# Magnetic dichroism in XAS and its application

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1. Introduction.

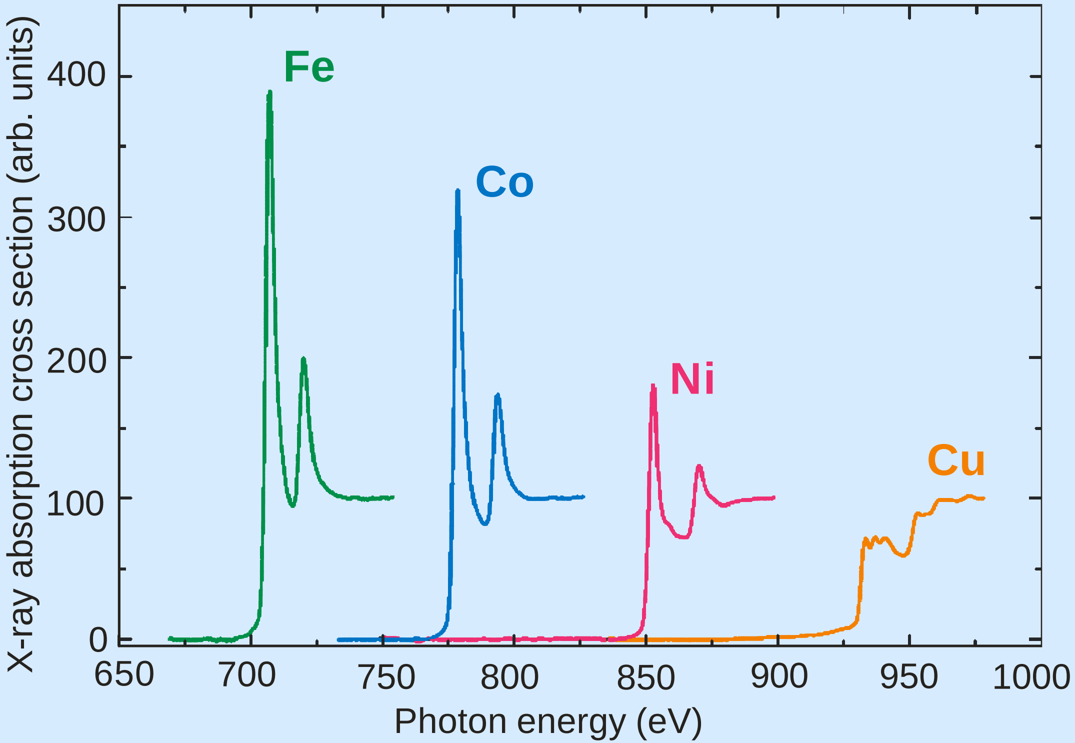
2. Element specific magnetizations in trilayers.

3. Determination of orbital- and spin- magnetic moments;
XMCD sum rules.
Magnetic Anisotropy Energy (MAE) and anisotropic $\mu_{orb}$.

4. Induced magnetism at interfaces.

# 1. Introduction: XAS

X-ray Absorption Spectroscopy is the most appropriate technique for element specific investigations.




$L_{3,2}$ edges of 3d elements

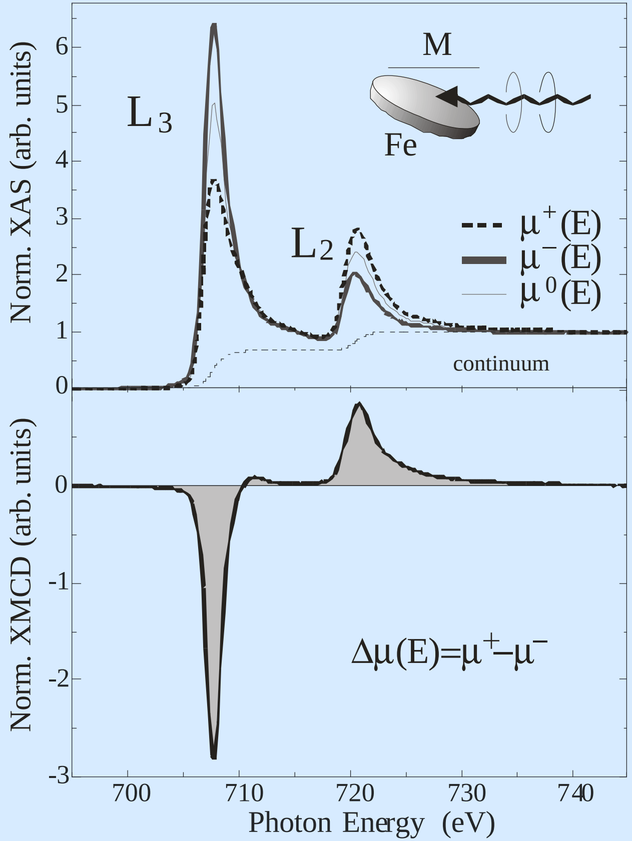
Note: the intensity of the 2p → 3d dipole transitions (E1) is proportional to the number of unoccupied final state (i.e. 3d- holes).

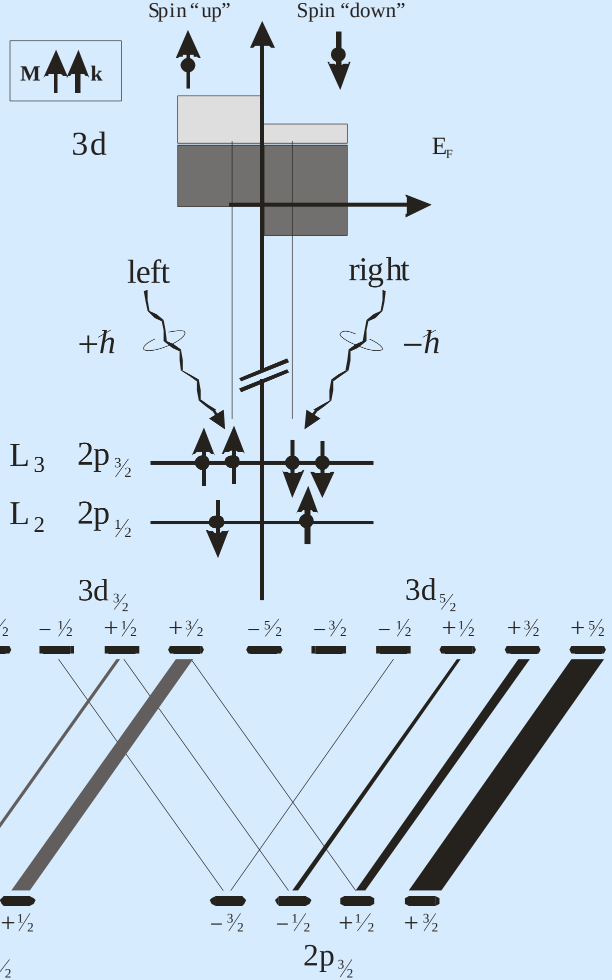
# Appendices/References

- In this lecture we will not discuss the equipment/apparatus, but rather focus on application of XAS in magnetism.
- Concerning XAS-technique there exist many review articles e.g. J. Stöhr: *NEXAFS Spectroscopy*, Springer Series in Surface Science **25**, 1992; H. Wende: *Recent advances in the x-ray absorption spectroscopy*, Rep. Prog. Physics **67**, 2105 2004.
- In the soft X-ray regime (VUV) one needs to work in vacuum. For nanomagnetism one wants to prepare and work anyway in UHV (*in situ* experiments).

# X-ray Magnetic Circular Dichroism

Faraday – effect in the X-ray regime (Gisela Schütz, 1987)



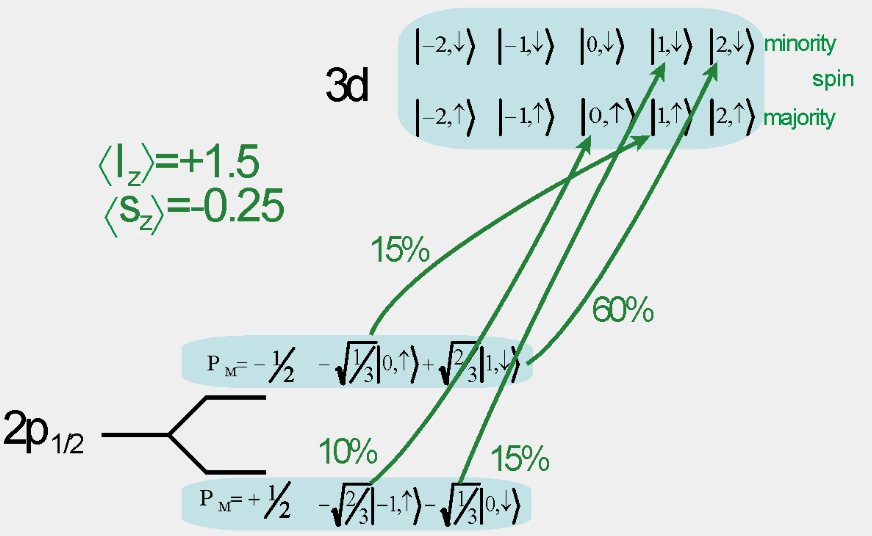




XMCD signal is a measure of the magnetization

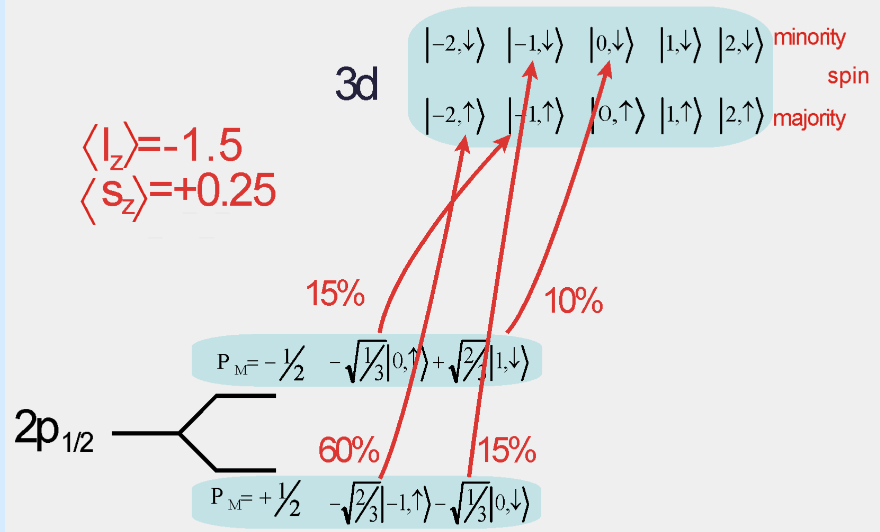
Many Reviews, e.g. H. Ebert Rep. Prog. Phys. **59**, 1665 (1996)

# The origin of MCD (after K. Fauth, Univ. Würzburg)

absorption of left circular pol. photon Δl=1, Δs=0

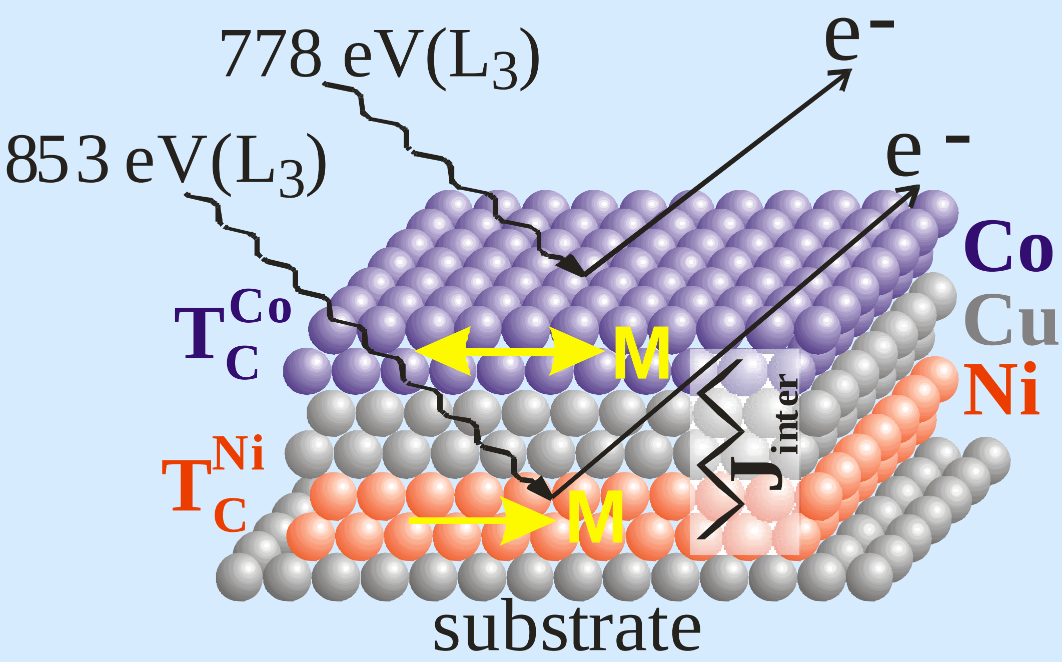



absorption of right circular pol. photon Δl=-1, Δs=0




There are many reviews e.g.: Lecture Notes in Physics Vol. 466 by H. Ebert, G. Schütz

# 2. Element specific magnetizations in trilayers




A trilayer is a prototype to study magnetic coupling in multilayers.

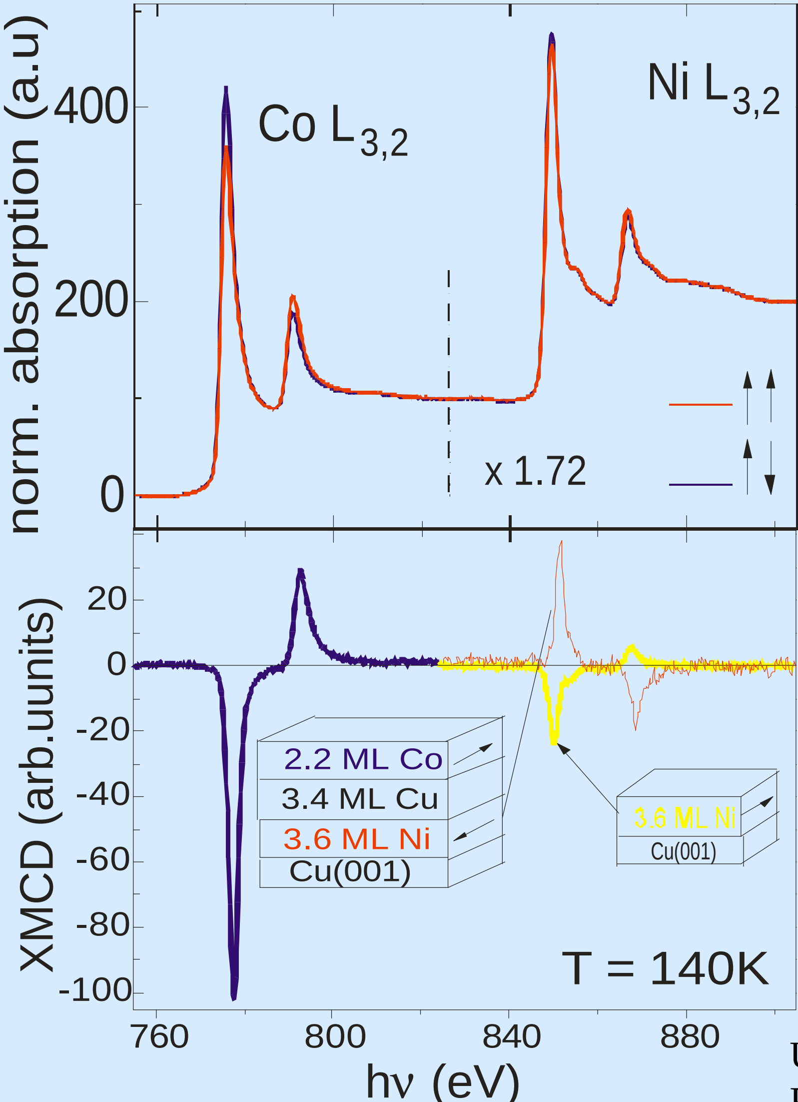
What about element specific Curie-temperatures ?

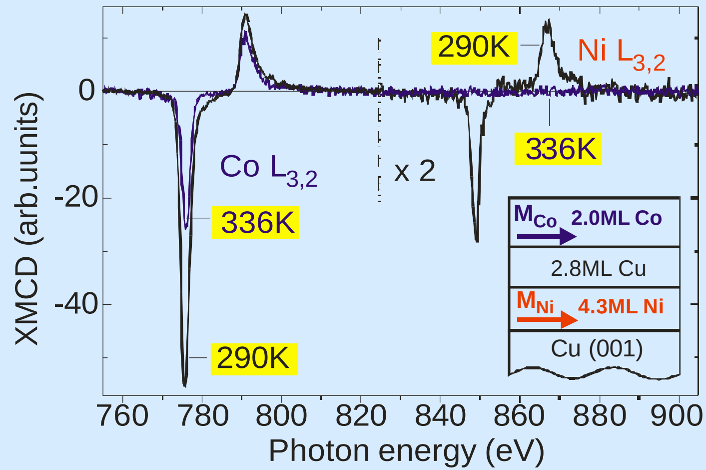
Two trivial limits: (i) $d_{Cu} = 0$ ⇒ direct coupling like a Ni-Co alloy

(ii) $d_{Cu}$ = large ⇒ no coupling, like a mixed Ni/Co powder

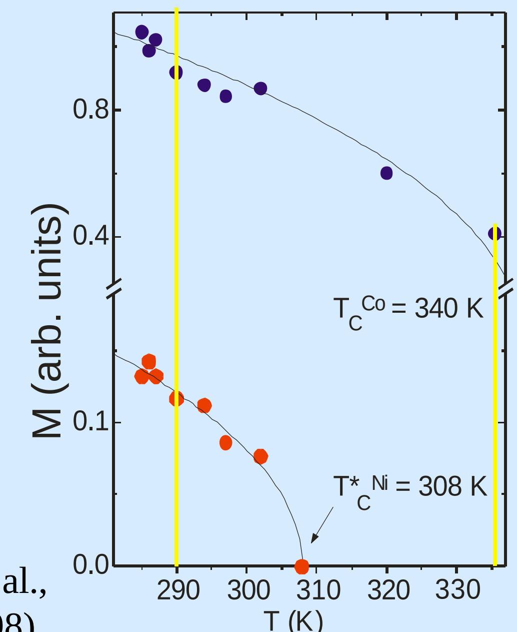
**BUT** $d_{Cu} \approx 2$ ML ⇒ **?**

# Ferromagnetic trilayers

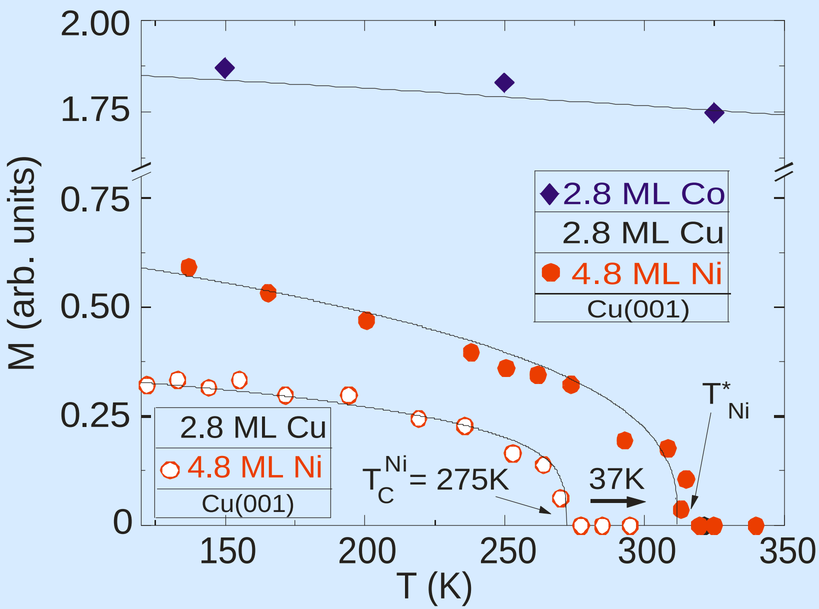


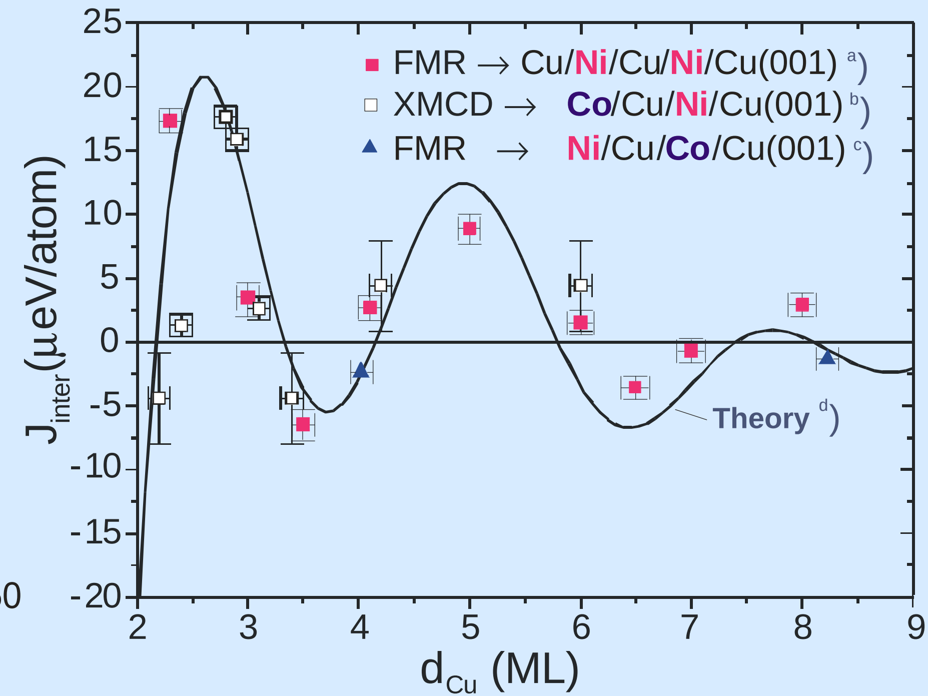




U. Bovensiepen et al.,
PRL **81**, 2368 (1998)



# Interlayer exchange coupling

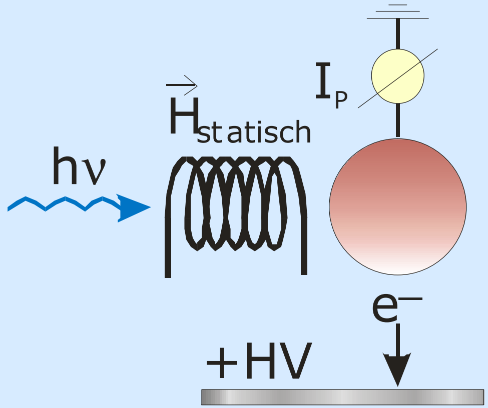


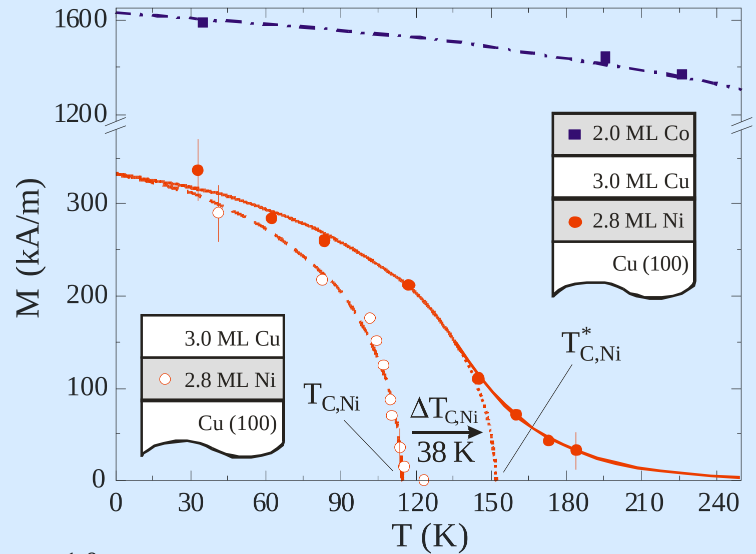

P. Poulopoulos, K. B., Lecture Notes in Physics **580**, 283 (2001)

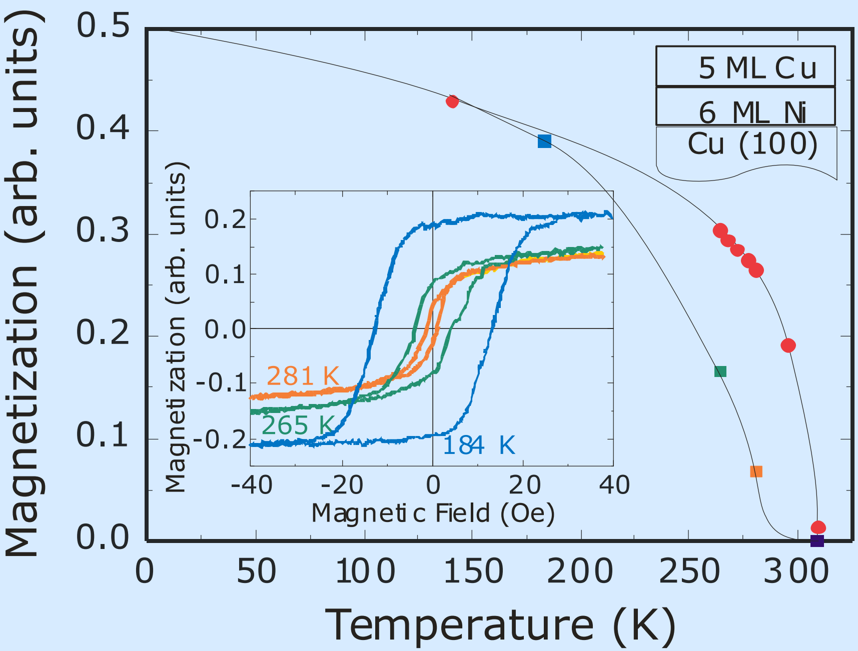
a) J. Lindner, K. B., J. Phys. Condens. Matter **15**, S465 (2003)
b) A. Ney et al., Phys. Rev. B **59**, R3938 (1999)
c) J. Lindner et al., Phys. Rev. B **63**, 094413 (2001)
d) P. Bruno, Phys. Rev. B **52**, 441 (1995)

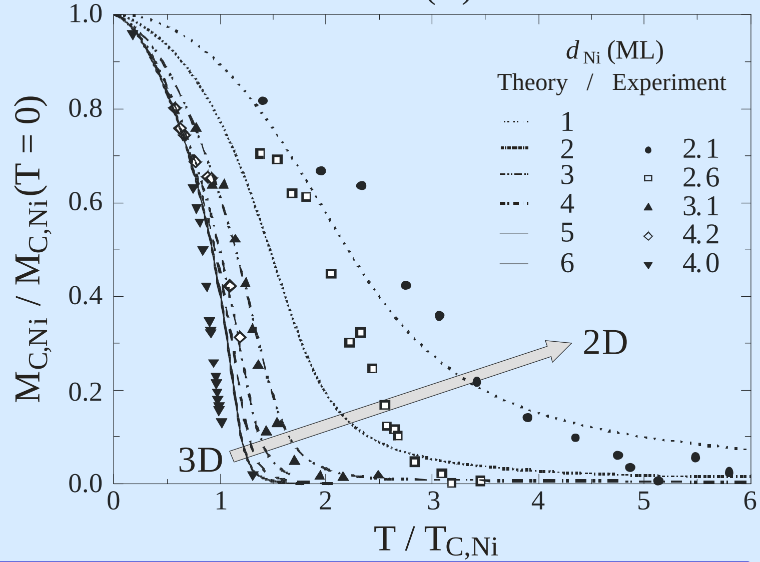
# Remanence and saturation magnetization




C. Sorg et al.,
XAFS XII, June 2003
Physica Scripta 2005

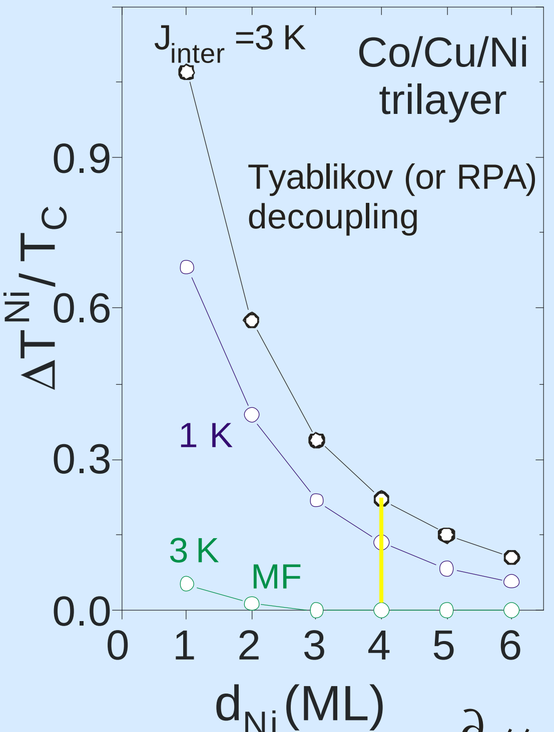


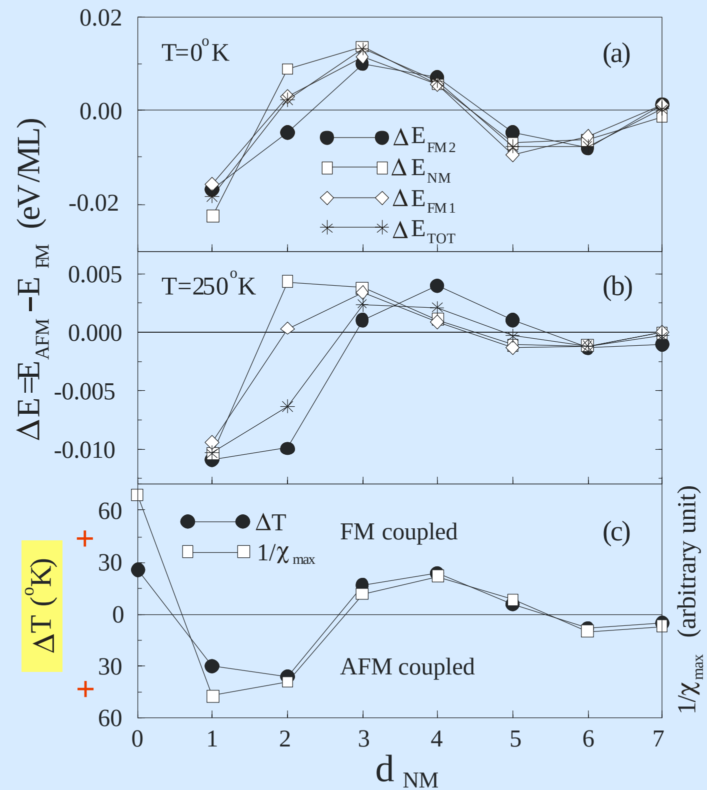



# Enhanced spin fluctuations in 2D (theory)

P. Jensen et al. PRB **60**, R14994 (1999)




J.H. Wu et al. J. Phys.: Condens. Matter **12** (2000) 2847




Spin-Spin correlation function $\frac{\partial}{\partial t}\langle\langle S_i^+ S_j^-\rangle\rangle \rightarrow$

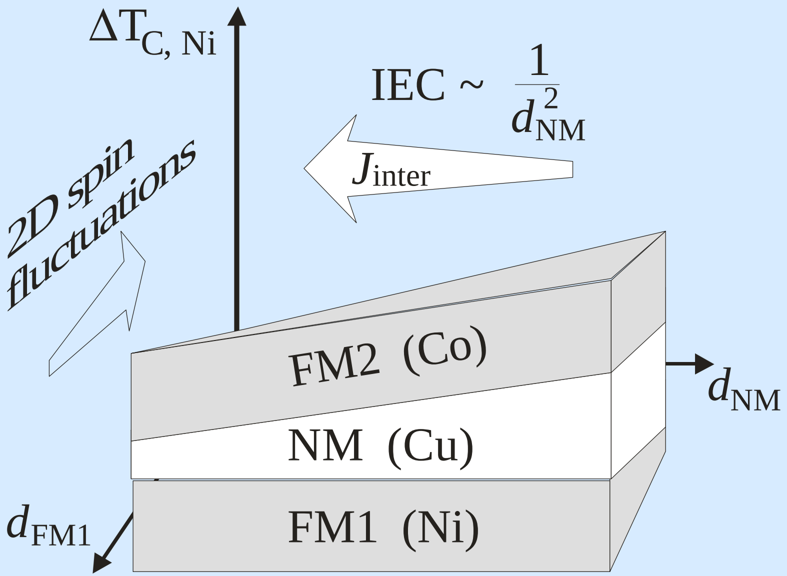
$$S_i^z S_j^+ \approx \langle S_i^z\rangle S_j^+ - \langle S_i^- S_i^+\rangle S_j^+ - \langle S_i^- S_j^+\rangle S_i^+ + \cdots$$

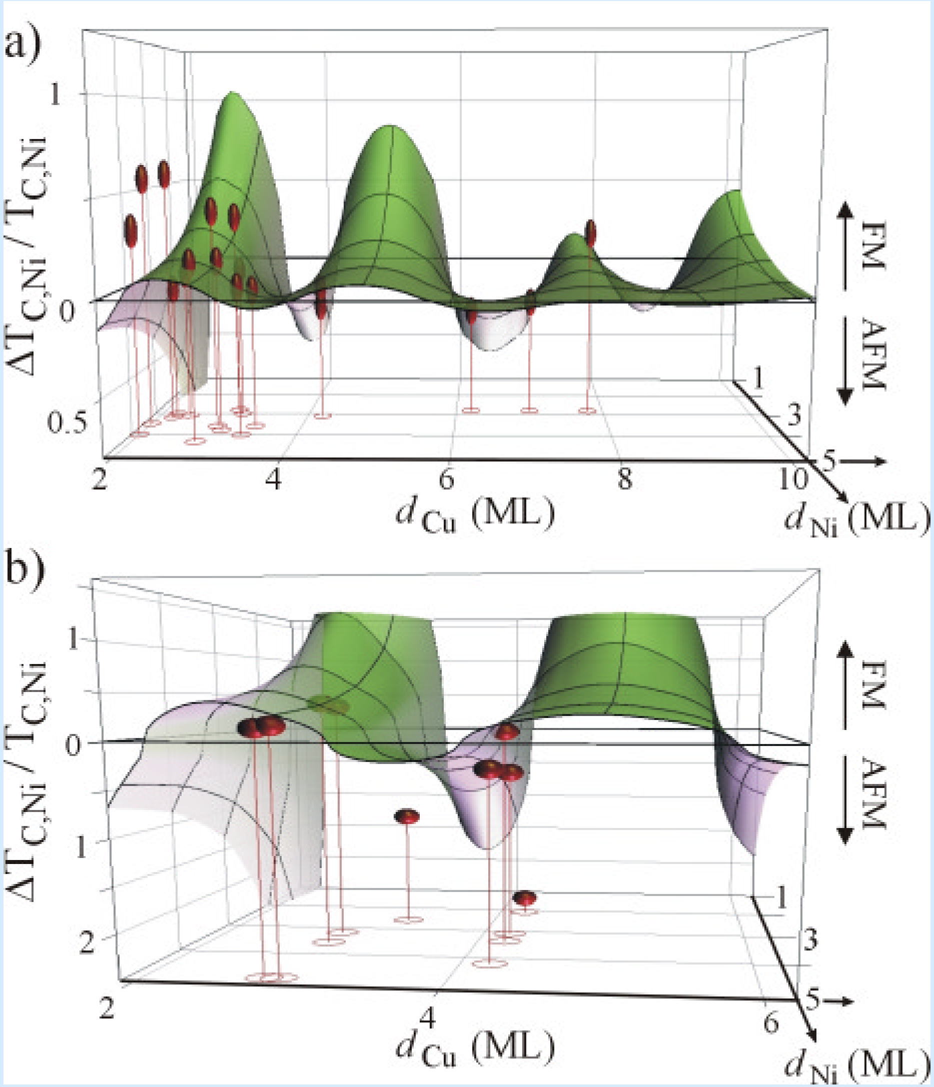
← RPA →

$\langle S_i^z\rangle S_j^+$, mean field ansatz (Stoner model) is insufficient to describe spin dynamics at interfaces of nanostructures

Single band Hubbard model:
Simple Hartree-Fock (Stoner) ansatz is insufficient
Higher order correlations are needed to explain $T_C$-shift

# Evidence for giant spin fluctuations (to be published)



# 3. Determination of orbital- and spin- magnetic moments

**Which technique measures what?**

**$\mu_L$ , $\mu_S$ in UHV-XMCD**

**$\mu_L + \mu_S$ in UHV-SQUID**

**$\mu_L / \mu_S$ in UHV-FMR**

For FMR see: J. Lindner and K. Baberschke
*In situ Ferromagnetic Resonance:*
*An ultimate tool to investigate the coupling in ultrathin magnetic films*
J. Phys.: Condens. Matter **15**, R193 (2003)

*per definition:*

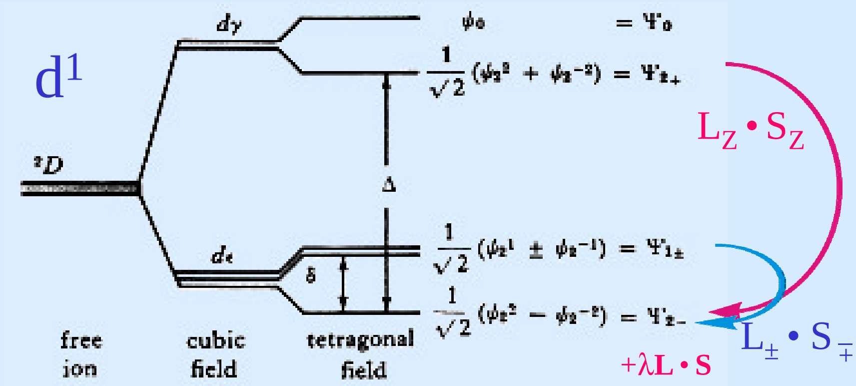
1) spin moments are isotropic

2) also exchange coupling $\mathbf{J\ S_1 \cdot S_2}$ is isotropic

3) so called anisotropic exchange is a (hidden) projection of the orbital momentum into spin space

# Orbital magnetism in second order perturbation theory

d[1]




Splitting of the ²D term by a tetragonally distorted cubic field.

$$y_{2-} \equiv (2)^{-1/2}\{|2\rangle - |-2\rangle\} \equiv |2-\rangle$$

The orbital moment is quenched in cubic symmetry

$\langle 2- | L_Z | 2- \rangle = 0,$

but not for tetragonal symmetry

effective Spin Hamiltonian

$g_{exp}$ $\qquad B_2^0 \rightarrow K_2^0$

$$\mathscr{H} = \sum_{i,j=1}^{3} [\beta g_e(\delta_{ij} - 2\lambda\Lambda_{ij})S_iH_j - \lambda^2\Lambda_{ij}S_iS_j]$$
$$+ \text{ diamagnetic terms in } H_iH_j \qquad (3\text{-}23)$$

where $\Lambda_{ij}$ is defined in relation to states $(n > 0)$ as

$$\Lambda_{ij} = \sum_{n \neq 0} \frac{(0|L_i|n)(n|L_j|0)}{E_n - E_0} \qquad (3\text{-}24)$$

$\langle o|\mu_B \mathbf{H}\cdot\mathbf{L}|n\rangle \langle n|\lambda \mathbf{L}\cdot\mathbf{S}|o\rangle$ $\qquad$ $\langle o|\lambda \mathbf{L}\cdot\mathbf{S}|n\rangle \langle n|\lambda \mathbf{L}\cdot\mathbf{S}|o\rangle$

In the principal axis system of a crystal with axial symmetry, the $\underline{\mathbf{\Lambda}}$ tensor is diagonal with $\Lambda_{zz} = \Lambda_\parallel$ and $\Lambda_{xx} = \Lambda_{yy} = \Lambda_\perp$. Under these conditions, $\mathscr{H}$ of (3-23) can be simplified, since

$$S_x^2 + S_y^2 = S(S+1) - S_z^2$$

to give

$$\mathscr{H} = g_\parallel \beta H_z S_z + g_\perp \beta(H_xS_x + H_yS_y) + D[S_z^2 - \tfrac{1}{3}S(S+1)] \qquad (3\text{-}25)$$

where

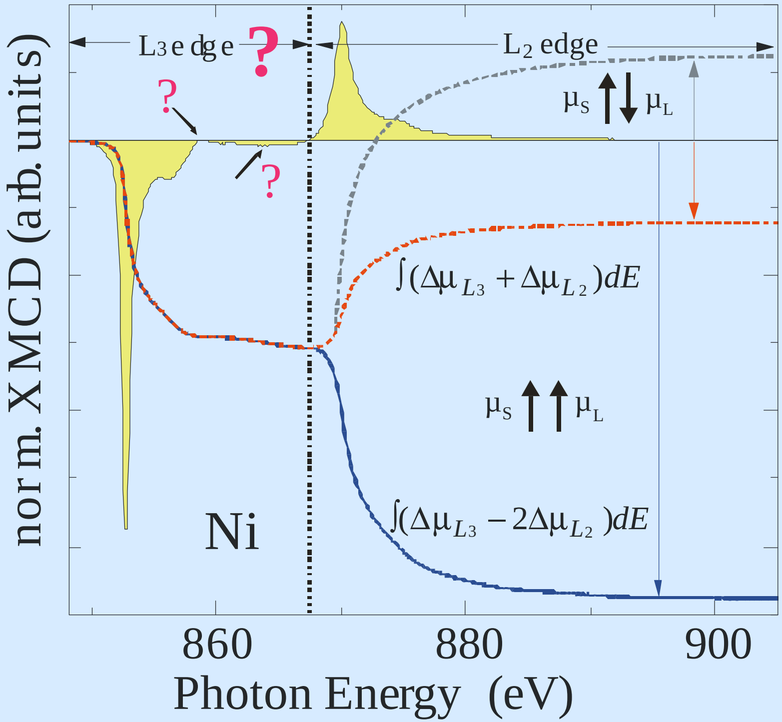
$$g_\parallel = g_e(1 - \lambda\Lambda_\parallel)$$
$$g_\perp = g_e(1 - \lambda\Lambda_\perp) \qquad (3\text{-}26)$$
$$D = \lambda^2(\Lambda_\perp - \Lambda_\parallel)$$

G.E. Pake, p.66

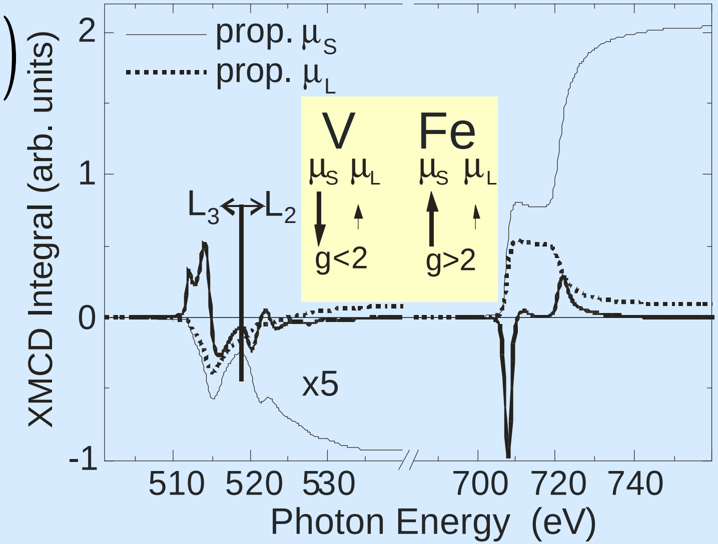
# Orbital and spin magnetic moments deduced from XMCD

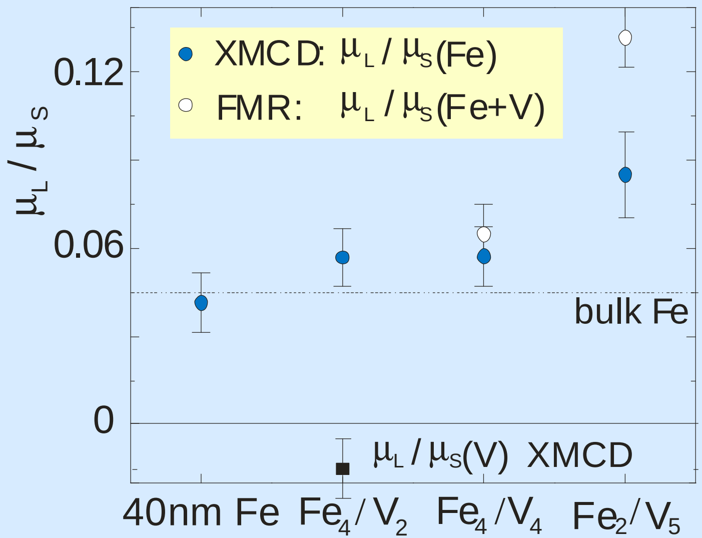
$$\int\left(?\,\mu_{L3} - 2\cdot ?\,\mu_{L2}\right)dE = \frac{N}{3N_h^d}\left(2\langle S_z\rangle^d + 7\langle T_z\rangle^d\right)$$

$$\int\left(?\,\mu_{L3} + ?\,\mu_{L2}\right)dE = \frac{N}{2N_h^d}\langle L_z\rangle^d$$

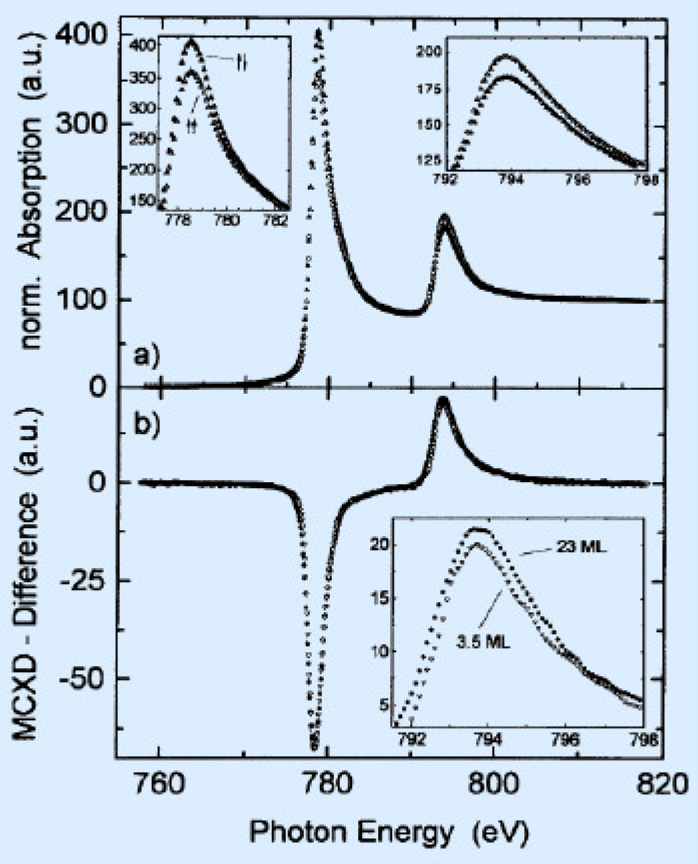


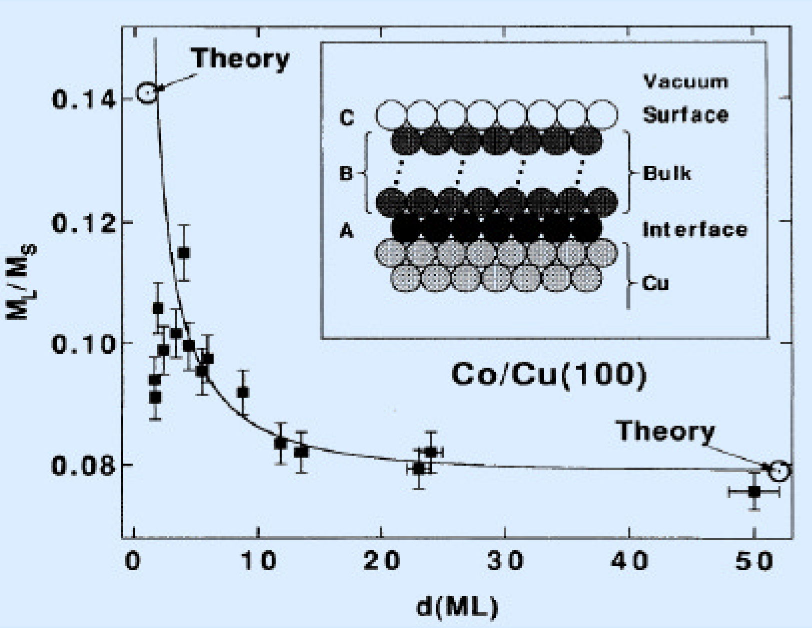

H. Ebert Rep. Prog. Phys. **59**, 1665 (1996)



# Enhancement of Orbital Magnetism at Surfaces: Co on Cu(100)



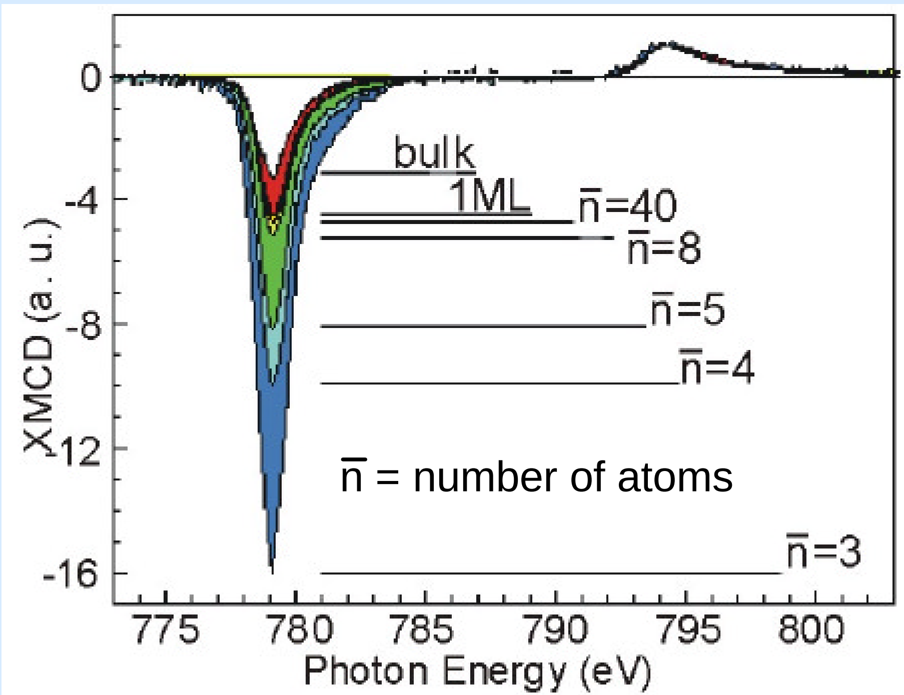




$$\left(\frac{M_L}{M_S}\right)_{\exp} = \frac{Ae^{-D(d-1)/l} + B\Sigma_{n=3}^{d} e^{-D(n-2)/l} + C}{\Sigma_{n=0}^{d-1} e^{-nD/l}}$$

M. Tischer et al., Phys. Rev. Lett. **75**, 1602 (1995)

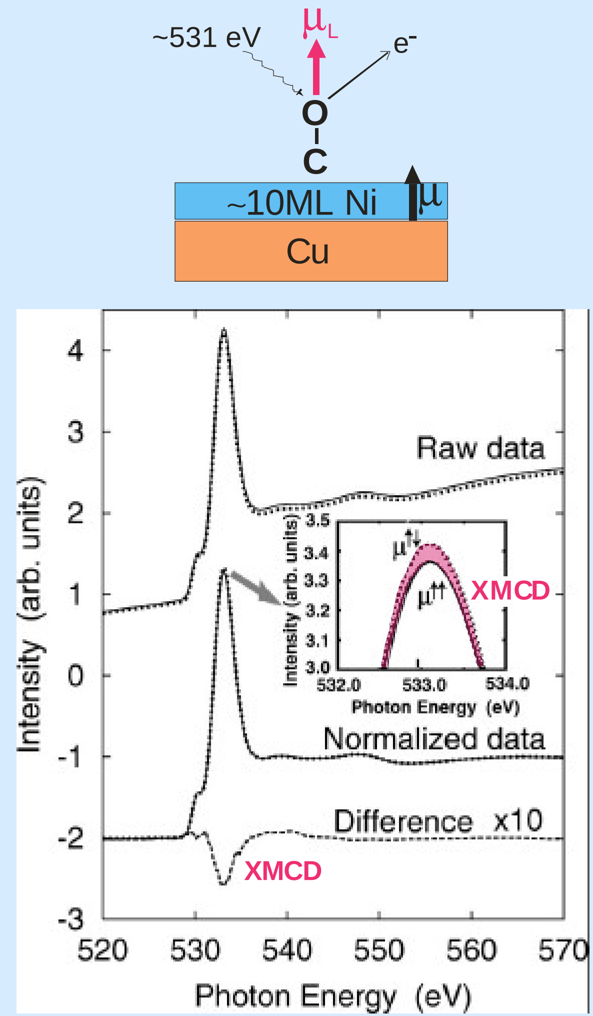
# Giant Magn. Anisotropy of Single Co Atoms and Nanoparticles

P. Gambardella et al., Science **300**, 1130 (2003)




# Induced magnetism in molecules

T. Yokoyama et al., PRB **62**, 14191 (2000)



# Magnetic Anisotropy Energy (MAE) and anisotropic $\mu_{orb}$

$$g_{||} - g_{\perp} = g_e \lambda(\Lambda_{\perp} - \Lambda_{||})$$

anisotropic $\mu_L \leftrightarrow$ MAE

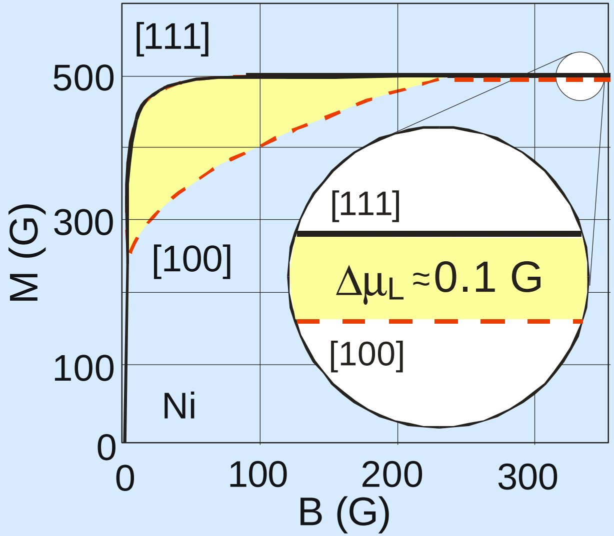
$$D = \frac{\lambda}{g_e} \Delta g$$

$\updownarrow$

$$MAE \propto \frac{\xi_{LS}}{4\mu_B} \Delta\mu_L$$

Bruno ('89)

1. Magnetic anisotropy energy = f(T)
2. Anisotropic magnetic moment ≠ f(T)




*Characteristic energies of metallic ferromagnets*

| | |
|---|---|
| binding energy | 1 - 10 eV/atom |
| exchange energy | 10 - $10^3$ meV/atom |
| cubic MAE (Ni) | 0.2 μeV/atom |
| uniaxial MAE (Co) | 70 μeV/atom |

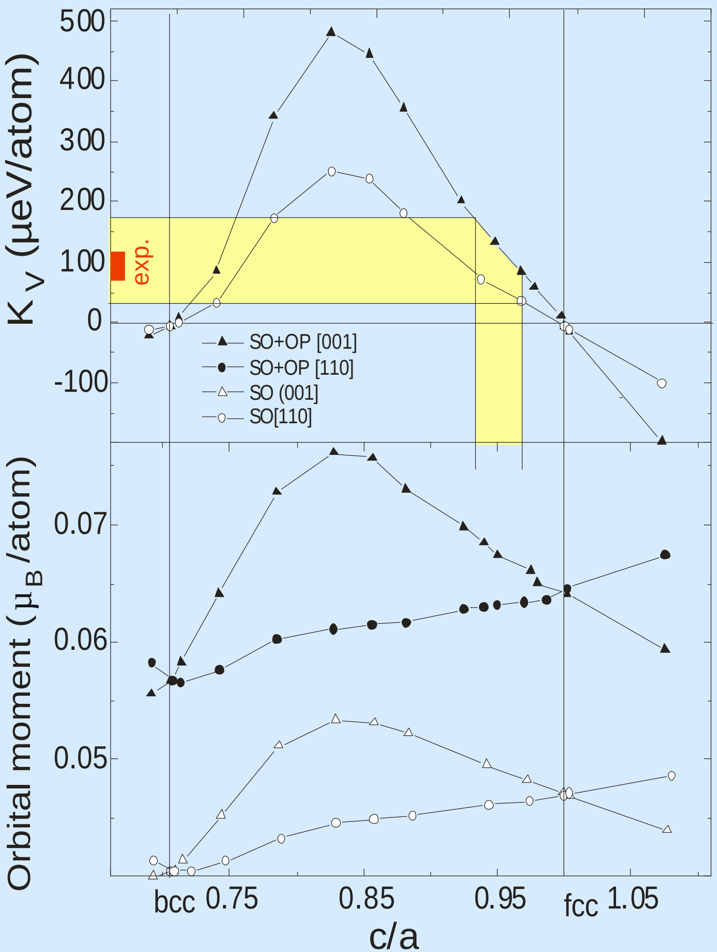
MAE = ?M·dB ˜ ½ ?M·?B ˜ ½ 200 · 200 $G^2$

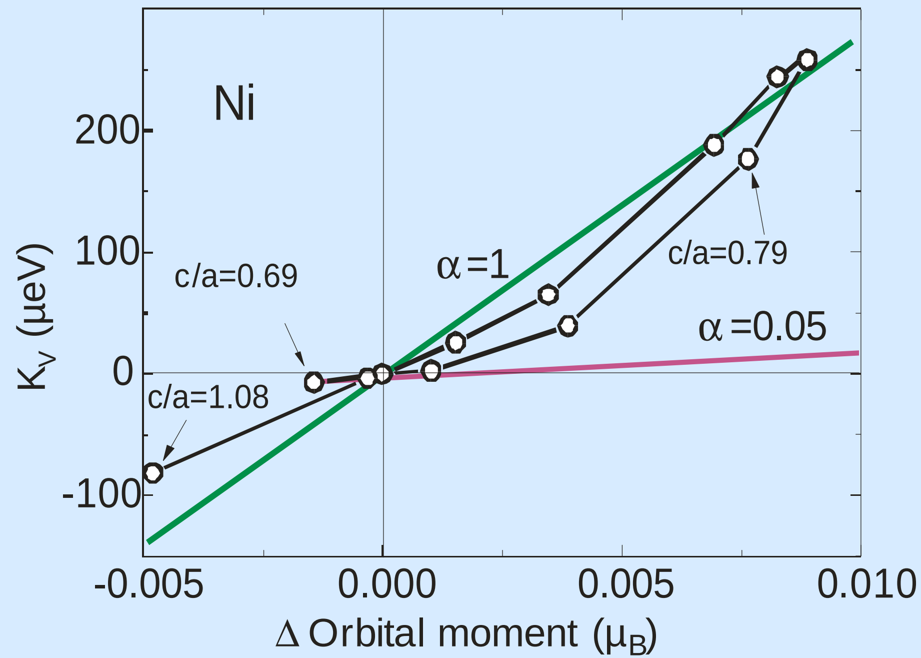
MAE ˜ $2 \cdot 10^4$ erg / $cm^3$ ˜ 0.2 μeV / atom

≈ 1μeV/atom is very small compared to
≈ 10 eV/atom total energy but all important

K. Baberschke, Lecture Notes in Physics, Springer **580**, 27 (2001)

# Magnetic Anisotropy Energy MAE and anisotropic $\mu_{orb}$




O. Hjortstam, K. B. et al. PRB **55**, 15026 ('97)

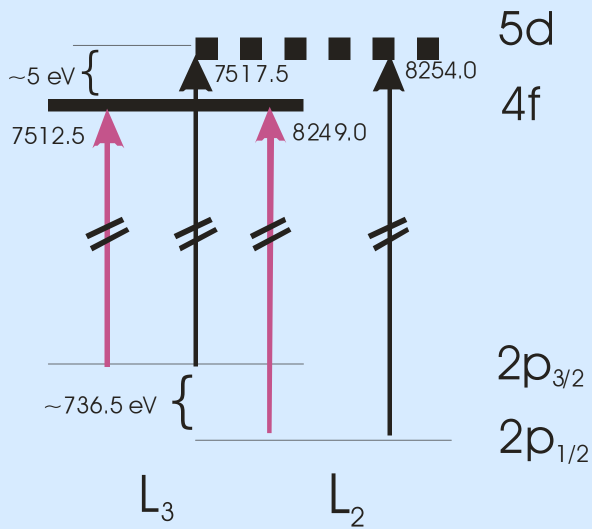
# Quadrupolar effects (E2)

Rare earth metals: • highly localized 4f states
• ordering by exchange interaction via magnetized conduction band (5d)

XMCD at $L_{3,2}$ edges:

electric dipolar transitions E1: 2p → **5d** ($\Delta l = 1$)

and electric quadrupolar transitions E2: 2p → **4f** ($\Delta l = 2$)




**Literature:** rare earth compounds (3d,4f)

- Bartolomé, Tonnerre et al., Phys. Rev. Lett. **79**, 3775 (1997)
- Giorgetti, Dartyge et al., Appl. Phys. A, **73**, 703 (2001)

**Tb foil:** G. Schütz et al: Z. Phys. B: Condens. Matter **73** (1988) 67

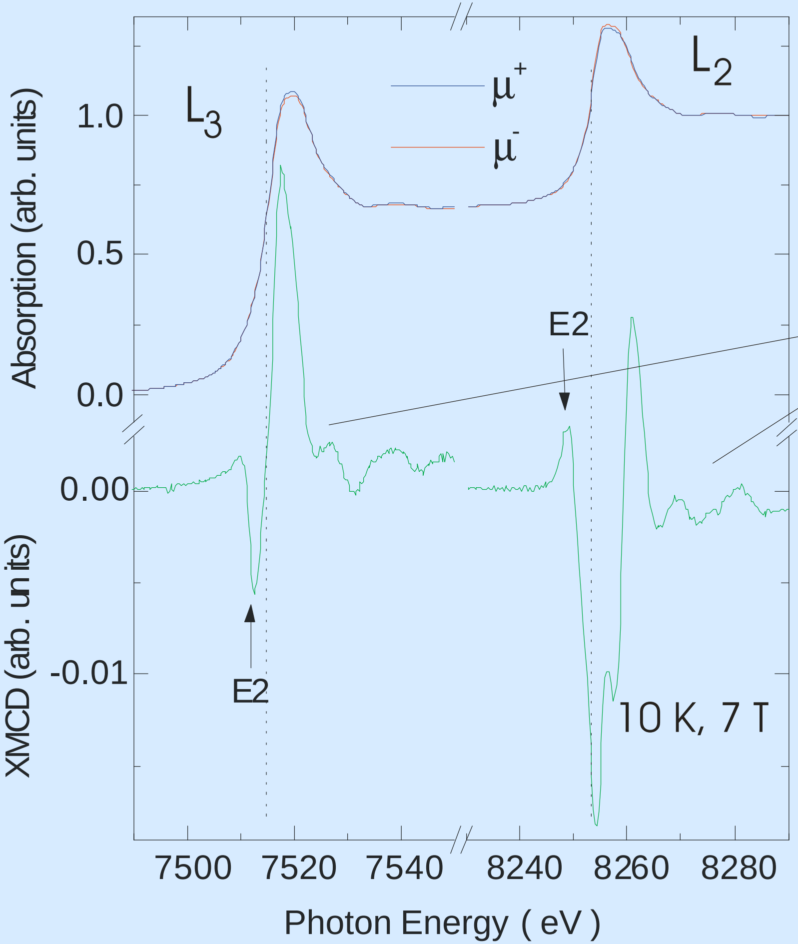
$$\mu(E) \propto |M_{fi}(E)|^2 \rho(E)$$

**but:** E2 contributions neglected

**Theory:** • H. Ebert et al., Solid State Comm. **76** (1990) 475
• Xindong Wang, P. Carra et al., Phys. Rev. B**47** (1993) 9087

⇒"simple model for interpretation in terms of spin-polarization of the d-band - in contrast to transition metals - is not justified"

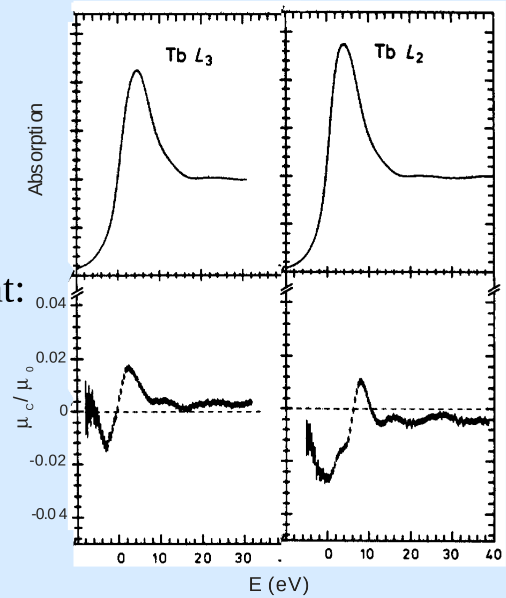
# Tb XMCD at $L_{3,2}$-edges




- XMCD of single element magnet
  Tb single crystal
- High resolution at ESRF (ID 12A):

fine structure
free of noise

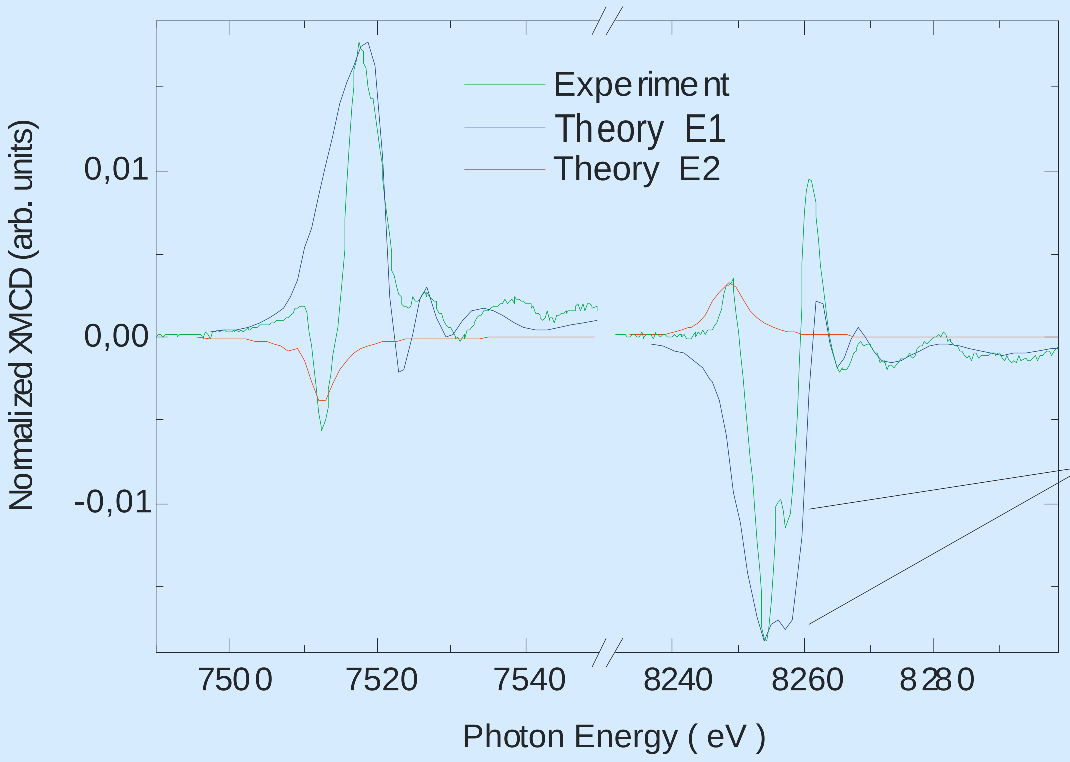
Pioneer experiment:
G. Schütz et al.
Z. Phys. B:
Condens Matter
**73** (1988) 67



# Separation E1 « E2 contributions

- FEFF8*: - self-consistent
  - full multiple scattering in real-space

- Separation of E1 and E2 contributions by switching on/off the E1,2 contribution in the calculation

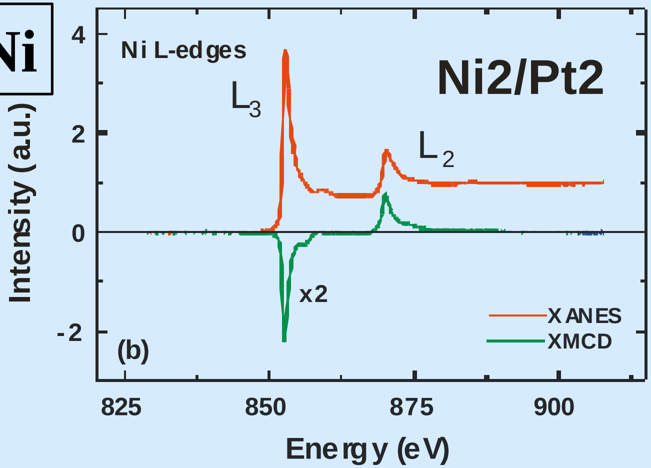


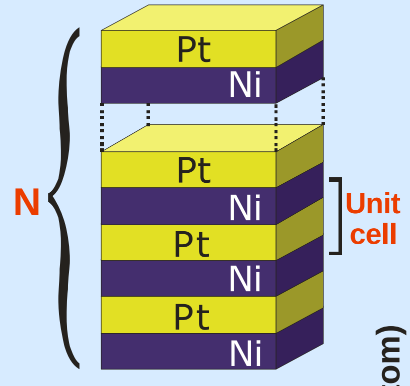

details of
5d spin polarized DOS

H. Wende et al., and J.J. Rehr et al., J. Appl. Phys. **91**, 7361 (2002)
and Highlights ESRF p. 84 (2003)

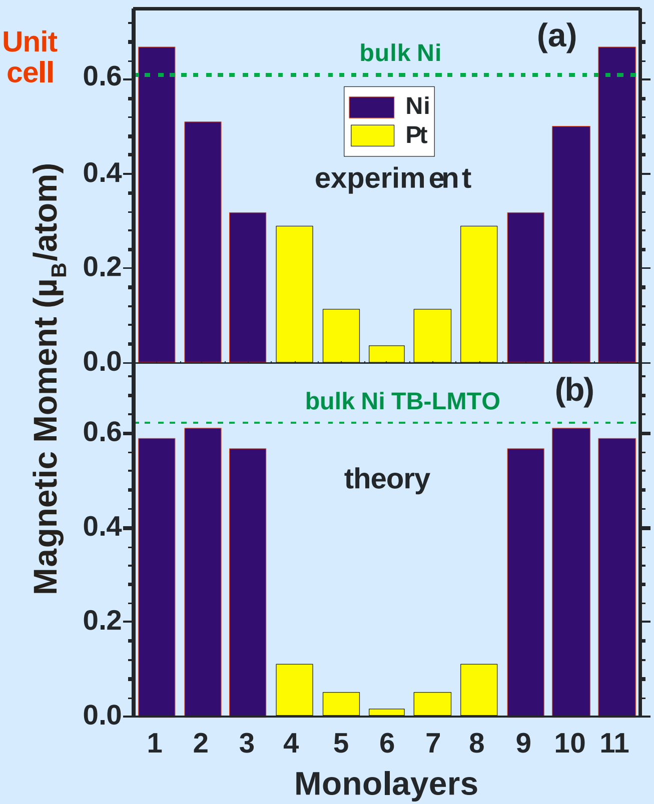
# 4. Induced magnetism at interfaces

**Ni**



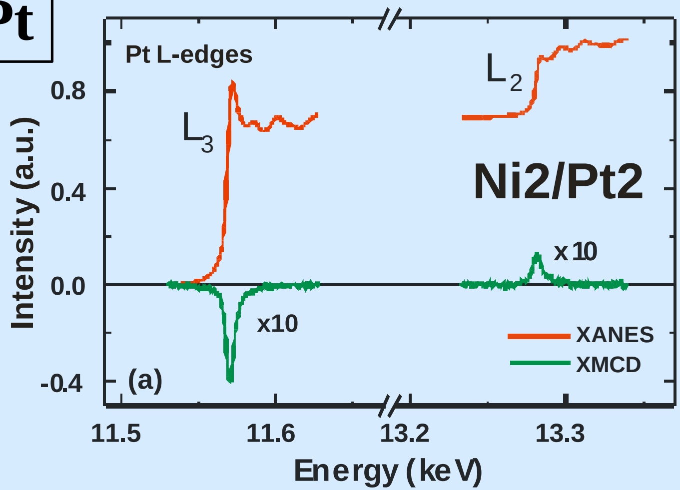




Magnetic moment profile for a $Ni_6/Pt_5$ ML




Ni and Pt carry a magnetic moment

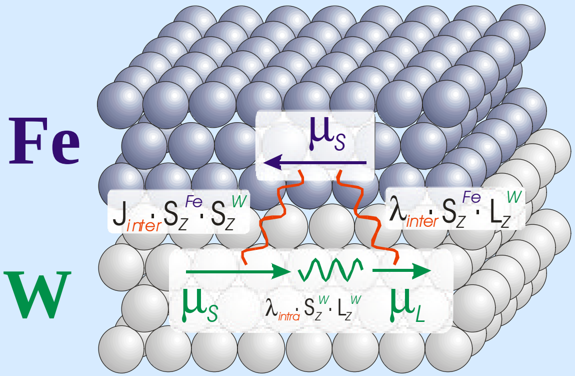
**Pt**

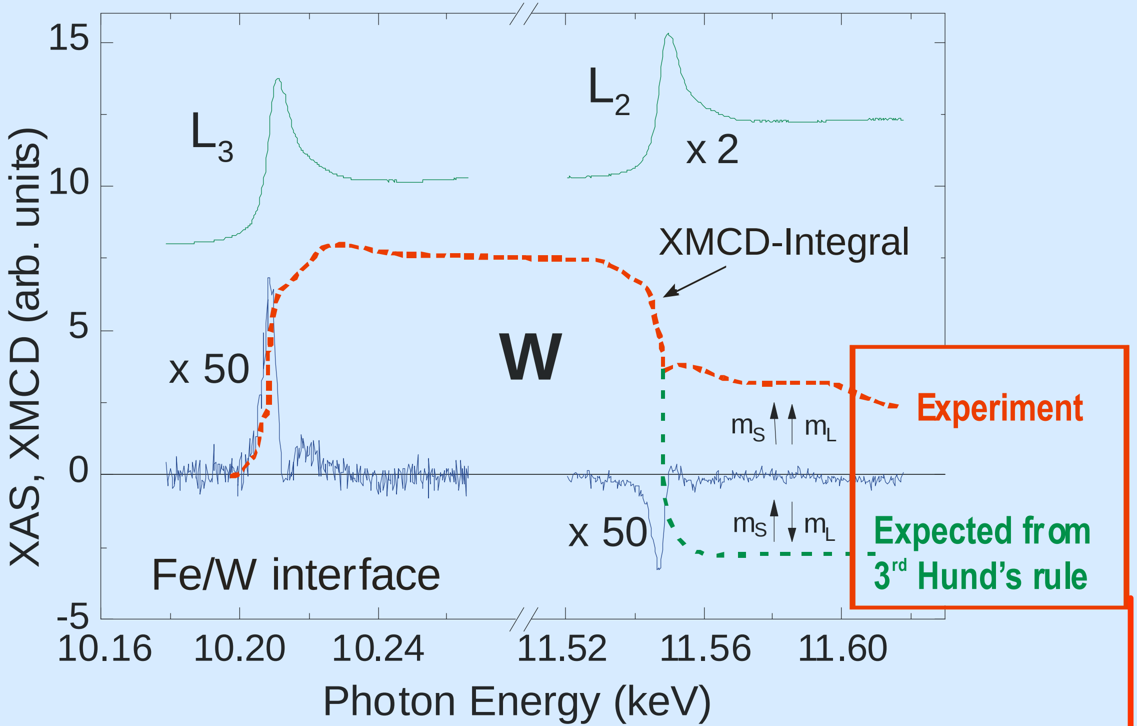



No ‘dead’ Ni layers

F. Wilhelm et al. PRL **85**, 413 (2000) and ESRF Highlights 2000

# Induced magnetism in 5d-transition metals breakdown of 3rd Hund's rule ?



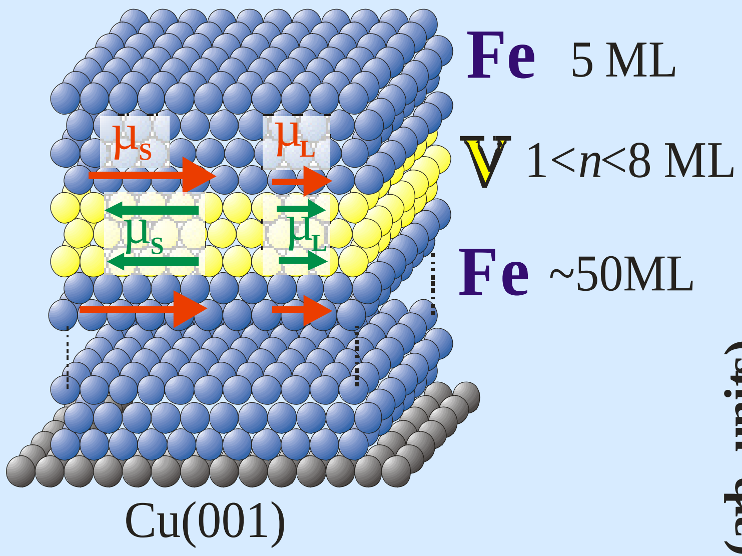




$$J_{inter}\cdot S_Z^{Fe}\cdot S_Z^{W} > \lambda_{inter}\cdot S_Z^{Fe}\cdot L_Z^{W} > \lambda_{intra}\cdot S_Z^{W}\cdot L_Z^{W}$$

alternatively: details of SP-DOS; hybridization

F. Wilhelm et al.,
Phys. Rev. Lett. **87**, 207202 (2001)

# Fe/V/Fe(110)Trilayer




Advantages:

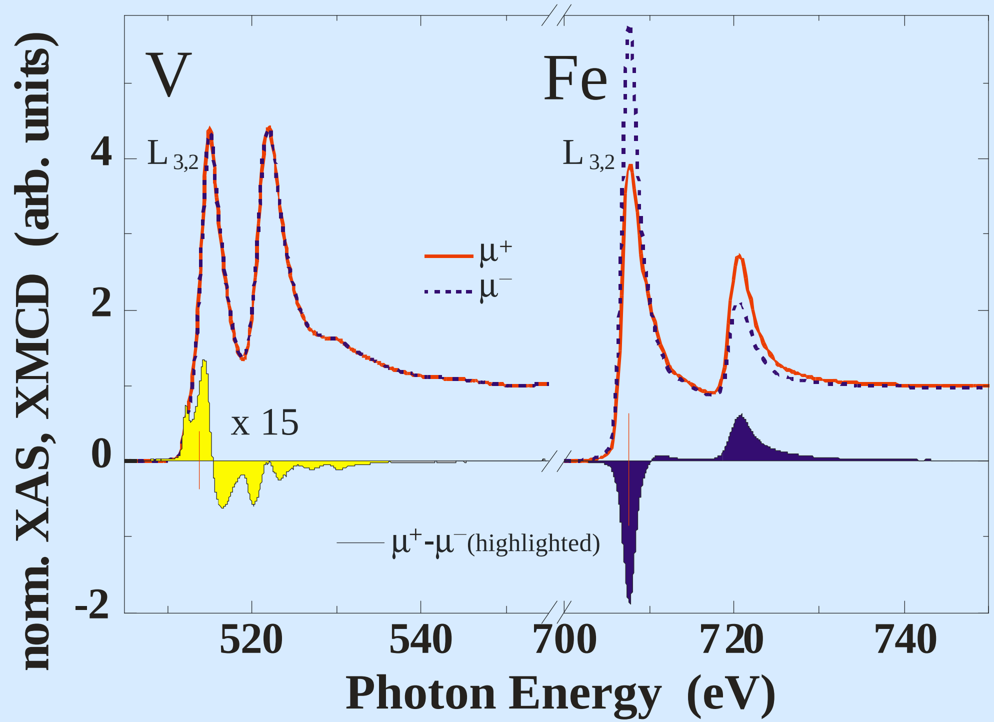
controlable growth

- annealing to reduce surface roughness
- preparation at 300 K

In-situ experiment, i.e. no capping layers

XMCD measurements:

- BESSY II, third generation synchrotron source in Berlin
- Newly developed 'gapscan' mode: High degree of circularly polarized light and high photon flux

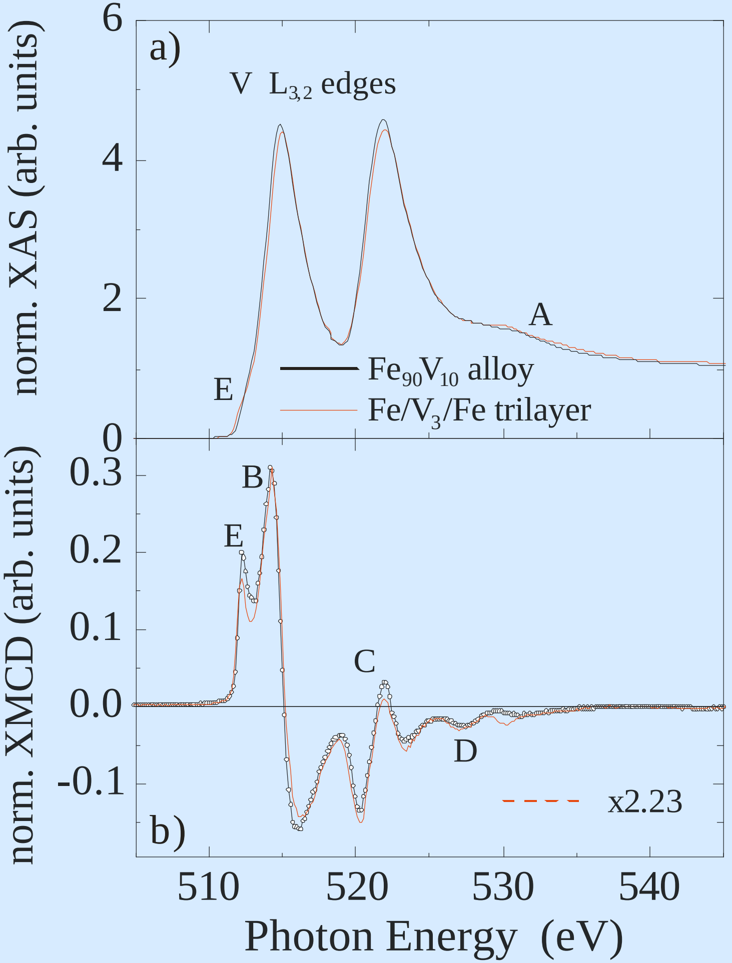


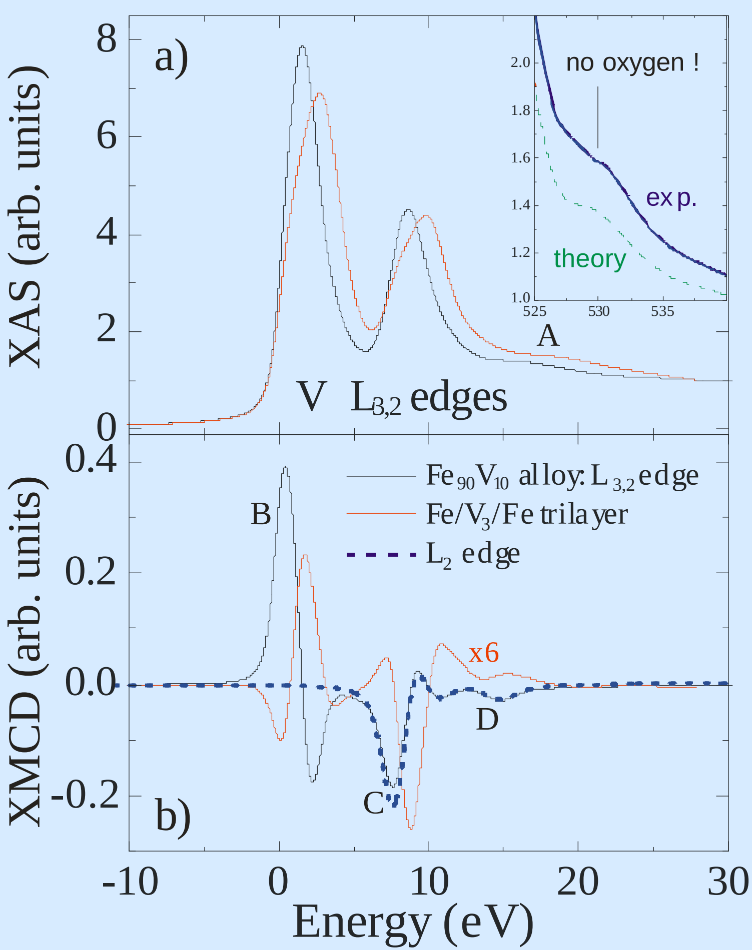
# Beyond sum rules: full calculation of μ(E)

***Experiment***
A. Scherz et al.

PRB **66**, 184401 (2002)

***Theory***
J. Minar, D. Benea, H. Ebert, LMU



# Beyond sum rules: full calculation of μ(E)

PHYSICAL REVIEW B **66**, 184401 (2002)

**Relation between $L_{2,3}$ XMCD and the magnetic ground-state properties for the early 3$d$ element V**

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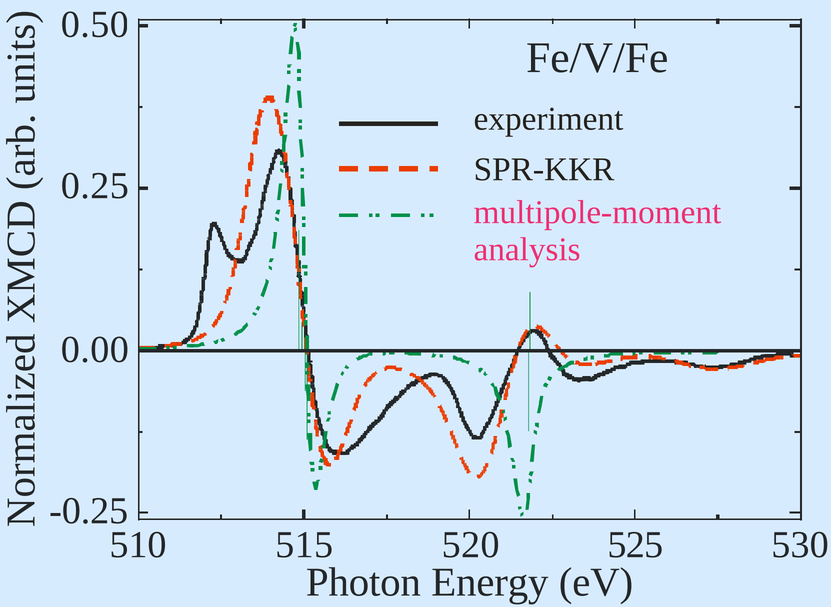


**Sum rules and beyond**

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*Abstract*
Sum rules relate the integrated signals of the spin-orbit split core levels to ground state properties, such as the spin-orbit coupling, magnetic moments and charge distribution. These rules give the zeroth moments of the spectral distribution. It is shown how this can be generalized to higher-moment statistical analysis … .

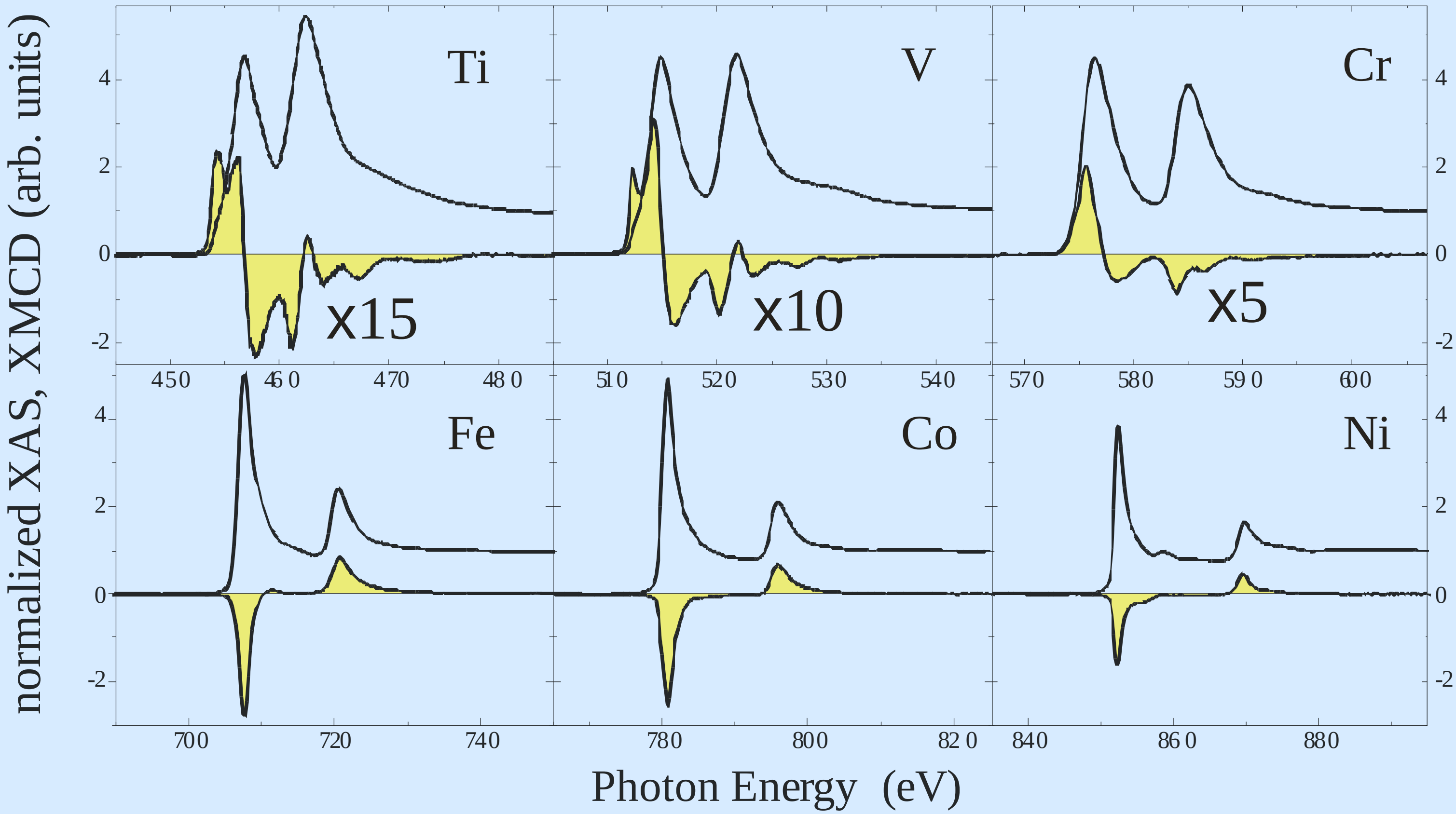
*Journal of Electron Spectroscopy and Related Phenomena 101–103 (1999) 859–868*

- All spectroscopic techniques do not restrict themselves to measure the intensity (area under the resonance) only. i.e.: integral sum rules.
- A resonance signal contains a resonance position, a width, an asymmetry profile, etc.
- The optimum is given if theory can calculate the full profile of the resonance, in our case μ(E) i.e. the spectral density.

***Recent advances in x-ray absorption spectroscopy***
H. Wende , Rep. Prog. Phys. **67**, 2105 (2004)

# $L_{2,3}$ XAS and XMCD of *3d* TM's

| Sc | Ti | V | Cr | Mn | Fe | Co | Ni | Cu | Zn |
|---|---|---|---|---|---|---|---|---|---|




A. Scherz et al., XAFS XII June 2003 Sweden, Physica Scripta;
A. Scherz et al., BESSY Highlights p. 8 (2002)

# Conclusion, Future

During last few years: enormous progress in

Theory:
- calculate $\mu(E)$, spin dependent spectral distribution
- full relativistic calculations
- real (not ideal) crystallographic structures

Experiment:
- higher $\Delta E/E$, detailed dichroic fine structure
- undulator, gap-scan technique, constant high $P_C$
- element-selective microscopy, probe of “non-magnetic” constituents

Future:
- core hole effects:

  $\rightarrow$ change of branching ratio for early 3d elements

  $\rightarrow$ effect on XMCD unknown!
- correct determination of $\Delta \mu_L$ and MAE with XMCD (x30)
- correction for spin- and energy-dependence of matrix elements