Methoden moderner Röntgenphysik I + II: Struktur und Dynamik kondensierter Materie

Vorlesung zum Haupt/Masterstudiengang Physik SS 2009

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Materials Science

7. 5.	Martin v. Zimmermann	correlated electron materials – structural properties
12. 5.	Hermann Franz	glasses I
14. 5.	Martin v. Zimmermann	correlated electron materials – magnetic properties
19. 5.	Hermann Franz	glasses II
26.5.	Hermann Franz	exercises

correlated electron materials: overview

- phase transitions
- structural phase transition of $SrTiO_3$
- x-ray diffraction to investigate phase transitions
- structural aspects of transition metal oxides
- orbital and charge order in $La_{1-x}Ca_xMnO_3$
- resonant scattering to study orbital/charge order

- magnetic interactions in transition metal oxides
- Mott insulator
- colossal magneto resistance (CMR) effect
- resonant / non-resonant magnetic scattering

exchange interactions

combination of Coulomb interaction and Pauli principle

$$J \sim -\int \Psi^*_x(\mathbf{r}_1)\Psi_y(\mathbf{r}_1)\ (e^2/r_{12})\ \Psi^*_y(\mathbf{r}_2)\Psi_x(\mathbf{r}_2)$$

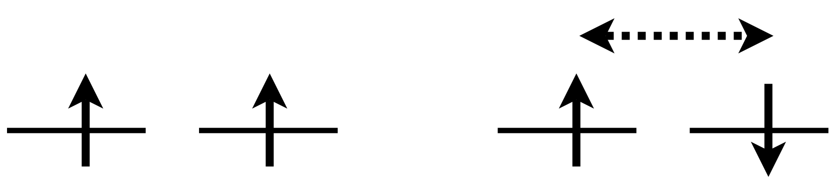
one-band Hubbard model:

$$H = -\sum t_{ij}\ (c_{i\sigma}^{+}\ c_{j\sigma}) + U \sum n_{i\uparrow}n_{i\downarrow}$$

$$= H_{kin} + H_U$$

t_{ij} hopping amplitude between nn sites $\langle ij \rangle$
$c_{i\sigma}^{+}$ creates an electron with spin σ at lattice site i
U Coulomb repulsion
$n_{i\sigma}$ number of electrons at site i with spin σ

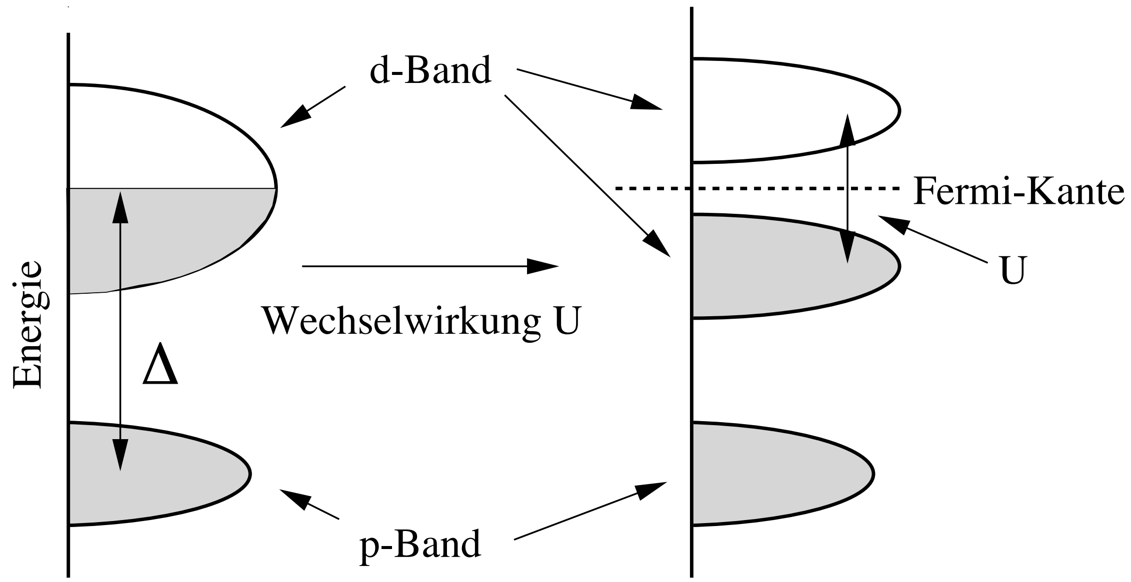
$t >> U$: metallic system
$t << U$: insulator with one electron per site



$\Delta E = 0$ $\Delta E = -2t^2/U$

superexchange:
antiferromagnetic

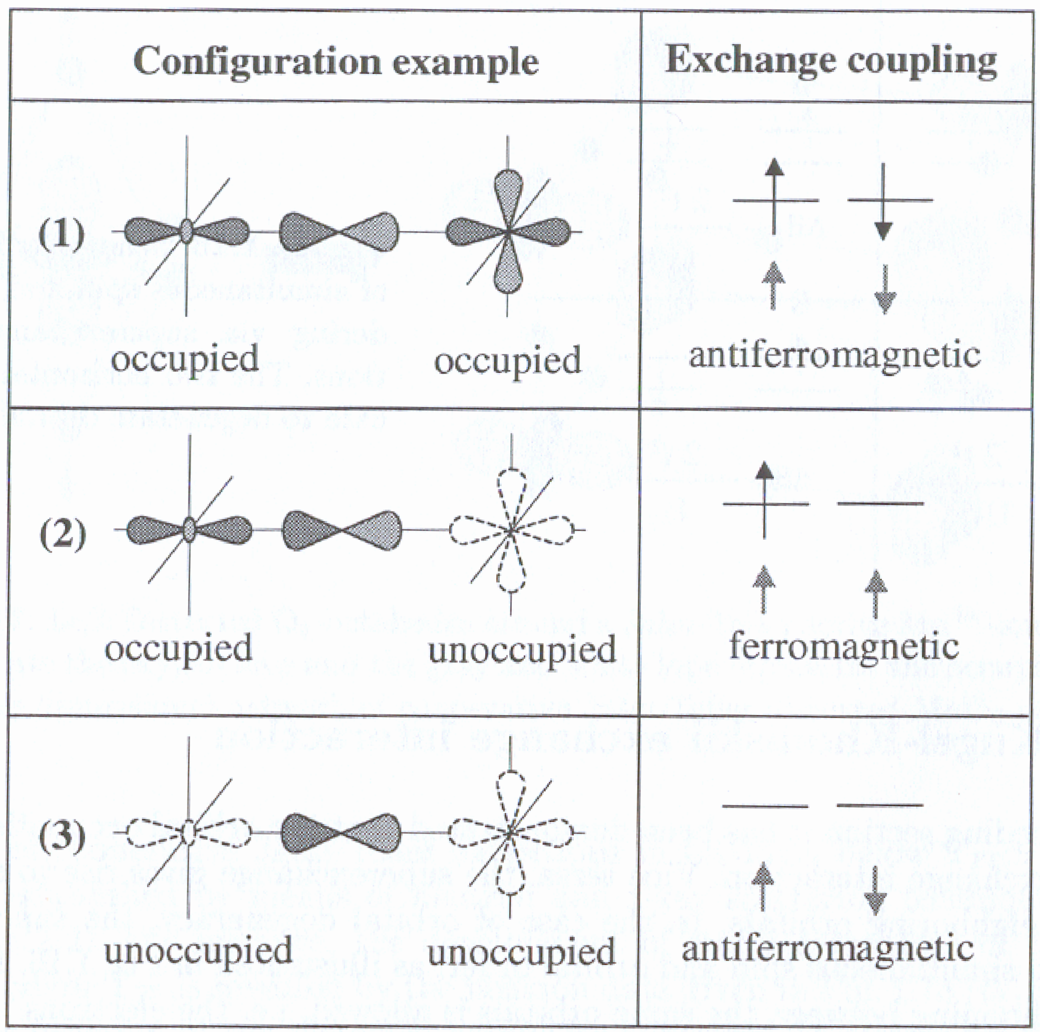
Mott insulator



strongly correlated electron systems: transition metal oxides
high-Tc superconductors
CMR-manganites ...

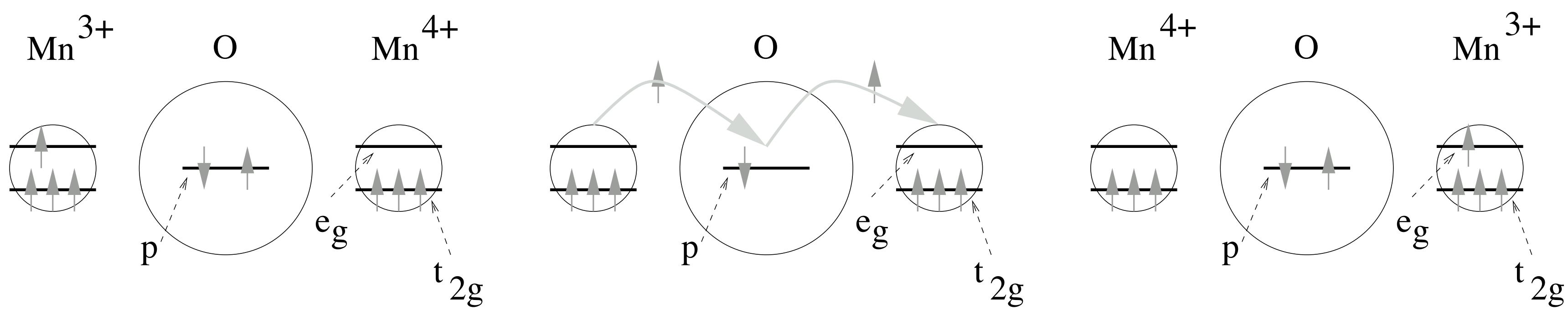
GKA-rules (Goodenough-Kanamori-Andersen)

orbital dependent exchange interaction

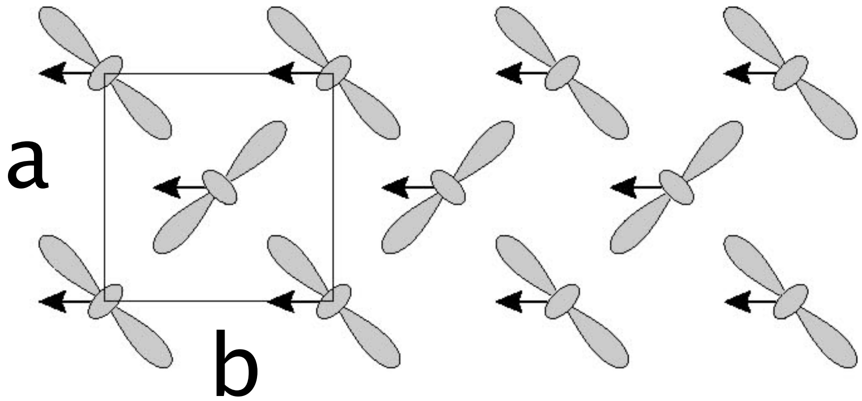


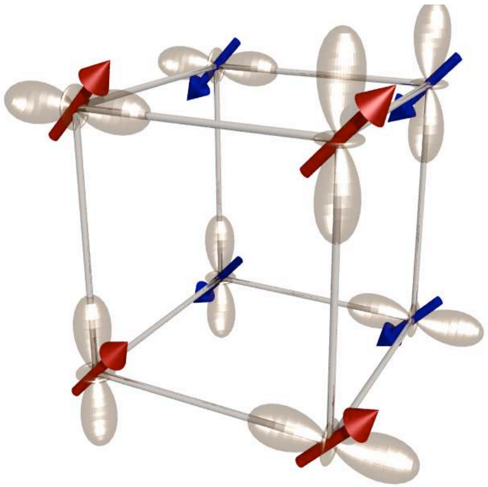
double exchange interaction

ferromagnetic interaction between different ions due to Hund's coupling

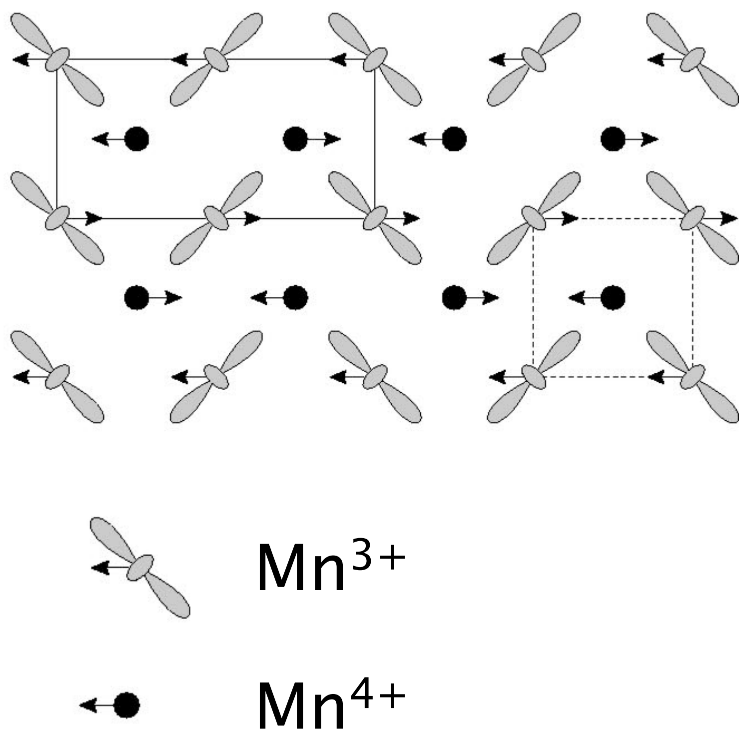


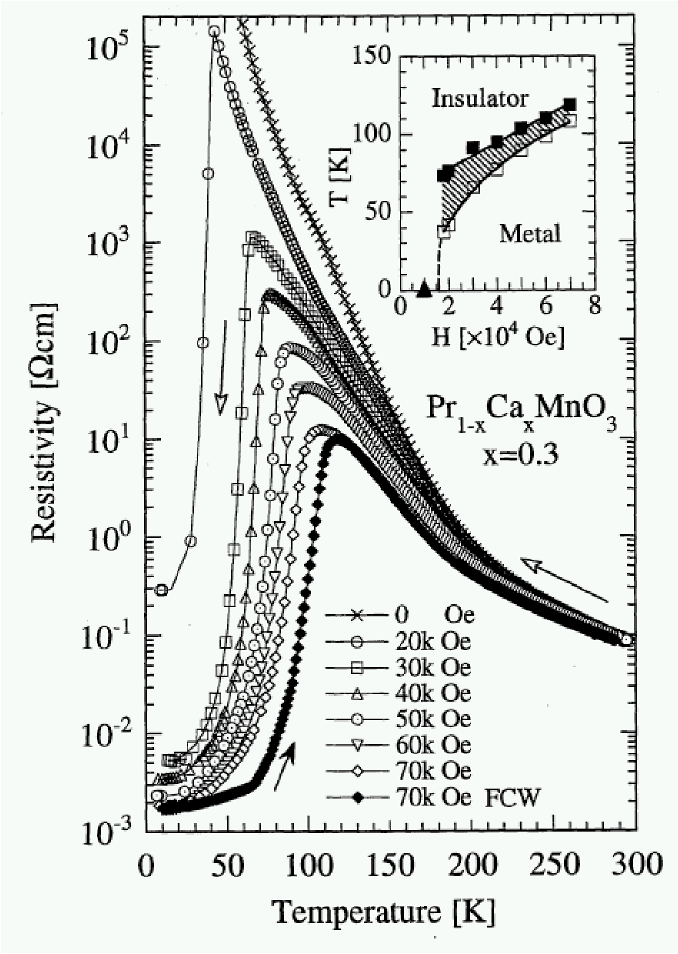
magnetism of $LaMnO_3$





magnetism of $La_{0.5}Ca_{0.5}MnO_3$ and colossal magneto resistance (CMR) effect



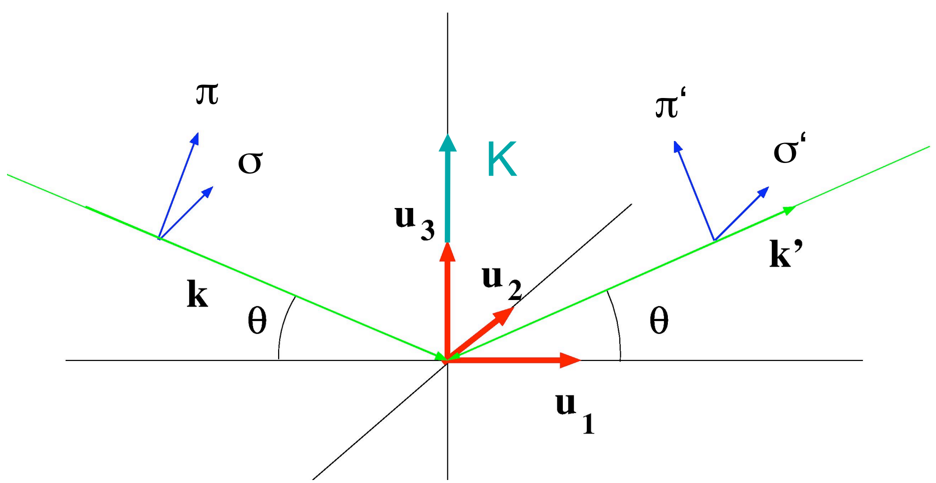


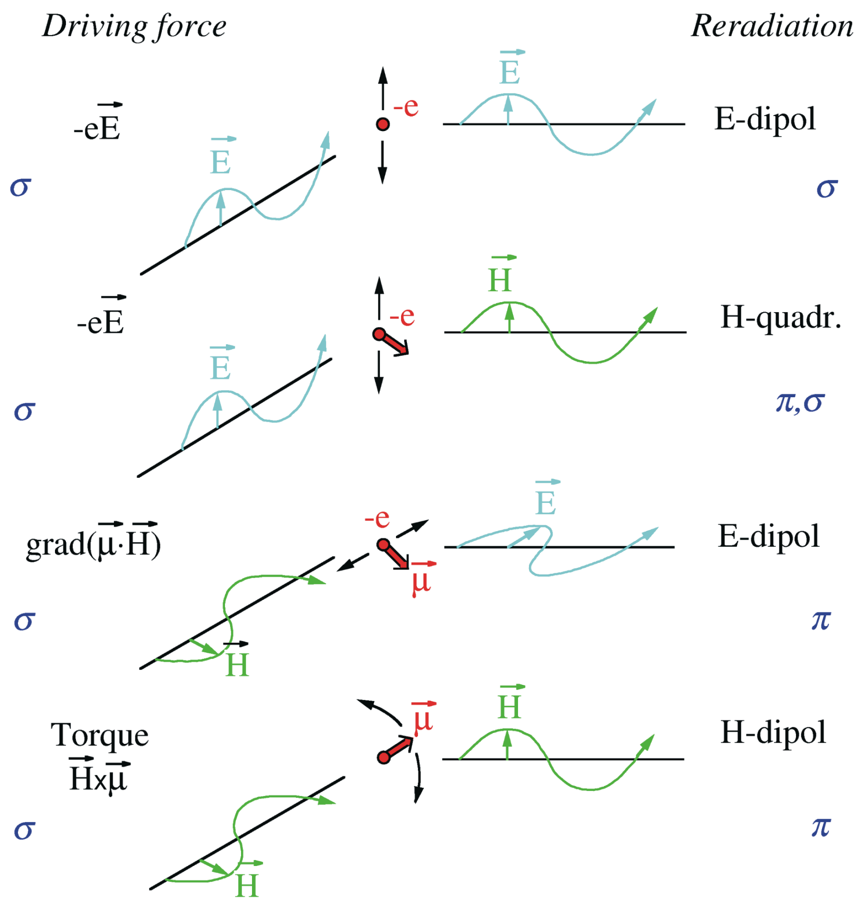
magnetic x-ray scattering

Synchrotronstrahlung linear polarisiert in Ringebene

Streugeometrie vertikal

➔ σ – polarisierte einfallende Strahlung





resonant magnetic x-ray scattering

Röntgenstreuung an periodischen Strukturen

(Blume, J. Appl. Phys. **57**, 3615 (1985); Blume and Gibbs, PRB **37**, 1779 (1988)):

$$\frac{d\sigma}{d\Omega} = r_o^2 \left| \sum_n e^{i\vec{Q}\cdot\vec{r}_n} f_n\left(\vec{k},\vec{k}',\hbar\omega\right) \right|^2$$

Streuamplitude:

$$f\left(\vec{k},\vec{k}',\omega\right) = f^{charge}\left(\vec{Q}\right) + f^{spin}\left(\vec{k},\vec{k}',\omega\right) + f'\left(\vec{k},\vec{k}',\omega\right) + if''\left(\vec{k},\vec{k}',\omega\right)$$

f charge $\rightarrow$ Thomsonstreuung

f ´und f ˝ $\rightarrow$ Energieabhängige Beiträge

f spin $\rightarrow$ Streuung an Spins

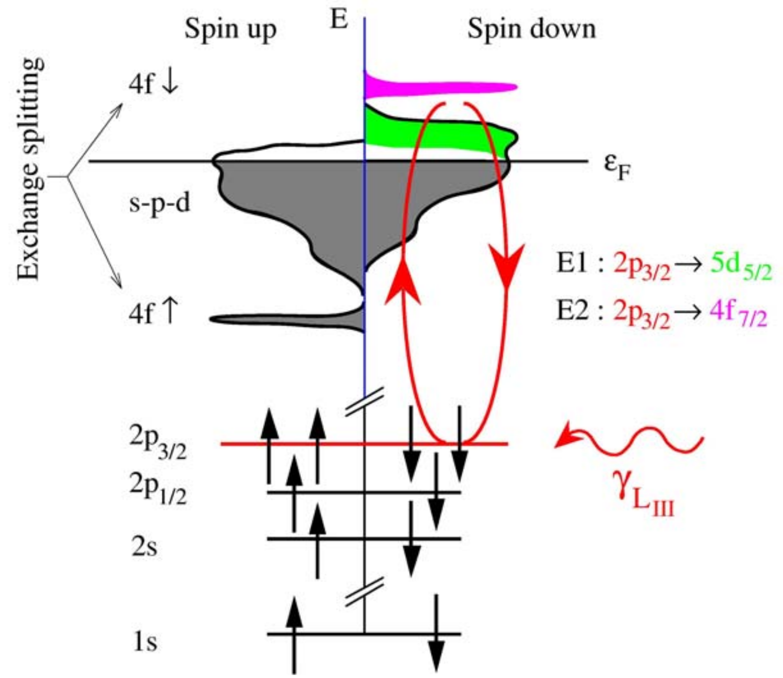
Bei 10 keV:

$$\frac{f^{spin}}{f^{charge}} = 0.02$$

resonant magnetic x-ray scattering

Störungstheorie 1. und 2. Ordnung

$$
\begin{aligned}
\left(\frac{d^2\sigma}{d\Omega dE}\right)^2 = r_0^2 \Bigg| & \left\langle b \middle| \sum_j e^{i\vec{K}\vec{r}_j} \middle| a \right\rangle \hat{\varepsilon}\cdot\hat{\varepsilon}' - i\frac{\hbar\omega}{mc^2}\left\langle b \middle| \sum_j e^{i\vec{K}\vec{r}_j}\vec{s} \middle| a \right\rangle \hat{\varepsilon}\times\hat{\varepsilon}' \\
& + \frac{\hbar^2}{m}\sum_c \sum_{ij} \left(\frac{\left\langle b \middle| \left(\frac{\vec{\varepsilon}\cdot\vec{P}_i}{\hbar} - i\left(\vec{k}'\times\vec{\varepsilon}'\right)\cdot\vec{s}_i\right) e^{-i\vec{k}'\vec{r}_i} \middle| c \right\rangle \left\langle c \middle| \left(\frac{\vec{\varepsilon}\cdot\vec{P}_j}{\hbar} + i\left(\vec{k}\times\vec{\varepsilon}\right)\cdot\vec{s}_j\right) e^{i\vec{k}\vec{r}_j} \middle| a \right\rangle}{E_a - E_c + \hbar\omega_{k'} - i\Gamma_c/2} \right. \\
& \left. + \frac{\left\langle b \middle| \left(\frac{\vec{\varepsilon}\cdot\vec{P}_j}{\hbar} + i\left(\vec{k}\times\vec{\varepsilon}\right)\cdot\vec{s}_i\right) e^{-i\vec{k}\vec{r}_i} \middle| c \right\rangle \left\langle c \middle| \left(\frac{\vec{\varepsilon}\cdot\vec{P}_j}{\hbar} - i\left(\vec{k}'\times\vec{\varepsilon}'\right)\cdot\vec{s}_j\right) e^{i\vec{k}'\vec{r}_j} \middle| a \right\rangle}{E_a - E_c - \hbar\omega_{k'}} \right) \Bigg|^2 \delta(E_a - E_b + \hbar\omega_k - \hbar\omega_{k'})
\end{aligned}
$$



K-Kante:	geringe Verstärkung
L-Kante:	Faktor 50-1000
M-Kante:	mehrere Größenordnungen bei Actiniden

resonant magnetic x-ray scattering

$$f\left(\vec{k},\vec{k}',\omega\right)=f^{charge}\left(\vec{Q}\right)+\underbrace{f'\left(\vec{k},\vec{k}',\omega\right)+if''\left(\vec{k},\vec{k}',\omega\right)}_{}+f^{spin}\left(\vec{k},\vec{k}',\omega\right)$$

abseits der Resonanzen

2. Ordnung

1. Ordnung

$$\hbar\omega >> E_c - E_a$$

$$\left(\frac{d^2\sigma}{d\Omega dE}\right)^2 = r_0^{\,2}\left|<b|\sum_j e^{i\vec{K}\vec{r}_j}\,|a>\hat{\varepsilon}\cdot\hat{\varepsilon}'-i\frac{\hbar\omega}{mc^2}<b|\sum_j e^{i\vec{K}\vec{r}_j}\left(\frac{i}{\hbar K}\vec{K}\times\vec{P}_j\cdot\vec{B}_L+\vec{s}_j\cdot\vec{B}_S\right)|a>\right|^2\delta(E_a-E_b+\hbar\omega_k-\hbar\omega_{k'})$$

$$\rightarrow \rho(K)\cdot B_\rho$$

$$\rightarrow -\frac{\hbar\omega}{mc^2}\left(\frac{1}{2}L(K)\cdot B_L+S(K)\cdot B_S\right)$$

polarisationsabhängige Faktoren

Neutronenstreuung

$$\left(\frac{d^2\sigma}{d\Omega dE}\right)^2 = (\gamma r_0)^2\,\frac{k'}{k}\left|<b|\sum_j e^{i\vec{K}\vec{r}_j}\left(-\frac{i}{\hbar k}\vec{K}\times\vec{P}_j+\vec{K}\times(\vec{s}_j\times\vec{K})\right)|a>\right|^2\delta\left(E_a-E_b+\frac{\hbar^2k^2}{2m_0}-\frac{\hbar^2k'^2}{2m_0}\right)$$

$$\rightarrow \left(\frac{1}{2}L(K)+S(K)\right)\cdot(K\times(\sigma\times K))$$

resonant magnetic x-ray scattering

$$<f_M> = -\frac{\hbar\omega}{mc^2} \begin{array}{c|cc|c} & \sigma & \pi & \\ \hline & (k \times k') \cdot S(Q) & \frac{Q^2}{2k^2}\left(\left(\frac{1}{2}L(Q)+S(Q)\right)\cdot k' + \frac{1}{2}L(Q)\cdot k\right) & \sigma' \\ & \frac{Q^2}{2k^2}\left(\left(\frac{1}{2}L(Q)+S(Q)\right)\cdot k + \frac{1}{2}L(Q)\cdot k'\right) & \left(\frac{Q^2}{2k^2}L(Q)+S(Q)\right)\cdot(k \times k') & \pi' \end{array}$$

Linear polarisierter einfallender Strahl

$$<f_M> = -i\frac{\lambda_C}{d}\begin{vmatrix} S_2\cos\theta & [(L_1+S_1)\cos\theta + S_3\sin\theta]\sin\theta \\ -[(L_1+S_1)\cos\theta + S_3\sin\theta]\sin\theta & [2L_2\sin^2\theta + S_2]\cos\theta \end{vmatrix}$$

Ladungsstreuung $<f_c> = \begin{vmatrix} 1 & 0 \\ 0 & \cos 2\theta \end{vmatrix}$

Hohe Energien (kleine Winkel)

$$\boxed{<f_M> = -i\frac{\lambda_C}{d}\begin{vmatrix} S_2 & 0 \\ 0 & S_2 \end{vmatrix}}$$ ← polarisationsunabhängig

Magnetischer Streuquerschnitt (E > 80 keV)

$$\boxed{\frac{d\sigma}{d\Omega} = r_0^2\left(\frac{\lambda_C}{d}\right)^2 |S_2|^2}$$

resonant magnetic x-ray scattering

E<4keV	E=4-20keV	E=30-60keV	E>80keV
„weiche“ Röntgenstrahlung	„harte“ Röntgenstrahlung		„hochenergetische“ Röntgenstrahlung

4d: Ca_2RuO_4

L-Kanten der 3d und 4d Übergangsmetalle

4f: $Eu_xGd_{1-x}S$

M-Kanten der Actinide

L-Kanten der 4f-(Seltene-Erd)-systeme
K-Kanten der 3d Übergangsmetalle

Übergangsmetall-Difluoride
MF_2

Resonante und nichtresonante Beugung mit Polarisationsanalyse

nichtresonante Beugung

Resonant:	Elementspezifisch / Phasenübergänge	
Nichtresonant:	L/S Verhältnis	S absolut / Phasenübergänge

Scattering scheme with polarization analysis

Non-resonant magnetic scattering amplitude [Blume & Gibbs]

$$f^{mag} = -i\frac{\hbar\omega}{mc^2}\begin{pmatrix} f^{\sigma\sigma'} & f^{\sigma\pi'} \\ f^{\pi\sigma'} & f^{\pi\pi'} \end{pmatrix}$$

$$= -i\frac{\hbar\omega}{mc^2}\begin{pmatrix} S_2 \sin 2\theta & -2\sin^2\theta\left[\cos\theta\left(L_1+S_1\right)-S_3\sin\theta\right] \\ 2\sin^2\theta\left[\cos\theta\left(L_1+S_1\right)+S_3\sin\theta\right] & \sin 2\theta\left[2L_2\sin^2\theta+S_2\right] \end{pmatrix}$$

Determination of L/S ratio
Magnetic structure determination

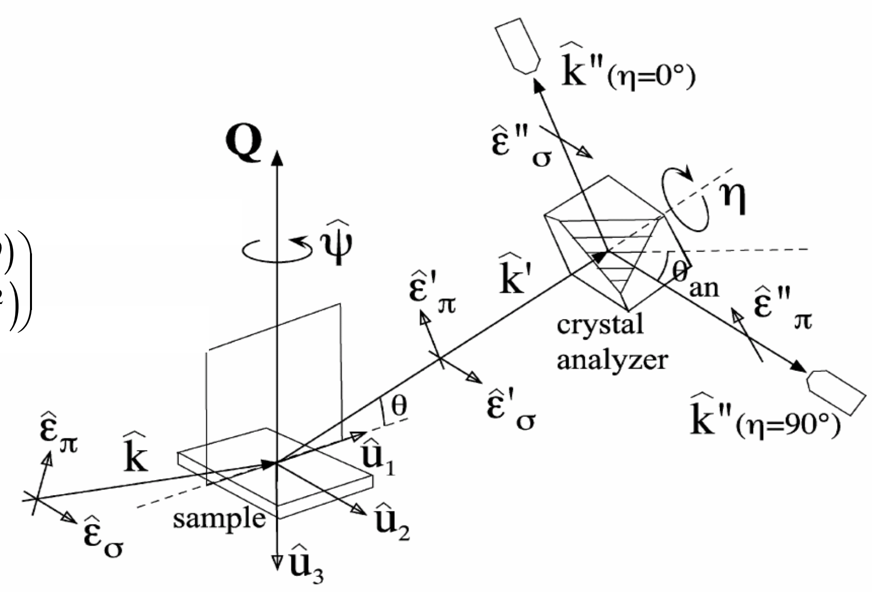
Resonant magnetic scattering amplitude (dipole transitions) [Hill & McMorrow]

$$f_{E1}^{res-mag} = \begin{pmatrix} f^{\sigma\sigma'} & f^{\sigma\pi'} \\ f^{\pi\sigma'} & f^{\pi\pi'} \end{pmatrix}$$

$$= F^0 - iF^1\begin{pmatrix} 0 & m_1\cos\theta + m_3\sin\theta \\ m_3\sin\theta - m_1\cos\theta & -m_2\sin 2\theta \end{pmatrix}$$

$$+ F^2\begin{pmatrix} m_2^2 & m_2\left(m_1\sin\theta - m_3\cos\theta\right) \\ m_2\left(m_1\sin\theta + m_3\cos\theta\right) & -\cos^2\theta\left(m_1^2\tan\theta + m_3^2\right) \end{pmatrix}$$

Strong intensities due to resonance enhancement
Element sensitivity at absorption edges
Magnetic structure determination



L/S determination

Non-resonant magnetic diffraction from $KCuF_3$
(Ciaruffo et al., PRB 65, 174455 (2002))

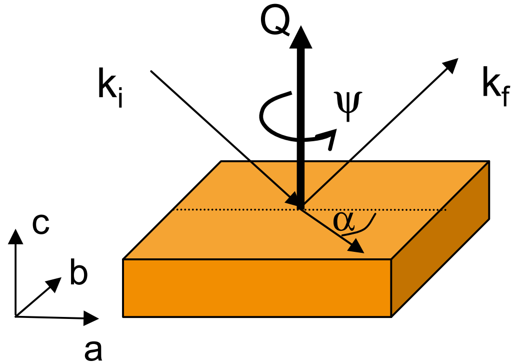
Magnetic moment in basal-plane (S_3=0)
L and S collinear: $S_1 = S\cos\alpha$

$S_2 = S\sin\alpha$

Scattering amplitude:

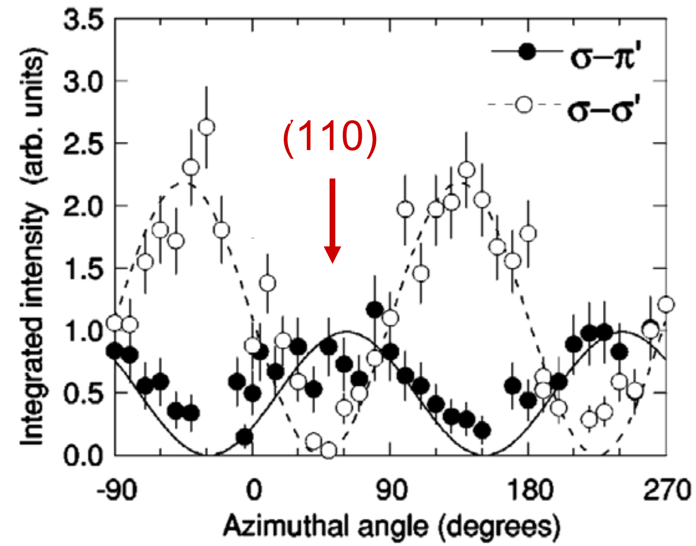
$$\begin{pmatrix} f_{\sigma\sigma'} \\ f_{\sigma\pi'} \end{pmatrix} = \begin{pmatrix} S\sin\alpha\sin 2\theta \\ -2(L+S)\cos\alpha\cos\theta\sin^2\theta \end{pmatrix}$$

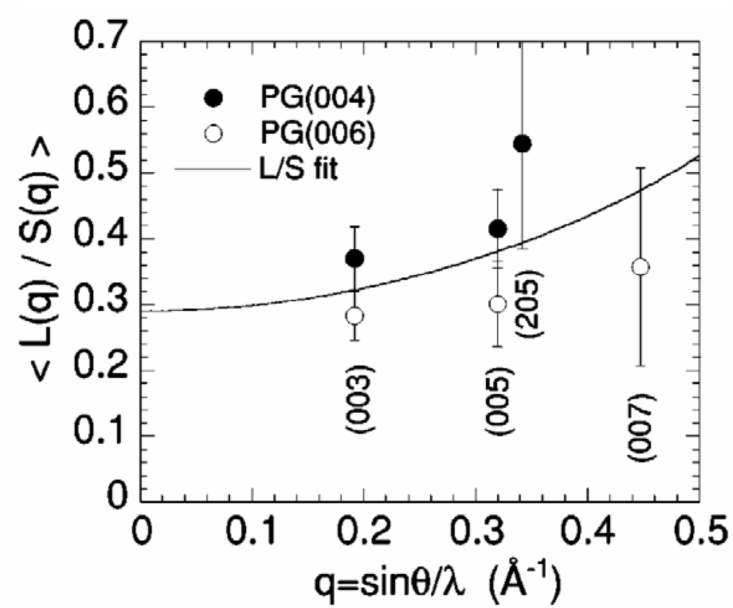
$$\frac{L(Q)}{S(Q)} = \frac{\tan\alpha}{\sin\theta}\sqrt{\frac{I_{\sigma\pi'}}{I_{\sigma\sigma'}}}$$



Magnetic moment || (110)

→ L/S=0.29(5)





exercises

Is it possible to observe resonant scattering from orbital order (magnetic order) in $LaMnO_3$ (lattice parameter 5.4 Angstroem) at the Mn L-edge?

At which position of (h,k,l) can magnetic scattering and scattering from orbital order be measured in $LaMnO_3$ and $La_{0.5}Ca_{0.5}MnO_3$?

Explain the principle of a polarization analyzer.