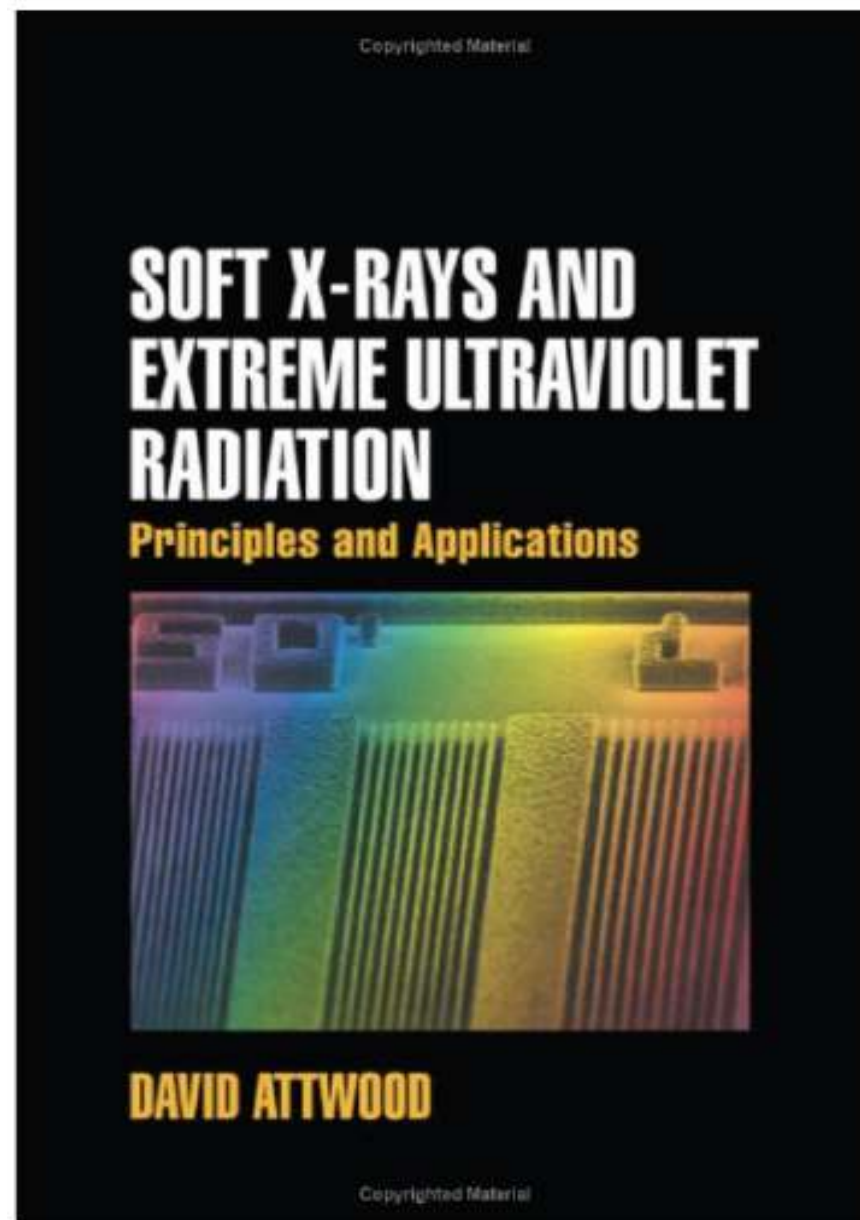


Jan Lüning

Sorbonne University, Paris (France) and Helmholtz-Zentrum Berlin (Germany)
jan.luning@helmholtz-berlin.de

Lecture topics:

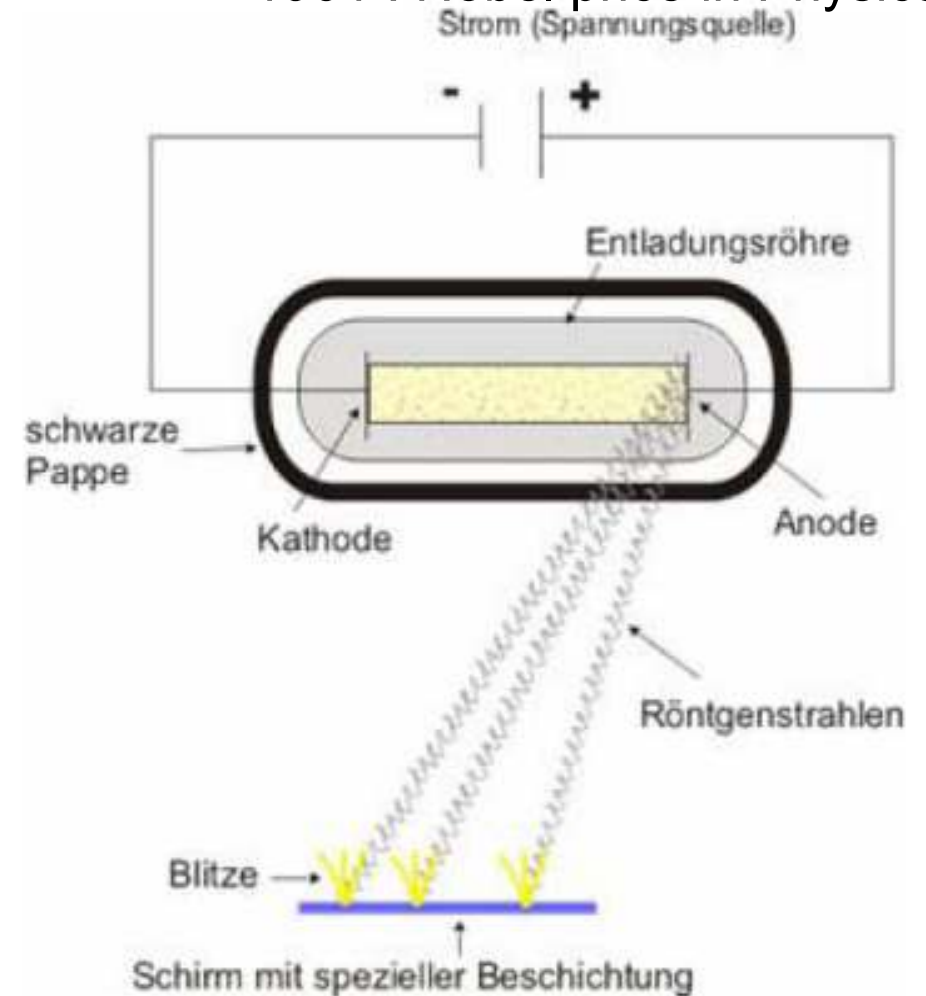
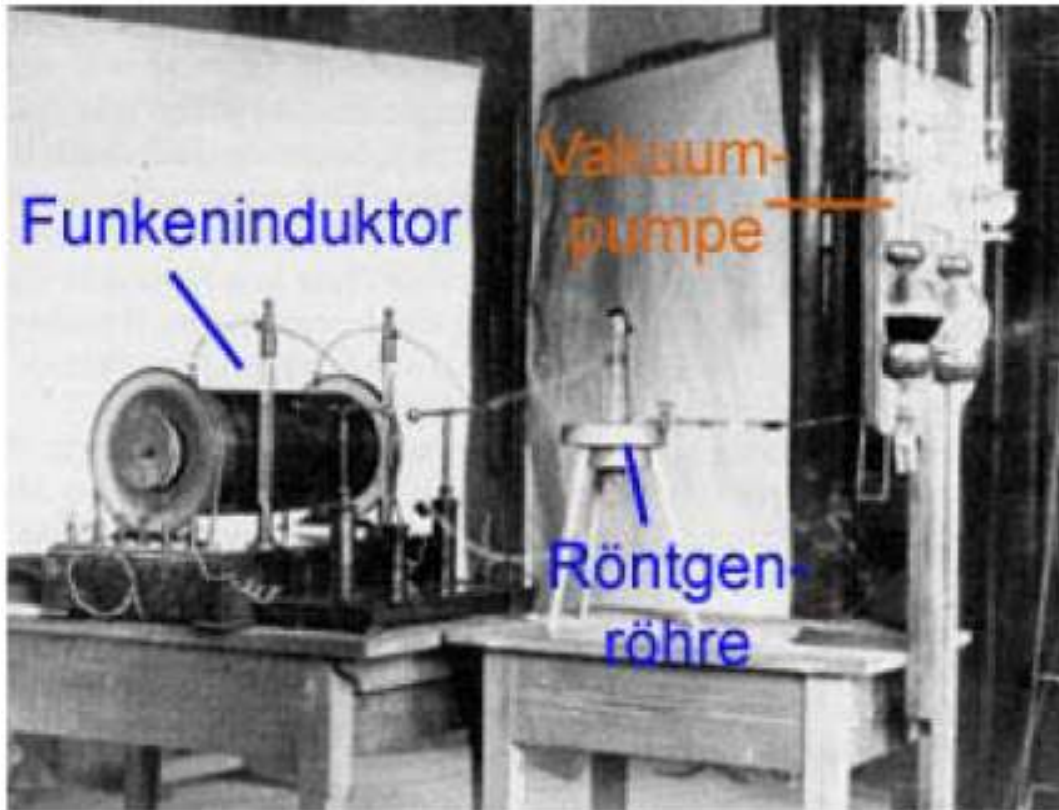
- 1) A brief introduction to X-rays
 - The basics of the interaction of X-rays with matter
 - Origin and properties of synchrotron radiation
- 2) X-ray based techniques
 - X-ray absorption spectroscopy
 - Types of magnetic dichroism
 - XMCD and Sum Rules
 - XMLD
 - Resonant magnetic scattering
- 3) X-ray microscopy
 - STXM
 - XPEEM
 - Lensless microscopy

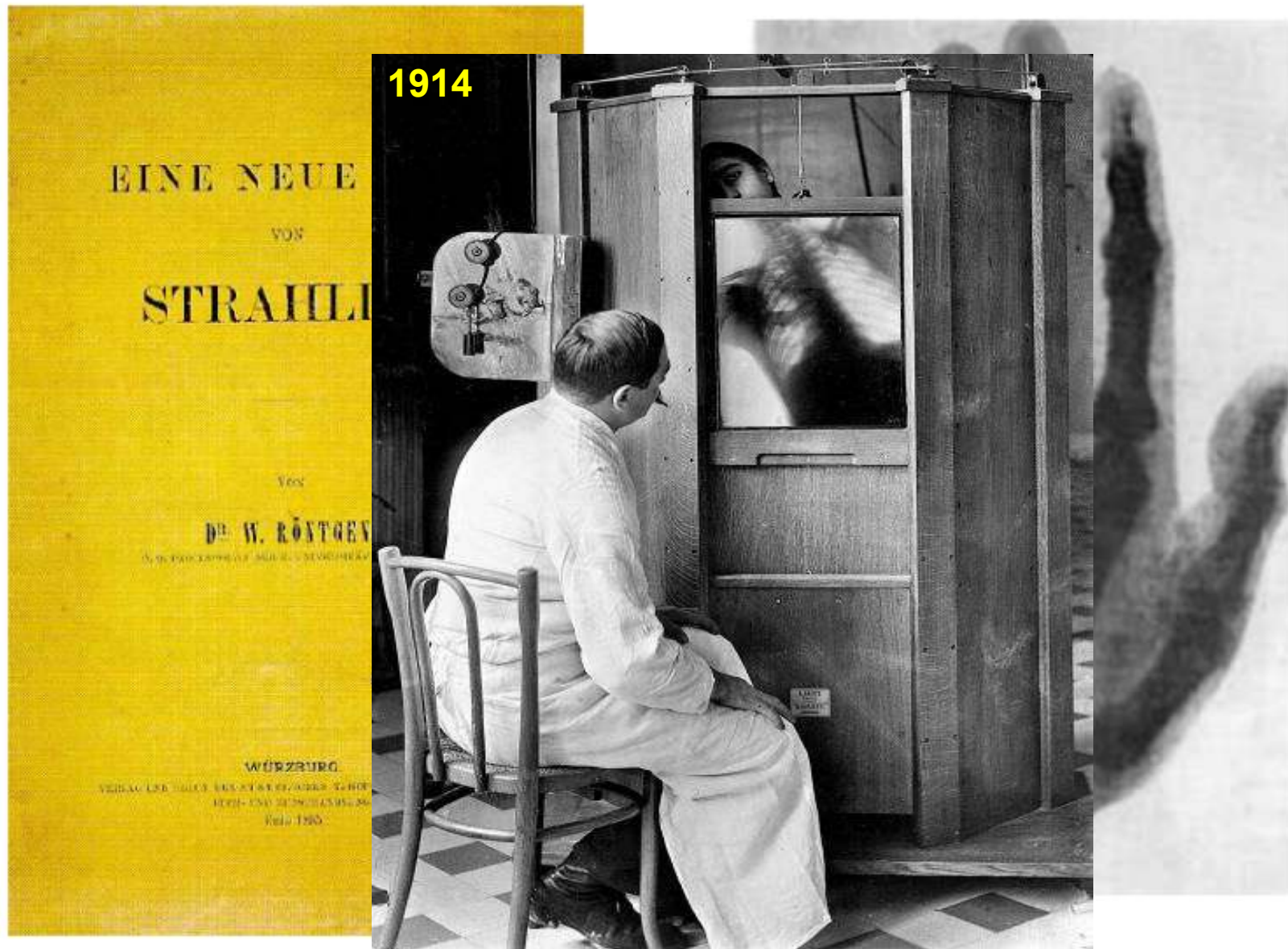


Discovery of X-rays

8 November 1895
(Würzburg, Germany)

1901 : Nobel price in Physics

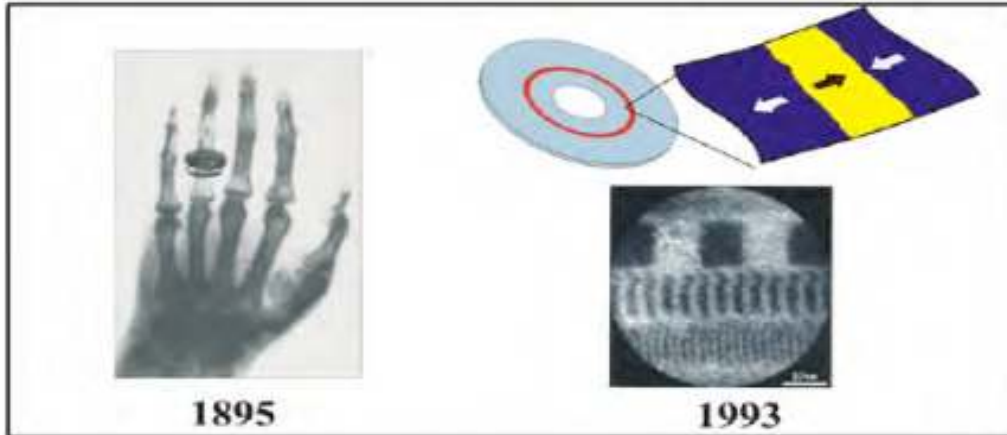




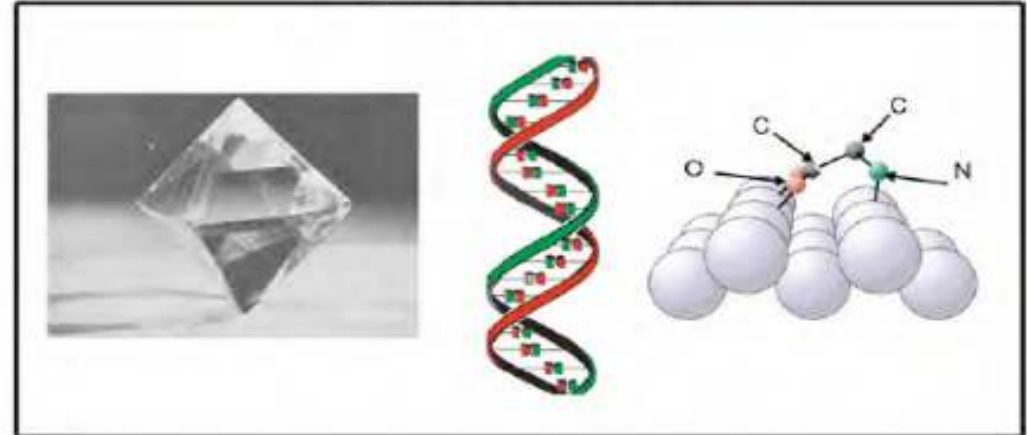
A chest X-ray in progress at Professor Menard's radiology department at the Cochin hospital, Paris, 1914. (Jacques Boyer/Roger Viollet—Getty Images)

Why are X-rays so useful

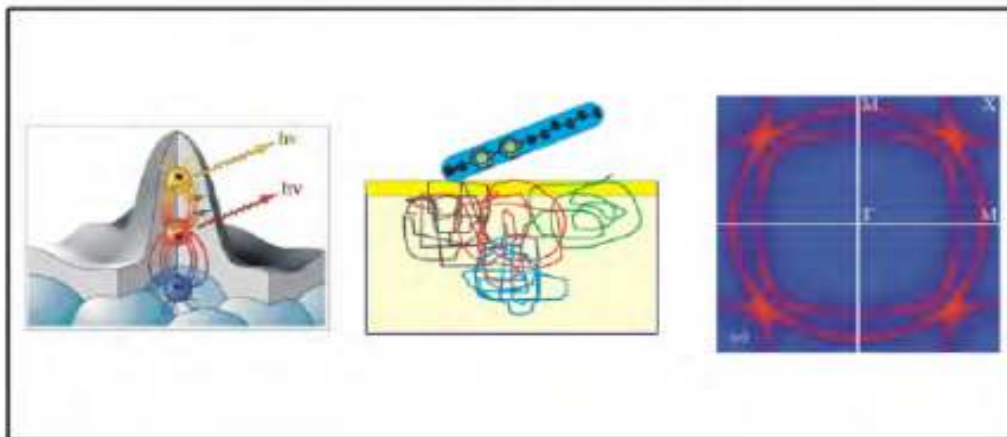
Imaging - Seeing the Invisible



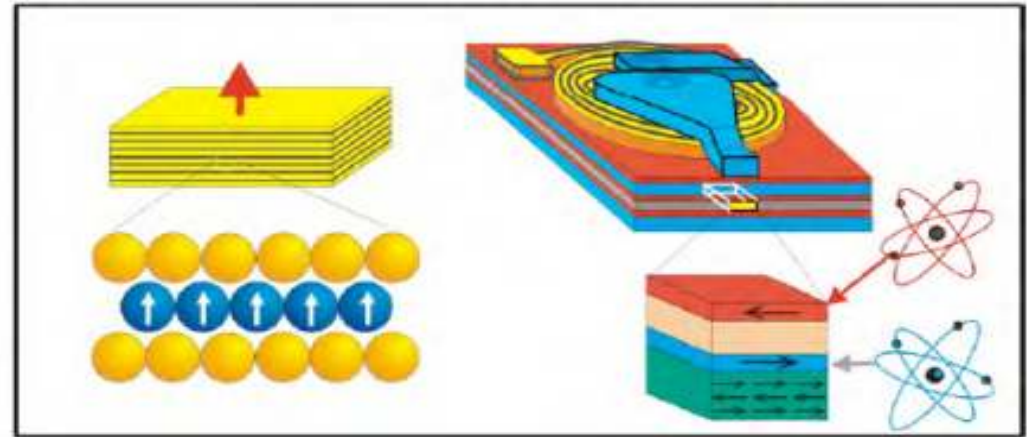
Atomic and Molecular Structure - where are the **atoms** -



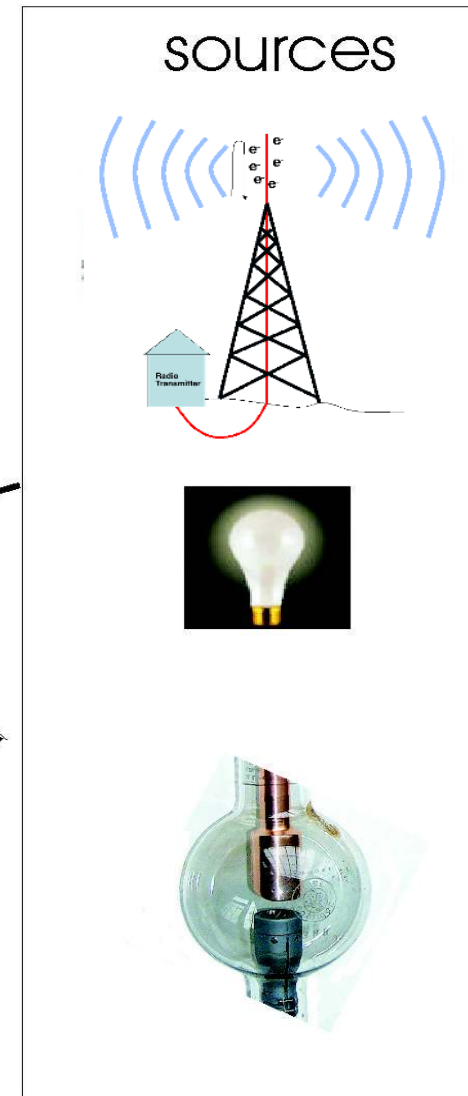
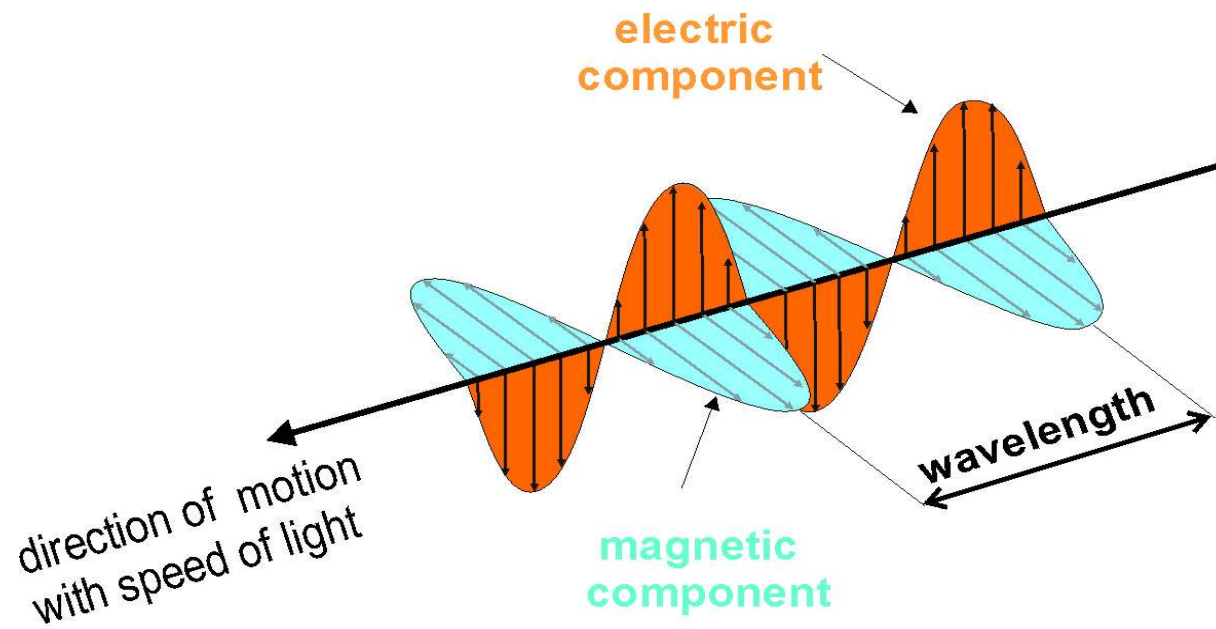
Electronic Structure and Bonding - where are the **electrons** -



Magnetic Structure and Properties - where are the **spins** -



Only difference: the wavelength!



TV

Radio

Radar

Microwaves

Light

X-Rays

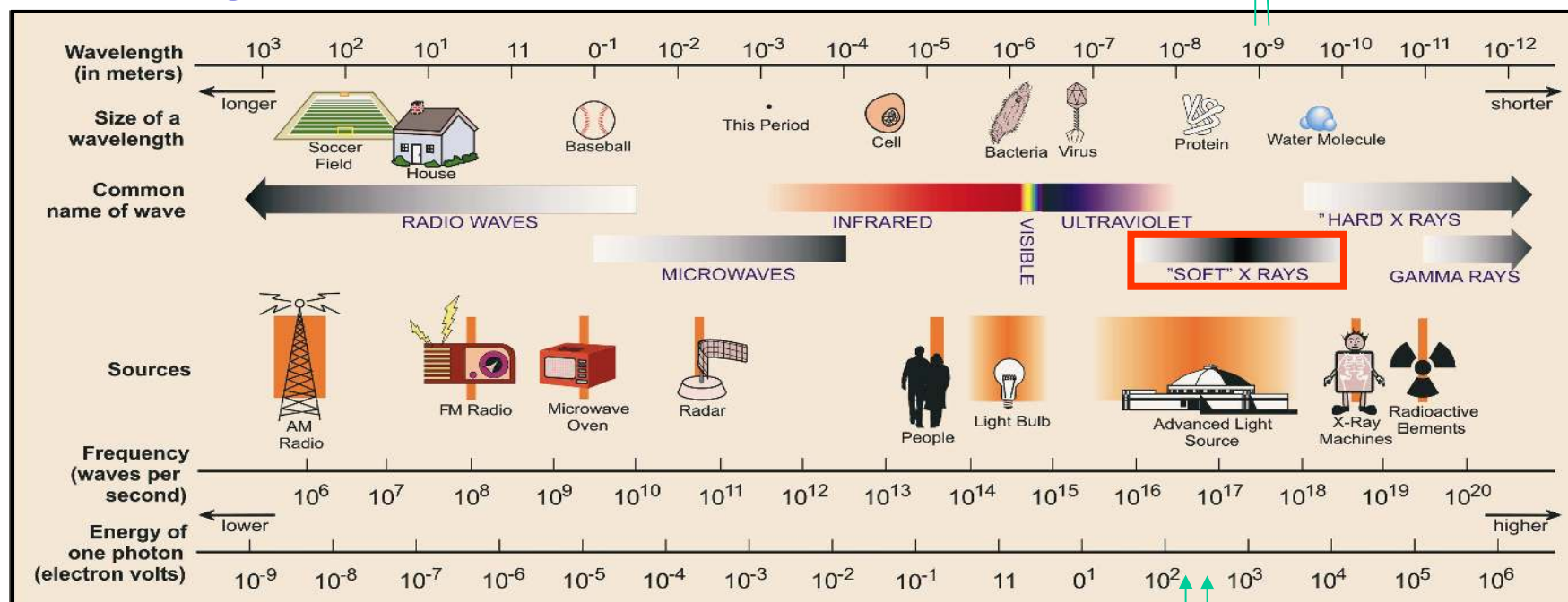
Gamma-Rays

Soft & Hard X-rays as part of the electromagnetic spectrum

Three 'common' definitions:

Soft: Grating UHV electronic structure
Hard: Crystal 'Air' crystal structure

Wavelength

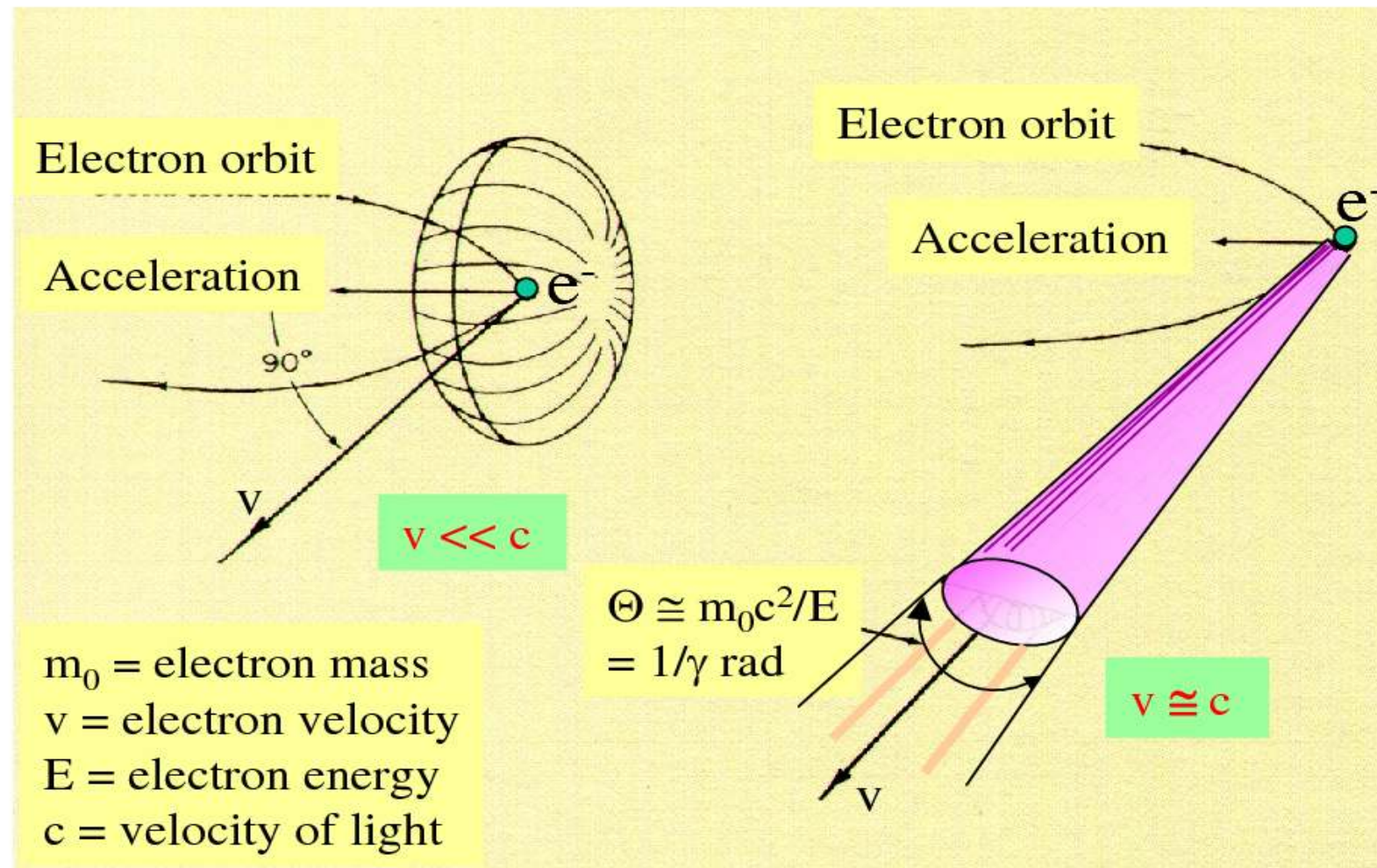


Energy

C, N, O
K-edges

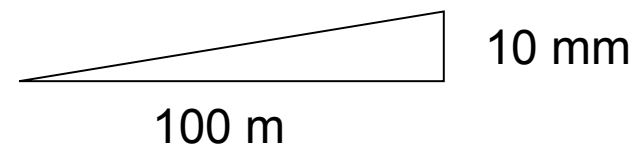
3d TM: strong magnetic
L-edge resonances

Origin of synchrotron radiation

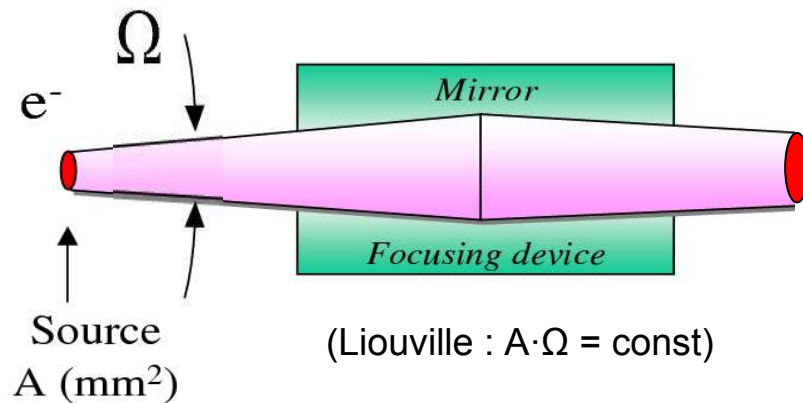


Emission angle $\Theta = 1/\gamma \text{ rad} \sim 100 \mu\text{rad}$

Small angle: $\tan (\Theta [\text{rad}]) = \Theta [\text{rad}]$



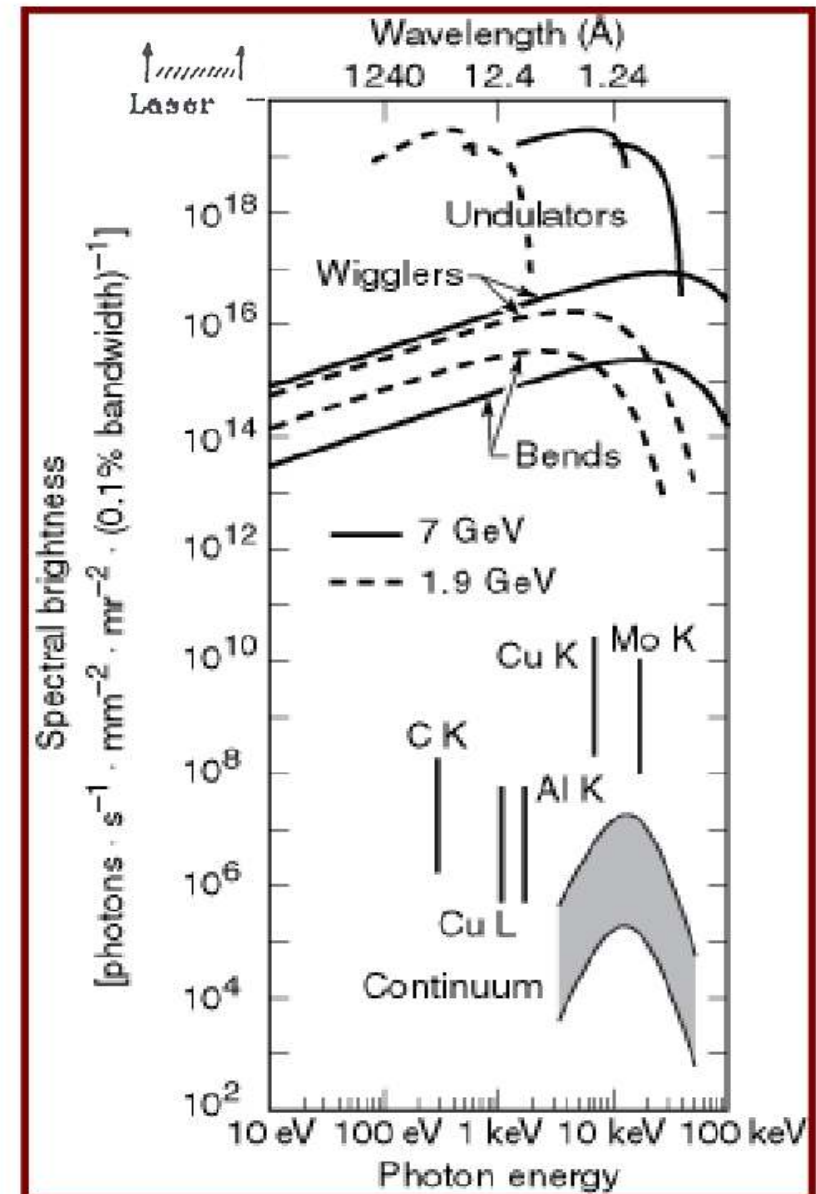
UNE grandeur caractéristique pour évaluer la qualité d'une source est la luminance.

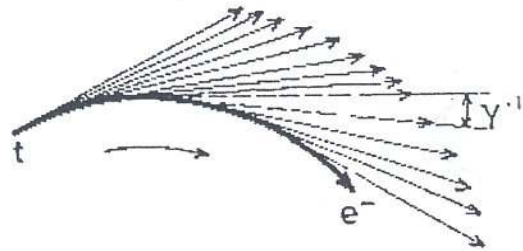


$$\text{Brilliance} = \frac{\text{Photons/second}}{\text{Source size} \times \text{Divergence} \times \Delta E}$$

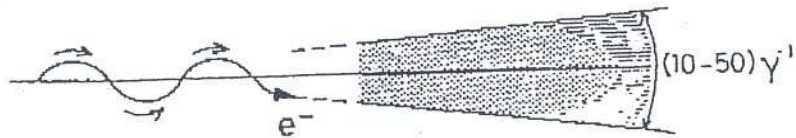
$$\left(\frac{\text{Photons / sec}}{\text{mm}^2 \cdot (\text{m rad})^2 (0.1\% \text{ ban})} \right)$$

Note: The coherence of a source is proportional to its brilliance!

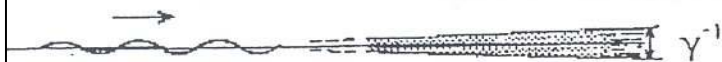




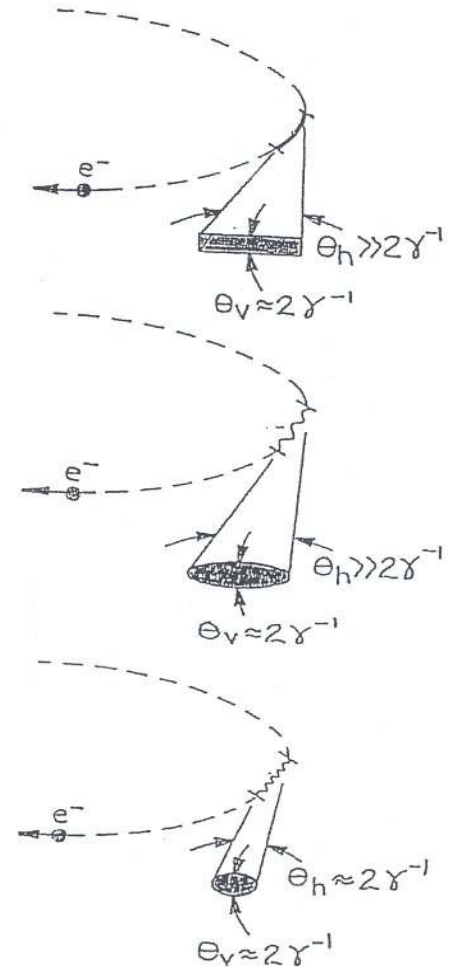
Bend magnet



Wiggler



Undulator



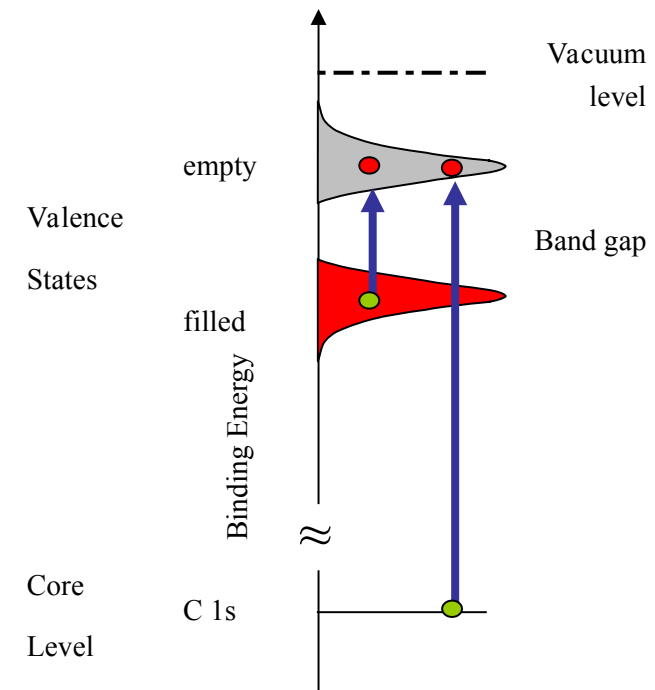
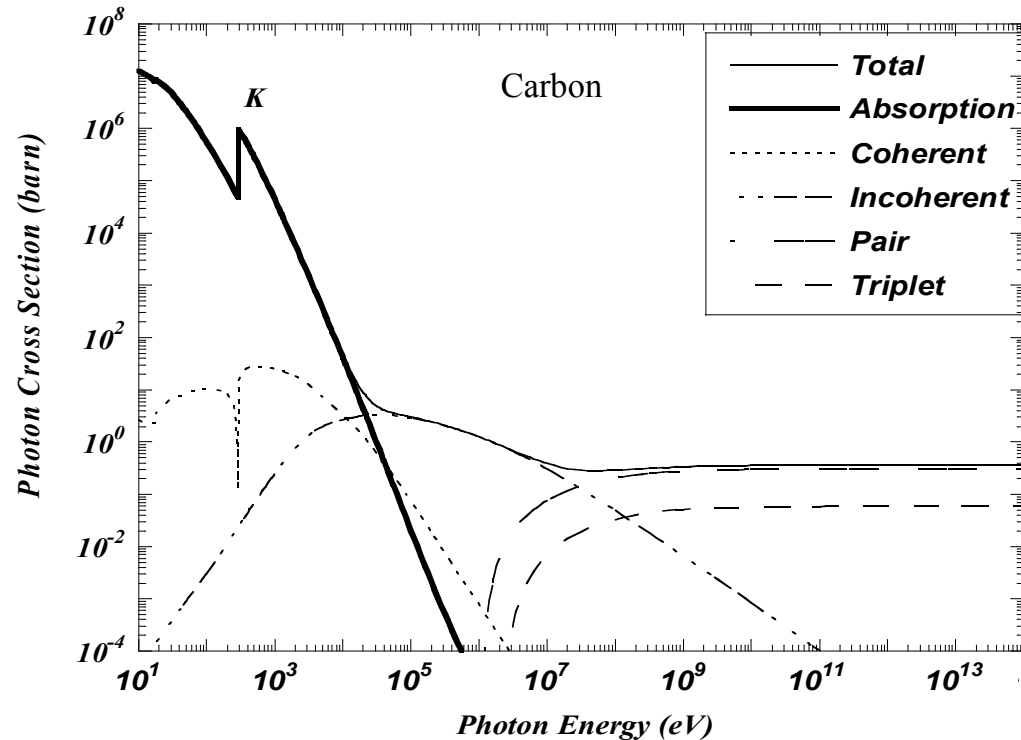
13 SR sources in Europe



... and 4
XUV/X-FELs



Photon – Matter Interaction



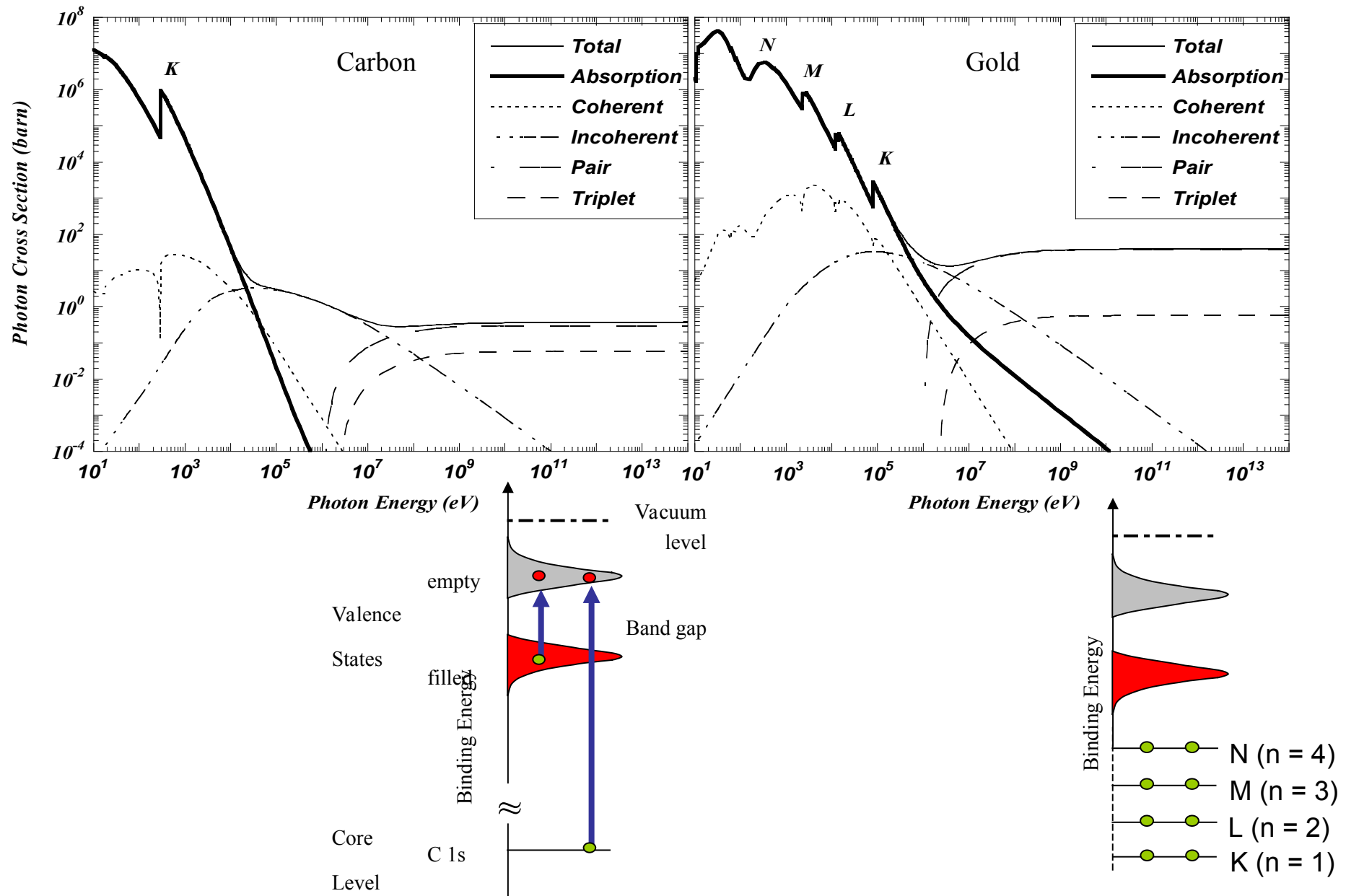
Absorption → dominates in X-ray photon energy range

Coherent = Thomson scattering → X-ray scattering/diffraction
Incoherent = Compton scattering

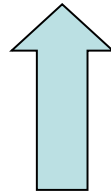
Photon Absorption Cross Section:

$$\sigma_X = \frac{\text{\# of absorbed photons per second}}{\text{\# of incident photons per second per area}}$$

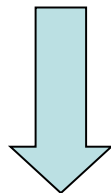
$$[\sigma] = \text{barn} = 10^{-24} \text{ cm}^2$$



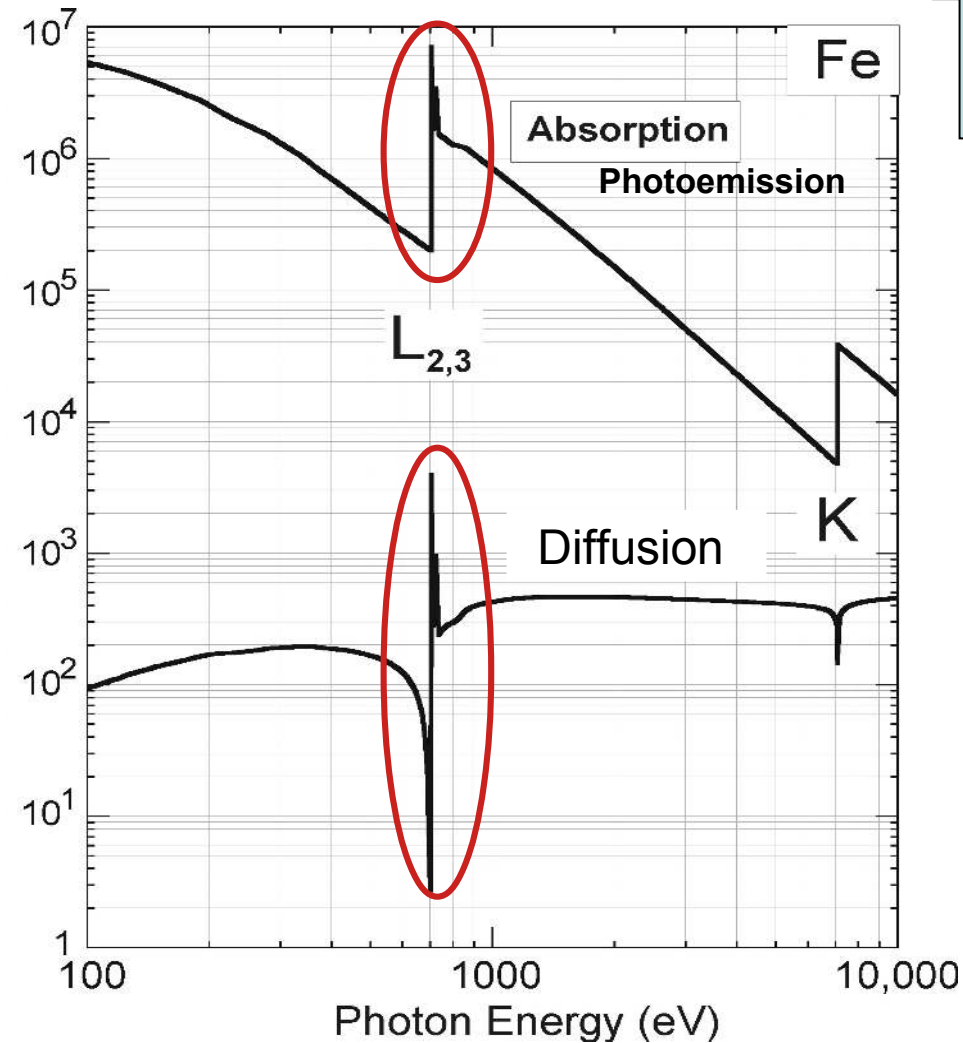
Visible light
(Metals)



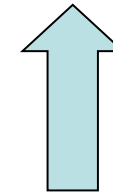
Visible light
(Insulators)



Section efficace (barn / atome)



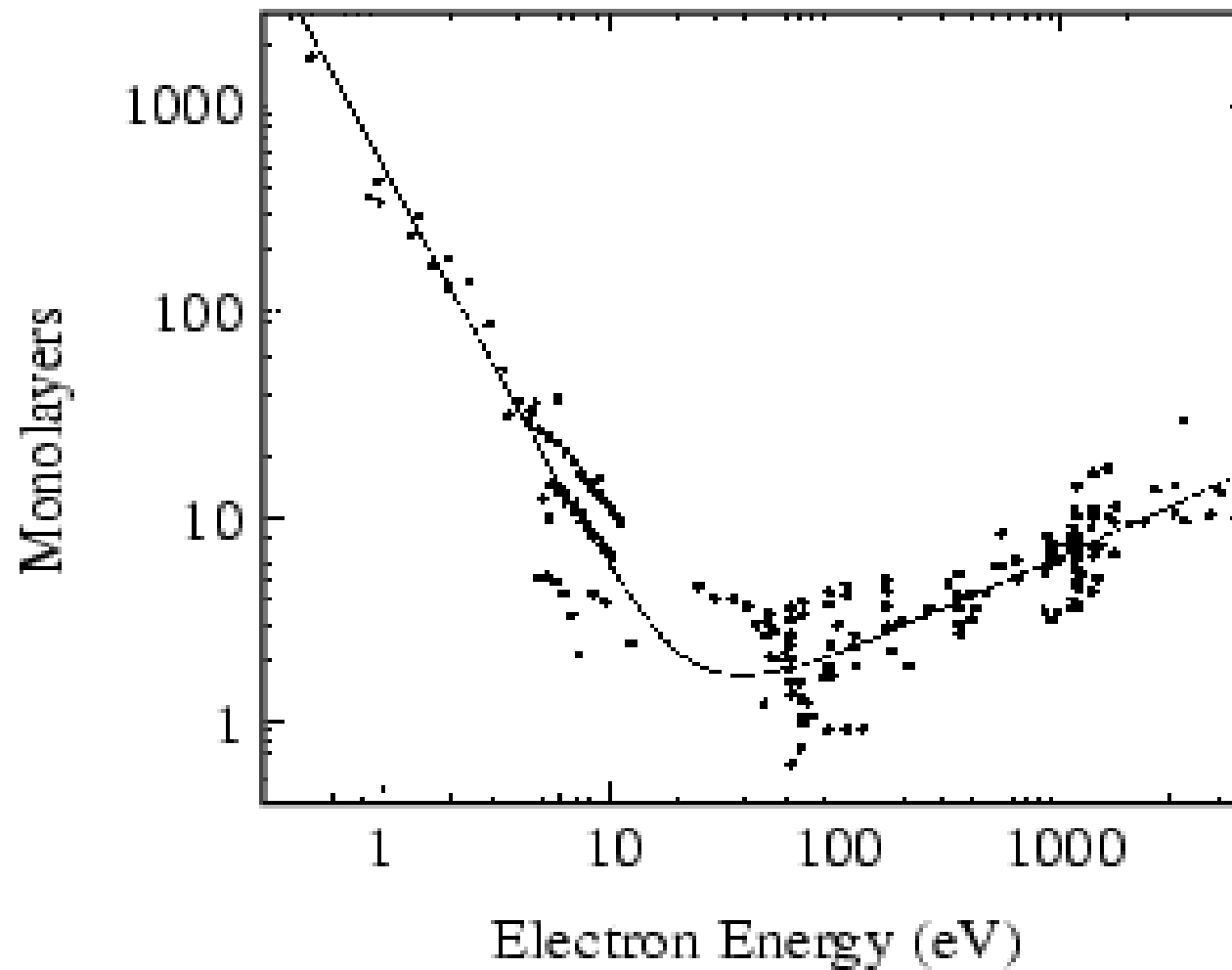
Electrons
Charged probe



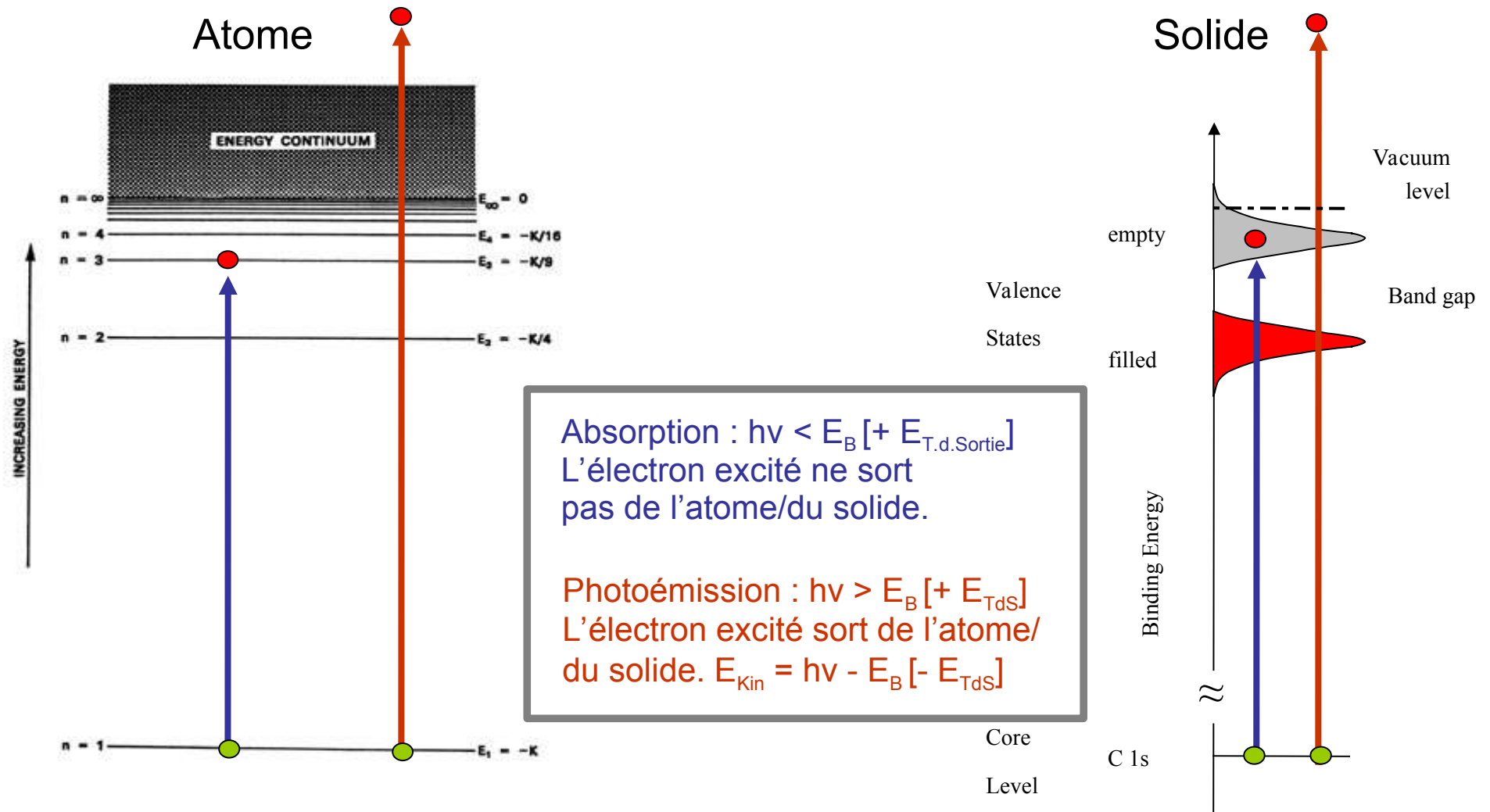
Neutrons

No charge,
but a spin

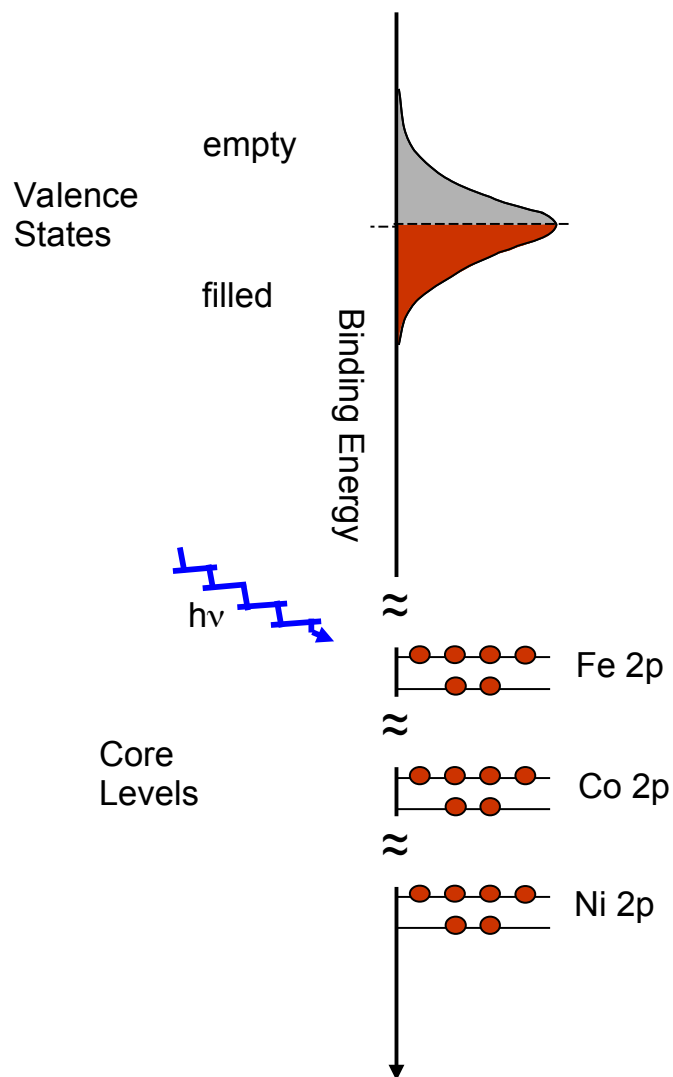




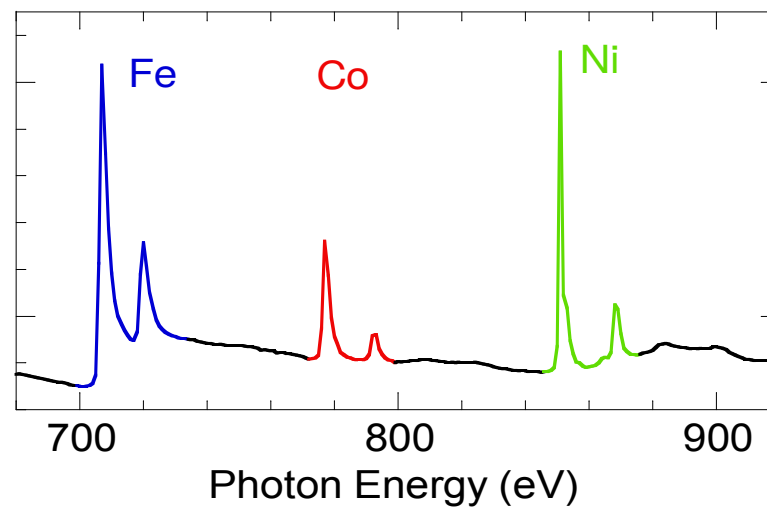
Absorption and Photoemission in atoms and solids



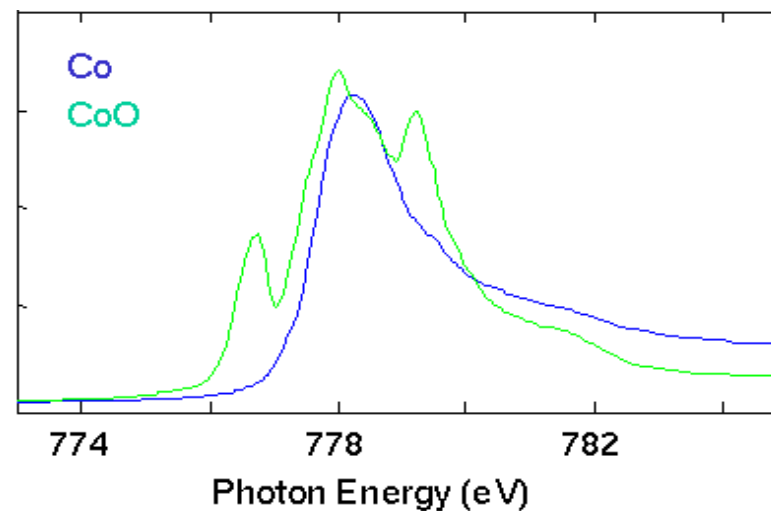
X-ray absorption spectroscopy



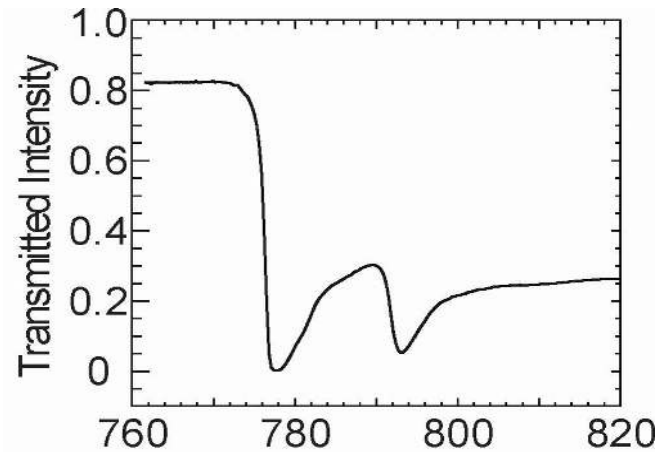
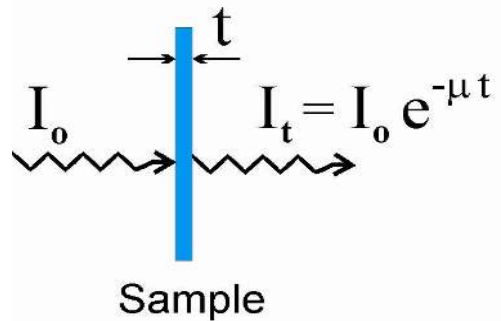
Elemental Specificity



Chemical Sensitivity



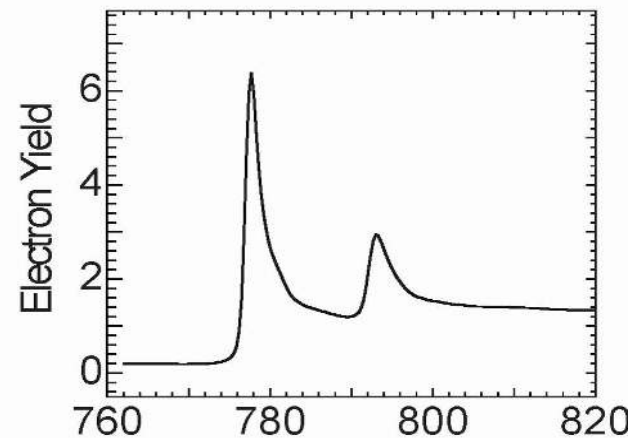
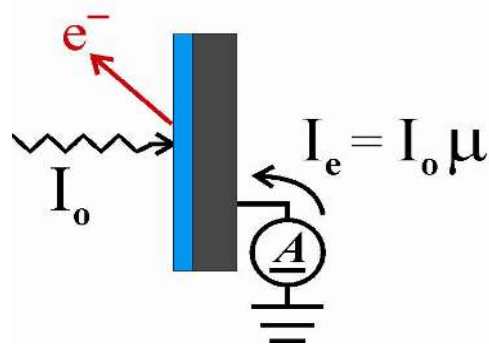
Transmission



“Photons lost”

Volume
($< 1 - 10$ microns)

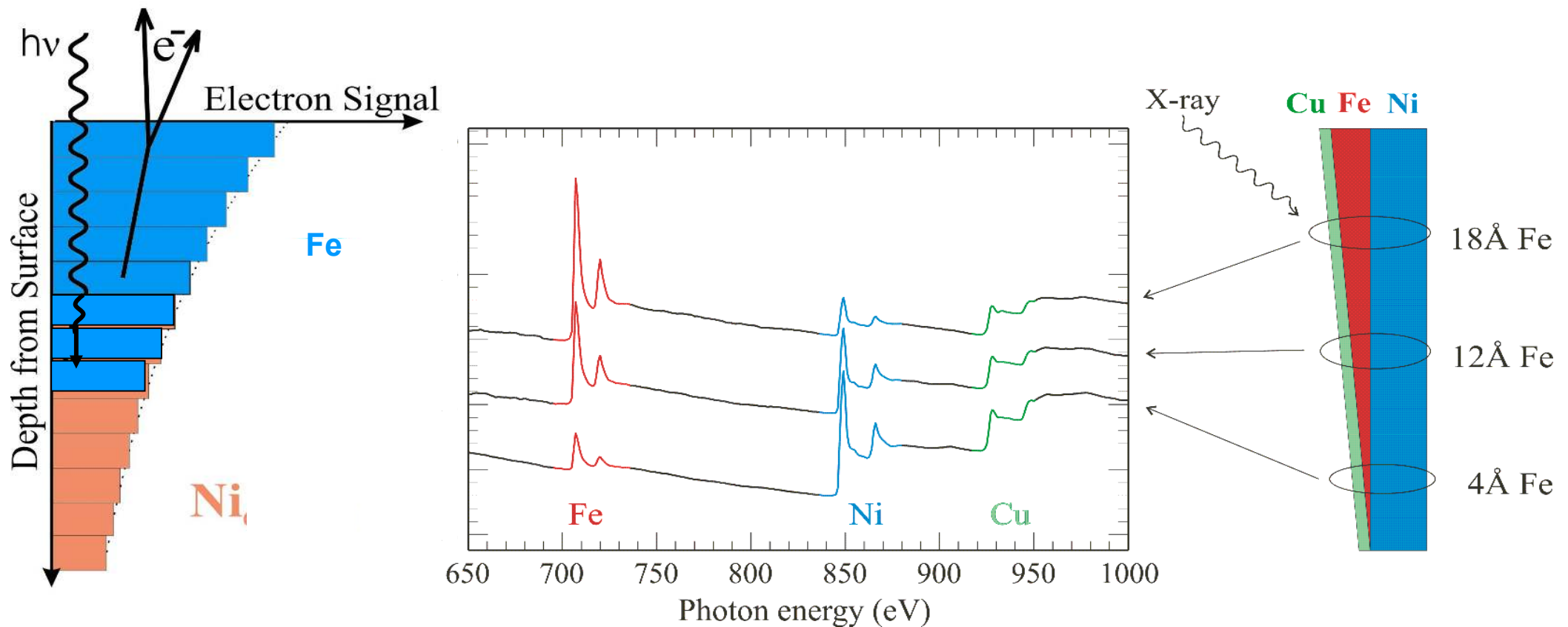
Electron Yield

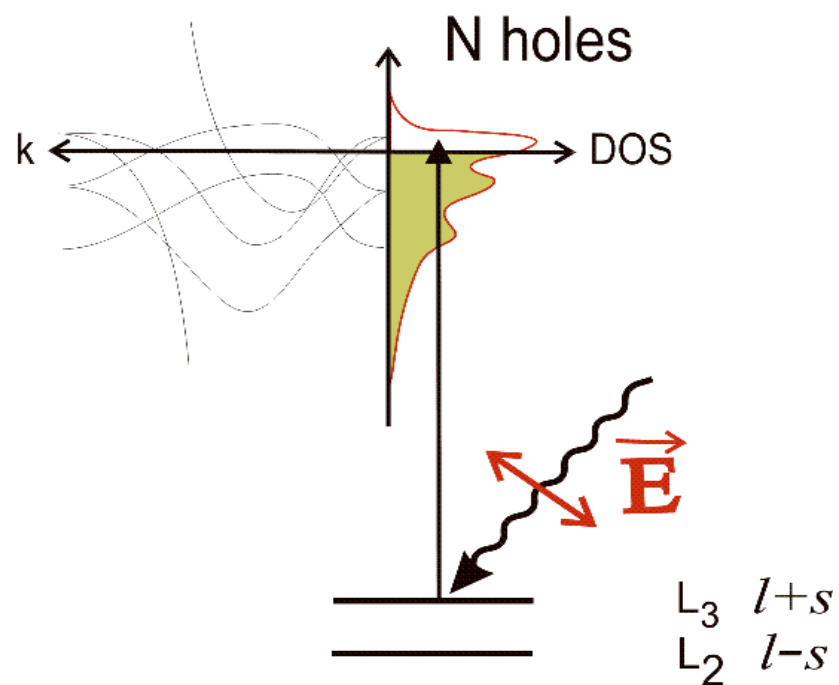


“Electrons generated”

Surface
($1 - 2$ nanometer)

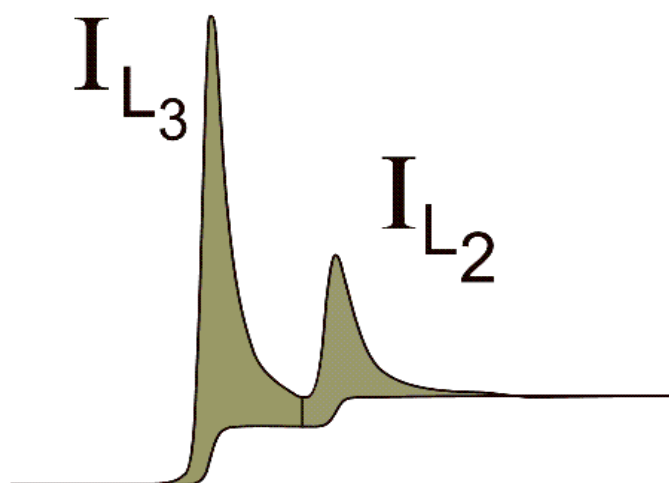
Total electron yield detection to render X-ray absorption spectroscopy surface sensitive

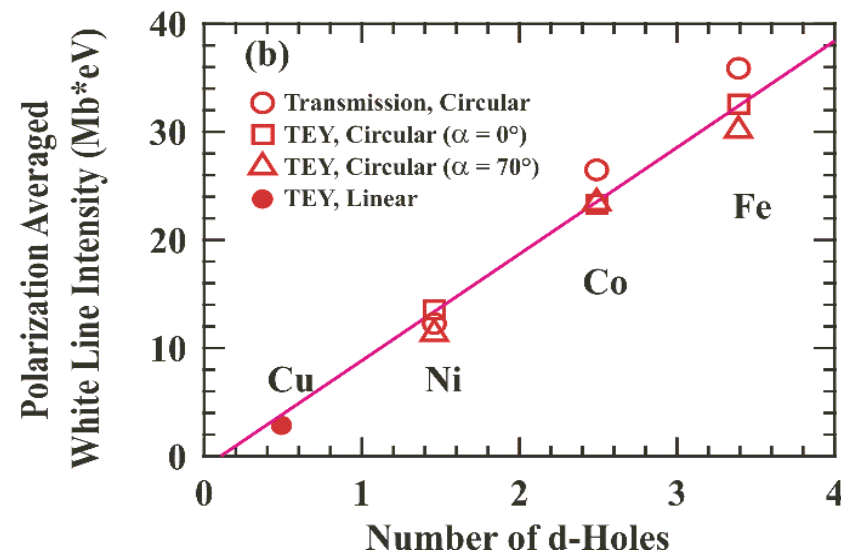
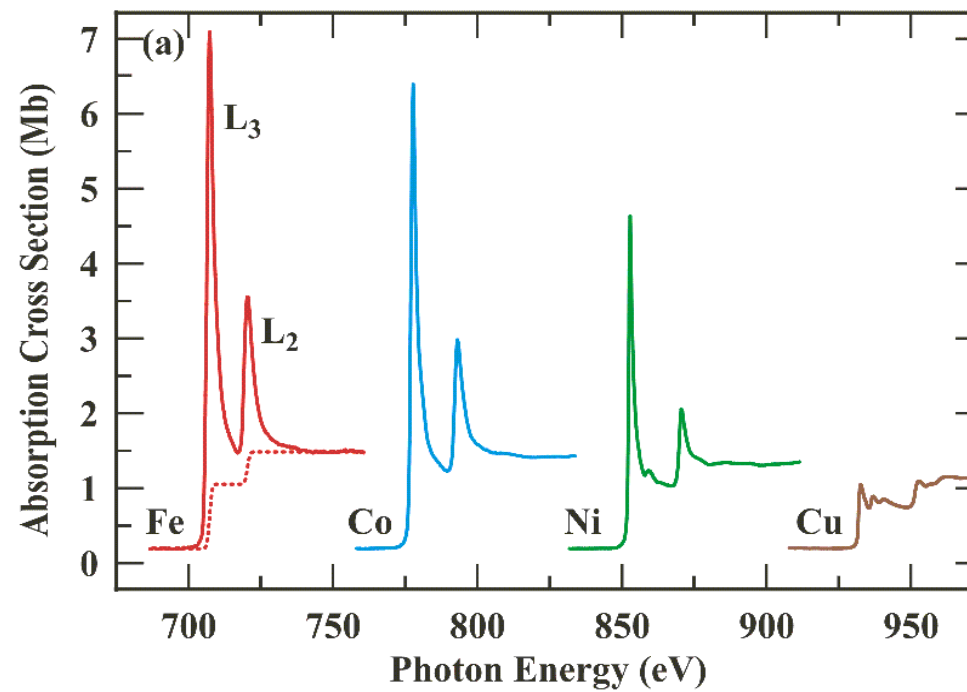




Fe
Co
Ni

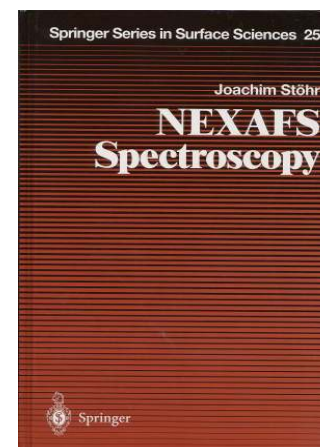
holes
3.4
2.5
1.5



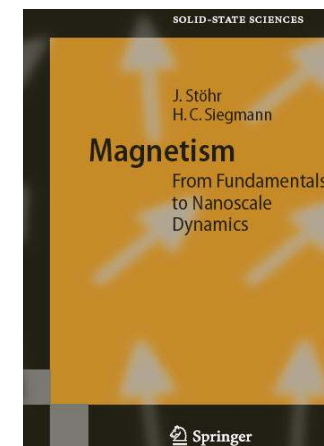


X-ray magnetic circular dichroism (XMCD) in X-ray absorption spectroscopy

J. Stöhr,
NEXAFS SPECTROSCOPY,
Springer Series in Surface Sciences 25,
Springer, Heidelberg, 1992.



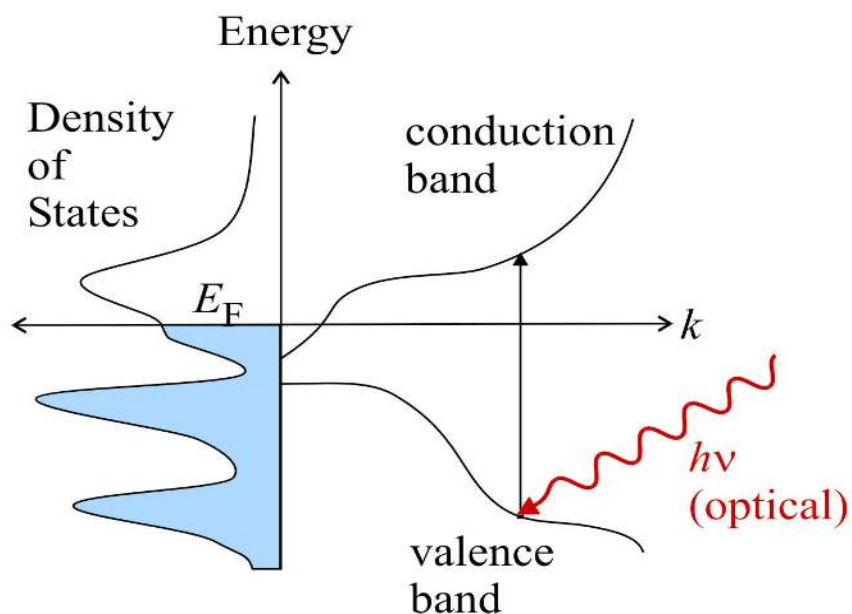
J. Stöhr and H. C. Siegmann
*MAGNETISM: FROM FUNDAMENTALS
TO NANOSCALE DYNAMICS*,
Springer Series in Solid State Sciences 152,
Springer, Heidelberg, 2006



Many (!) transparencies are taken from Jo Stöhr's 2007 presentation on 'X-rays and magnetism'

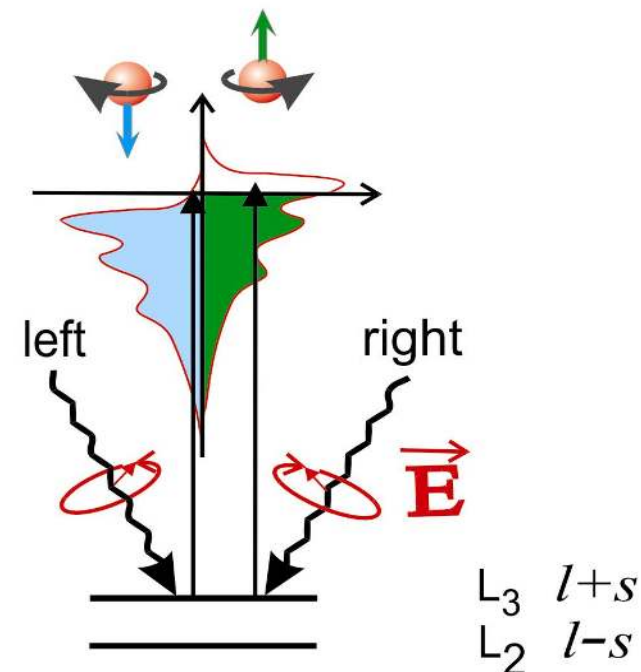
www-ssrl.slac.stanford.edu/stohr

Faraday and Kerr effect

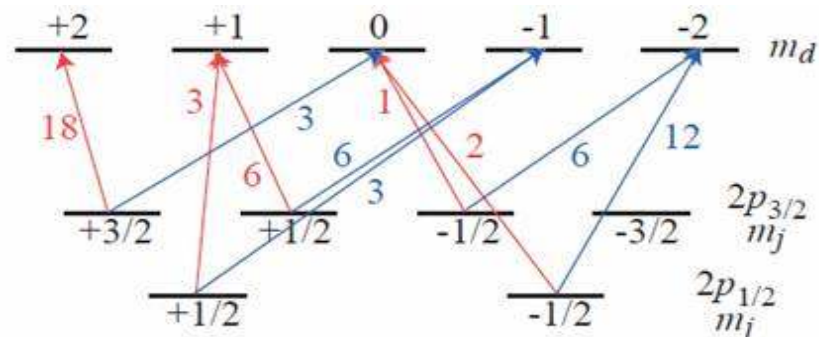


Magneto-optical response:
weak, k -dependent

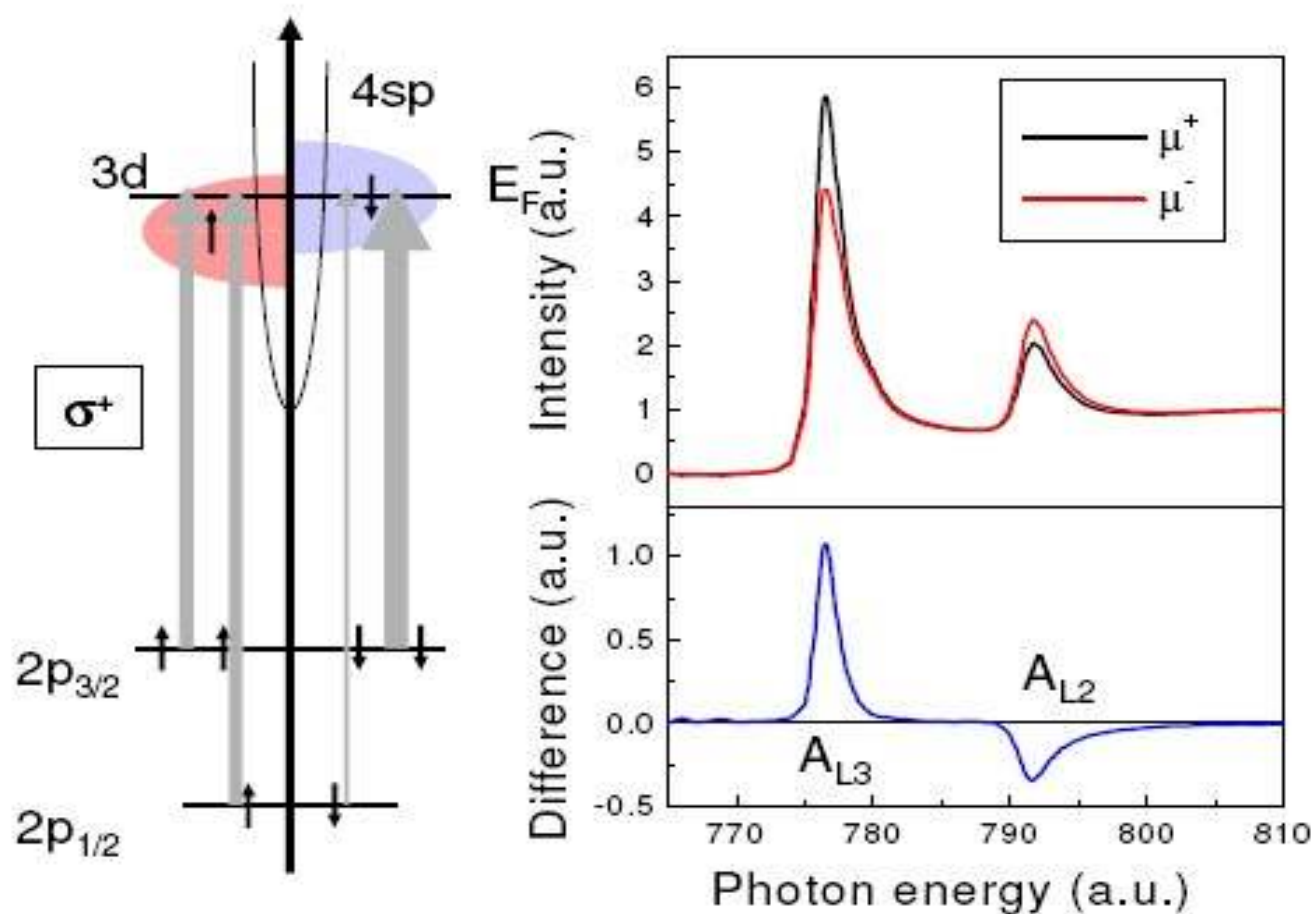
X-ray Magnetic Dichroism

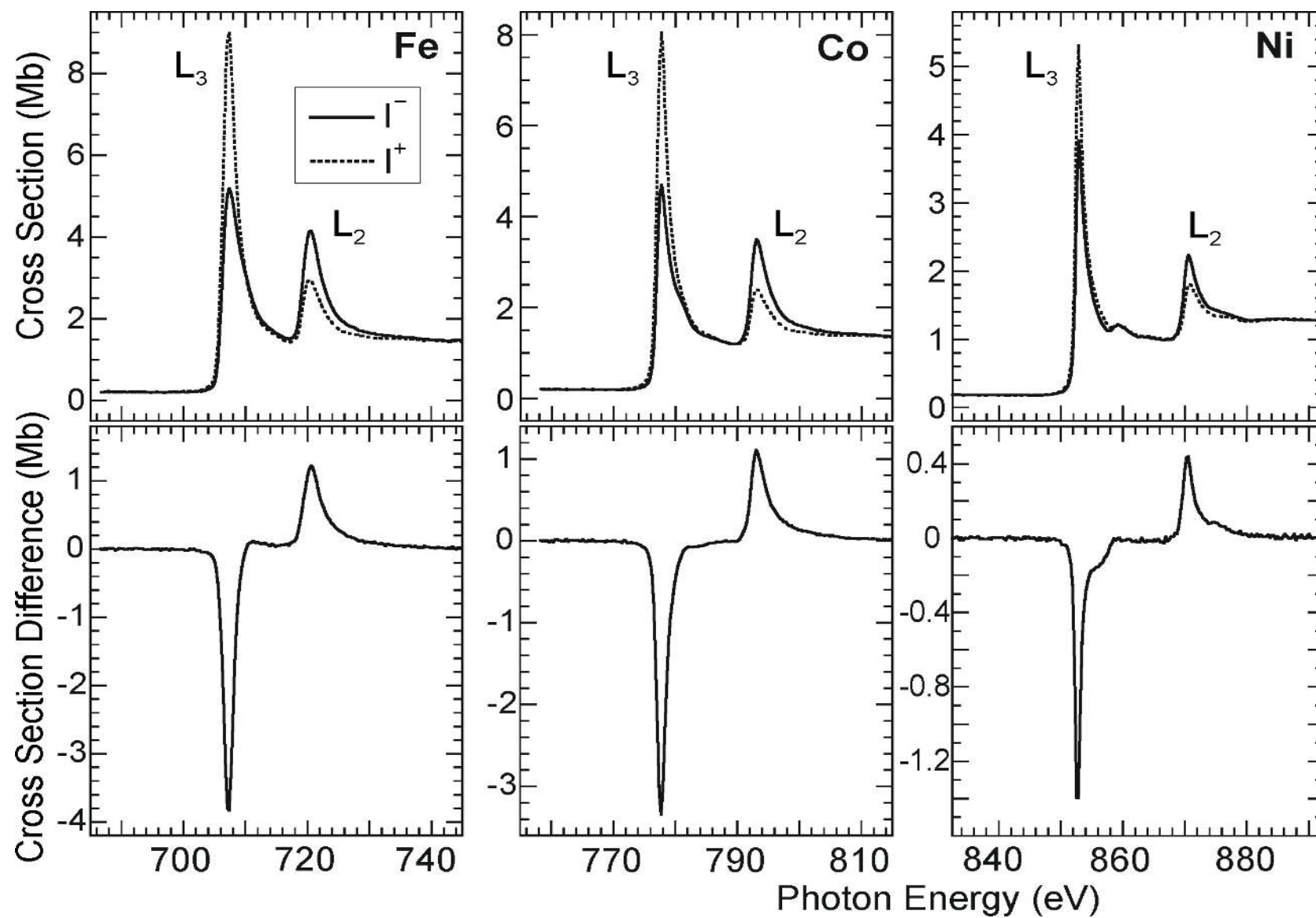


X-ray response:
strong, k -integrated quantities
number of holes, spin moment, orbital moment

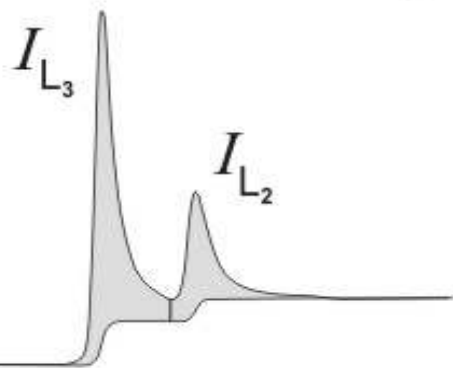
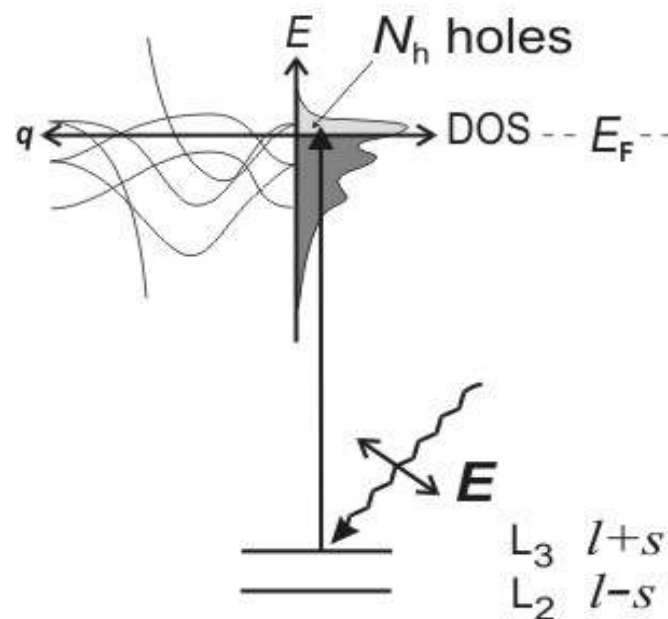


Relative transition amplitudes are given by the respective Clebsch Gordon coefficients





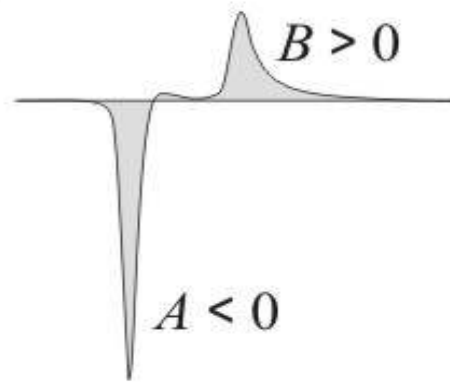
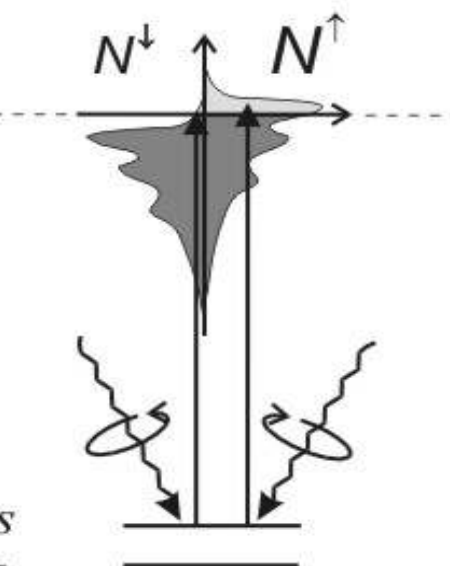
(a) *d*-Orbital occupation



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

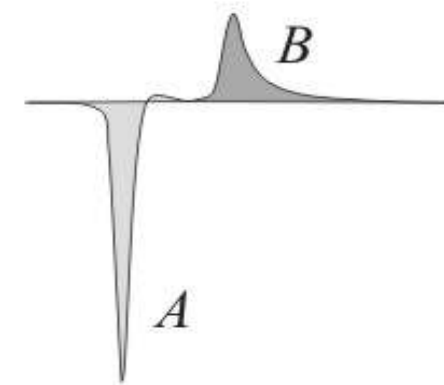
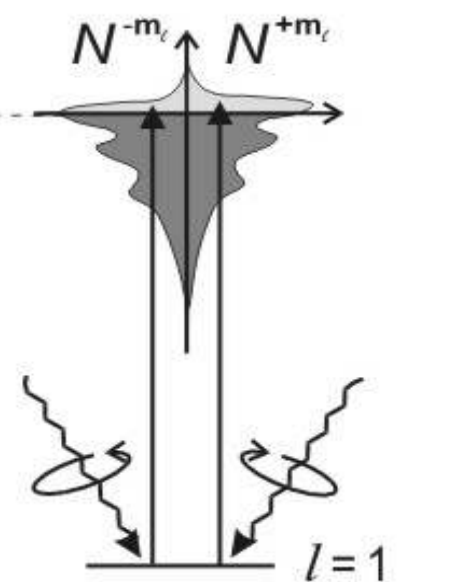
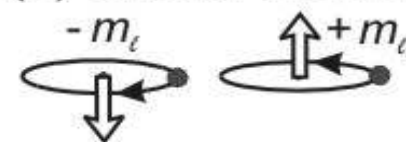
(b) Spin moment

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

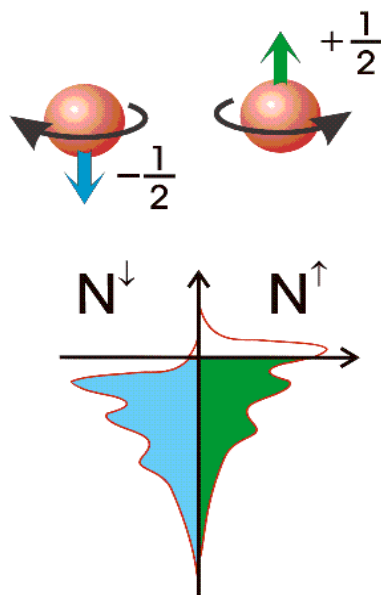


$$m_s = \mu_B \langle -A + 2B \rangle / C$$

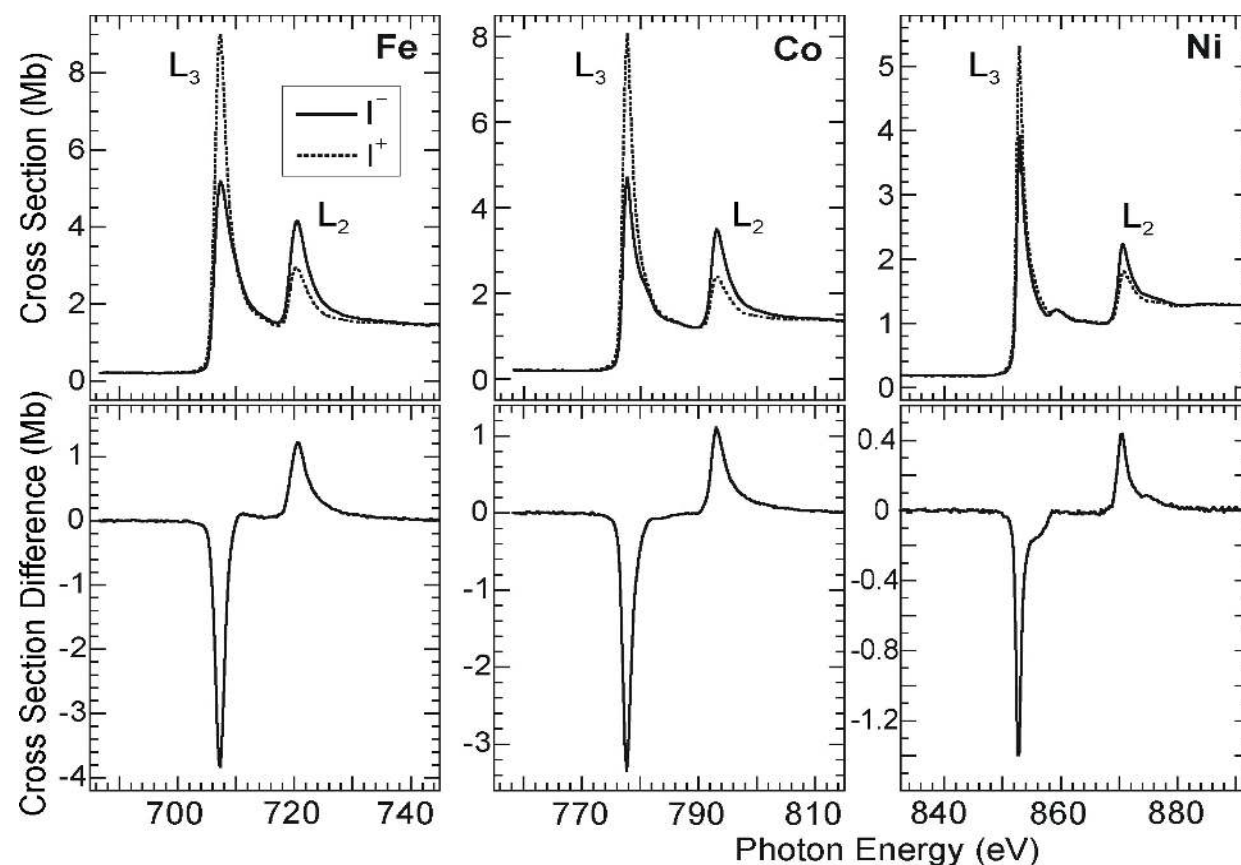
(c) Orbital moment



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



	$m_{\text{spin}}^{\text{tot}}$	$m_{\text{spin}}^{\text{d}}$	$m_{\text{spin}}^{\text{sp}}$
Fe	2.19	2.26	-0.07
Co	1.57	1.64	-0.07
Ni	0.62	0.64	-0.02

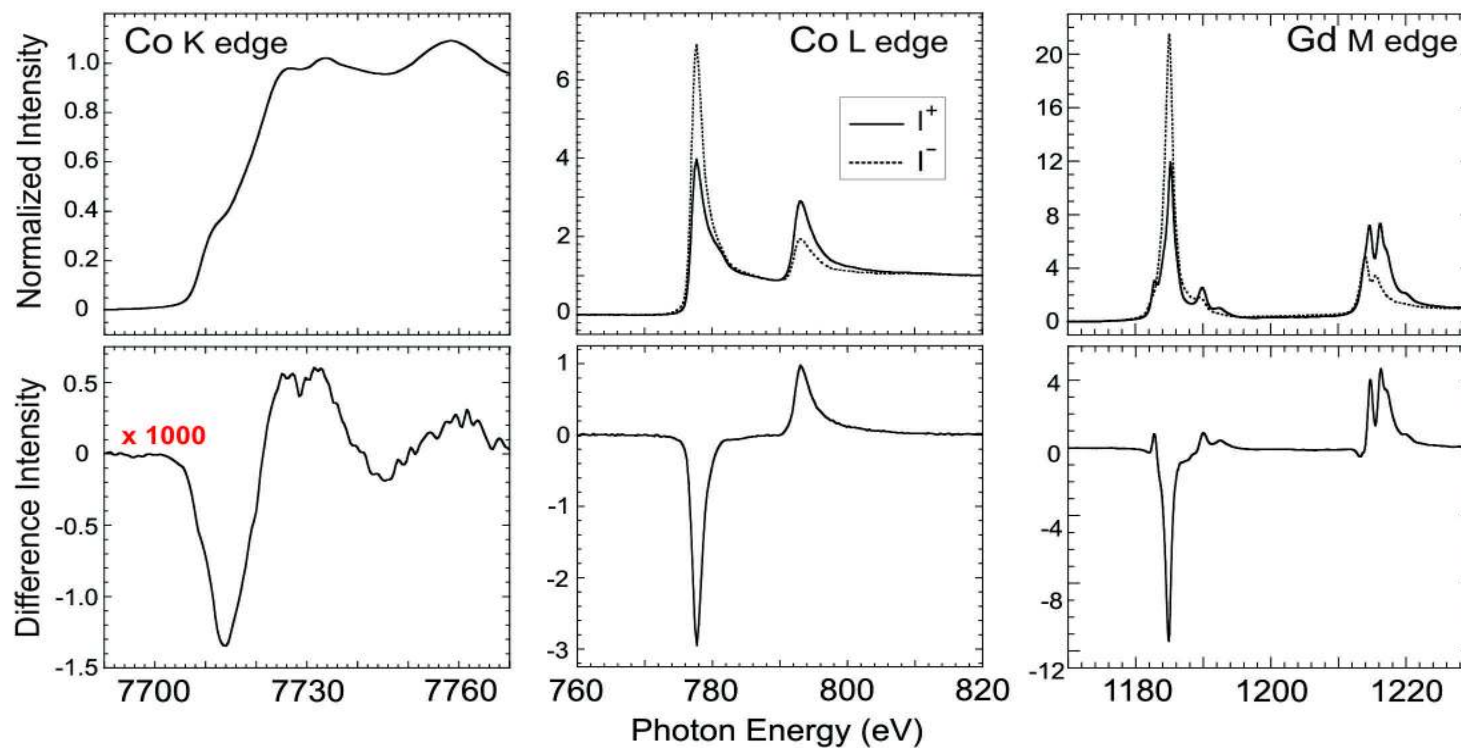
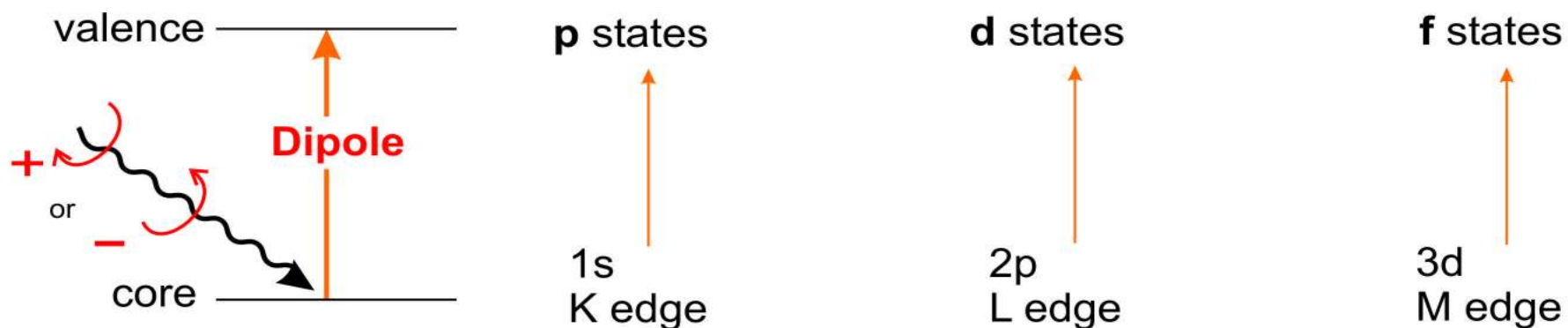


(b) Spin moment

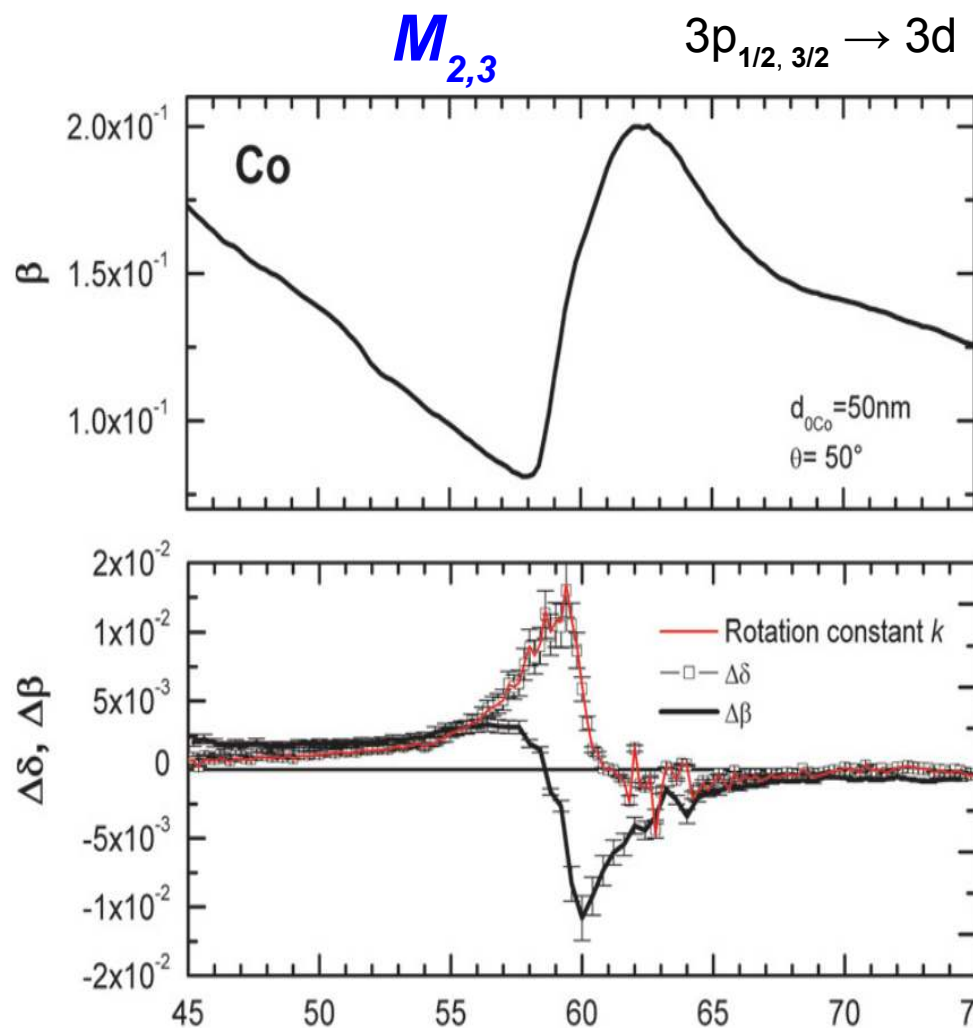
(c) Orbital moment

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

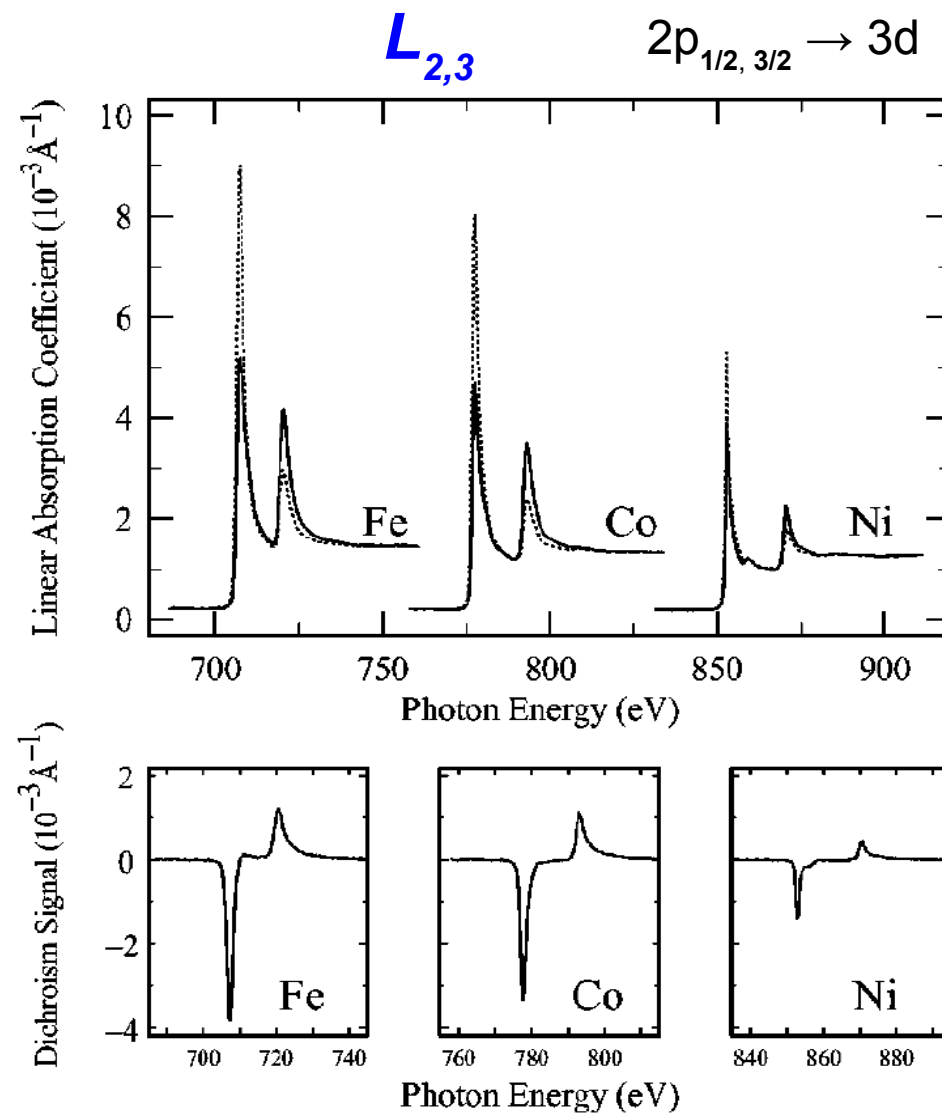
$$m_o = -2\mu_B \langle A + B \rangle / 3C$$



XMCD of transition metal $M_{2,3}$ and $L_{2,3}$ edges



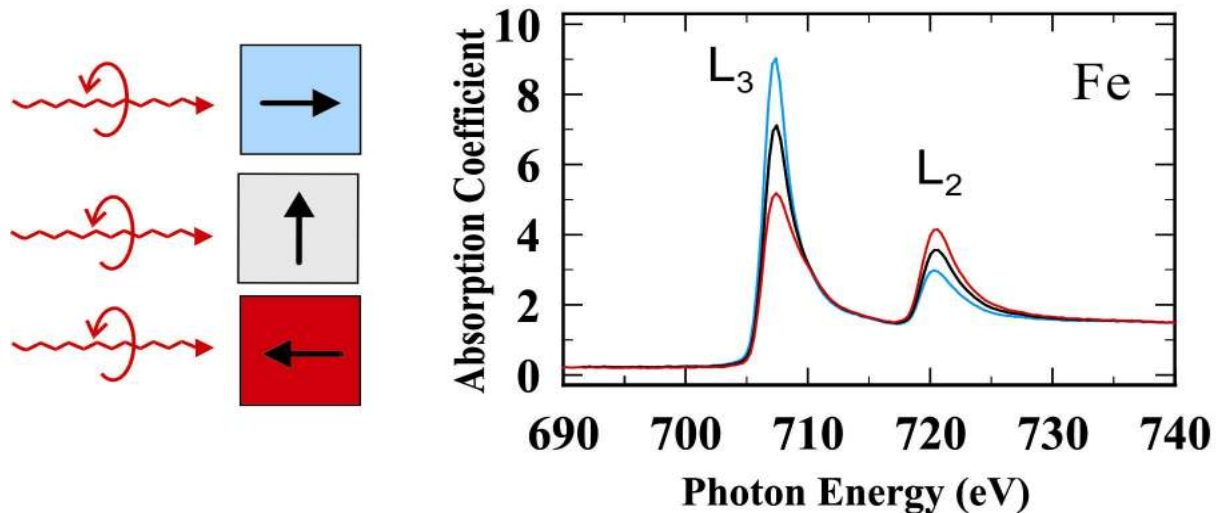
S. Valencia et al., New J. Phys. 8, 254 (2006)



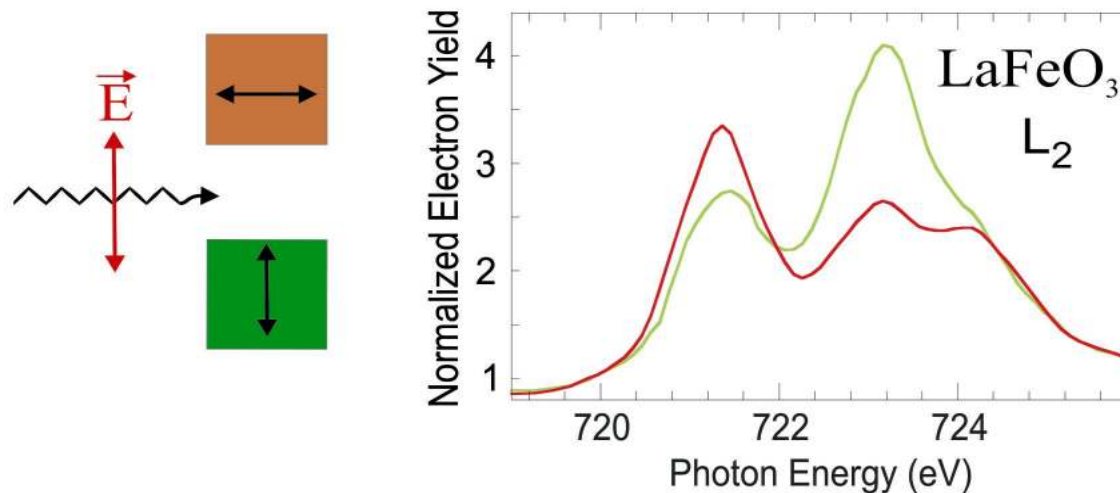
R. Nakajima et al., Phys. Rev. B 59, 6421 (1999)

X-ray magnetic LINEAR dichroism (XMLD) in X-ray absorption spectroscopy

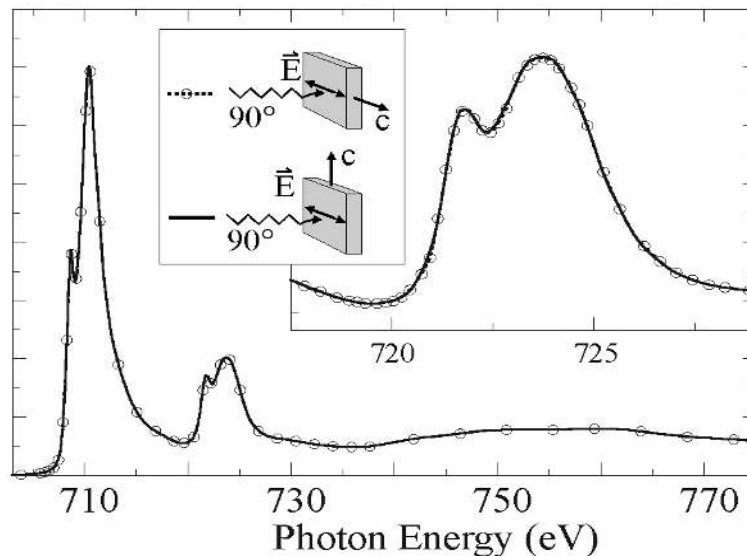
XMCD : X-ray Magnetic Circular Dichroism: **Ferromagnets**



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



XMLD: Linear dichroism and presence of AFM order



$\text{La}_{0.6}\text{Sr}_{0.4}\text{FeO}_3 / \text{SrTiO}_3$ (110)

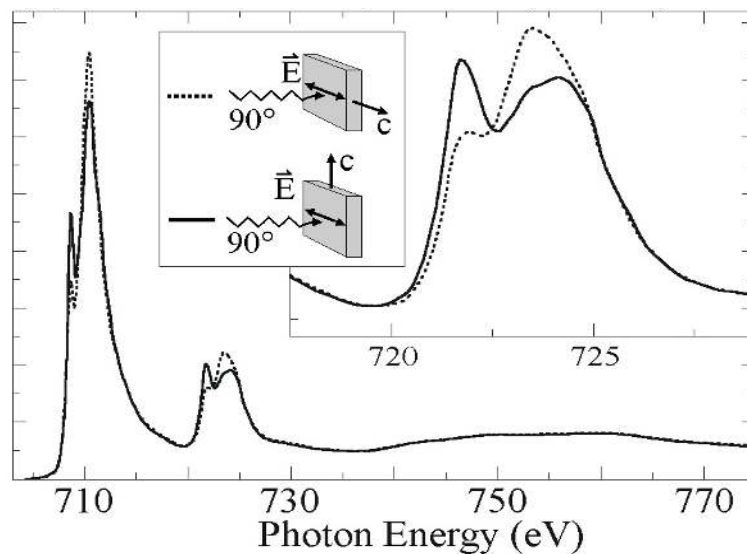
Above Néel temperature

No linear dichroism

Warnings:

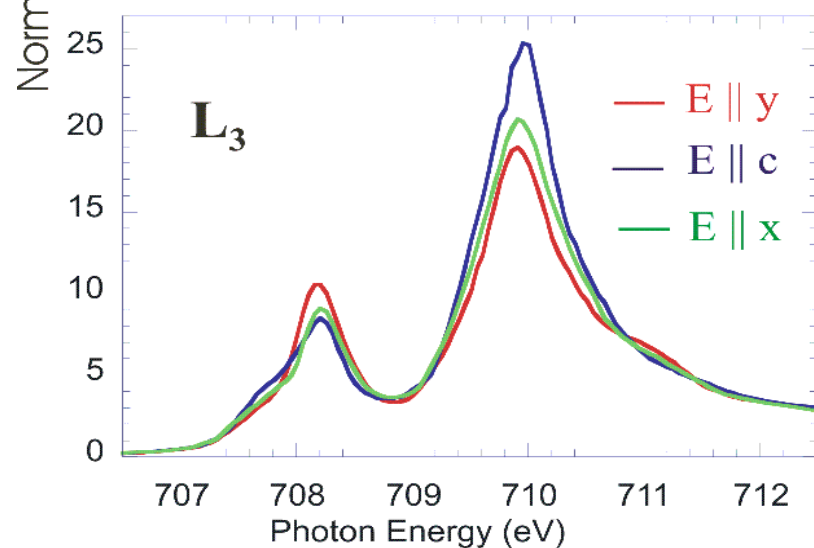
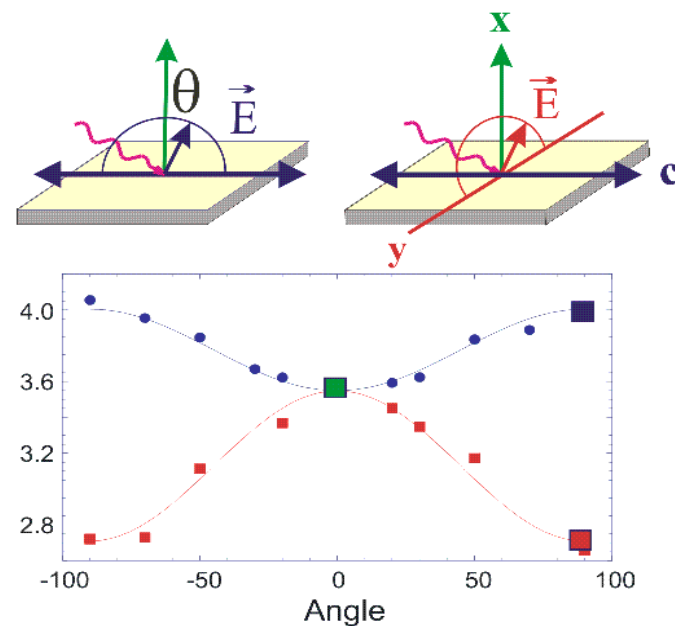
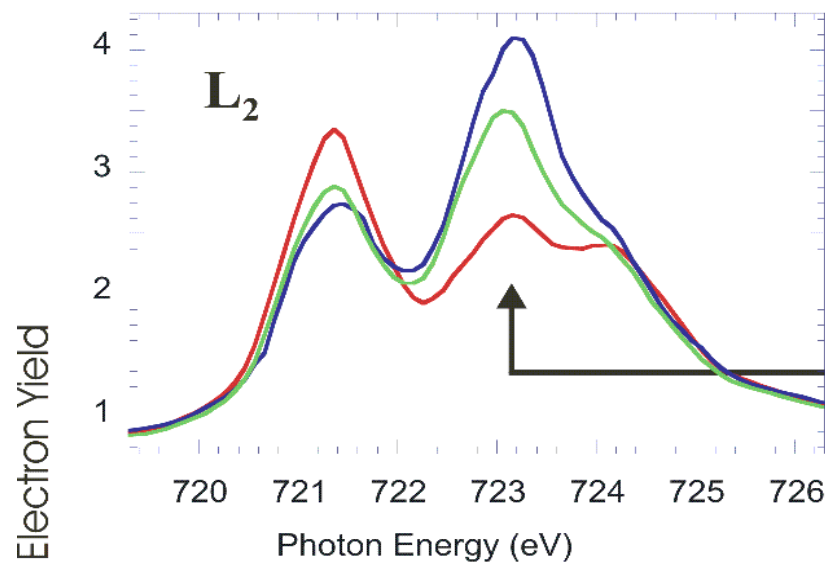
Crystal fields can cause linear dichroism

Relationship between orientation of AFM axis and dichroic ratio can depend on crystal orientation

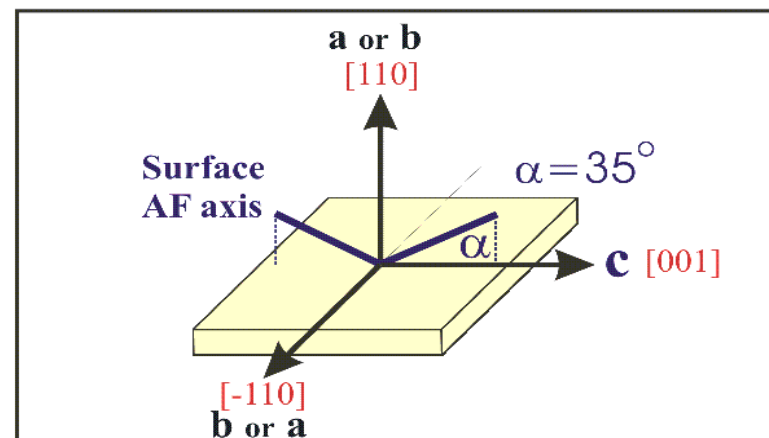


Below Néel temperature

Strong linear dichroism



AF axis is rotated from bulk



Adding spatial resolution to x-ray spectroscopy

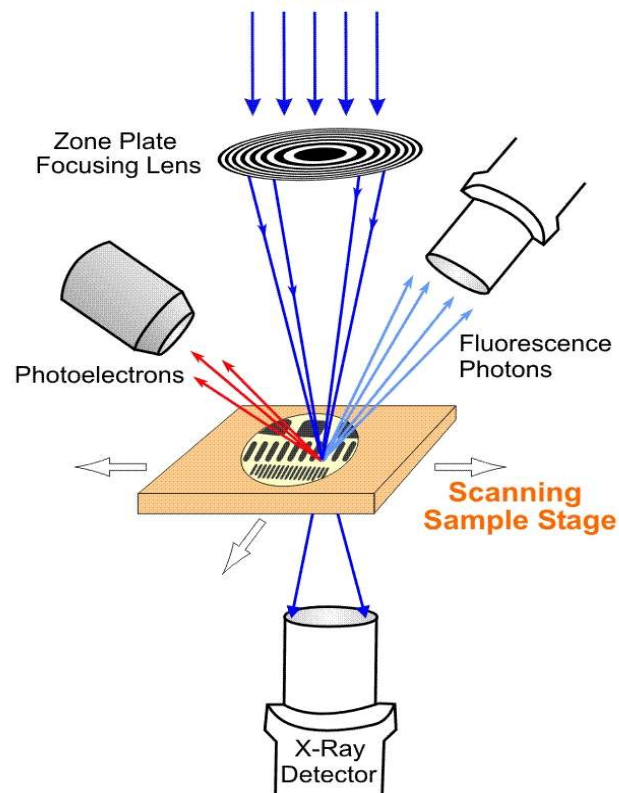
X-ray spectro-microscopy

Application: Co / NiO

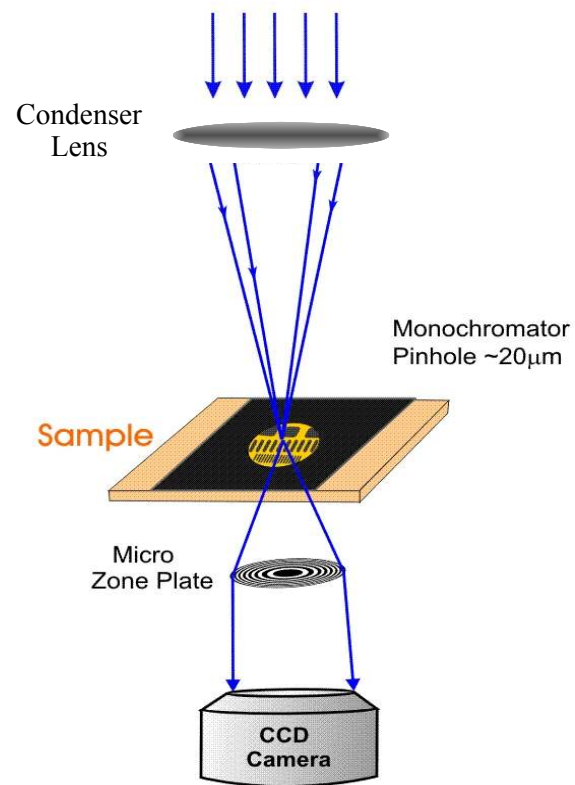
The interface between a ferromagnetic metal
and an antiferromagnetic metal oxide

Quantitative imaging with sensitivity to
elemental and chemical distribution and charge/spin ordering

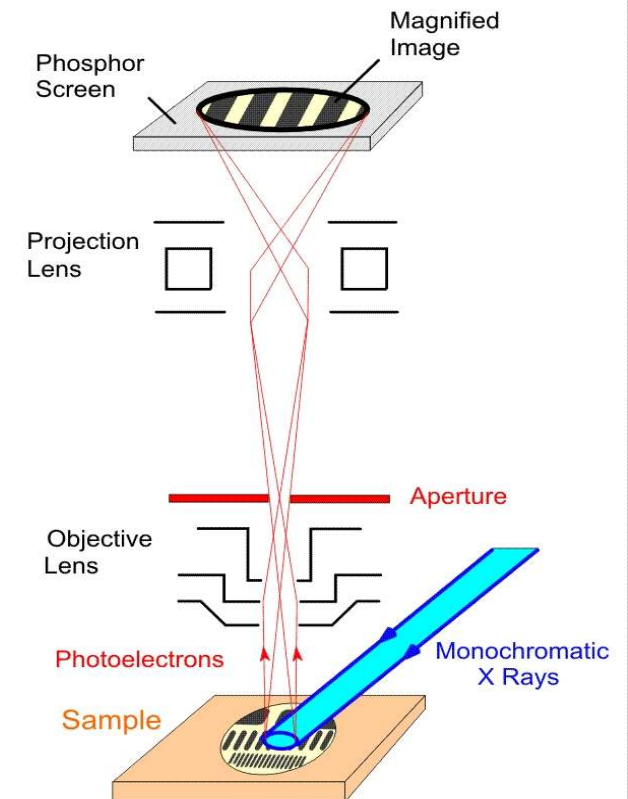
Scanning Transmission X-ray Microscopy STXM



Transmission X-ray Microscopy TXM



X-Ray Photoemission Electron Microscopy XPEEM

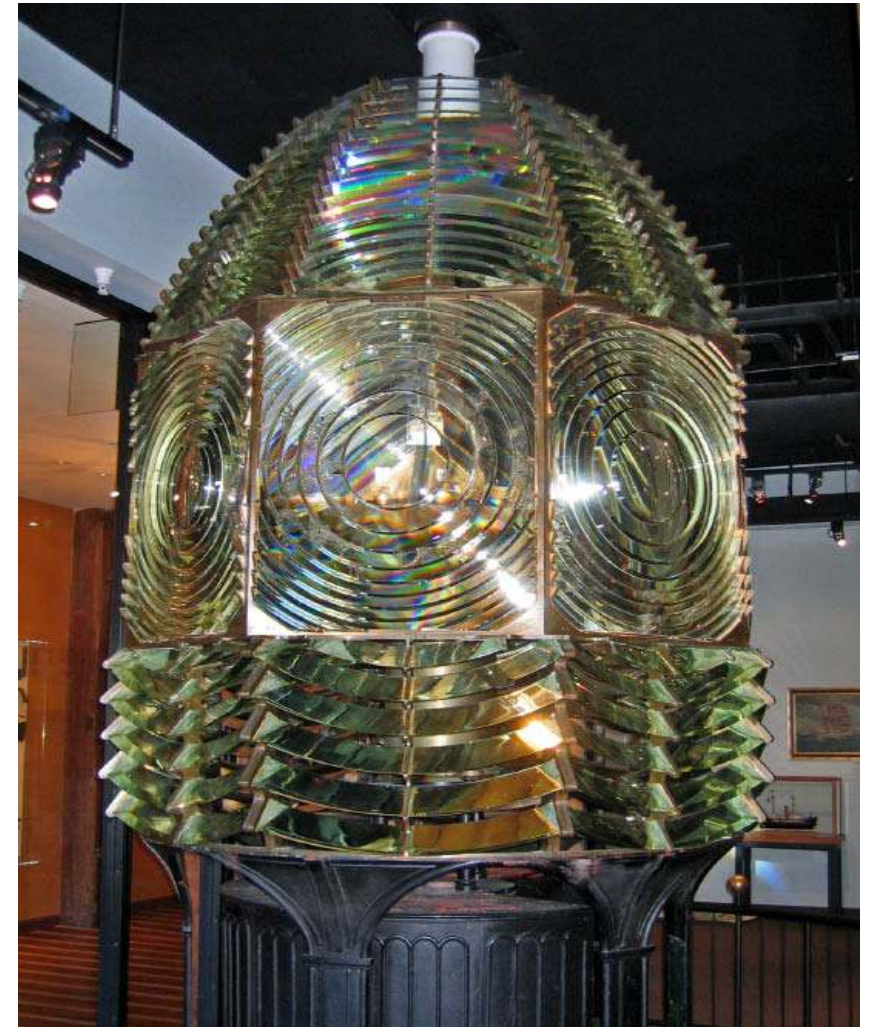
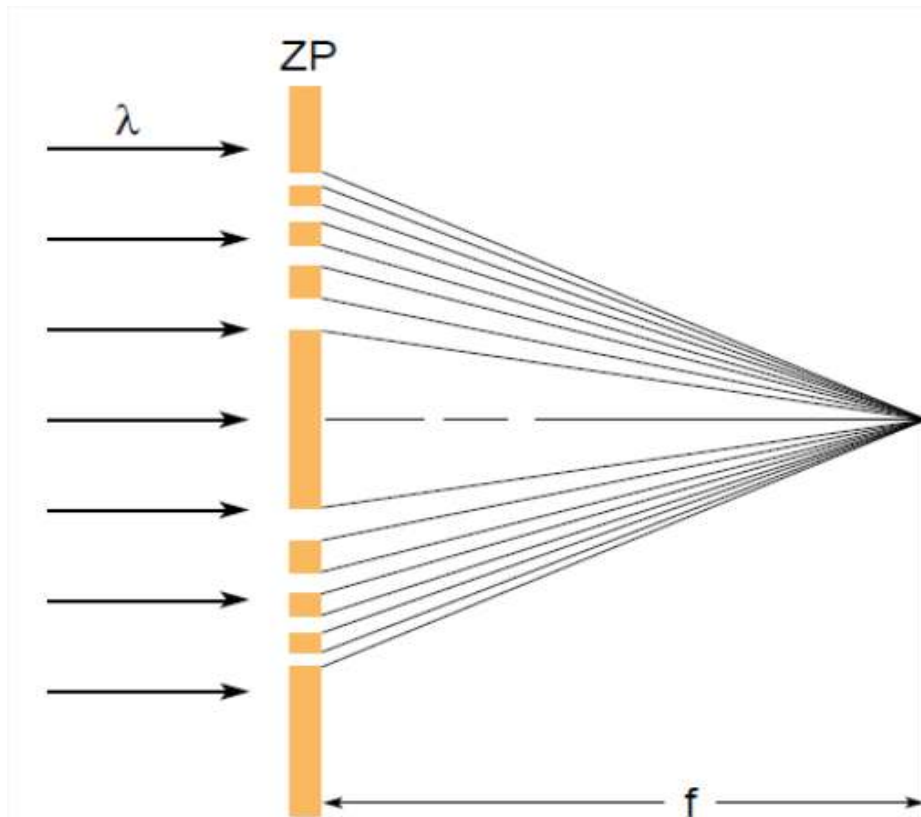


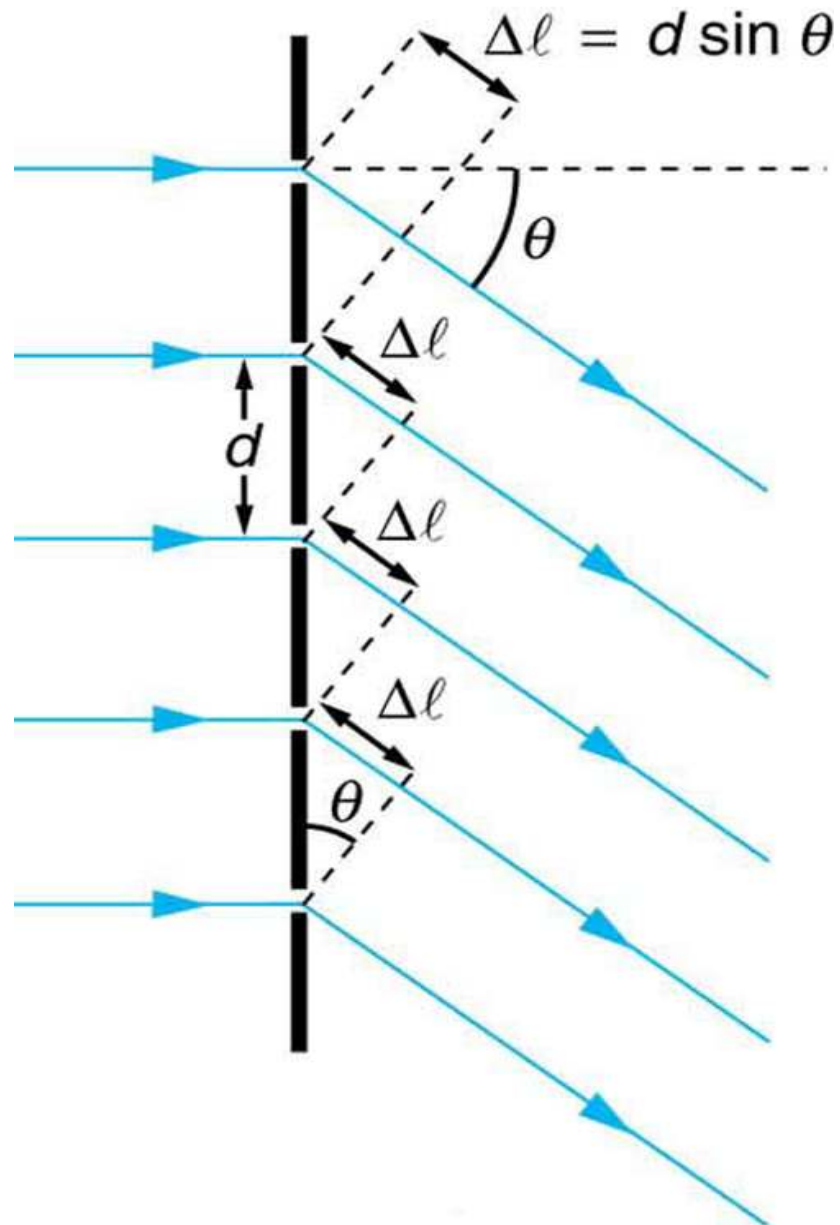


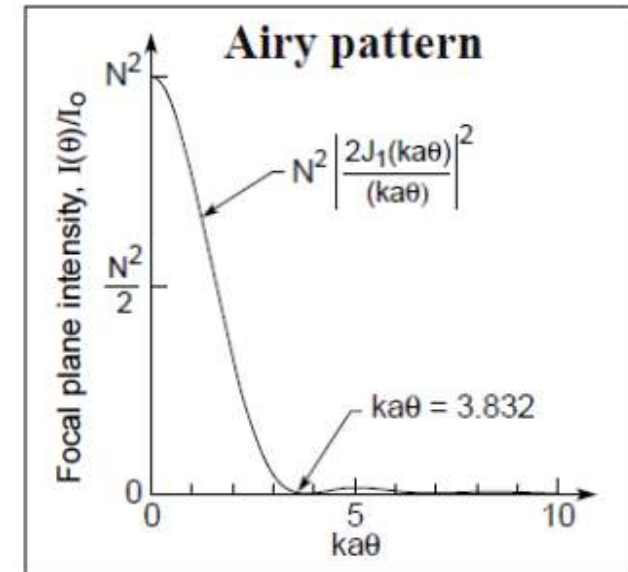
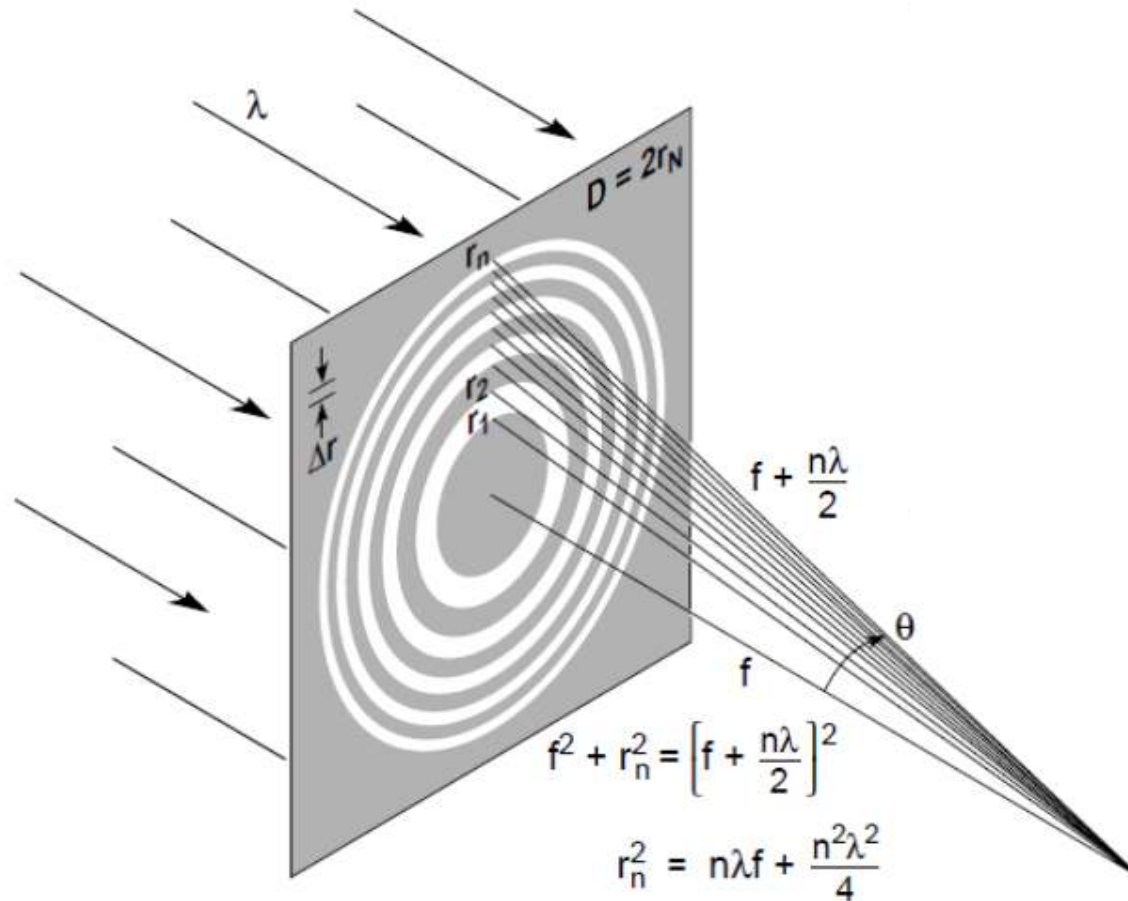
- spatial resolution $\sim \Delta r$
- focal length $\sim N(\Delta r)^2/\lambda$
- spectral bandwidth $\Delta\lambda/\lambda \sim 1/N$

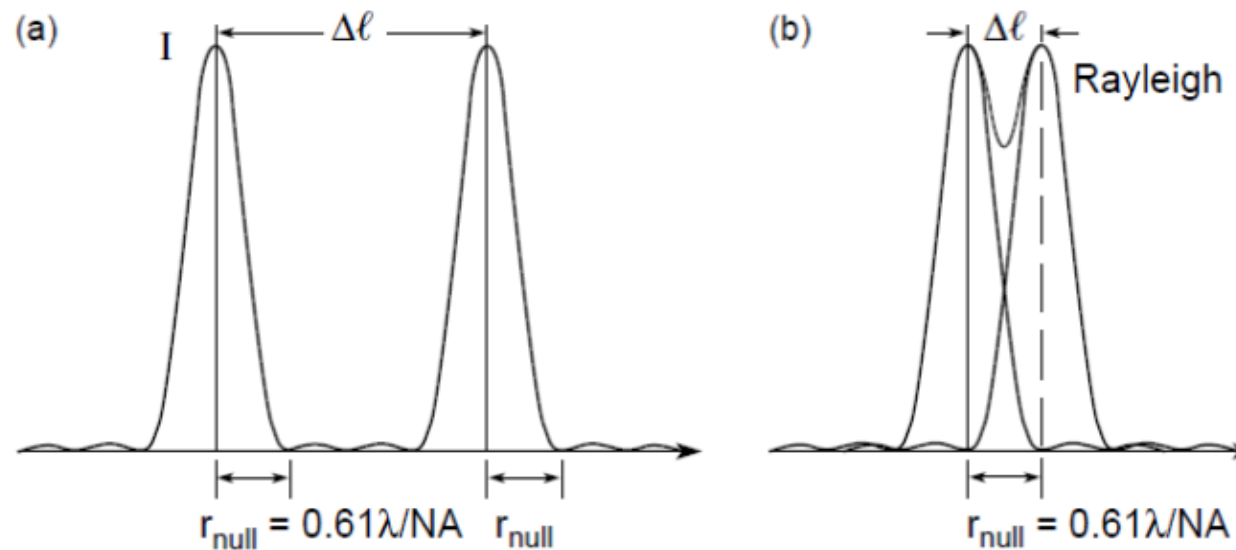
Fresnel Zone Plate Principle

Spherical Grating with varying line density







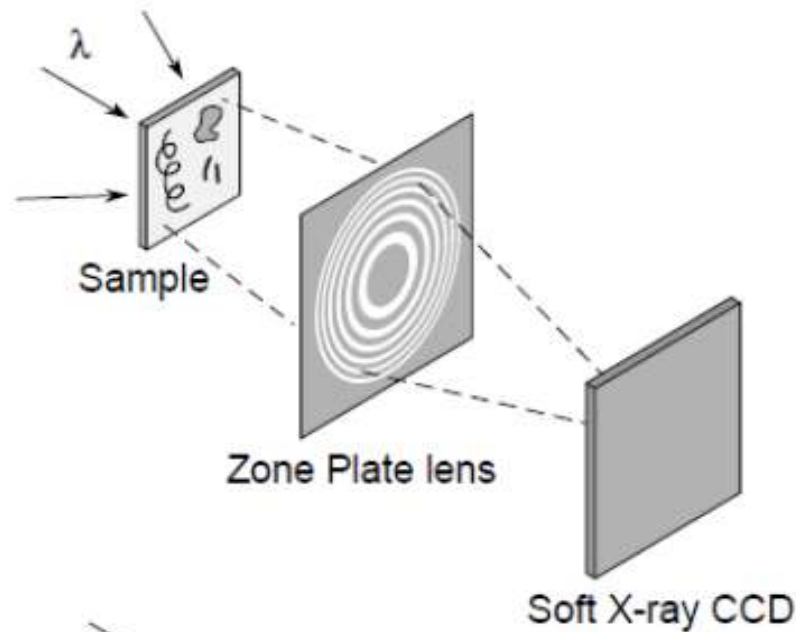


- Point sources are spatially coherent
- Mutually incoherent
- Intensities add
- Rayleigh criterion (26.5% dip)

Conclusion: With spatially coherent illumination, objects are “just resolvable” when

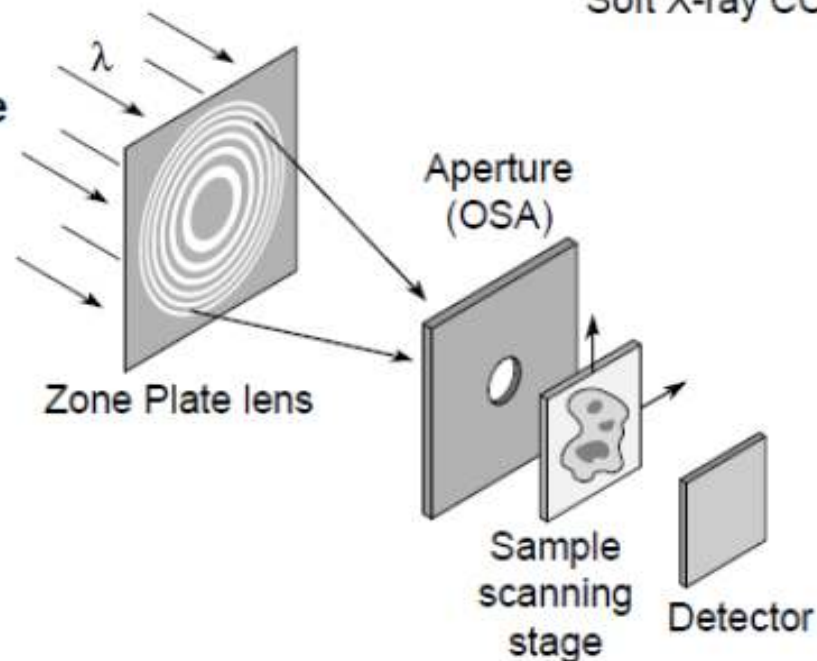
$$\text{Res}|_{\text{coh}} = \frac{0.61 \lambda}{\text{NA}} = 1.22 \Delta r$$

Full-Field Microscope



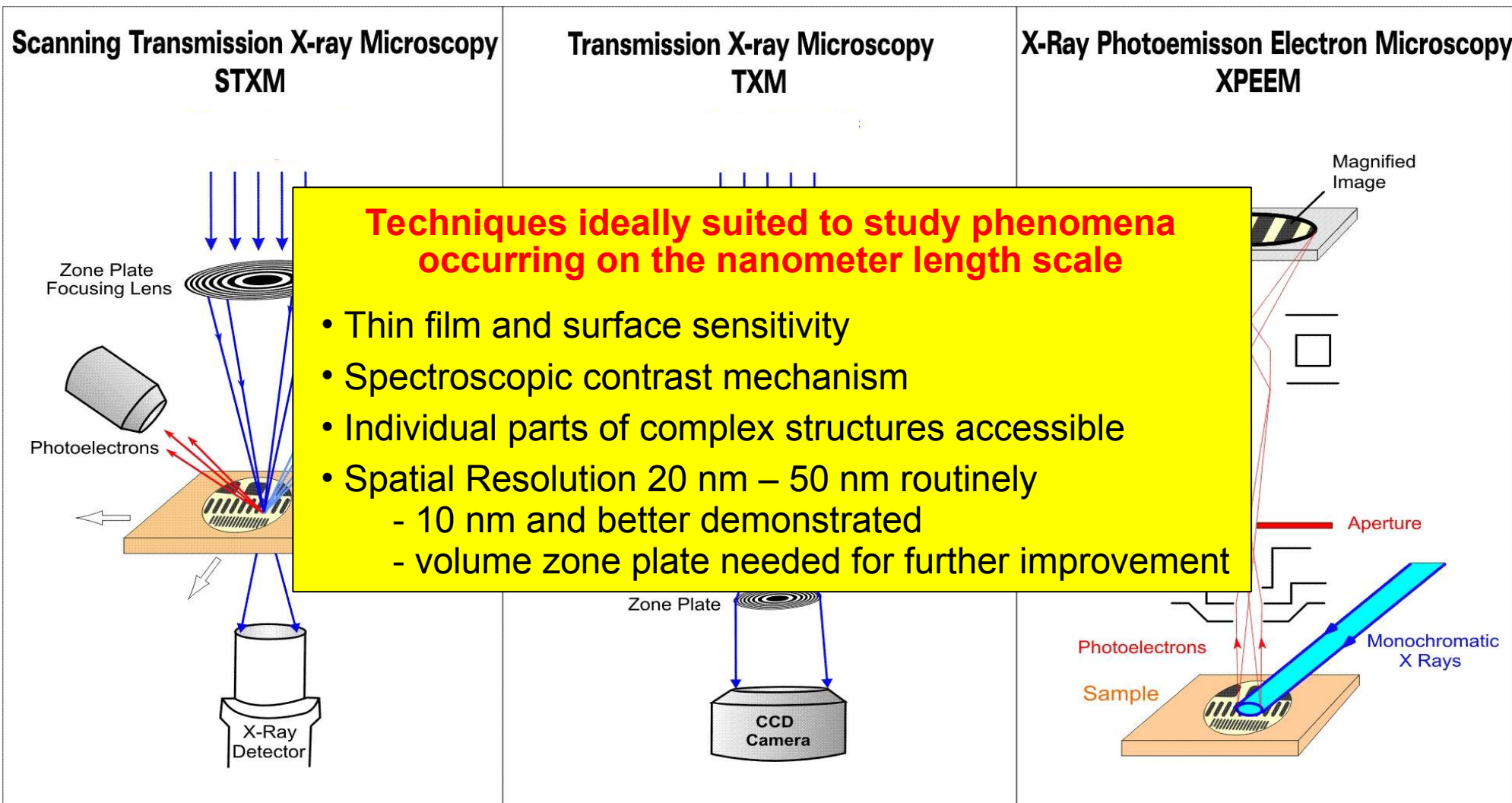
- Best spatial resolution
- Modest spectral resolution
- Shortest exposure time
- Bending magnet radiation
- Higher radiation dose
- Flexible sample environment (wet, cryo, labeled magnetic fields, electric fields, cement, ...)

Scanning Microscope



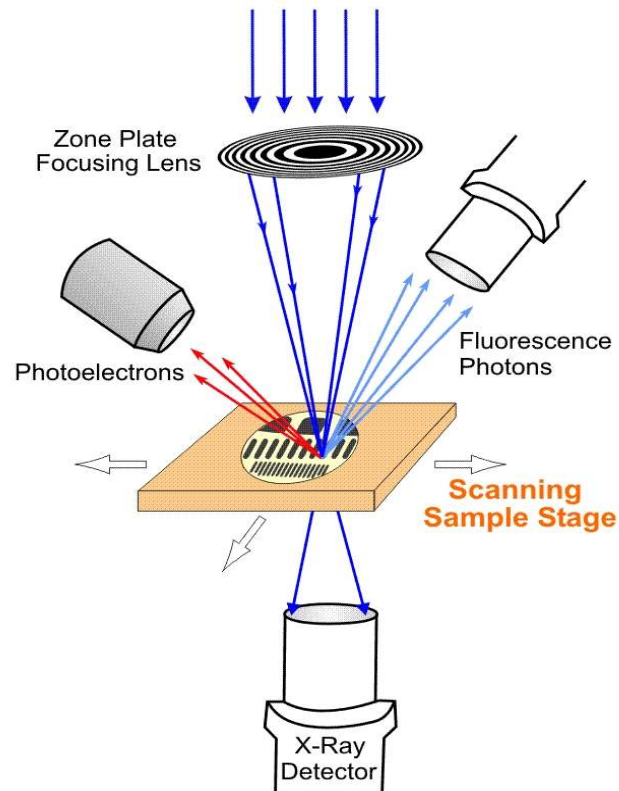
- Least radiation dose
- Next best spatial resolution
- Best spectral resolution
- Requires spatially coherent radiation
- Long exposure time
- Flexible sample environment
- Photoemission (restricted magnetic fields), fluorescence imaging

Quantitative imaging with sensitivity to
elemental and chemical distribution and charge/spin ordering

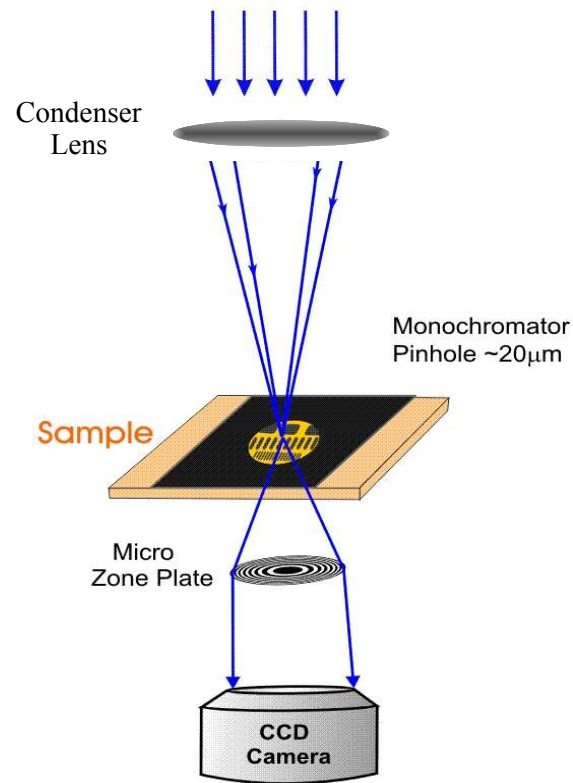


Quantitative imaging with sensitivity to
elemental and chemical distribution and charge/spin ordering

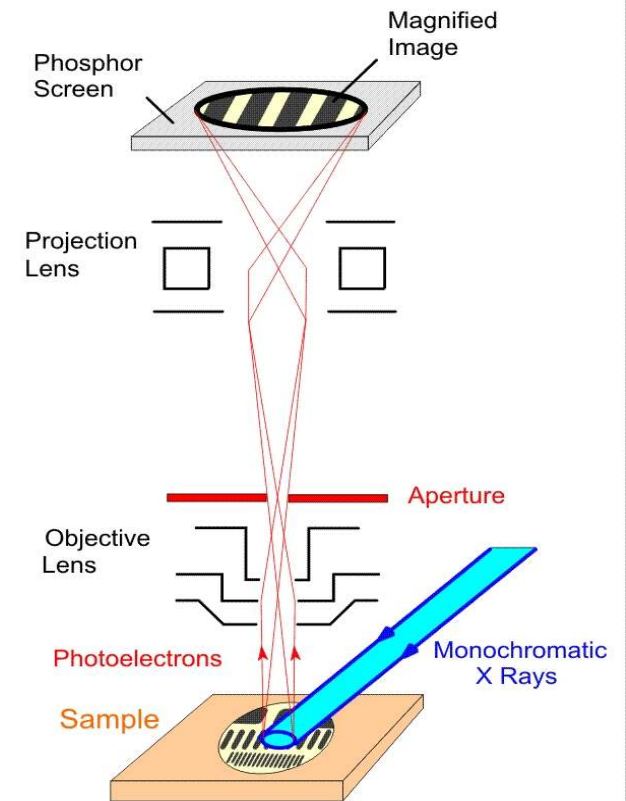
Scanning Transmission X-ray Microscopy STXM



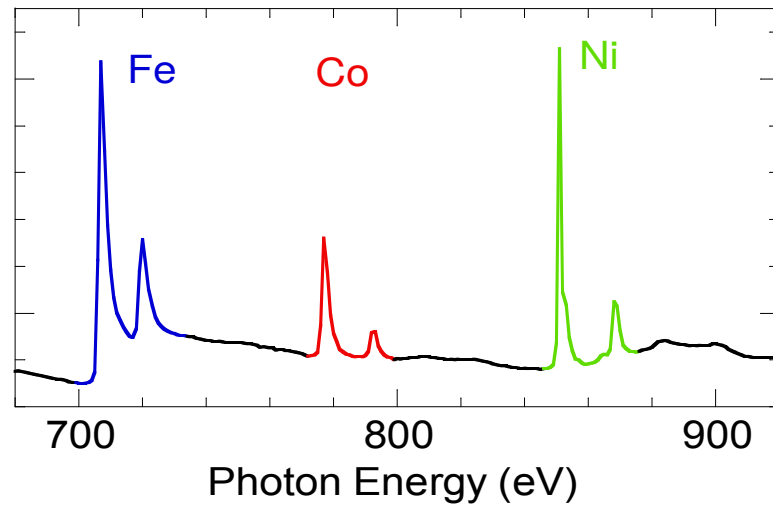
Transmission X-ray Microscopy TXM



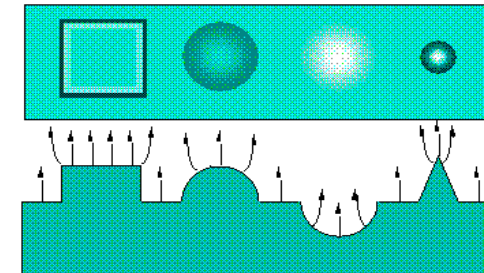
X-Ray Photoemission Electron Microscopy XPEEM



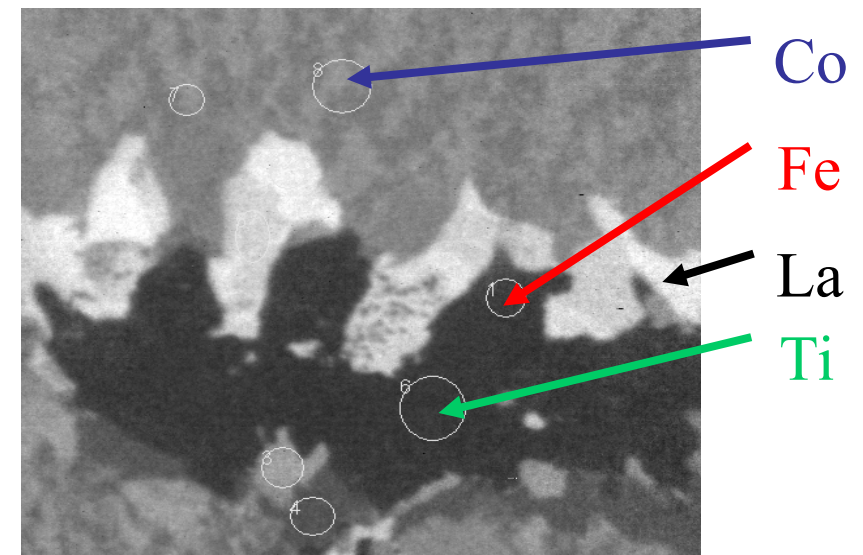
Elemental Specificity



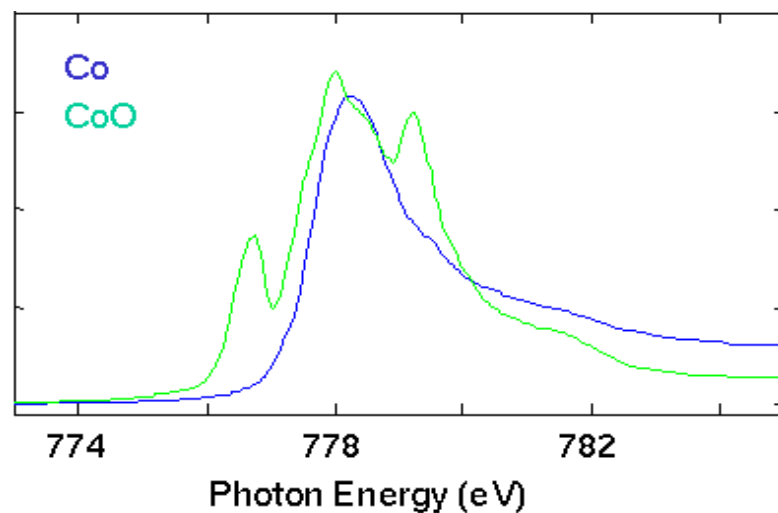
Topographical Contrast



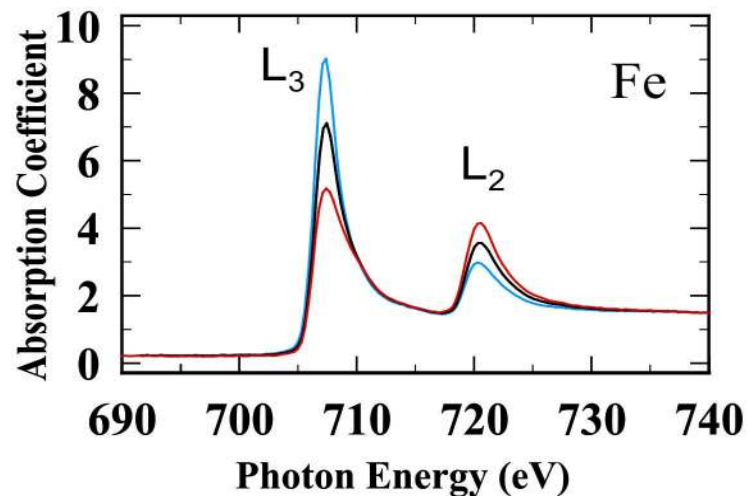
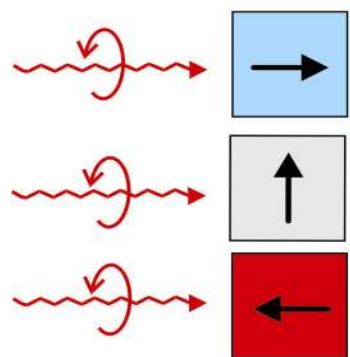
Elemental Contrast



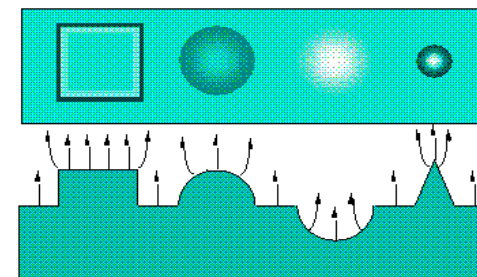
Chemical Sensitivity



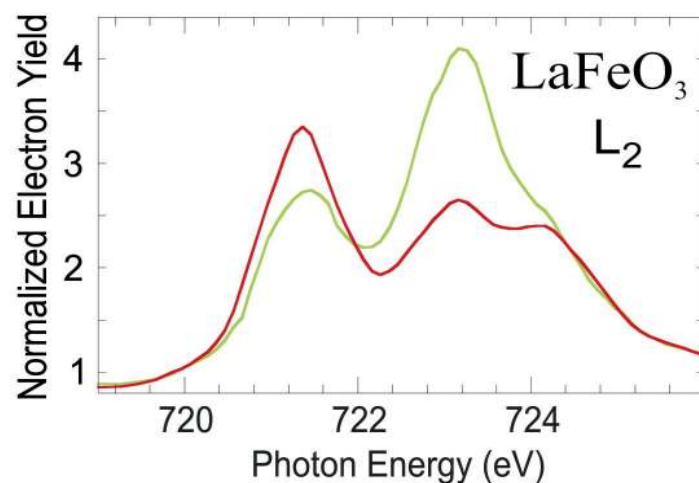
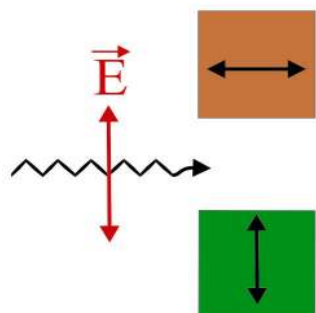
X-ray Magnetic Circular Dichroism: **Ferromagnets**



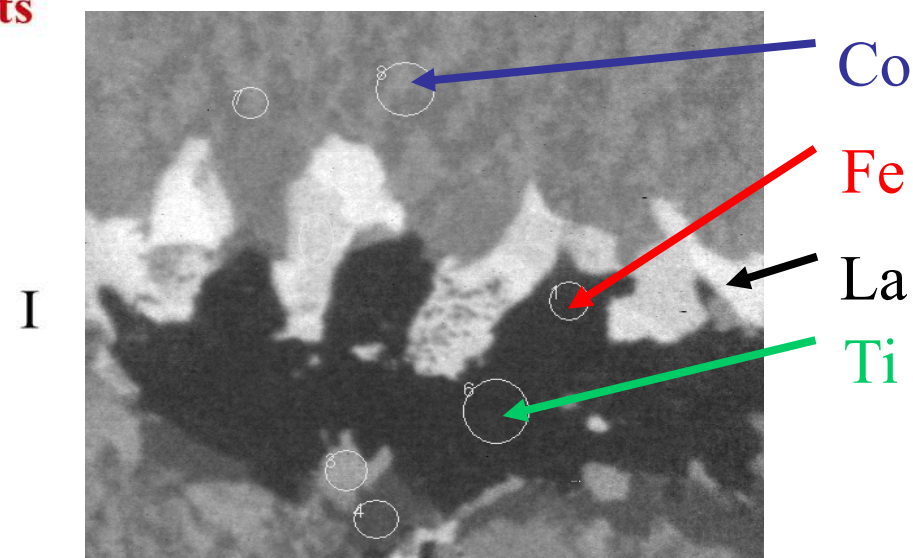
Topographical Contrast

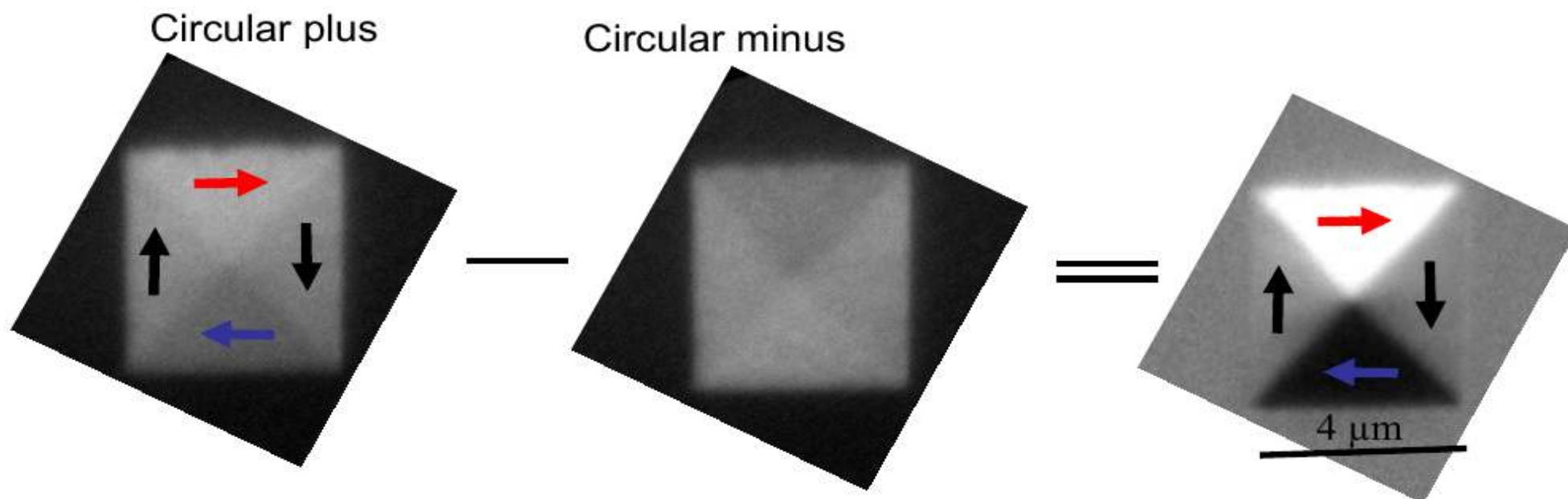
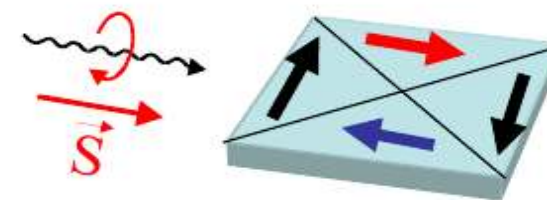
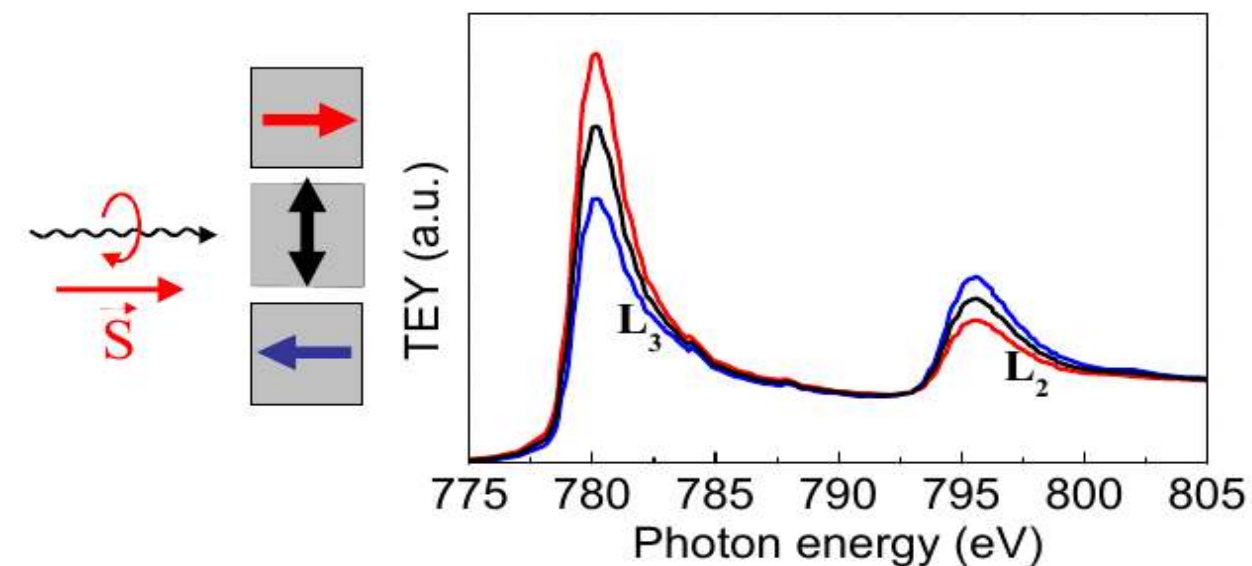


X-ray Magnetic Linear Dichroism: **Antiferromagnets**

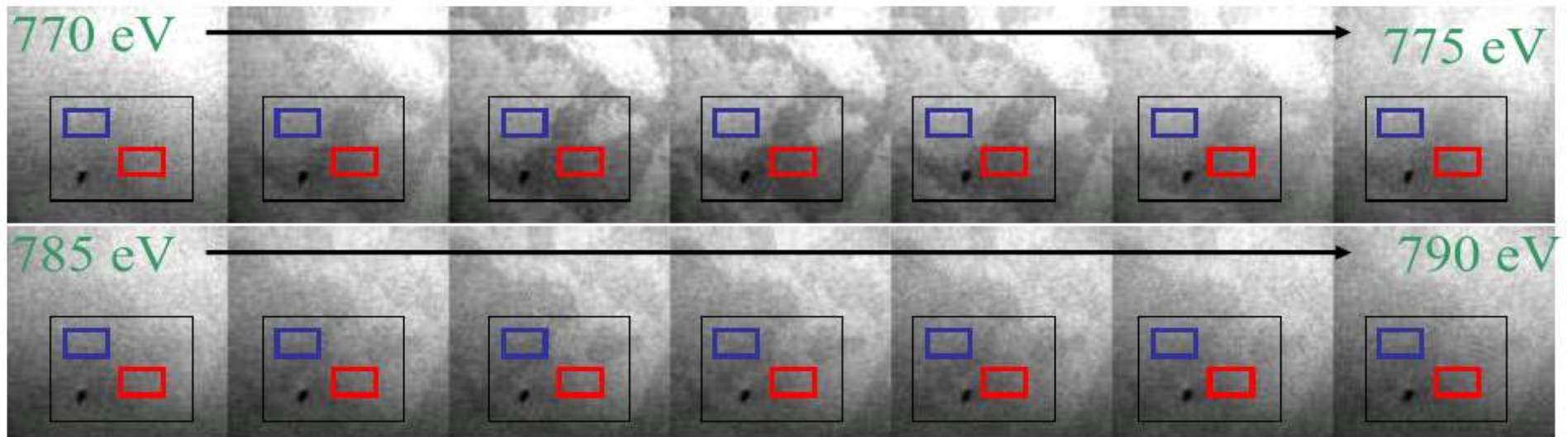
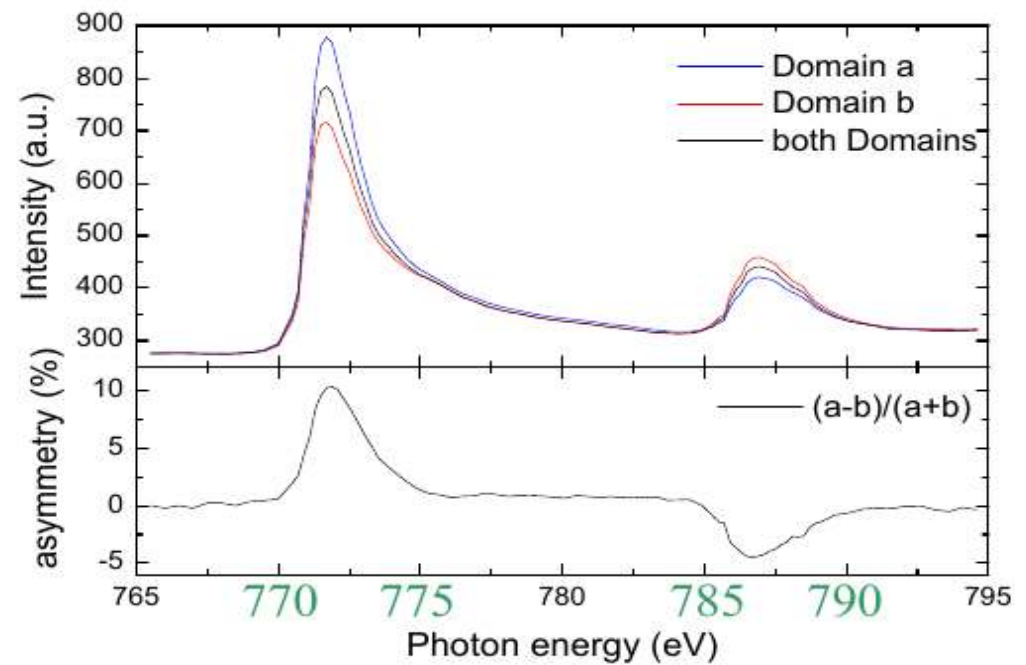
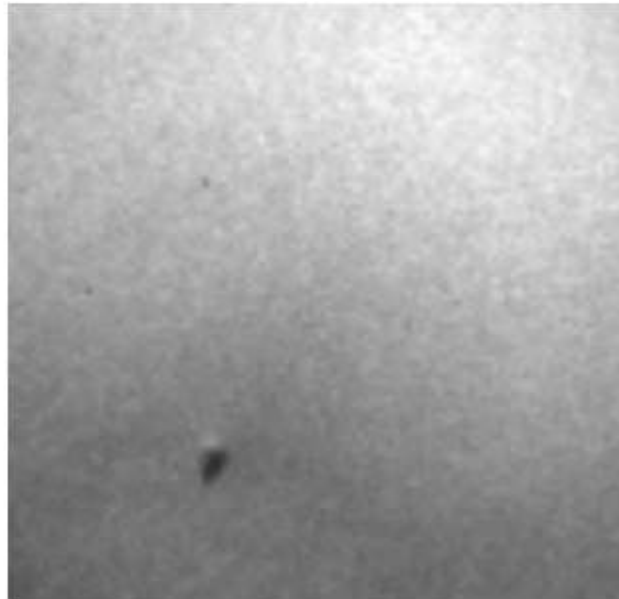


Elemental Contrast



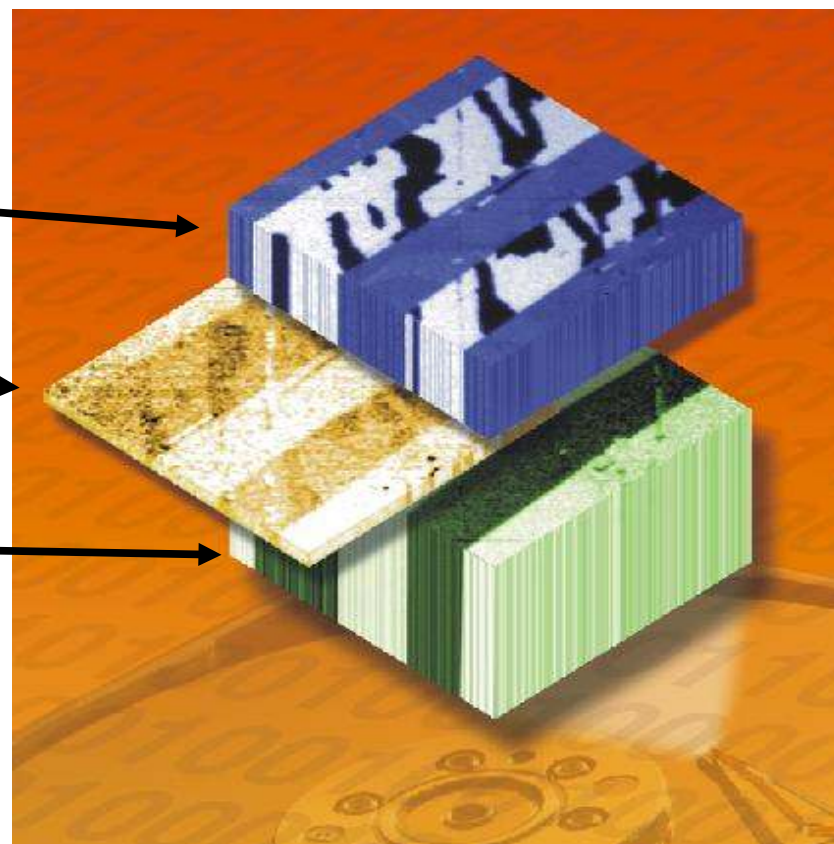
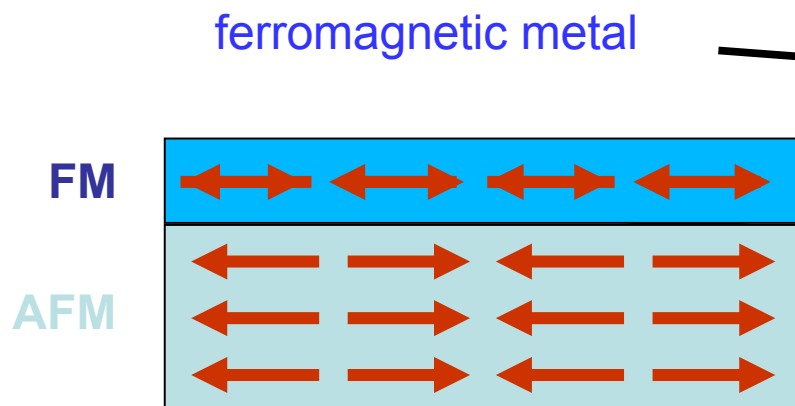


5 μm

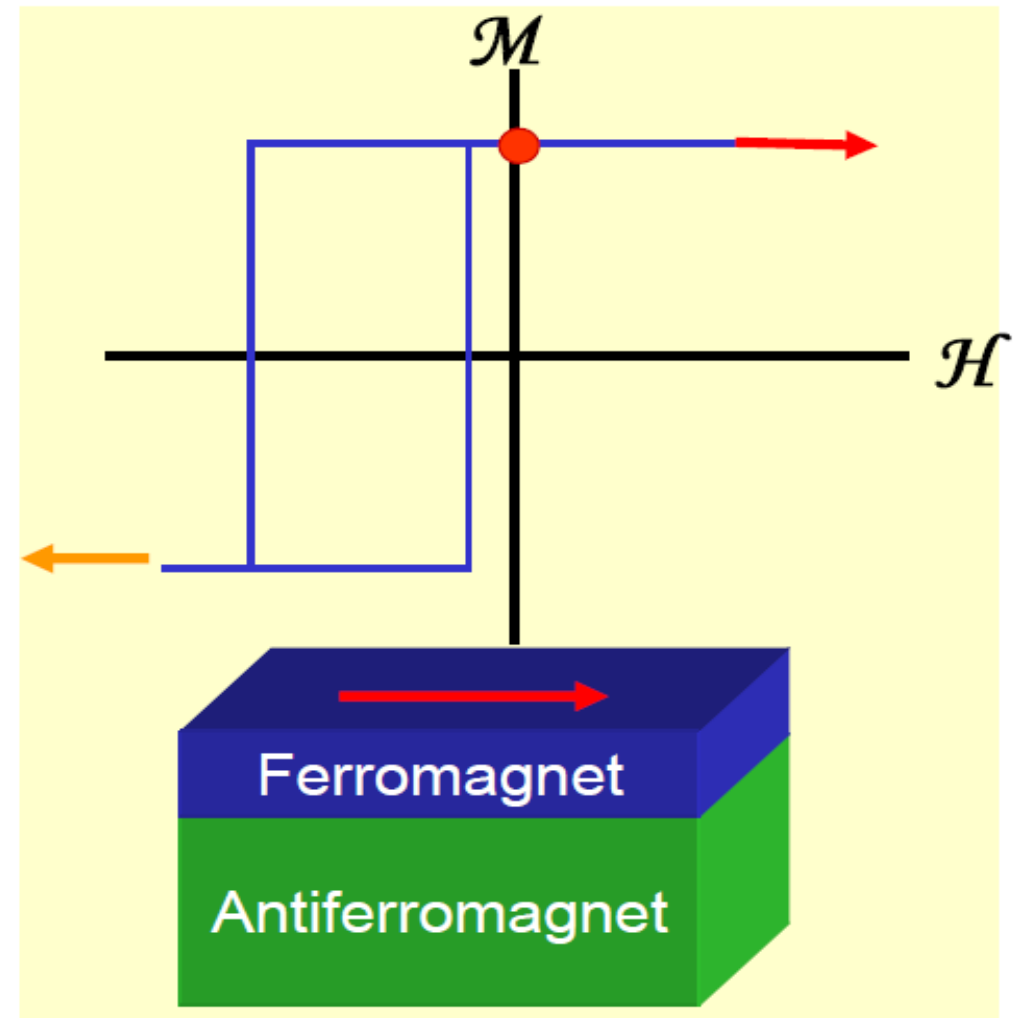
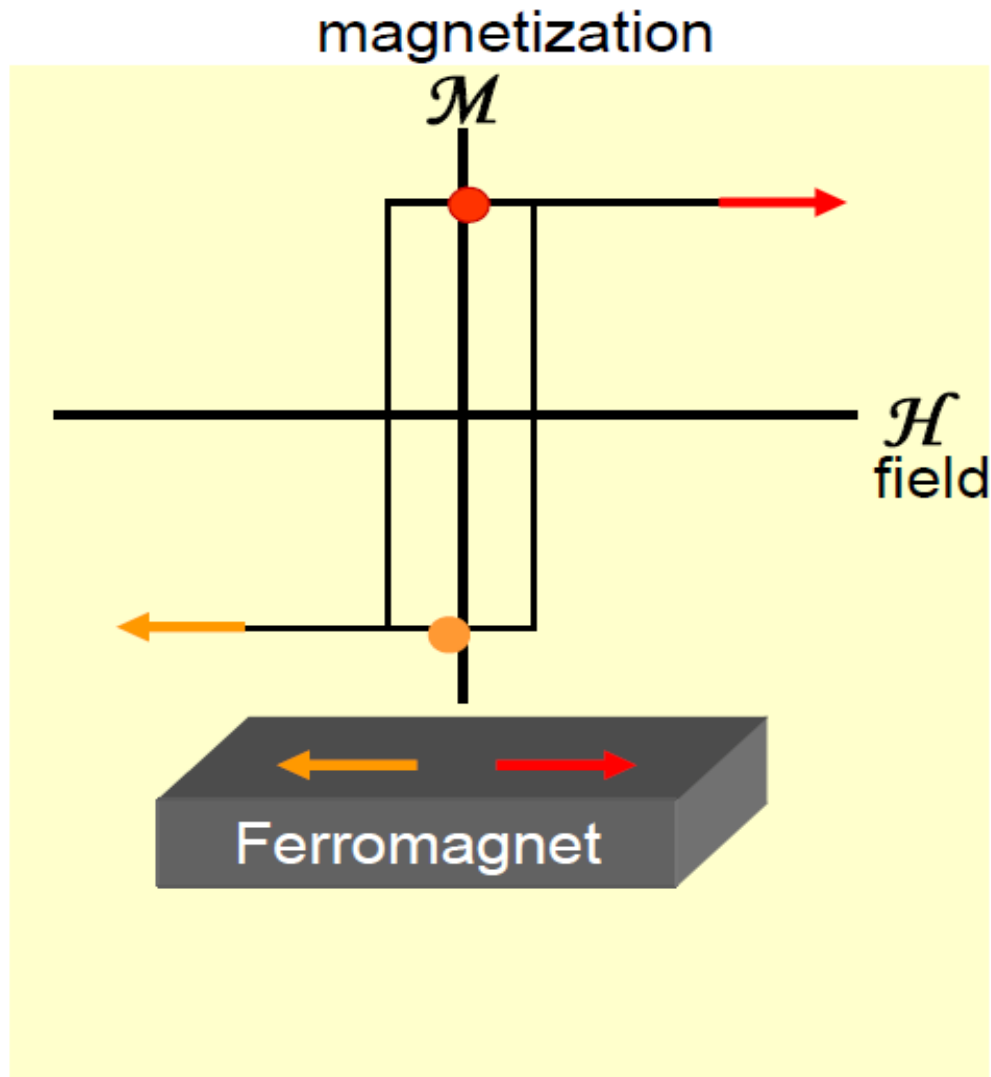


“Exchange bias” magnetic multilayer

Magnetic domain structure in:



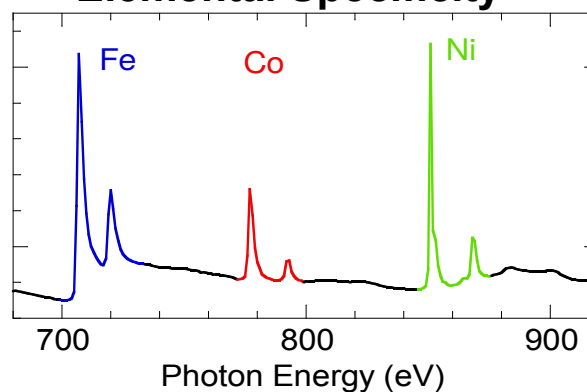
Spectroscopic Identification and Direct Imaging of Interfacial Magnetic Spins
H. Ohldag et al., Phys. Rev. Lett **87**, 247201 (2001).



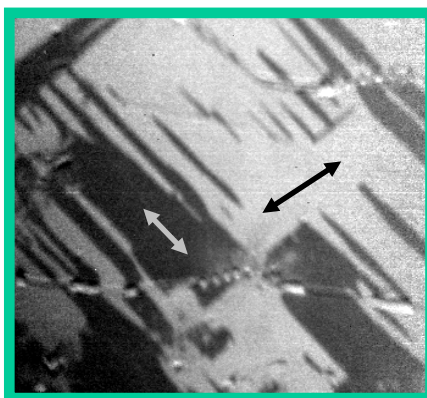
\mathcal{M} of blue layer is “pinned”
or “exchange biased”



Elemental Specificity



NiO XMLD



XMLD for imaging of **antiferromagnetic** spin order in NiO substrate

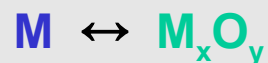
Co XMCD



XMCD for imaging of **ferromagnetic** spin order in Co film

Parallel alignment of spins on both sides of the FM – AFM interface

Chemical environment
influences NEXAFS



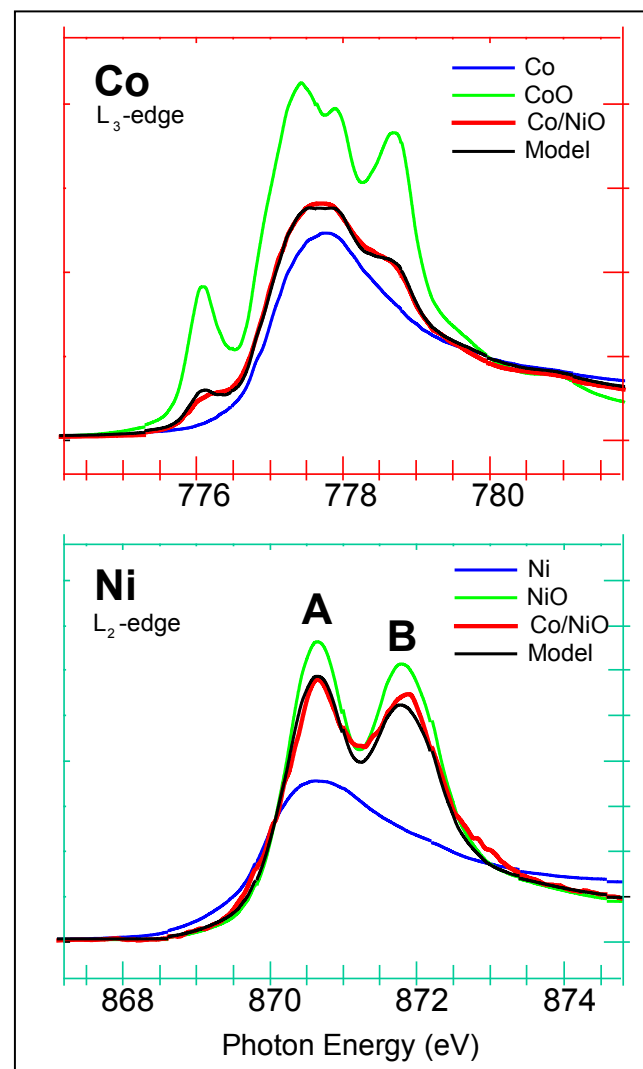
Upon deposition of
2 ML of Co on NiO

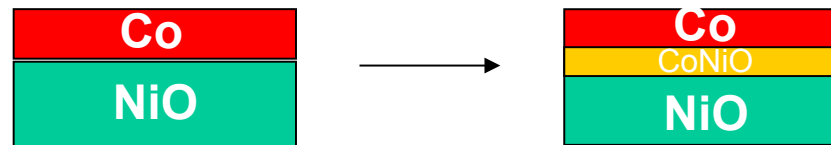
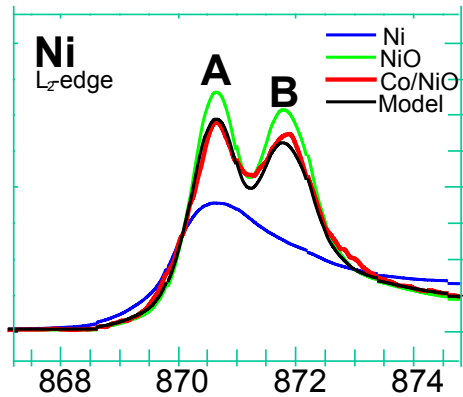
2 ML CoO (Co oxidized)
2 ML Ni (Ni reduced)



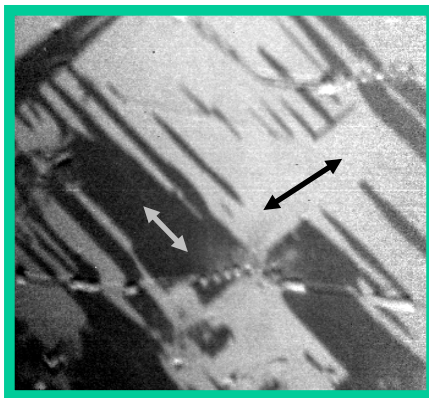
linear combination of metal
and oxide spectra possible

X-ray absorption spectroscopy



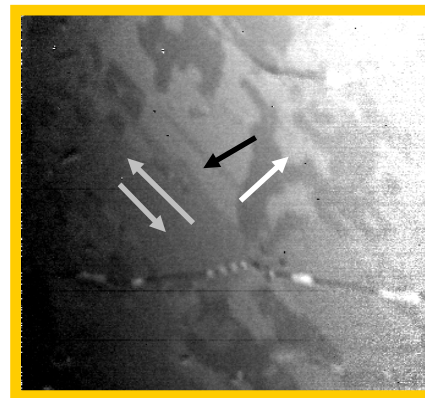


NiO **XMLD**



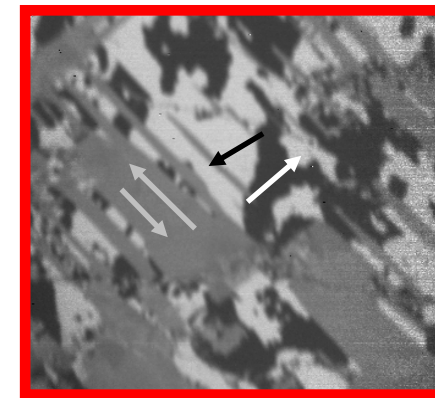
XMLD for imaging of **antiferromagnetic** spin order in NiO substrate

Ni(O) **XMCD**



XMCD for imaging of **ferromagnetic** spin order in NiO substrate

Co **XMCD**



XMCD for imaging of **ferromagnetic** spin order in Co film

Interfacial
chemical
reaction



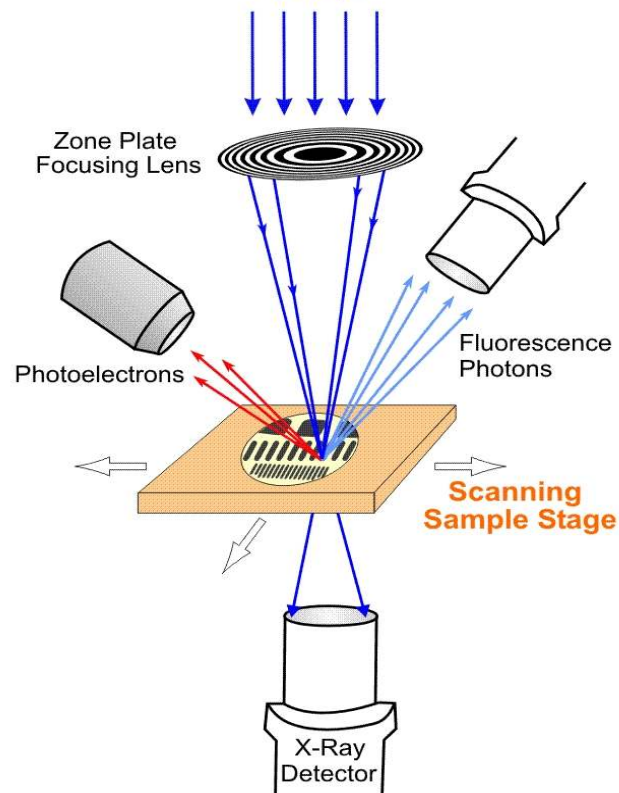
uncompensated
ferromagnetic Ni spins
at interface

Adding **time resolution** to X-ray spectro-microscopy

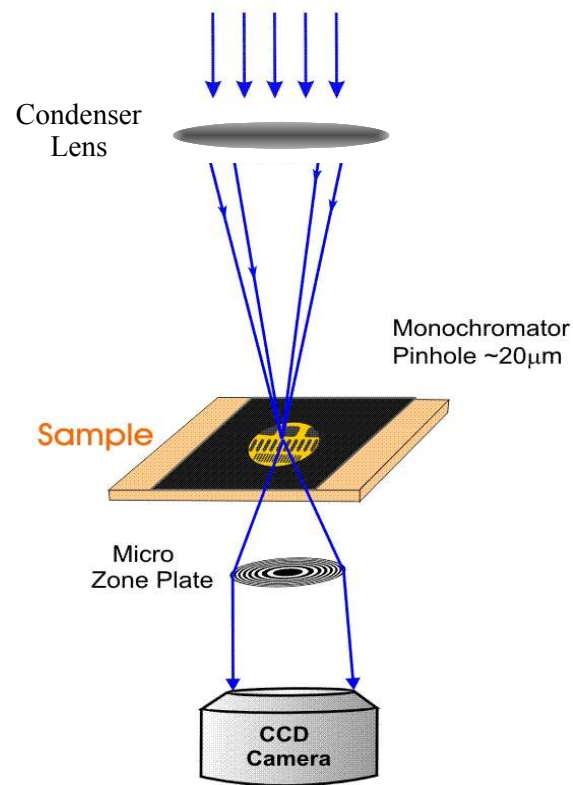
Example: Magnetization switching by spin injection

Quantitative imaging with sensitivity to
elemental and chemical distribution and charge/spin ordering

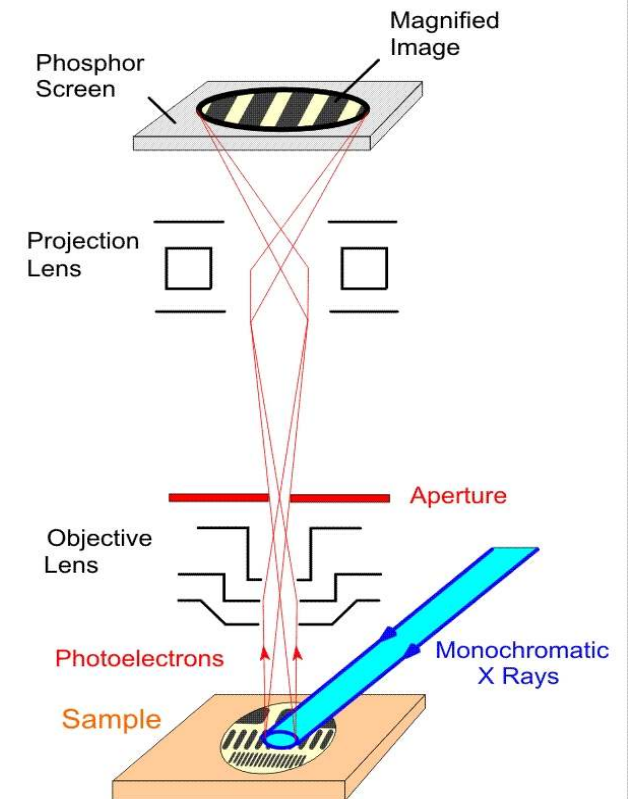
Scanning Transmission X-ray Microscopy STXM



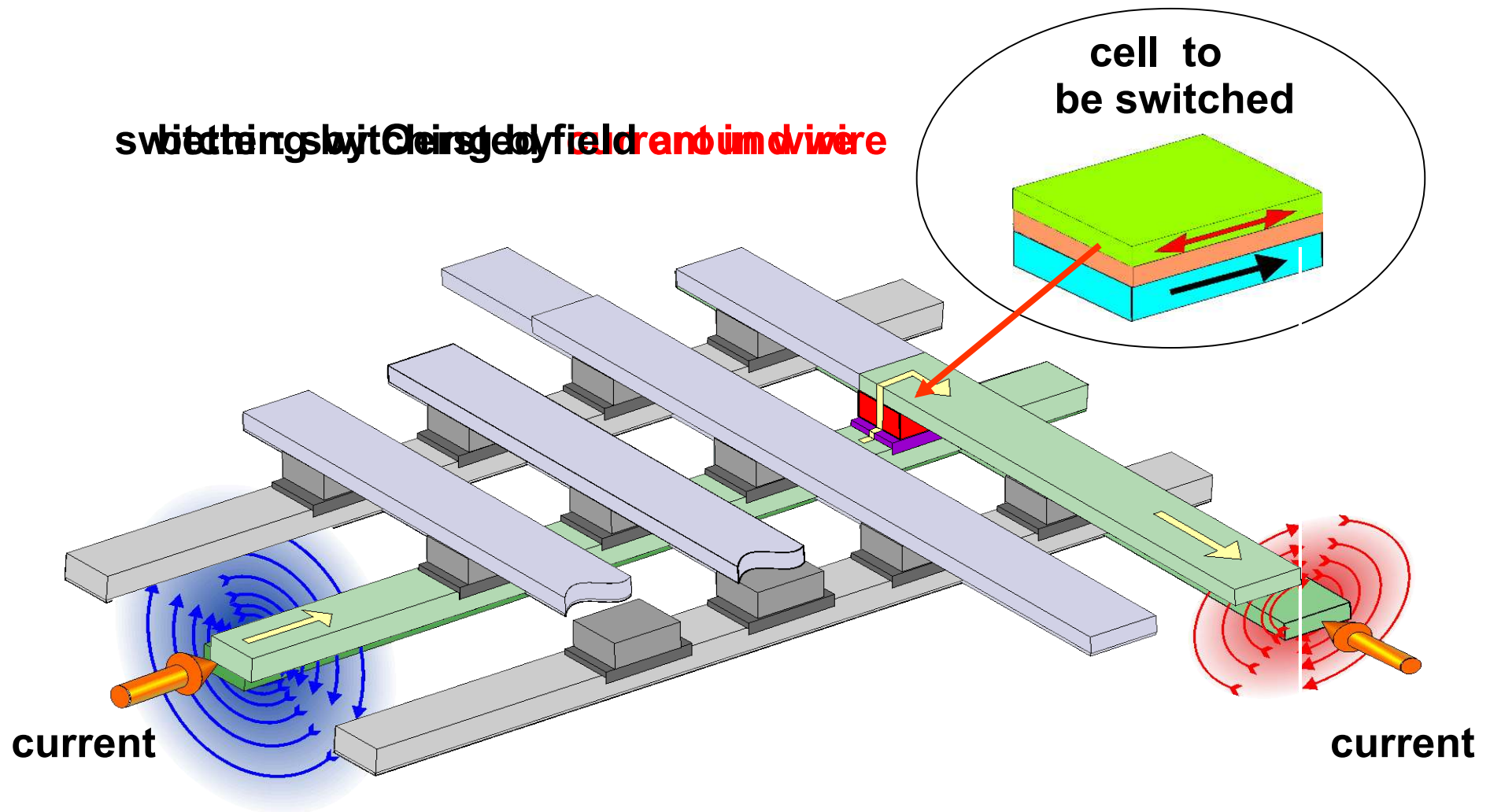
Transmission X-ray Microscopy TXM



X-Ray Photoemission Electron Microscopy XPEEM

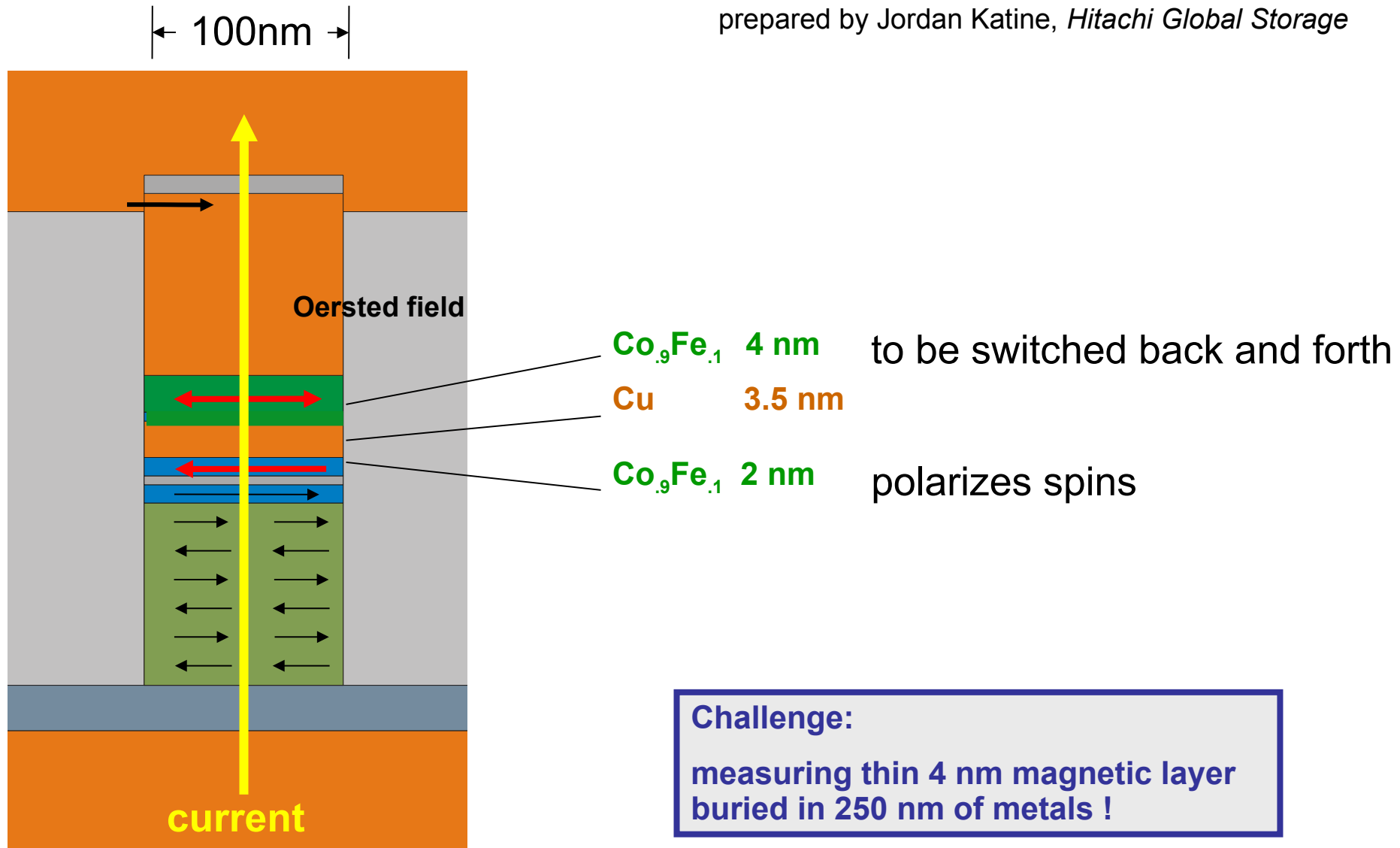


Motivation: Switching of magnetic memory cells (MRAM)

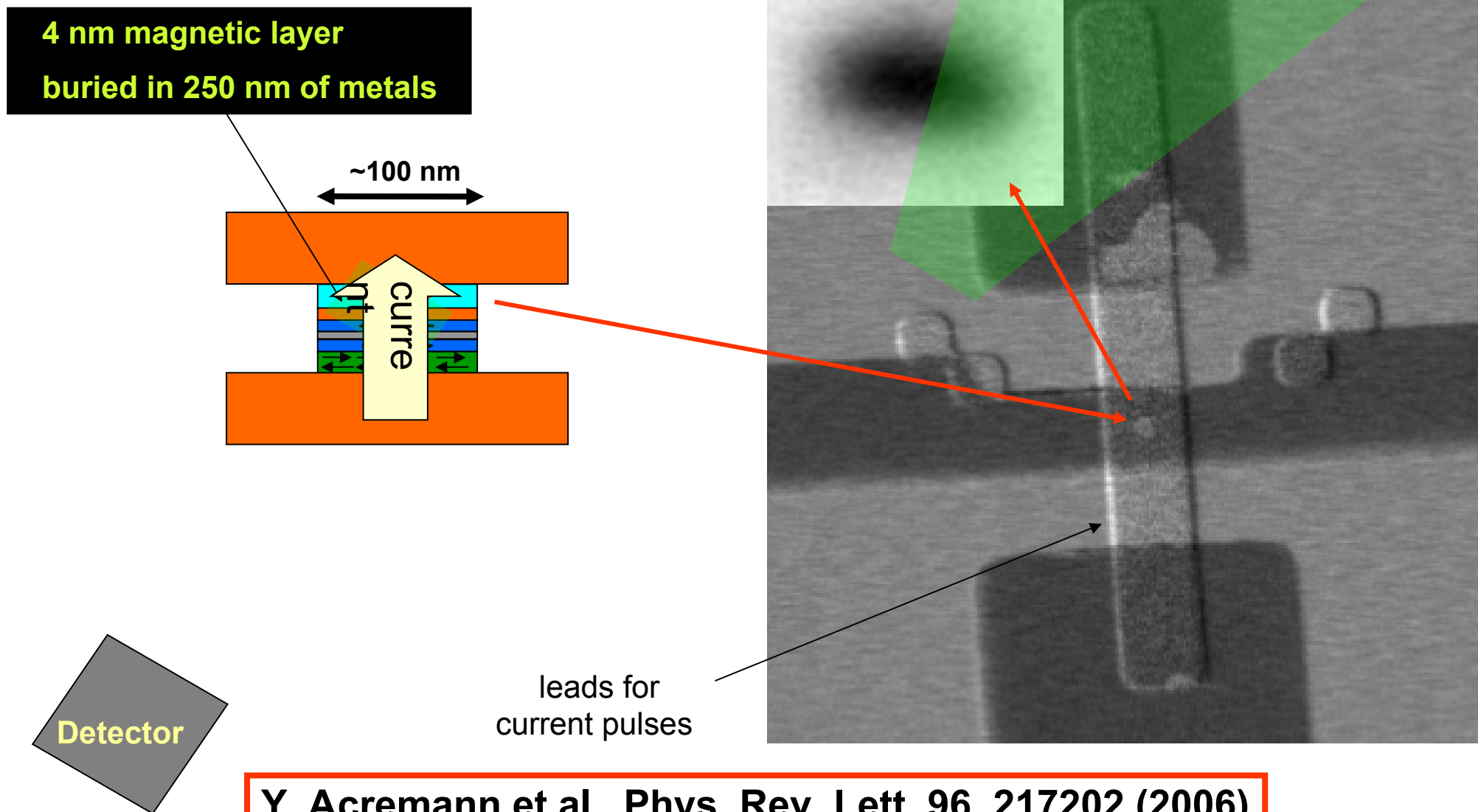


Sample realization for spin-injection experiment

prepared by Jordan Katine, *Hitachi Global Storage*

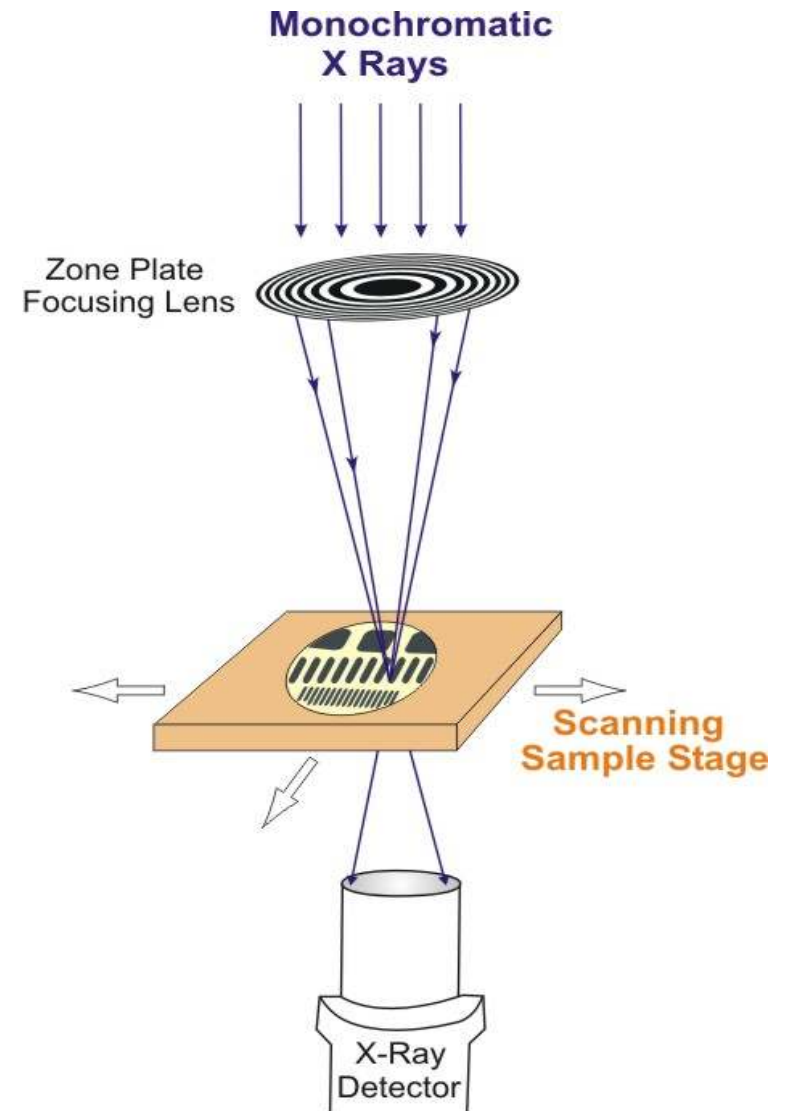


STXM image of spin injection structure

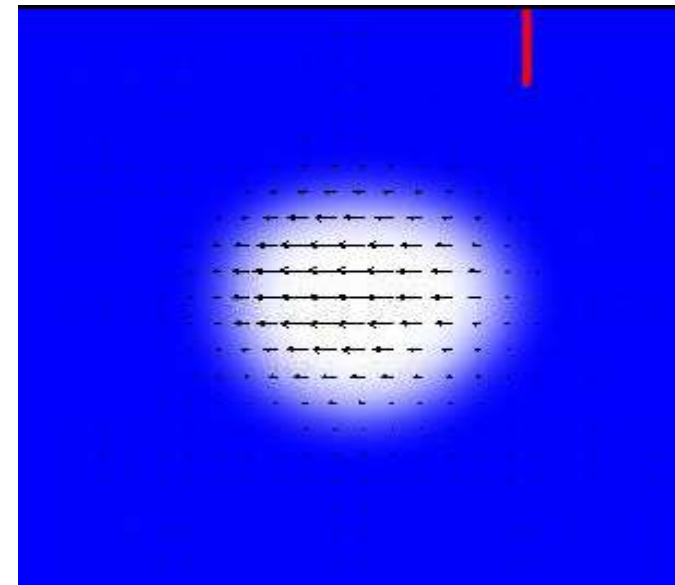
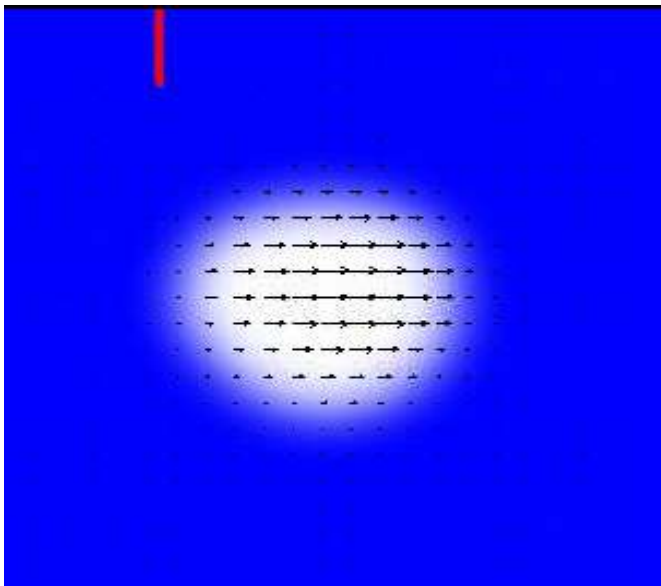


Y. Acremann et al., Phys. Rev. Lett. 96, 217202 (2006)

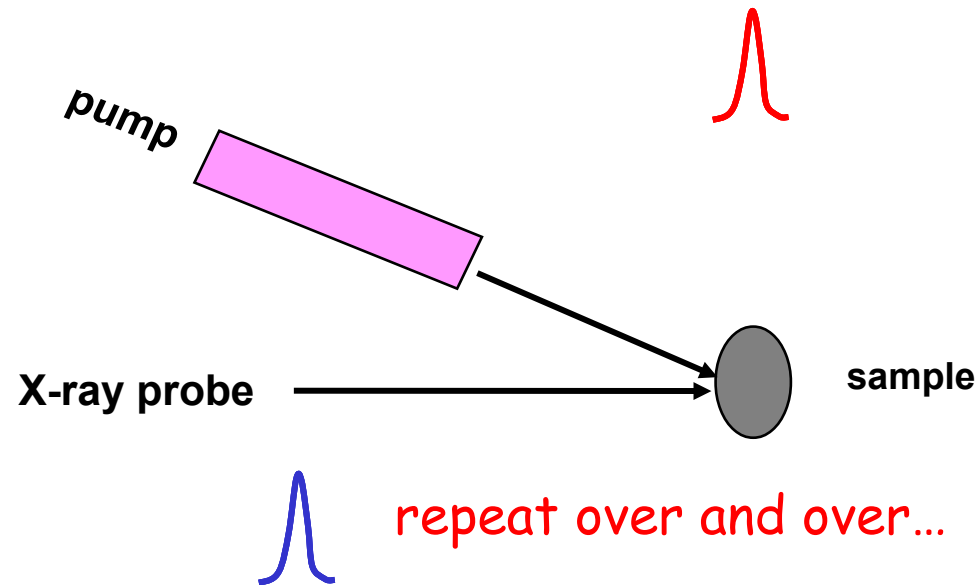
- **Sensitivity to buried thin layer (4 nm)**
Cross section just right - can see signal from thin layer X-rays can distinguish layers, tune energy to Fe, Co, Ni or Cu L edges
- **Resolving nanoscale details (< 100 nm)**
Spatial resolution, x-ray spot size ~ 30 nm
- **Magnetic contrast**
Polarized x-rays provide magnetic contrast (XMCD)
- **Sub-nanosecond timing**
Synchronize spin current pulses with ~ 50 ps x-ray pulses



Fast detector for
X-ray pulse selection



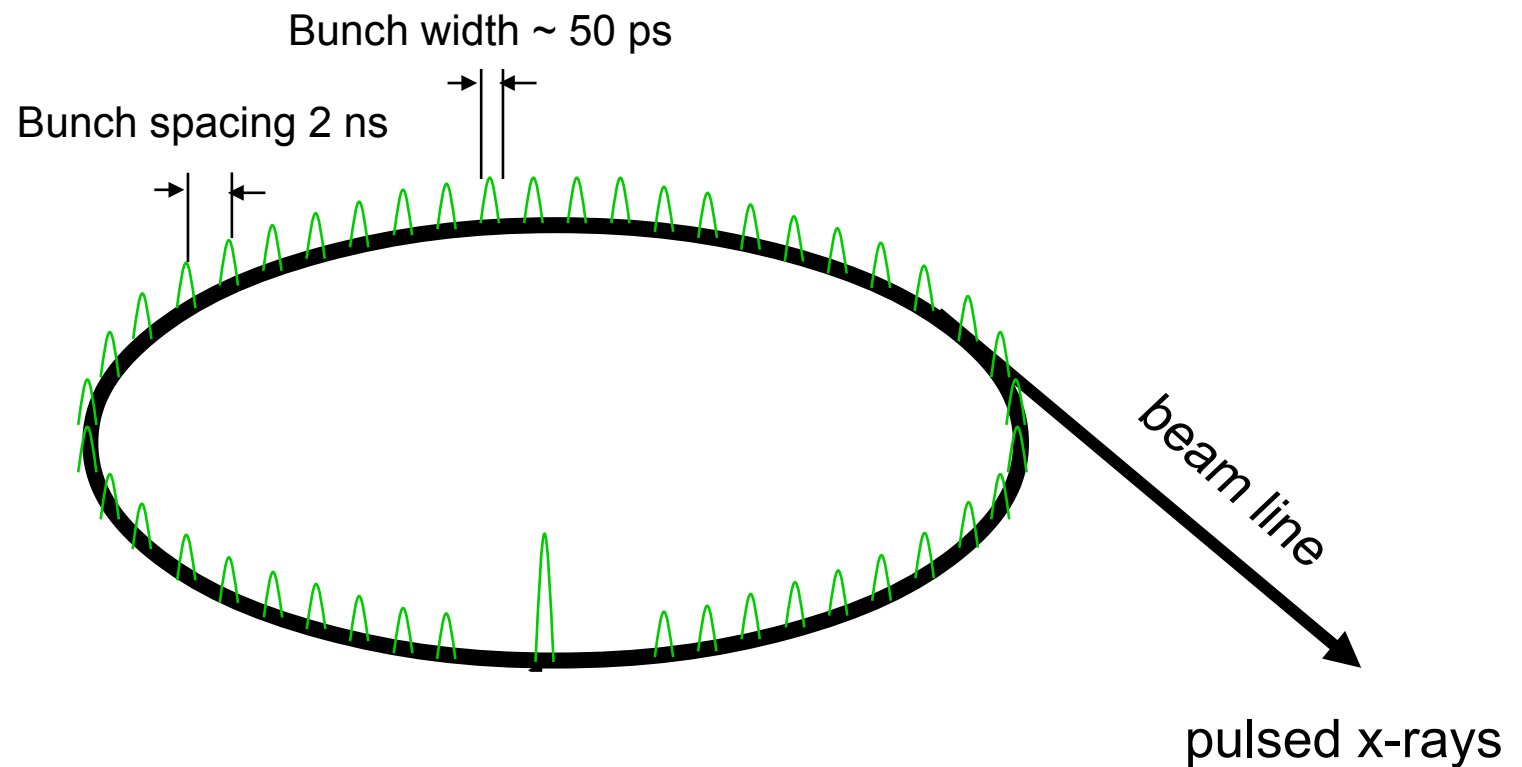
Problem: Today not enough intensity for single shot experiments with nanometer spatial and picosecond time resolution



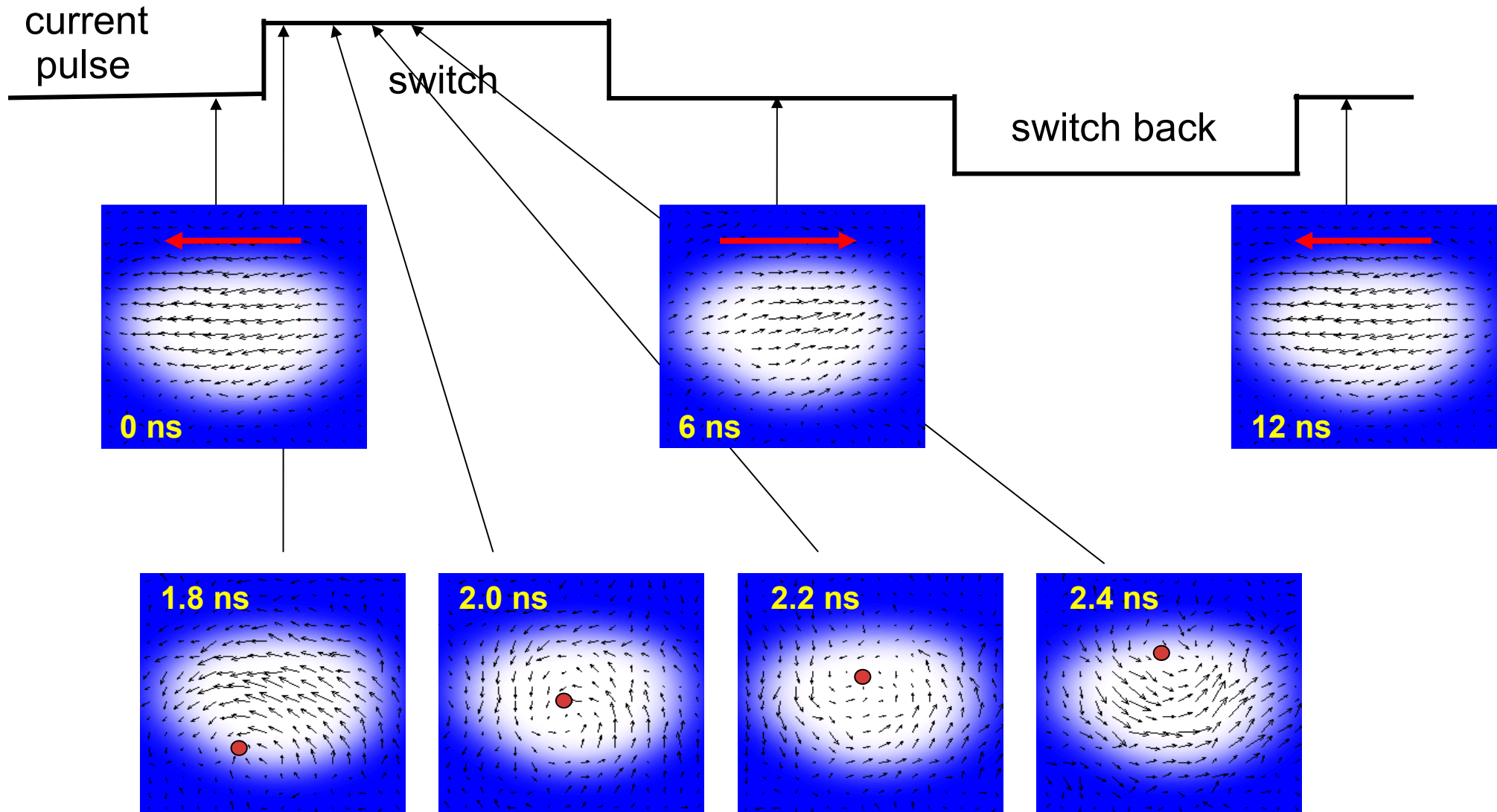
Limitation:

Process has to be repeatable

Storage ring is filled with electron bunches → emission of X-ray pulses



Magnetization reversal dynamics by spin injection



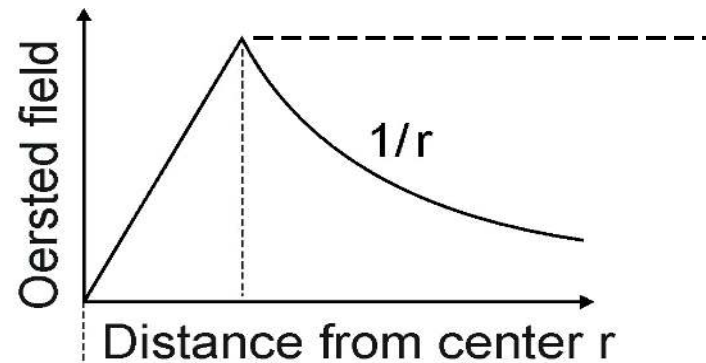
Switching best described by movement of vortex across the sample!

CHARGE CURRENT:

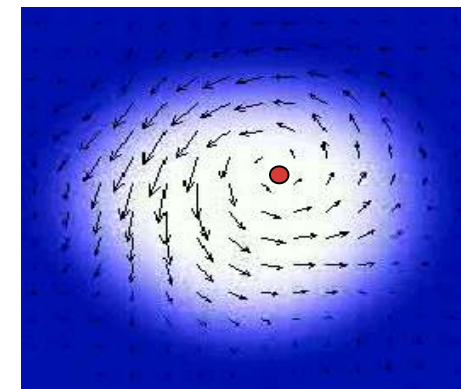
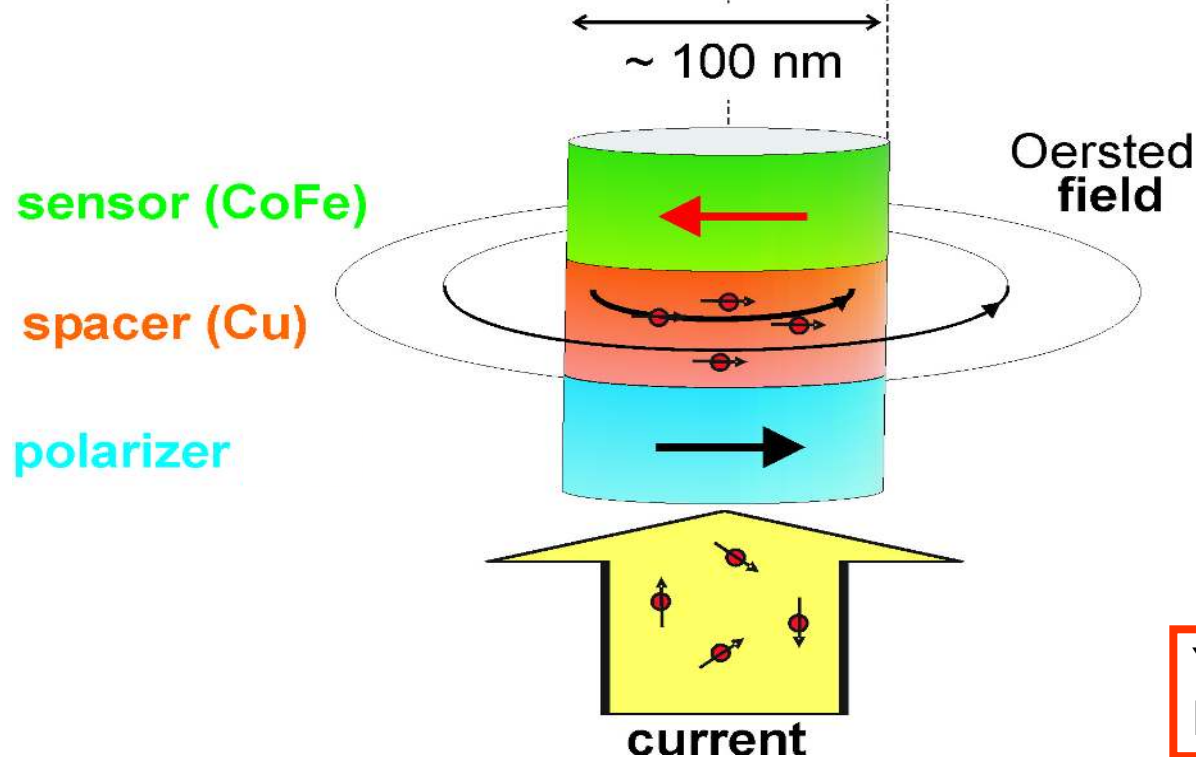
creates vortex state

SPIN CURRENT:

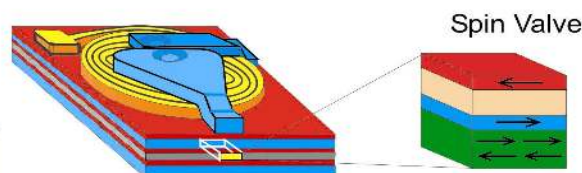
drives vortex across sample



= 950 Oersted
for $150 \times 100 \text{ nm}$,
 $j = 2 \times 10^8 \text{ A/cm}^2$



Y. Acremann et al.,
Phys. Rev. Lett. 96, 217202 (2006)



General requirements:

distinguish components

study thin films and interfaces

look below the surface

see the invisible

resolve dynamic motions

Technique requirements:

elemental (chemical) specificity

large cross section for “signal”

depth sensitivity

nanoscale spatial resolution

time resolution < 1 nanosecond

separate spin and orbital contributions **sensitive to s-o coupling**

x-rays cover them all

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

But:

Never ignore the power of other experimental techniques, because:

- *Good* argument: Each technique has specific strengths
- *Good but dangerous* argument: More readily accessible for you