Methoden moderner Röntgenphysik II: Streuung und Abbildung

Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2019

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Location Lecture hall INF, Physics, Jungiusstraße 11

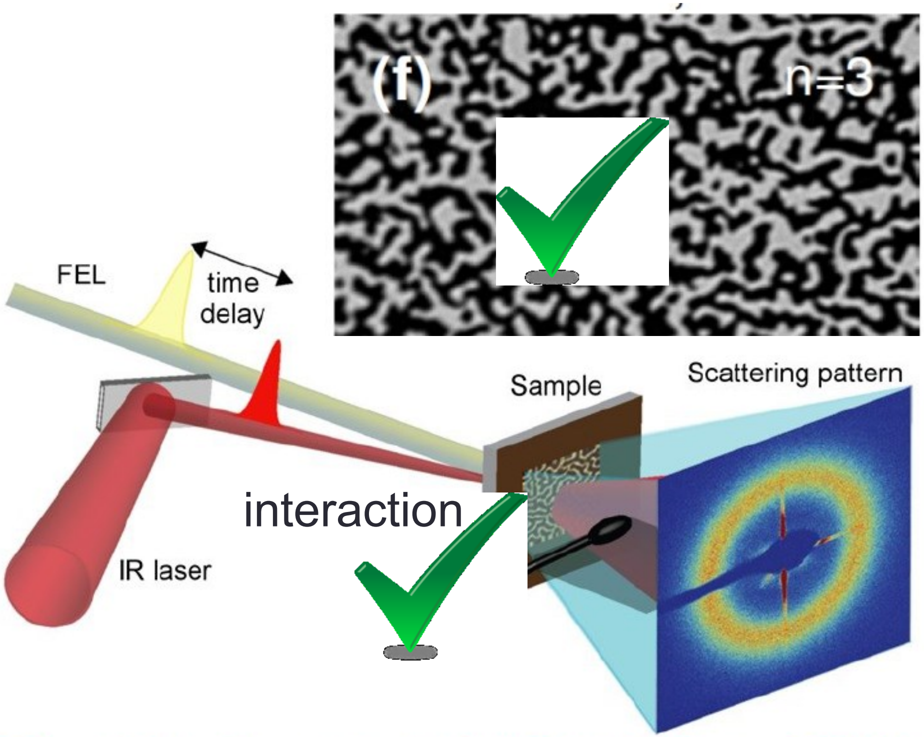
Time Tuesday 12:30 - 14:30
Thursday 8:30 - 10:00

Outline

Part II/1:

Studies on Magnetic Nanostructures

by Leonard Müller



B. Pfau et al., Nature Communications, Vol. 3, 11; DOI:doi:10.1038/ncomms2108 (2012)
L.Müller et al., Rev. Sci. Instrum. 84, 013906 (2013)

[15.5.] **Ferromagnetism in a Nutshell**

- Introduction to Magnetic Materials
- Magnetic Phenomena
- Magnetic Free Energy
- Perpendicular Magnetic Anisotropy
- Magnetic Domains and Domain Walls

[17.5.] **Interaction of Polarized Photons with Ferromagnetic Materials**

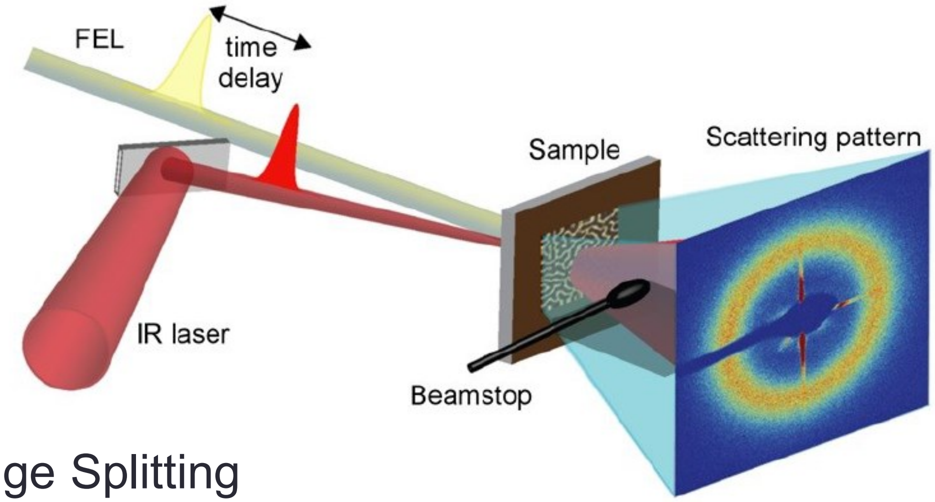
- Charge and Spin X-ray Scattering by a Single Electron
- Absorption and Resonant Scattering of Ferromagnets (Semi-Classical and Quantum-Mechanical Concepts)

Outline

Part II/2:

Studies on Magnetic Nanostructures

by Leonard Müller



[6.6.] **X-ray Magnetic Circular Dichroism (XMCD) & Resonant Magnetic Small Angle X-ray Scattering (mSAXS)**

- Role of Spin-Orbit Coupling and Exchange Splitting
- Sum Rules
- XMLD and Natural Dichroisms
- mSAXS of Magnetic Domain Patterns

Interaction of **polarized** photons with matter

> Absorption & Resonant scattering (qm concept, Fermi's Golden rule)

- Time-dependent pertubation theory (up to second order) = „Fermi's Golden rule"

$$T_{if} = \frac{2\pi}{\hbar} \left| \langle f|\mathcal{H}_{\text{int}}|i\rangle + \sum_n \frac{\langle f|\mathcal{H}_{\text{int}}|n\rangle\langle n|\mathcal{H}_{\text{int}}|i\rangle}{\varepsilon_i - \varepsilon_n} \right|^2 \delta(\varepsilon_i - \varepsilon_f)\rho(\varepsilon_f)$$

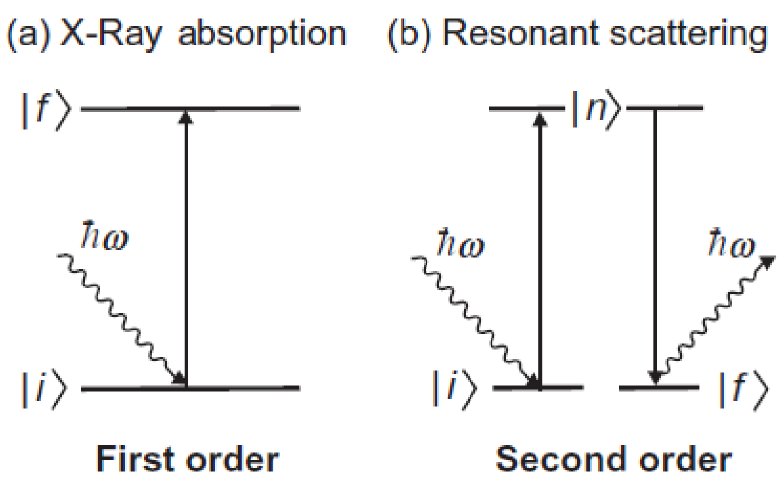
T_{if}: transition rate from state i to f; $[T_{if}] = s^{-1}$; i and f are initial and final states of the combined electron and photon system

$\rho(\varepsilon_f)$: density of final states

ε_n: energy of all possible intermediate states n

- Total cross-section given by $\sigma = \frac{T_{if}}{\Phi_0}$

Incident photon flux



Interaction of **polarized** photons with matter

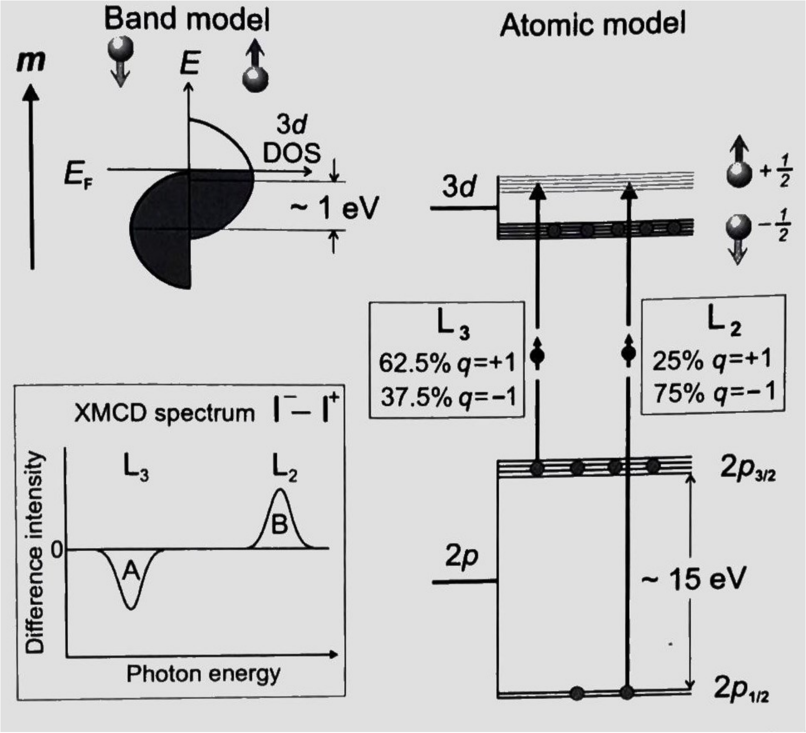
> X-ray magnetic circular dicroism (XMCD) effect

- Strong ferromagnet: one subband is completely filled
- Spin in conserved during transition
- Weak spin-orbit interaction ignored

➢ $m = 5\mu_B$ per atom
➢ Spin down electrons cannot be excited

➔ XMCD: $\Delta I = I^{\uparrow\downarrow} - I^{\uparrow\uparrow} \neq 0$

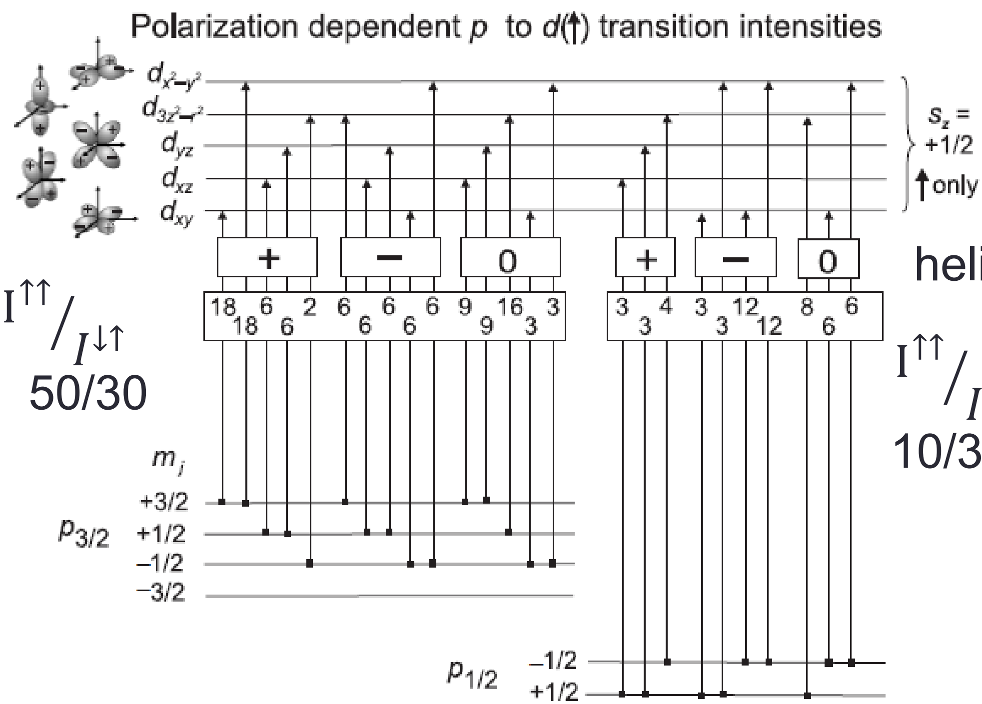
➔Calculate transition matrix elements for **Spin-Up** electrons & helicity q = ± 1 (RCP and LCP)
Sum over all possible transitions…



Interaction of **polarized** photons with matter

> X-ray magnetic circular dicroism (XMCD) effect

Crystal-field-split-d-states



$I^{\uparrow\uparrow}/I^{\downarrow\uparrow}$

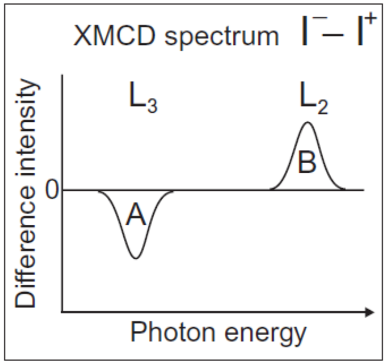
50/30

helicity

$I^{\uparrow\uparrow}/I^{\downarrow\uparrow}$

10/30

In this special case, L3 and L2 edges have the same ΔI



$$\frac{I_{L3}}{I_{L2}} = \frac{80}{40} = 2:1$$

$$\Delta I_{L_3} = \mathcal{A}R^2 \sum_{n,m_j} |\langle d_n,\chi^+|C_{-1}^{(1)}|p_{3/2},m_j\rangle|^2 - |\langle d_n,\chi^+|C_{+1}^{(1)}|p_{3/2},m_j\rangle|^2 \quad = -\frac{2}{9}AR^2$$

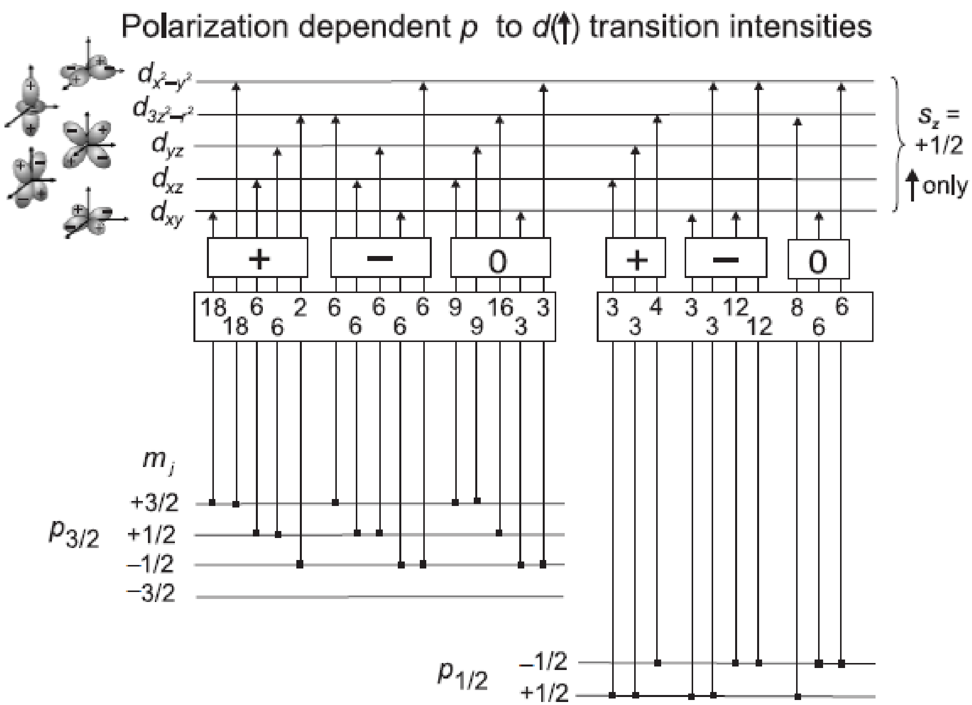
$$\Delta I_{L_2} = \mathcal{A}R^2 \sum_{n,m_j} |\langle d_n,\chi^+|C_{-1}^{(1)}|p_{1/2},m_j\rangle|^2 - |\langle d_n,\chi^+|C_{+1}^{(1)}|p_{1/2},m_j\rangle|^2 \quad = +\frac{2}{9}AR^2$$

$A = 4\pi^2\alpha_f\hbar\omega$

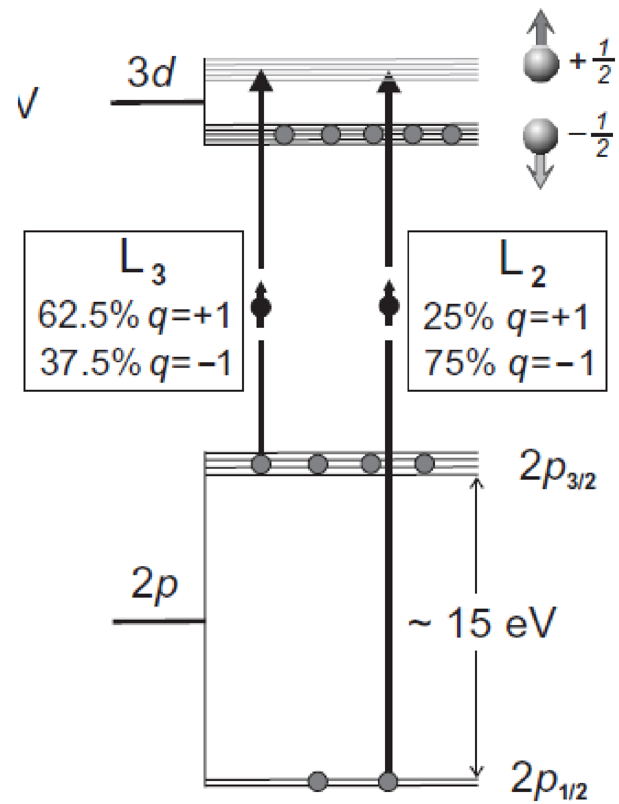
R =radial matrix element

Interaction of **polarized** photons with matter

> X-ray magnetic circular dicroism (XMCD) effect



Crystal-field-split d-states



Atomic-exchange-split d-states (w/o SOC)

Same results for $I_{L_{3,total}} : I_{L_{2,total}} = 2:1,$

$$\Delta I_{L_{3,total}} : \Delta I_{L_{2,total}} = 1:-1$$

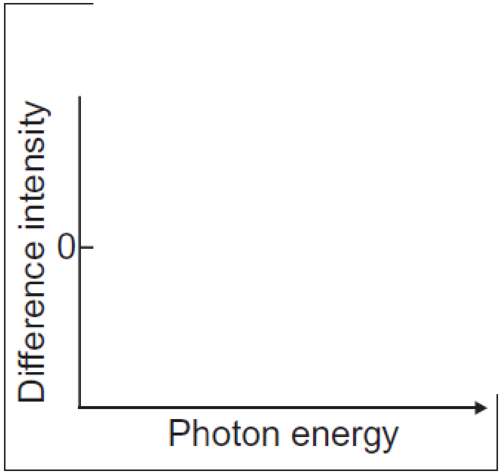
when using atomic d-states (w/o SOC); today's lecture

DESY

Interaction of **polarized** photons with matter

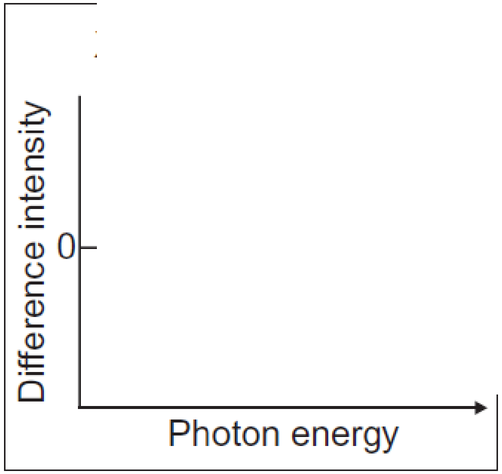
> X-ray magnetic circular dicroism (XMCD) effect

What is happening in a paramagnet?



→ No XMCD

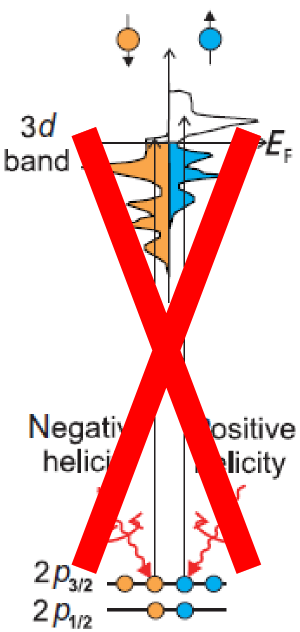
What is happening w/o Spin-Orbit-Coupling for the p-states?

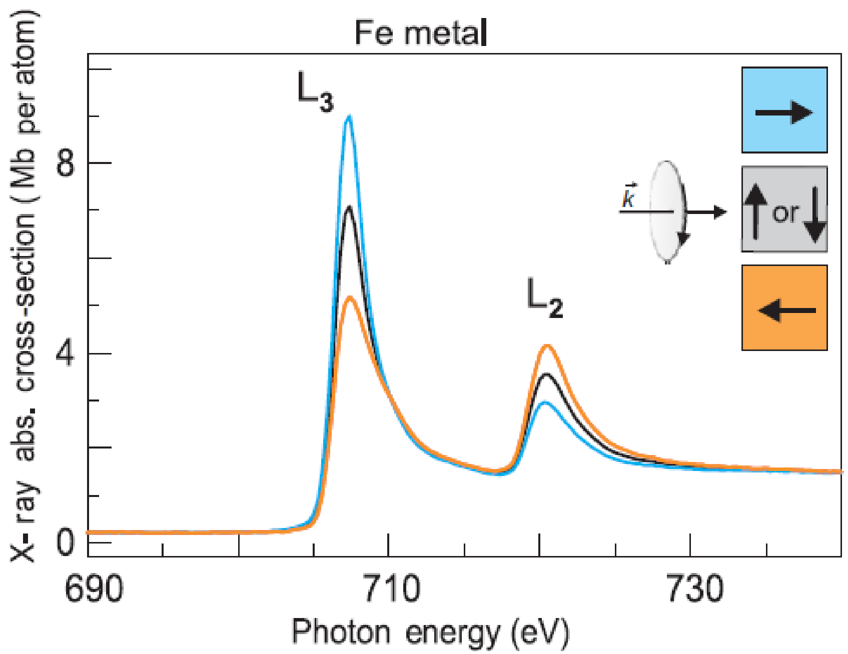


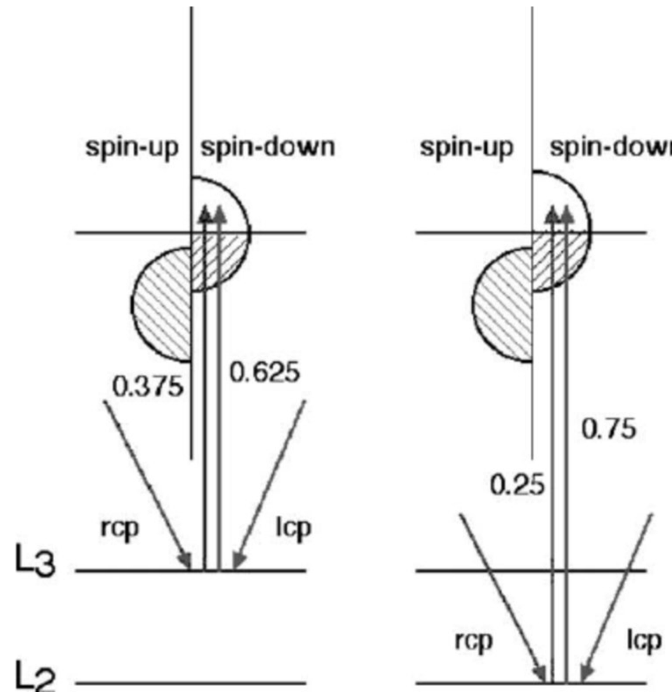
→ No XMCD

Interaction of **polarized** photons with matter

- X-ray magnetic circular dichroism (XMCD) effect







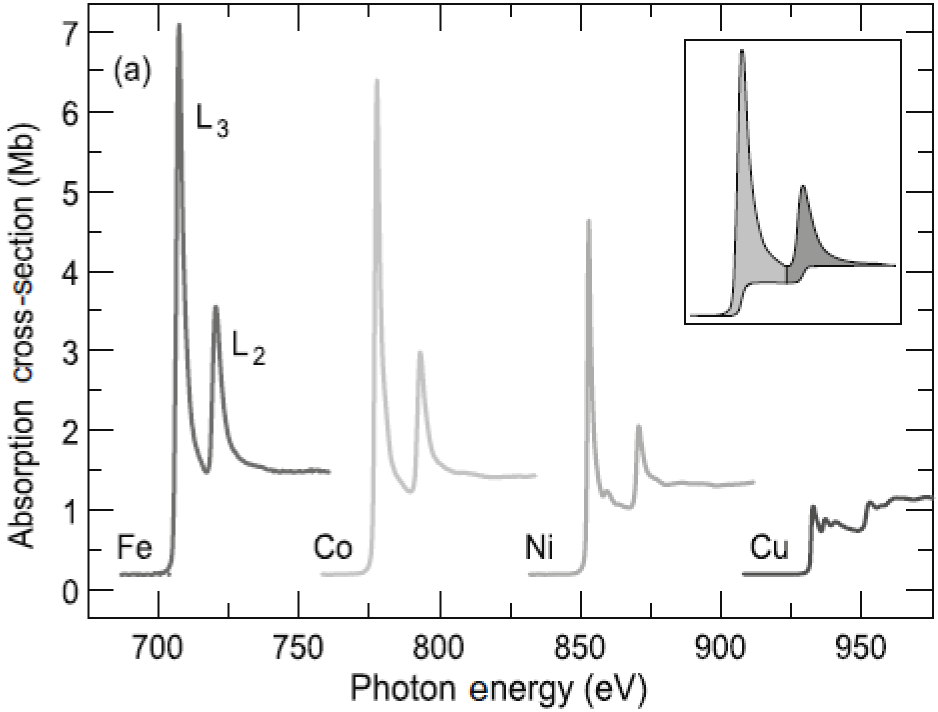
(sketches in textbooks can be misleading!)

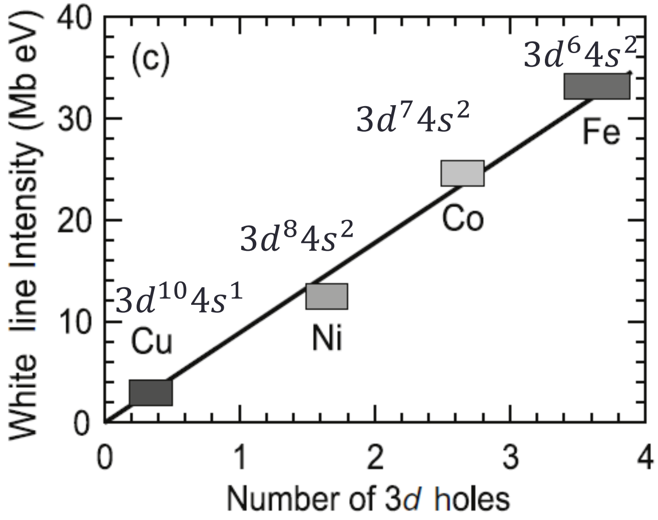
$$\Delta I_{\mathrm{XMCD}} \propto \vec{\mathrm{M}} \cdot \vec{L}_{\gamma} \propto \cos\theta, \qquad \theta \measuredangle(\vec{M}, \vec{L}\gamma)$$

Interaction of **polarized** photons with matter

- (Orientation averaged) Sum rules $\langle I\rangle = \frac{1}{3}\left(I_\alpha^{-1} + I_\alpha^{0} + I_\alpha^{+1}\right)$ ($\alpha = z$)

Density of d-states at E_F $\sigma^{\text{abs}} = 4\pi^2 \frac{e^2}{4\pi\epsilon_0 \hbar c} \hbar\omega\, |\langle b|\,\epsilon\cdot r\,|a\rangle|^2\, \delta[\hbar\omega - (E_b - E_a)]\, \rho(E_b)$



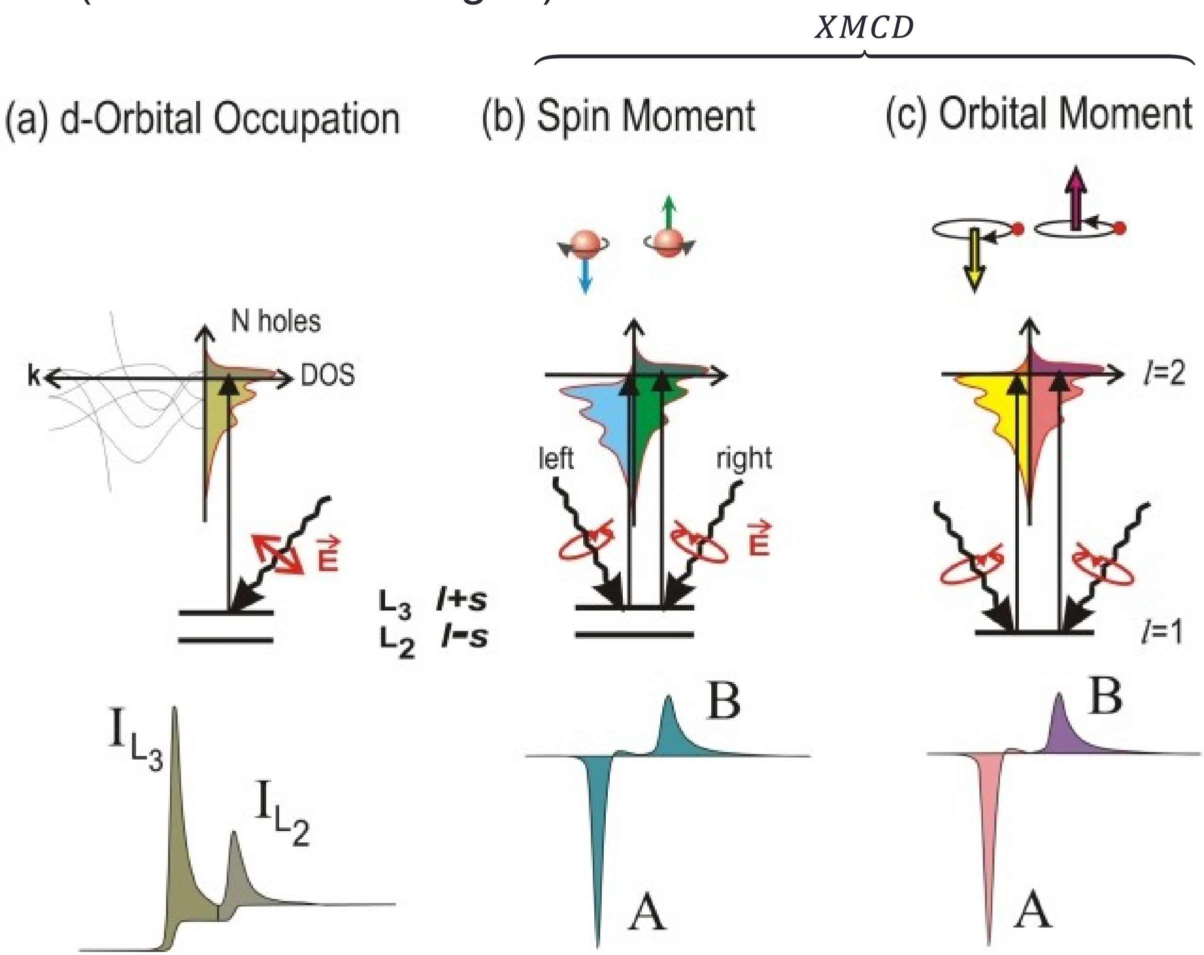


$D_d(E_F) = \frac{\langle I_{L3} + I_{L2}\rangle}{C}$ with $C = AR^2 \frac{L}{3(2L+1)}$

Orientation averaged refers to polycrystalline samples, such that anisotropic charge and spin order is eliminated

Interaction of **polarized** photons with matter

> (Orientation averaged) Sum rules



SOC ~50 meV
For d electrons

$$m_l \ll m_s$$

Angular moment

$$m_l = \frac{2\mu_B\langle A+B\rangle}{3C}$$

Spin moment

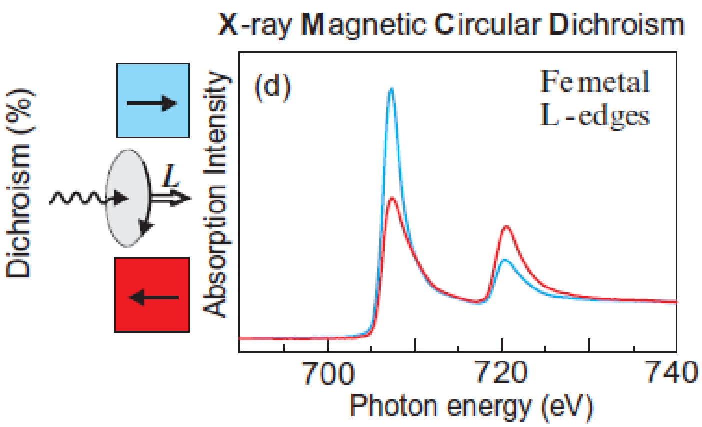
$$m_s = \frac{\mu_B\langle -A+2B\rangle}{C}$$

$$N_h = D(E_f) = \frac{\langle I_{L_3} + I_{L_2}\rangle}{C}$$

Interaction of **polarized** photons with matter

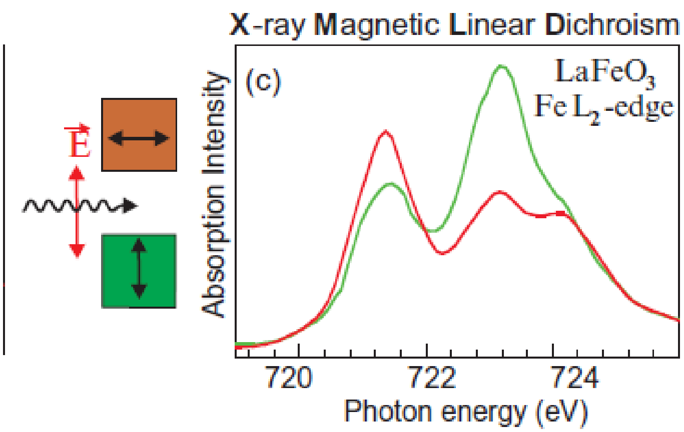
> XMCD and XMLD effect

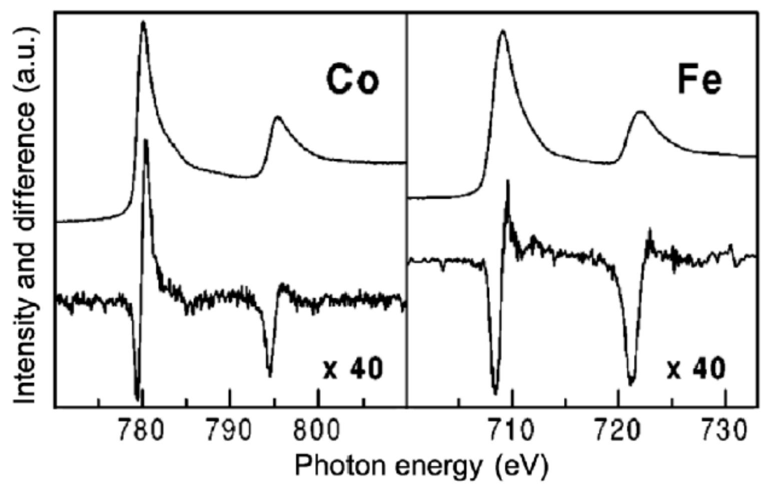
X-ray "magnetic" dichroism is due to spin alignment and the spin–orbit coupling.

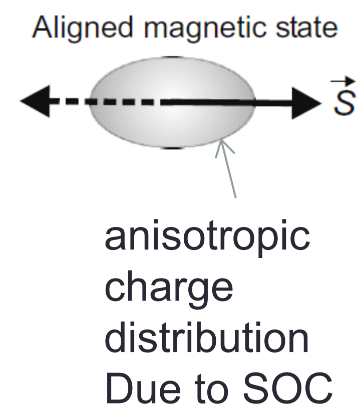


– X-ray magnetic circular dichroism – XMCD – arises from *directional* spin alignment. The effect is parity even and time odd.

– X-ray magnetic linear dichroism – XMLD – arises from a charge anisotropy induced by *axial* spin alignment. The effect is parity even and time even.

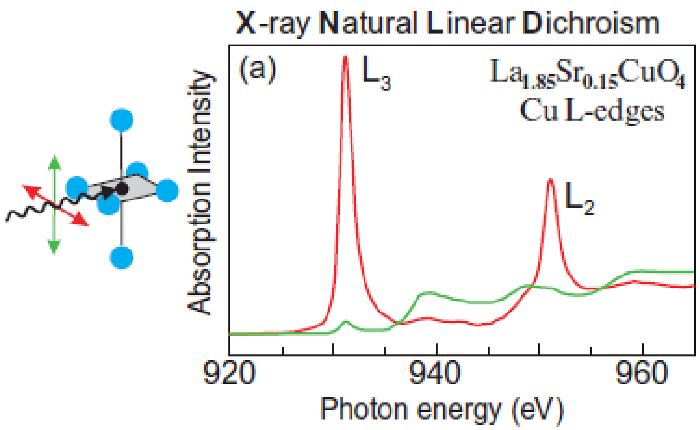


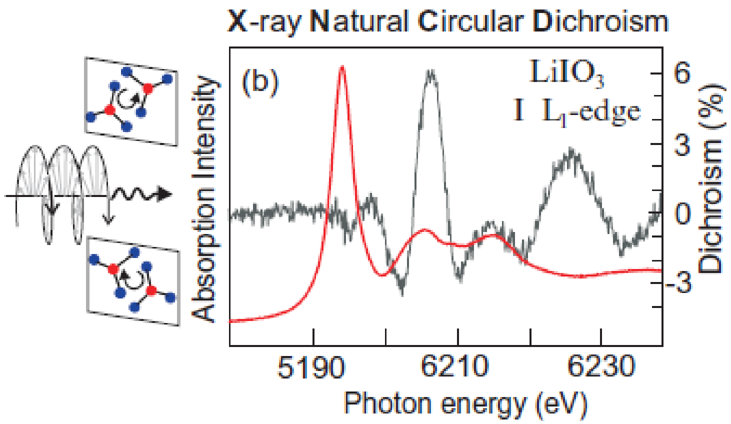




Interaction of **polarized** photons with matter

> XNLD and XNCD effect





X-ray "natural" dichroism refers to the absence of spin alignment.

- X-ray natural linear dichroism – XNLD – is due to an anisotropic charge distribution. The effect is parity even and time even.
- X-ray natural circular dichroism – XNCD – may be present for anisotropic charge distributions that lack a center of inversion. The effect is parity odd and time even.

Interaction of **polarized** photons with matter

> Application of XMCD

J. Appl. Phys. 67 (9), 1 May 1990

DORIS II at HASYLAB, DESY, Hamburg.

Spin-dependent x-ray absorption in Co/Pt multilayers and $Co_{50}Pt_{50}$

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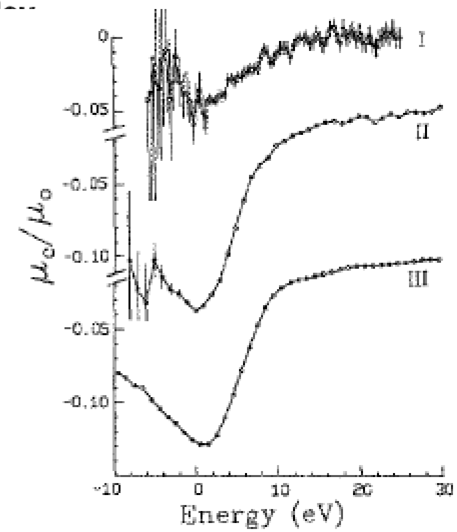
The spin dependence of $L_{2,3}$ absorption in 5*d* atoms oriented in a ferromagnetic matrix contains information on the spin density of the empty *d*-projected states of the absorbing atom. Spin-dependent absorption spectroscopy using circularly polarized synchrotron radiation was applied to study the polarization of the Pt atoms in the binary alloy $Co_{50}Pt_{50}$ and Pt/Co layered structures, which are promising candidates for magneto-optical recording. The spin-dependent absorption signals for vapor-deposited 250(4 Å Co + 18 Å Pt) and 250(6 Å Co + 18 Å Pt) multilayers indicate a ferromagnetic coupling on Pt and Co atoms with a significant Pt polarization. This is reduced on average by about 60% with respect to the Pt polarization in the $Co_{50}Pt_{50}$ alloy. The experimental results are discussed on the basis of spin-polarized band-structure calculations.



XMCD, CoPt

XMCD FePt

$$|\vec{m}_s|_{\mathrm{Pt}}^{\mathrm{CoPt}} = 0.35\mu_B/\text{atom}$$
$$|\vec{m}_s|_{\mathrm{Pt}}^{\mathrm{FePt}} = 0.08\mu_B/\text{atom}$$

„Historic example at hard x-ray energies (~11.5 keV) corresponding to the Platinum $L_{2,3}$ edge

Interaction of **polarized** photons with matter

> From Absorption to Resonant Scattering (exp. approach):

$f'' = -(k/4\pi)\,\sigma_a\,(E)$

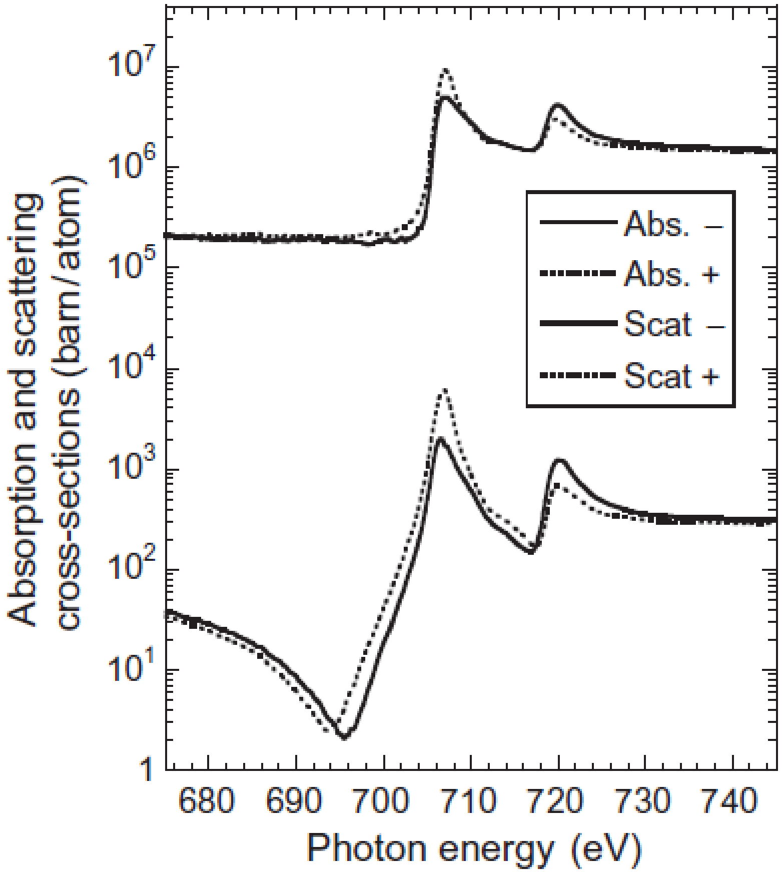
Measure absorption cross-section for both helicities

↓ Kramers-Kronig relation

f'

↓

$$\sigma_{\text{scattering}} = f^2$$
$$= [Z + f'(\omega, \boldsymbol{\varepsilon})]^2 + [f''(\omega, \boldsymbol{\varepsilon})]^2$$



Interaction of **polarized** photons with matter

- Resonant scattering (qm concept): 2. Term of Fermi's Golden rule in dipole approx.

$$T_{if} = \frac{2\pi}{\hbar} \left| \langle f|\mathcal{H}_{\rm int}|i\rangle + \sum_n \frac{\langle f|\mathcal{H}_{\rm int}|n\rangle\langle n|\mathcal{H}_{\rm int}|i\rangle}{\varepsilon_i - \varepsilon_n} \right|^2 \delta(\varepsilon_i - \varepsilon_f)\rho(\varepsilon_f) \qquad \sigma = \frac{T_{if}}{\Phi_0}$$

Dipol approximation etc. (as done for absorption term)

$$\frac{\hbar^2\omega^4}{c^2}\,\alpha_{\rm f}^2 \left| \sum_n \frac{\langle a|\boldsymbol{r}\cdot\boldsymbol{\epsilon}_2^*|n\rangle\langle n|\boldsymbol{r}\cdot\boldsymbol{\epsilon}_1|a\rangle}{(\hbar\omega - E_R^n) + \mathrm{i}(\Delta_n/2)} \right|^2$$

Δ_n: line width of intermediate states

J. P. Hannon et al., Phys. Rev. Lett **61**, 1245 (1988)

$$\begin{aligned}\langle a|\boldsymbol{r}\cdot\boldsymbol{\epsilon}_2^*|n\rangle\langle n|\boldsymbol{r}\cdot\boldsymbol{\epsilon}_1|a\rangle = \frac{\mathcal{R}^2}{2} \Big[&(\boldsymbol{\epsilon}_2^*\cdot\boldsymbol{\epsilon}_1)\left\{|C_{+1}|^2 + |C_{-1}|^2\right\} \\ &+ \mathrm{i}(\boldsymbol{\epsilon}_2^*\times\boldsymbol{\epsilon}_1)\cdot\hat{\boldsymbol{m}}\left\{|C_{-1}|^2 - |C_{+1}|^2\right\} \\ &+ (\boldsymbol{\epsilon}_2^*\cdot\hat{\boldsymbol{m}})(\boldsymbol{\epsilon}_1\cdot\hat{\boldsymbol{m}})\left\{2|C_0|^2 - |C_{-1}|^2 - |C_{+1}|^2\right\}\Big]\end{aligned}$$

Interaction of **polarized** photons with matter

> Resonant scattering: 2. Term of Fermi's Golden rule in dipole approximation

$$T_{if} = \frac{2\pi}{\hbar} \left| \langle f|\mathcal{H}_{\text{int}}|i\rangle + \sum_n \frac{\langle f|\mathcal{H}_{\text{int}}|n\rangle\langle n|\mathcal{H}_{\text{int}}|i\rangle}{\varepsilon_i - \varepsilon_n} \right|^2 \delta(\varepsilon_i - \varepsilon_f)\rho(\varepsilon_f)$$

with $\sigma = \frac{T_{if}}{\Phi_0}$ and $\sigma_{\text{scattering}} = f^2$

➔ The *elastic resonant magnetic scattering factor* in units [number of electrons] is given by

$$f(\omega,\boldsymbol{\varepsilon}_1) = \frac{\hbar\omega^2\alpha_f\mathcal{R}^2}{2cr_0}\left[\underbrace{(\epsilon_2^*\cdot\epsilon_1)\,G_0}_{\text{charge}} + \underbrace{\mathrm{i}(\epsilon_2^*\times\epsilon_1)\cdot\hat{m}\,G_1}_{\text{XMCD}} + \underbrace{(\epsilon_2^*\cdot\hat{m})(\epsilon_1\cdot\hat{m})\,G_2}_{\text{XMLD}}\right]$$

$$G_1 = \sum_n \frac{|\langle a|C_{-1}^{(1)}|n\rangle|^2 - |\langle a|C_{+1}^{(1)}|n\rangle|^2}{(\hbar\omega - E_R^n) + \mathrm{i}(\Delta_n/2)}$$

For circularly polarized light

$$\mathrm{i}\left[(\boldsymbol{\epsilon}^{\pm})^* \times \boldsymbol{\epsilon}^{\pm}\right] = \mp\,\mathbf{e}_z$$

Charge scattering/XNLD

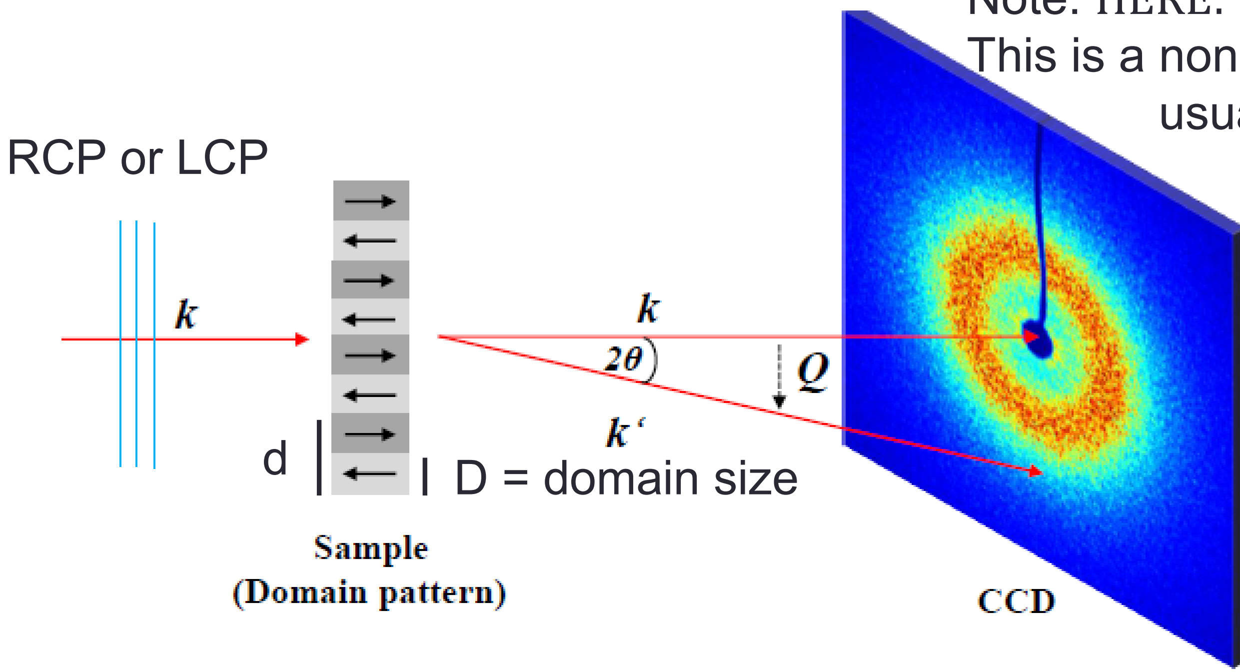
$$G_0 = \sum_n \frac{|\langle a|C_{+1}^{(1)}|n\rangle|^2 + |\langle a|C_{-1}^{(1)}|n\rangle|^2}{(\hbar\omega - E_R^n) + \mathrm{i}(\Delta_n/2)}$$

XMLD

$$G_2 = \sum_n \frac{2|\langle a|C_0^{(1)}|n\rangle|^2 - |\langle a|C_{-1}^{(1)}|n\rangle|^2 - |\langle a|C_{+1}^{(1)}|n\rangle|^2}{(\hbar\omega - E_R^n) + \mathrm{i}(\Delta_n/2)}$$

mSAXS of magnetic domain patterns

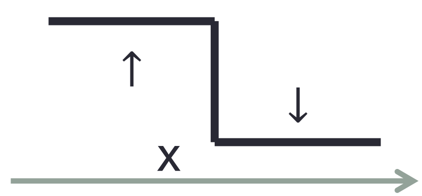
Note: $\text{HERE: } 2\theta =$ total scattering angle
This is a non-standard nomenclature!
usually the scattering angle is called θ! [for SAXS]



„magnetic grating/lattice“ = stripe domain pattern with equal domain size *D* (periodicity of *d* = 2*D*)

➔Scattering factor $f_m = M_z F^m$ varies in x-direction due to XMCD effect & alternating M_z

$$f_m(x) = \underbrace{f_m^0}_{\text{unit cell}} \otimes \underbrace{\sum_n \delta(x - nd)}_{\text{lattice}}$$



Domain wall thickness =0

mSAXS of magnetic domain patterns

Scattering amplitude (Fourier transform of scattering factor):

$$A(\vec{Q}) = FT\big(f(r)\big) = \underbrace{\tilde{f}(Q)}_{\text{unit cell}} \underbrace{\textstyle\sum_n e^{-iQnd}}_{\text{lattice sum}}$$ (for a regular magnetic lattice, e.g., stripes)

θ = full scattering angle!

With scattering vector = momentum transfer $Q = k - k' = \frac{4\pi}{\lambda} \cdot \sin\frac{\theta}{2}$,

Scattering intensity:

$$I(Q) = |A(Q)|^2 = \begin{cases} |\tilde{f}(Q)|^2 \cdot N_d^2 & \text{for } e^{iQnd} = 1 \\ \sim 0 & \text{else} \end{cases}$$

Intensity for $Q \cdot d = 2\pi \cdot n \Rightarrow Q = \frac{2\pi}{d}$, typically $d = 200\text{nm}, \lambda_{L\ \text{edge}} \approx 1.5\text{nm}$

$$\theta = 0.46° \ \Rightarrow \text{first max. at 4.8mm distance from } Q = 0 \text{ for distance of 600 mm}$$

mSAXS of magnetic domain patterns

What happens when the magnetic domains are disordered?

The discrete Fourier sum (lattice) becomes an integral over the magnetic structure

$$I(\boldsymbol{q}) = \left| \int_V F \exp(\mathrm{i}\boldsymbol{q}\boldsymbol{r})\, \mathrm{d}\boldsymbol{r} \right|^2 = \left| \int_V (\boldsymbol{k} \cdot \boldsymbol{m})\, G_1 \exp(\mathrm{i}\boldsymbol{q}\boldsymbol{r})\, \mathrm{d}\boldsymbol{r} \right|^2$$

Assuming homogeneous magnetization through the thickness of the film = k direction:

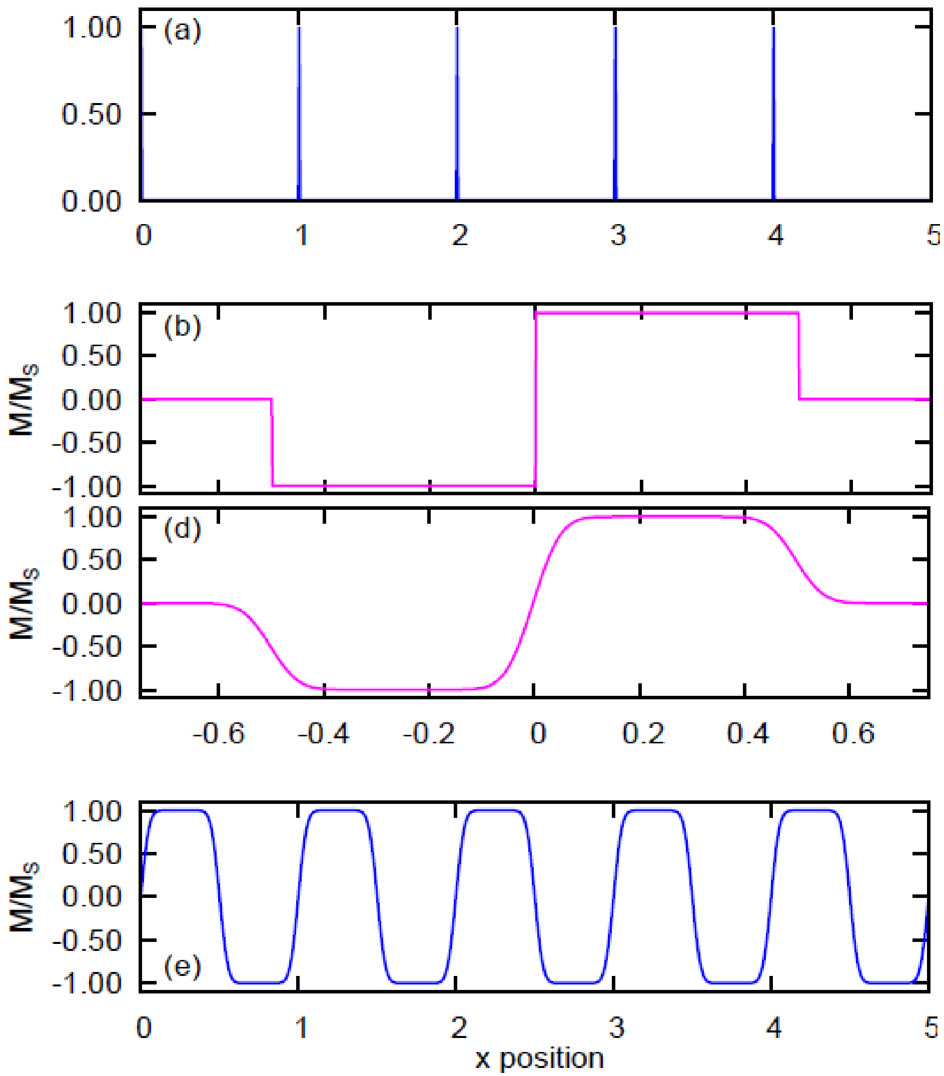
$$I(\boldsymbol{q}) \propto \left| \int_A M_z(\boldsymbol{r}) \exp(\mathrm{i}\boldsymbol{q}\boldsymbol{r})\, \mathrm{d}\boldsymbol{r} \right|^2$$

With V, the probed volume and A, the probed area, respectively

Note: We measure the Fourier transform absolute square of the z component of the magnetization. In-plane components we cannot measure (easily)

mSAXS of magnetic domain patterns

More realistic domains model (in 1d)



Basic lattice

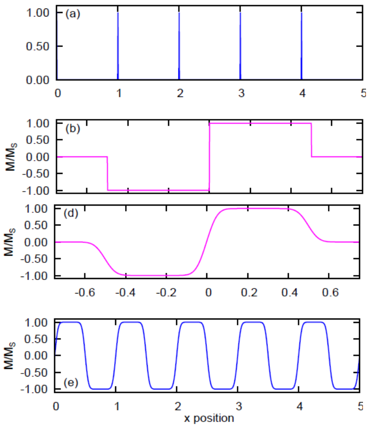
Unit cell

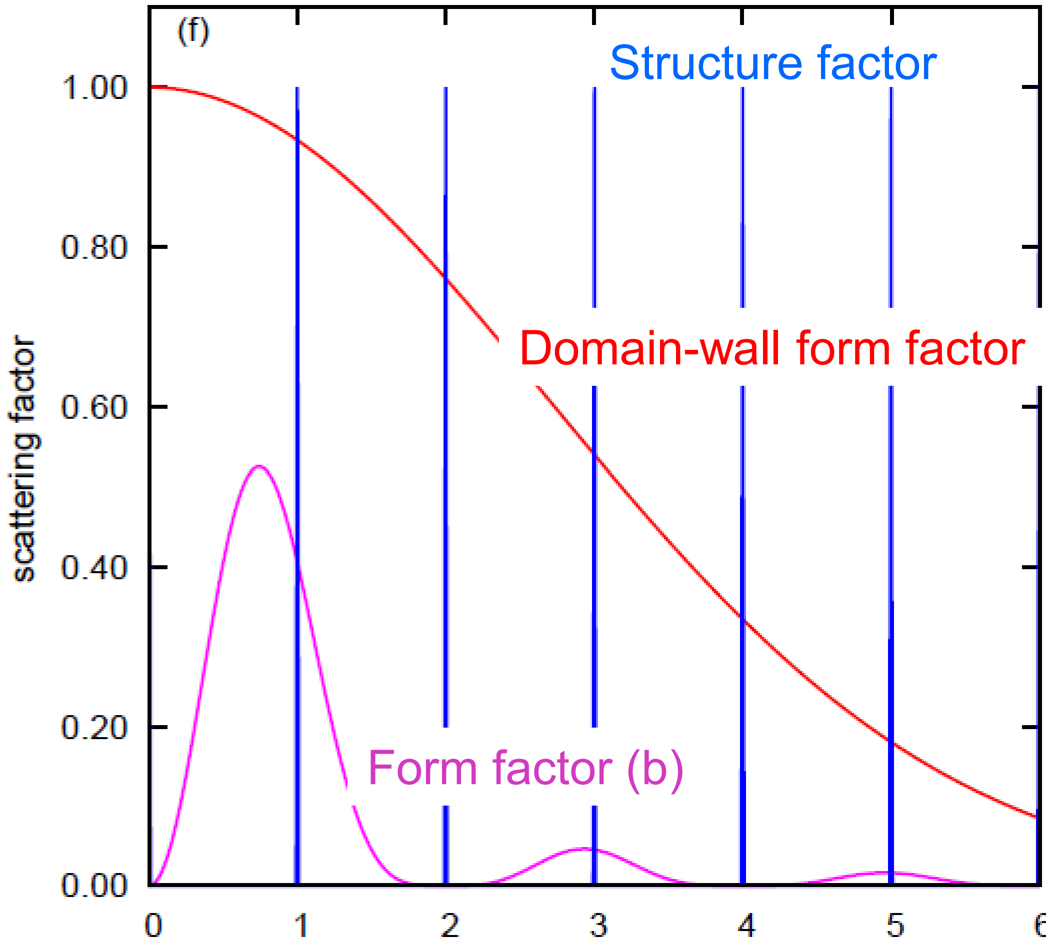
Unit cell with realistic domain walls

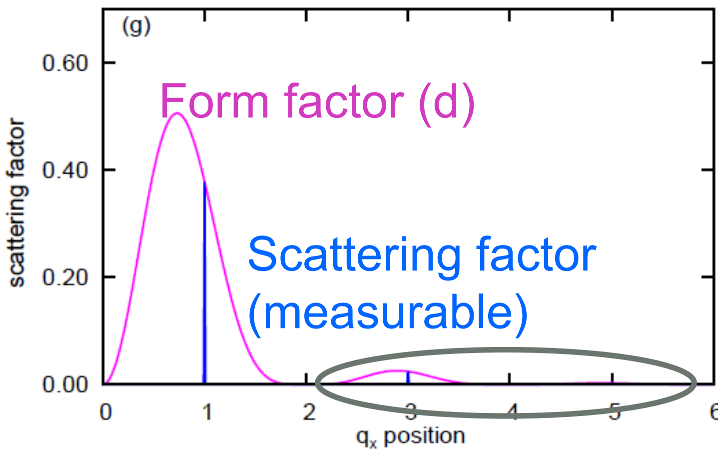
Magnetic lattice (unit cells at basic lattice points) [convolution of a and d]

mSAXS of magnetic domain patterns

More realistic domain model (in 1d)



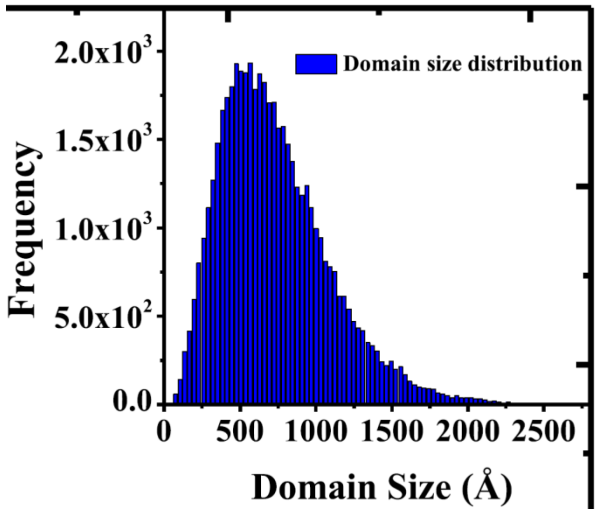




Note: Finite domain-wall width ℓ decreases the peak intensities $\propto e^{-\ell^2 q^2}$,
i.e., like a Debye-Waller factor [there: $\Delta\, r$ caused by thermal movement].
Impact of changes in ℓ are huge

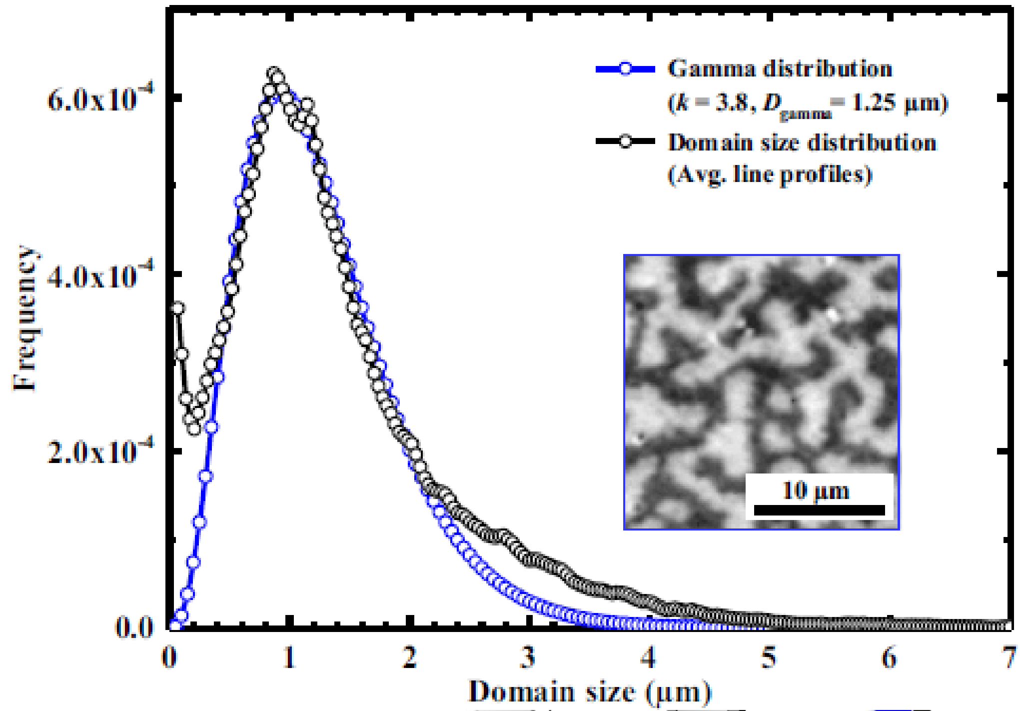
mSAXS of magnetic domain patterns

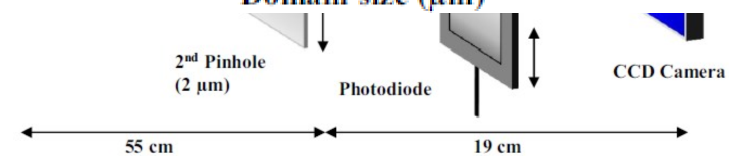
Even more realistic domain model (in 1d)



$$g(x) = \frac{x^{k-1} \exp\left(-\frac{x}{\vartheta}\right)}{\vartheta^k \Gamma(k)}, \quad x > 0,$$

Assume domains in 1d with a size distribution according to the gamma distribution (and 40 nm domain walls = average value)

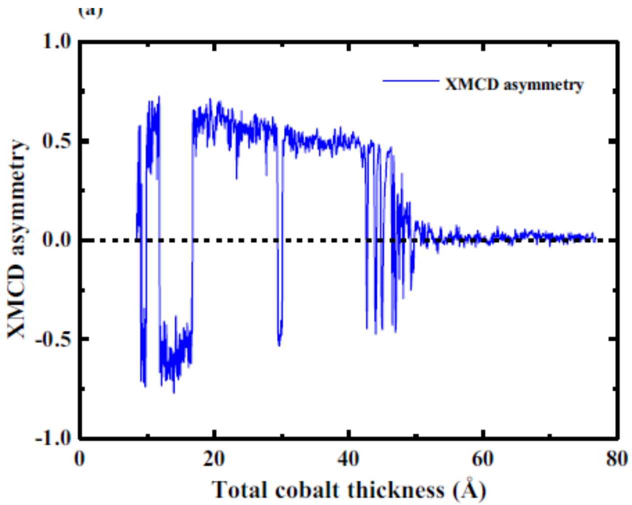




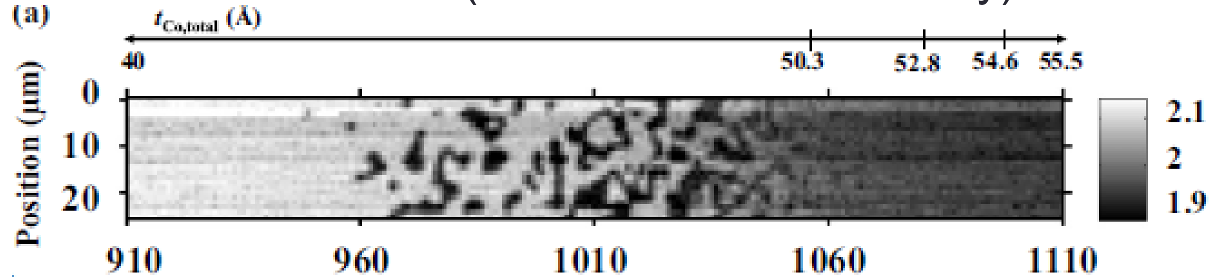
Compare experimental data from Petra III P04
To simulation

K. Bagschik et al., PRB 94, 134413

mSAXS of magnetic domain patterns

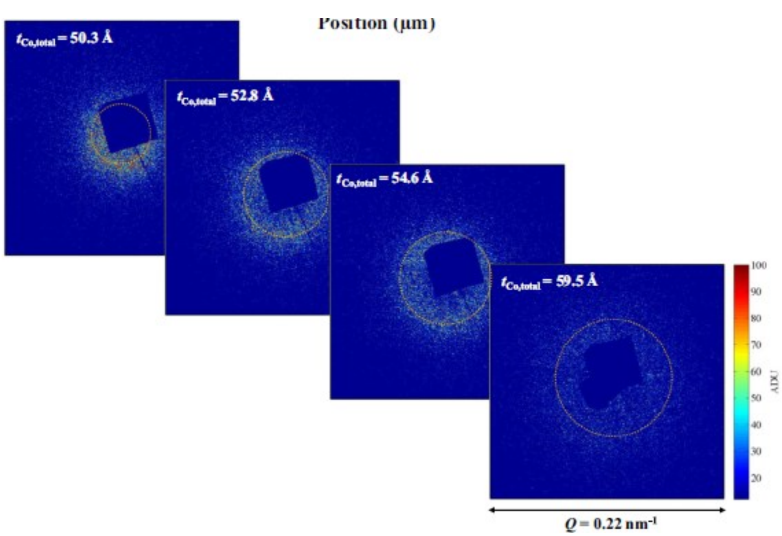


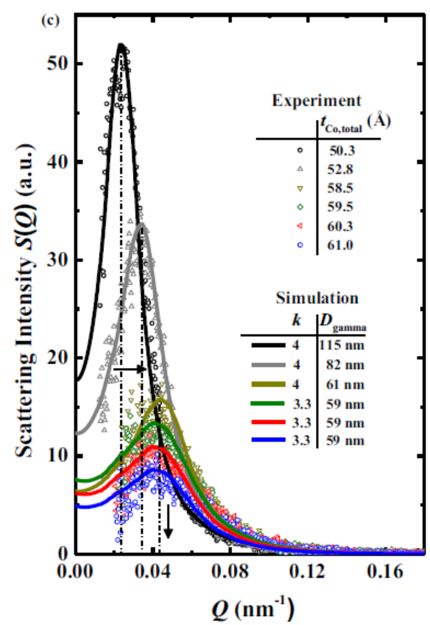
STXM (transmission of 1 helicity)



Position µm

Large domains for thin Co, small domains for thicker Co

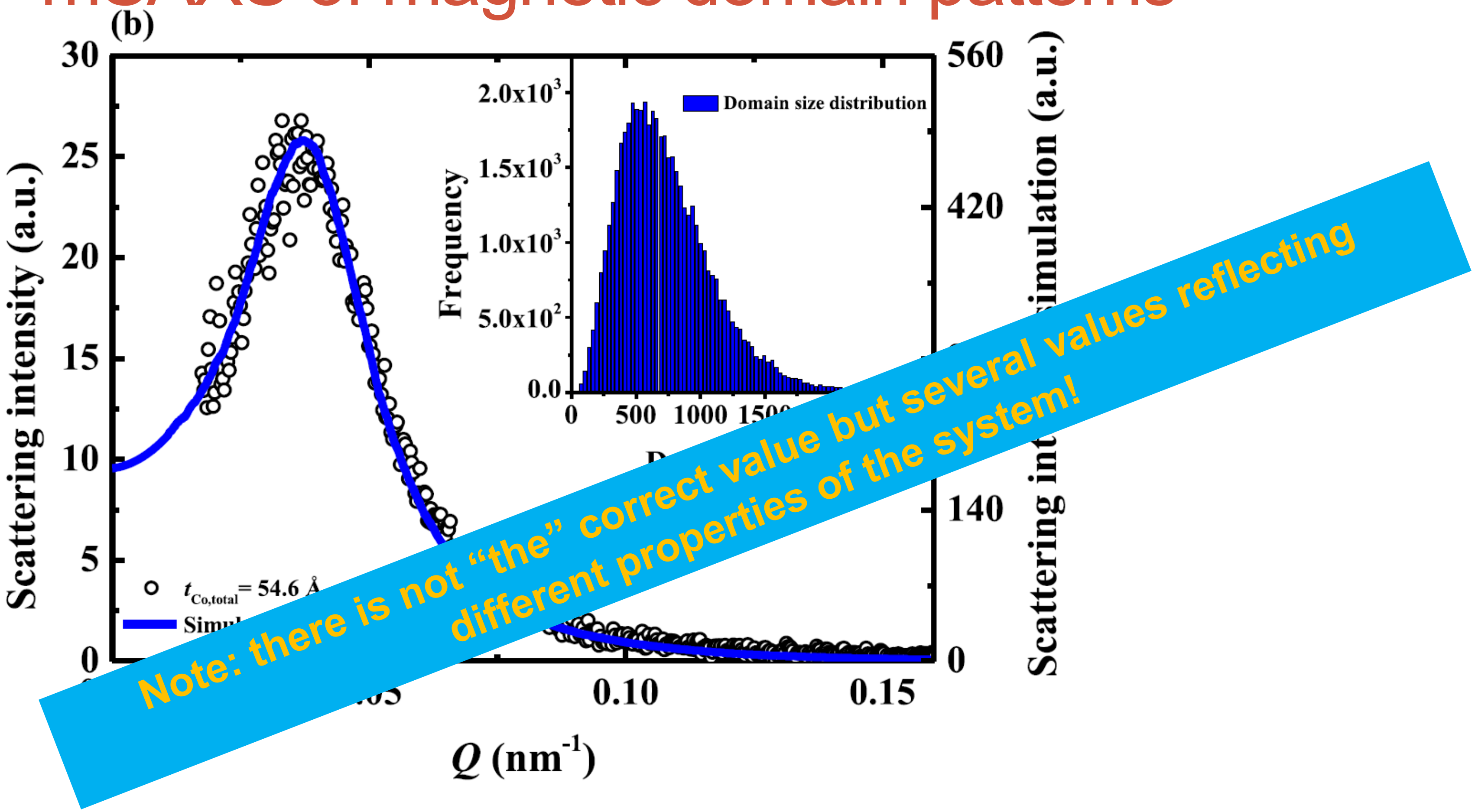




Good agreement between data and simulation

Note: the position of the peak is not the domain width but a measure for the average length scale

mSAXS of magnetic domain patterns

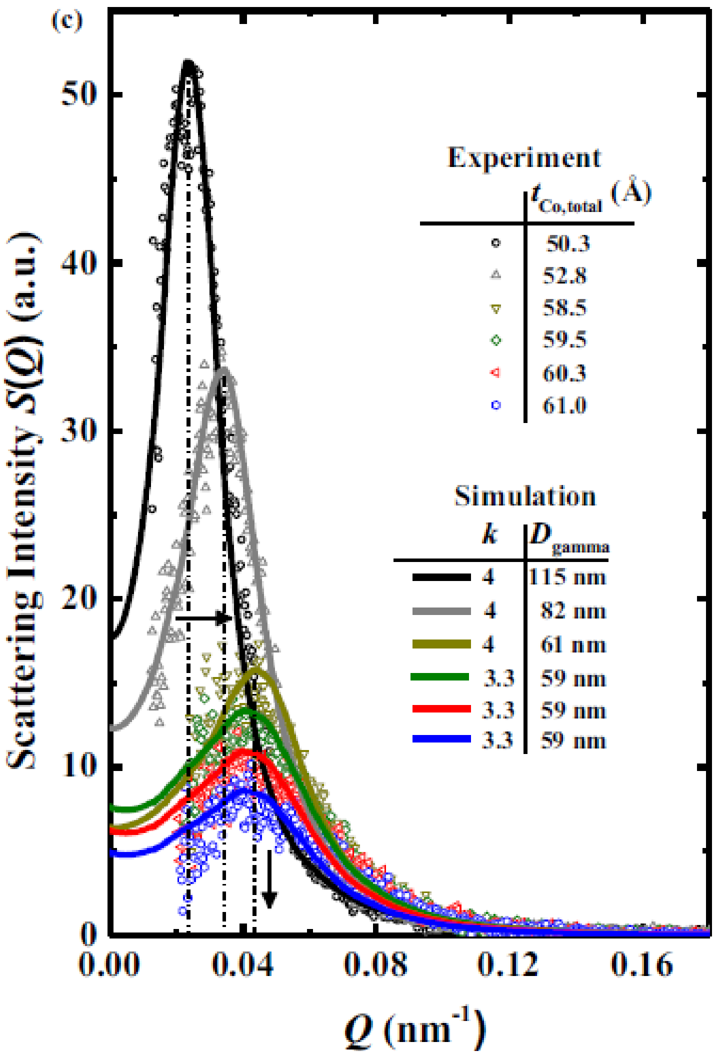


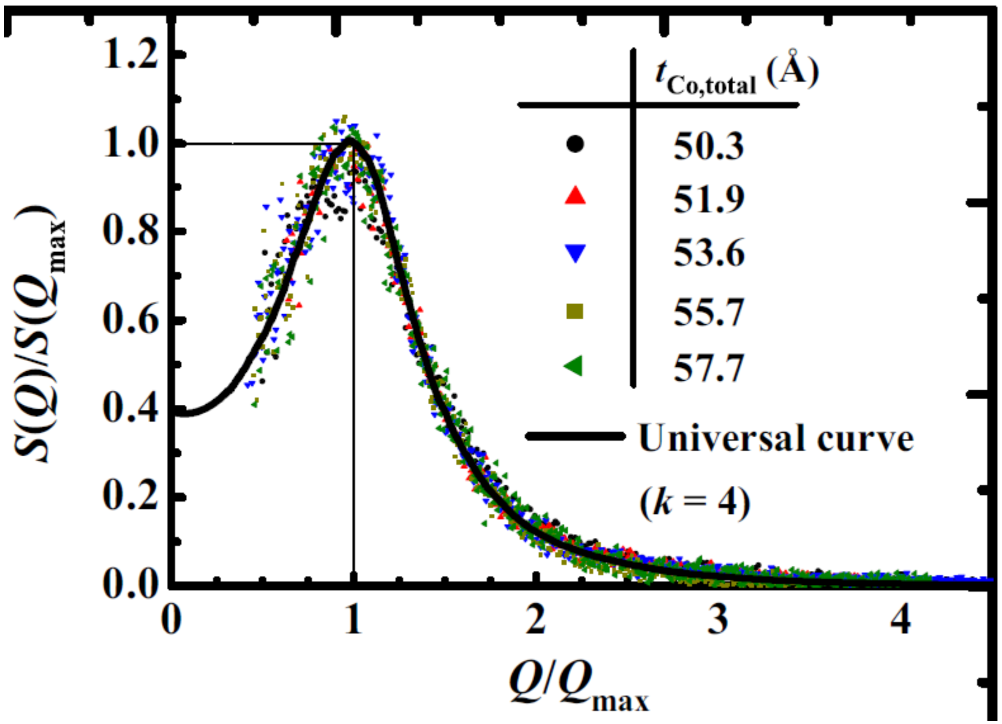
Domain width from scattering peak $D_{Q_{max}} = \frac{\frac{1}{2}2\pi}{Q_{max}} = 82.5\ \mathrm{nm}$

Domain width from gamma distribution $D_\gamma = k\vartheta = 73\ \mathrm{nm}$

Deviation of 12.6%

mSAXS of magnetic domain patterns





While the effective domain width decreases with cobalt thickness, the (accessible) spatial properties stay the same as all curves can be normalized to one universal curve.

mSAXS of magnetic domain patterns

- Linear polarization is not an eigenstate of magnetig scattering

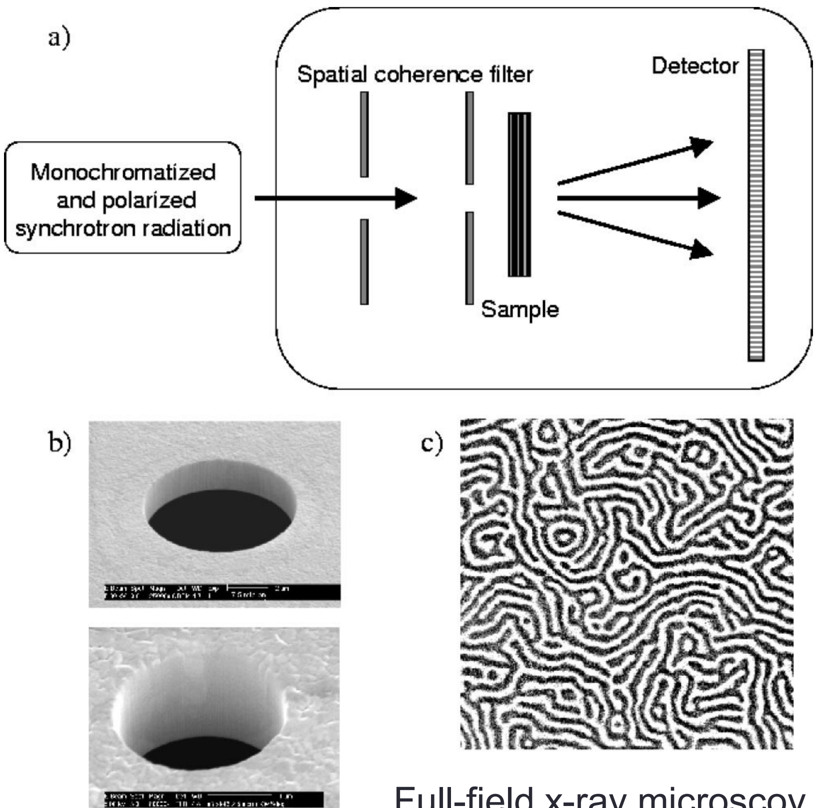
$$F(\hbar\omega) = \frac{\hbar\omega^2 \alpha_f R^2}{2cr_0}\left[\quad + i\underbrace{(\epsilon_2^* \times \epsilon_1)\cdot \hat{m}\, G_1}_{\text{XMCD}} + \quad\right]$$

Rotation of polarization when linear polarized light is scattered

a)

b)

c)



Full-field x-ray microscoy (5x5 µm)

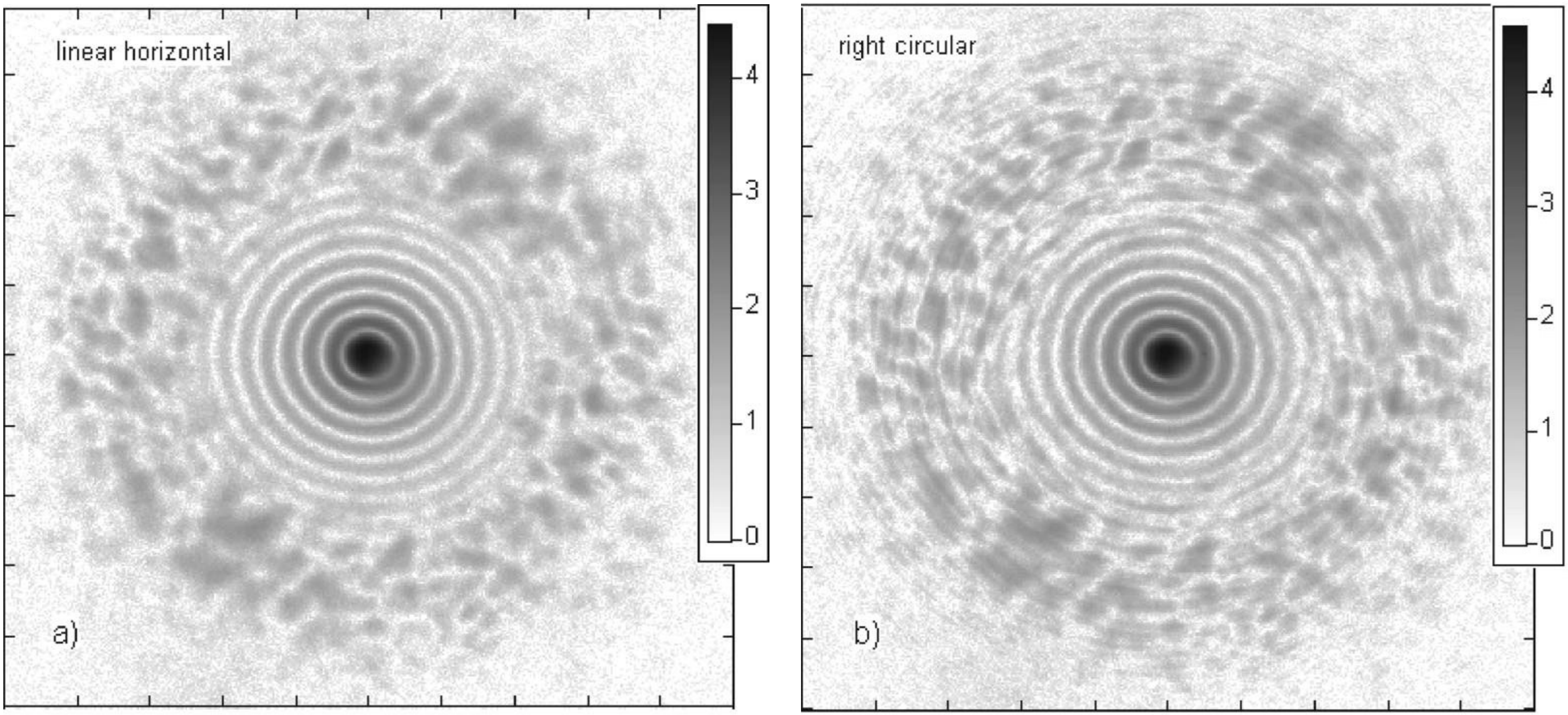
A gold mask ensures coherent illumination of a sample region, so that the detector records a speckle pattern.

The details of this pattern are directly related to the domain arrangement in the illuminated area.

S. Eisebitt et al., PRB 68, 104419 (2003)

mSAXS of magnetic domain patterns

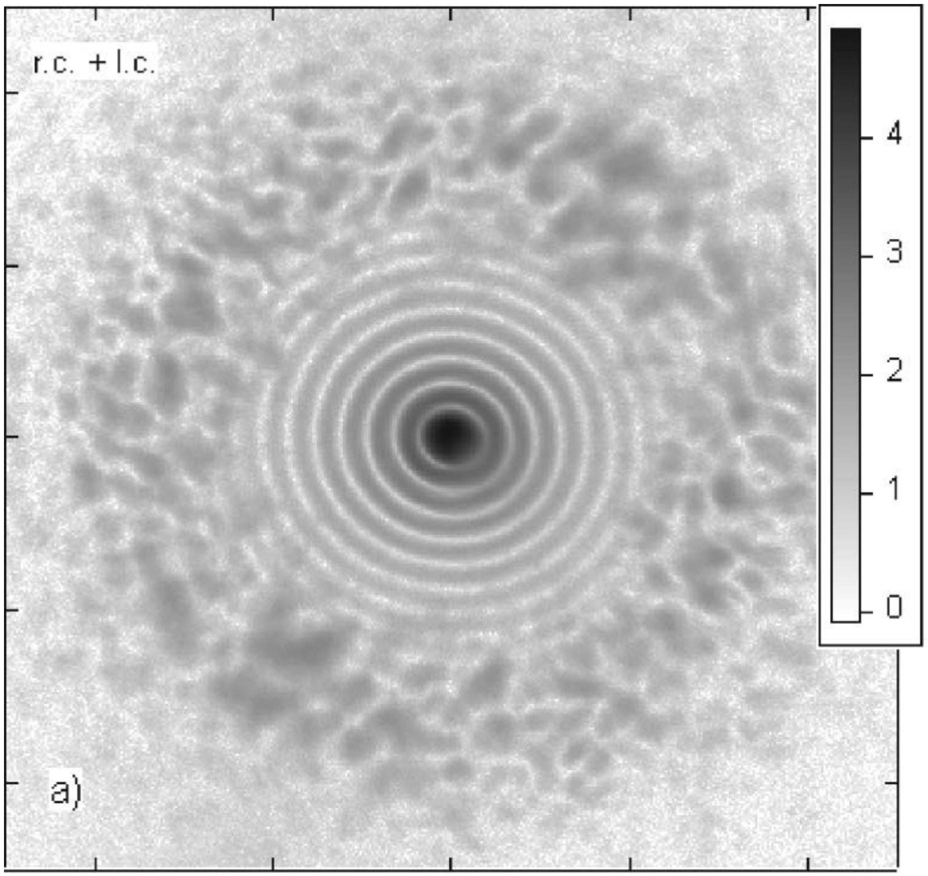
- Linear polarization is not an eigenstate of magnetig scattering



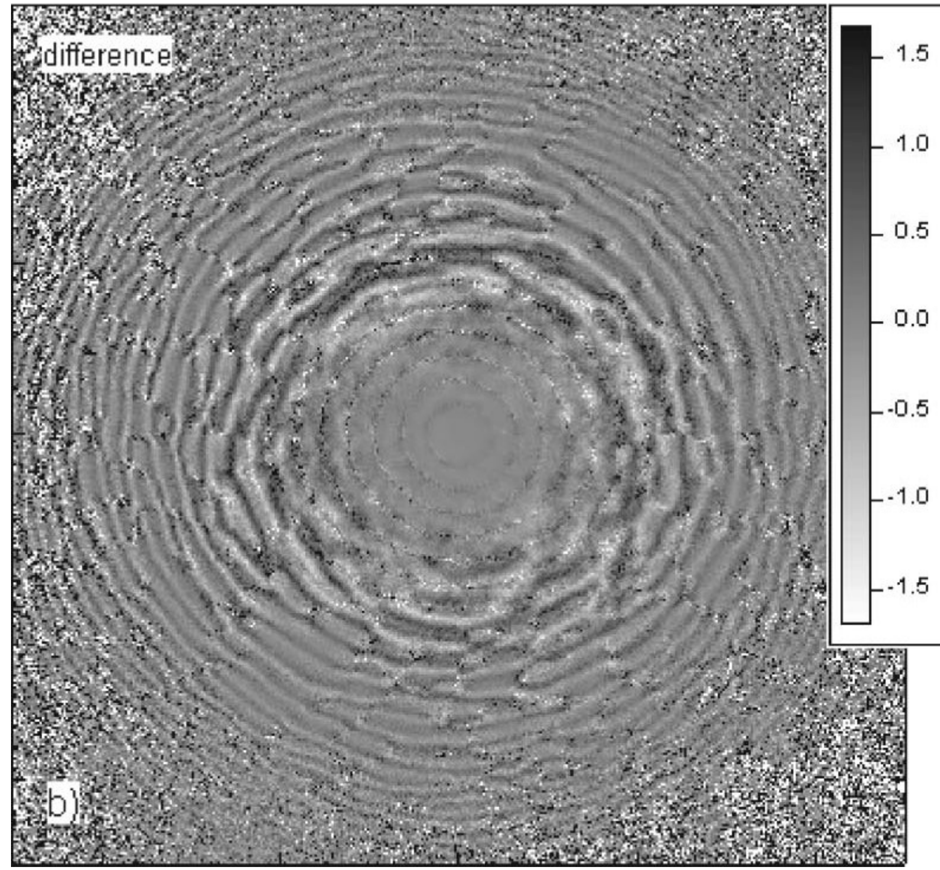
While in case of linear polarization the Airy pattern of the pinhole simply adds up with the magnetic speckles for circular polarization the Airy pattern modulates the speckle intensity

mSAXS of magnetic domain patterns

- Linear polarization is not an eigenstate of magnetic scattering



RCP+LCP = „LP image“



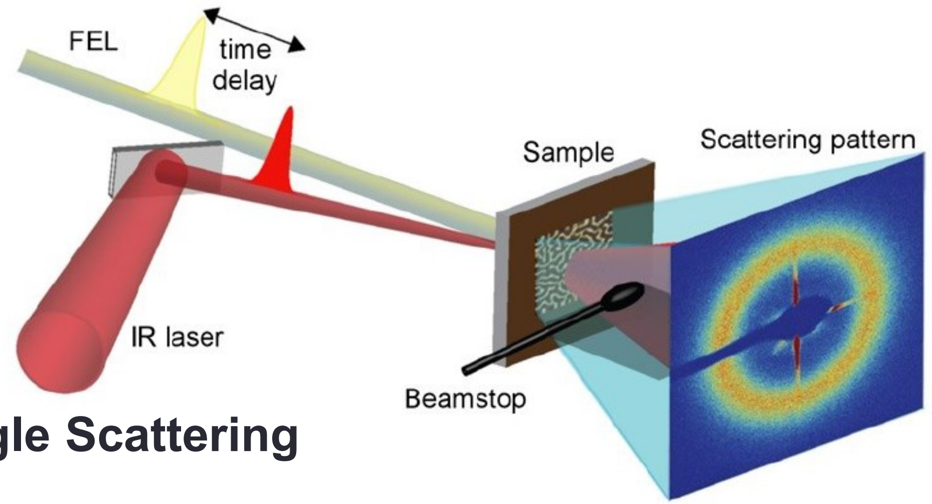
RCP-LCP=purely magnetic contrast

Outline

Part II/2:

Studies on Magnetic Nanostructures

by Leonard Müller

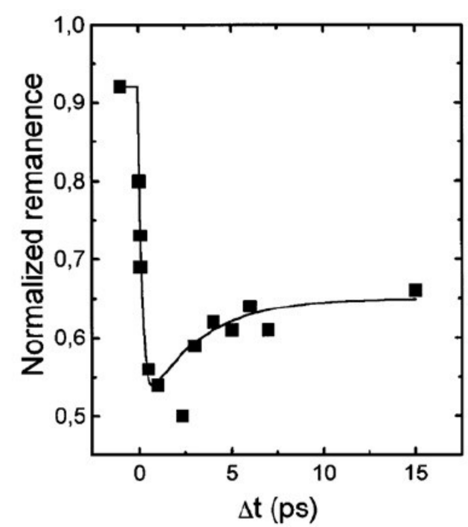


[6.6.] **X-ray Magnetic Circular Dichroism (XMCD) & Resonant Magnetic Small-Angle Scattering (mSAXS)**

- Role of Spin-Orbit Coupling and Exchange Splitting
- Sum Rules
- XMLD and Natural Dichroism
- mSAXS of Magnetic Domain Patterns

[~~18.6.~~ 20.6.] Femtomagnetism

- Introduction to Ultrafast Magnetization Dynamics Induced by Femtosecond Infrared Pulses
- Pump-Probe Experiments of Nano-Scale Magnetic Domain Patterns
- All-Optical Switching
- Manipulating Magnetism by XUV and THz Pulses



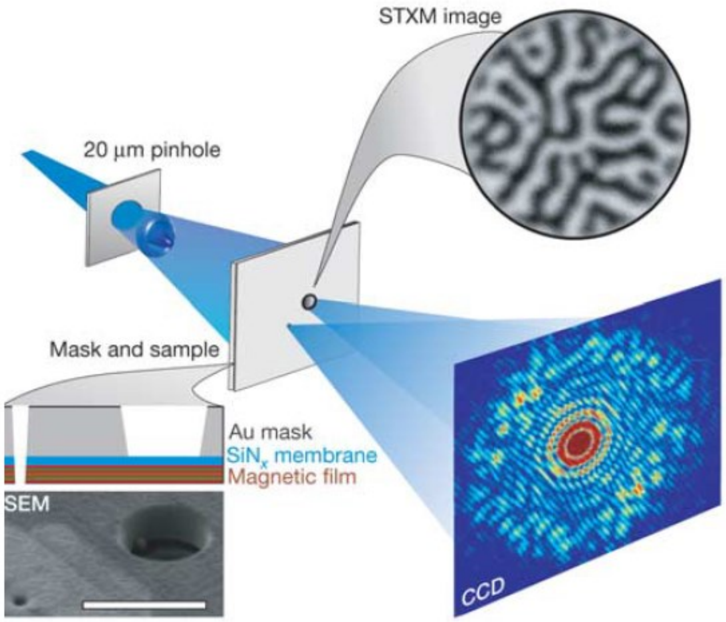
Part II/3:

Studies on Magnetic Nanostructures

by Leonard Müller

[18.6.] Imaging of Magnetic Domains

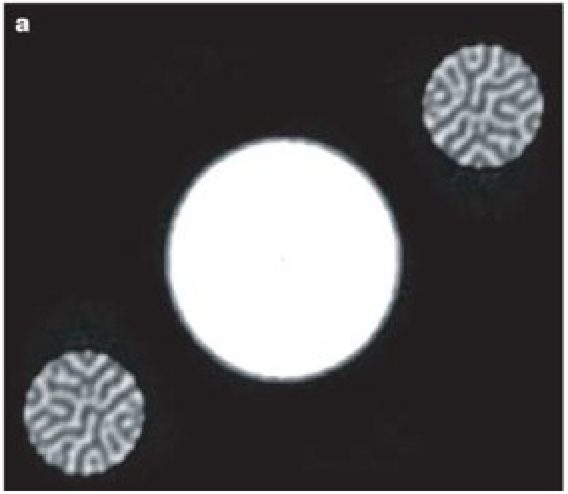
- **Fourier Transform Holography (FTH)**
- Scanning Transmission X-ray Microscopy (STXM)
- Coherent Diffraction Imaging (CDI)



Lensless imaging of magnetic nanostructures by X-ray spectro-holography

S. Eisebitt[1], J. Lüning[2], W. F. Schlotter[2,3], M. Lörgen[1], O. Hellwig[1,4], W. Eberhardt[1] & J. Stöhr[2]

NATURE | VOL 432 | 16 DECEMBER 2004 |



FTH