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

Vorlesung zum Haupt/Masterstudiengang Physik SS 2012

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# Materials Science

10. 5. correlated electron systems - structural properties

15. 5. correlated electron systems - magnetic properties

22. 5. high-Tc superconductors

24. 5. charge density waves

# 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
- magnetic x-ray scattering

# exchange interactions

combination of Coulomb interaction and Pauli principle

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

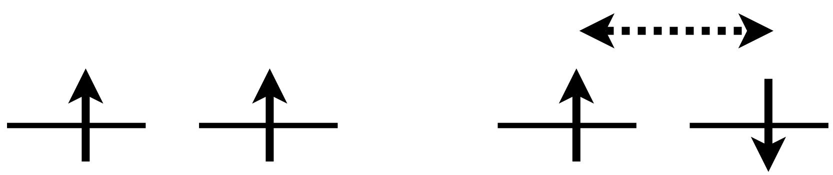
one-band Hubbard model:

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

$\quad = H_{kin} + H_U$

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

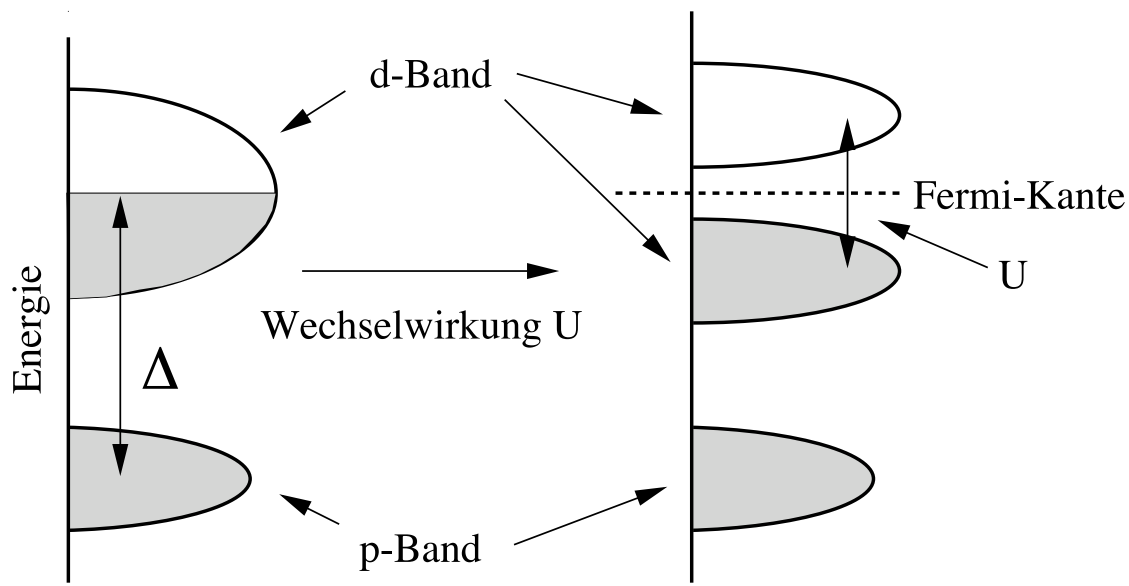
t >> U : metallic system
t << U : insulator with one electron per site



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

superexchange:
antiferromagnetic

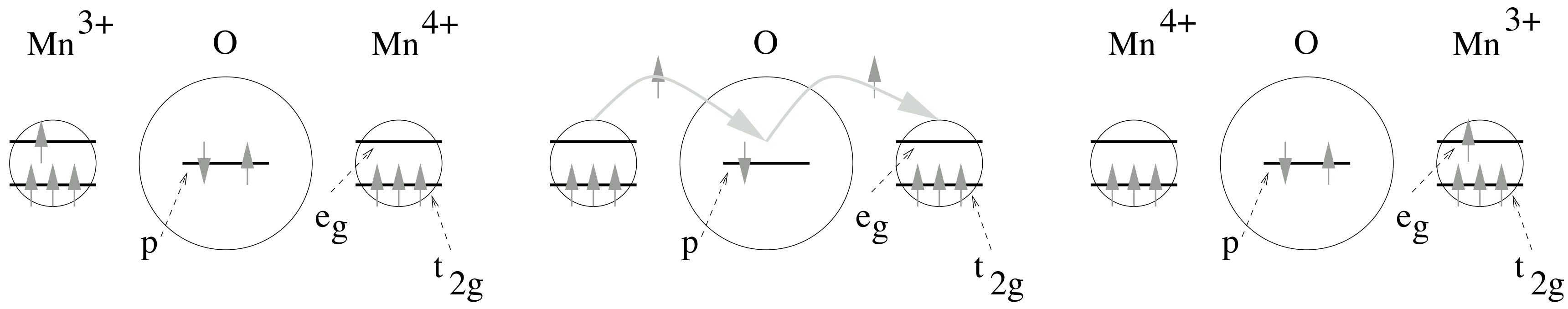
# Mott insulator




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

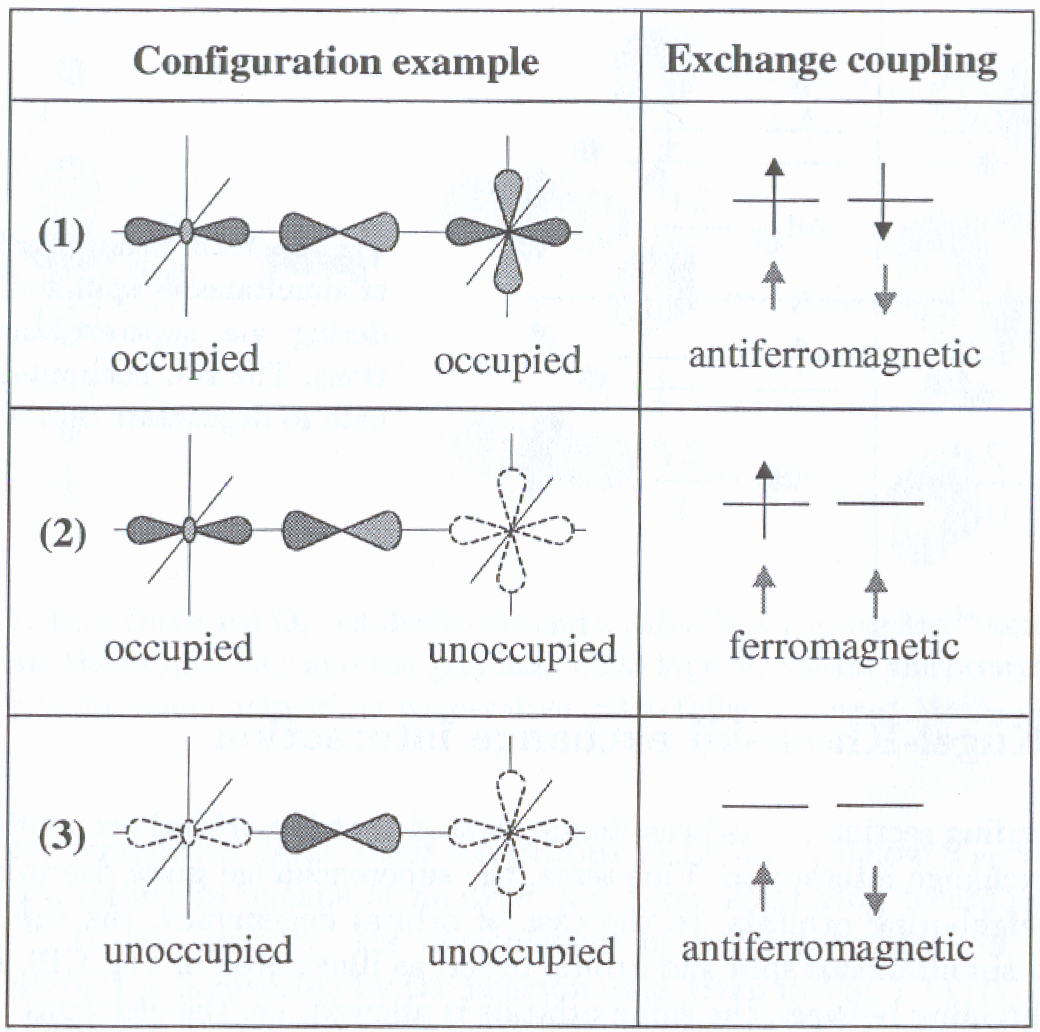
# double exchange interaction

ferromagnetic interaction between different ions due to Hund's coupling

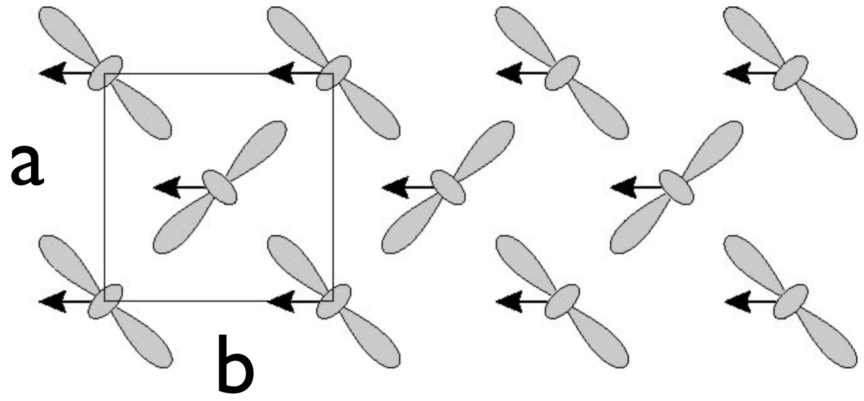


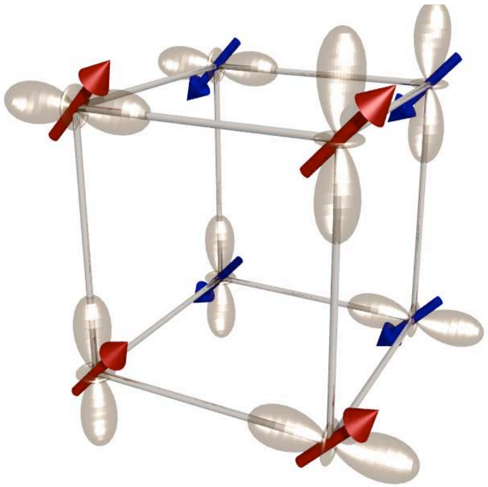
# GKA-rules (Goodenough-Kanamori-Andersen)

orbital dependent exchange interaction

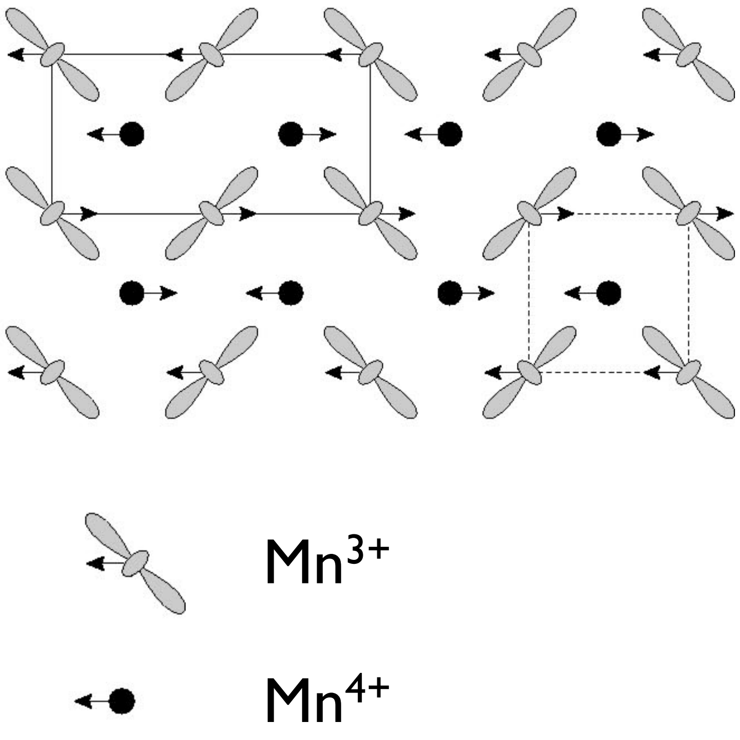


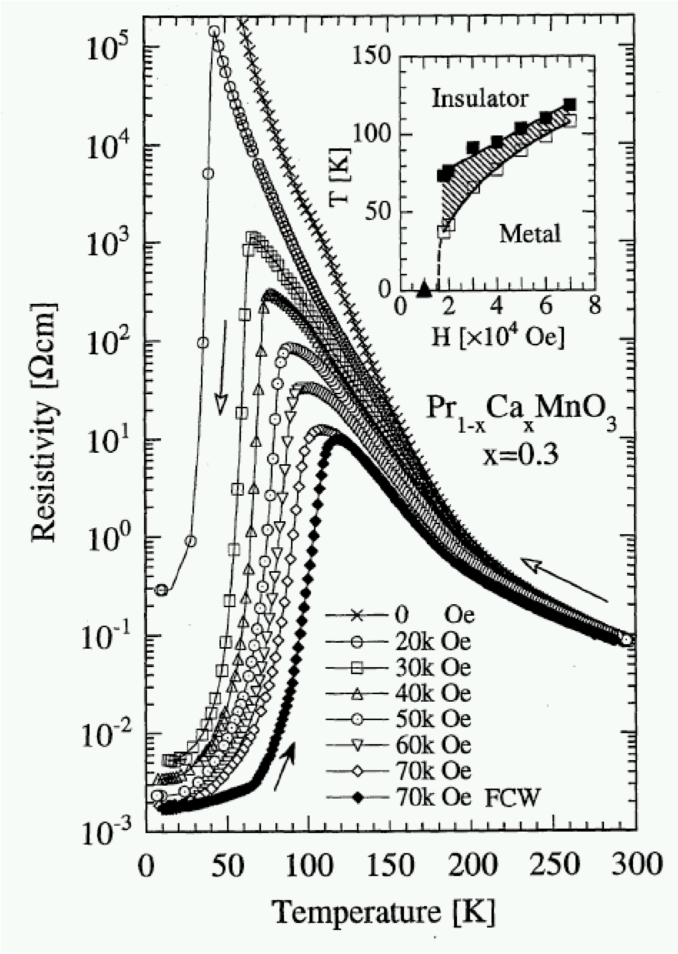
# magnetism of $LaMnO_3$



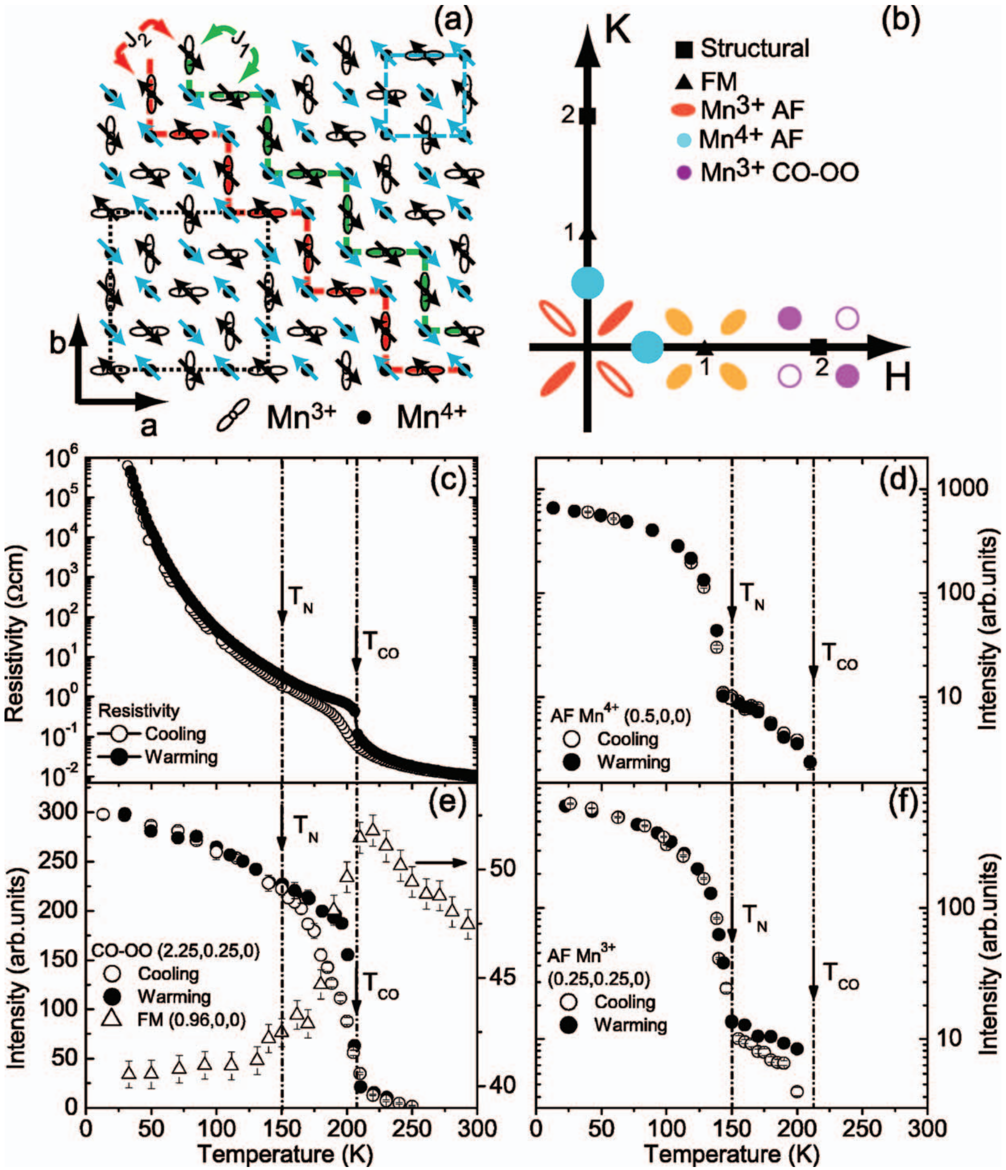



# magnetism of $La_{0.5}Ca_{0.5}MnO_3$ and colossal magneto resistance (CMR) effect



# magnetism of manganites



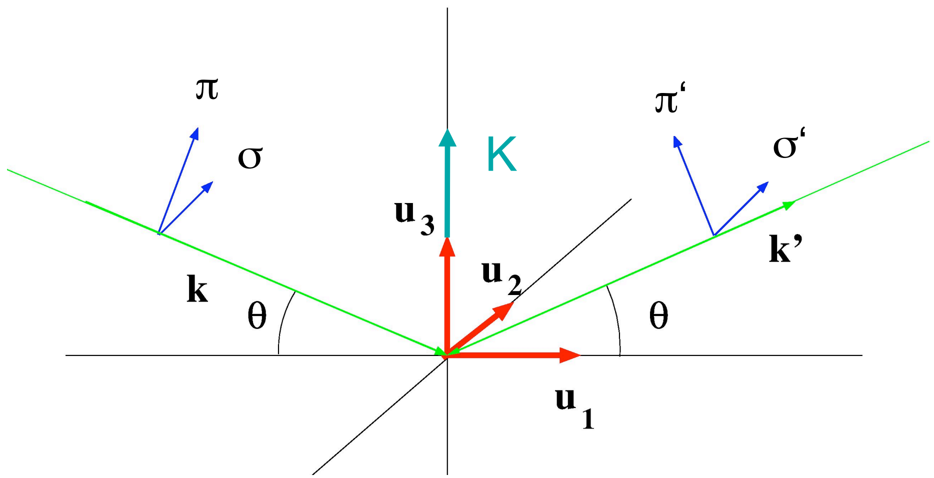

F. Ye et al.
Phys. Rev. B 72, 212404 (2005)

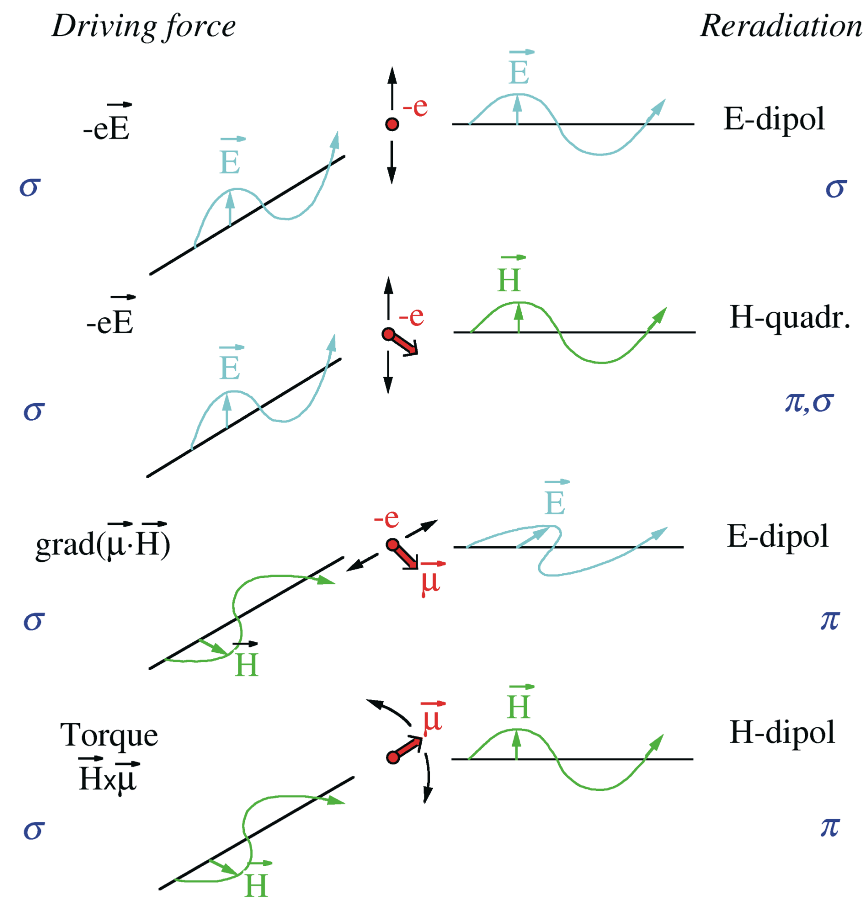
# magnetic x-ray scattering

Synchrotronstrahlung linear polarisiert in Ringebene

Streugeometrie vertikal

➔ σ – polarisierte einfallende Strahlung



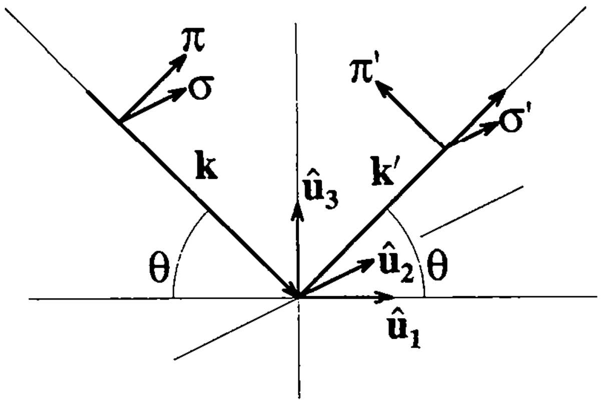
# magnetic x-ray scattering

$$d\sigma/d\Omega\big|_{\varepsilon\to\varepsilon'} = [e^2/mc^2]^2 \left|\langle M_C\rangle_{\varepsilon'\varepsilon} + i(\lambda_C/d)\langle M_M\rangle_{\varepsilon'\varepsilon}\right|^2$$

$\lambda_c = h/mc = 0.024$ Å electron Compton length

$M_M$:

| to \ from | $\sigma$ | $\pi$ |
|---|---|---|
| $\sigma'$ | $S_2\cos\theta$ | $[(L_1+S_1)\cos\theta + S_3\sin\theta]\sin\theta$ |
| $\pi'$ | $[-(L_1+S_1)\cos\theta + S_3\sin\theta]\sin\theta$ | $[2L_2\sin^2\theta + S_2]\cos\theta$; |




$M_C$:

| to \ from | $\sigma$ | $\pi$ |
|---|---|---|
| $\sigma'$ | $\rho(\mathbf{Q})$ | 0 |
| $\pi'$ | 0 | $\rho(\mathbf{Q})(\cos 2\theta)$. |

spin moment: $S = S_1\cdot\hat{u}_1 + S_2\cdot\hat{u}_2 + S_3\cdot\hat{u}_3$

orbital monment: $L = L_1\cdot\hat{u}_1 + L_2\cdot\hat{u}_2 + L_3\cdot\hat{u}_3$

charge density: $\rho(Q)$

# magnetic x-ray scattering

- ratio charge scattering / magnetic scattering = $(\lambda_c/d)^2 \sim 10^{-6}$
- x-ray diffraction allow to distinguish between orbital and spin contribution by polarization.
- Components S2 and L2 perpendicular to the scattering plane preserve the photon polarization.
- Components in the scattering plane components alter the polarization $\sigma \leftrightarrow \pi$.

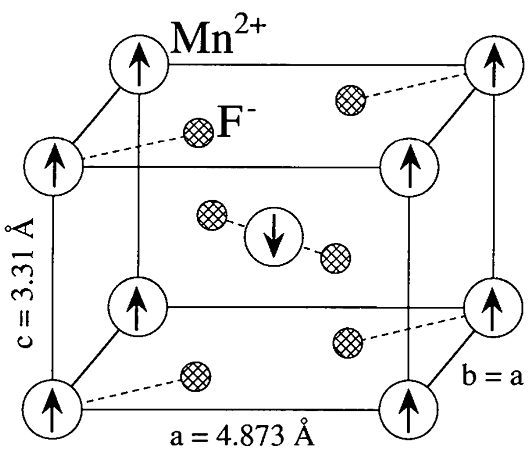
- high energy x-ray limit ( $\cos(\theta) \rightarrow 0$; E < 80 keV )

$$\mathrm{d}\sigma/\mathrm{d}\Omega\big|_{\varepsilon\to\varepsilon'} = r_0^2\left|\langle M_C\rangle_{\varepsilon\varepsilon'} + i(\lambda_C/d)\langle M_M\rangle_{\varepsilon\varepsilon'}\right|^2.$$

$$\langle M_M\rangle = \begin{array}{c|cc} & \sigma & \pi \\ \hline \sigma' & S_2 & 0 \\ \pi' & 0 & S_2 \end{array}.$$

$$(\mathrm{d}\sigma/\mathrm{d}\Omega)_{\mathrm{magnetic}} = r_0^2(\lambda_C/d)^2|S_2^2|.$$

# magnetic x-ray scattering: example $MnF_2$

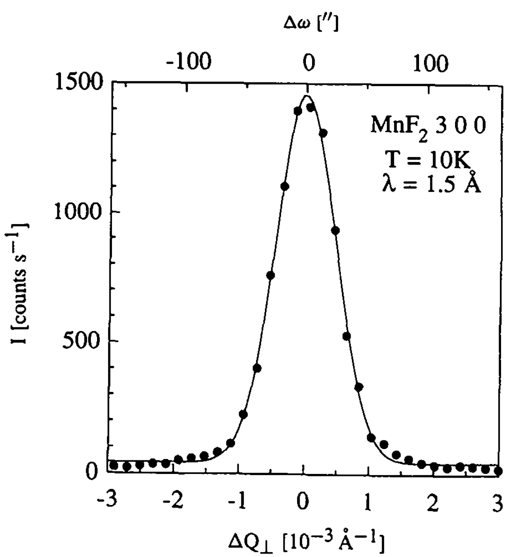



magnetic ordering wave vector: (1, 0, 0)

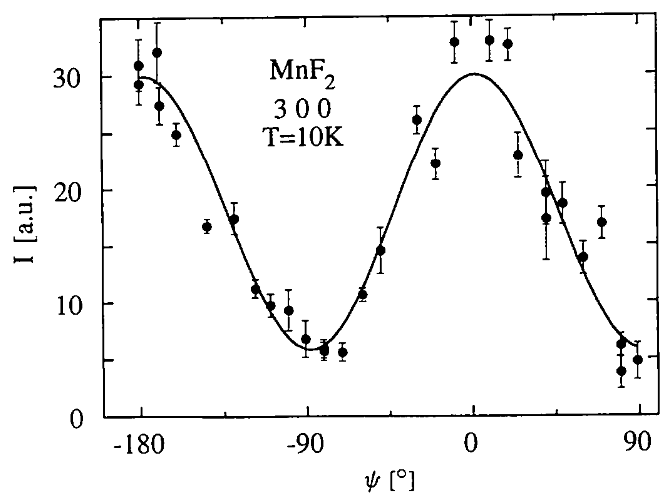
moment direction: || c-axis

T. Brückel et al.
Acta Cryst. (1996) A52, 427

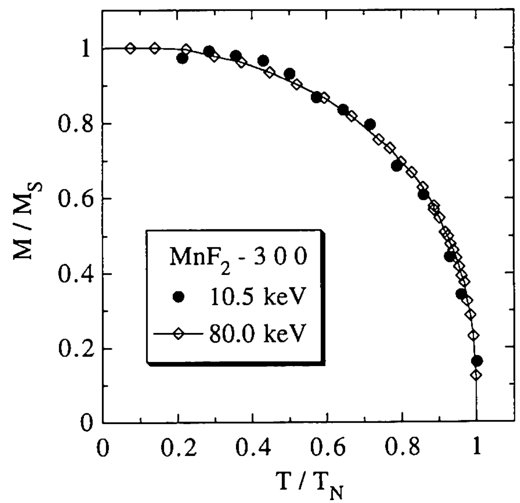
magnetic reflection




azimuthal dependence




temperature dependence



# 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

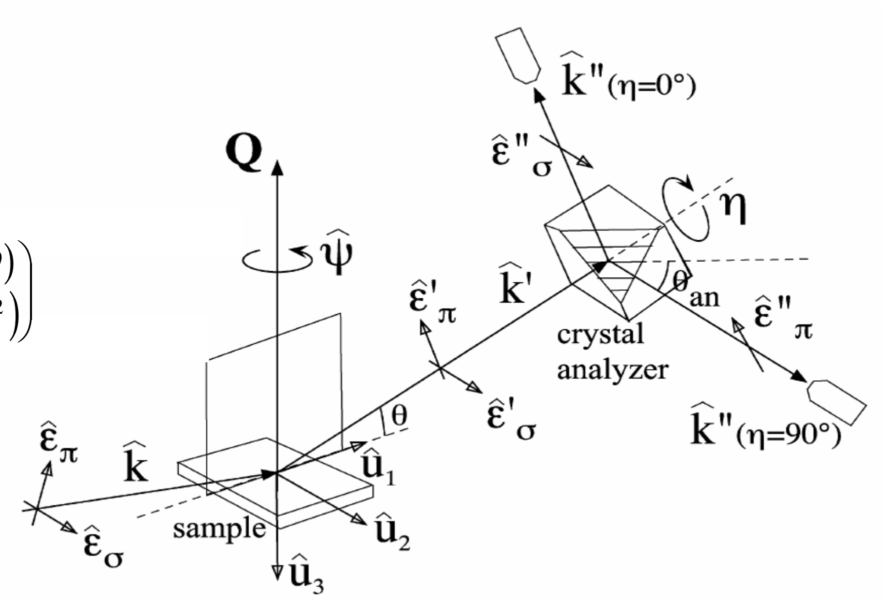
## Resonant magnetic scattering amplitude (dipole transitions) [Hill & McMorrow]

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

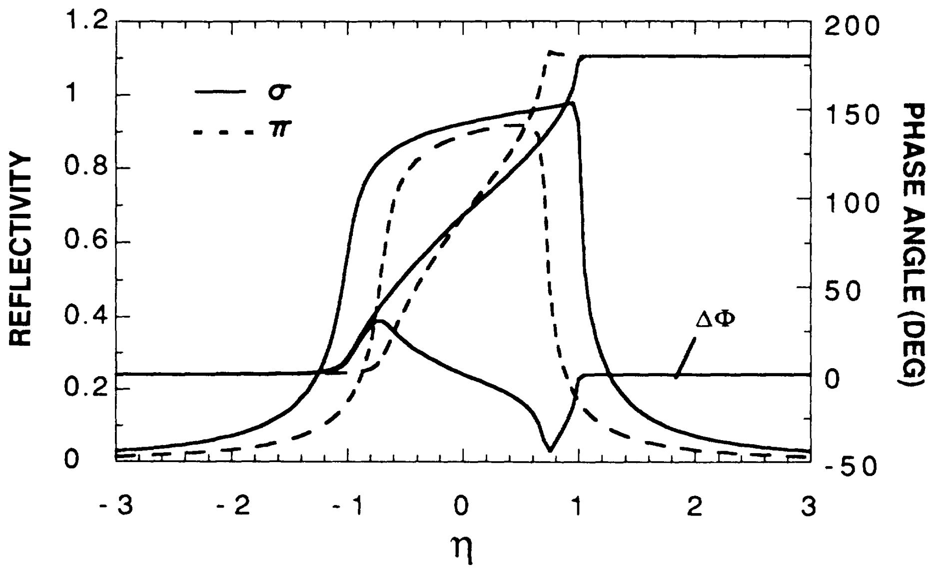
$$= F^0 - iF^1\begin{pmatrix} 0 & m_1\cos\theta + m_3\sin\theta \\ m_3\sin\theta - m_1\cos\theta & -m_2\sin 2\theta \end{pmatrix}$$

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

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



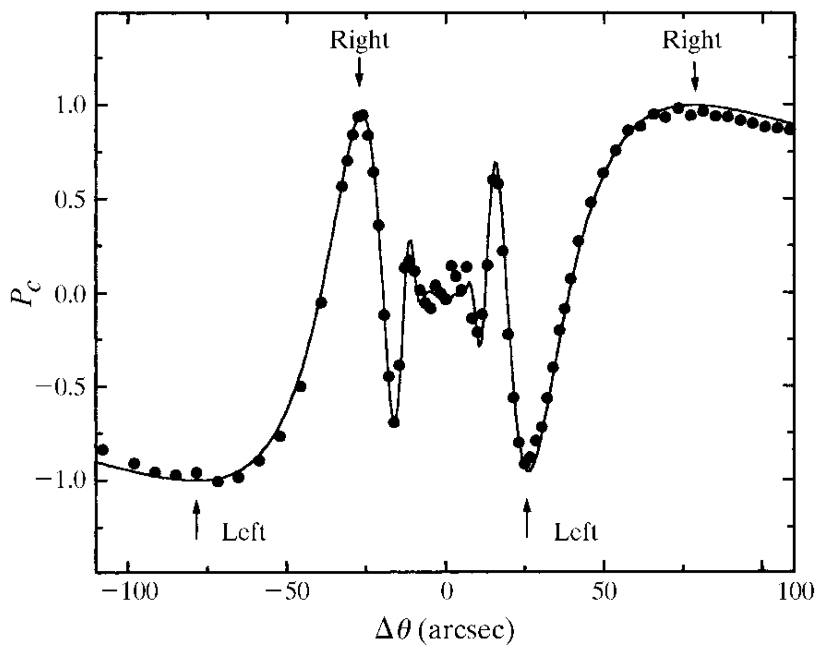
# quarter wave phase plate - circular polarization

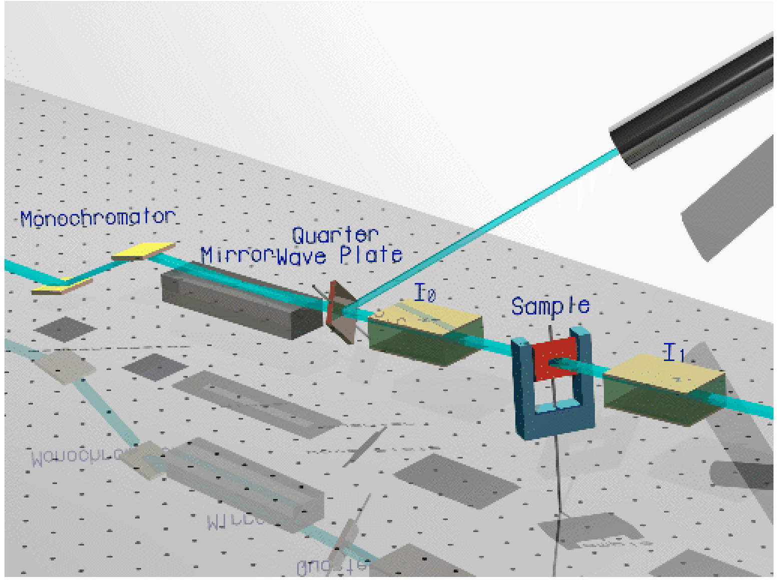



control of incident polarization:

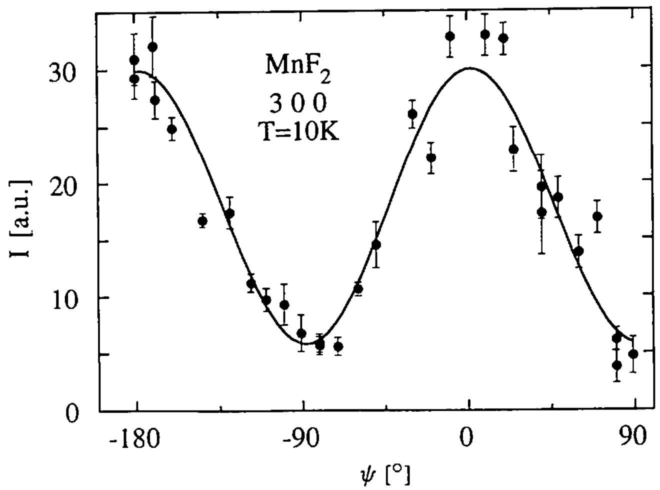
circular: left/right
linear: σ / π

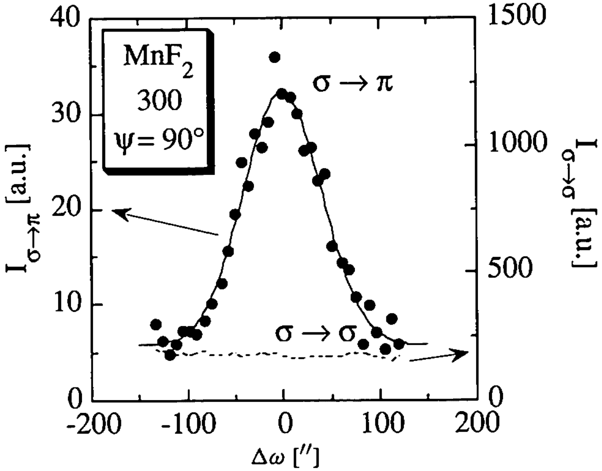
Battermann
Phys. Rev. B 45, 12677 (1992)



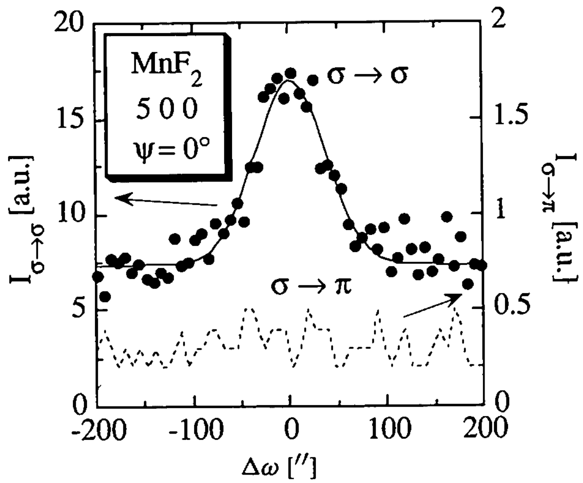



# polarization resolved magnetic scattering




(a)




T. Brückel et al.
Acta Cryst. (1996) A52, 427

# summary

- magnetic interactions in transition metal oxides
- Mott insulator
- colossal magneto resistance (CMR) effect
- magnetic x-ray scattering

# 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 $La_{0.5}Ca_{0.5}MnO_3$?

What does Erna need to do, if she wants to do polarization resolved x-ray diffraction on her rotating anode source.