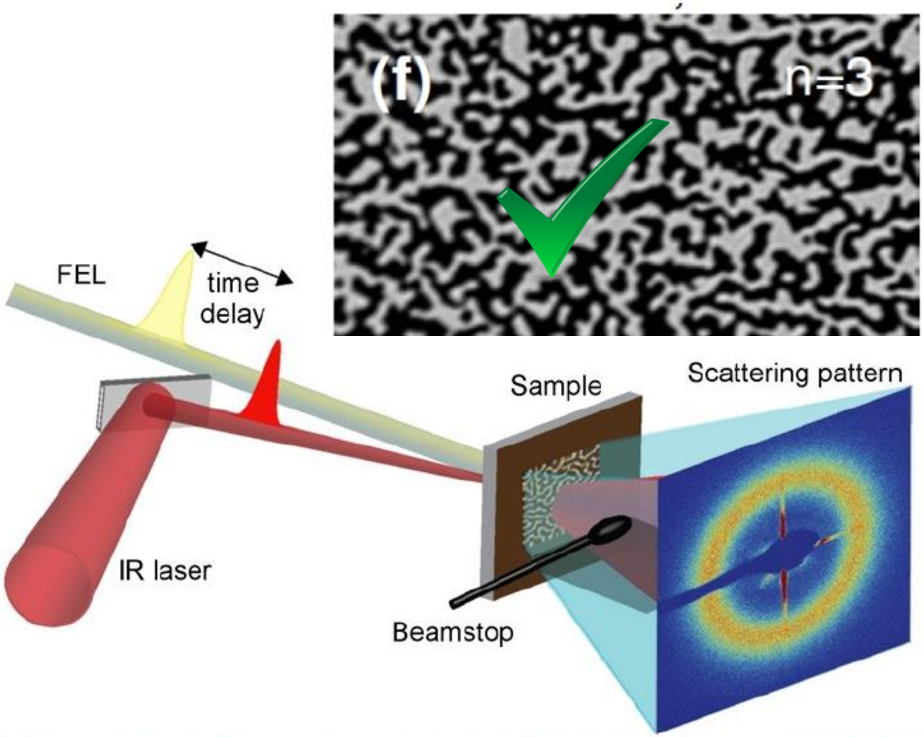
Location	online		
Date	Tuesdays	12:30 - 14:00	(starting 6.4.)
	Thursdays	8:30 - 10:00	(until 8.7.)

Outline

Part III/1:

Studies on Magnetic Nanostructures

by André Philippi-Kobs



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)

[22.6.] **Ferromagnetism in a Nutshell**

- Introduction to Magnetic Materials
- Magnetic Phenomena
- Magnetic Free Energy
- Perpendicular Magnetic Anisotropy
- Magnetic Domains and Domain Walls

[24.6.] **Interaction of Polarized Photons with Ferromagnetic Materials**

- Charge and Spin X-ray Scattering by a Single Electron
- Absorption and Resonant Scattering of Ferromagnets (Semi-Classical and Quantum-Mechanical Concepts)

Interaction of **polarized** photons with matter

2.) Interaction of **polarized** photons with ferromagnetic materials

- Recap: Interaction of X-rays 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

Interaction of **polarized** photons with matter

> Recap: Interaction of X-rays with matter (**consider also light's polarization $\boldsymbol{\varepsilon}$**)

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

$f(\mathbf{q}, \omega, \boldsymbol{\varepsilon}) = f^0(\mathbf{q}) + f'(\omega, \boldsymbol{\varepsilon}) - if''(\omega, \boldsymbol{\varepsilon})$ 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 3)

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

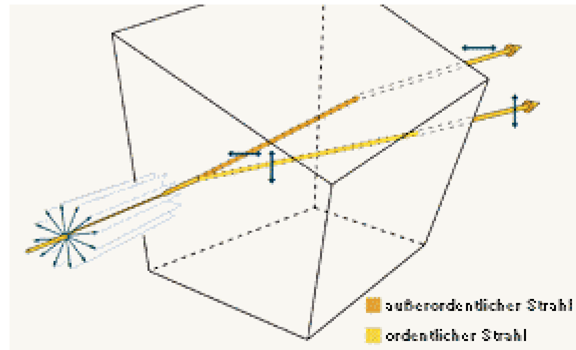
atomic density

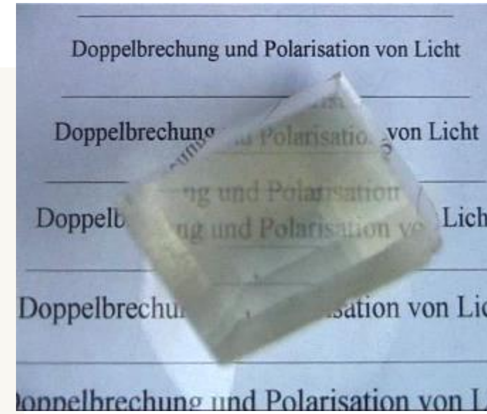
$$\delta(\omega, \boldsymbol{\varepsilon}) = \frac{r_0 \lambda^2}{2\pi} \rho \left(Z + f'(\omega, \boldsymbol{\varepsilon})\right)$$

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

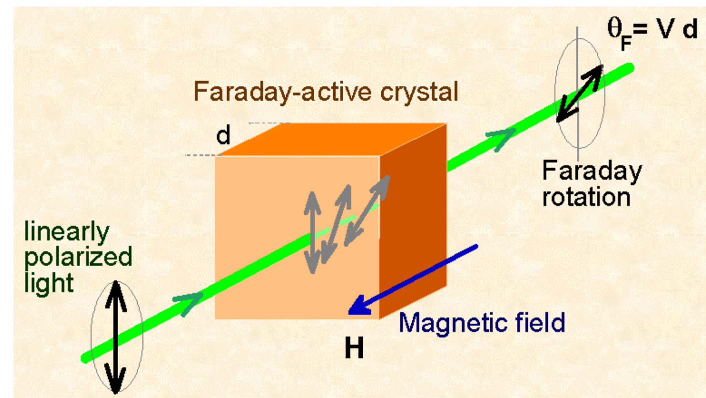
Interaction of **polarized** photons with matter

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





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



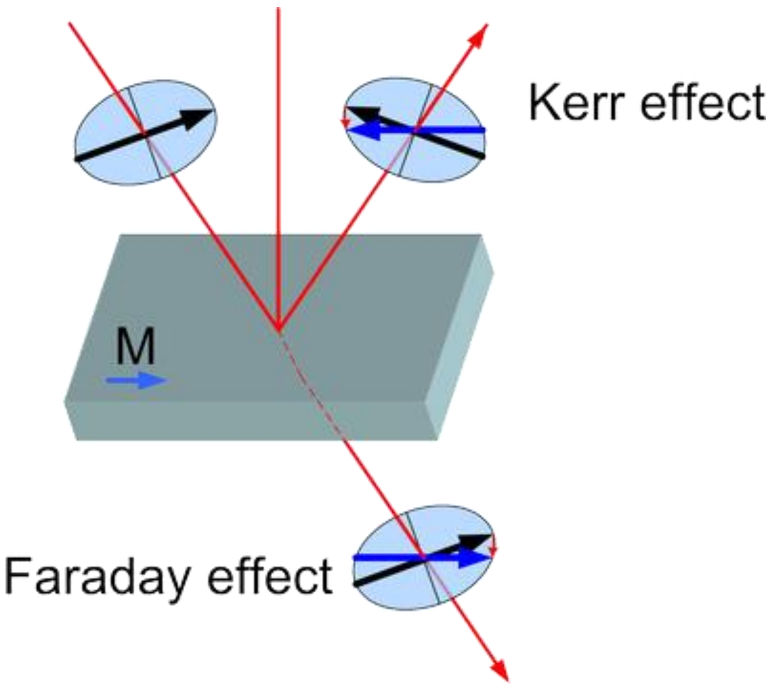
- The change of polarization **ε** is called optical rotation (Faraday effect in case of magnetic materials)

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

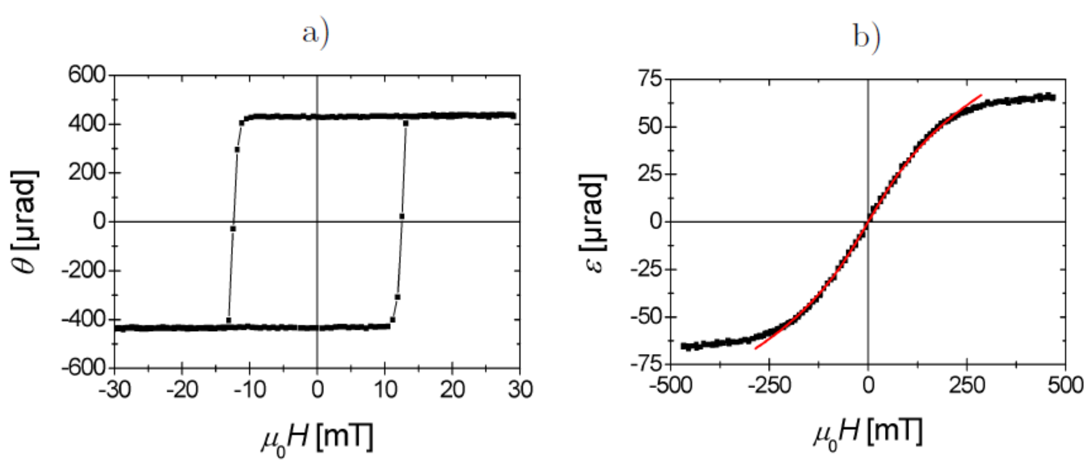
Interaction of **polarized** photons with matter

> History of the interaction between light and ferromagnets

Faraday (1845) and magneto-optical Kerr effect (MOKE) (1876):
Polarization of visible light changes when transmitted/ reflected by a ferromagnetic material



→ Magnetic hysteresis



Interaction of **polarized** photons with matter

> History of the interaction between light and ferromagnets

XMCD effect:

Erskine and Stern (1975):
First theoretical formulation of XMCD for the excitation from a core to valence state for the M2,3 edge of Ni

G. Schütz et al. (1987):
First experimental demonstration of the XMCD effect at the K-edge of Fe at DORIS at DESY

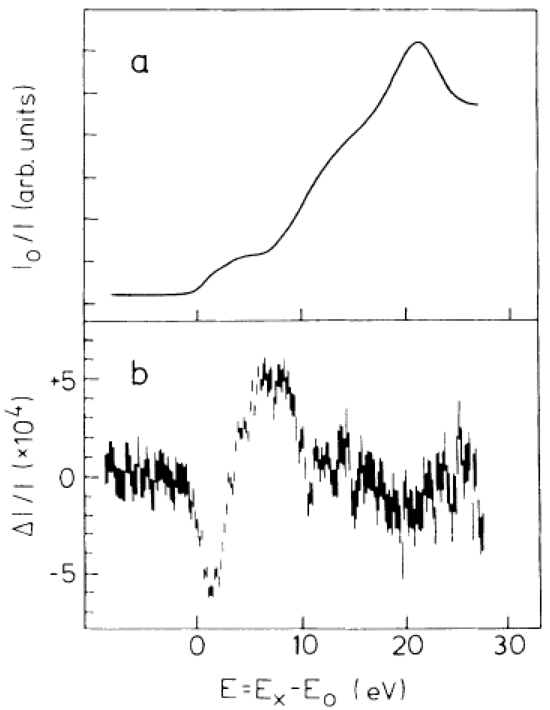


FIG. 1. (a) Absorption I_0/I of x rays as function of the energy E above the K edge of iron and (b) the difference of the transmission $\Delta I/I$ of x rays circularly polarized in and opposite to the direction of the spin of the magnetized d electrons.

VOLUME 58, NUMBER 7 PHYSICAL REVIEW LETTERS 16 FEBRUARY 1987

Absorption of Circularly Polarized X Rays in Iron

G. Schütz, W. Wagner, W. Wilhelm, and P. Kienle[a]
Physik Department, Technische Universität München, D-8046 Garching, West Germany

R. Zeller
Institut für Festkörperforschung der Kernforschungsanlage Jülich, D-5175 Jülich, West Germany

and

R. Frahm and G. Materlik
Hamburger Synchrotronstrahlungslabor am Deutsches Elektronen-Synchrotron DESY, D-2000 Hamburg 52, West Germany
(Received 22 September 1986)

May 2019 814 citations! (~20 in the last year)

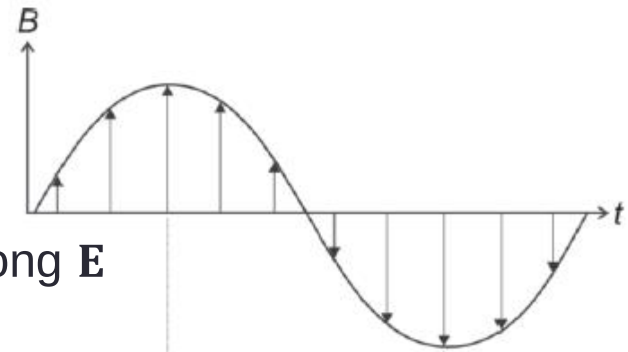
Interaction of **polarized** photons with matter

- Scattering of X-rays by a single electron (lecture 2); also **consider spin of electron ➔ magnetic XRD**

Incoming plane wave

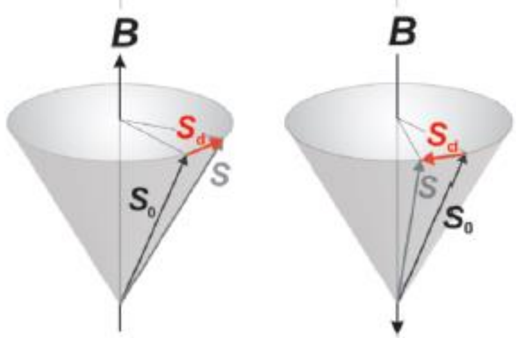
$$\mathbf{E}(\mathbf{r},t) = \boldsymbol{\varepsilon} \cdot E_0 e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})}$$

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



Electric dipole moment (charge movement) oscillates along $\mathbf{E}$

$$\mathbf{p}(t) = -\frac{e^2}{m_e \omega^2}\mathbf{E}_0 e^{-i\omega t}$$



Spin of electron precesses around magnetic field according to

$$\frac{\mathrm{d}\mathbf{S}_d(t)}{\mathrm{d}t} = -\frac{e}{m_\mathrm{e}}\mathbf{S}_0(t) \times \mathbf{B}(t)$$

Landé-factor

With definition of magnetic moment $m = -2\mu_\mathrm{B} s_\mathrm{d}$

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

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

Note: different oscillation directions of **p** and **m**

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):

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

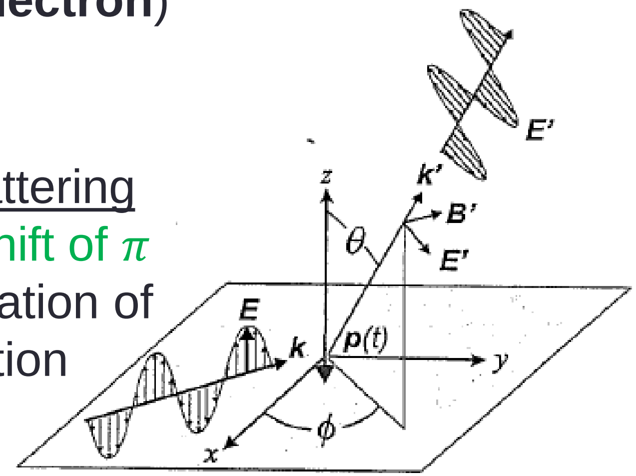
$$E'(t) = -\frac{1}{4\pi\epsilon_0}\cdot\frac{e^2}{m_e c^2}\cdot\frac{e^{ik'r}}{r}[\boldsymbol{k}_0' \times \boldsymbol{E}(t)] \times \boldsymbol{k}_0'$$

- magnetic dipole (Jackson text book):

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

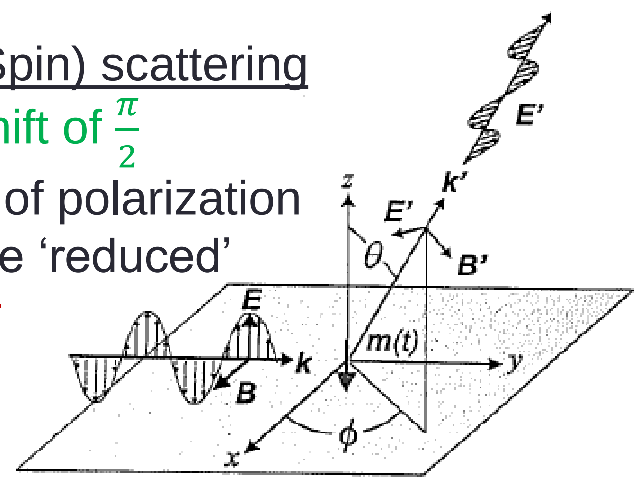
$$\boldsymbol{E}'(t) = \mathrm{i}\frac{1}{4\pi\epsilon_0}\frac{e^2}{m_e c^2}\frac{\hbar\omega}{m_e c^2}\frac{\mathrm{e}^{ik'r}}{r}[\boldsymbol{s} \times (\boldsymbol{k}_0 \times \boldsymbol{E}(t))] \times \boldsymbol{k}_0'$$

Electric dipole scattering



Charge scattering
- Phase shift of π
- Conservation of polarization for **k||k'**

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: $f(\epsilon, \epsilon') = -\frac{re^{-ik'r}}{E} E' \cdot \epsilon'$

$$f_e(\epsilon, \epsilon') = \frac{1}{4\pi\epsilon_0} \frac{e^2}{m_e c^2} \epsilon \cdot \epsilon' = r_0 \underbrace{\epsilon \cdot \epsilon'}$$

$$f_s(\epsilon, \epsilon') = -\mathrm{i}\, r_0 \frac{\hbar\omega}{m_e c^2} s \cdot (k_0 \times \epsilon) \times (k'_0 \times \epsilon')$$

Remember: Polariazion factor $P = \sin\theta$

Differential scattering cross-section: $\frac{d\sigma}{d\Omega} = |f(\epsilon, \epsilon')|^2 = r_0^2 \sin^2\theta$ für $f = f_e$

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

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

$$\sigma_s = \frac{8\pi}{3} \frac{1}{4} \left(\frac{\hbar\omega}{m_e c^2}\right)^2 r_0^2 = \frac{\sigma_e}{4} \left(\frac{\hbar\omega}{m_e c^2}\right)^2$$

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

Interaction of **polarized** photons with matter

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

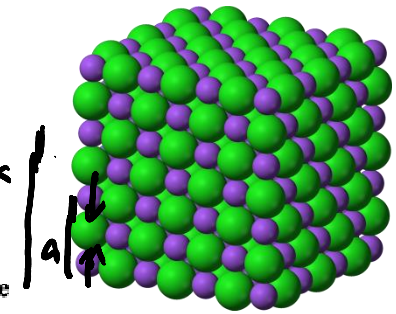
Take-home messages:

- Charge scattering causes a phase shift of π ('-') between incident and scattered field
- Spin scattering causes a phase shift of $\frac{\pi}{2}$ ('*i*') between incident and transmitted field
- Charge scattering conserves the polarization while spin scattering causes a rotation of the polarization
- The spin-scattering amplitude is different by a factor of $\frac{\hbar\omega}{m_e c^2}$ relative to charge scattering amplitude, with $m_e c^2 = 511\mathrm{keV}$, i.e., for extremely high photon energies spin scattering is stronger (these energies are [usually] not relevant for X-ray physics)
- Spin scattering is generally weak and can only be measured in special circumstances

Interaction of **polarized** photons with matter

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

Example: **magnetic XRD** of antiferromagnetic NiO



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

intensity of the (111) reflection. 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. The r

Reflex aufgrund
- Ladungsdichte bei $(\frac{2\pi}{a}, \frac{2\pi}{a}, \frac{2\pi}{a})$ ≙ (1 1 1) (h k l)
- Spindichte bei (h k l) $(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})$

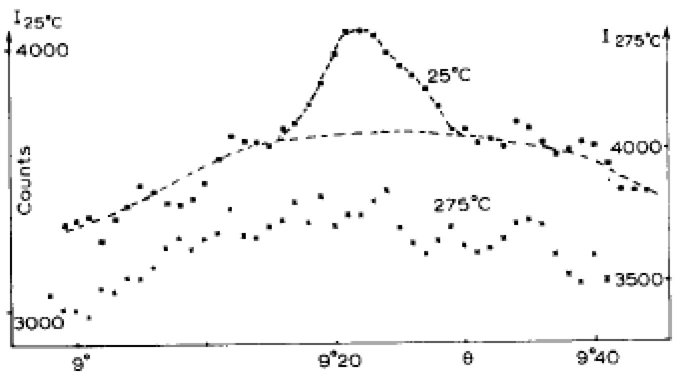


Fig. 1. Intensity $I_t(\theta)$ near the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ position at $t = 25°$ C and 275° C in counts/225 min. The hump which cover the interval could be due to some impurity.

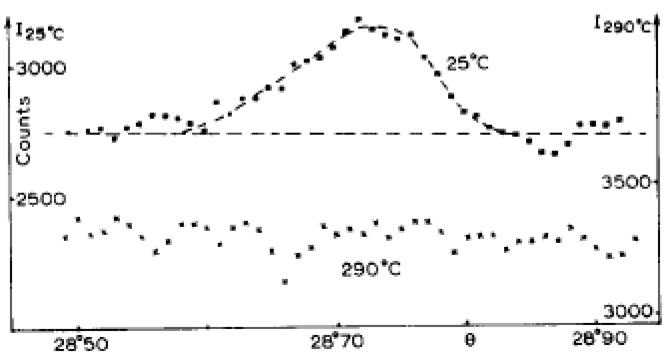


Fig. 2. Intensity $I_t(\theta)$ near the $(\frac{3}{2}\frac{3}{2}\frac{3}{2})$ position at $t = 25°$C and 290°C in counts/225 min.

Interaction of **polarized** photons with matter

> Recap: Absorption and Resonant Scattering (classical concept, lecture 6)

Picture: Electrons are bound to atoms

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

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

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)

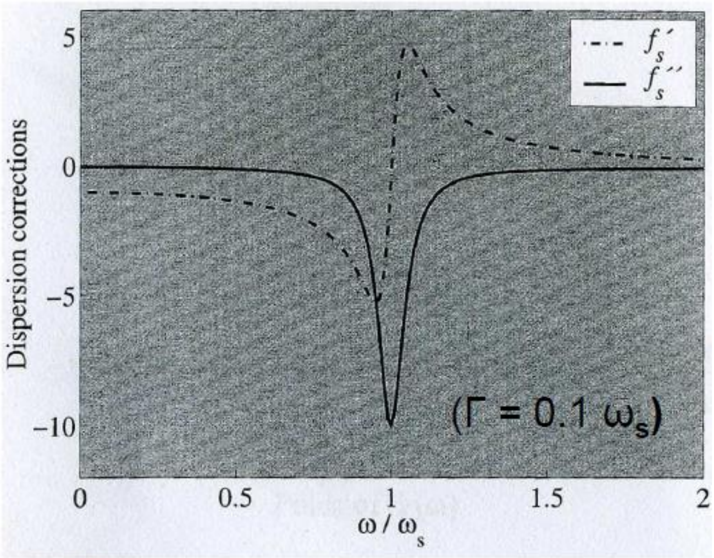
$$\sigma_T = \left(\frac{8\pi}{3}\right) \frac{\omega^4}{(\omega^2 - \omega_s^2)^2 + (\omega\Gamma)^2} r_0^2$$

(scattering cross-section)

DESY

Interaction of **polarized** photons with matter

> Recap: Absorption and Resonant Scattering (classical concept)



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

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

Measure absorption cross-section in experiment

Use Kramers-Kronig relations to obtain *f* ‘

with:

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

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

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

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

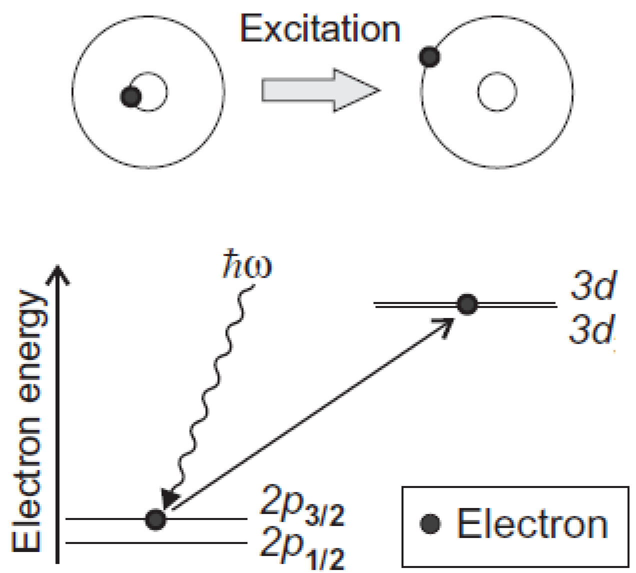
Interaction of **polarized** photons with matter

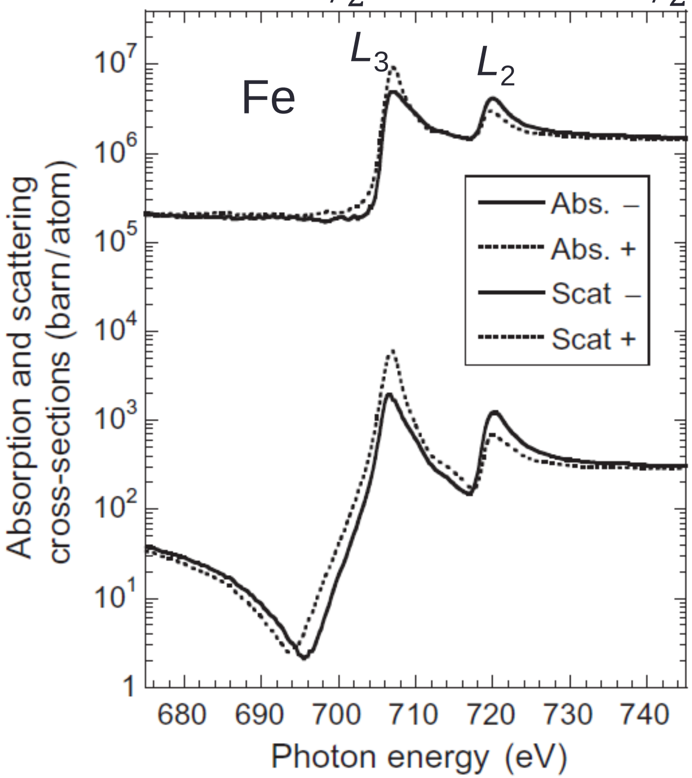
> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

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

$L_3: 2p_{3/2} \rightarrow 3d; L_2: 2p_{1/2} \rightarrow 3d$

$(2p^1 \rightarrow 3d^1$ 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"

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

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

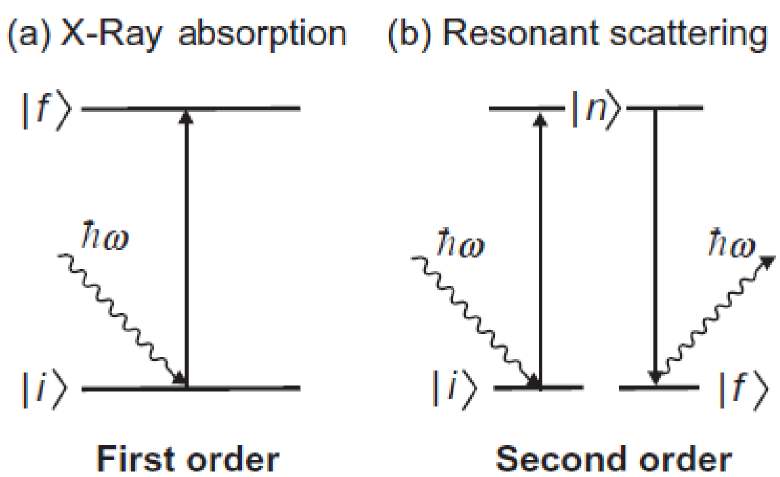
$\rho(\varepsilon_f)$: density of final states

ε_n: energy of all possible intermediate states n

$\mathcal{H}_{\text{int}}$: Interaction Hamiltonian

- 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)

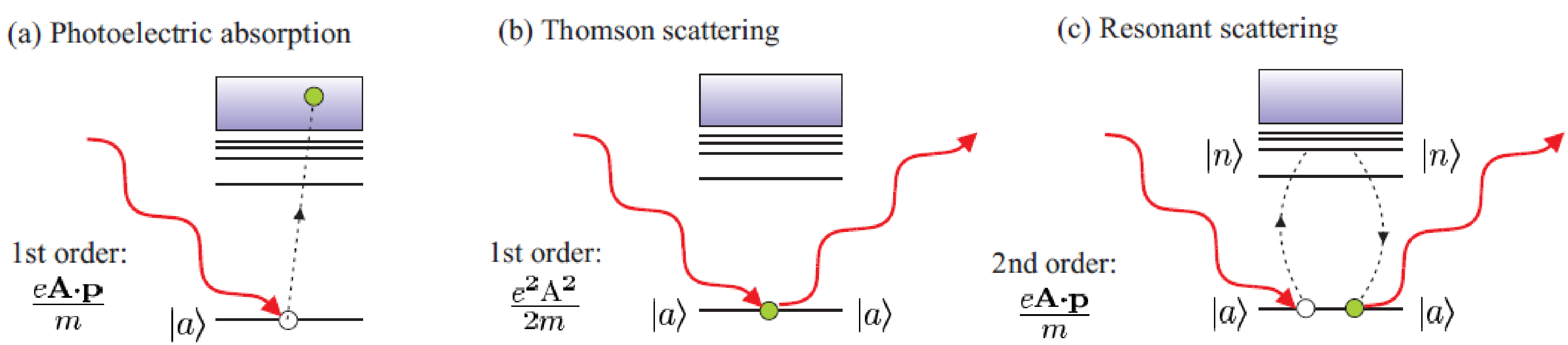
- Time-dependent pertubation theory (up to second order)

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

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

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

p: momentum of electrons
A: vector potential of EM-wave



Interaction of **polarized** photons with matter

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

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

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

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

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

p: momentum of electrons
A: vector potential

With $\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t}$ and assumption of a plane wave $\mathbf{E}(\mathbf{r}) = \boldsymbol{\epsilon}E_0 \cdot e^{i\mathbf{k}\cdot\mathbf{r}}$:

Transition-matrix element $M \propto \langle b|\mathbf{p}\cdot\boldsymbol{\epsilon}e^{i\mathbf{k}\cdot\mathbf{r}}|a\rangle$, $|a\rangle, |b\rangle$ atomic states

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

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

Dipole-Approximation: eliminate the **k** dependence in *M*

Expansion of $\mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}} = 1 + i\mathbf{k}\cdot\mathbf{r} - \frac{(i\mathbf{k}\cdot\mathbf{r})^2}{2} + \cdots$

Size of electron-radius $\mathbf{r}$: $|\mathbf{r}| \approx 0.1\text{Å} = \ell$ for 2*p*-core shells

Soft X rays : $E_\gamma \leq 1\text{keV} \rightarrow \lambda \geq 1\text{nm} \approx 100 \cdot \ell;\ |\mathbf{k}| = \frac{2\pi}{\lambda} \leq 5 \cdot 10^9\ \text{m}^{-1}$

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

$\rightarrow M = \langle b|\vec{p}\cdot\vec{\epsilon}|a\rangle$, i.e., 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

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

Reformulation of Matrix-elements

$$\begin{aligned} M &= \langle b|\mathbf{p}\cdot\boldsymbol{\epsilon}|a\rangle = \frac{im_e}{\hbar}\langle b|[H,\mathbf{r}]\boldsymbol{\epsilon}|a\rangle \\ &= \frac{im_e}{\hbar}(\langle b|H\mathbf{r}\cdot\boldsymbol{\epsilon}|a\rangle - \langle b|\mathbf{r}H\boldsymbol{\epsilon}|a\rangle) \\ &= \frac{im_e}{\hbar}(E_b\,\langle b|\mathbf{r}\cdot\boldsymbol{\epsilon}|a\rangle - E_a\langle b|\mathbf{r}\cdot\boldsymbol{\epsilon}|a\rangle) \\ &= \frac{im_e}{\hbar}(E_b - E_a)\langle b|\mathbf{r}\cdot\boldsymbol{\epsilon}|a\rangle = im_e\omega\langle b|\vec{r}\vec{\epsilon}|a\rangle \end{aligned}$$

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 $\mathbf{r} = x\mathbf{e}_x + y\mathbf{e}_y + z\mathbf{e}_z$

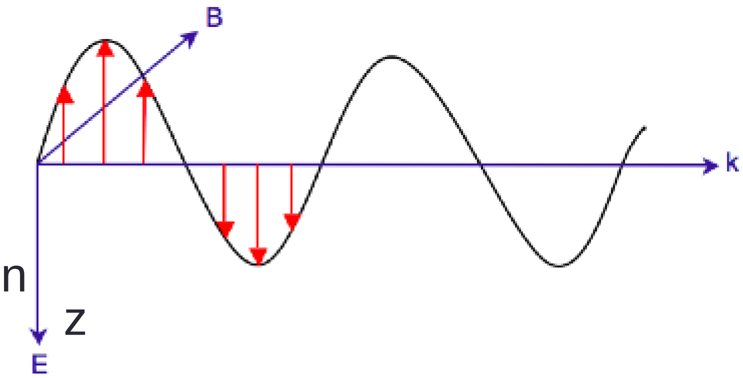
Linear polarized light $\epsilon_x^0 = \epsilon_x = e_x$ $\epsilon_y^0 = \epsilon_y = e_y$ $\epsilon_z^0 = \epsilon_z = e_z$

('extreme cases')

$$P_z^0 = \boldsymbol{\epsilon} \cdot \mathbf{r} = z = r\cos\theta$$
$$= r\sqrt{\frac{4\pi}{3}} \cdot Y_{1,0}$$

$Y_{l,m}$: spherical harmonics = Kugelflächenfuktionen

Linearly polarized



Interaction of **polarized** photons with matter

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

Polarization dependent dipole operator $P = \boldsymbol{\epsilon} \cdot \boldsymbol{r}$

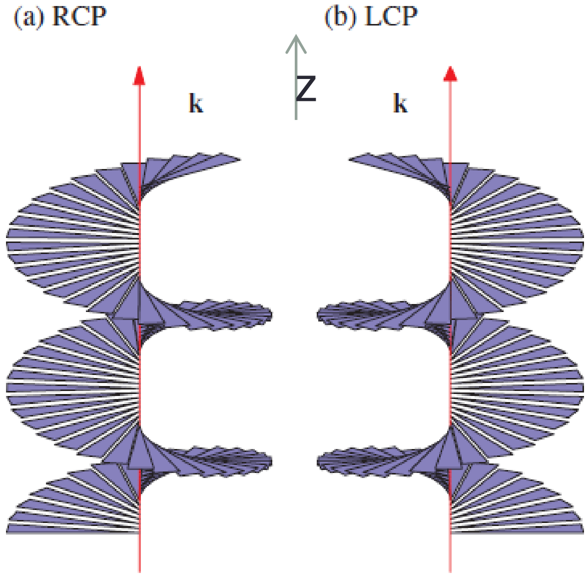
Right and left circularly polarized light $(\mathbf{k}||\mathbf{e}_z)$

$\boldsymbol{\epsilon}^{\pm} = \pm\frac{1}{\sqrt{2}}(\boldsymbol{\epsilon}_x \pm i\boldsymbol{\epsilon}_y)$, i.e., supposition of linear light with a phase shift of $\frac{\pi}{2}$

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

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

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



Interaction of **polarized** photons with matter

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

➔ Polarization dependent Dipole Operator for circularly polarized light:

$$P^{\pm} = \boldsymbol{\epsilon}^{\pm} \cdot \mathbf{r} = \mp \frac{1}{\sqrt{2}}(x + iy) = \mp \mathrm{r} \cdot \sin\theta \, e^{\pm i\phi}$$

$$= r\sqrt{\frac{4\pi}{3}} Y_{1,\pm 1} \;= r\sqrt{\frac{4\pi}{3}} Y_{\ell,m_l}$$

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: $P_z^0 = rC_0^{(1)}$ for linear polarization

$P_z^{\pm} = rC_{\pm 1}^{(1)}$ for RCP (+) and LCP (-)

Interaction of **polarized** photons with matter

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

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

$$|a\rangle = \left|R_{n,l}(r); l, m_l, s, m_s\right\rangle = R_{n,l}(r) Y_{l,m_l} \chi_{s,m_s}$$
$$|b\rangle = |R_{n',l'}(r); l', m_{l'}, s', m'_s\rangle$$

$$\left\langle b \middle| P_z^q \middle| a \right\rangle = \underbrace{\delta(m_s', m_s)}_{\text{Spin}} \cdot \underbrace{\langle n', l' | r | n, l\rangle}_{\text{Radial}} \cdot \underbrace{\sum_{m_l, m_l', q} \langle l', m_l' \left| C_q^{(1)} \right| l, m_l\rangle}_{\text{Angular}}$$

Spin:	Electron-spin direction is conserved
Radial transition strength:	the same transition strength for all $2p \rightarrow 3d$ transitions
Angular:	Contains polarization dependence

Note: as core electrons are strongly localized, absorbtion spectroscopy is sensitive to the valence shell properties within the atomic volume

Interaction of **polarized** photons with matter

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

Non-vanishing matrix elements (use in excercise, 30.6.)

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

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

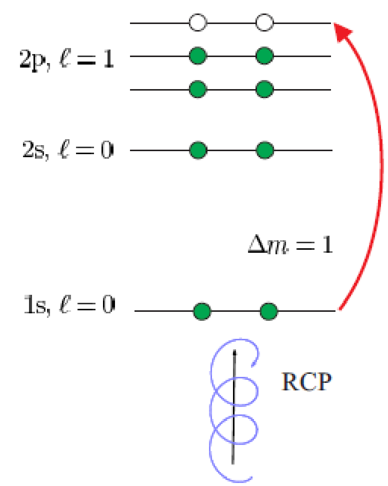
$$\langle l-1,m+1|\,C_1^{(1)}\,|l,m\rangle = -\sqrt{\frac{(l-m)(l-m-1)}{2(2l-1)(2l+1)}}$$

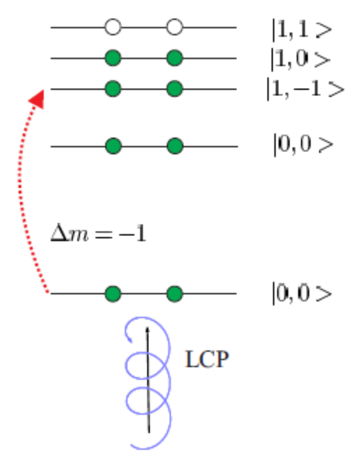
LCP

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

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

(a) Simplified energy level diagram





Interaction of **polarized** photons with matter

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

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

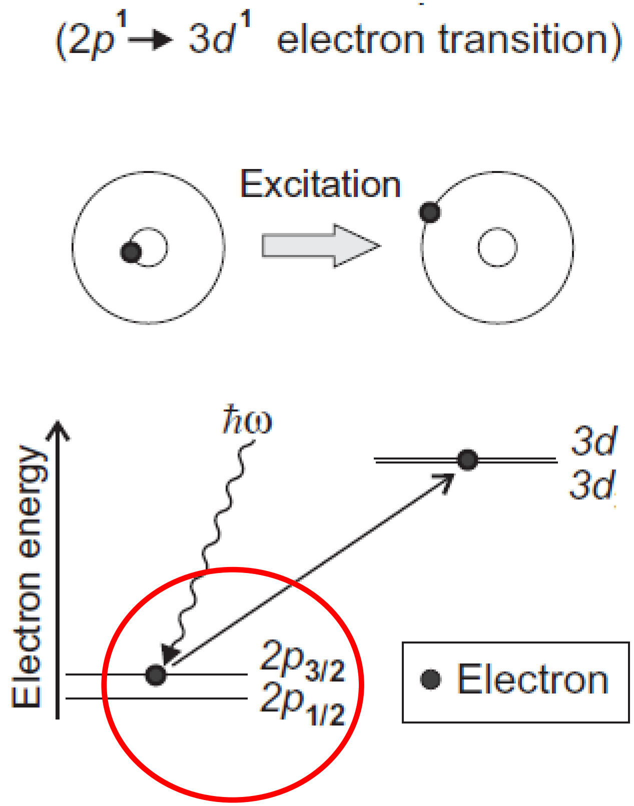
$$\Delta l \quad = l' - l = \pm 1$$
$$\Delta m_l = m_l' - m_l = q = 0, \pm 1$$
q = helicity of light (i.e., polarizations lin., RCP, LCP)
$$\Delta s \quad = s' - s = 0$$
$$\Delta m_s = m_s' - m_s = 0$$

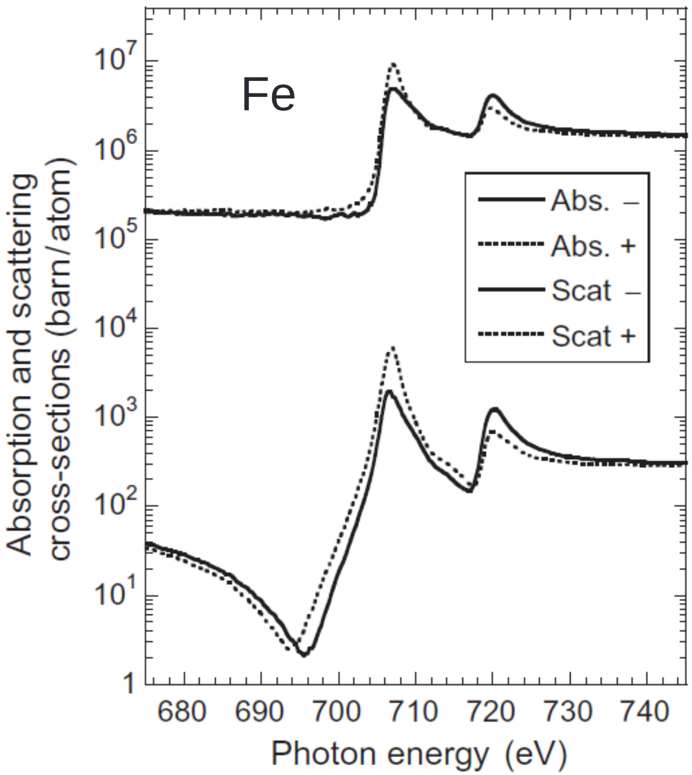
Note again: electron-spin direction is conserved

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





Spin-orbit coupling: $|n, l, s, j, m_j >$ are good quantum numbers!

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 next lecture)
➔ Clebsch-Gordon coefficients *C*

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

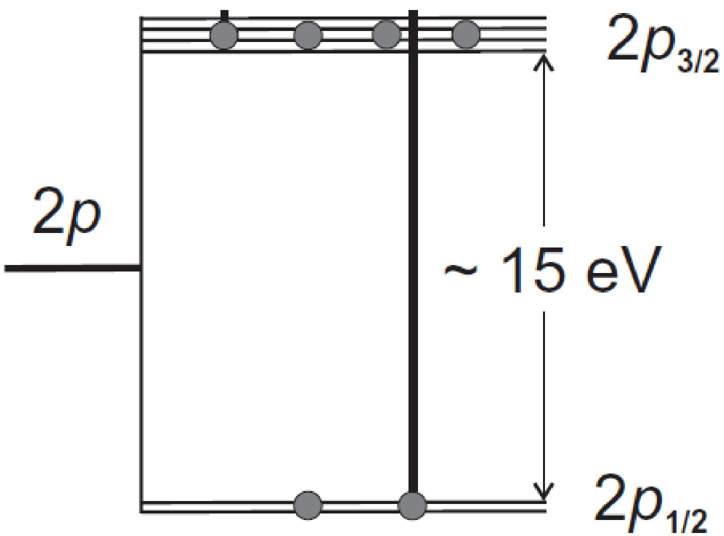


Table 2

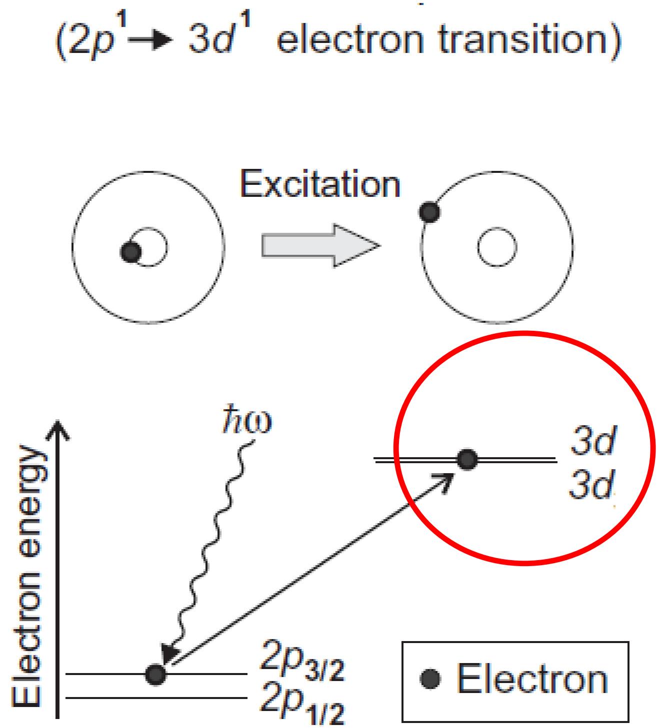
$\|l,s,j,m_j\rangle$ basis		$\|l,m_l,s,m_s\rangle$ basis
j	m_j	$Y_{l,m_l}\chi^{\pm}$
$\frac{1}{2}$	$+\frac{1}{2}$	$\frac{1}{\sqrt{3}}(-Y_{1,0}\,\alpha + \sqrt{2}\,Y_{1,+1}\,\beta)$
	$-\frac{1}{2}$	$\frac{1}{\sqrt{3}}(-\sqrt{2}\,Y_{1,-1}\,\alpha + Y_{1,0}\,\beta)$
$\frac{3}{2}$	$+\frac{3}{2}$	$Y_{1,+1}\,\alpha$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

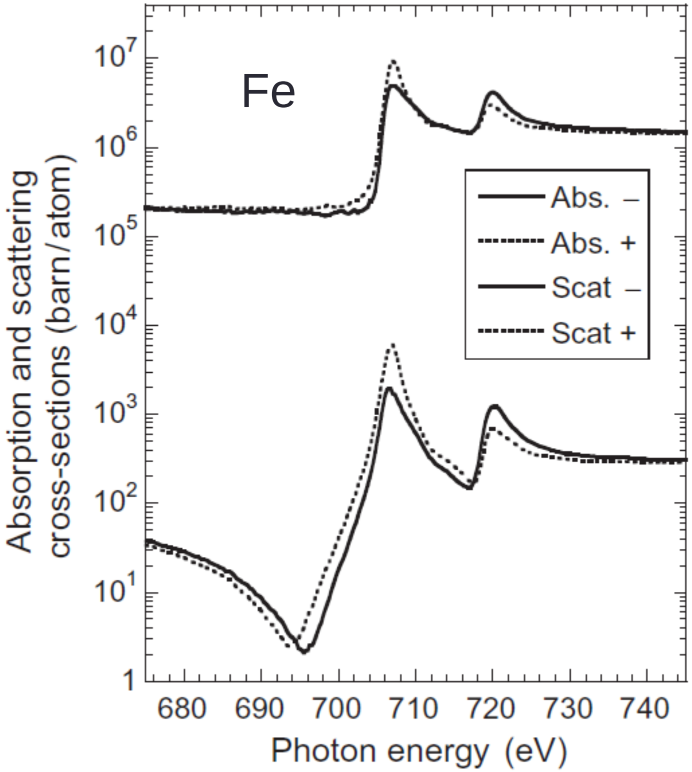
$\alpha \hat{=} \uparrow$
$\beta \hat{=} \downarrow$

Interaction of **polarized** photons with matter

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

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

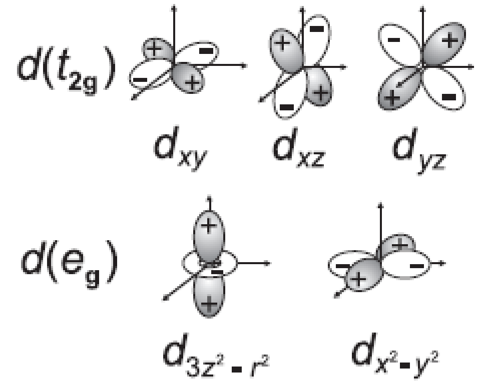




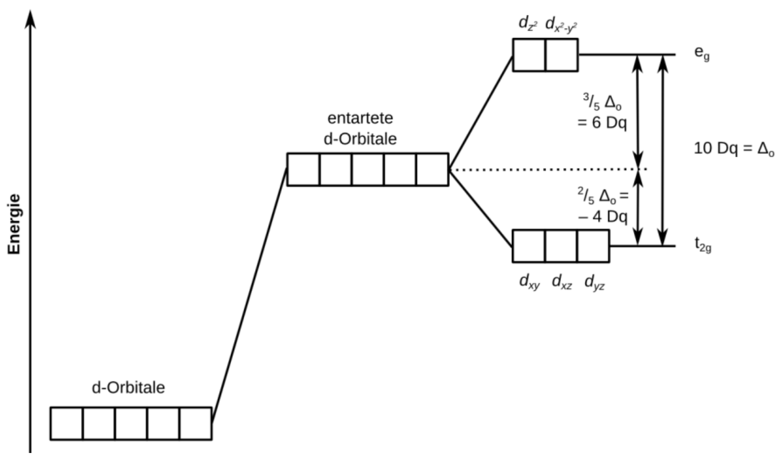
For *d*-states the spin-orbit coupling is small but there is crystal-field splitting

Interaction of **polarized** photons with matter

> <u>Absorption</u> (qm concept, Fermi's golden rule)



Itinerant *d*-states are split due to crystal field
(can be neglected in a good approximation as splitting is small
➔ use atomic wave functions without SOC; lecture on 30.6.)



crystal field split d-states | $|l, m_l\rangle$ basis

crystal field split d-states				$\|l, m_l\rangle$ basis
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}	$=$	$\sqrt{\frac{15}{4\pi}\frac{yz}{r^2}}$	$=$	$\frac{i}{\sqrt{2}}(Y_{2,-1} + Y_{2,+1})$
$d_{x^2-y^2}$	$=$	$\sqrt{\frac{15}{16\pi}\frac{(x^2-y^2)}{r^2}}$	$=$	$\frac{1}{\sqrt{2}}(Y_{2,-2} + Y_{2,+2})$
$d_{3z^2-r^2}$	$=$	$\sqrt{\frac{5}{16\pi}\frac{(3z^2-r^2)}{r^2}}$	$=$	$Y_{2,0}$