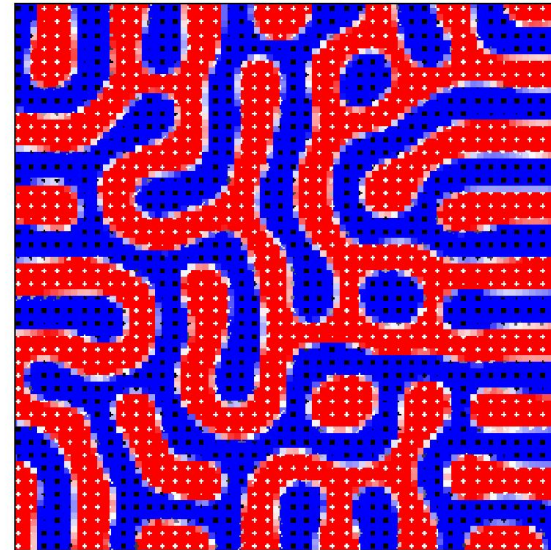
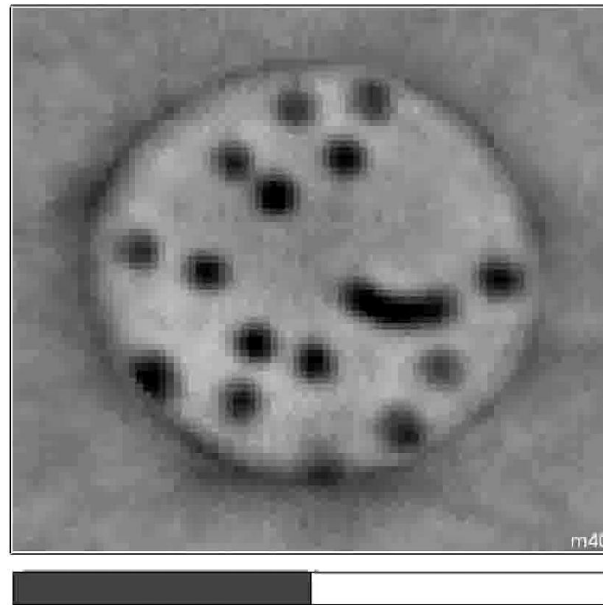


Magnetism at Nanoscale: Nano-small meets Ultra-fast



Jyoti Mohanty



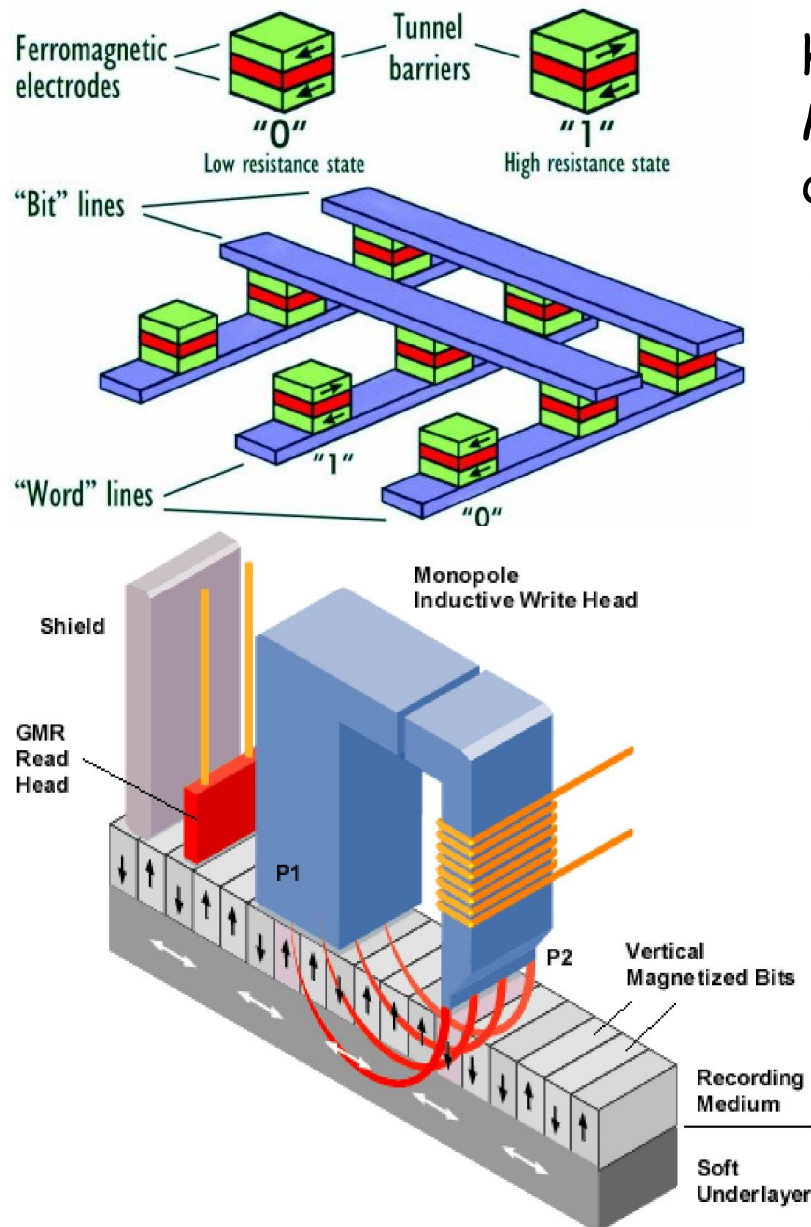
भारतीय प्रौद्योगिकी संस्थान हैदराबाद
Indian Institute of Technology Hyderabad

Motivation

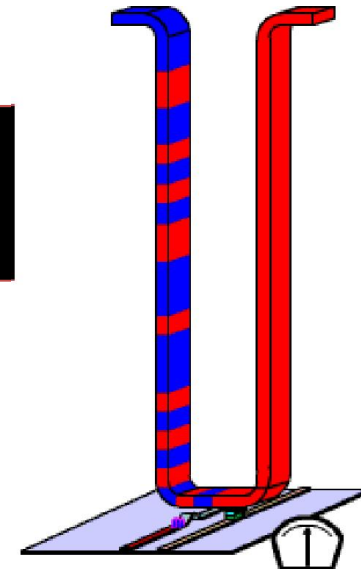
hard disks, read/write heads and
Magnetic Sensors, spintronics
devices

Thin film, Nanostructure, self-assembly

Magnetic domain & domain walls



Nano-small
Ultra-fast



Stuart S. P. Parkin, *et al.* *Science* 320, 190 (2008)

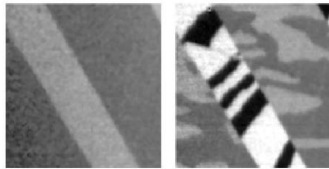
Space

10^{-3} m – 1 mm

100 μ m

10 μ m

AFM & FM domains

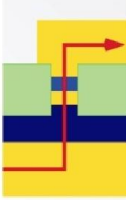


10^{-6} m – 1 μ m



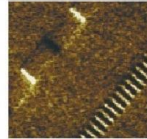
Recorded "bits"

Spin injection



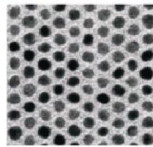
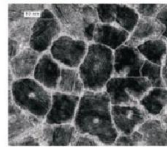
100 nm

Media grains



10 nm

Nano-particles



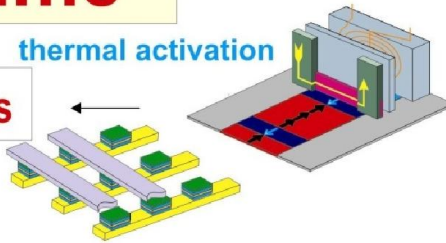
10^{-9} m – 1 nm

The Microworld

The Nanoworld

Time

10^{-9} s – 1 ns

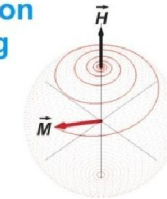


thermal activation

100 ps

spin precession and damping

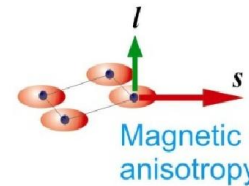
H = 1 Tesla



10 ps

10^{-12} s – 1 ps

100 fs

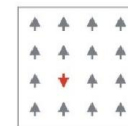


Spin-orbit coupling

Magnetic anisotropy

10 fs

10^{-15} s – 1 fs



Exchange interaction

Ultrafast

Note: Δt (fs) = 4 / ΔE (eV)

Space

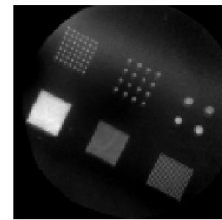
1 μ m

100 nm

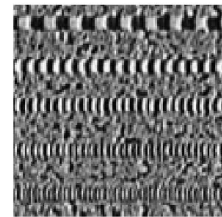
10 nm

1 nm

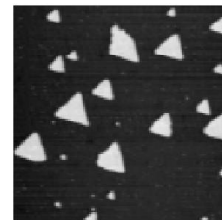
1 \AA



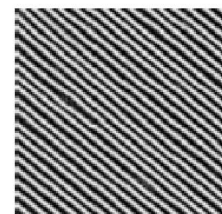
Patterned media



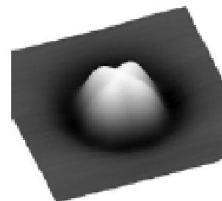
Granular/
multilayer media



Nanoparticles



Nanowires



Molecules
Atoms

Time

1 yr

1 s

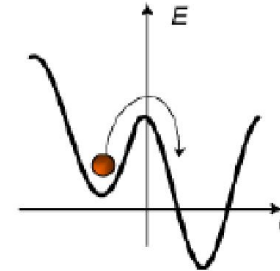
1 μ s

1 ns

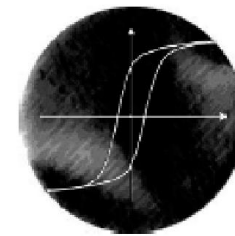
1 ps



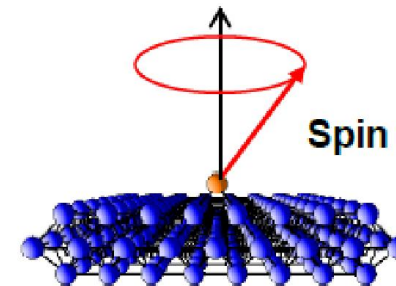
Permanent magnets



Magnetic relaxation

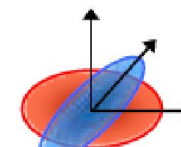


Domain wall propagation

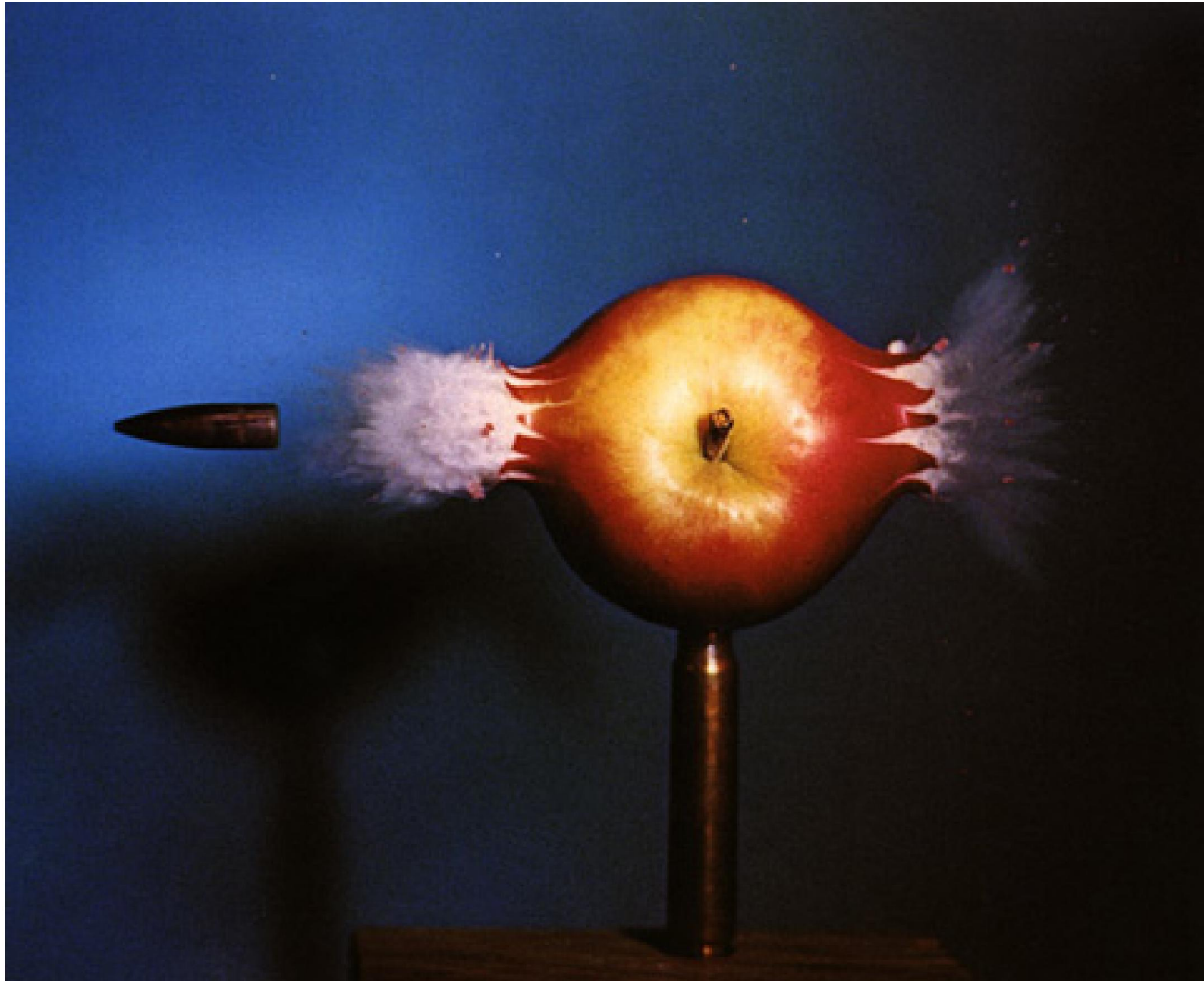


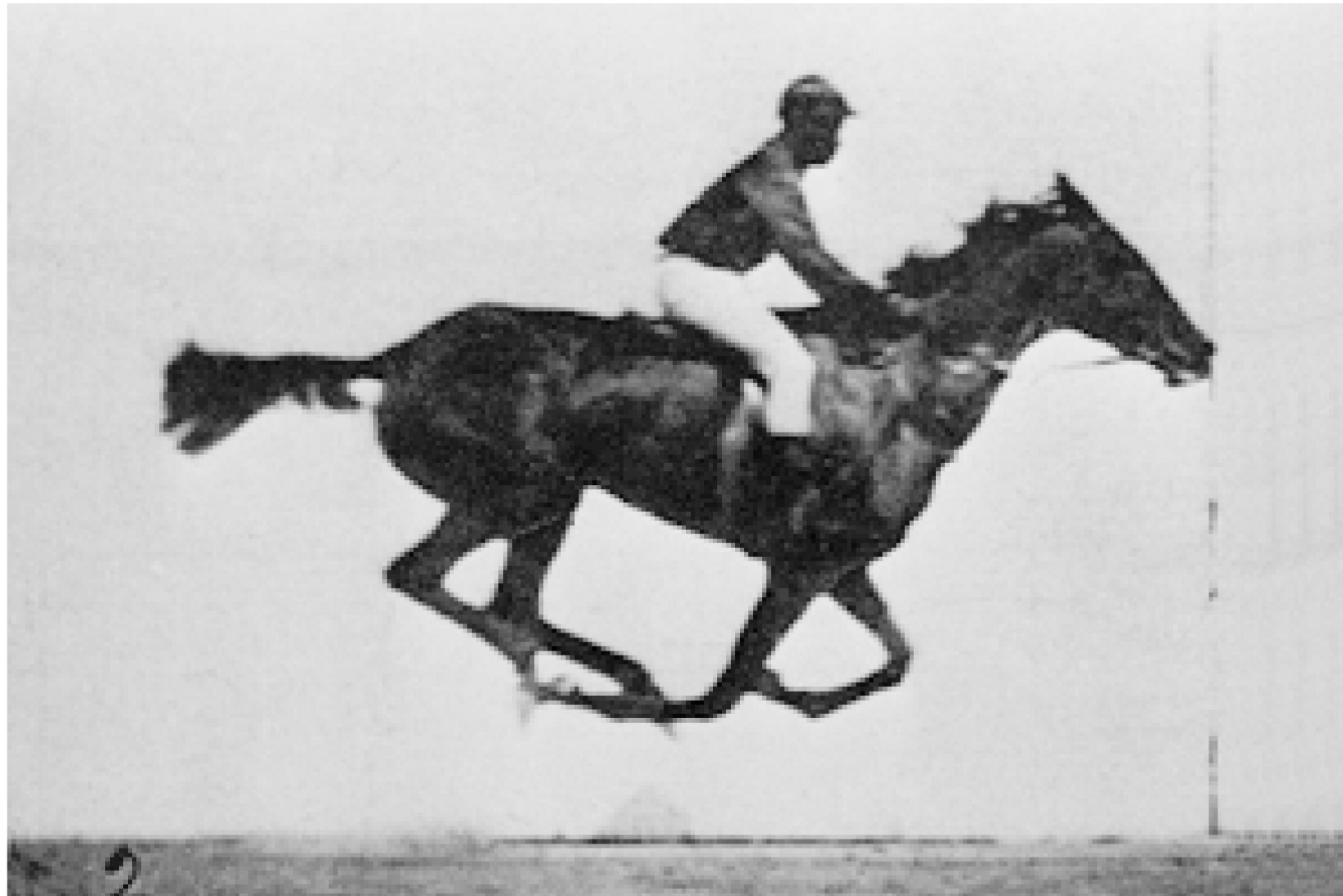
Spin reversal

Magnon excitations

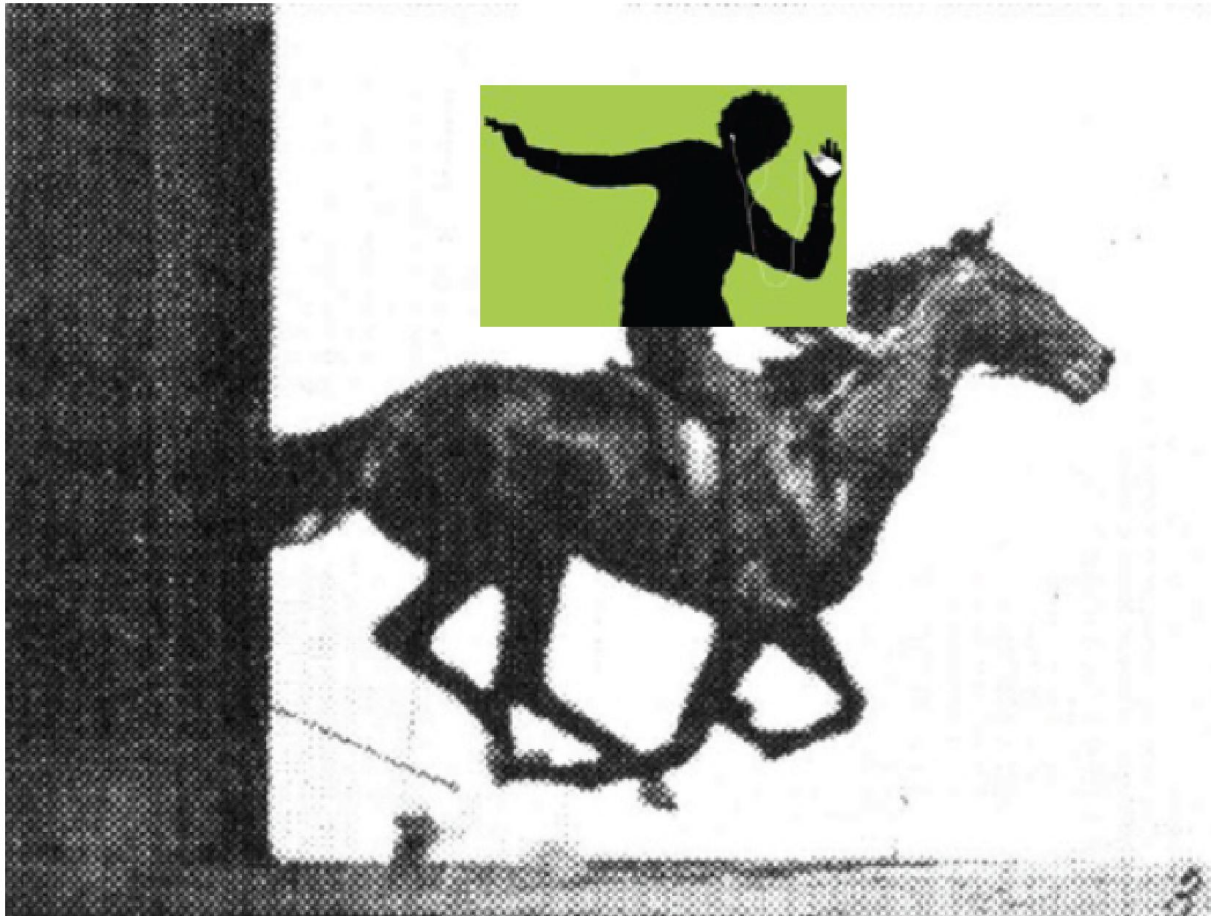


Spin-orbit coupling



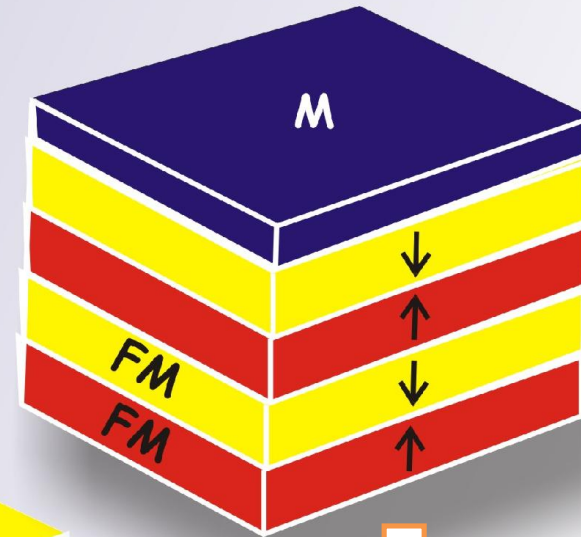
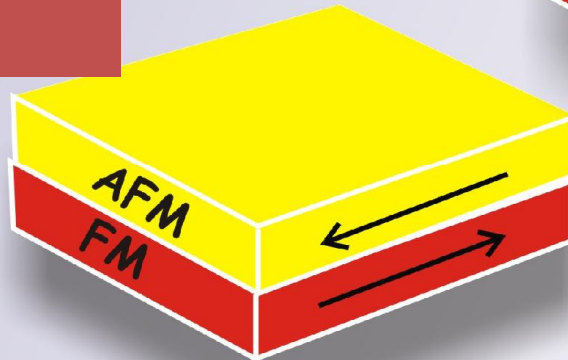
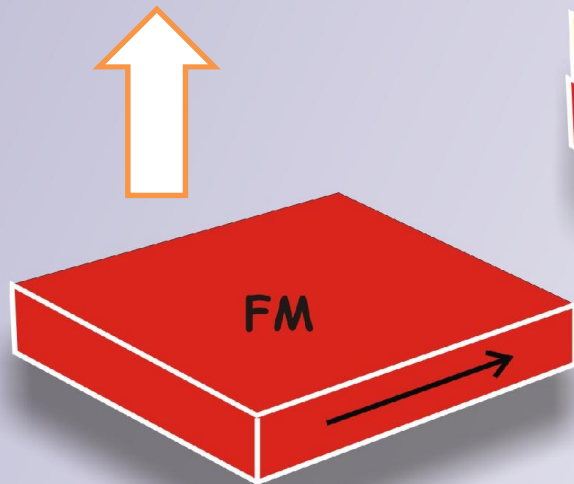


$(10^{-3} \text{ s}; 10^{-3} \text{ m}) \rightarrow (10^{-15} \text{ s}; 10^{-9} \text{ m})$



Nanomagnetism:

- Strain-Magnetism
 - FM-PM
 - Dipolar Coupling
 - M reversal (microscopic)
- MFM, X-PEEM



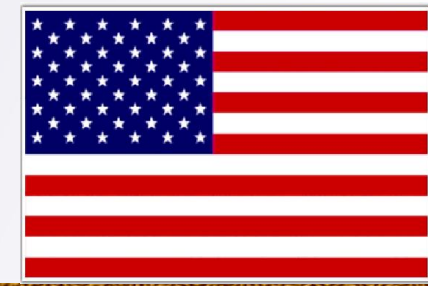
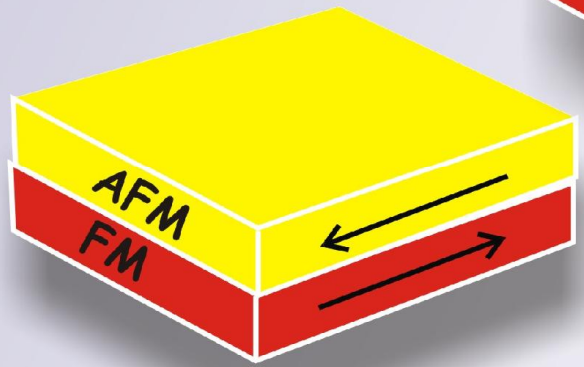
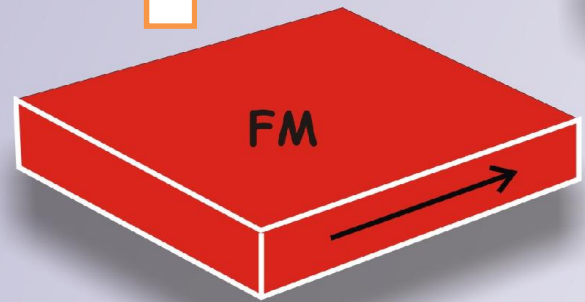
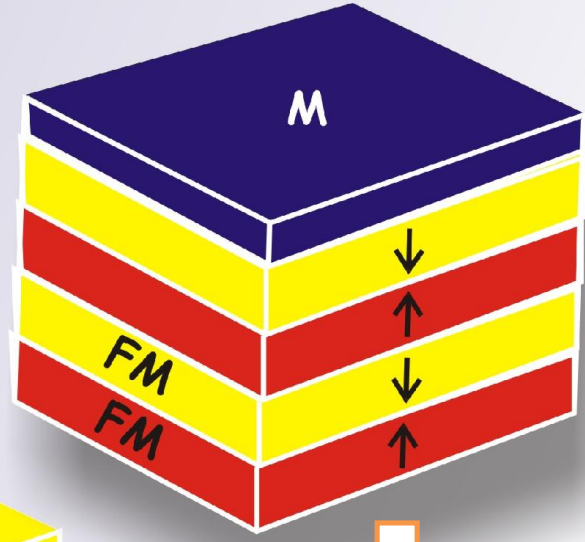
- Ferri-Magnet
 - Imaging without lens
 - Ultrafast M dynamics
- Coherent Magn Scattering,
X-ray Holography

- Exchange Bias
 - Interface Frozen spins
- XMCD, Element specific M-H loop, XMLD

Nanomagnetism:

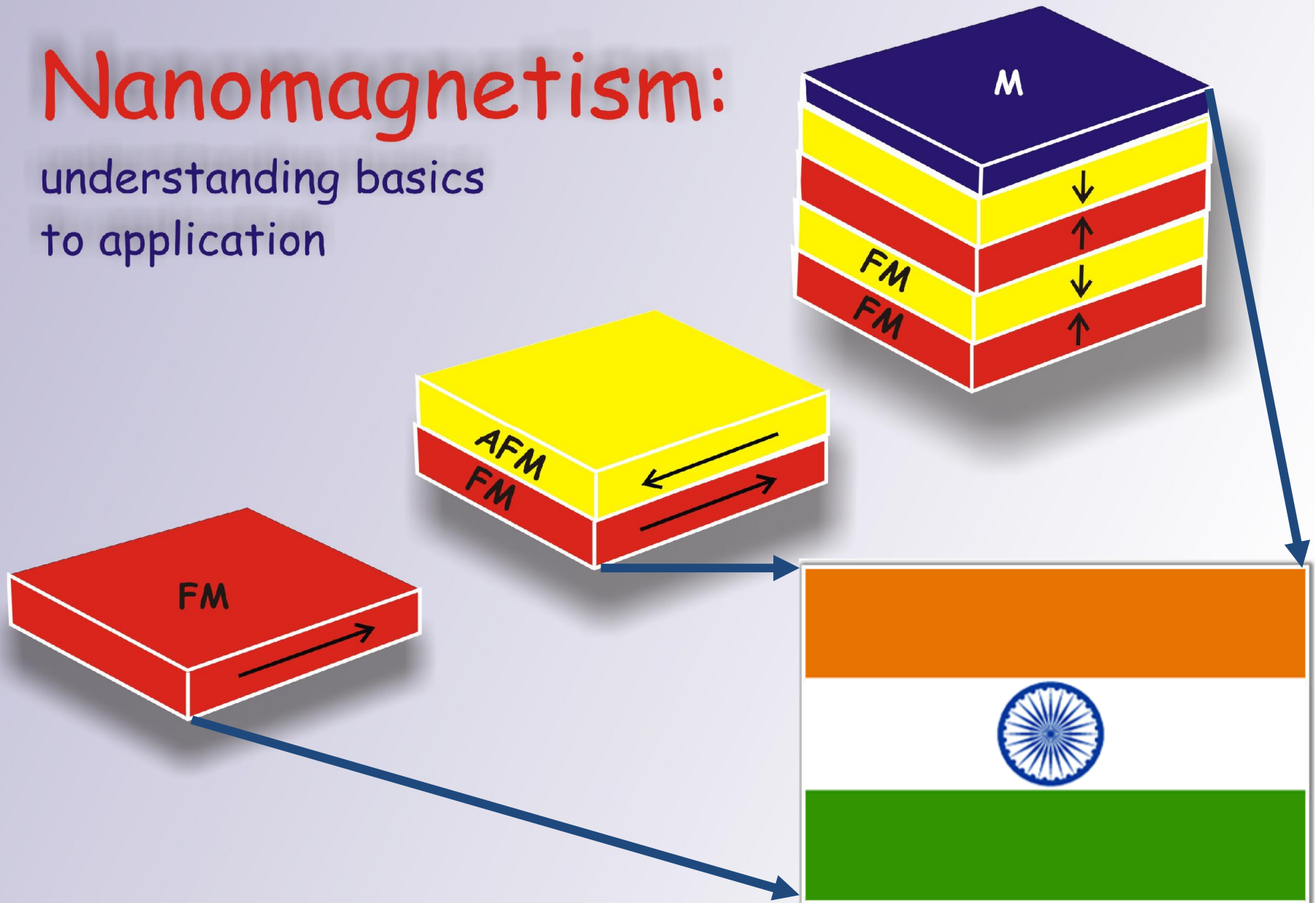


ing basics
n



Nanomagnetism:

understanding basics
to application



What we need

General requirements:

Technique requirements:

see the invisible

nanoscale spatial resolution

separate spin and orbital contributions

sensitive to s-o coupling

study thin films and interfaces

large cross section for "signal"

look below the surface

depth sensitivity

distinguish components

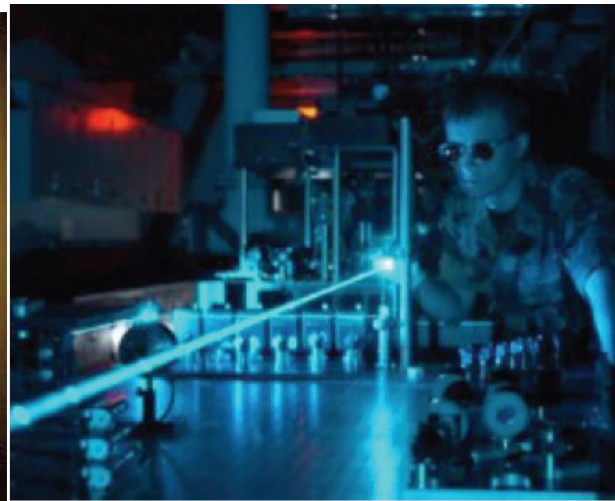
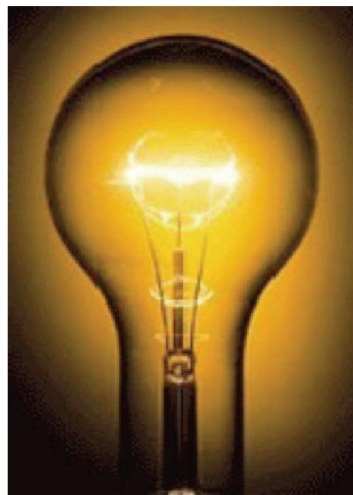
elemental (chemical) specificity

resolve dynamic motions

time resolution < 1 nanosecond

Why use X-rays?

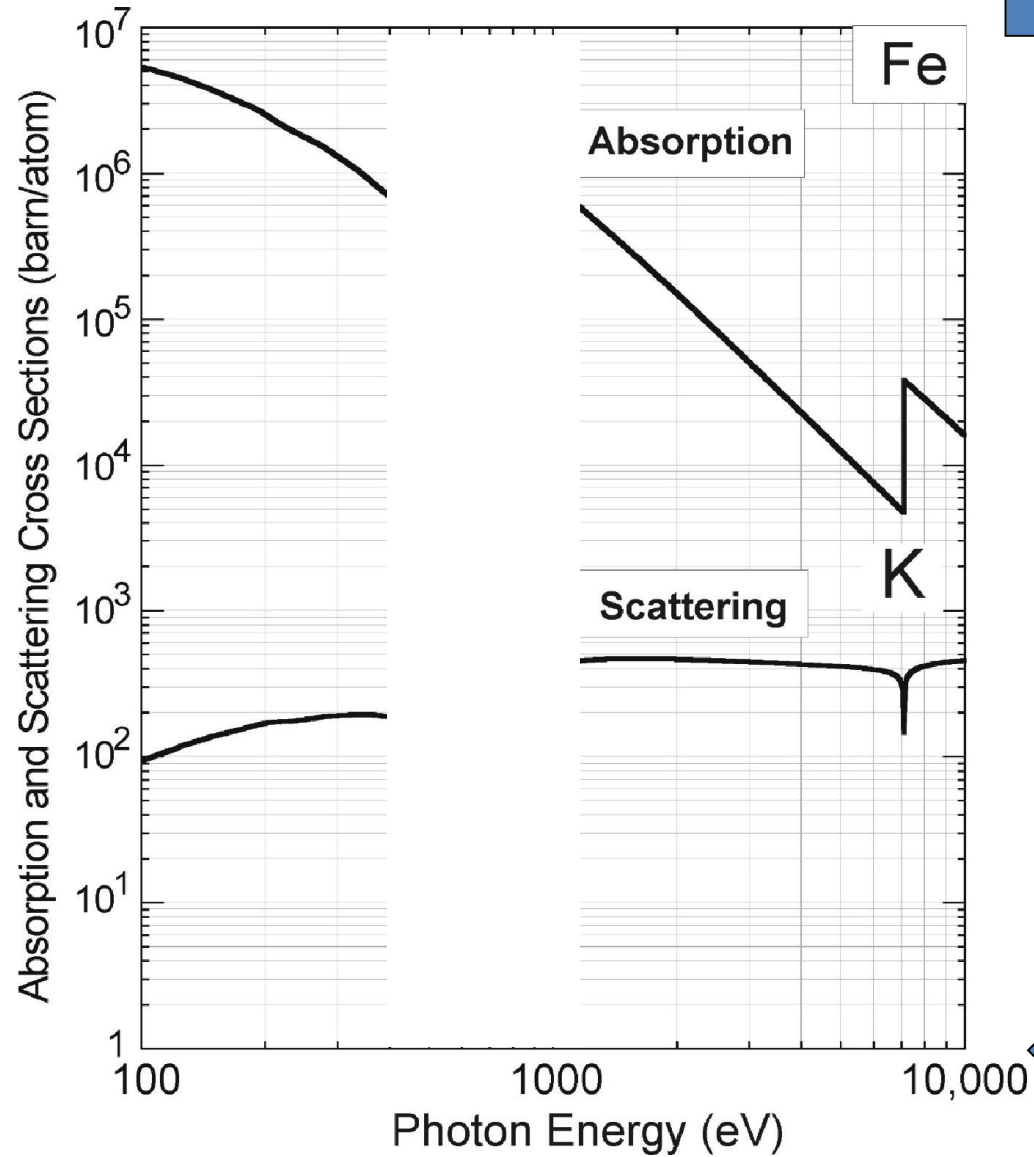
- x-ray cross section and flux
- x-ray tunability: resonances and sum rules
- x-ray polarization
- x-ray spatial resolution
- x-ray temporal resolution
- * Coherence Properties



Tunable x-rays offer large interaction cross sections

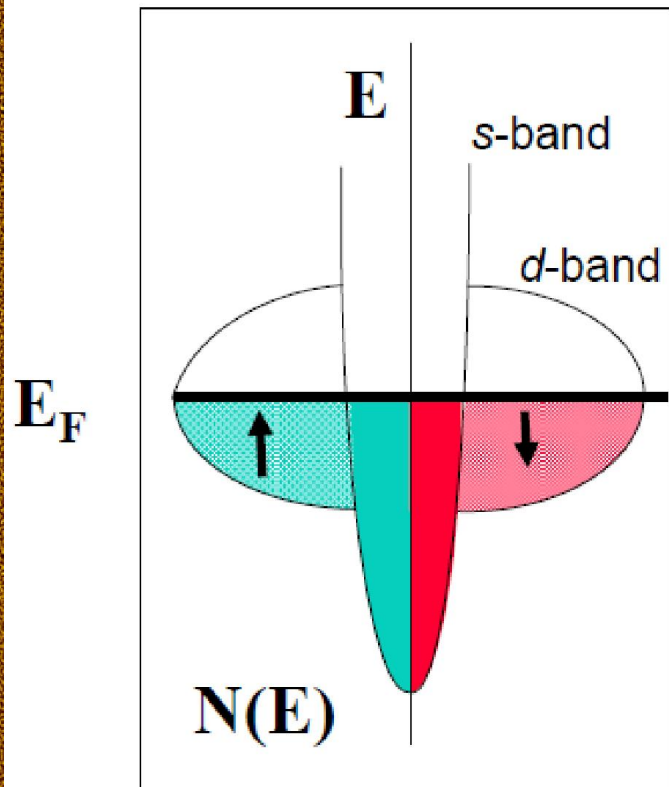
optical light

electrons

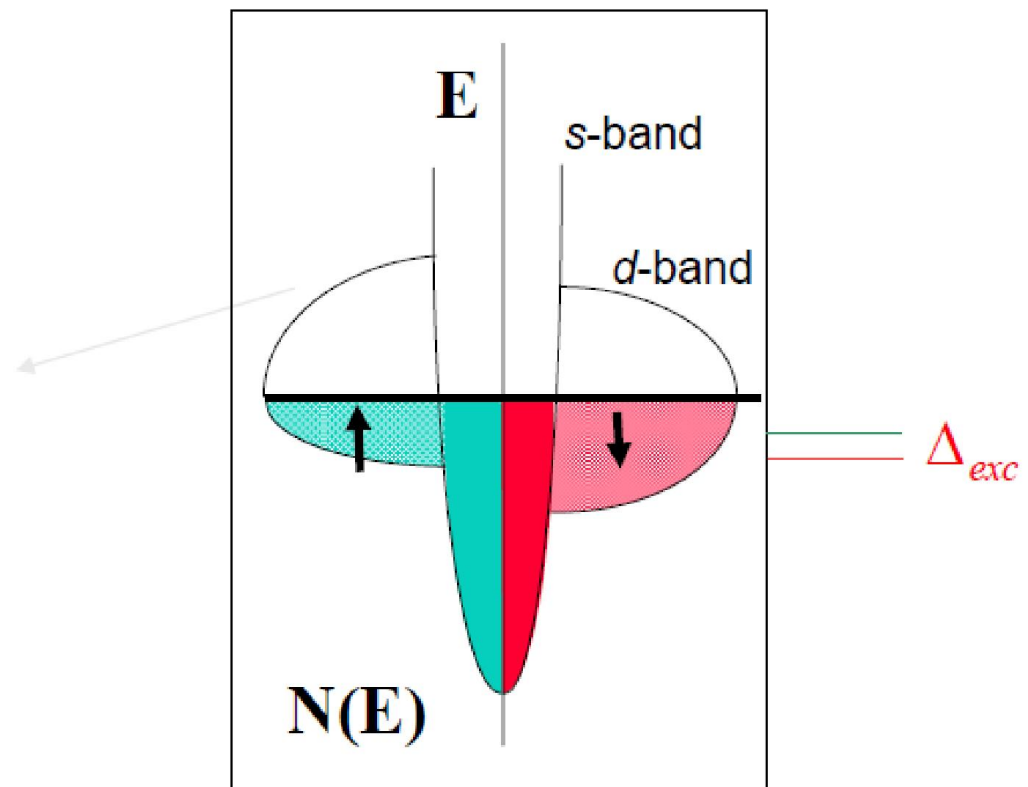


neutrons

nonmagnetic



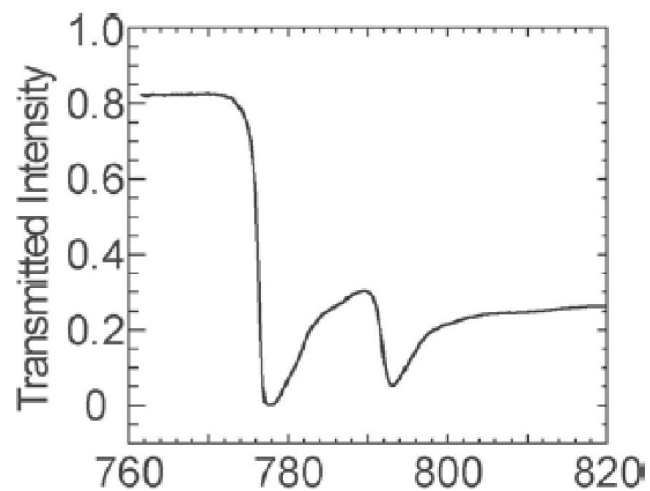
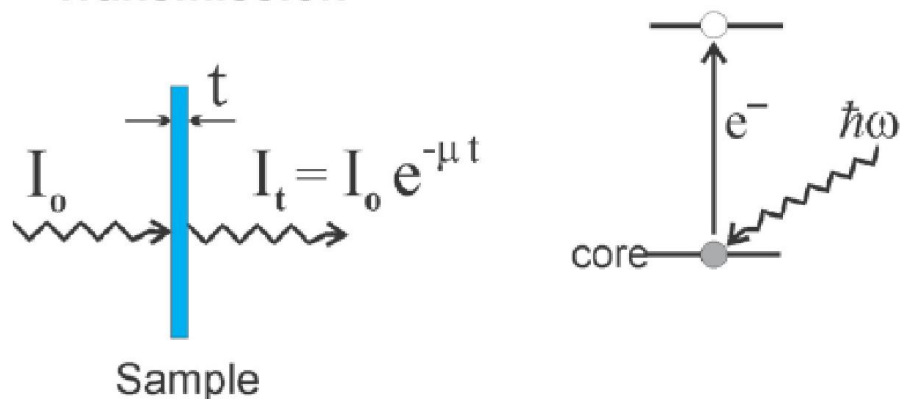
magnetic



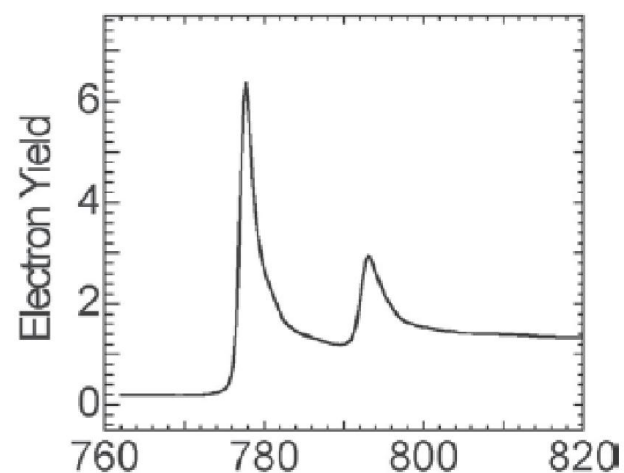
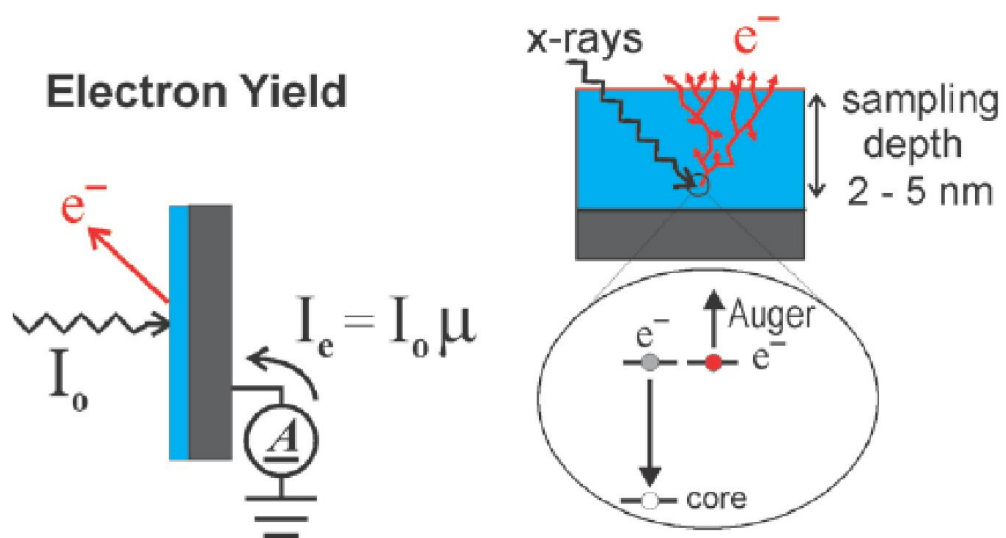
E.P. Wohlfarth, Iron, Cobalt and Nickel,
in *Ferromagnetic Materials*, vol.1, North Holland, Amsterdam, (1988)

X-ray absorption

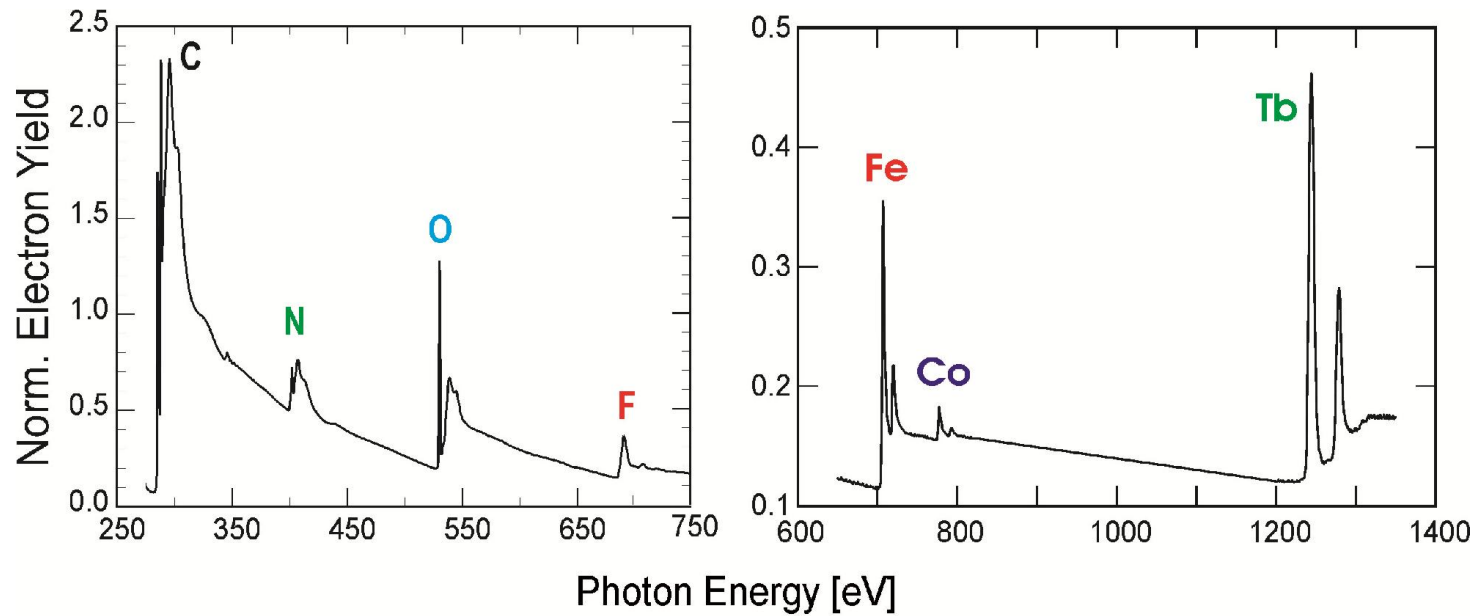
Transmission



Electron Yield



Tunable x-rays offer elemental specificity



Valence

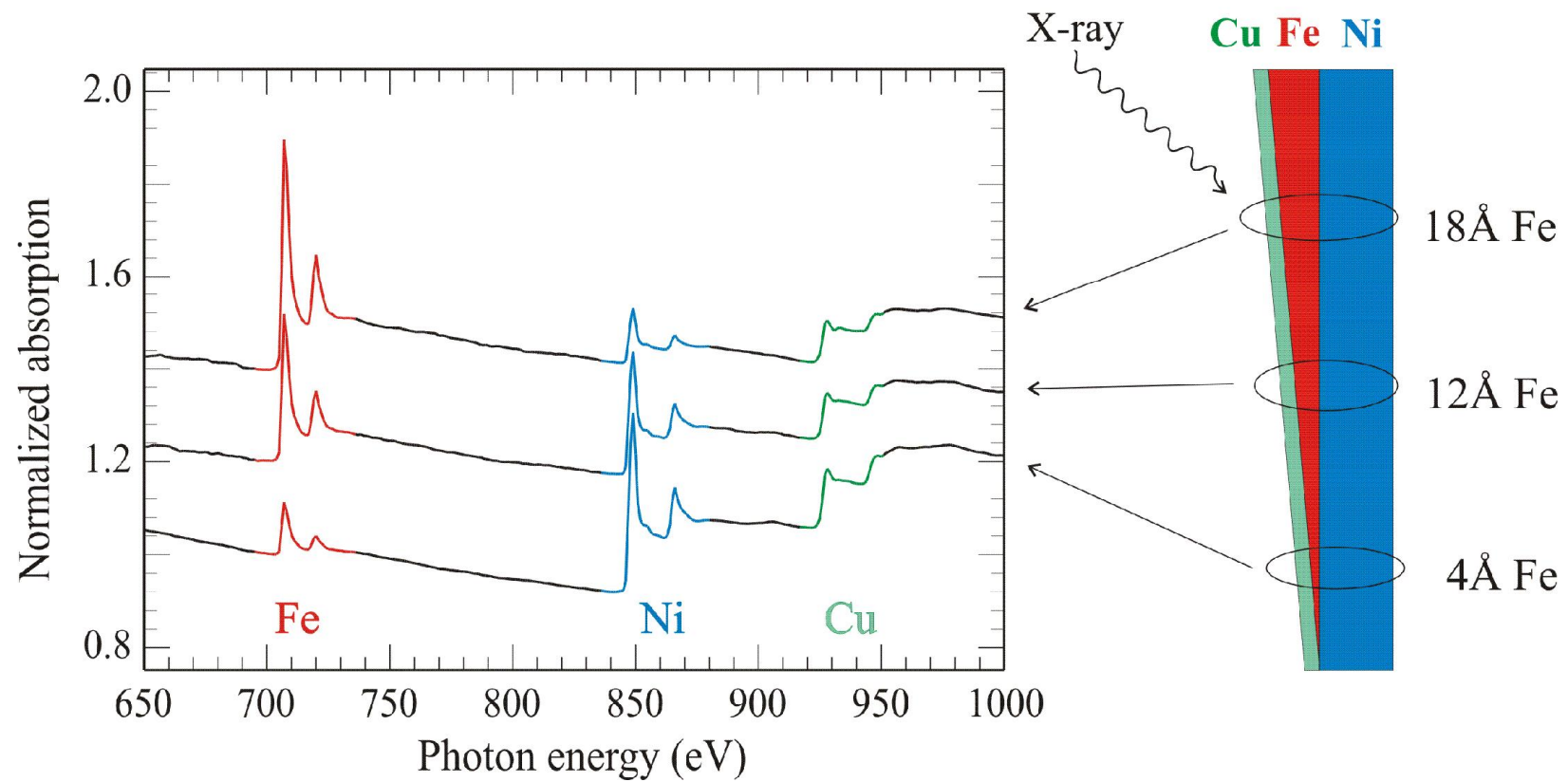
Dipole

Core

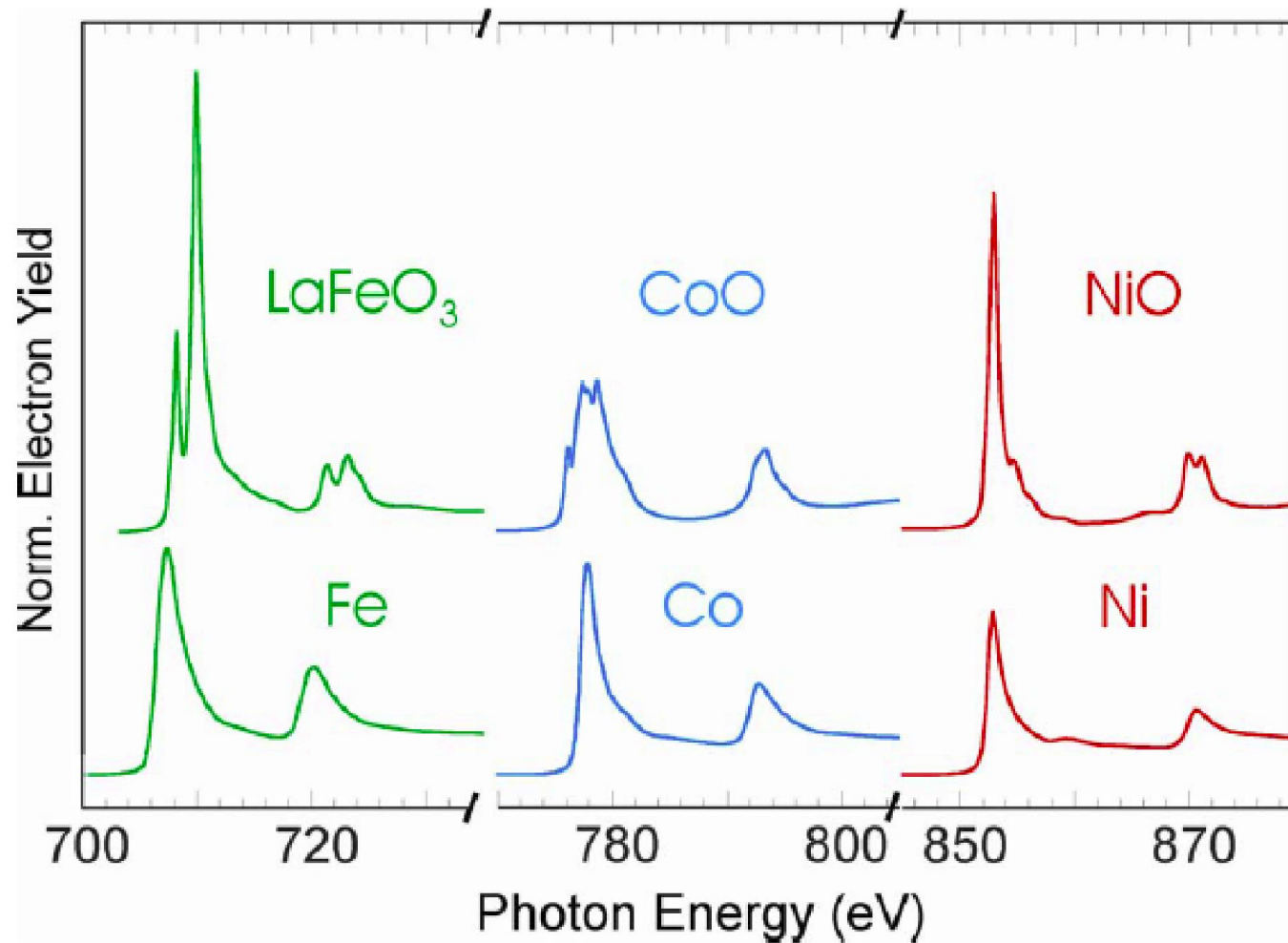
Polymers: p-orbitals
 Trans. Met.: d-orbitals
 Rare Earths: f-orbitals

1s K-edge
 2p L edge
 3d M edge

X-rays can pick materials apart: layer-by-layer

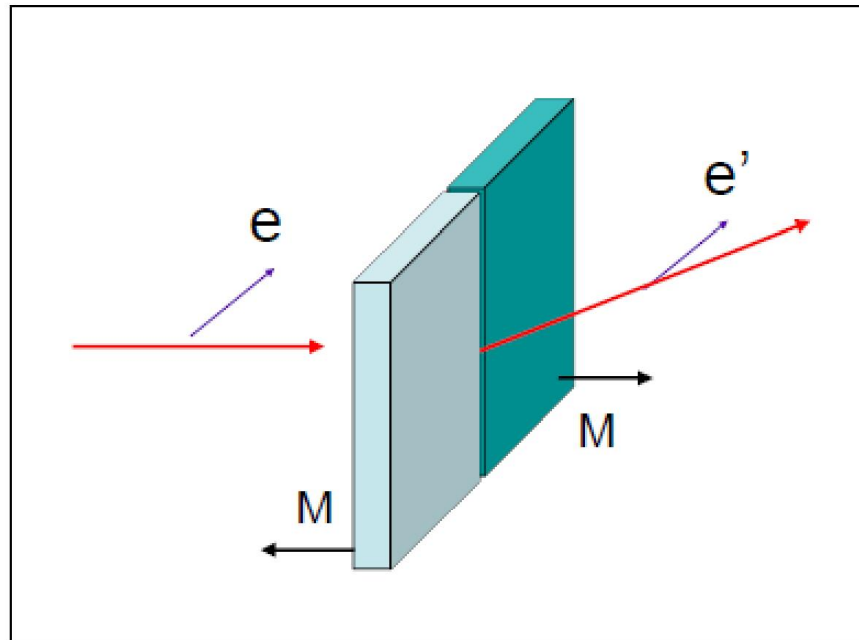


Rich “multiplet structure” reveals local bonding

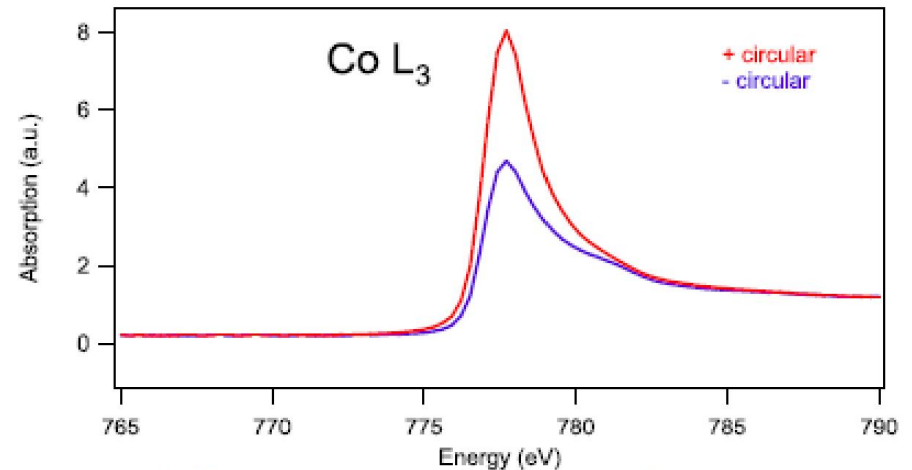


Resonant Magnetic Scattering

Photon-in photon-out: possible in applied fields



X-Ray Magnetic Circular Dichroism (XMCD)



$$I \propto \left| \sum_n \exp(i\mathbf{q} \cdot \mathbf{r}_n) f_n \right|^2$$

charge

magnetic

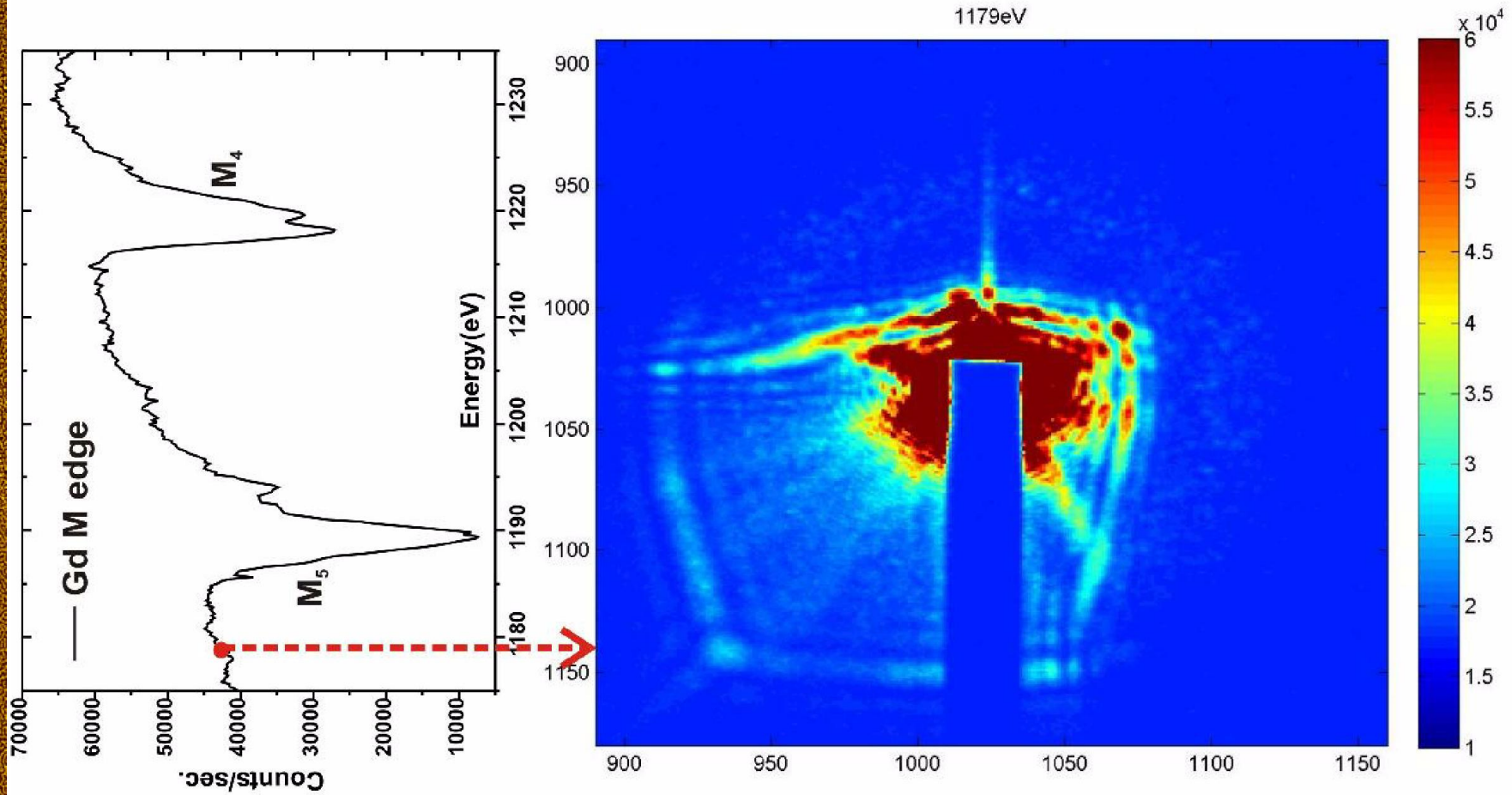
magnetic

$$f_n = \mathbf{e}' \cdot \mathbf{e} F_n^c - i(\mathbf{e}' \times \mathbf{e}) \cdot \mathbf{M}_n F_n^{m1} + (\mathbf{e}' \cdot \mathbf{M}_n)(\mathbf{e} \cdot \mathbf{M}_n) F_n^{m2}$$

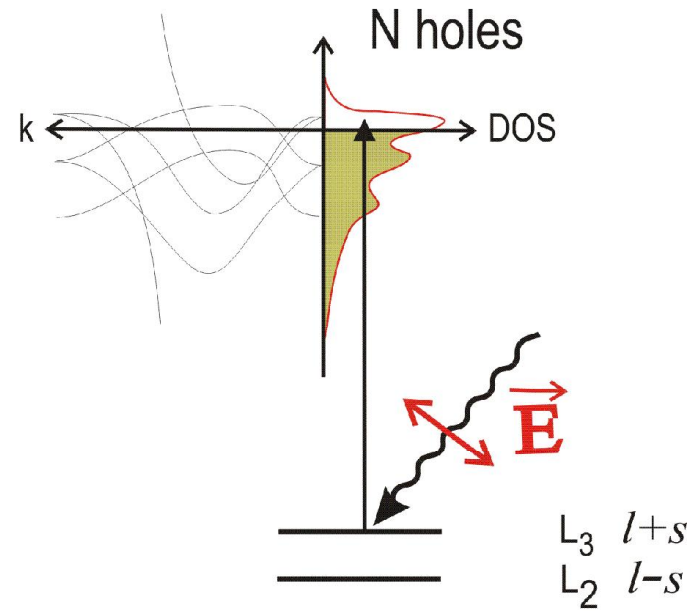
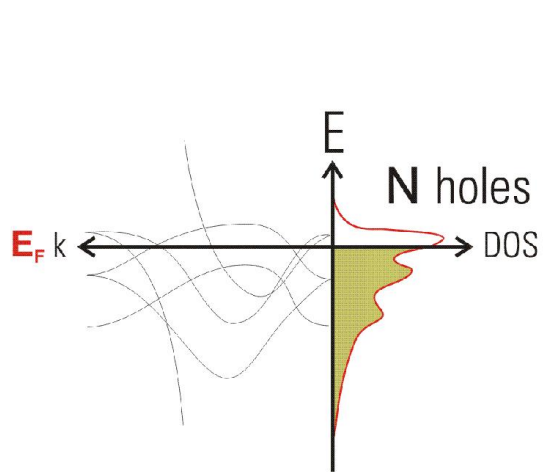
zero
in our geometry

J. P. Hannon, G. T. Trammell, M. Blume, D. Gibbs, Phys. Rev. Lett 61, 1245 (1988)

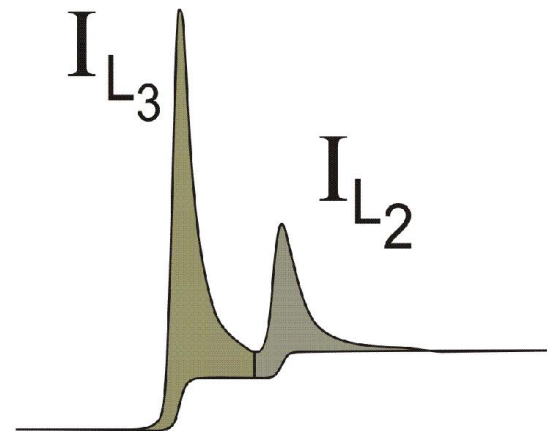
Resonant Magnetic Scattering

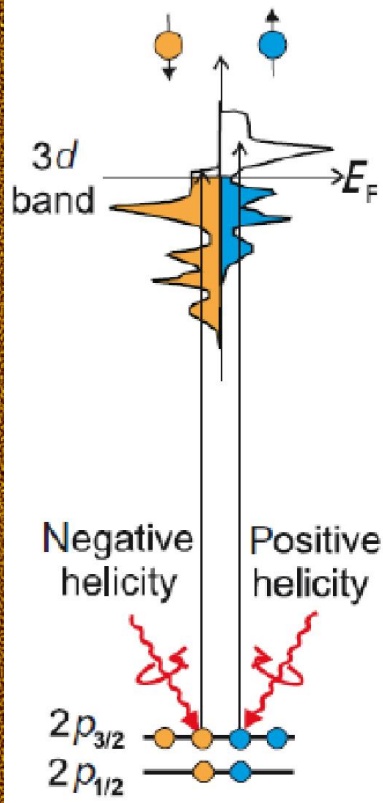


Properties of 3d Transition Metals

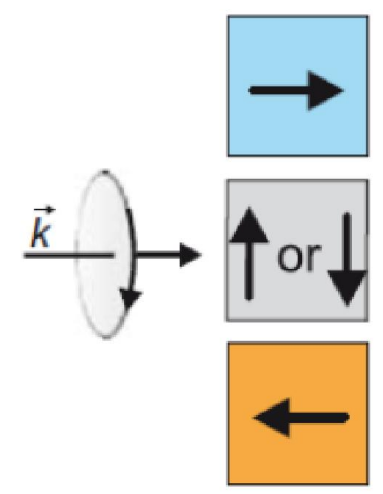
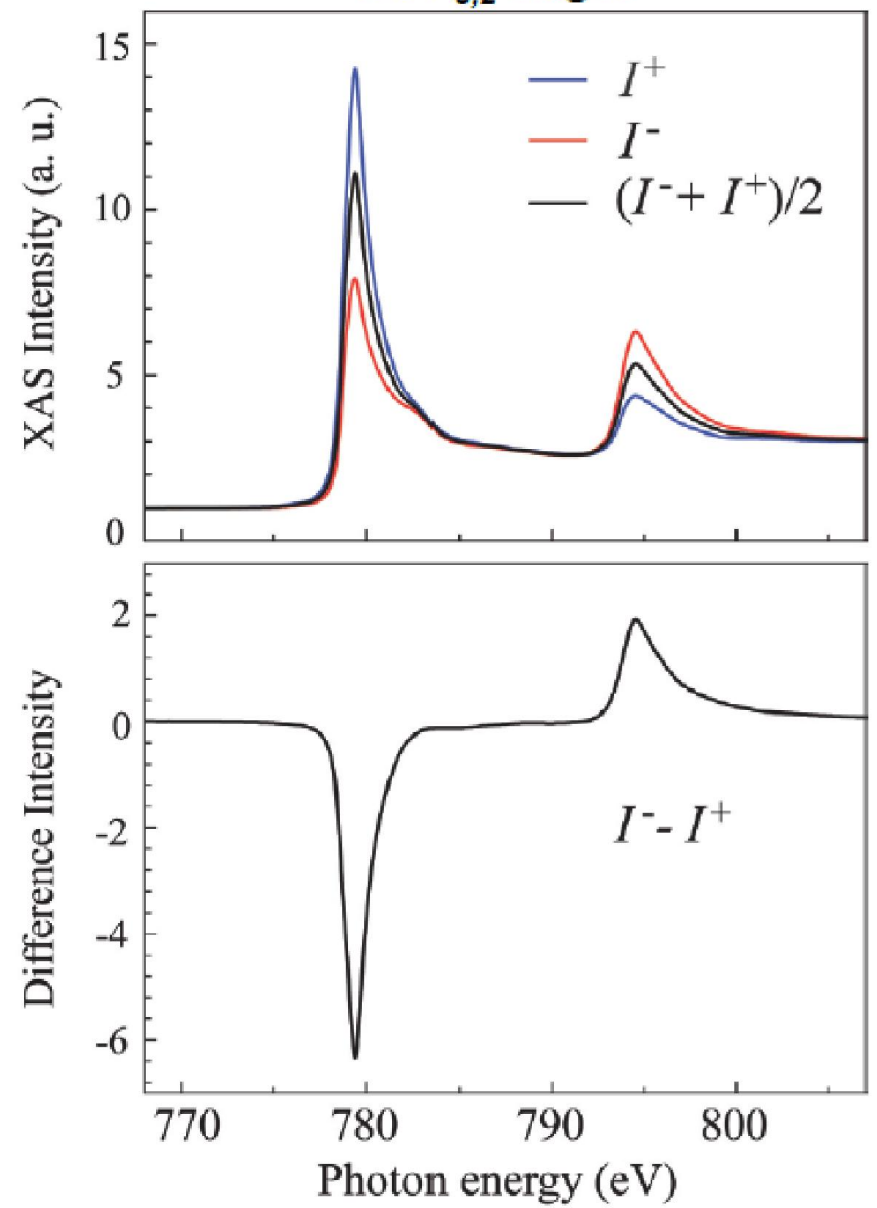


	holes
Fe	3.4
Co	2.5
Ni	1.5

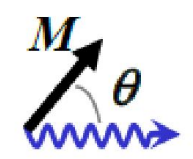




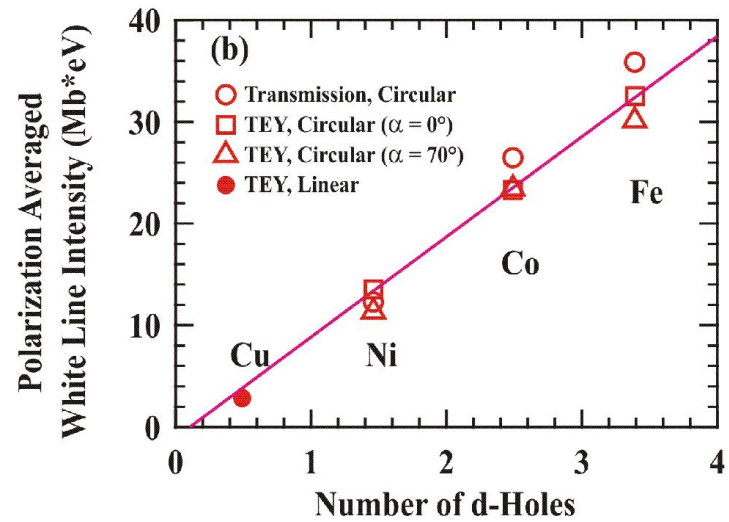
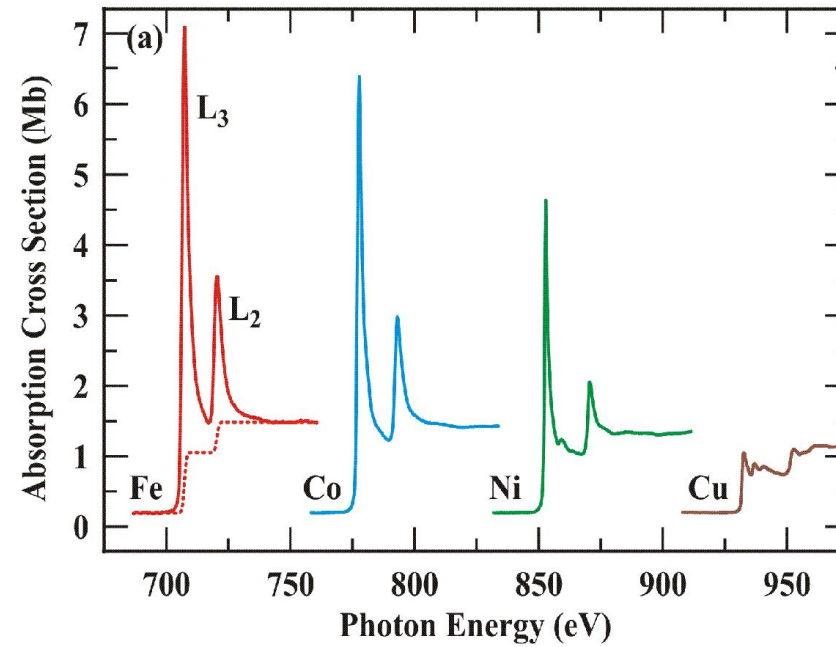
Co $L_{3,2}$ edges



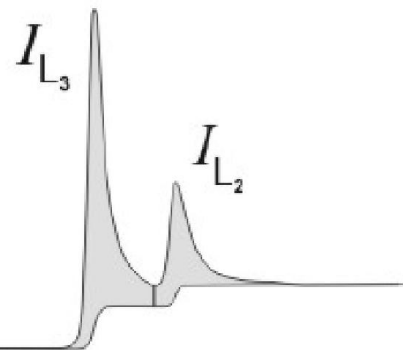
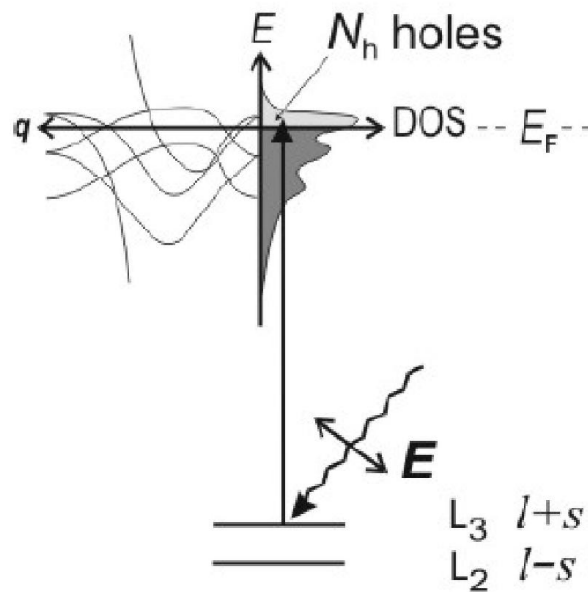
$$I_{XMCD} \propto M \cos \theta$$



White Line Intensity in 3d Metals



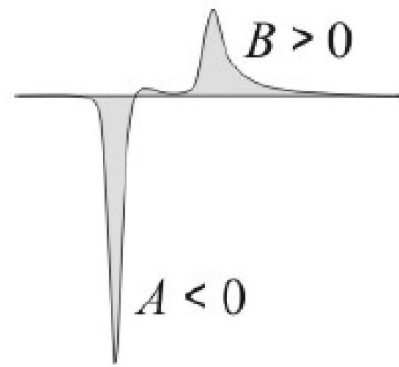
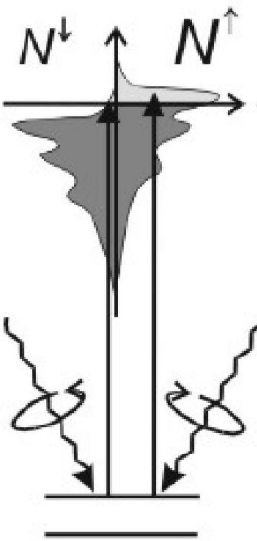
(a) *d*-Orbital occupation



$$N_h = \langle I_{L_3} + I_{L_2} \rangle / C$$

(b) Spin moment

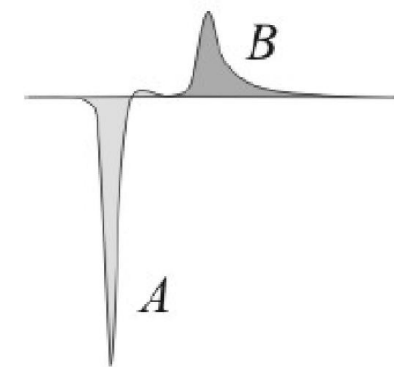
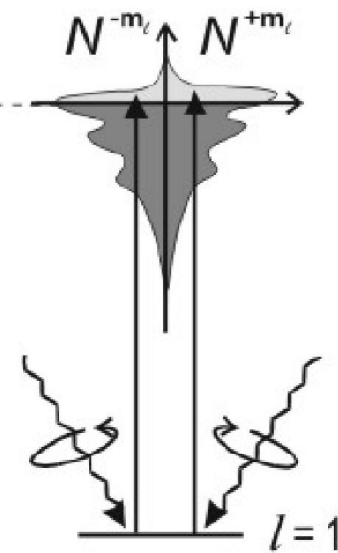
$$-\frac{1}{2} \downarrow \quad \uparrow + \frac{1}{2}$$



$$m_s = \mu_B \langle -A + 2B \rangle / C$$

(c) Orbital moment

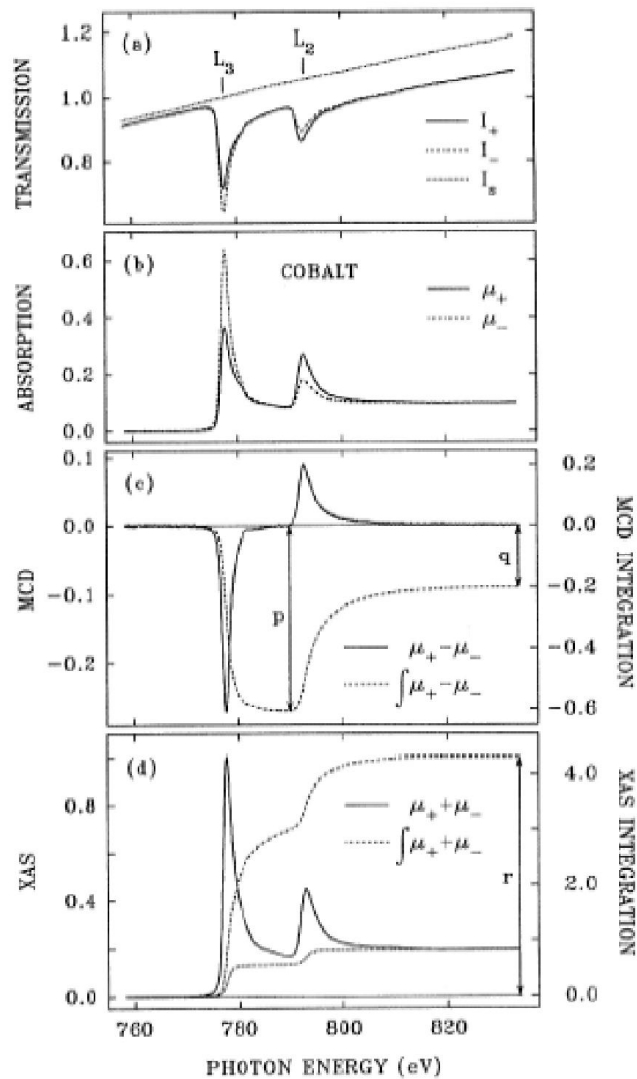
$$-m_l \quad +m_l$$



$$m_o = -2\mu_B \langle A + B \rangle / 3C$$

Carra, *et al.*, PRL 70, 694 (1993); Thole *et al.*, PRL 68, 1943 (1992);

Transmission through 50 Å Co on 1 μm parylene



C.T. Chen et al., PRL 75, 152 (1995)

$$m_L = 2n_h \frac{\int_{L_3+L_2} (\mu^+ - \mu^-) dE}{\int_{L_3+L_2} (\mu^+ + \mu^- + \mu^0) dE}$$

ORBITAL

$$m_S + m_T = 3n_h \frac{\int_{L_3} (\mu^+ - \mu^-) dE - 2 \int_{L_2} (\mu^+ - \mu^-) dE}{\int_{L_3+L_2} (\mu^+ + \mu^- + \mu^0) dE}$$

SPIN

where

$$m_L = -\langle L_\alpha \rangle$$

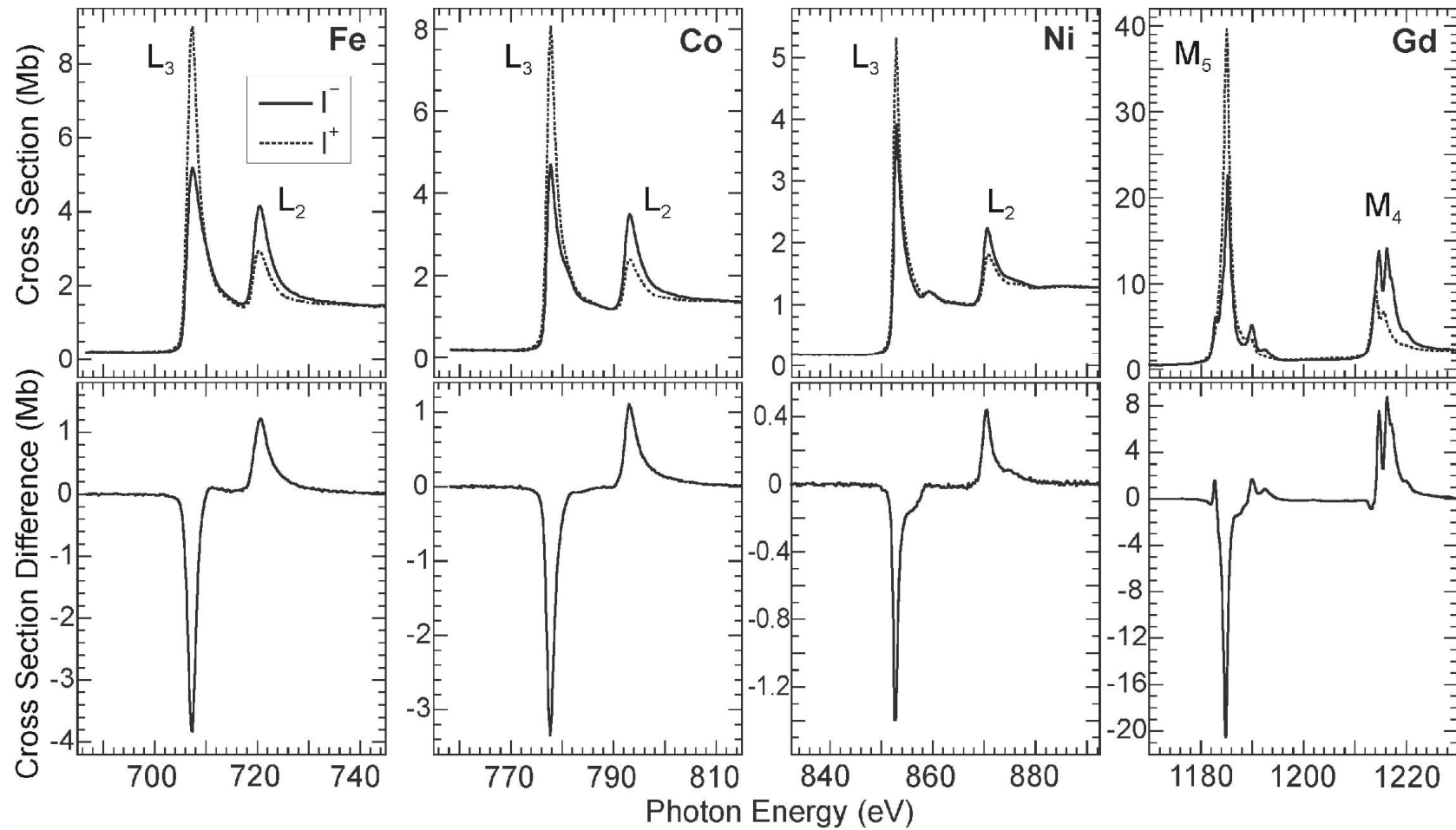
$$m_S = -\langle 2S_\alpha \rangle; \quad m_T = -7 \langle S_\alpha - 3\hat{r}(\hat{r} \cdot S_\alpha) \rangle$$

Sum-rules results:

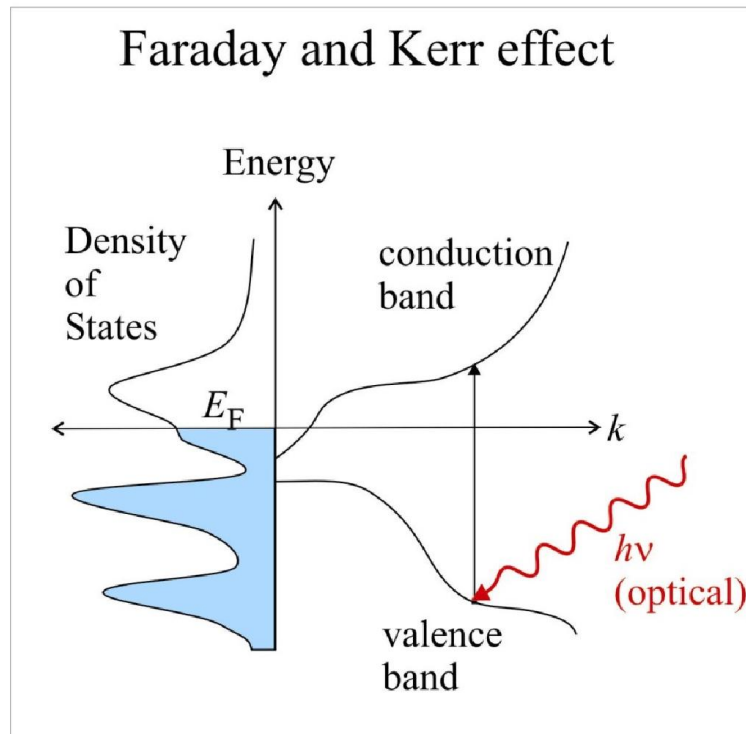
$$\text{Fe } (n_{3d} = 6.61): \quad m_{\text{orb}} = 0.086, \quad m_{\text{spin}} = 1.98$$

$$\text{Co } (n_{3d} = 7.51): \quad m_{\text{orb}} = 0.153, \quad m_{\text{spin}} = 1.55$$

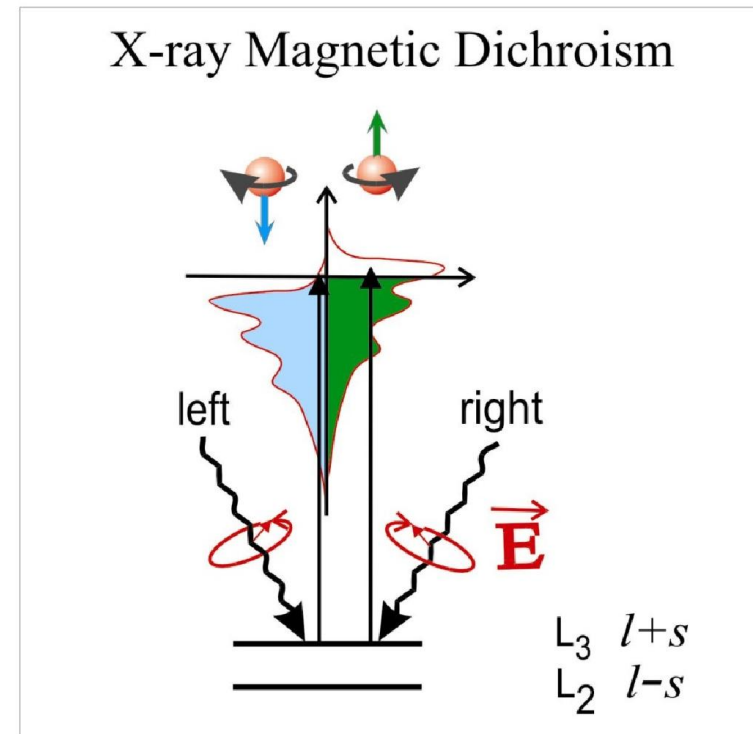
XMCD spectra of the pure ferromagnetic metals



Magnetic Circular Dichroism



Magneto-optical response:
weak, k -dependent



X-ray response:
strong, k -integrated quantities
number of holes, spin moment, orbital moment

- spin moments,
- orbital moments
- nanoscale resolution

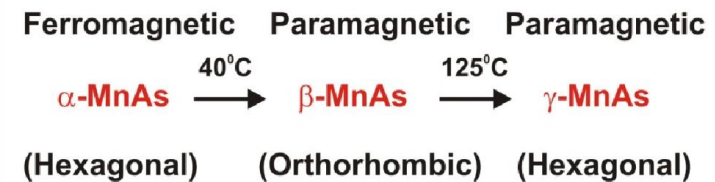
Magnetic Semiconductor: MnAs/GaAs

- MnAs on GaAs integrates magnetic and semiconducting properties
- room temperature ferromagnetism
- spin injection demonstrated

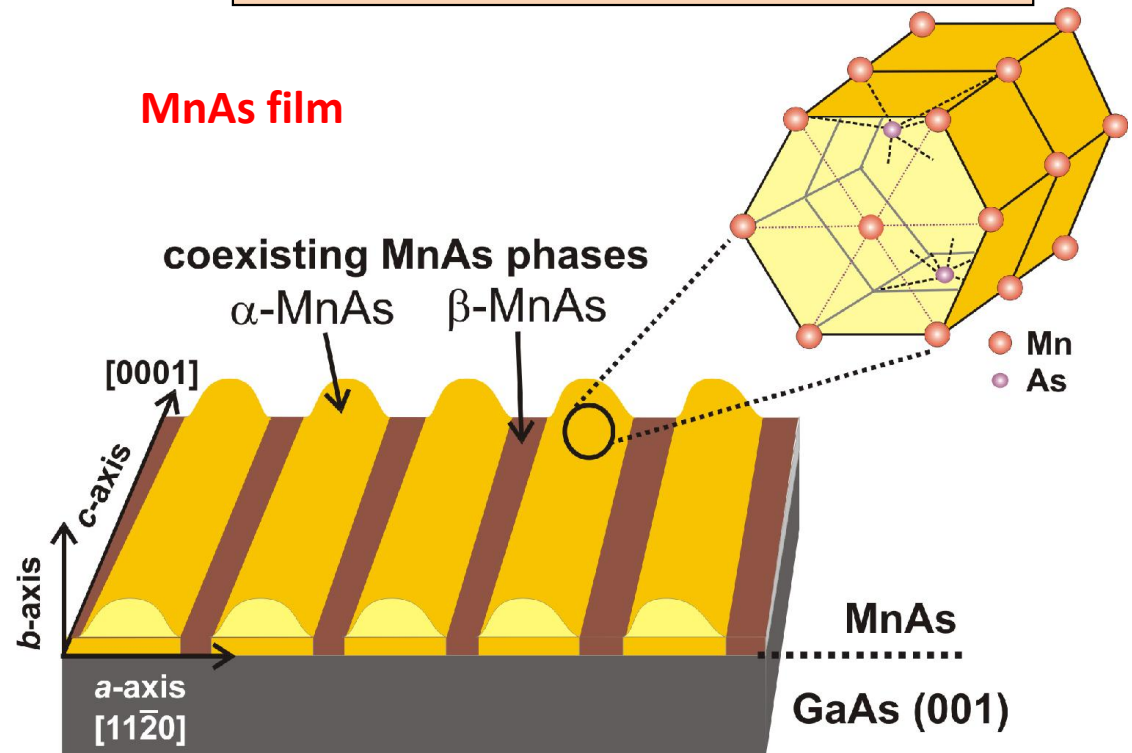
Ramsteiner *et.al*, Phys. Rev. B, 66, 081304(R), 2002.

MnAs bulk properties

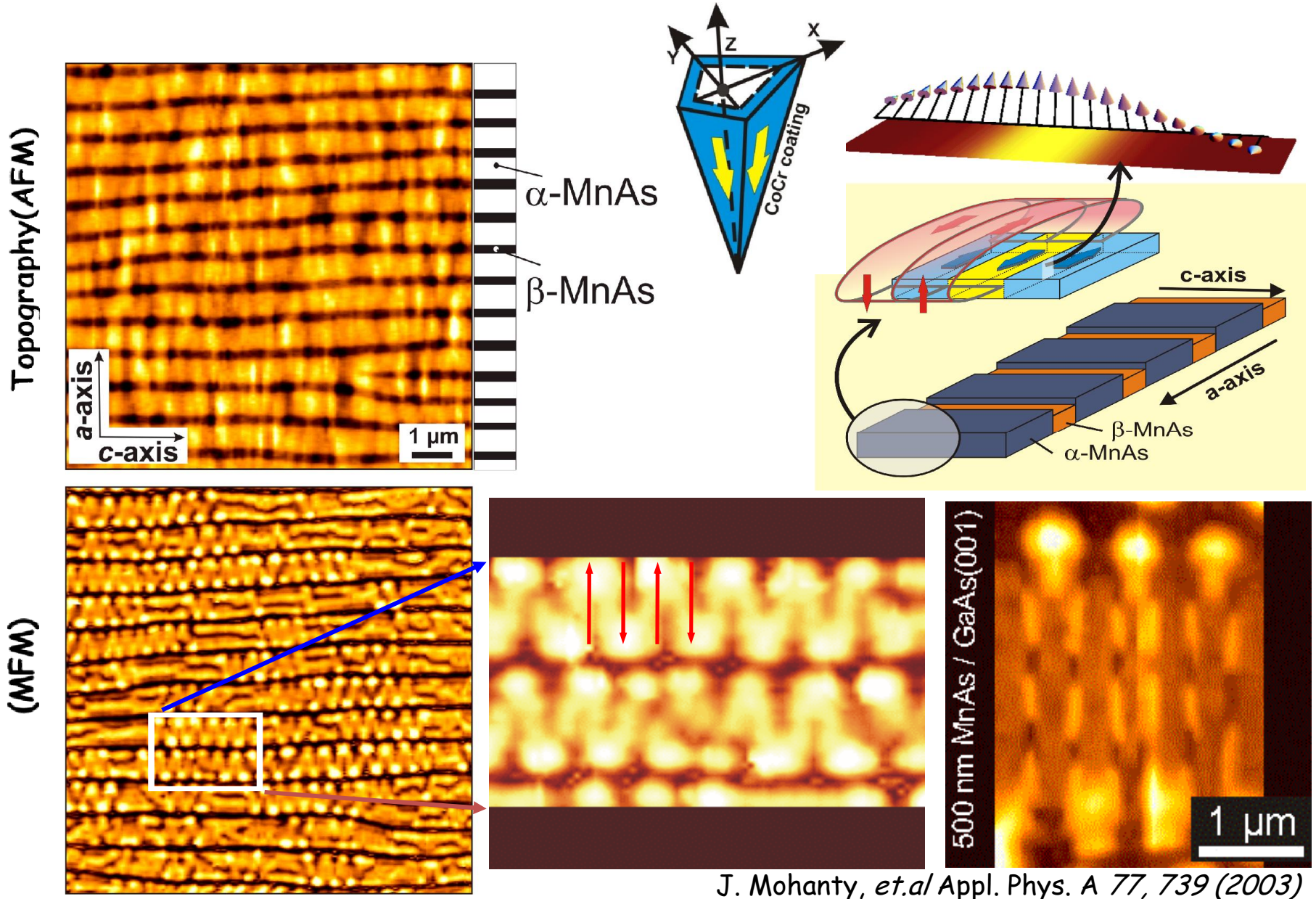
- phase transition: 1st order
- volume change ~ 2%



- strain induced phase coexistence
- thermal expansion mismatch between film and substrate
- stripe width function of temperature
- stripe periodicity function of film thickness
- magnetic easy and hard axes in the film plane

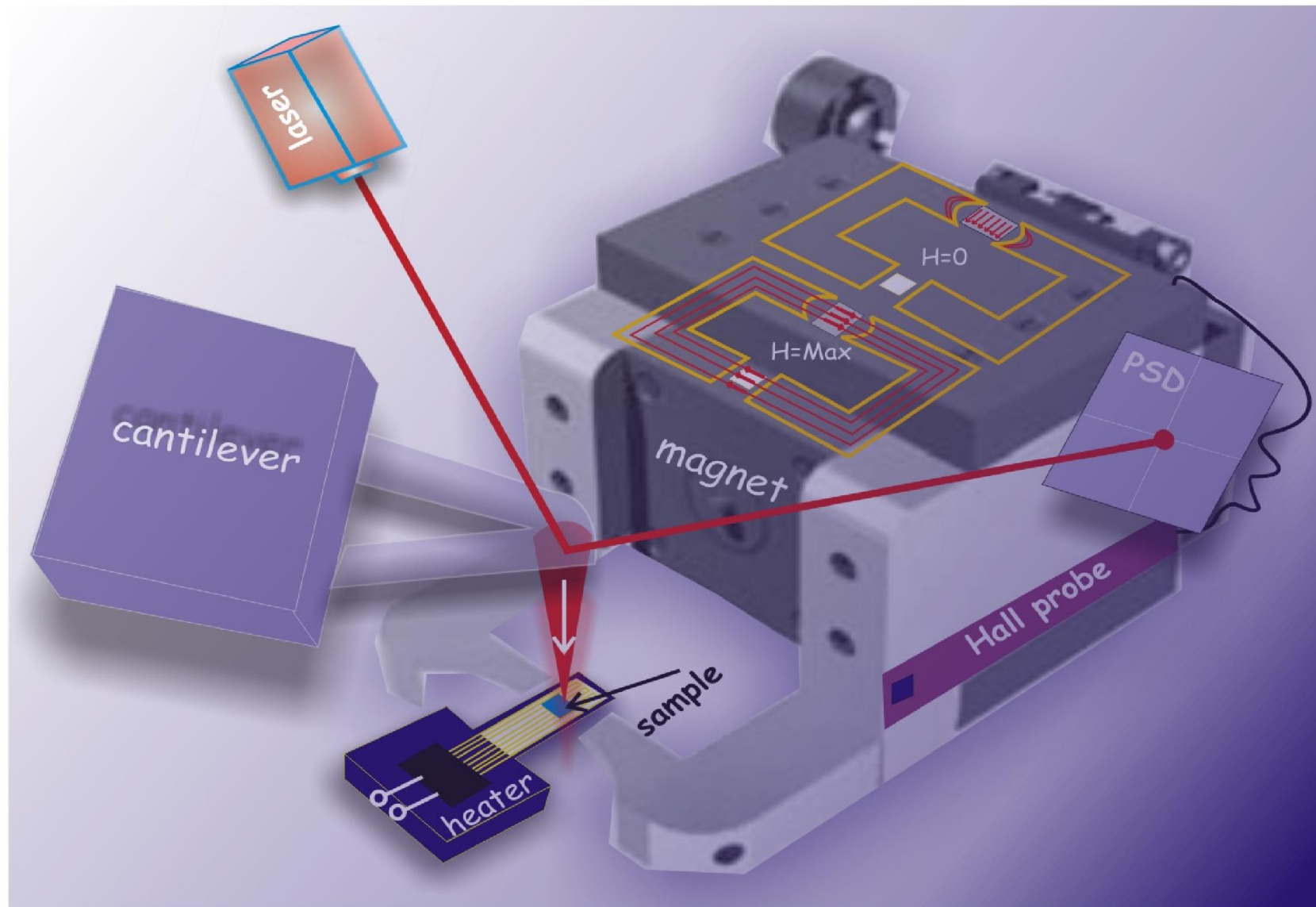


MFM Contrast of MnAs Films



J. Mohanty, *et.al* Appl. Phys. A 77, 739 (2003)

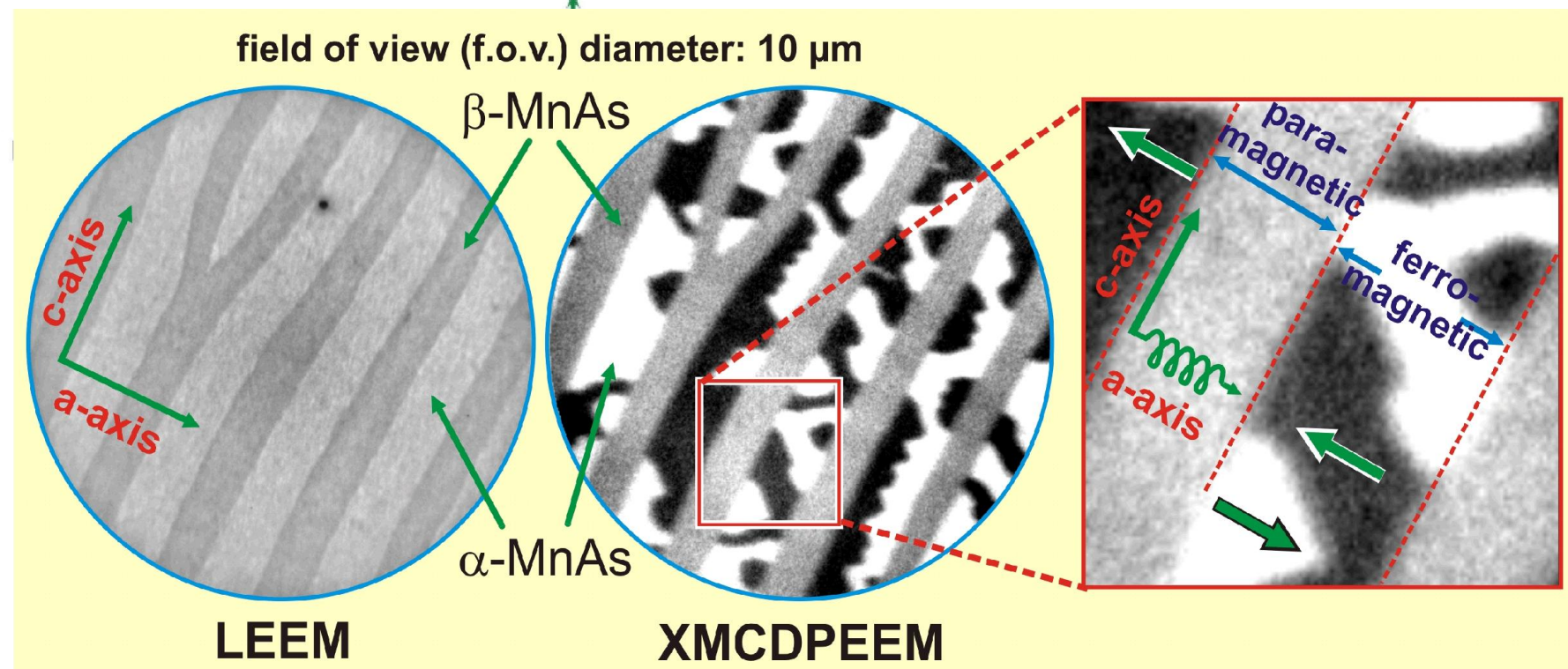
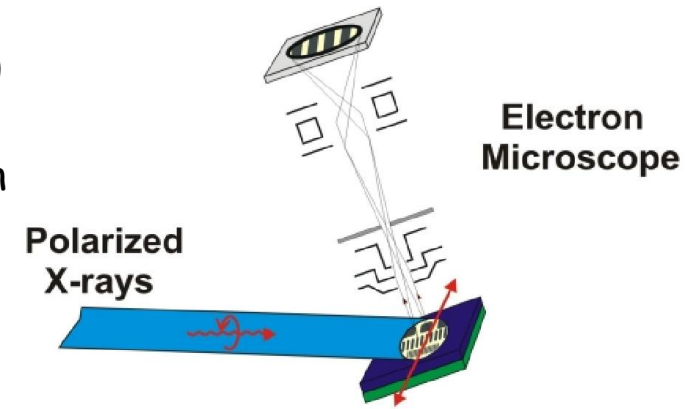
MFM Set-up



J. Mohanty, *et.al* Appl. Phys. A 81, 1359 (2005)

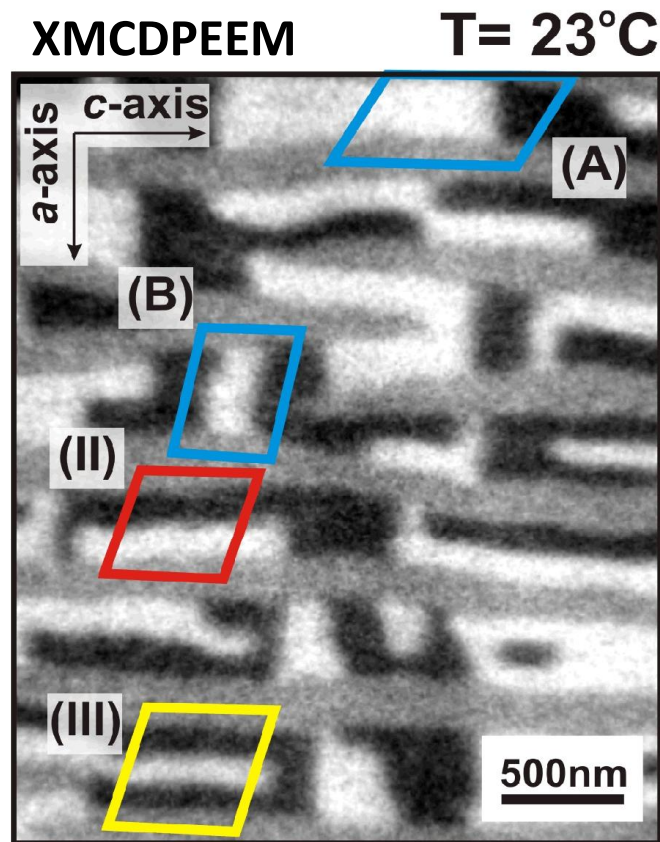
X-ray Magnetic Imaging

- Excitation: XMCD (X-ray magnetic circular dichroism)
- Imaging: PEEM (photoemission electron microscopy)
- SE yield = $\sigma \times M$, σ : incident photon, M : magnetization
- $C_{\text{XMCDPEEM}} = (I_+ - I_-) / (I_+ + I_-)$
- Element specific



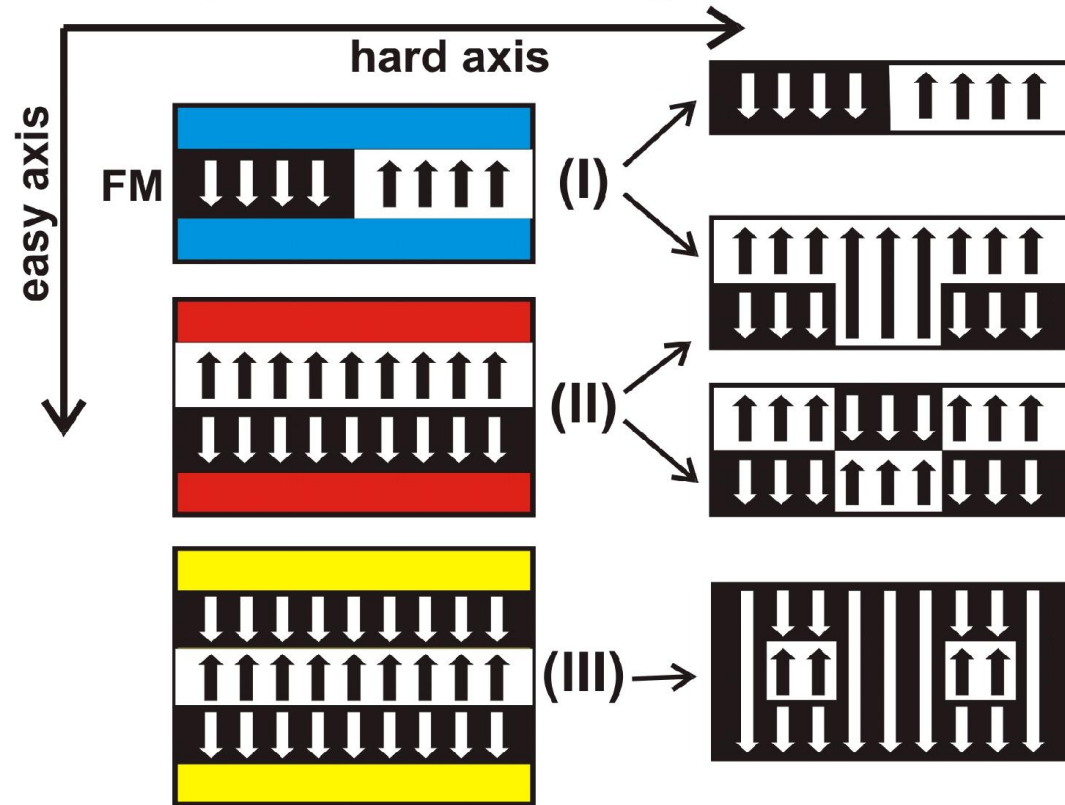
LEEM: Low energy electron microscopy

Domain Classification

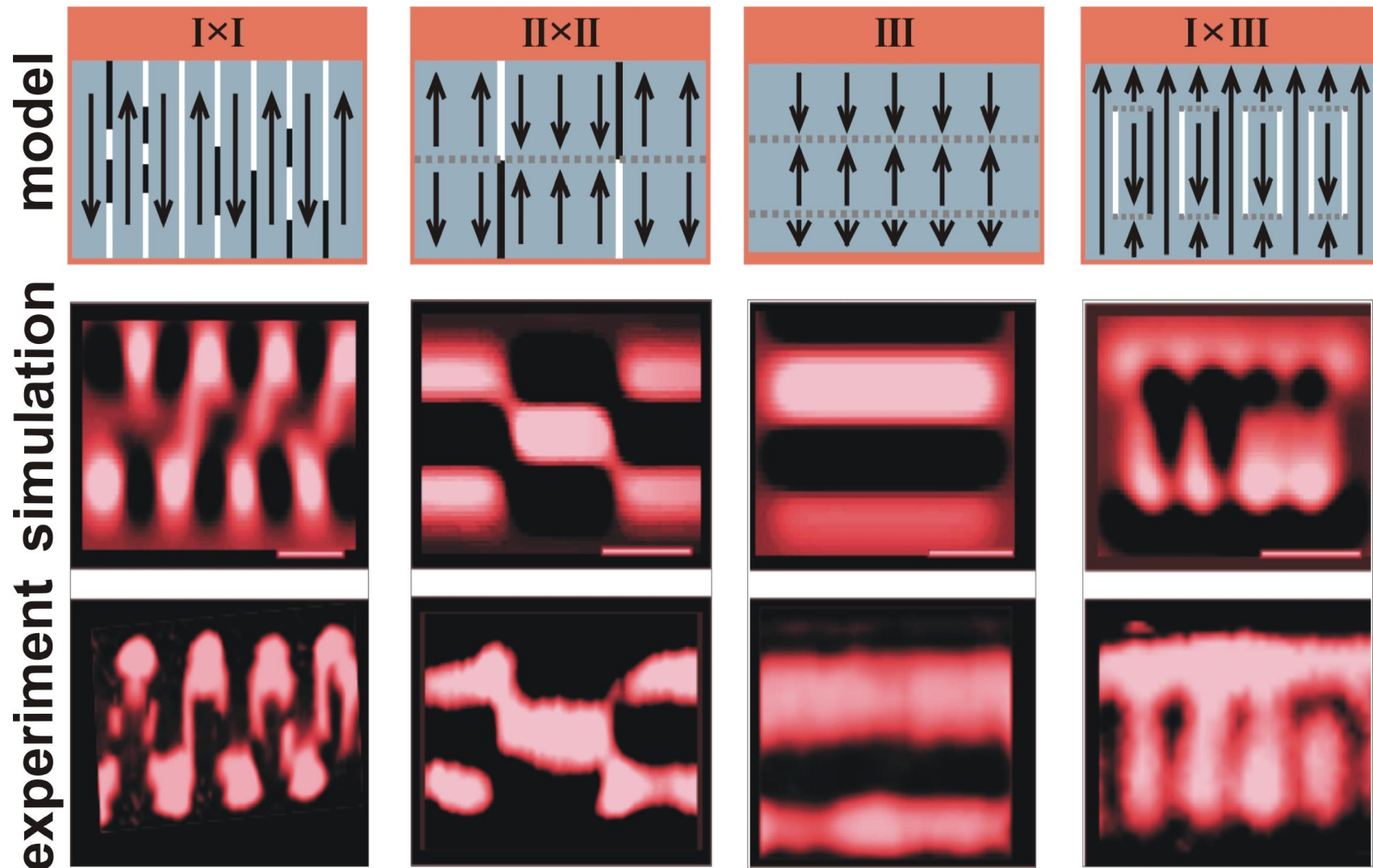


- I → (A) extended (I)
- II → (B) alternating (I)
- III film thickness = 120 nm

Simple domain configurations



Micromagnetic Modeling



MFM

Appl. Phys. Lett. 84, 1132 (2004).

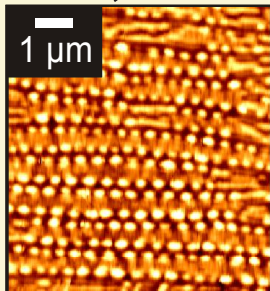
bar represents 500 nm

Structural and Magnetic Phase Transition

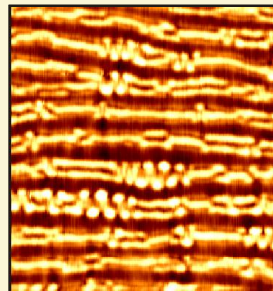
MFM

heating

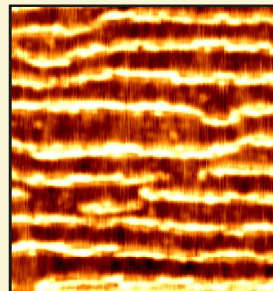
c-axis
a-axis
1 μm



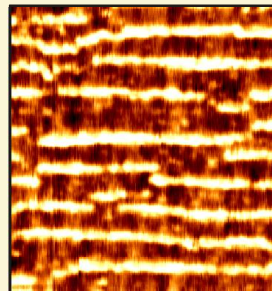
15°C



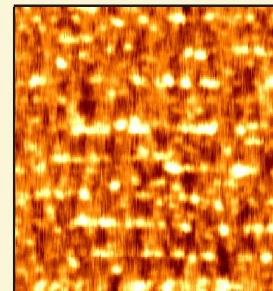
25°C



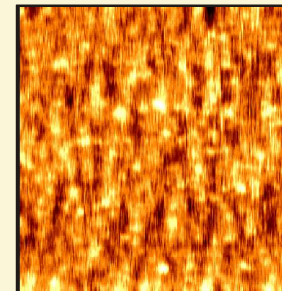
33°C



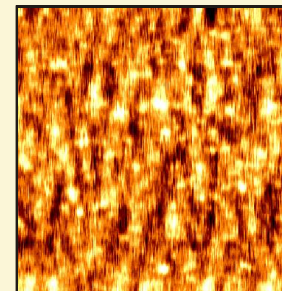
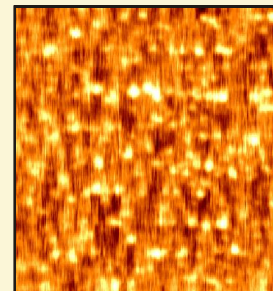
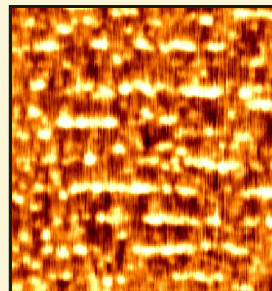
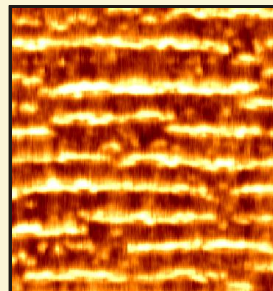
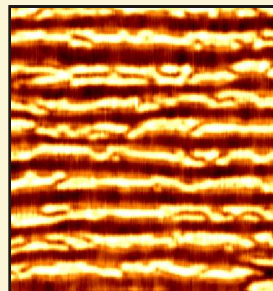
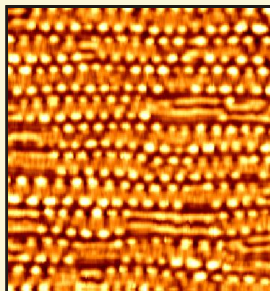
35°C



37°C



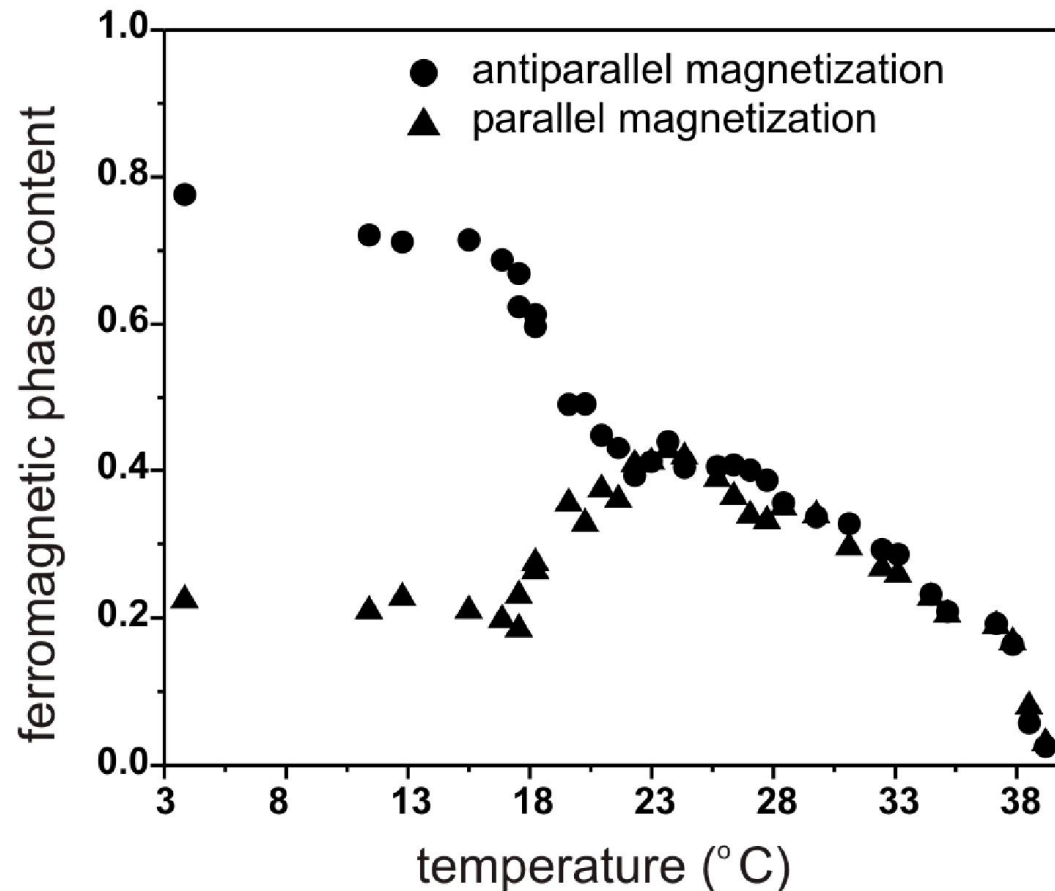
41°C



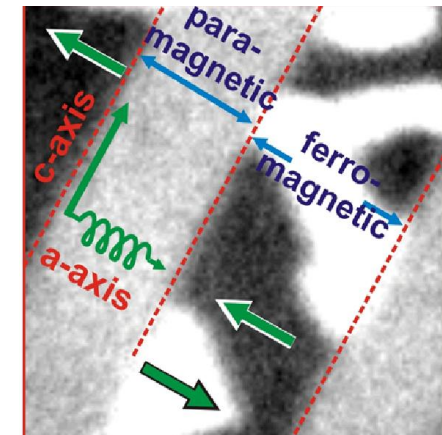
cooling

J. Mohanty, *et.al* Appl. Phys. A 77, 739 (2003),
Appl.Phys. Lett. 82, 2308-2310 (2003).

Temperature Dependence of the Domain Structure

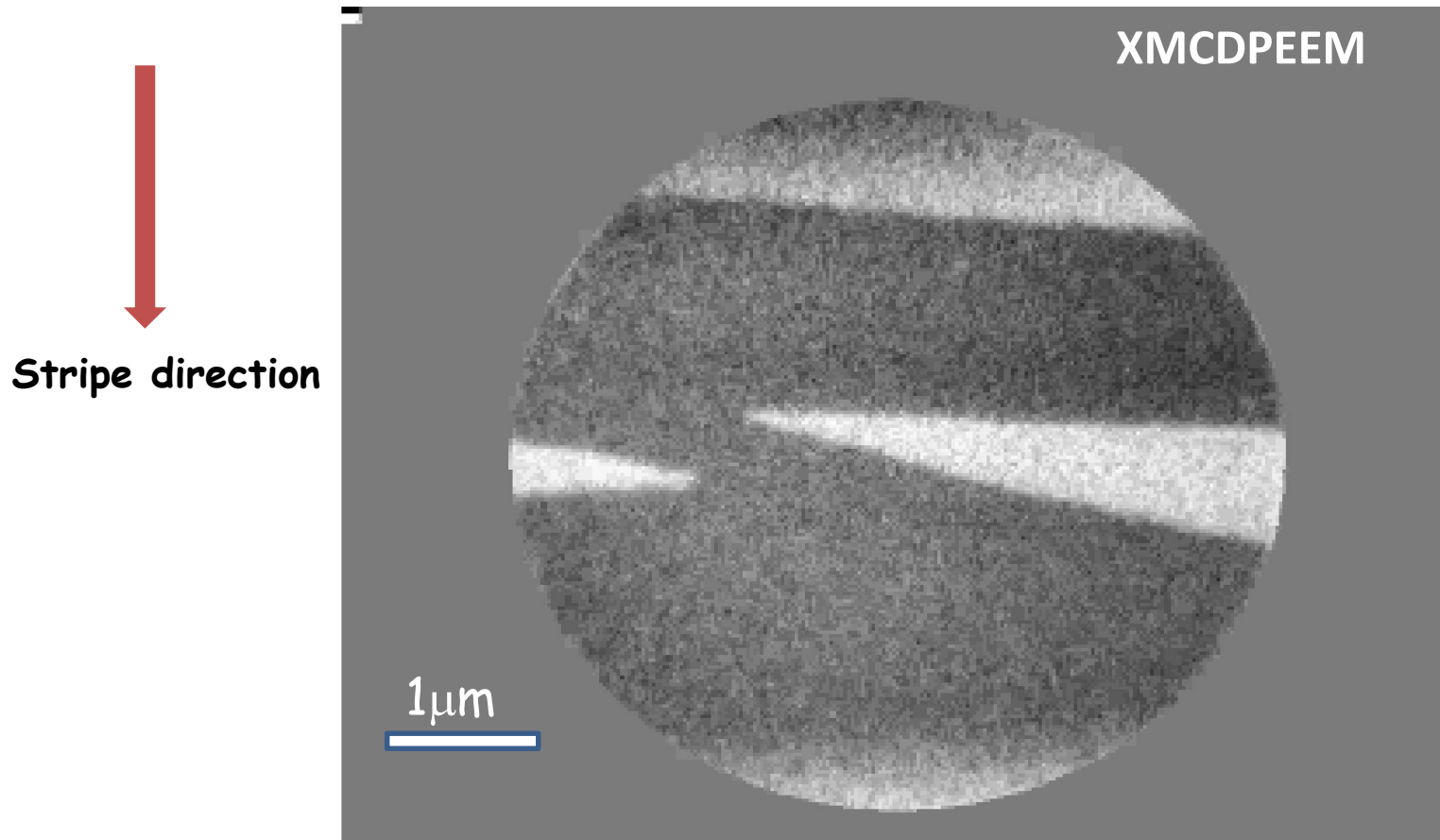


XMCDPEEM



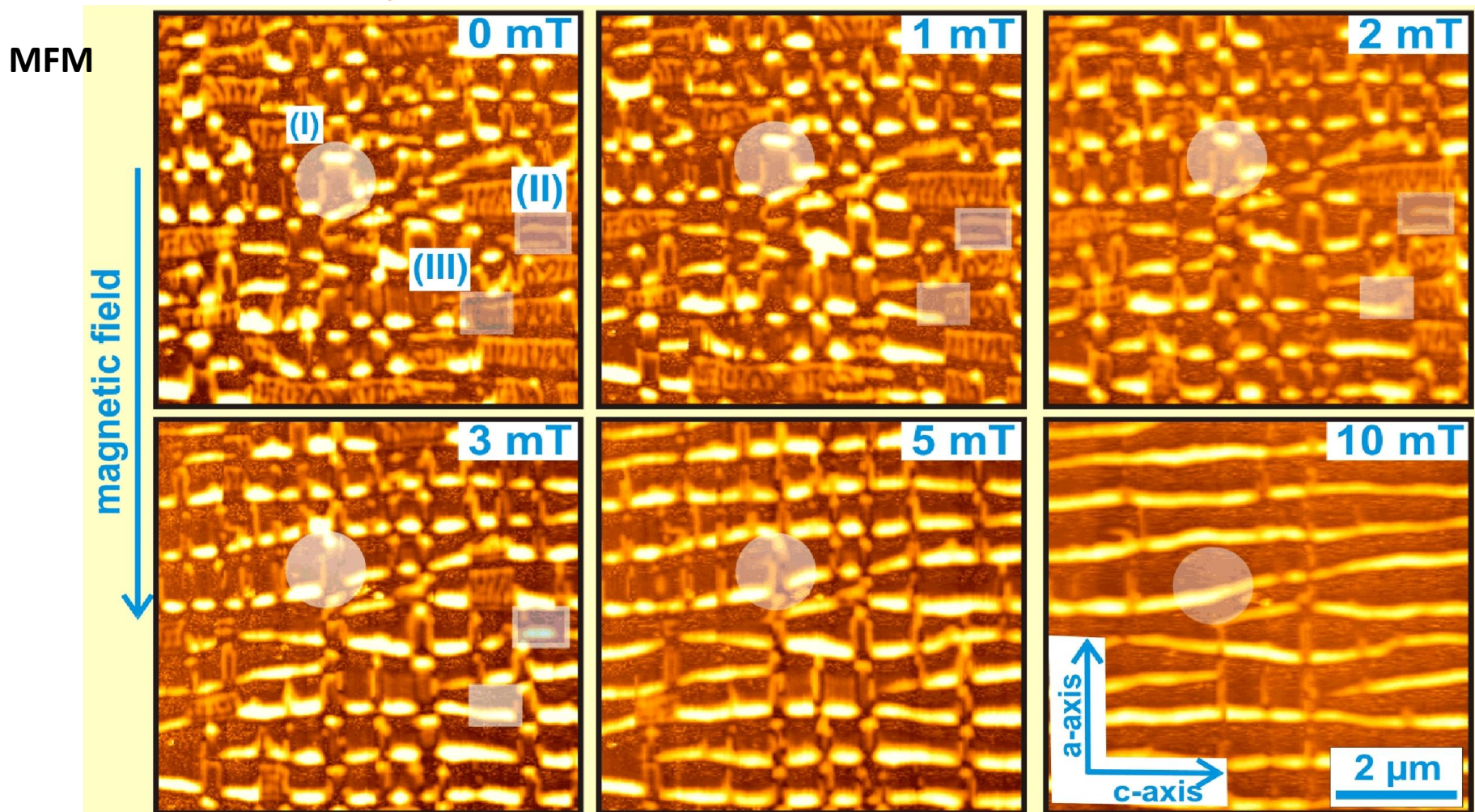
- magnetic stripe demagnetize at 22°C
- small distance favors inter-stripe dipole-dipole coupling

Temperature Dependence of the Domain Structure



MnAs/GaAs(001) f.o.v.diameter = 5 μm Thickness=120nm

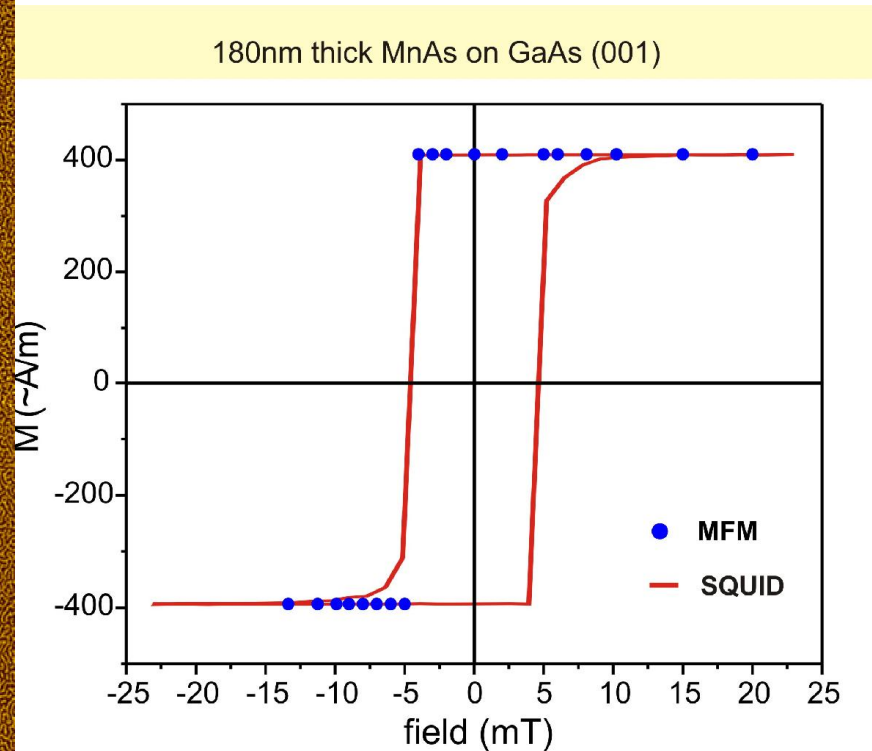
Field Dependence of Magnetic Domains



- type (III) domains **vanish first** (already at 3 mT)
- type (I) domains are the **most stable domains**

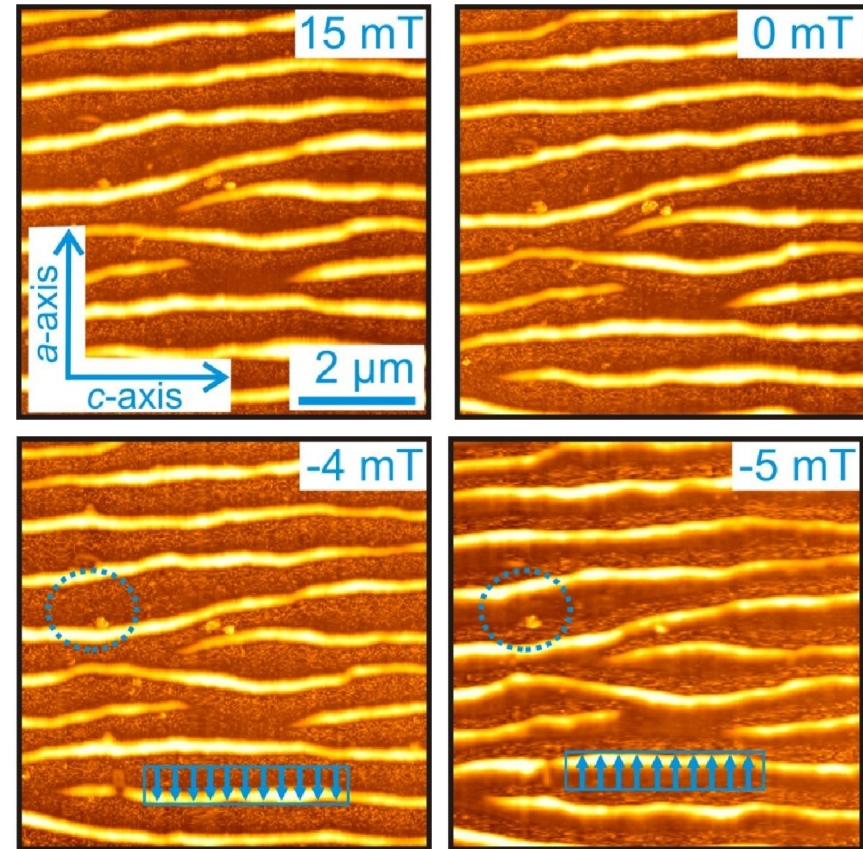
Microscopic Magnetization Reversal

MFM



measured along the easy magnetization direction of MnAs

Phys. Rev. B. 73, 104441 (2006)

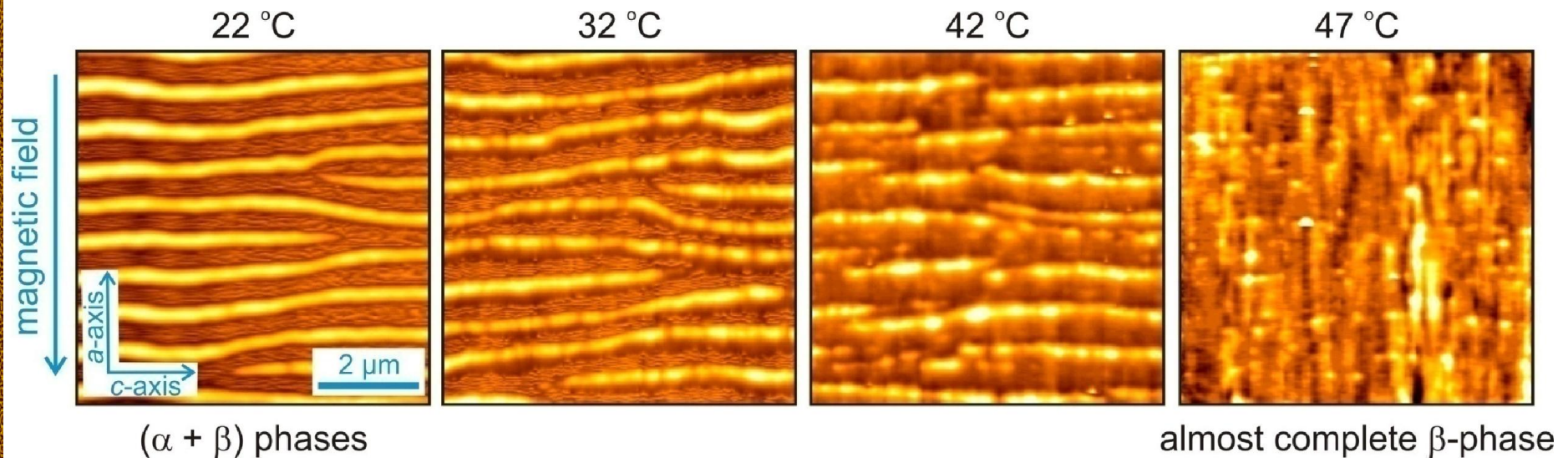
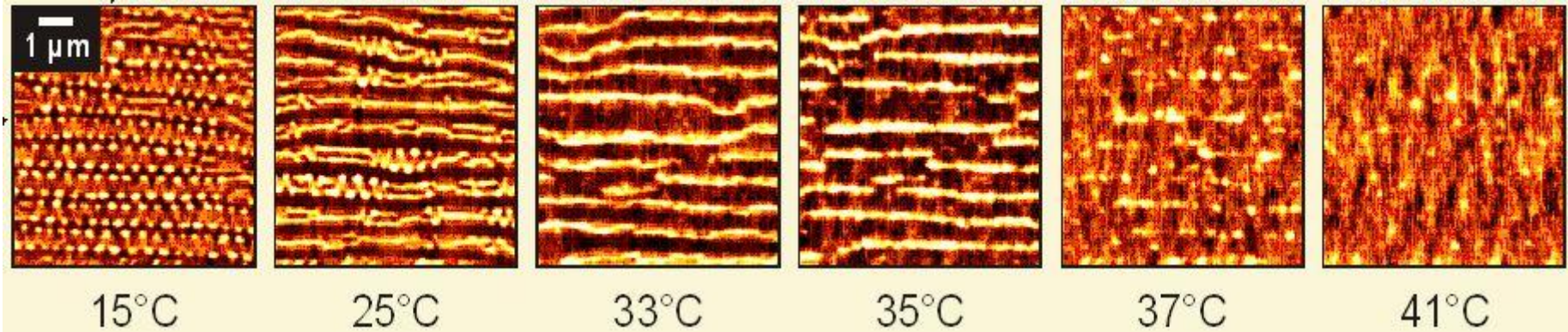


● **coercive field** (magnetization reversal):
5 mT

● **microscopic MFM** measurements agree well
with the **integral SQUID** measurements

Field Dependence of Phase Transition

MFM



Ferromagnetic phase stabilized above the bulk phase transition temperature by an external magnetic field: **Magnetism** → **Structure**

Strain Effect on the Phase Transition

XMCDPEEM

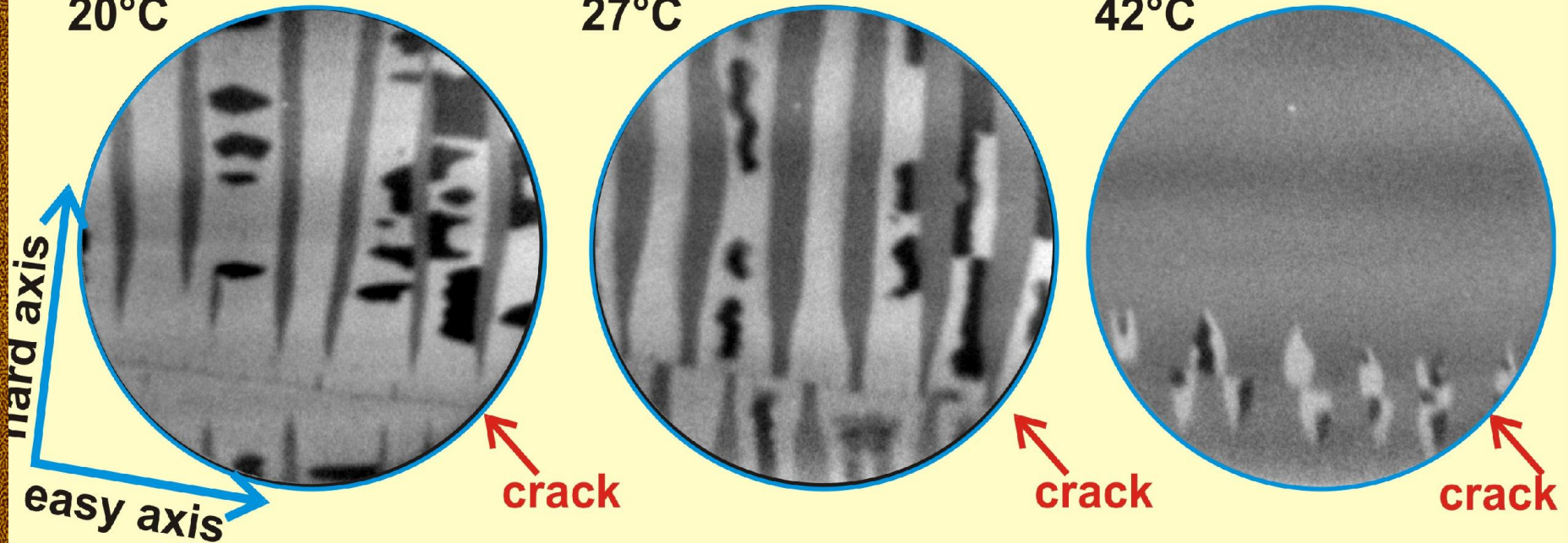
500 nm MnAs/ GaAs(001)

f.o.v. diameter = 10 μm

20°C

27°C

42°C

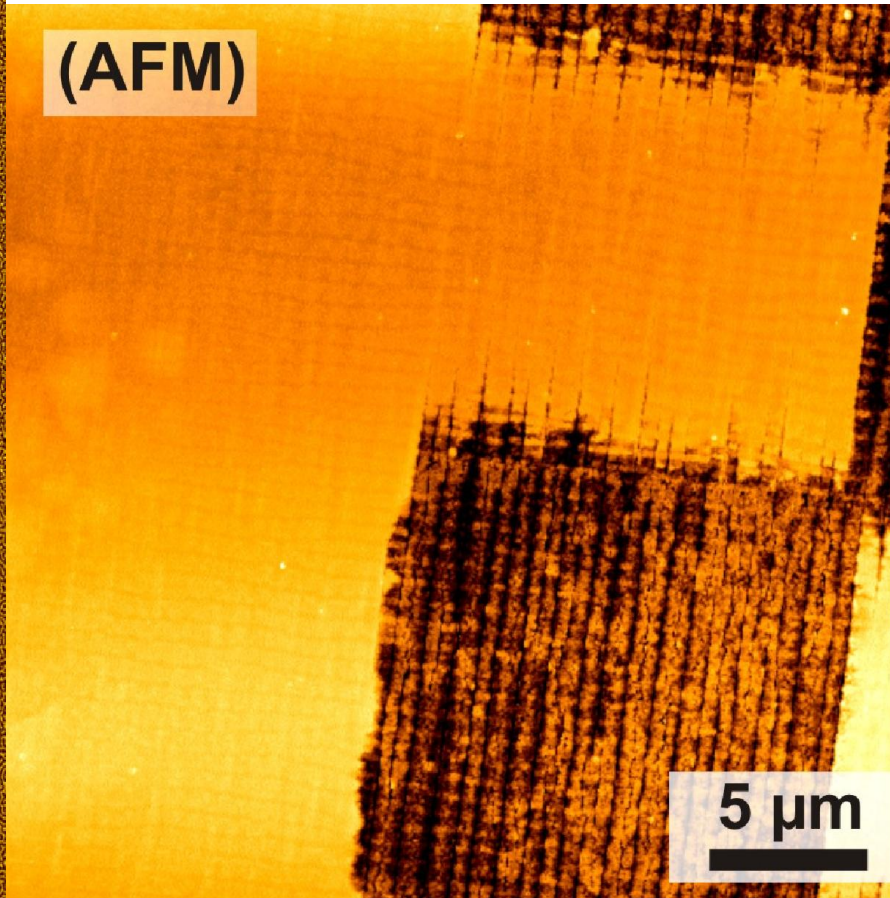


- Higher phase transition temperature due to strain relaxation: **Structure** \rightarrow **Magnetism**

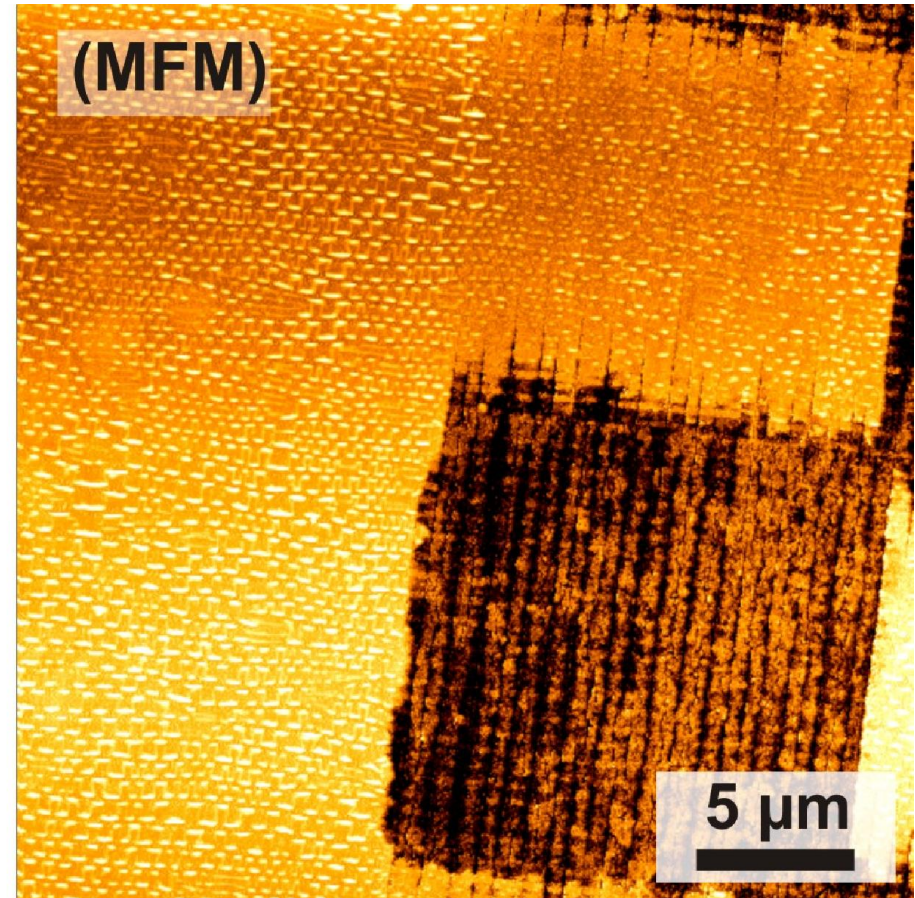
J. Mohanty, *et al* Appl. Phys. Lett. 83, 2829-2831 (2003)
J. Appl. Phys. 98, 013907(2005).

Etching

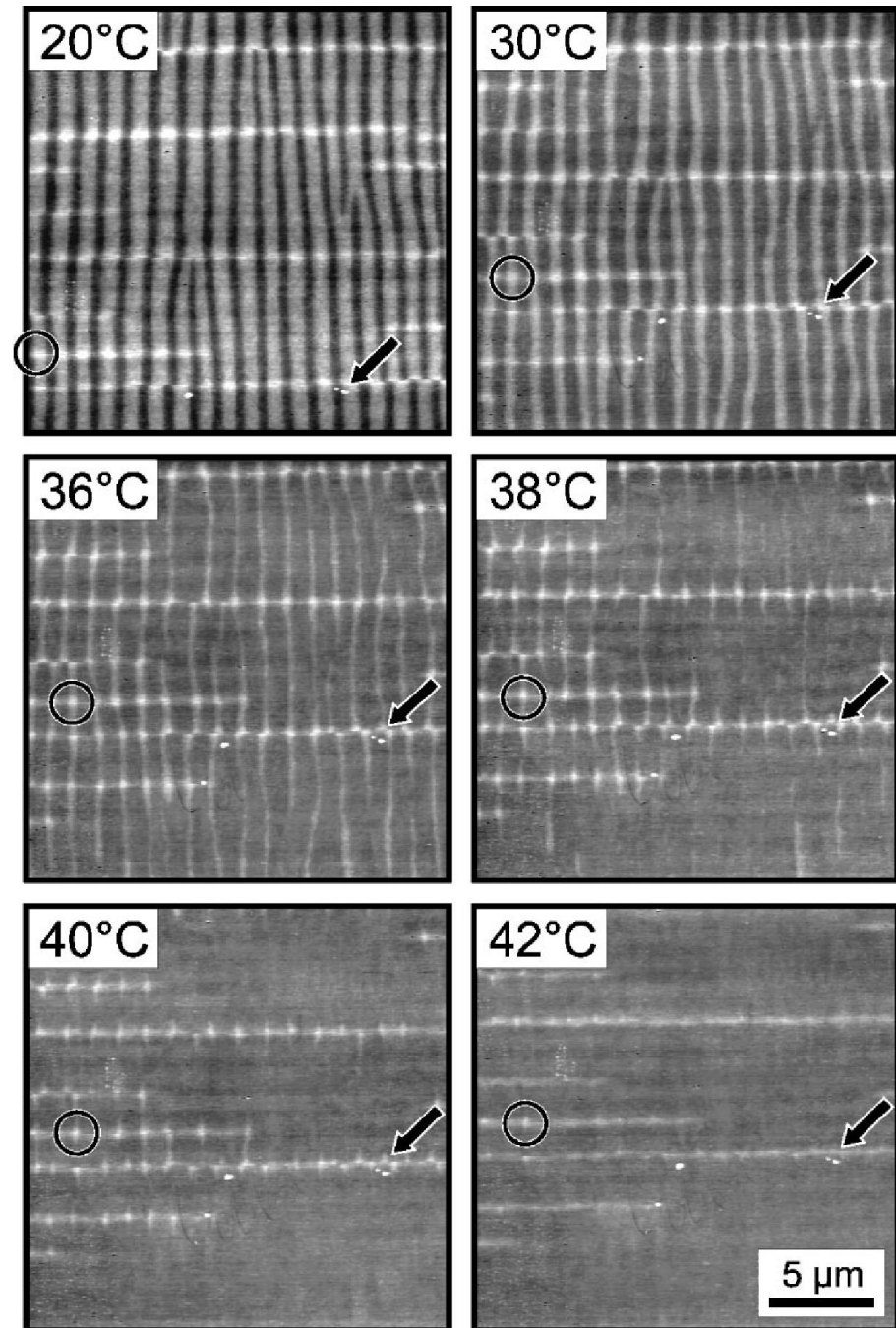
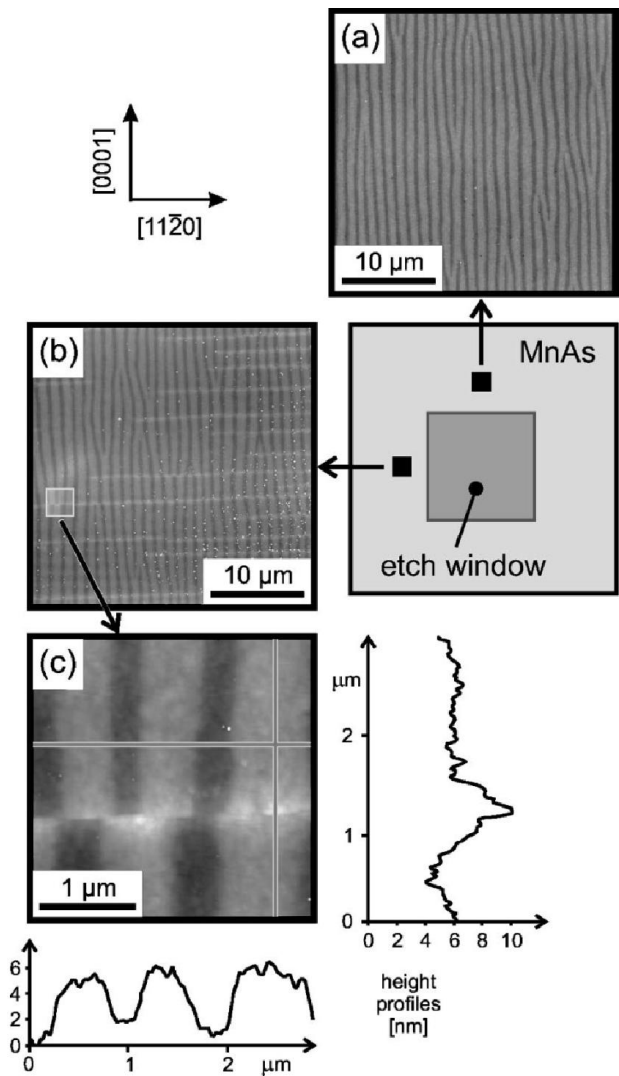
(AFM)



(MFM)

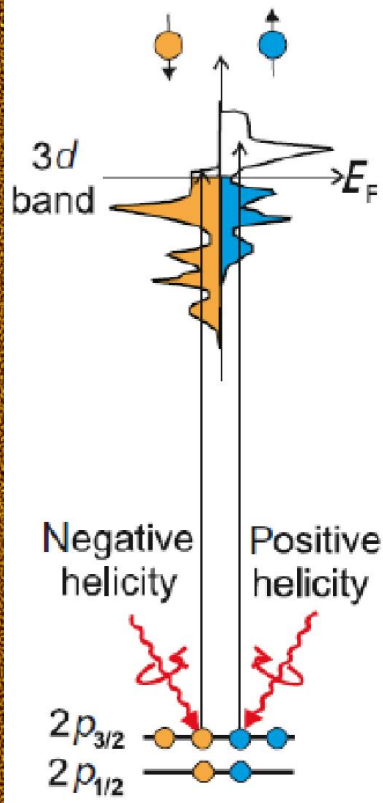


Etching and AFM Measurements

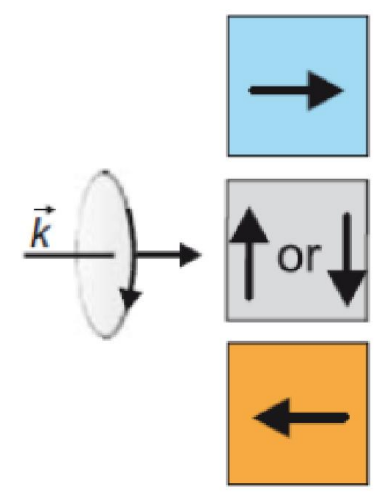
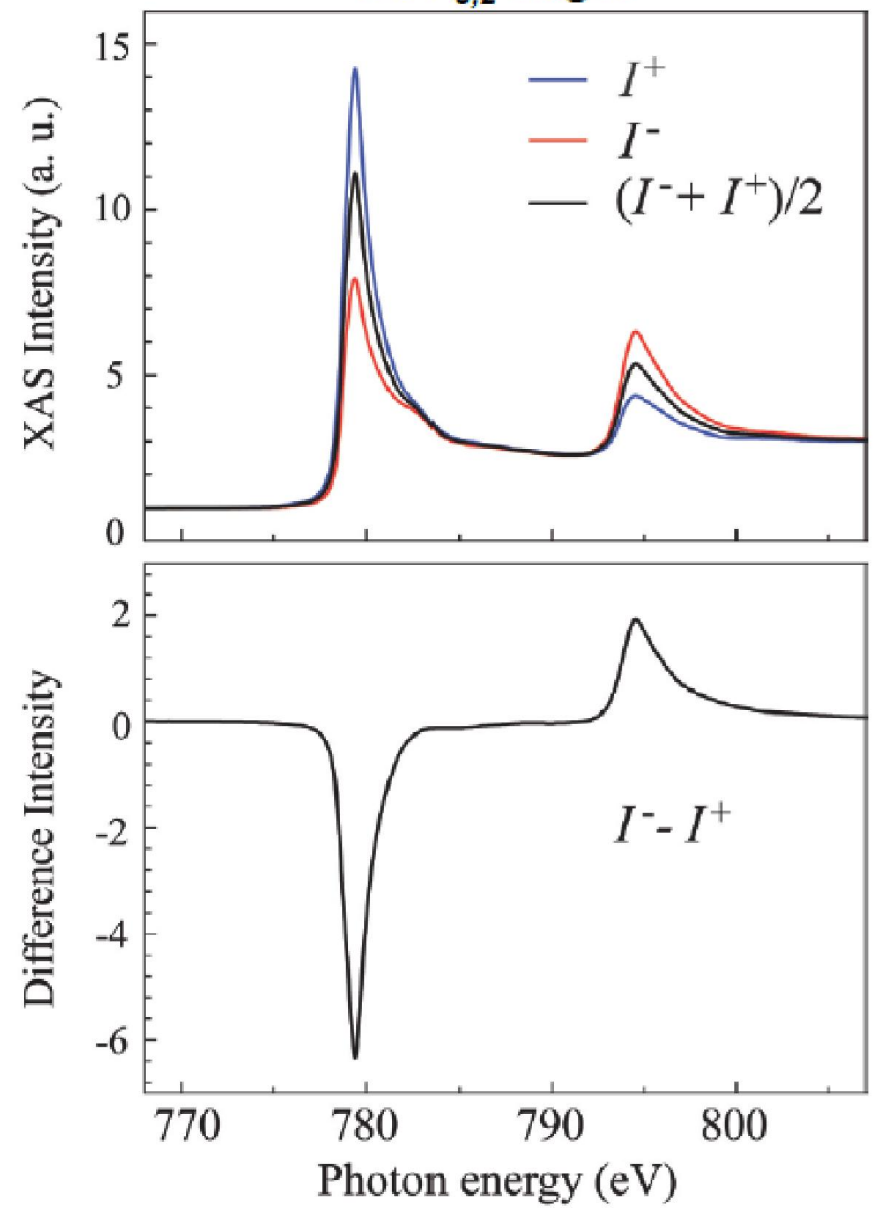


J. Mohanty, *et al* Appl. Phys. Lett. 83, 2829-2831 (2003)

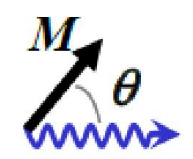
J. Appl. Phys. 98, 013907(2005).



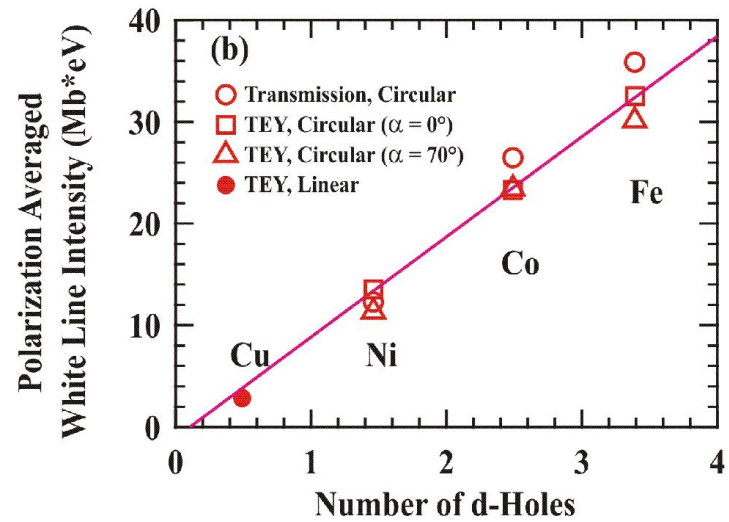
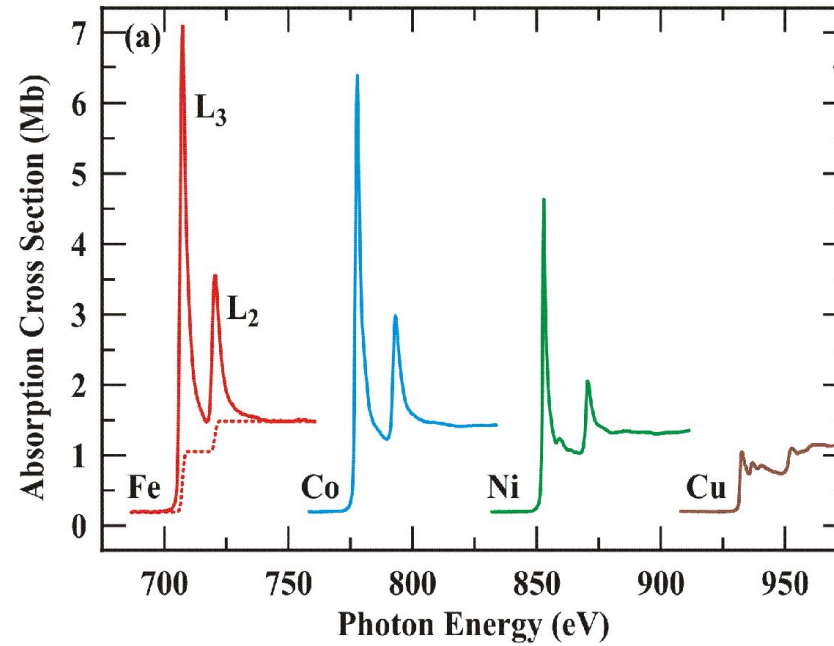
Co $L_{3,2}$ edges



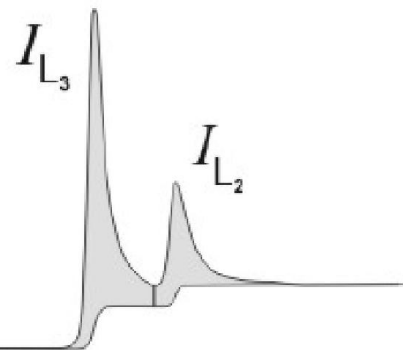
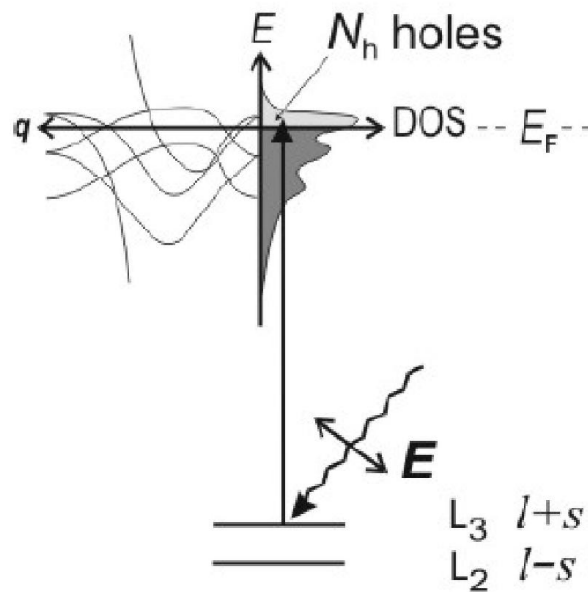
$$I_{XMCD} \propto M \cos \theta$$



White Line Intensity in 3d Metals



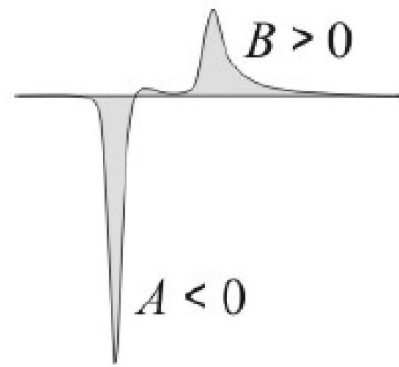
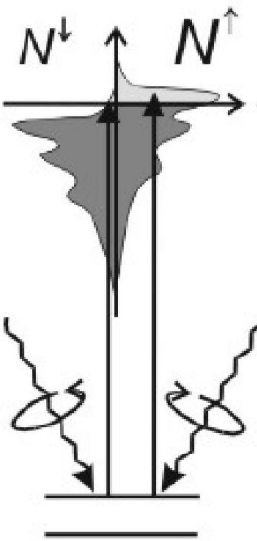
(a) *d*-Orbital occupation



$$N_h = \langle I_{L_3} + I_{L_2} \rangle / C$$

(b) Spin moment

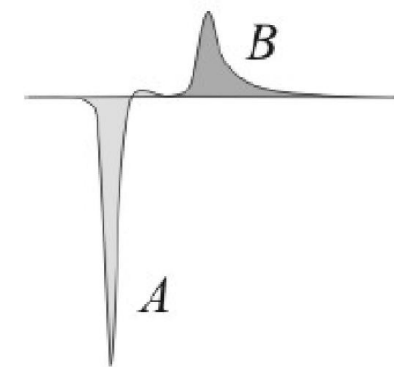
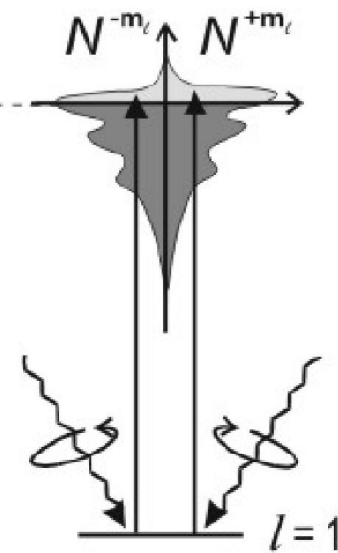
$-\frac{1}{2} \downarrow$ $\uparrow +\frac{1}{2}$



$$m_s = \mu_B \langle -A + 2B \rangle / C$$

(c) Orbital moment

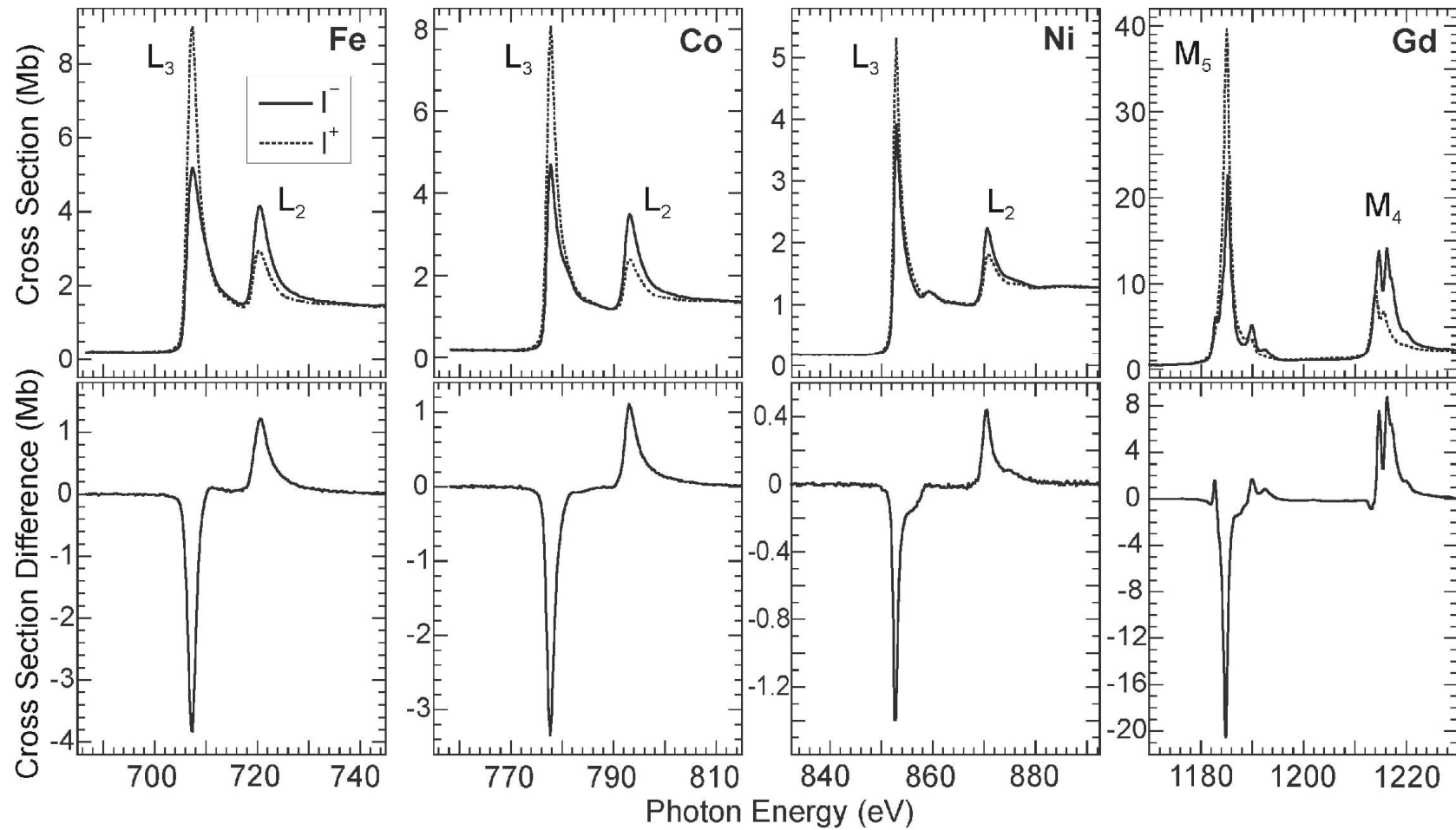
$-m_l$ $+m_l$



$$m_o = -2\mu_B \langle A + B \rangle / 3C$$

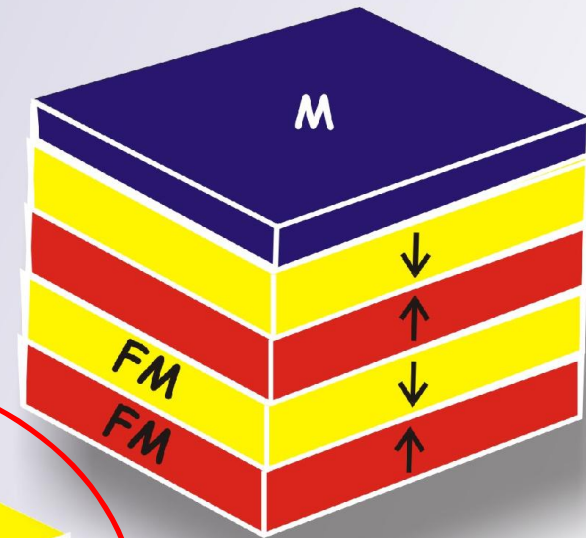
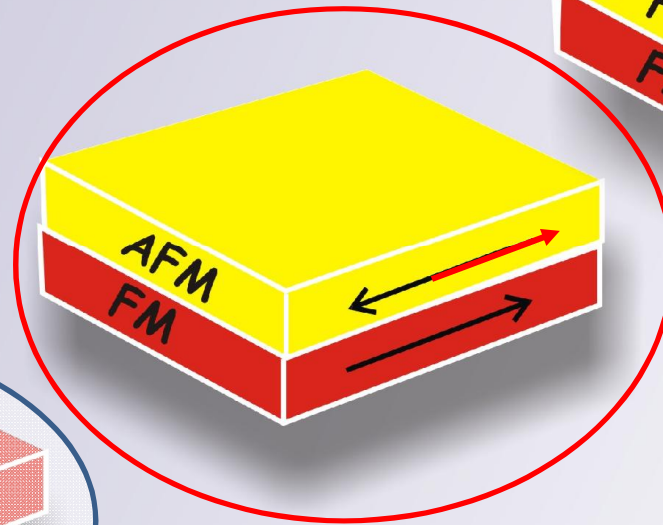
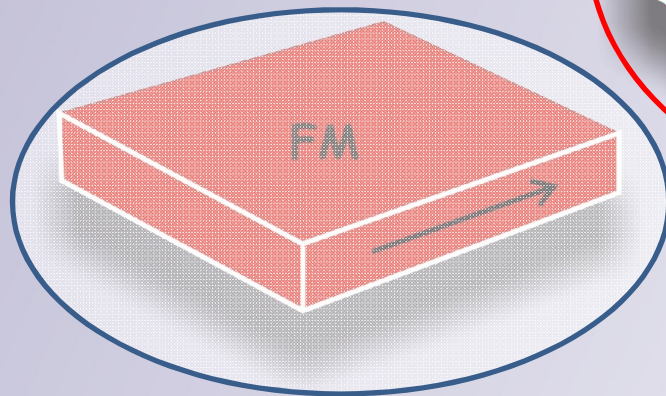
Carra, *et al.*, PRL 70, 694 (1993); Thole *et al.*, PRL 68, 1943 (1992);

XMCD spectra of the pure ferromagnetic metals

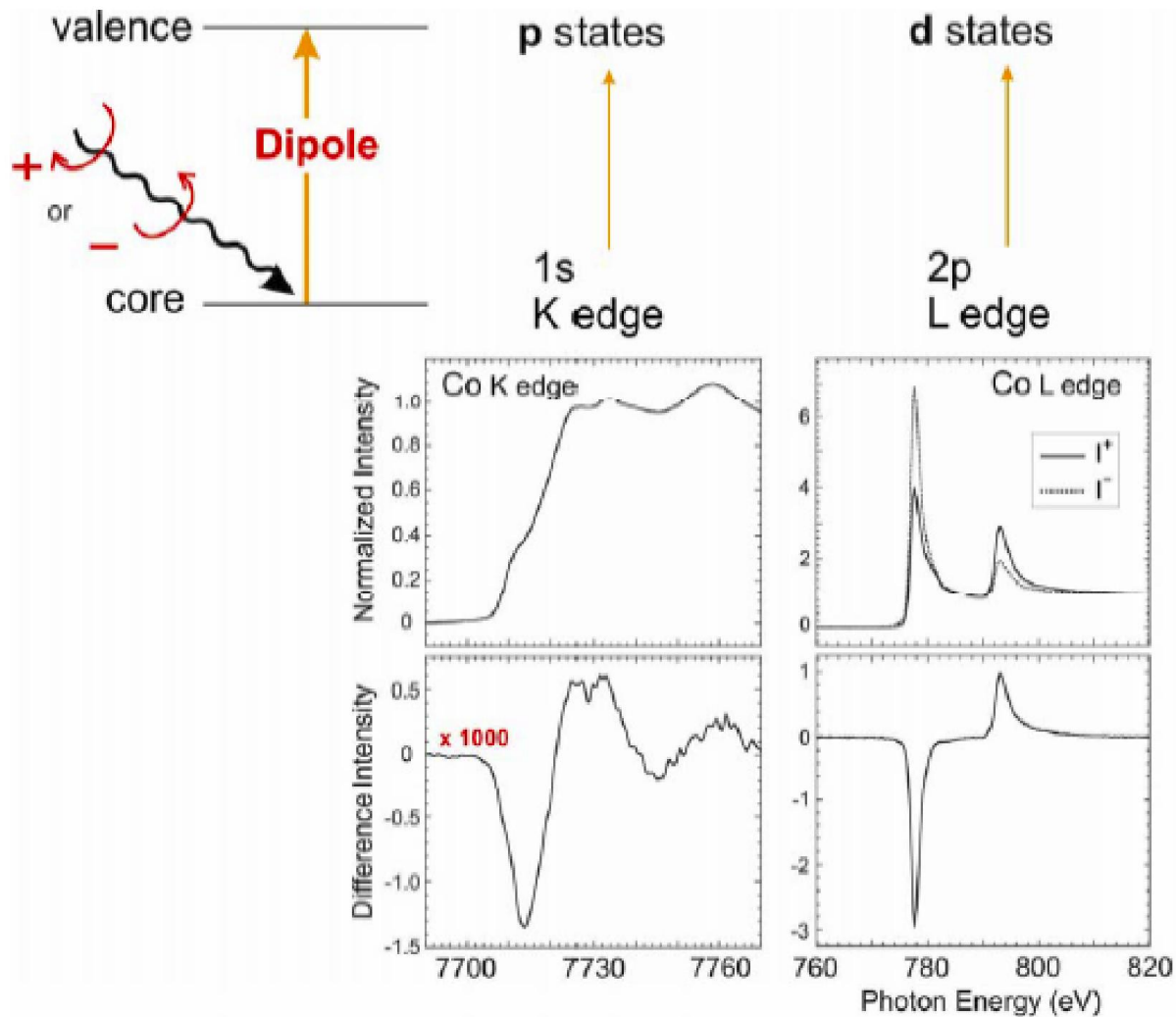


Nanomagnetism:

understanding basics
to application

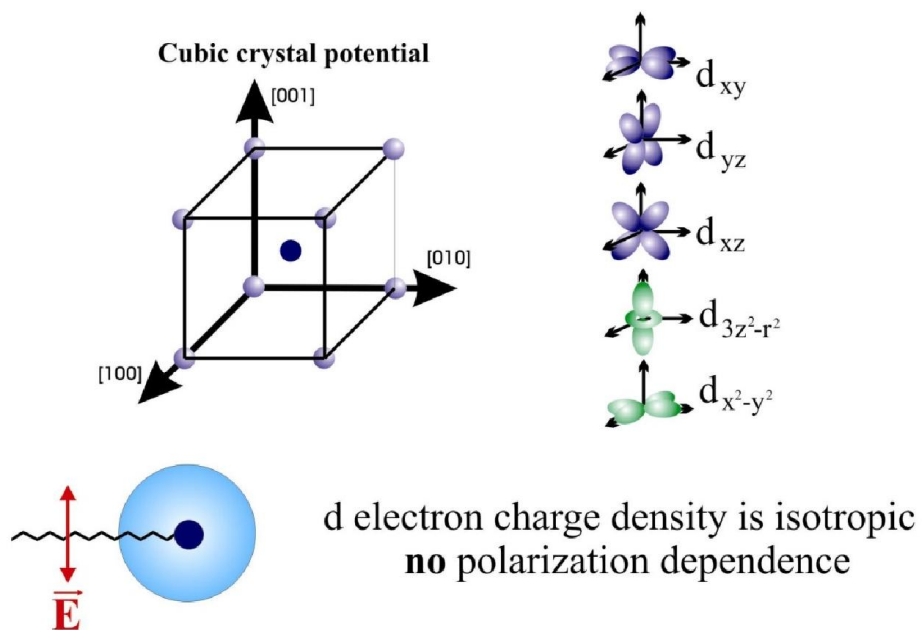


Soft x-rays are best for magnetism

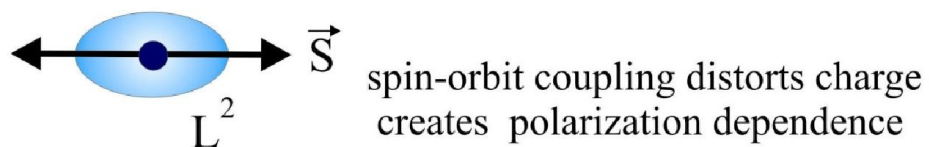


X-Ray **Magnetic Linear** Dichroism

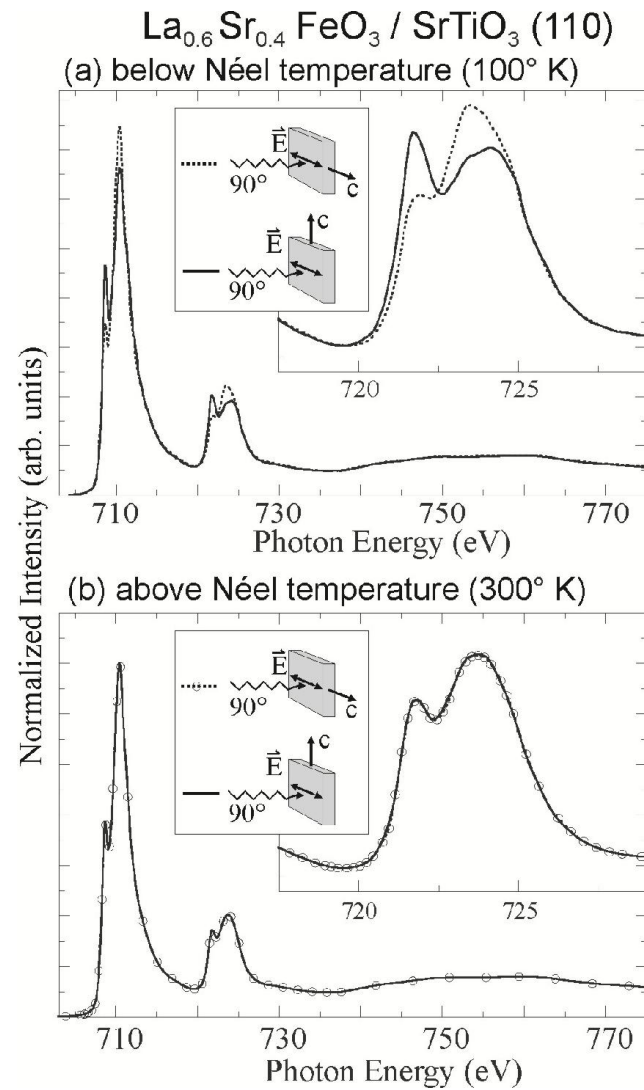
Non-magnetic state



Magnetic state - preferred spin axis

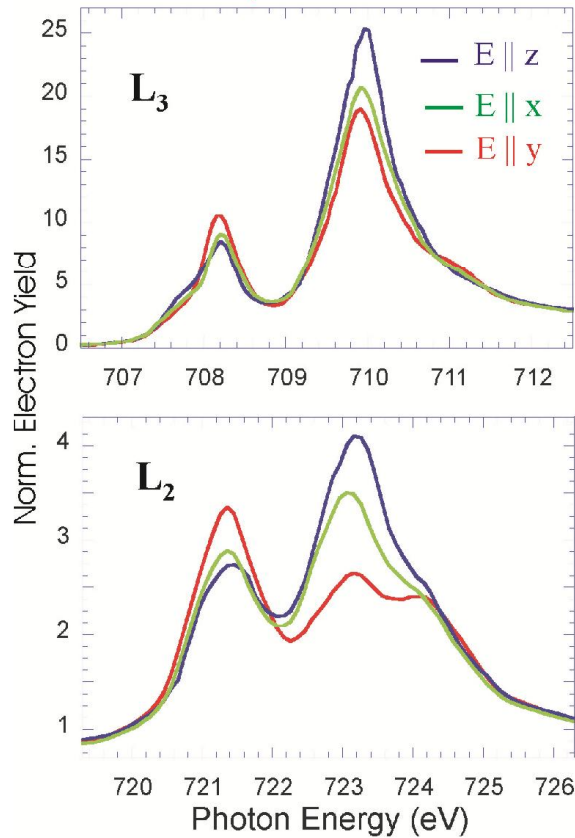
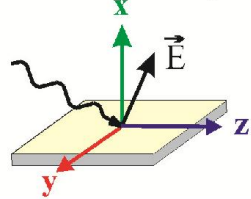


XMLD – spectra below and above T_N



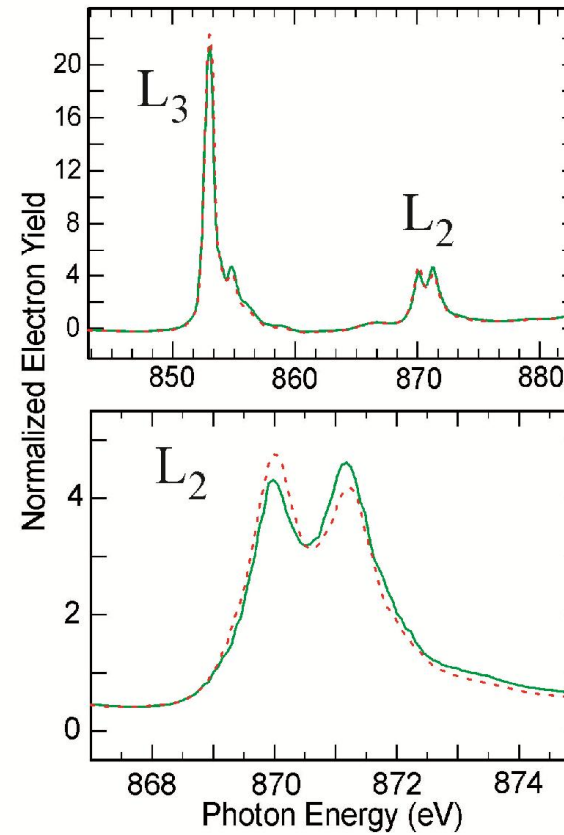
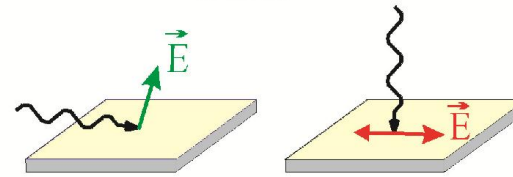
XMLD spectra of two oxides

LaFeO₃



Kuiper *et al.*, Phys. Rev. Lett. **70**, 1549 (1993)
 Lüning *et al.* Phys. Rev. B **67**, 214433 (2003)

NiO



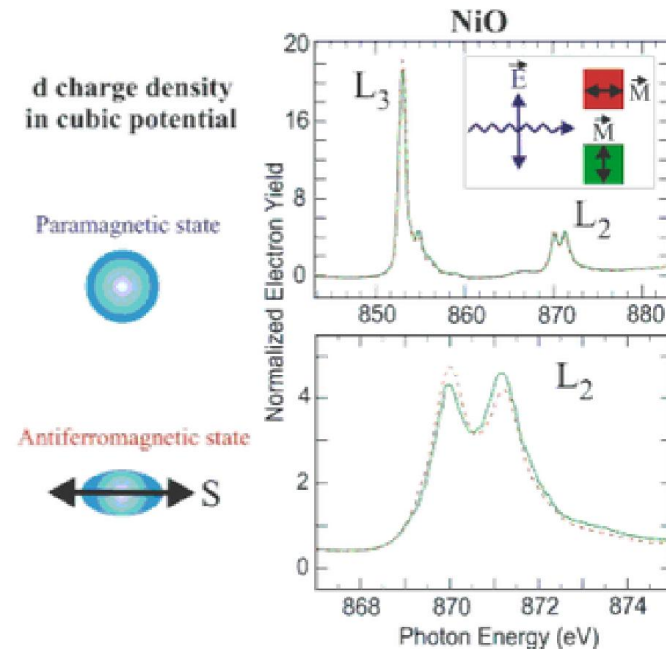
Alders *et al.*, Phys. Rev. B **57**, 11623 (1998)

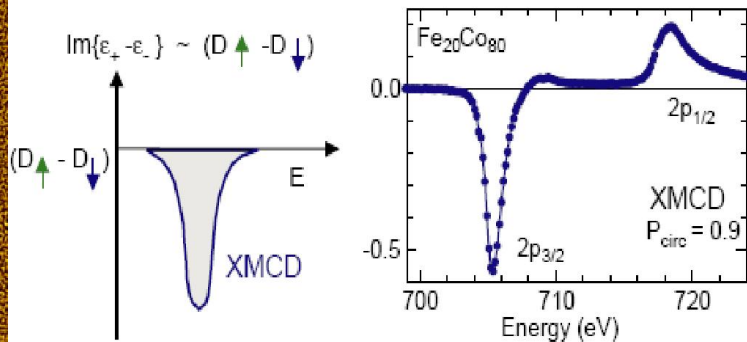
XMLD

- ❖ E of **linearly polarized x-rays** acts as a **search light** for the **number of valence holes** in different directions of the atomic volume.
- ❖ Charge in the atomic volume is caused by an **anisotropy in the bonding**, i.e. by the electrostatic potential. In the **absence of spin order**, linear dichroism can only determine **charge order** in systems where the absorbing atom has lower than cubic symmetry.
- ❖ In the presence of spin order the **spin-orbit coupling** leads to preferential charge order relative to the spin direction even in cubic systems.
- ❖ determination of the spin axis in ferromagnetic and especially AFM systems by means of XMLD spectroscopy.
- ❖ Since the electric field vector oscillates in time **along an axis** and the radiation may be absorbed at any time, linearly polarized x-rays are only **sensitive to axial not directional properties**. Hence one can determine the **orientation of the AFM** or FM axis, but the spin direction itself cannot be determined.

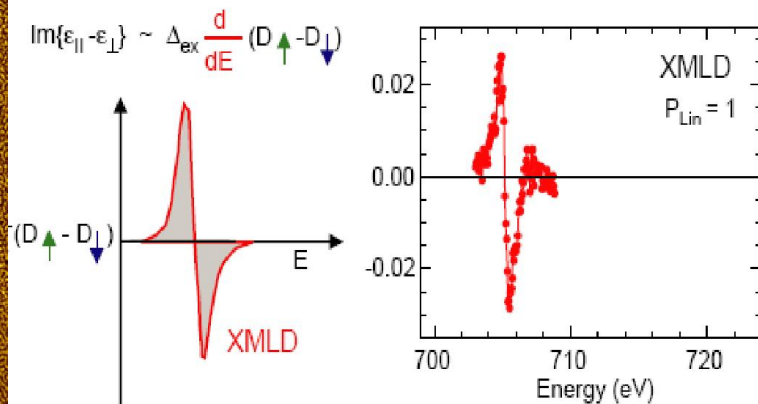
XMLD

- ❖ **E parallel** versus **E perpendicular** to the magnetic axis
- ❖ **$\cos^2\theta$ dependence**, where θ is the angle between E and the magnetic axis.
- ❖ **small in 3d metals** due to **small size of the spin orbit interaction** and the large band width, resulting in a small charge anisotropy when the d states are summed over the Brillouin zone.
- ❖ XMLD effect (10-30%) may be observed in the presence of **multiplet splitting**.
- ❖ At a particular multiplet energy only selected d valence states are probed through matrix element effects that enhance the XMLD effect.
- ❖ In a simple picture we can view each multiplet as a **strongly coupled spin-orbit** state whose **spatial extent is non-spherical**, giving rise to the large XMLD effect.
- ❖ Since XMLD can arise from electric and magnetic asymmetries, care needs to be taken to distinguish magnetic order effects from ligand field effects. This is typically done through temperature dependent measurements.





- ❖ The XMLD signal is much smaller than that of the widely used XMCD, therefore not many studies of the XMLD were carried out
- ❖ XMCD signal is odd in the magnetization M
- ❖ XMLD is even in M ; i.e., to lowest order quadratic in M



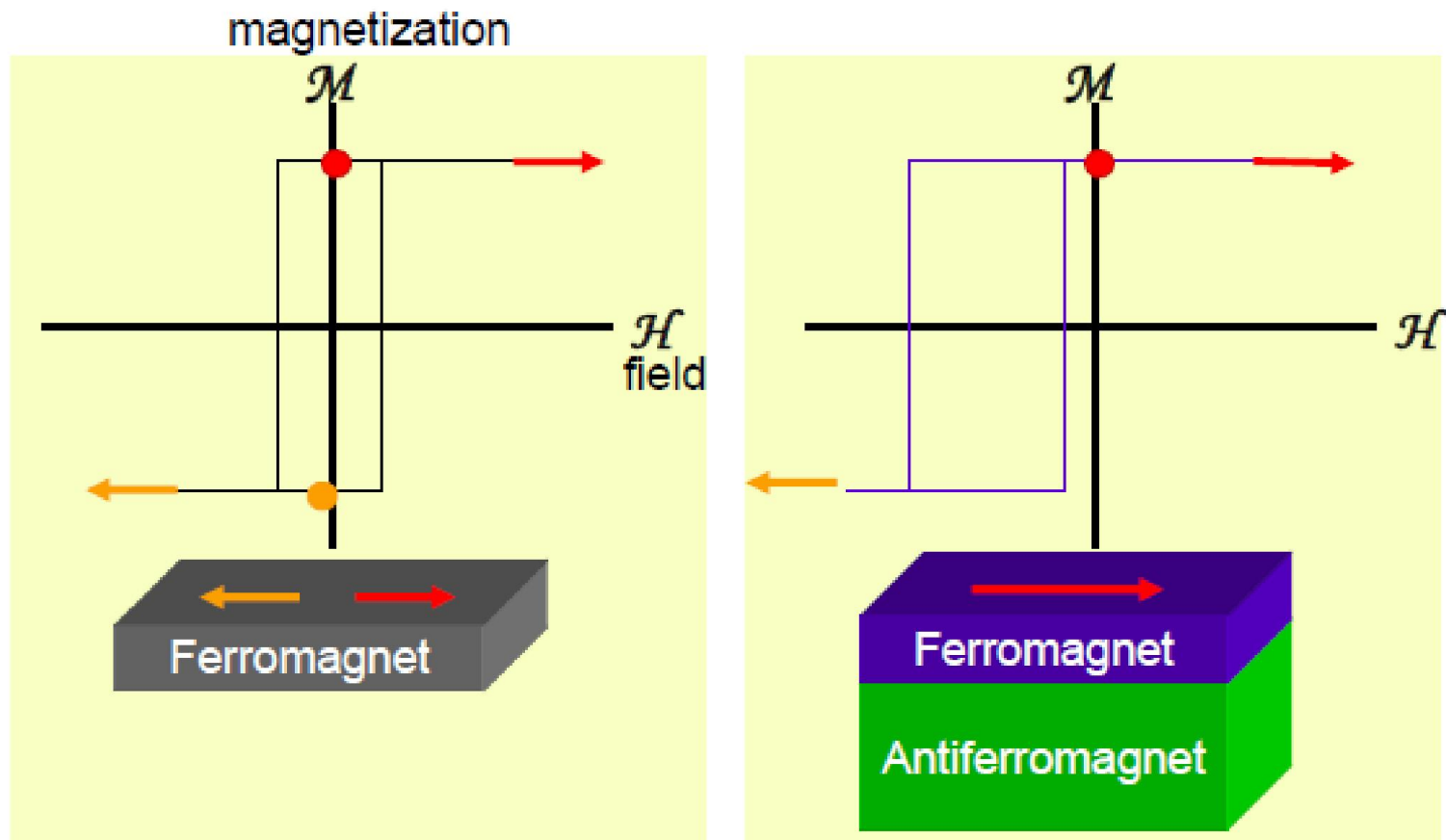
- ❖ The XMLD comes about as the intensity difference between resonant p-d transitions having $\Delta m = 0$ and the average of the transitions with $\Delta m = \pm 1$:
- ❖ The relatively small exchange splitting of the 2p core states is crucial: without this exchange splitting the XMLD signal vanishes

Fig. 1. Comparison of the predicted (left) and measured (right) types of XMCD and XMLD spectra. The experimental XMCD and XMLD spectra are measured at the Fe L₃-edge of an amorphous Fe₂₀Co₈₀ film (symbols).

- ❖ In a ferromagnetic transition metal the XMCD fraction of the absorption coefficient $\{(n_+ - n_-)/(n_+ + n_-)\}$ is larger than the XMLD fraction $\{(n_+ - n_-)/(n_+ + n_-)\}^2$.
 $n_{+(-)}$ is the hole occupation number of the spin up (down) bands.
 For example, in bulk Fe the peak XMCD at the 2p edge is 150% (normalized to the edge jump), while here the peak XMLD is only 8%.
- ❖ At the L_2 edge of Fe, both $2p_{1/2}$ multiplets contribute positively to the $\langle S_z \rangle$ component of the XMCD.
- ❖ These multiplets have equal and opposite contributions to the $\langle S_z \rangle$ component of the XMLD.
- ❖ To measure $\langle S_z \rangle$ with XMCD, one needs only to distinguish the L_3 and L_2 edges (separated by 13 eV in Fe). XMLD is more difficult, since it requires resolution of the individual multiplet states (separated by 1 eV).
- ❖ Each multiplet has an intrinsic broadening that is inversely proportional to the core-hole lifetime. In the solid state the atomic multiplets are further broadened into bands. Thus, if the core-hole lifetime is short or the hybridization strong, the $\langle S_z \rangle$ component of the XMLD spectrum is strongly suppressed. This is exactly the situation in transition metal systems, hence XMLD may not be measurable in a metal.
- ❖ Indeed, the two extant studies reporting nonzero TM XMLD have focused on metal oxides, where the multiplet peaks are narrow and well separated.

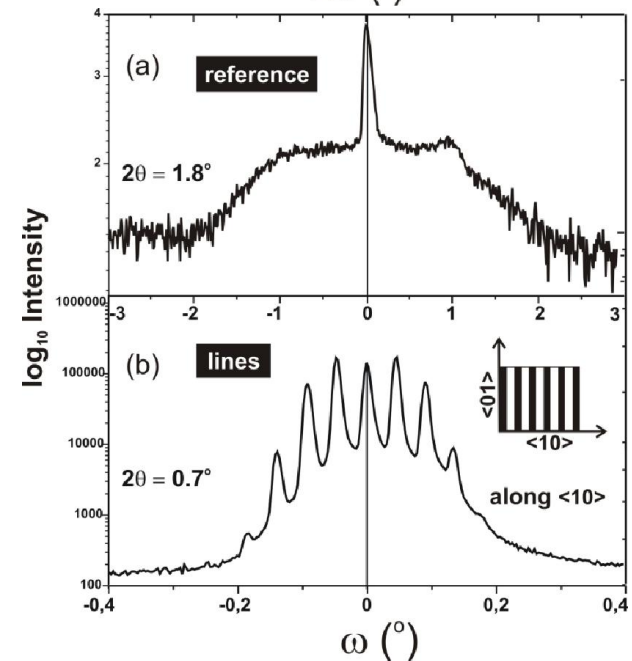
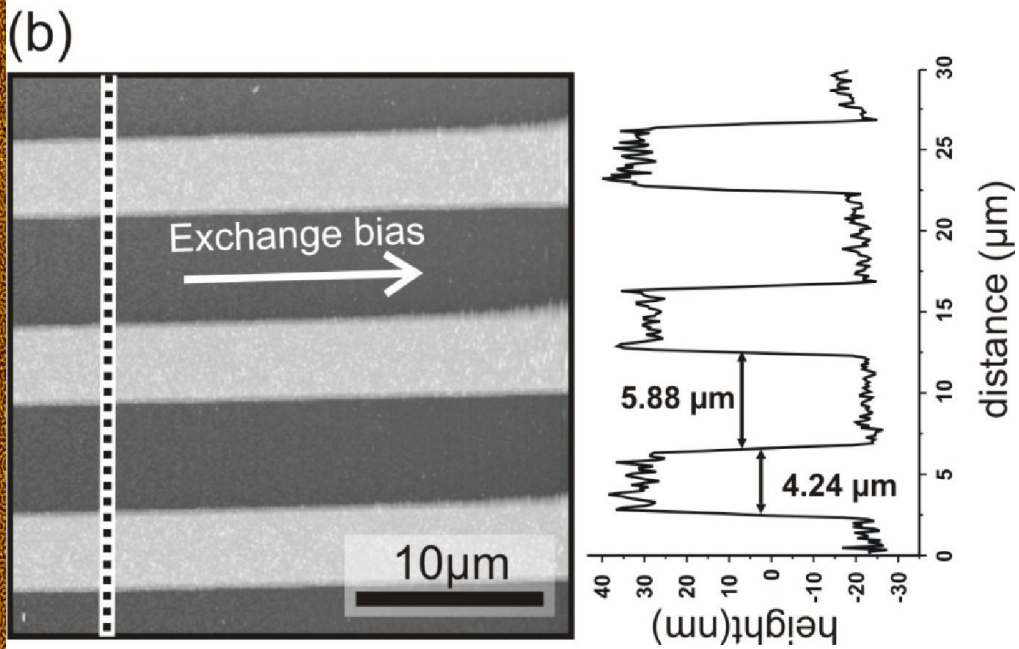
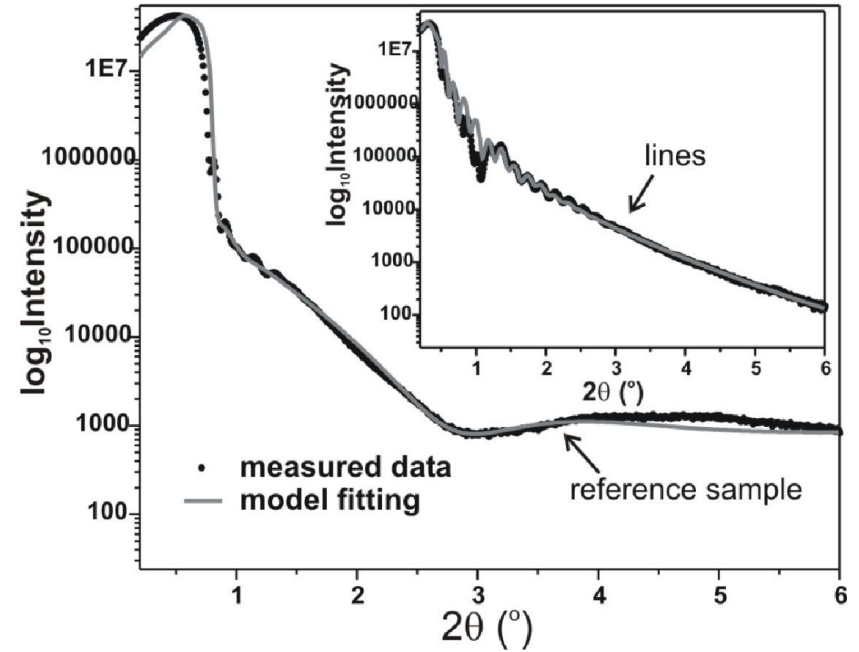
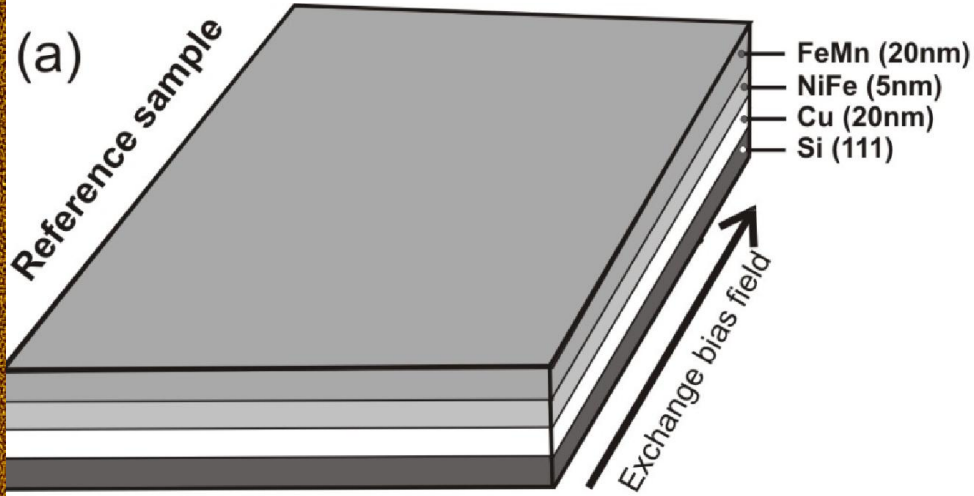
An important problem: Exchange Bias

discovered in 1956, technologically used for many years, ill understood

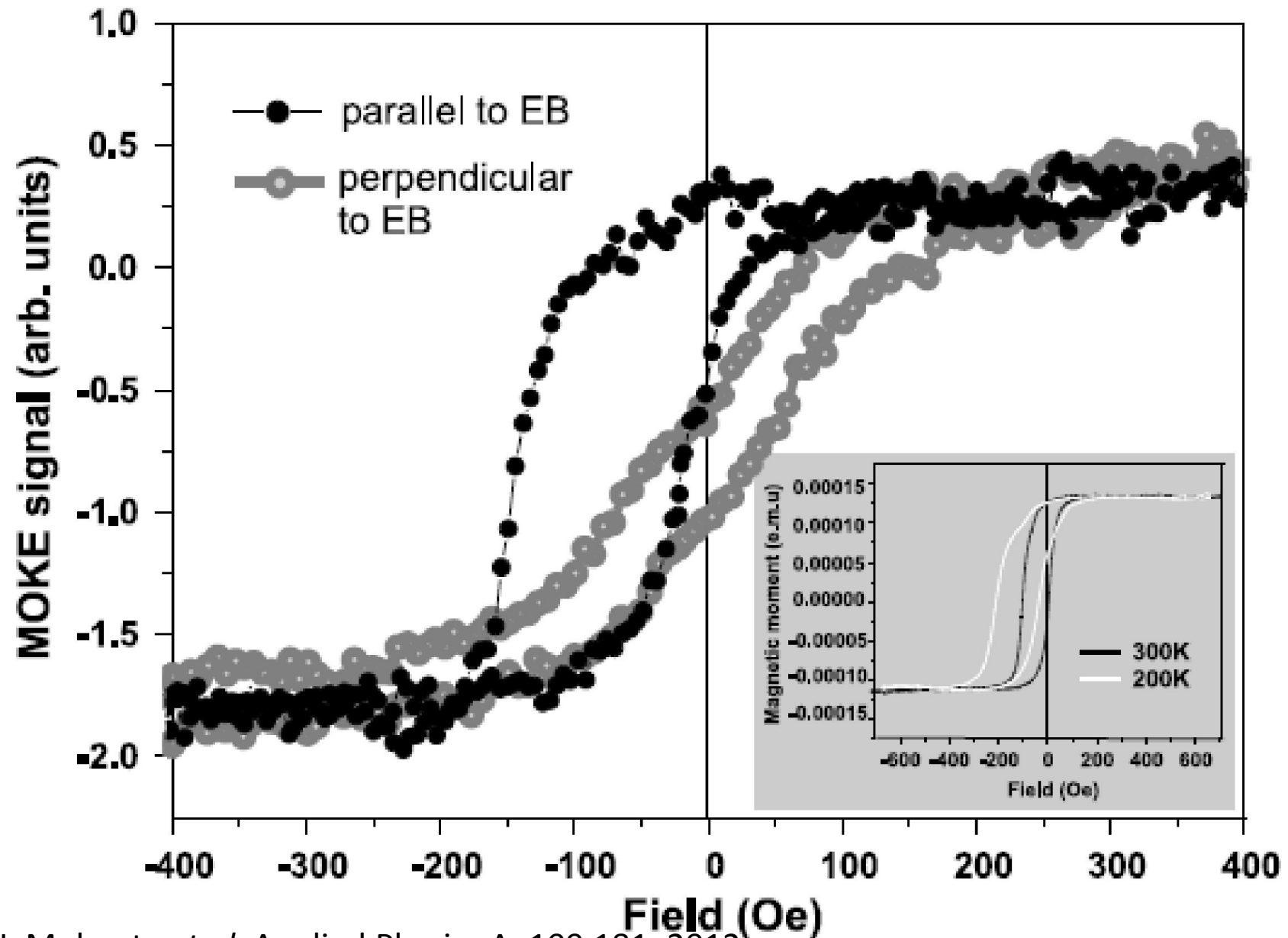


\mathcal{M} of blue layer is "pinned"
or "exchange biased"

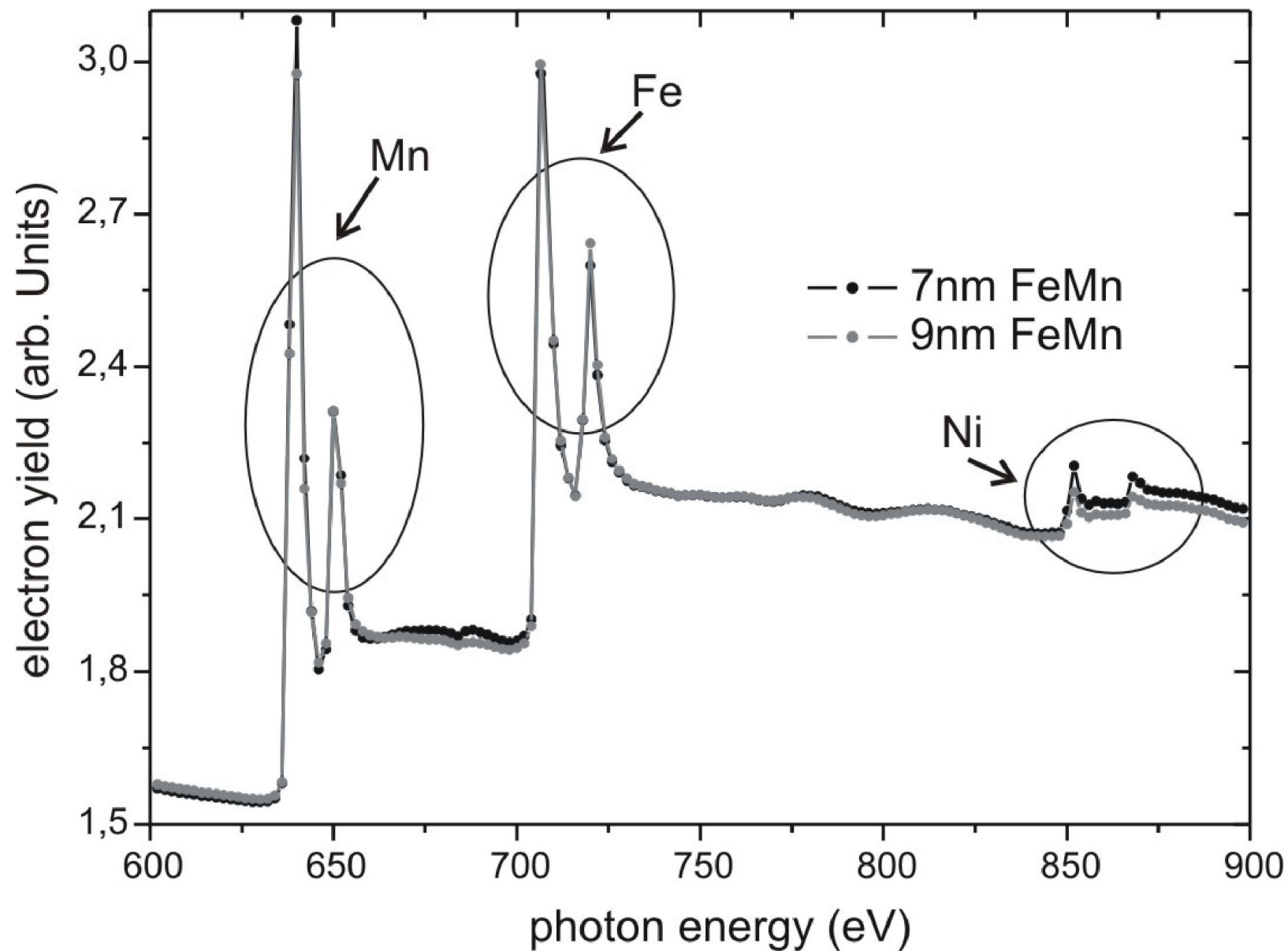
Structural Characterization



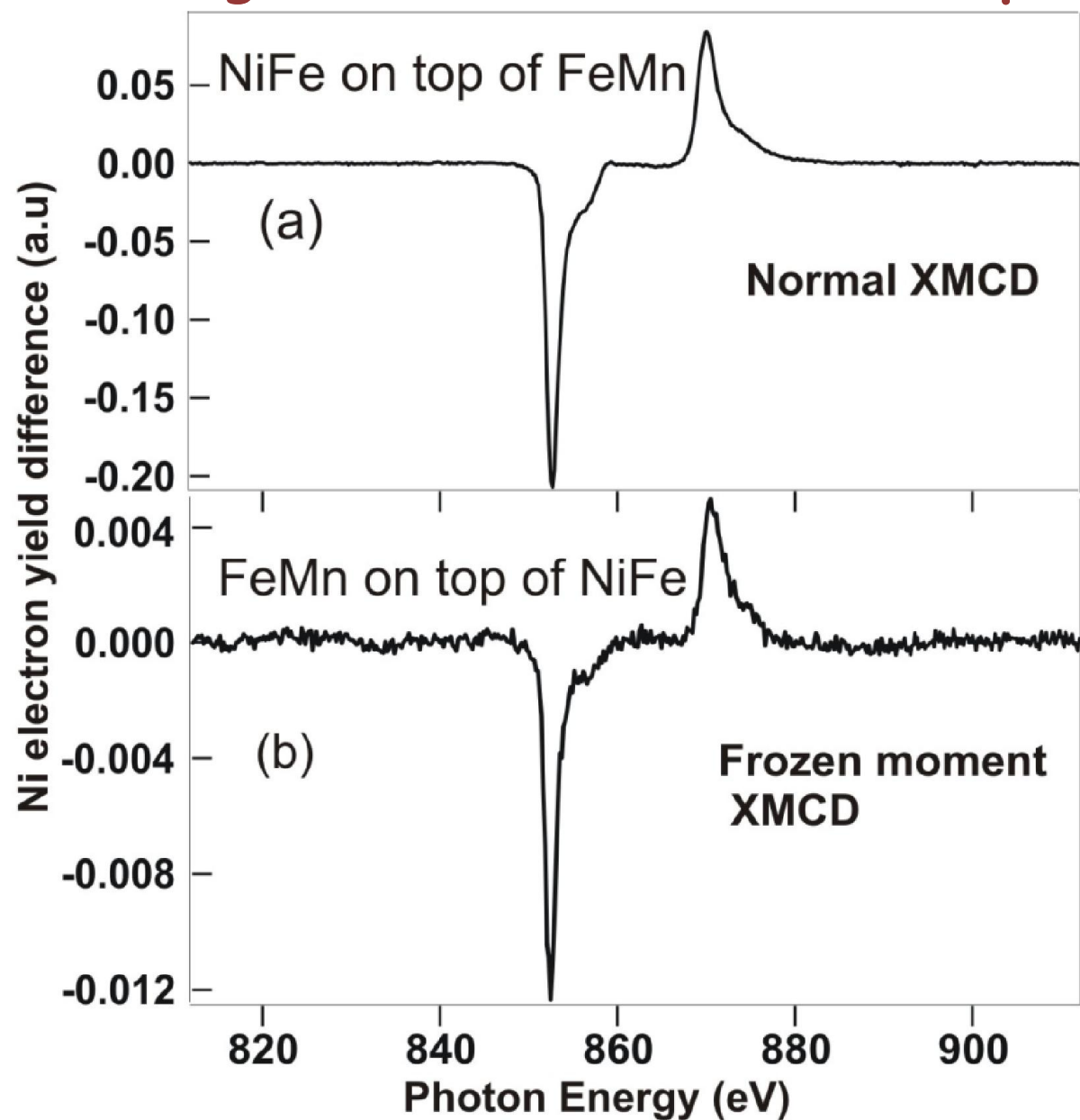
Magnetic Characterization



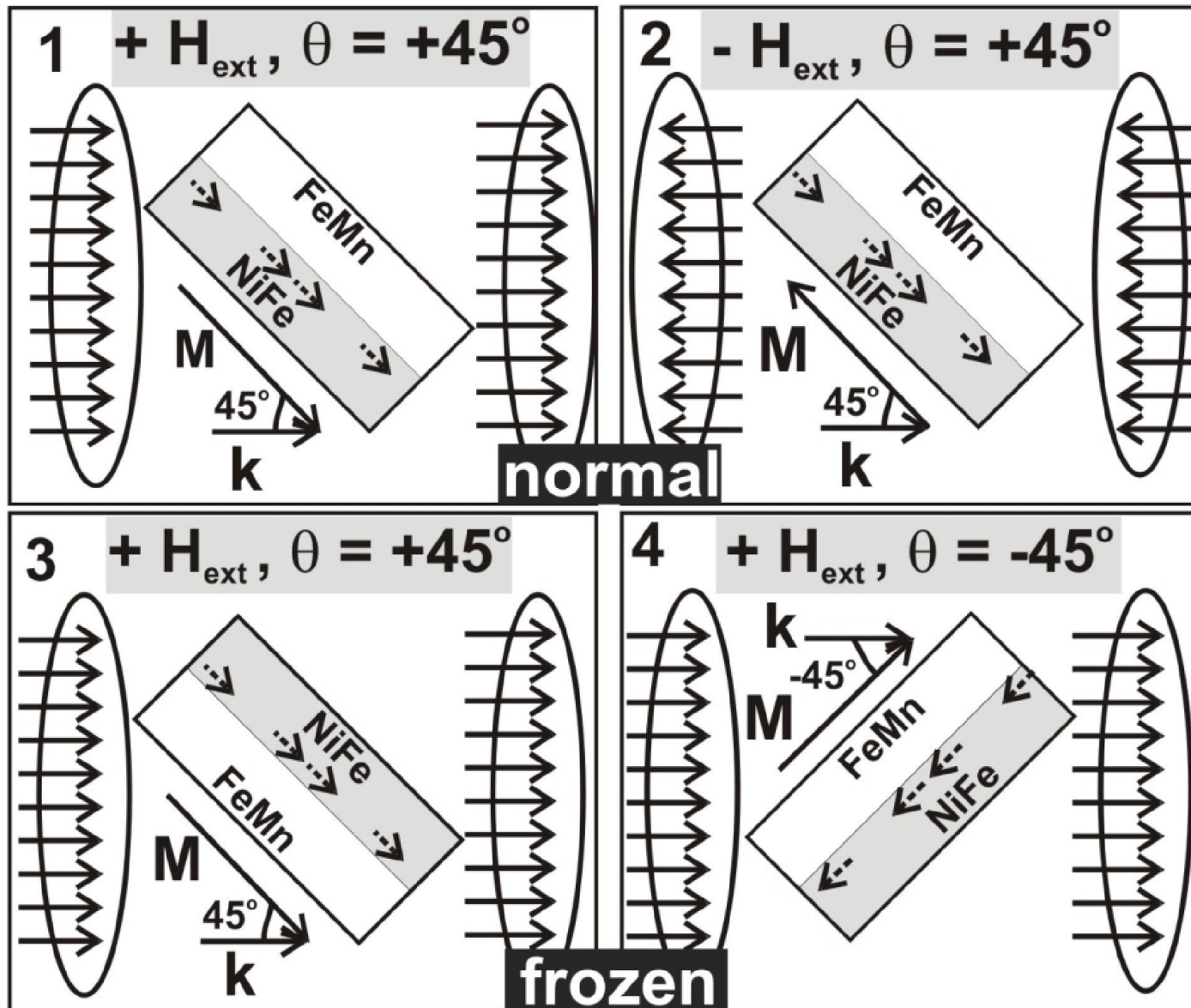
Element specific magnetic information



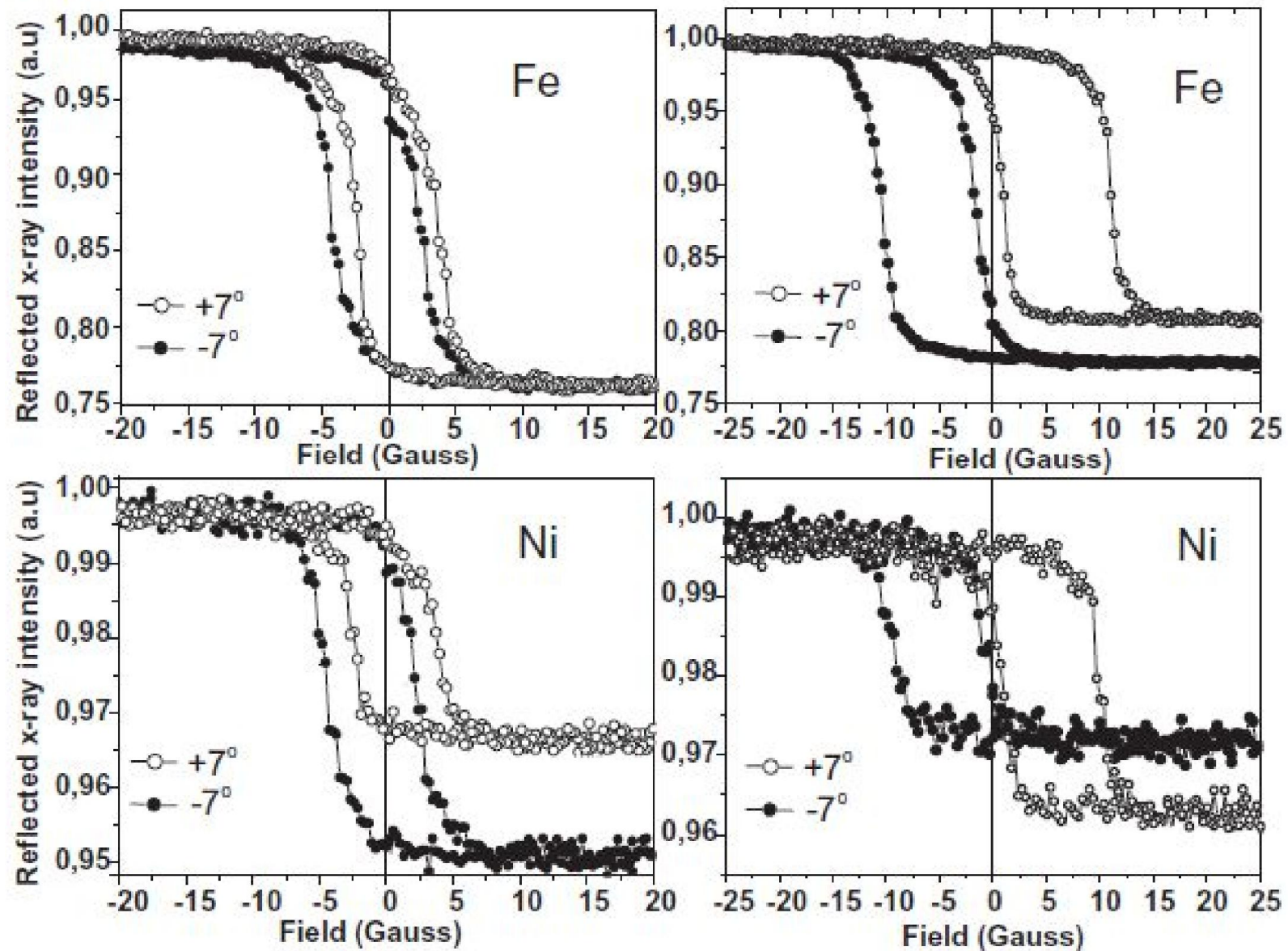
Looking at the frozen interface spins



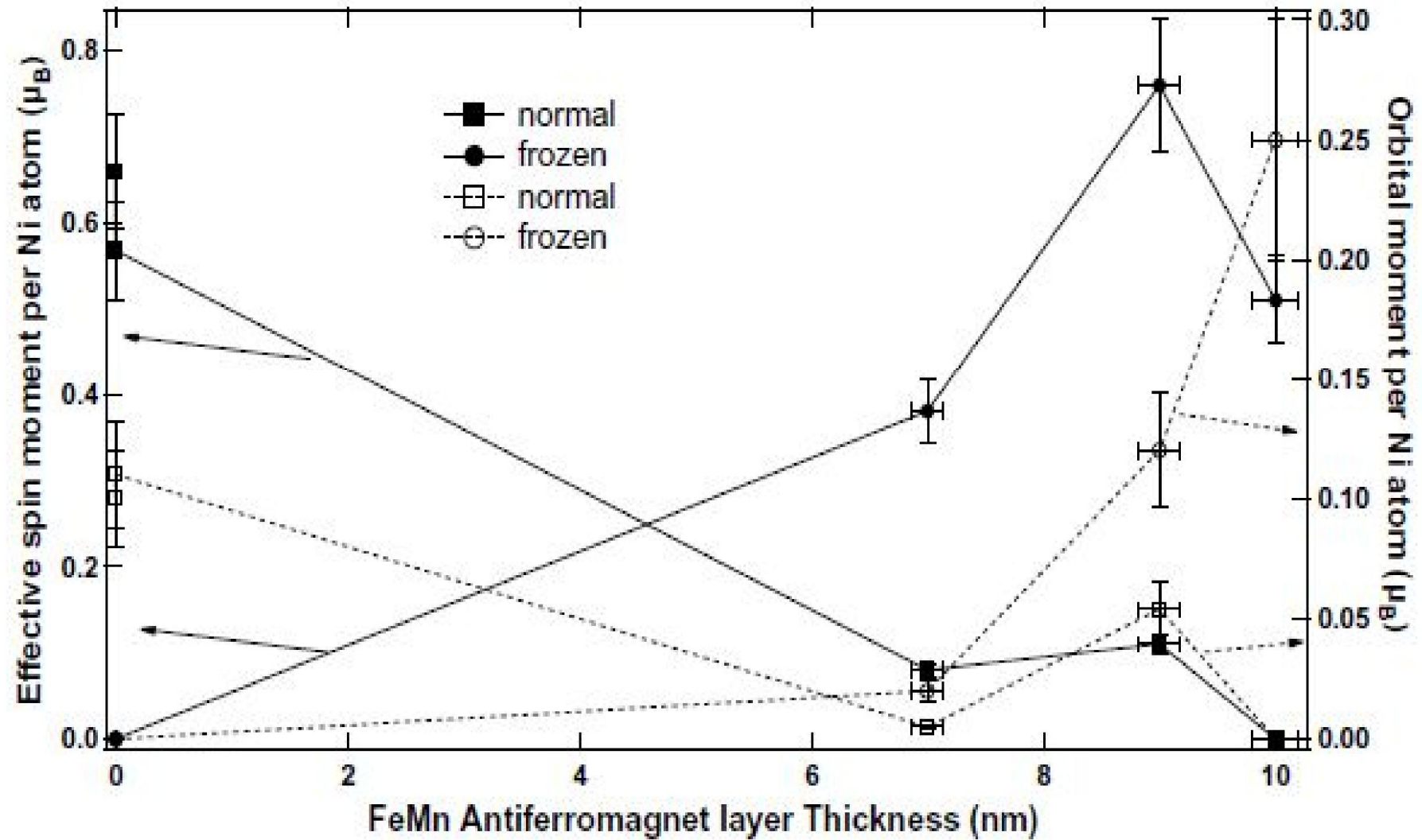
Looking at the frozen interface spins



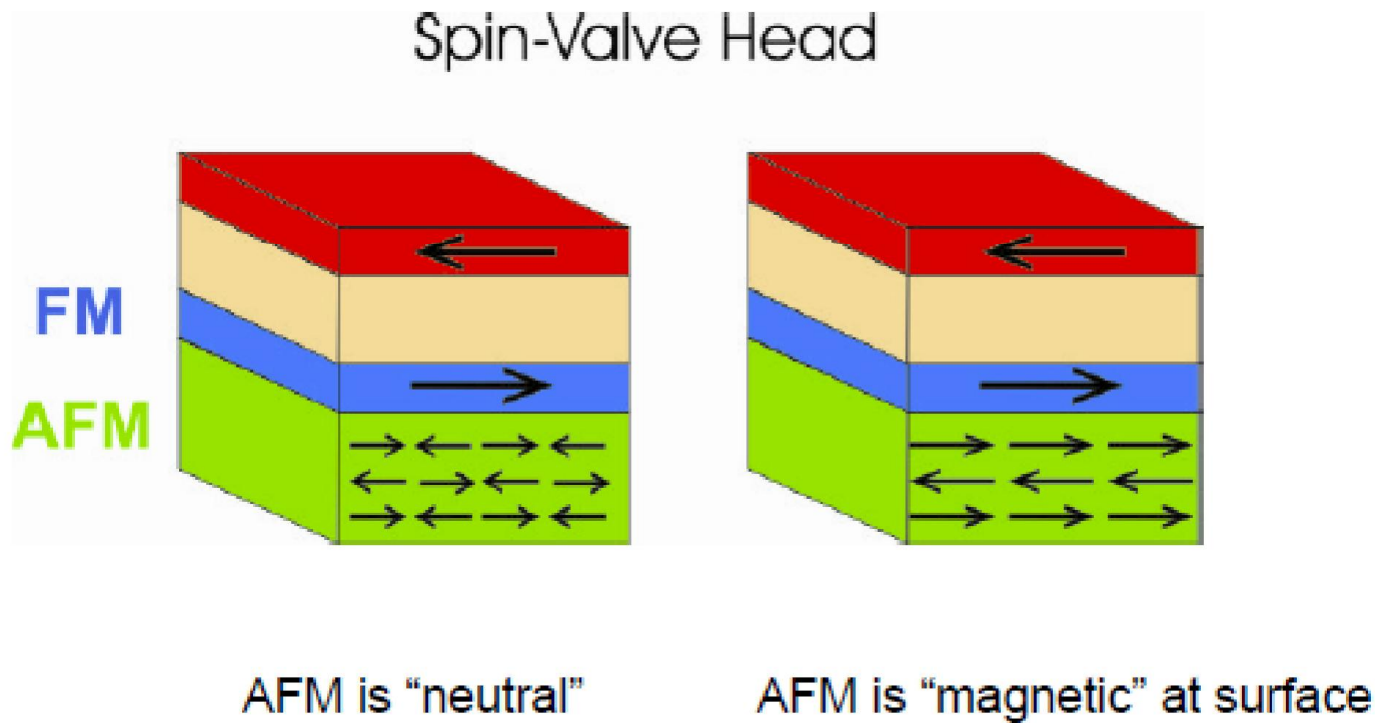
Where are the frozen spins?



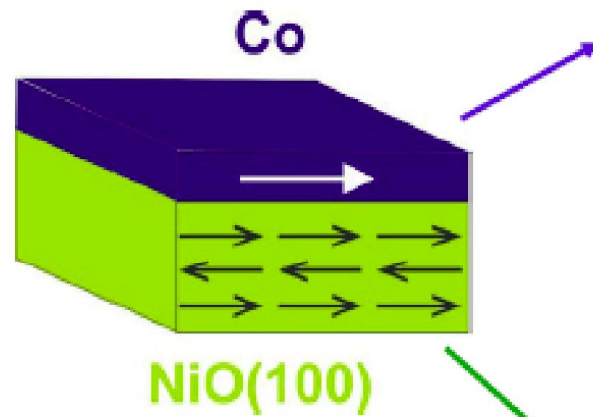
Our stand point: XMCD analysis



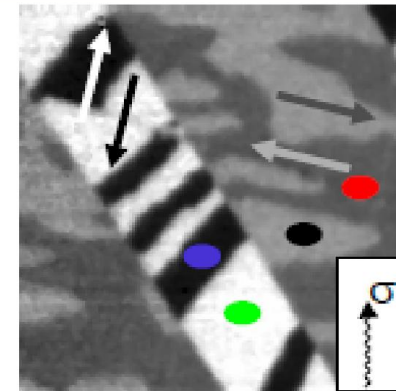
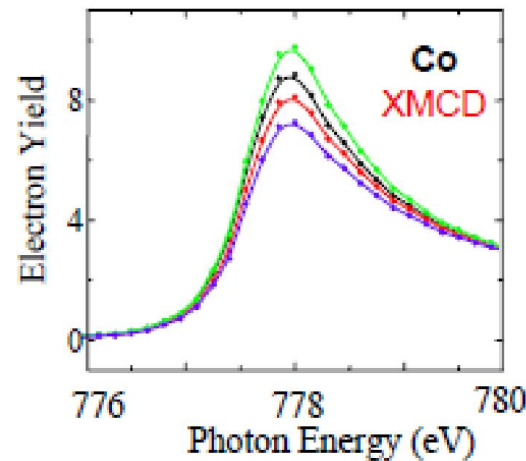
Example : The Spin Valve Sensor



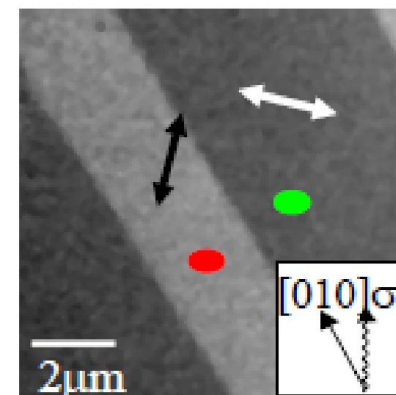
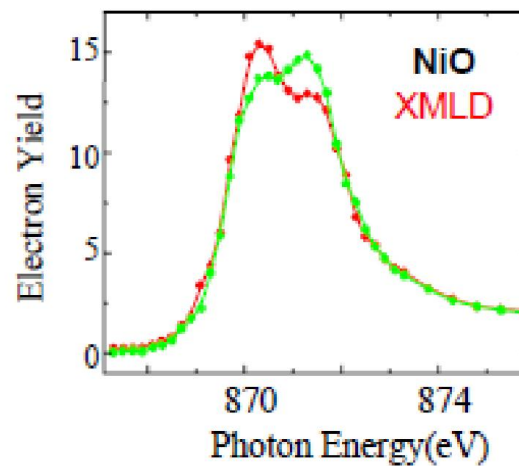
Coupling of ferromagnetic and antiferromagnetic domains



Co edge – use circular polarization – ferromagnetic domains

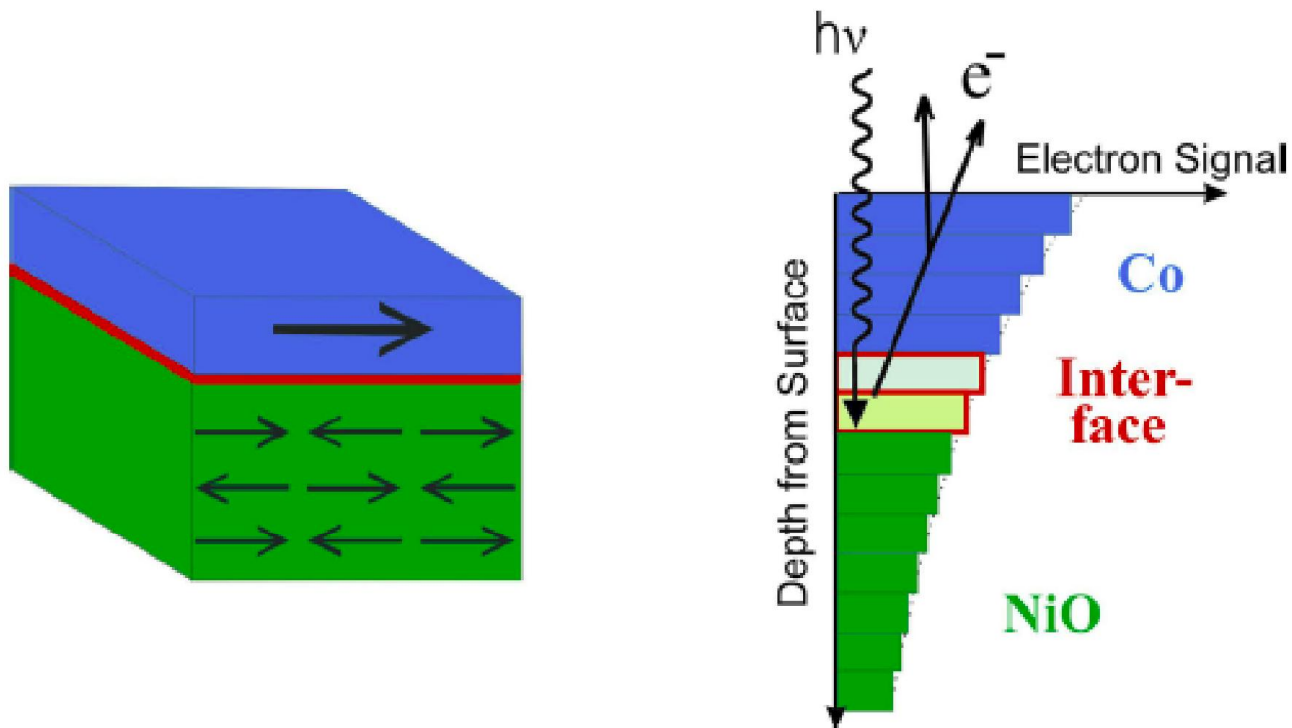


Ni edge – use linear polarization – antiferromagnetic domains



H. Ohldag *et al.*, PRL **86**, 2878 (2001)

X-Rays-in / Electrons-out - A way to study Interfaces

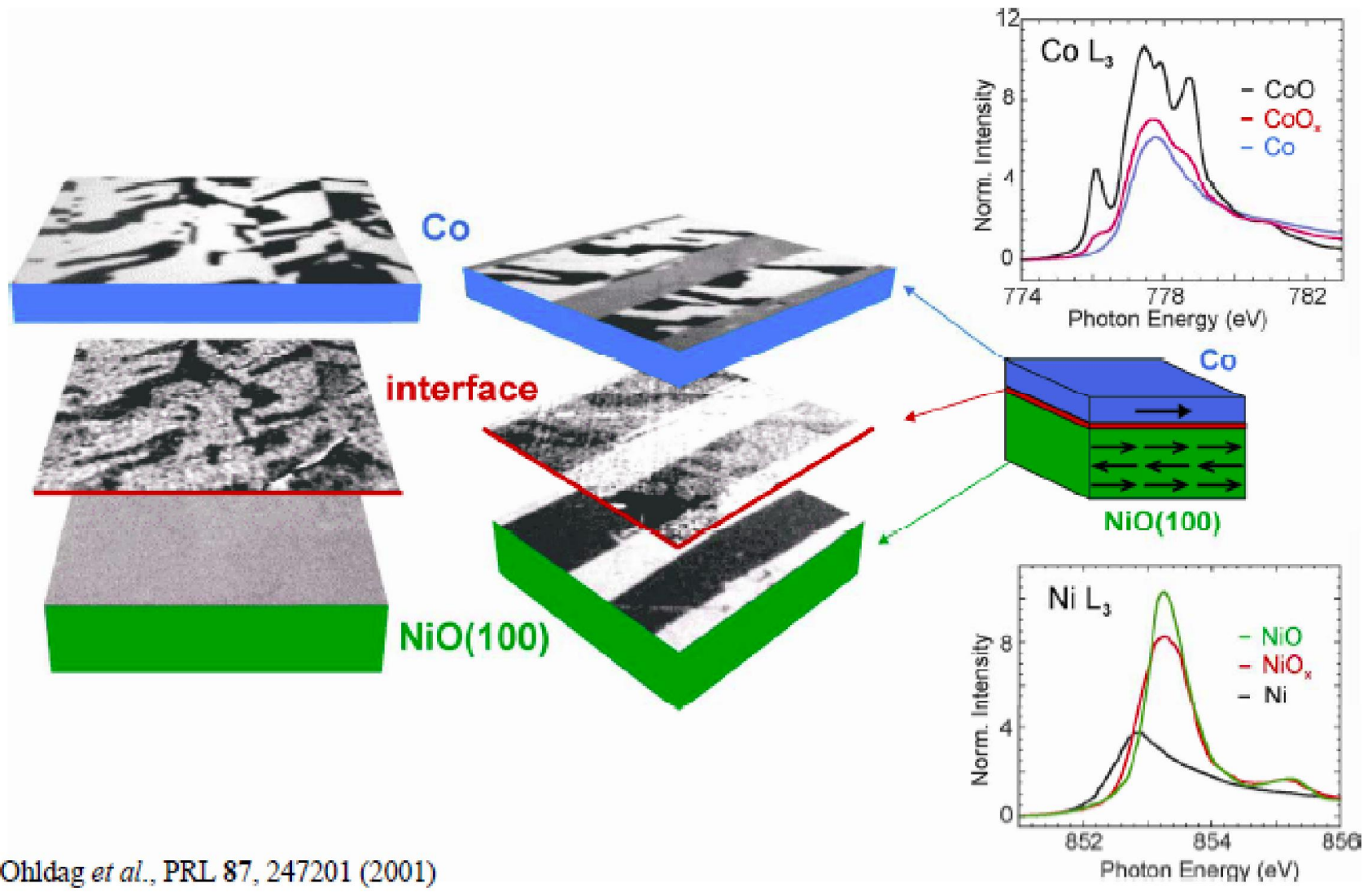


FM Co – tune to Co edge – circular polarization

AFM NiO – tune to Ni edge – linear polarization

FM Ni(O) – tune to Ni edge – circular polarization

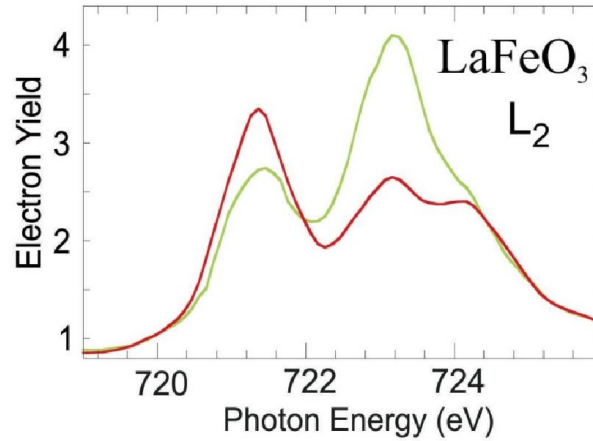
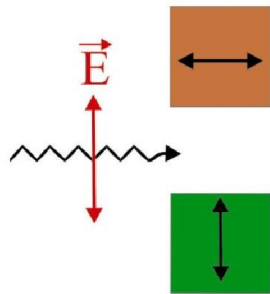
Images of the Ferromagnet-Antiferromagnet Interface



Ohldag *et al.*, PRL 87, 247201 (2001)

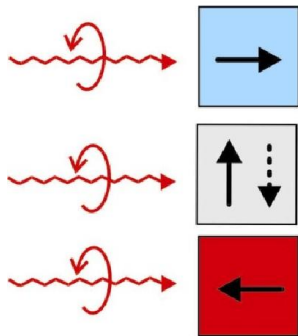
Magnetic Spectroscopy and Microscopy

X-ray Magnetic Linear Dichroism: **Antiferromagnets**

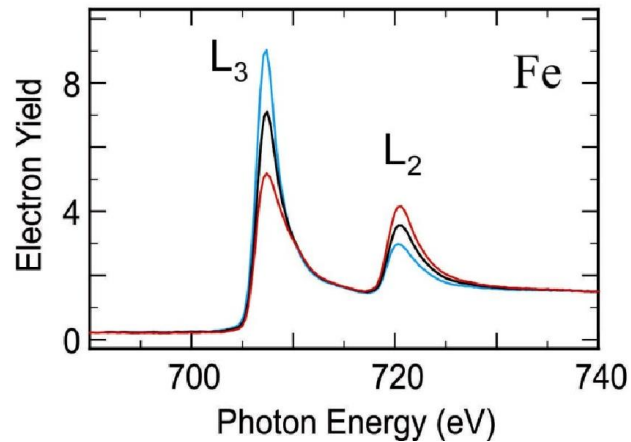


$$I \sim \cos^2 \theta$$

X-ray Magnetic Circular Dichroism: **Ferromagnets**



x-ray "spin"



$$I \sim \cos \theta$$

Soft X-Rays are best for magnetism!