Workshop on Nuclear Resonant Scattering and Data Analysis
November 16 – 19, 2017, Advanced Photon Source

Studies of Thin-Film Magnetism Using Nuclear Resonant Scattering

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Outline

1. **Hyperfine Interactions:**
   Temporal beat patterns as fingerprints of magnetic properties

1. **Magnetic structure of thin films, multilayers and nanostripes**
   a. Spin structure of exchange-spring magnets
   b. Magnetic order in Fe/Fe-oxide heterostructures
   c. Spin-structured multilayers for precision spintronics

1. **Magnetic dynamics in thin films and nanostructures**
   a. Spin precession at ferromagnetic resonance

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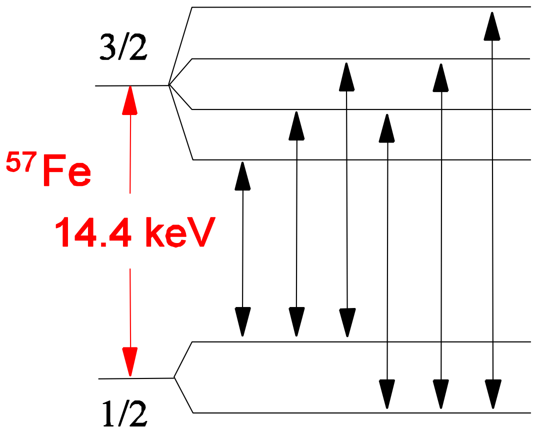
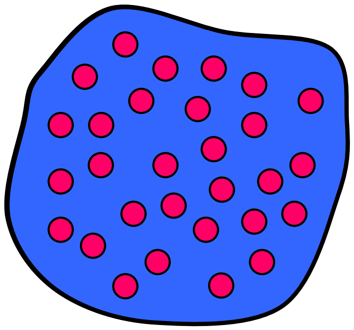
Nuclear Resonant Forward Scattering of Synchrotron Radiation

Pulsed broadband excitation of hyperfine-split nuclear levels



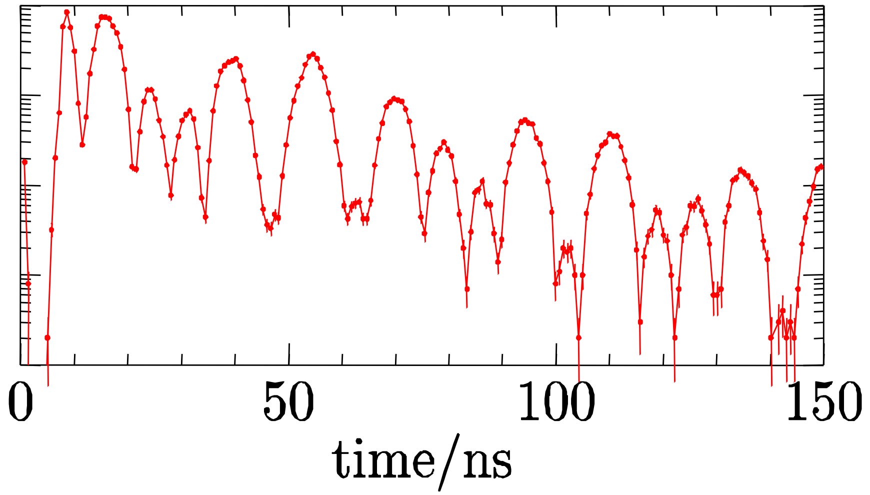
The 14.4 keV nuclear resonance of ^{57}Fe

τ_0 = 141 ns, Γ_0 = 4.7 neV



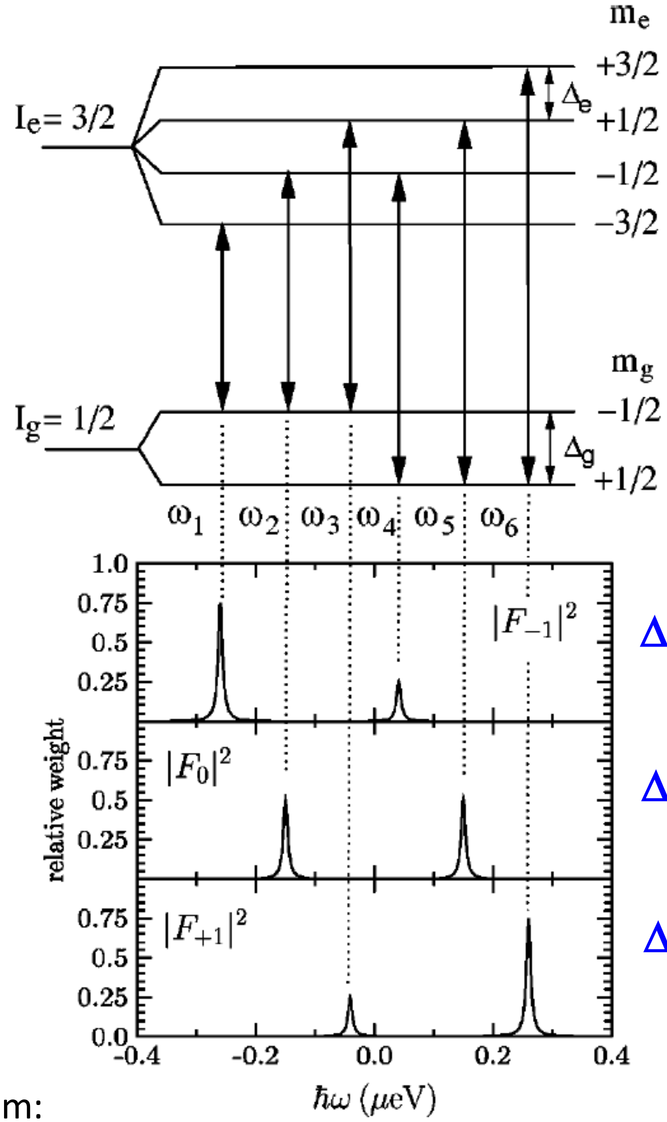
The beat pattern is a fingerprint of the magnetic structure of the sample:

Temporal beats



Magnetic Hyperfine Interaction

Energy dependence



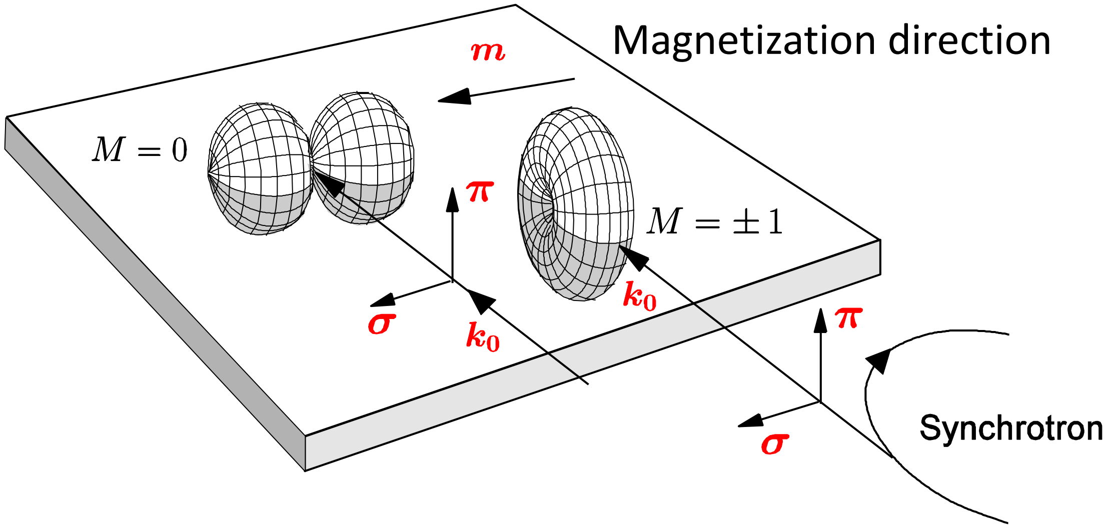
Δm = -1

Δm = 0

Δm = +1

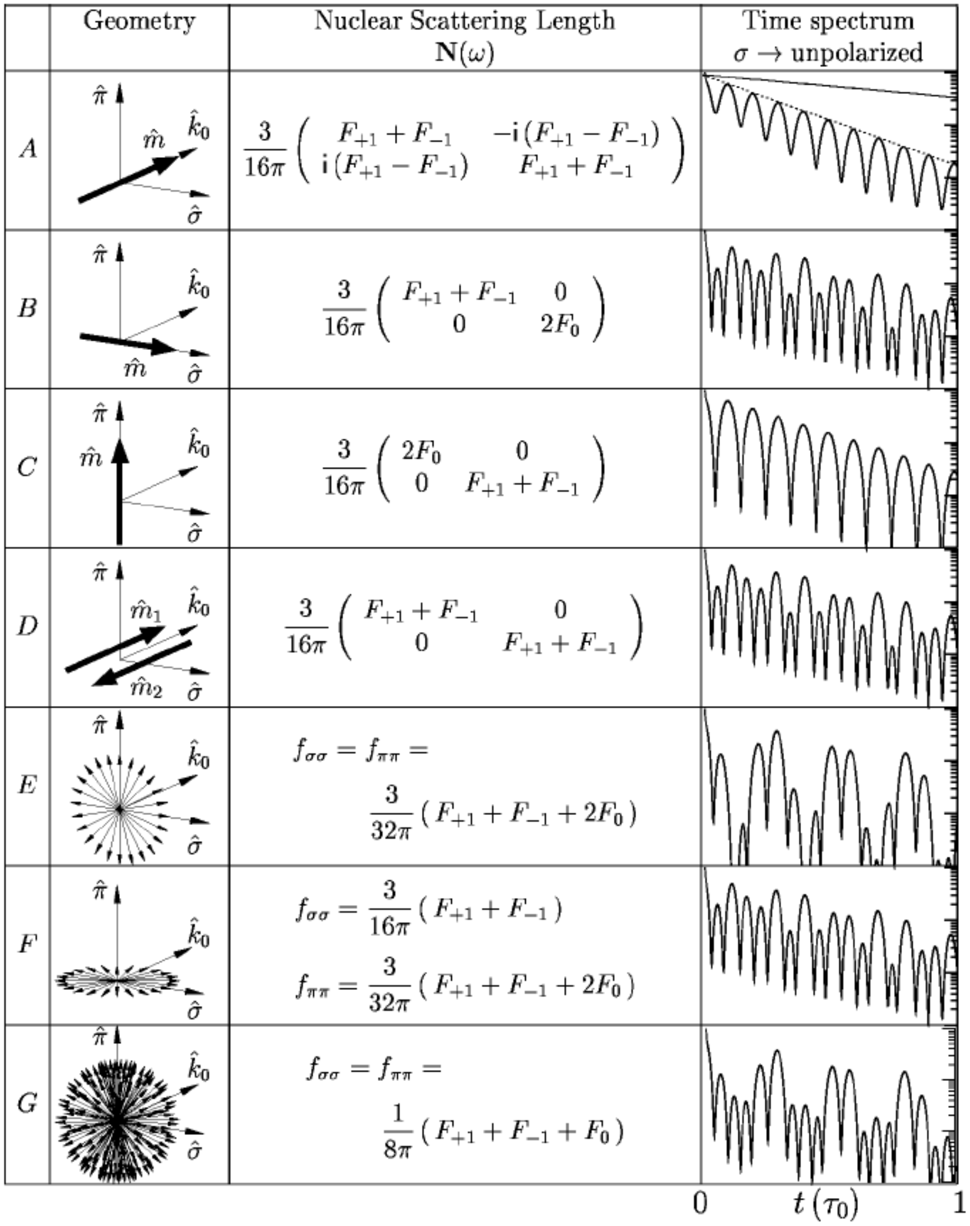
From:
Phys. Rev. B 67, 245412 (2003)

Directional dependence
→ Dipole emission characteristics, described by vector spherical harmonics



Magnetic Hyperfine Interaction

Directional dependence

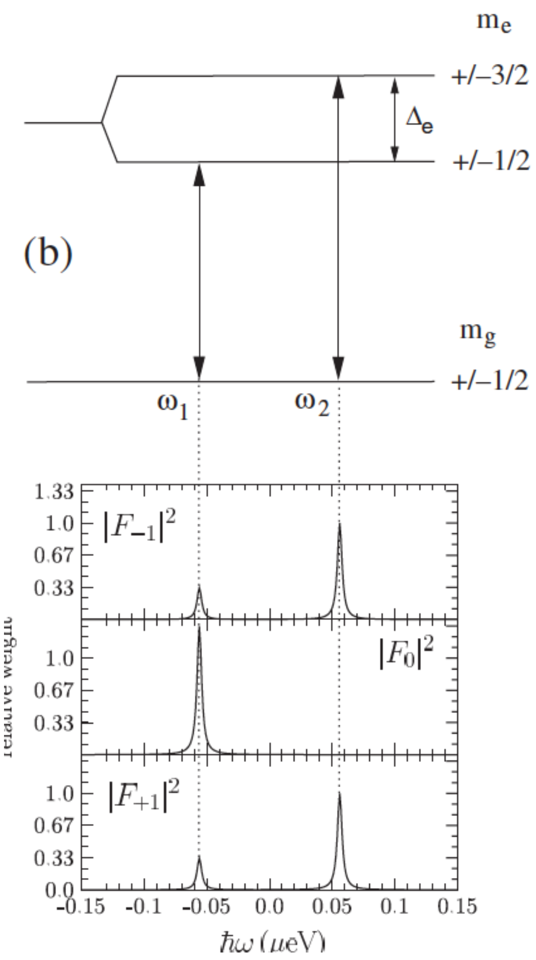


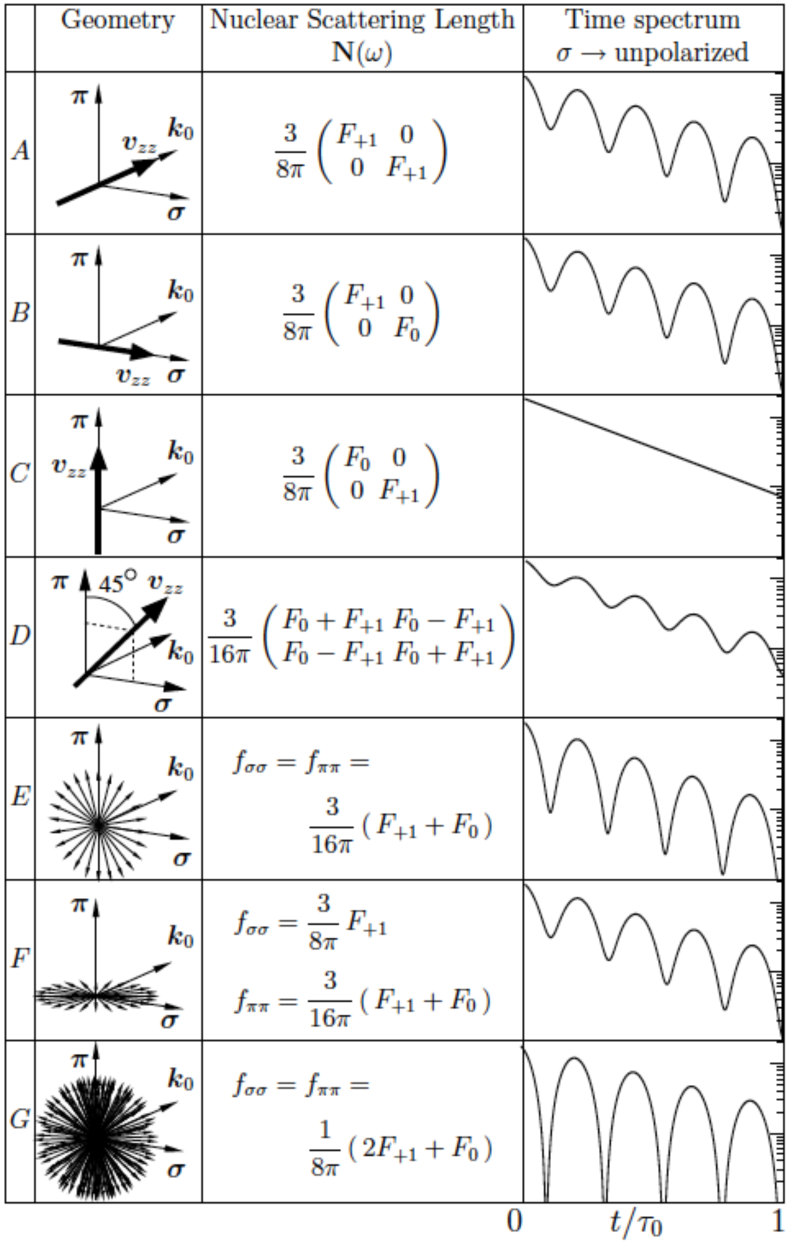
	Geometry	Nuclear Scattering Length $\mathbf{N}(\omega)$	Time spectrum $\sigma \to$ unpolarized
A		$\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & -\mathrm{i}\,(F_{+1}-F_{-1}) \\ \mathrm{i}\,(F_{+1}-F_{-1}) & F_{+1}+F_{-1} \end{pmatrix}$	
B		$\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & 0 \\ 0 & 2F_0 \end{pmatrix}$	
C		$\frac{3}{16\pi}\begin{pmatrix} 2F_0 & 0 \\ 0 & F_{+1}+F_{-1} \end{pmatrix}$	
D		$\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & 0 \\ 0 & F_{+1}+F_{-1} \end{pmatrix}$	
E		$f_{\sigma\sigma} = f_{\pi\pi} = \frac{3}{32\pi}(F_{+1}+F_{-1}+2F_0)$	
F		$f_{\sigma\sigma} = \frac{3}{16\pi}(F_{+1}+F_{-1})$ $f_{\pi\pi} = \frac{3}{32\pi}(F_{+1}+F_{-1}+2F_0)$	
G		$f_{\sigma\sigma} = f_{\pi\pi} = \frac{1}{8\pi}(F_{+1}+F_{-1}+F_0)$	

From:
Phys. Rev. B 67, 245412 (2003)

Electric Hyperfine Interaction

Directional dependence

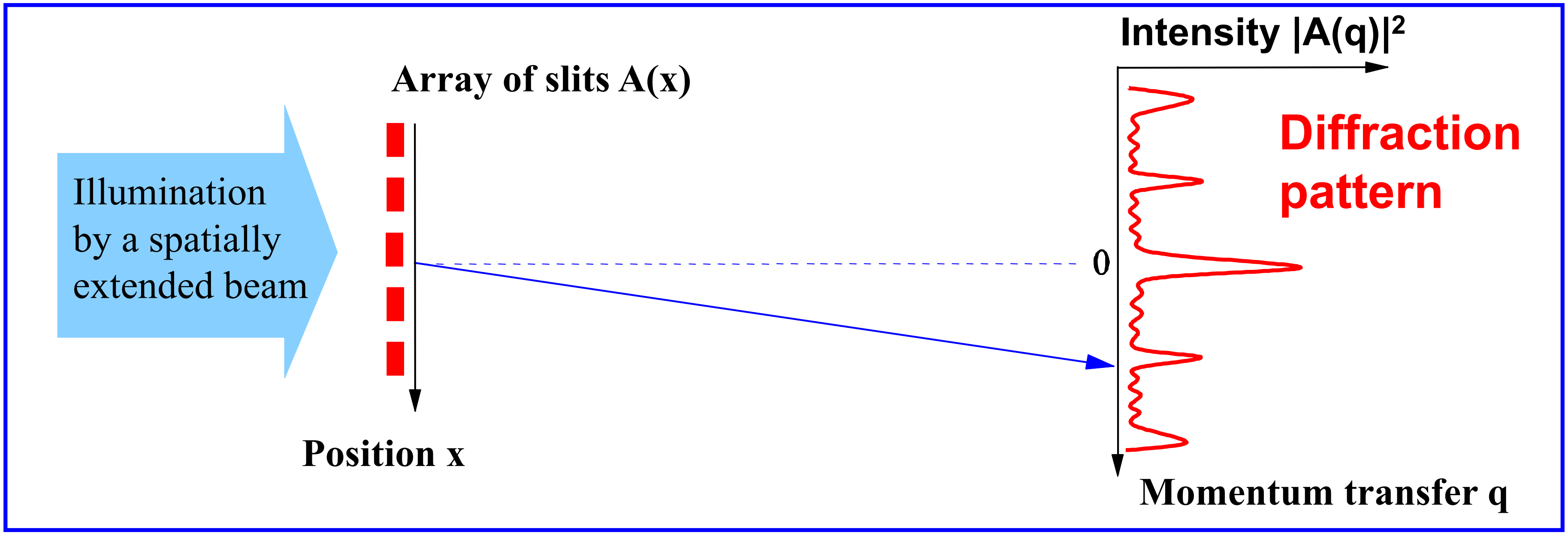




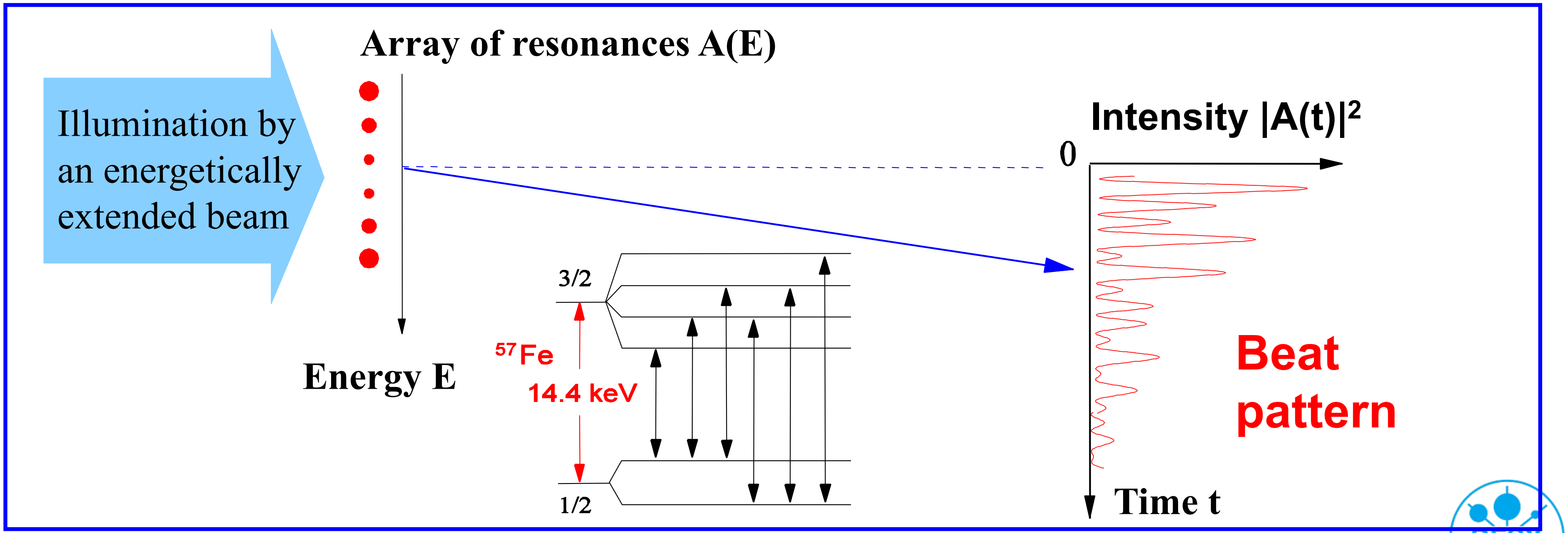
	Geometry	Nuclear Scattering Length $\mathbf{N}(\omega)$	Time spectrum $\sigma \rightarrow$ unpolarized
A	π, v_{zz}, k_0, σ	$\frac{3}{8\pi}\begin{pmatrix} F_{+1} & 0 \\ 0 & F_{+1} \end{pmatrix}$	
B	π, k_0, v_{zz}, σ	$\frac{3}{8\pi}\begin{pmatrix} F_{+1} & 0 \\ 0 & F_0 \end{pmatrix}$	
C	π, v_{zz}, k_0, σ	$\frac{3}{8\pi}\begin{pmatrix} F_0 & 0 \\ 0 & F_{+1} \end{pmatrix}$	
D	π, 45°, v_{zz}, k_0, σ	$\frac{3}{16\pi}\begin{pmatrix} F_0 + F_{+1} & F_0 - F_{+1} \\ F_0 - F_{+1} & F_0 + F_{+1} \end{pmatrix}$	
E	π, k_0, σ	$f_{\sigma\sigma} = f_{\pi\pi} = \frac{3}{16\pi}(F_{+1} + F_0)$	
F	π, k_0, σ	$f_{\sigma\sigma} = \frac{3}{8\pi}F_{+1}$ $f_{\pi\pi} = \frac{3}{16\pi}(F_{+1} + F_0)$	
G	π, k_0, σ	$f_{\sigma\sigma} = f_{\pi\pi} = \frac{1}{8\pi}(2F_{+1} + F_0)$	

From:
R. Röhlsberger, Springer Tracts in Modern Physics, Vol. 208 (2005)

Diffraction as Method of Structure Determination



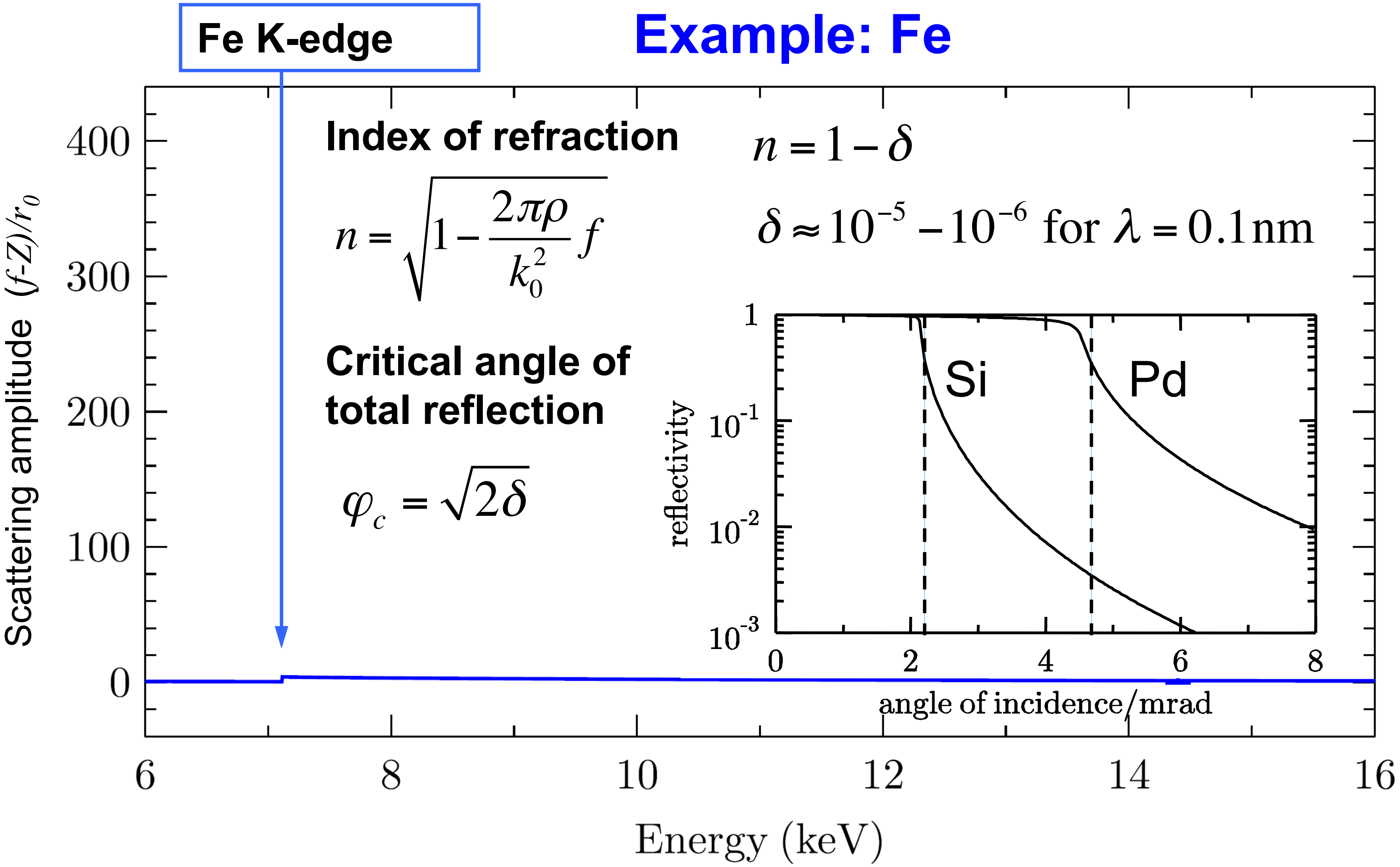
Illumination by a spatially extended beam



Illumination by an energetically extended beam

Optical Properties in the X-ray Regime

Example: Fe



Index of refraction

$$n = \sqrt{1 - \frac{2\pi\rho}{k_0^2} f}$$

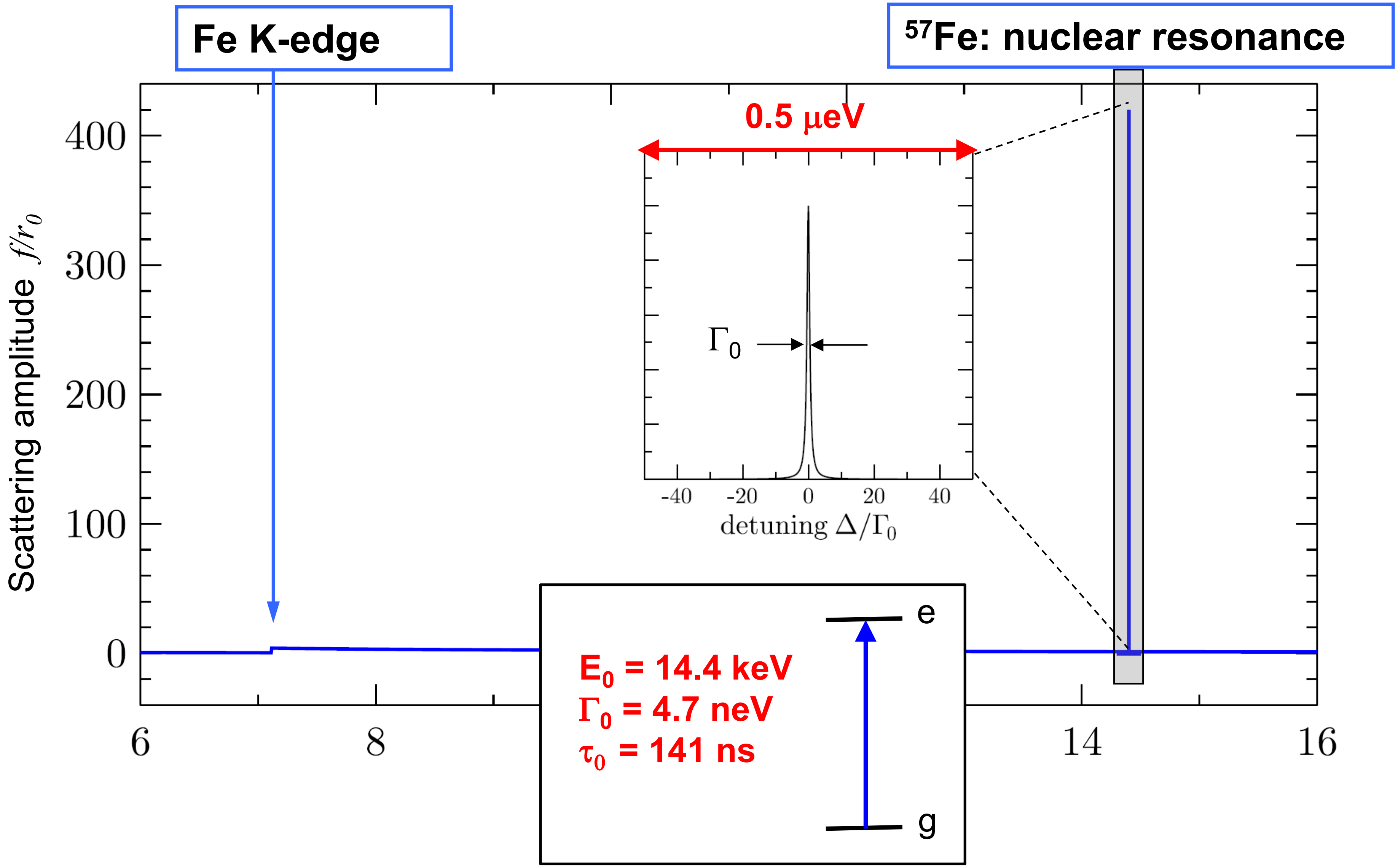
$$n = 1 - \delta$$

$$\delta \approx 10^{-5} - 10^{-6} \text{ for } \lambda = 0.1\,\text{nm}$$

Critical angle of total reflection

$$\varphi_c = \sqrt{2\delta}$$

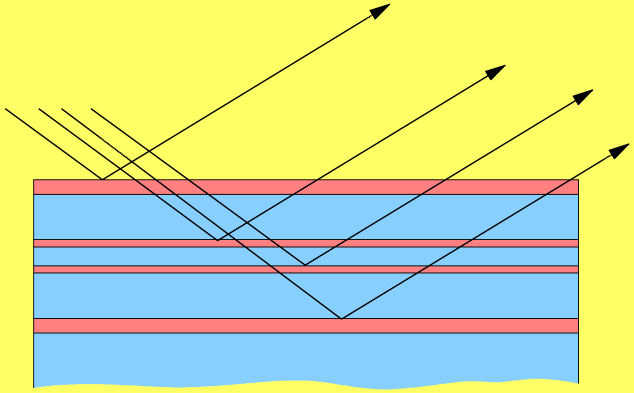
X-ray Optical Properties of Matter (Example: ^{57}Fe)

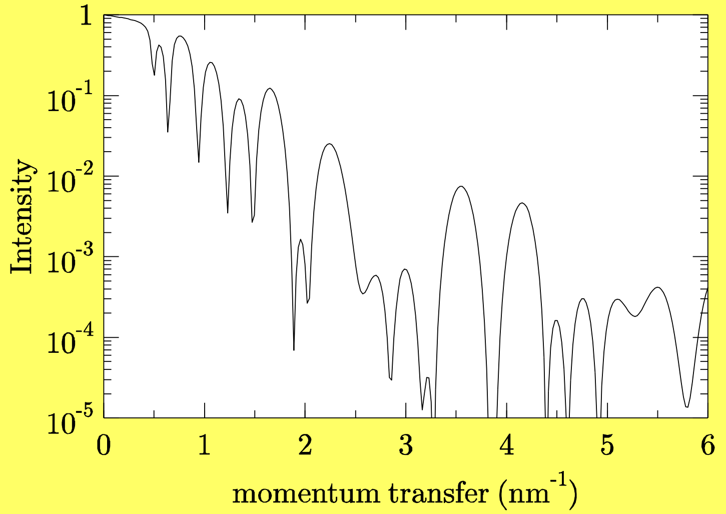


E_0 = 14.4 keV
Γ_0 = 4.7 neV
τ_0 = 141 ns

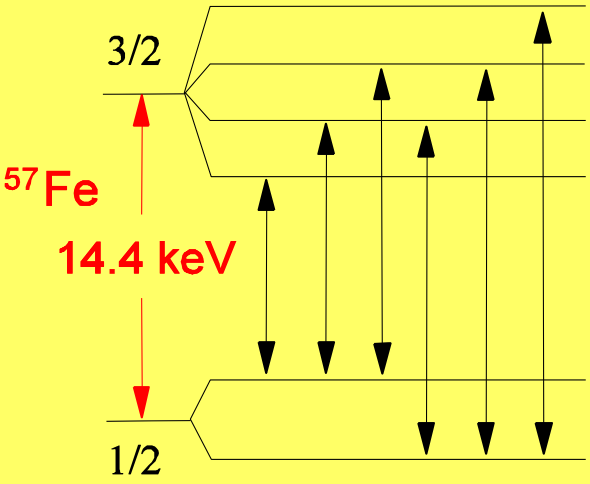
Structure Determination in SpaceTime (2)

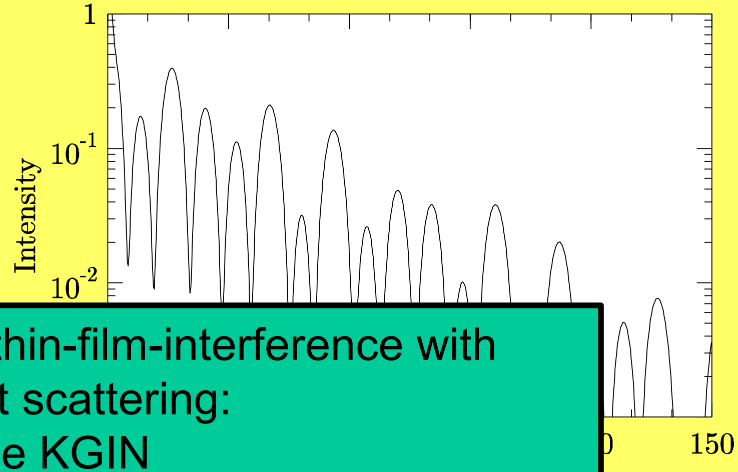
Real Space: Thin-film interfaces





Energy Space: Nuclear resonances





Combination of thin-film-interference with nuclear resonant scattering:
CONUSS module KGIN

Temporal beats and magnetization direction

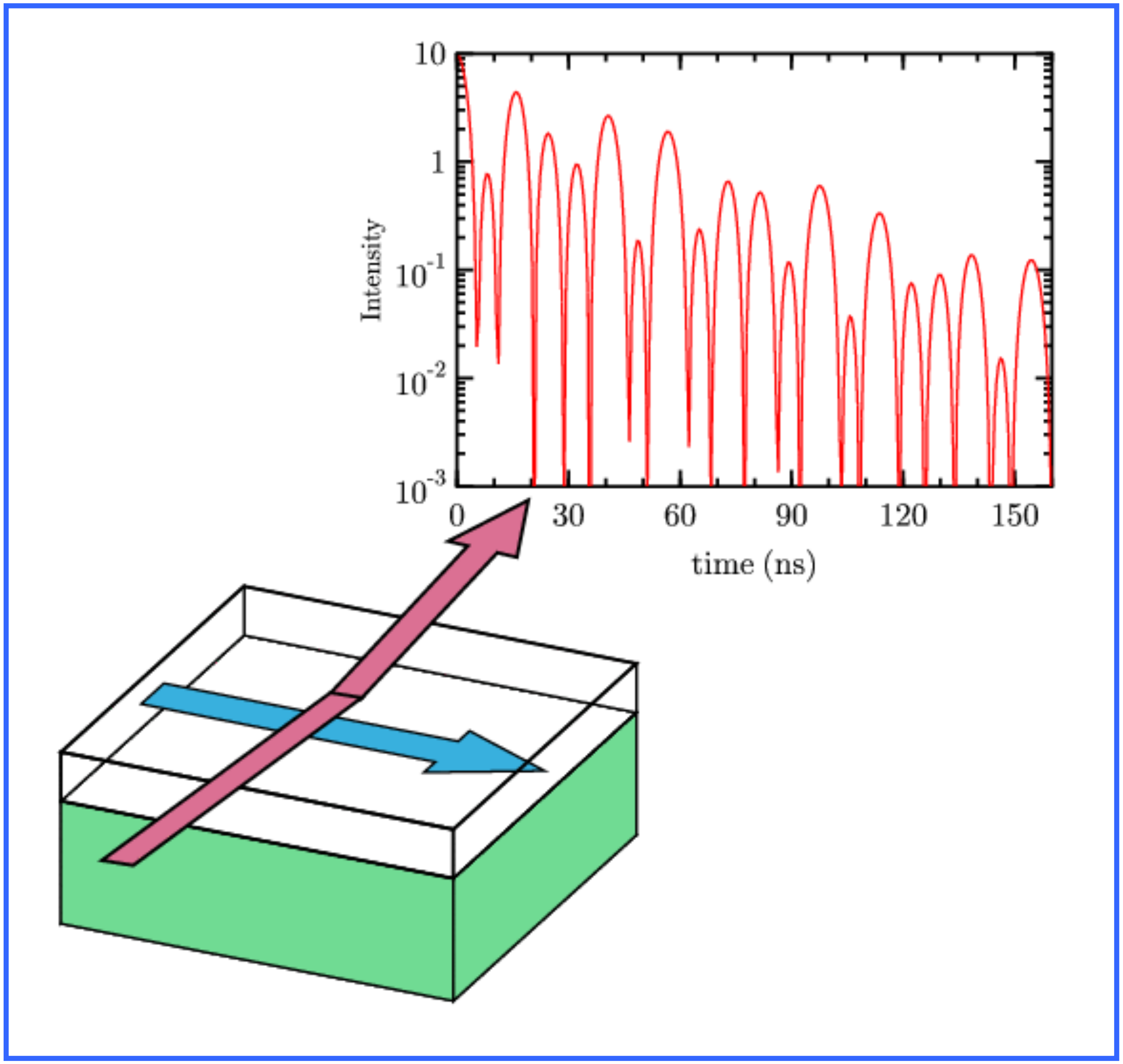
The temporal beat pattern sensitively depends on the orientation of the

Magnetization M

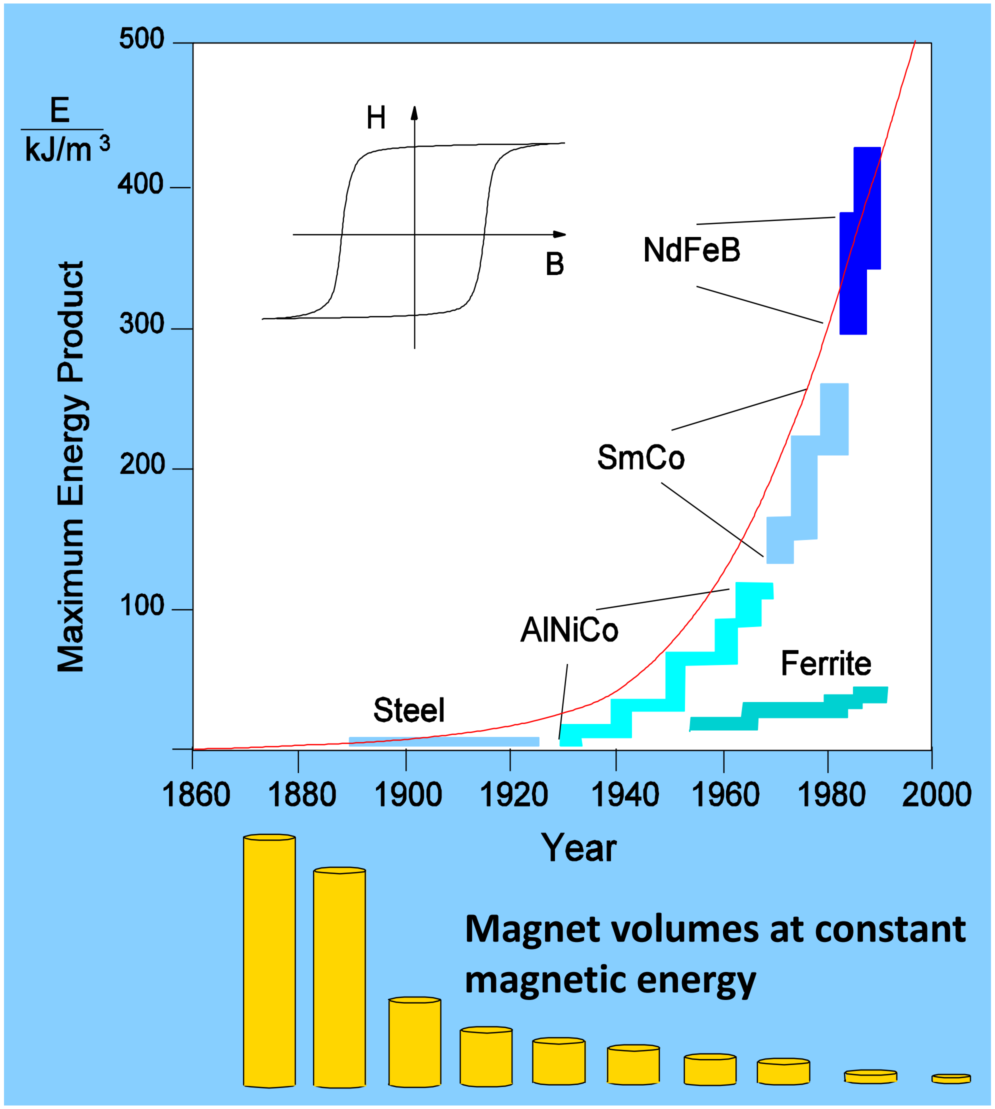
relative to the

Photon wavevector k_0

→ Use <u>isotopic probe layers</u> to investigate the depth dependence of magnetic properties



Permanent Magnets: Evolution of the Energy Product

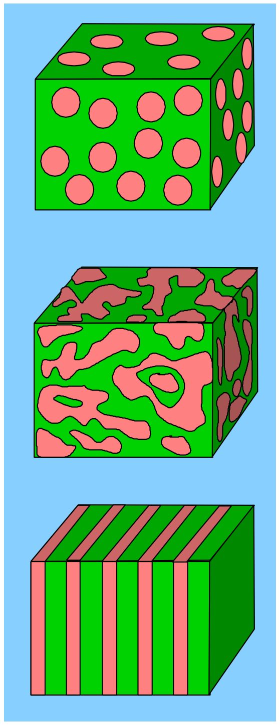


Magnet volumes at constant magnetic energy

Exchange hardening in nanostructured two-phase systems:

Hard phase with high coercivity

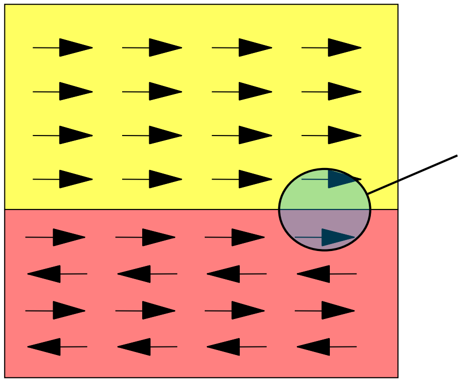
Soft phase with high magnetization



R. Skomski and J. Coey: PRB 48, 15812 (1993)

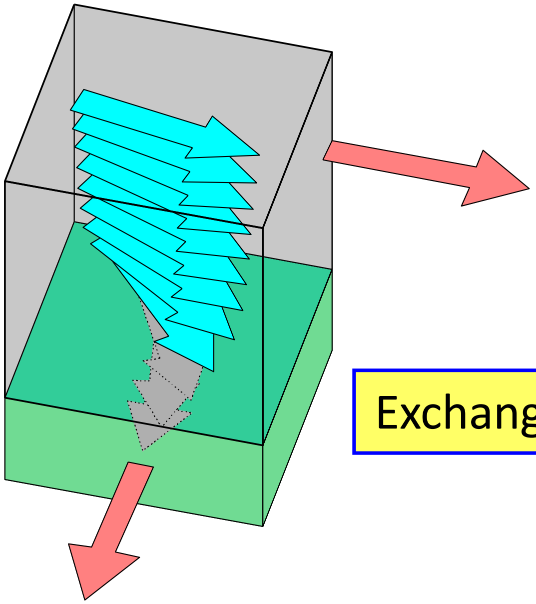
Exchange-Coupled Bilayers

Ferromagnet/Antiferromagnet



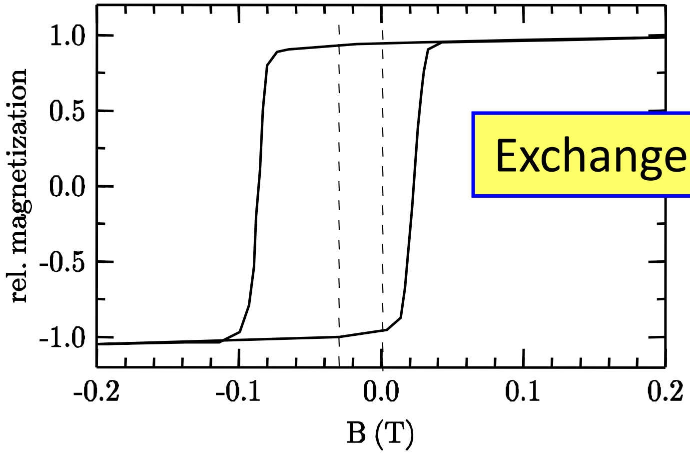
What happens at the interface ?

Hard magnet/Soft magnet



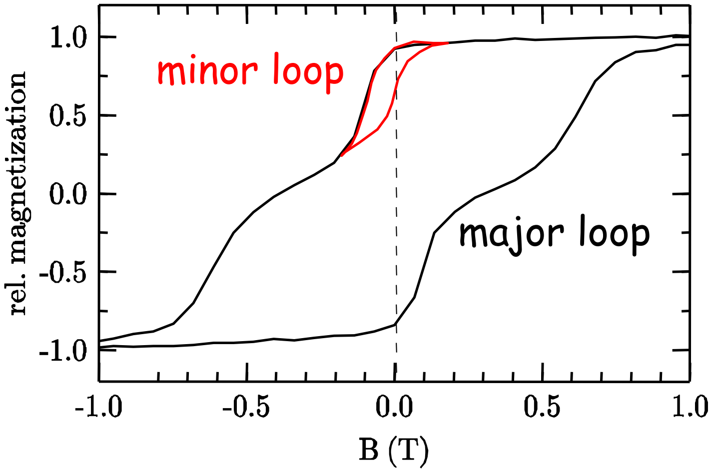
Exchange spring

Magnetic hysteresis



Exchange bias

Magnetic hysteresis



The Spin Structure of Magnetic Bilayers

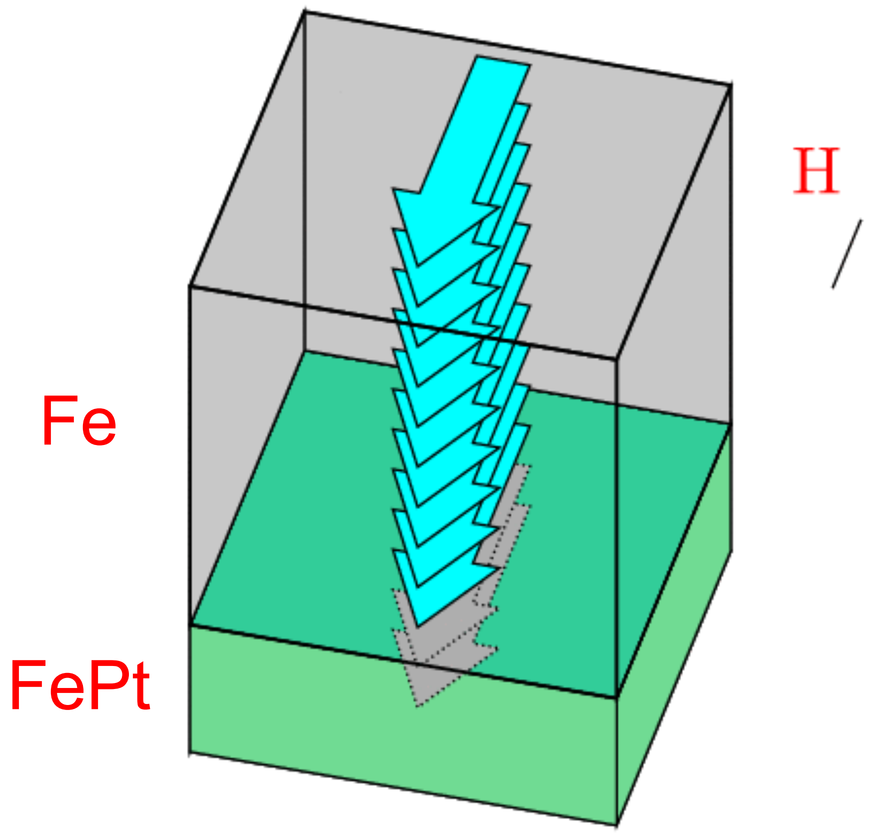
Fe on FePt

Soft – magnetic Fe

Hard – magnetic FePt
with uniaxial anisotropy

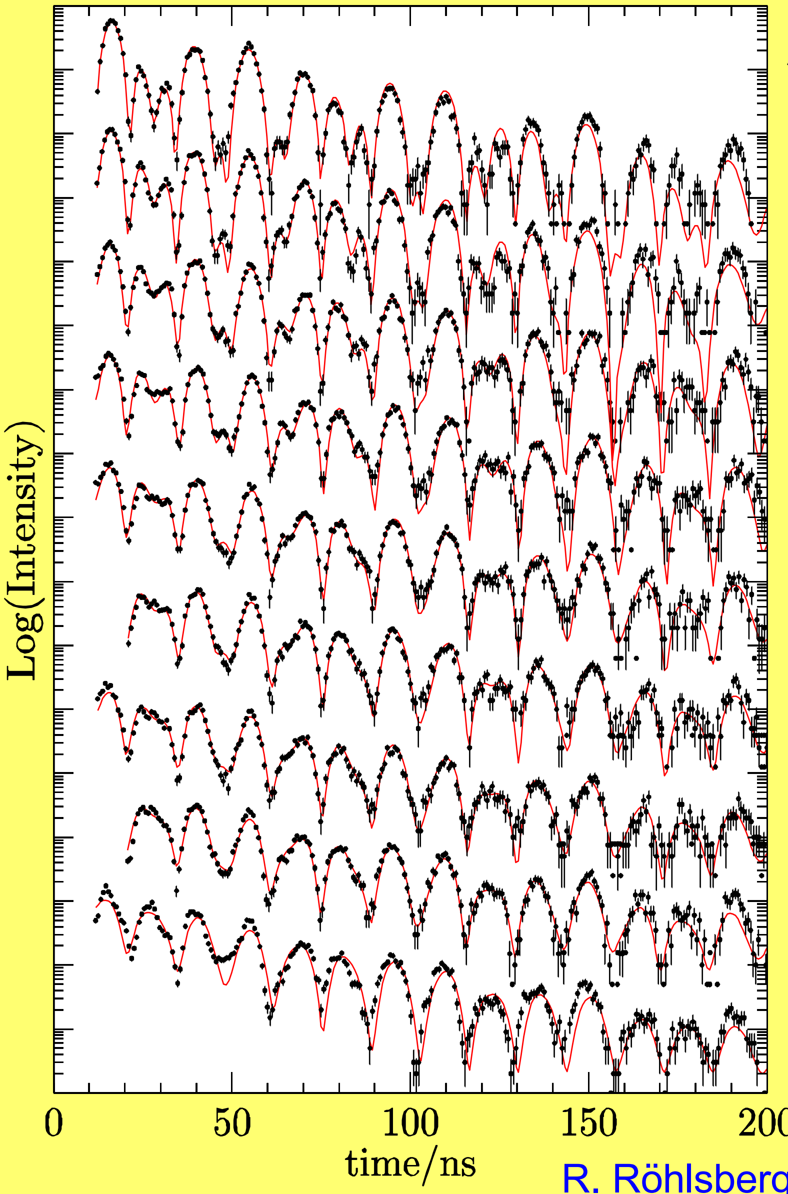
- Exchange coupling at the interface: Parallel alignment of Fe and FePt moments
- With increasing distance from the interface: Coupling becomes weaker
- External field H induces spiral magnetization
- Return to parallel alignment for H = 0

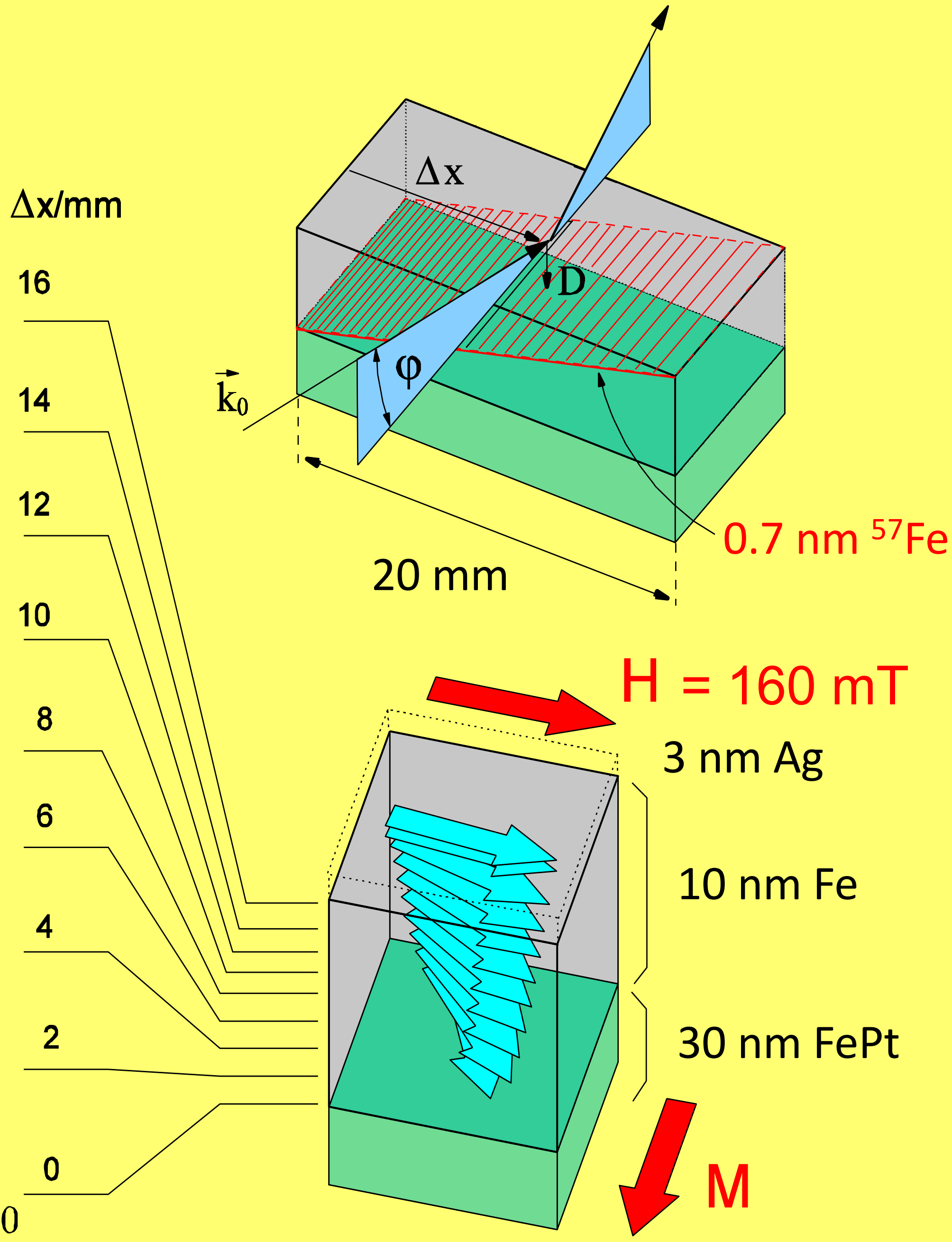
Exchange-Spring magnets



Imaging the Internal Spin Structure of Exchange-Spring Magnets

Time spectra of nuclear resonant scattering

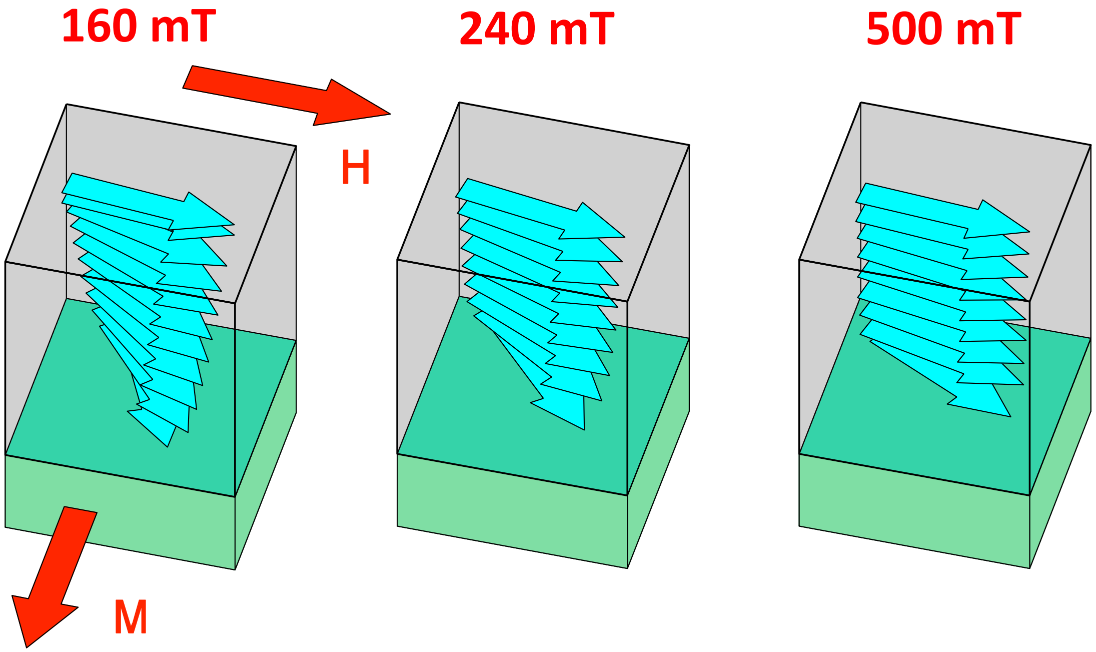


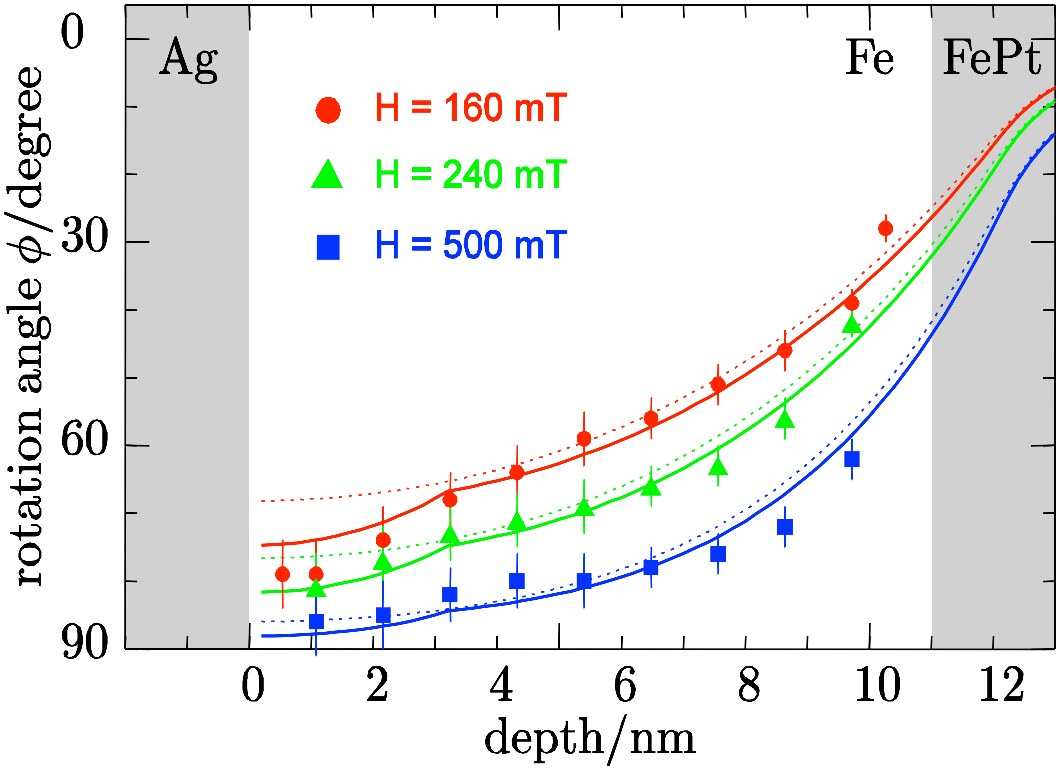


R. Röhlsberger et al., PRL 89, 237201 (2002)

Domain wall compression with increasing external field

Fe on FePt





Simulation of Exchange-Spring Layer Systems

E. Fullerton et al. PRB 58, 12193 (1998)

$$E = -\sum_{i=1}^{N-1} \frac{A_{i,i+1}}{d^2} \cos(\varphi_i - \varphi_{i+1})$$

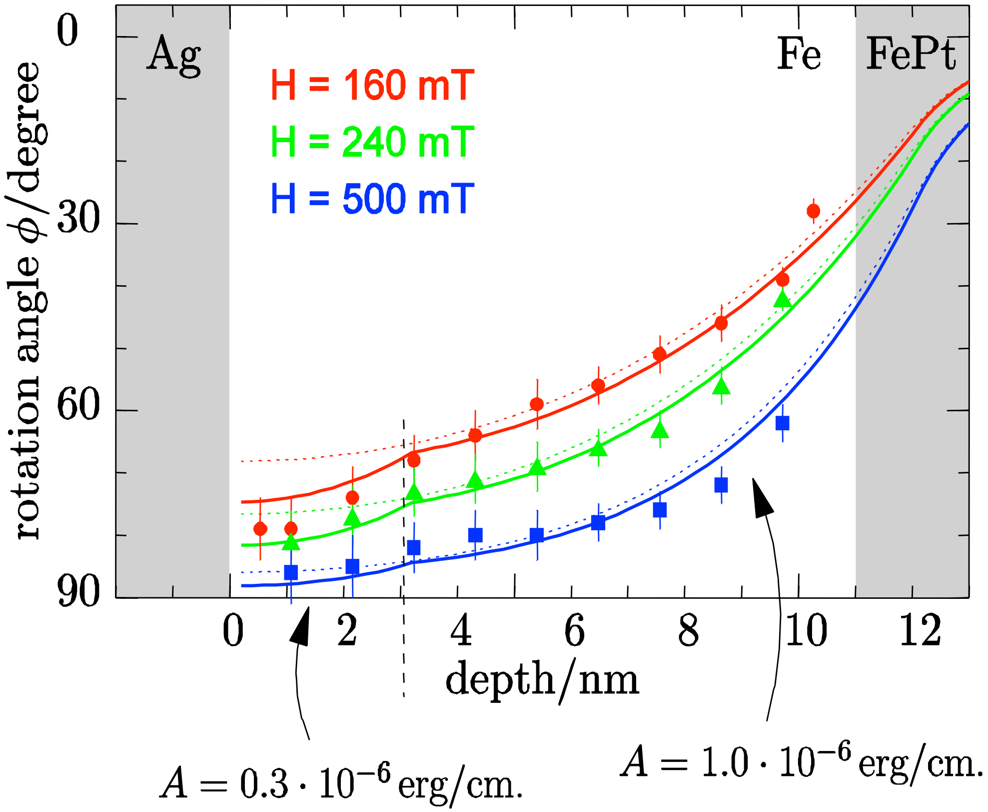
Exchange

$$-\sum_{i=1}^{N} K_i \cos^2 \varphi_i$$

Anisotropy

$$-\sum_{i=1}^{N} H M_i \cos(\varphi_i - \varphi_H)$$

Dipolar energy



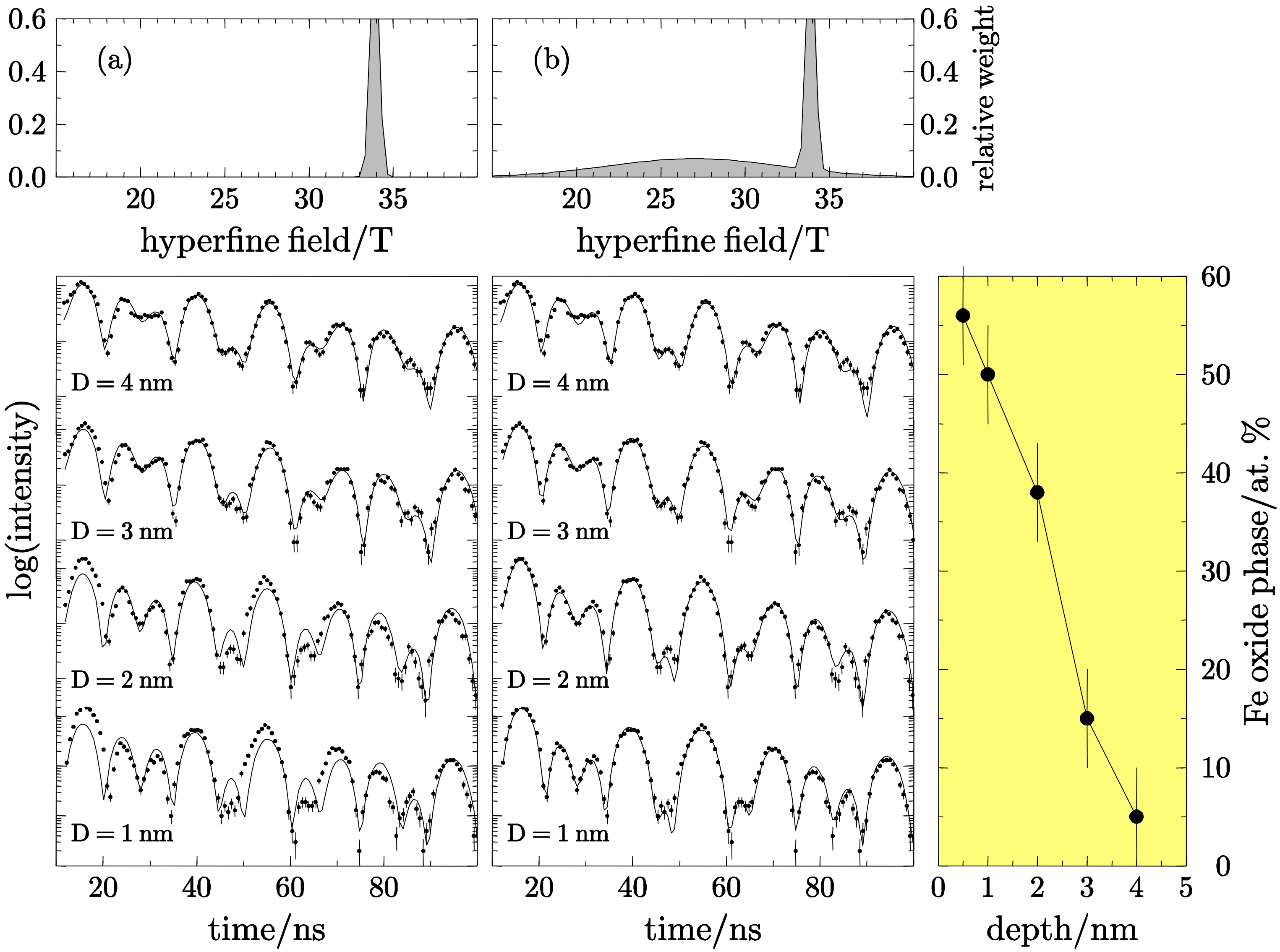
Divide the layer system into N sublayers of thickness d

Minimize the magnetic free energy

$$\frac{\partial E}{\partial \varphi_i} = 0$$

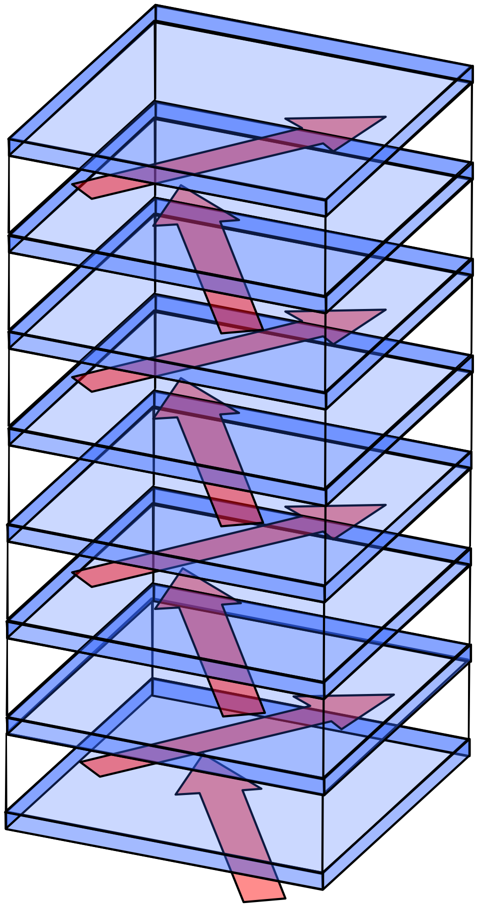
Depth profiling of magnetic properties

Depth Dependence of the Oxide Phase

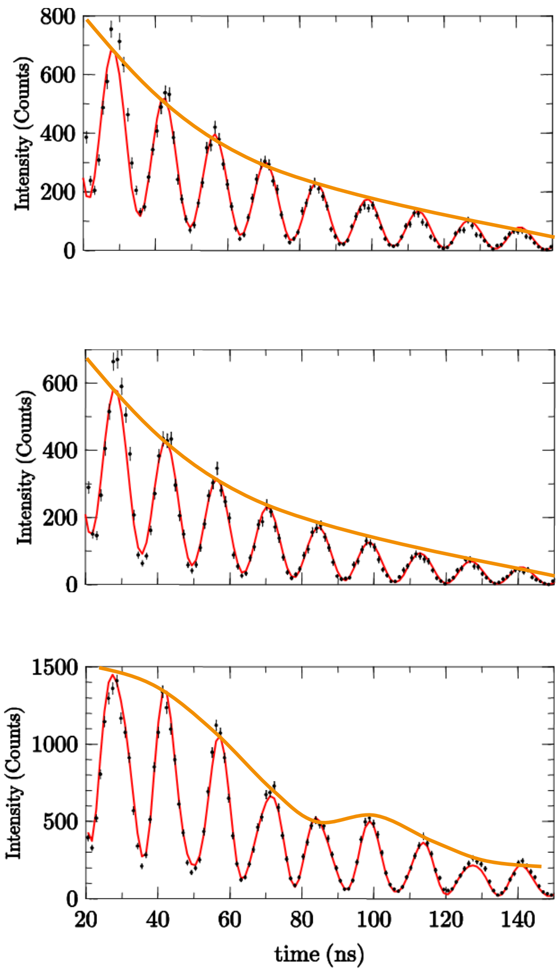


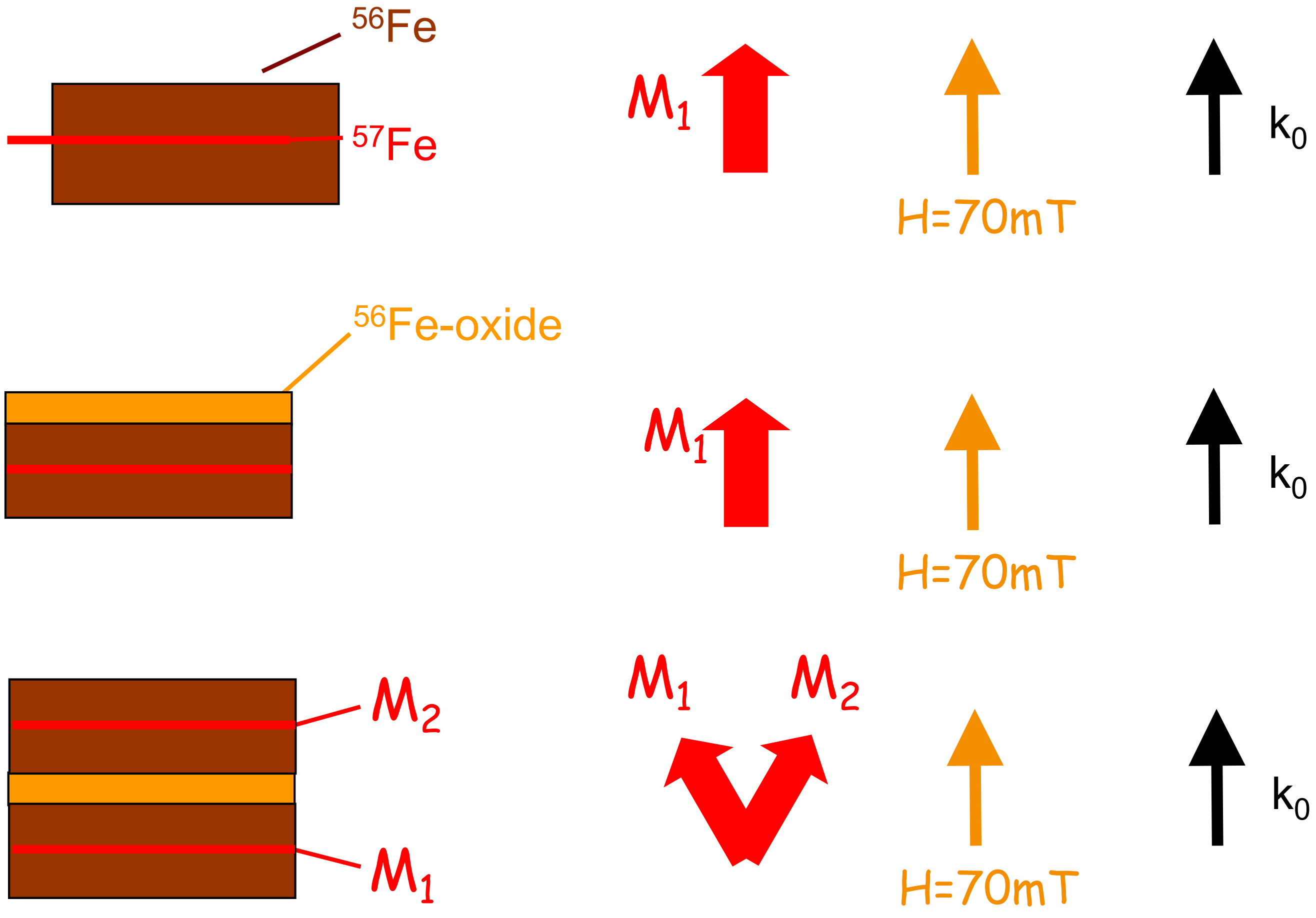
RR, H. Thomas, K. Schlage, T. Klein, J. Magn. Magn. Mater. 282, 329 (2004)

New magnetic order in Fe/Fe-oxide heterostructures



(1) New magnetic order in Fe/Fe-oxide heterostructures





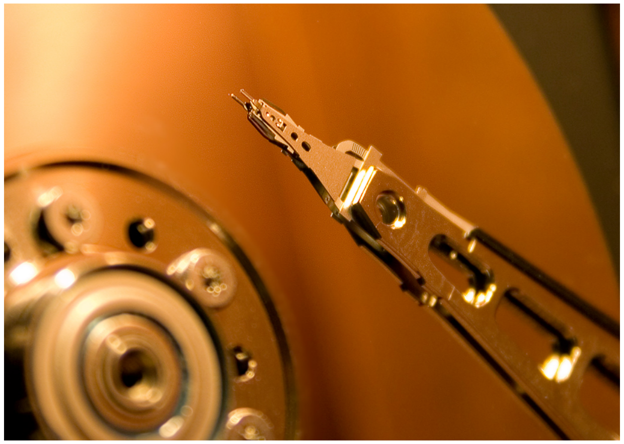
Δθ = 60° - 70 mT field

Δθ = 85° - in remanence

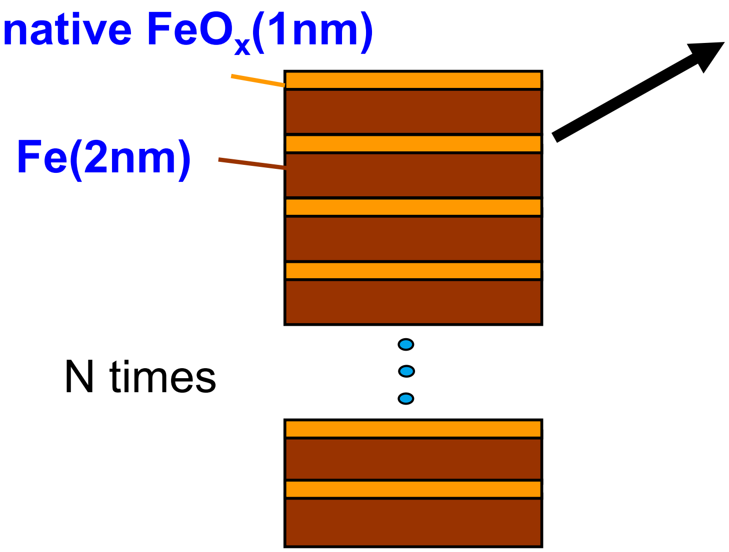
(1) New magnetic order in Fe/Fe-oxide heterostructures

Magnetic thin films and nanostructures for high-density magnetic recording

→ Read heads are subject to high-frequency magnetic fields



Eddy current problem at high frequencies : Possibilities with new materials?



Oxide / **insulating** spacer layer:

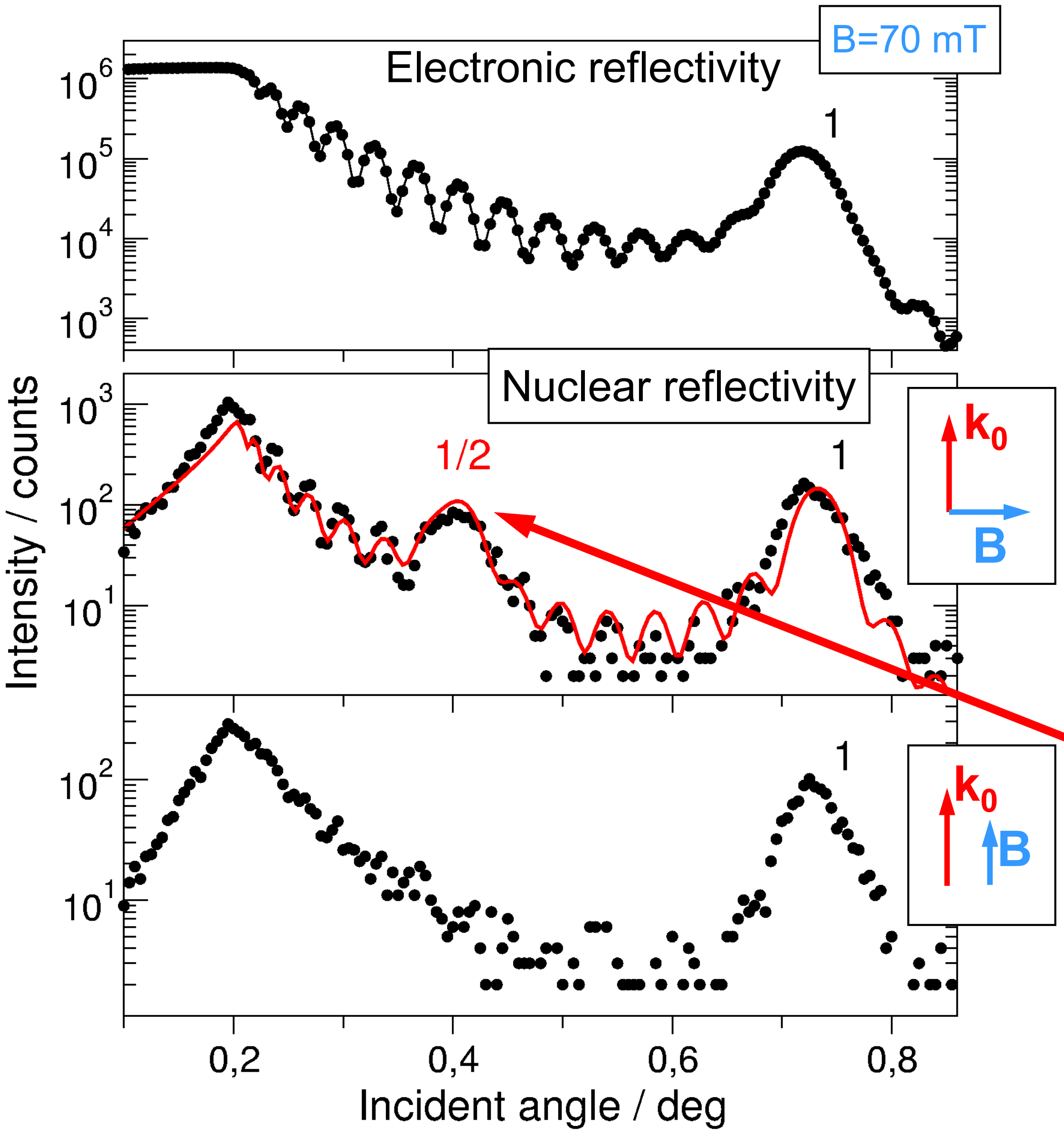
→ efficient eddy current damping[1]

Fe/Fe-oxide Heterostructures:

Materials with high magnetisation and low electrical conductivity

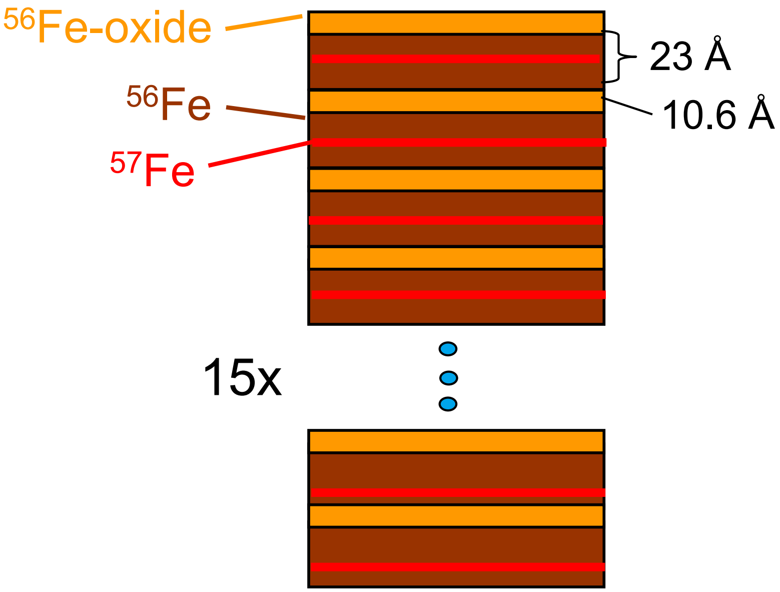
[1] G.S.D Beach and A.E. Berkowitz, PRL 91, 267201 (2003)

Fe/Fe-Oxide Multilayer: Magnetic Superstructure



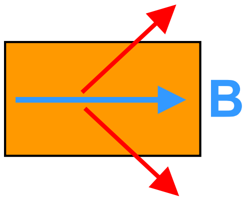
Th. Diederich et al., PRB 76, 54401 (2007)

Iron layers probed by ^{57}Fe

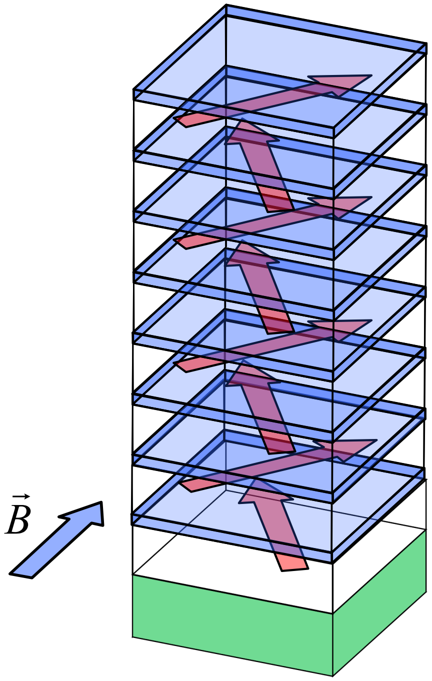


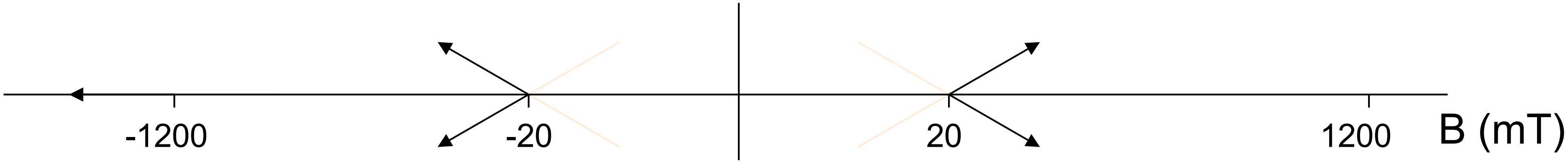
Superstructure peak

- Pure magnetic origin
- Canted moments of Fe-layers



Fe/Fe-oxide Multilayers Coupling strength and magnetization reversal





Th. Diederich, S. Couet, and RR, Phys. Rev. B 76, 54401 (2007)

What is the origin of the magnetic coupling ?

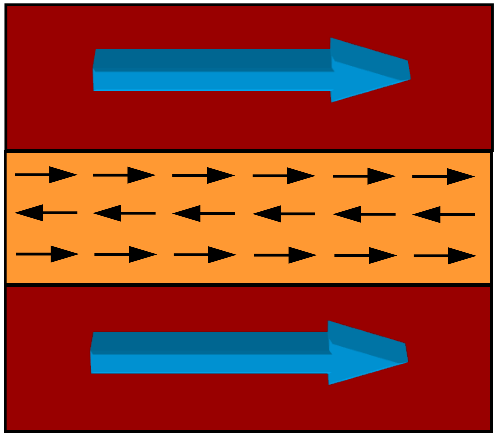
→ Investigate the nature of the buried native oxide !

Non collinear coupling in the system Fe/Fe-oxide/Fe

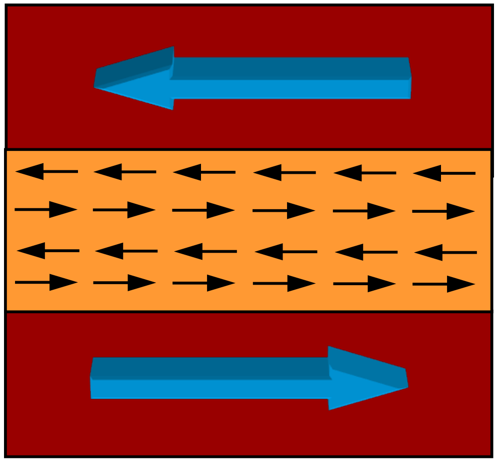
The strong coupling between the Fe layers excludes an RKKY type of interlayer coupling

→ **Coupling is mediated by an antiferromagnetically ordered oxide (at room temperature) !!**

Ideal systems, 2 possibilities:

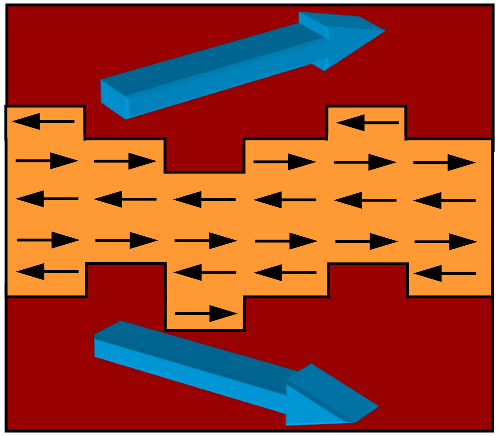


FM coupling



AFM coupling

Real system, roughness:



Spin frustration at the interfaces

↓

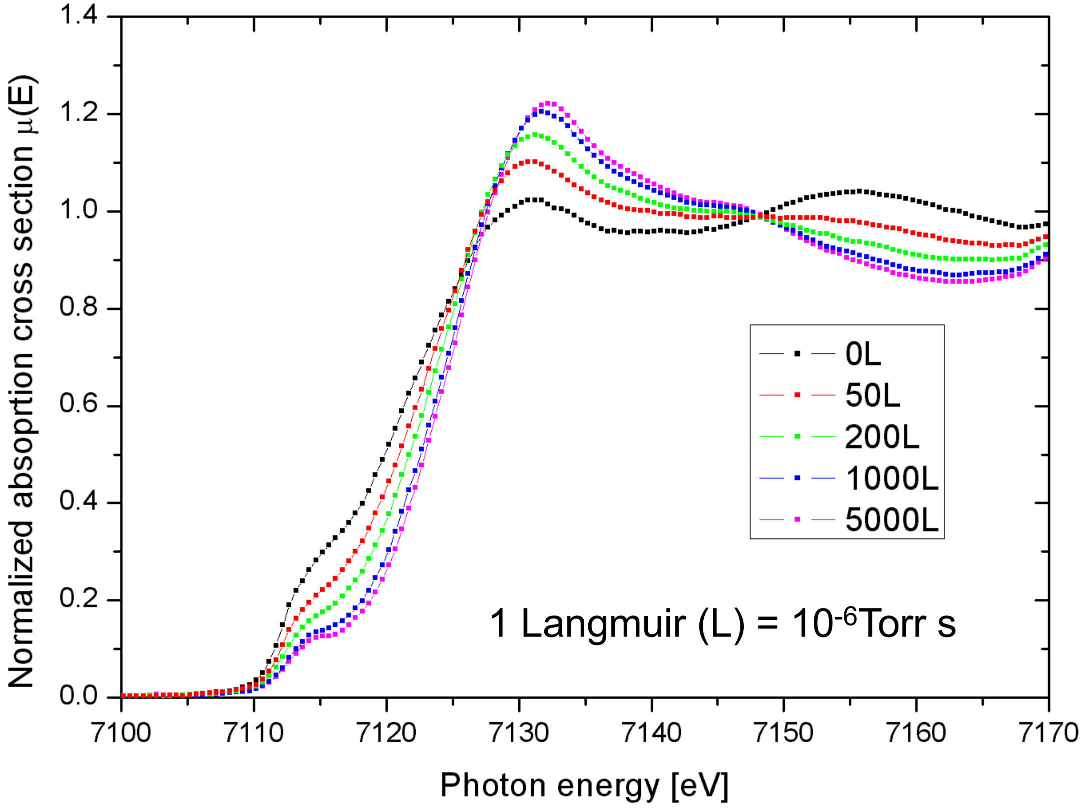
Non collinear coupling

QUESTION: How can the native oxide be antiferromagnetically ordered at room temperature?

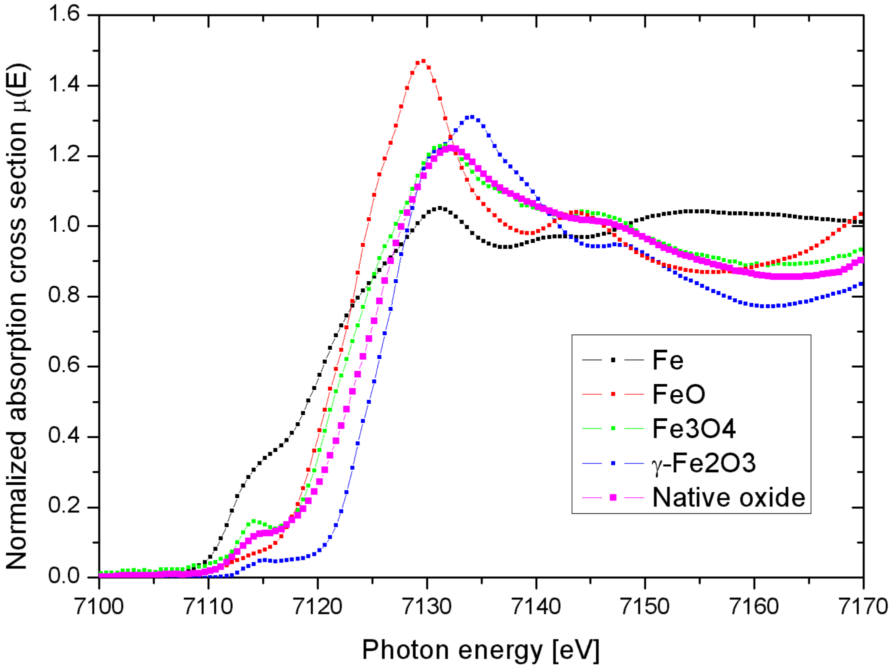
Evolution of the native oxide during growth

An in-situ XAS study

Oxidation of 8 Å Fe at 5x10^{-5} mbar



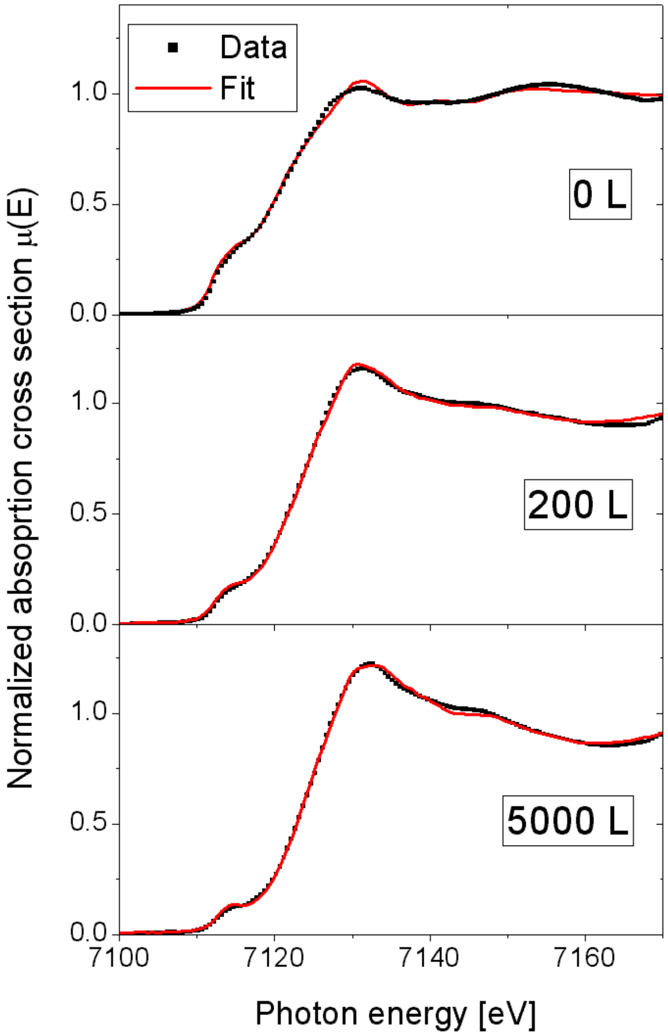
Reference spectra

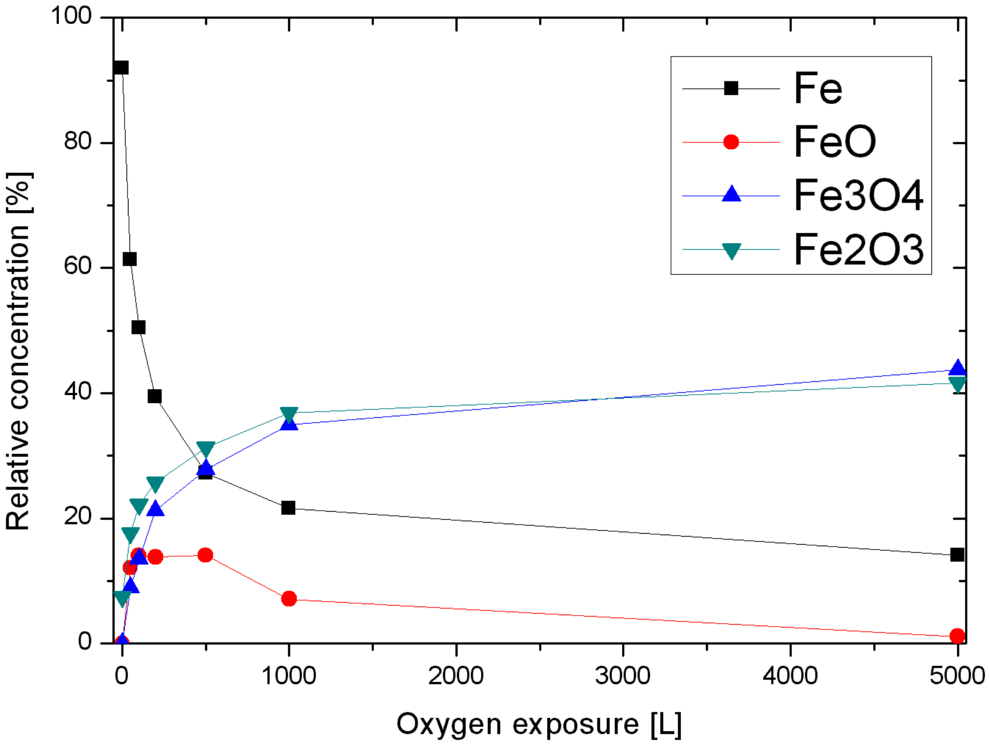


Mixture of phases at saturation

Chemistry of the *free* native oxide

Fitting XANES : **Linear combination of reference spectra** allows to extract the relative concentration of the different species

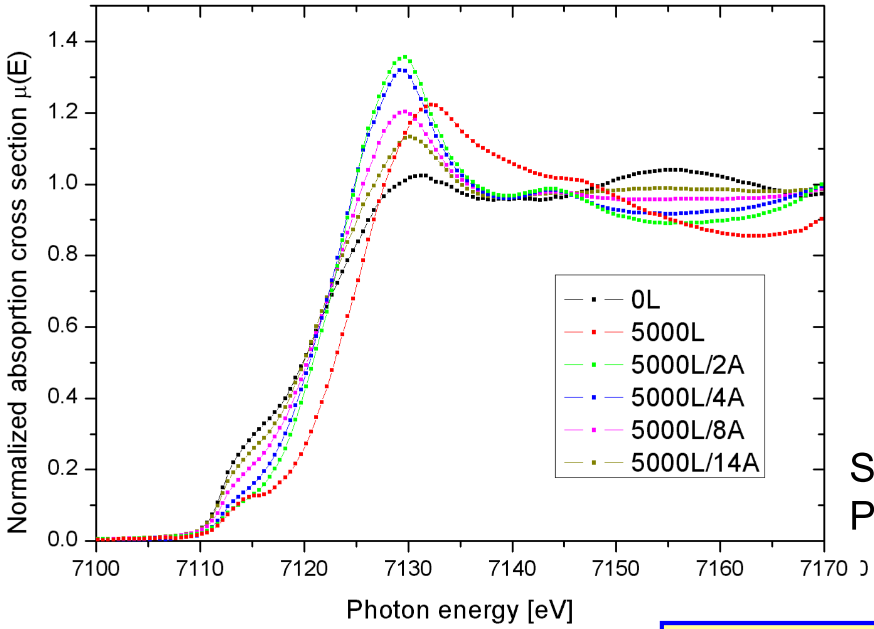




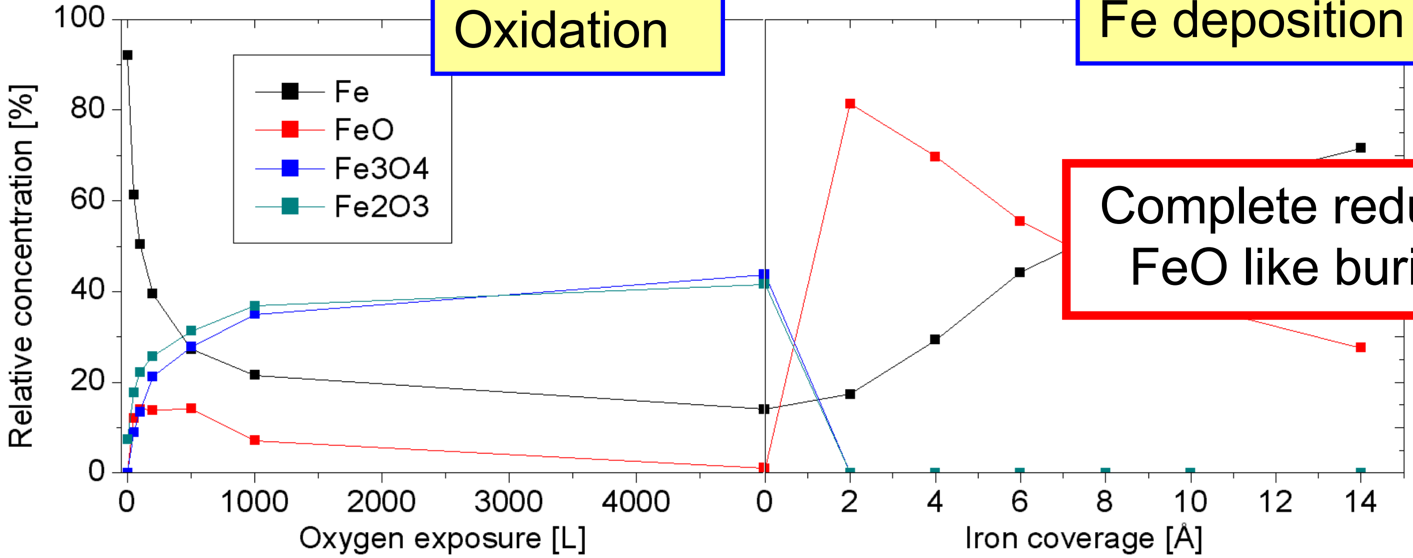
Some metallic iron exists even at saturation

Chemistry of the *buried* native oxide

Deposition of iron on its native oxide



S. Couet et al.,
Phys. Rev. Lett. 101, 056101 (2008)



Oxidation

Fe deposition

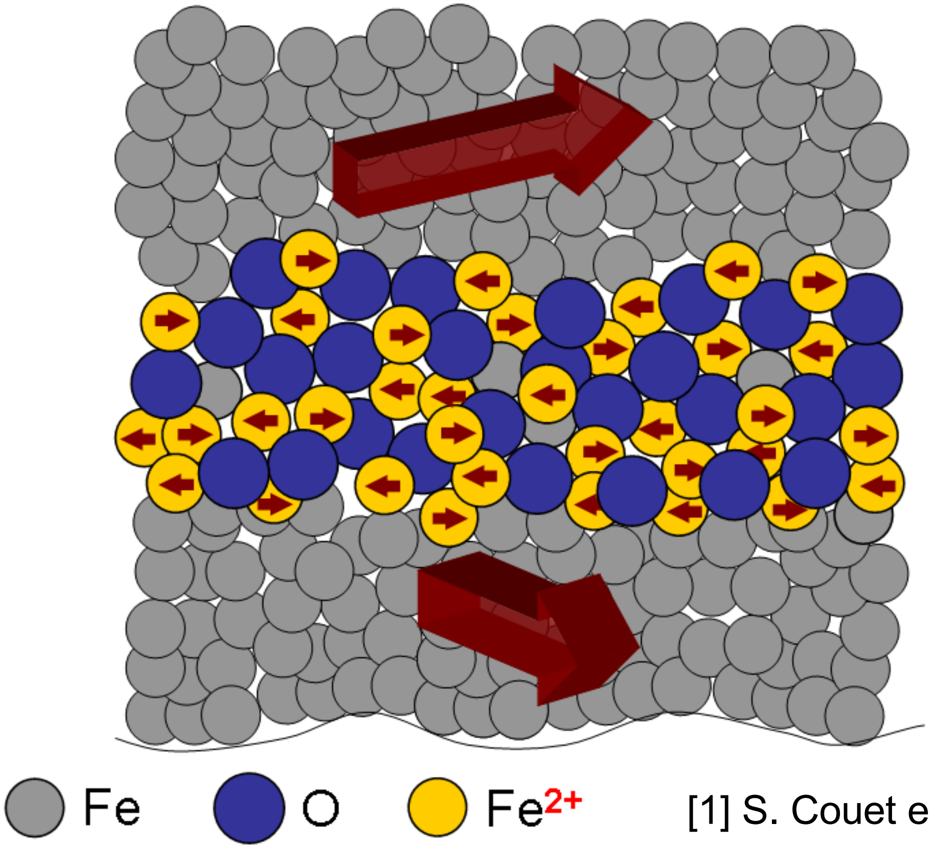
Complete reduction to an FeO like buried oxide!!

The Fe/Fe-Oxide Story: Summary

Stabilization of magnetism in the oxide

↓

Appearance of interlayer coupling



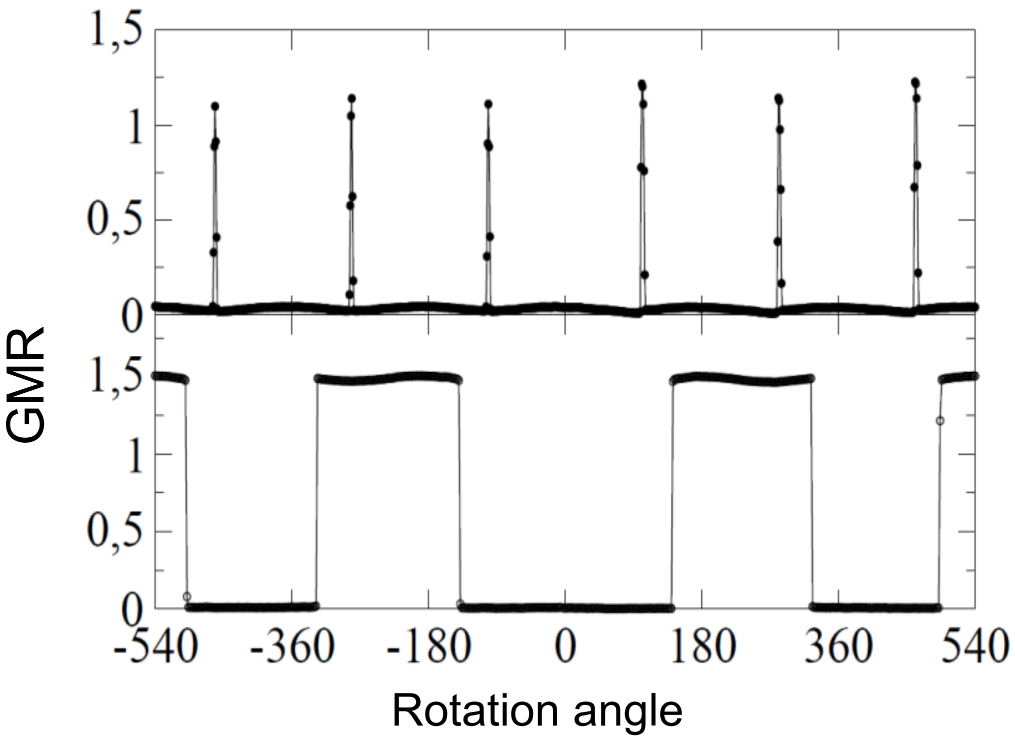
(Doctoral thesis of Sebastien Couet, Hamburg, 2008)

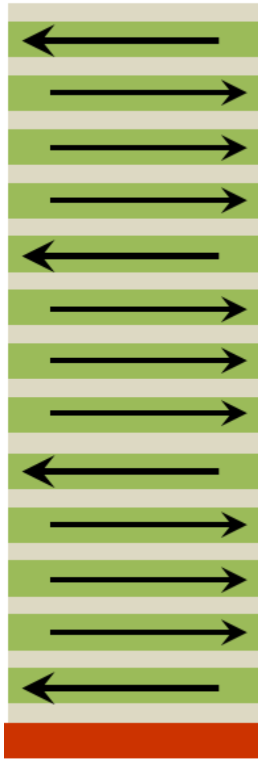
- Native oxide exhibits a mixture of chemical states – **not magnetic**
- Fe deposition leads to a FeO-like structure – **not magnetic [1]**
- FM order in the metal stabilizes **AFM order in the oxide at room temperature [2]**
- Exchange coupling at both interfaces of the oxide leads to a **non-collinear arrangement of the Fe layers**

[1] S. Couet et al., Phys. Rev. Lett. **101**, 056101 (2008)

[2] S. Couet et al., Phys. Rev. Lett. **103**, 097201 (2009)

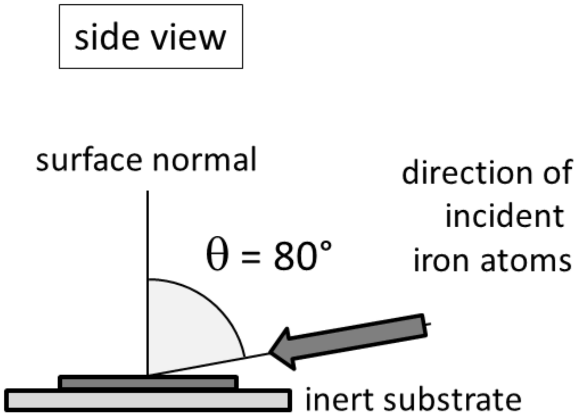
Spin-structured multilayers:
A new class of materials for precision spintronics





Tuning the Magnetic Anisotropy of Ultrathin Films via OID

OID = oblique incidence deposition



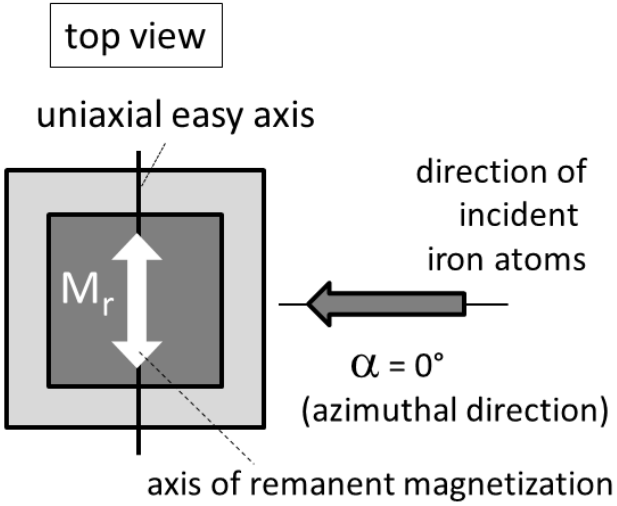
side view

surface normal

θ = 80°

direction of incident iron atoms

inert substrate



top view

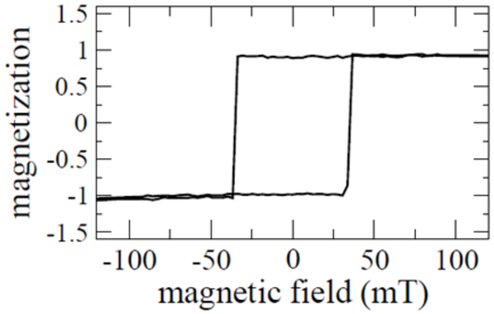
uniaxial easy axis

M_r

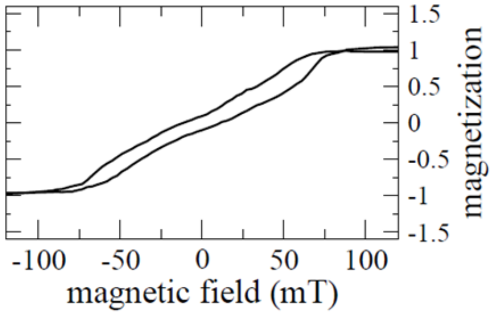
direction of incident iron atoms

α = 0° (azimuthal direction)

axis of remanent magnetization



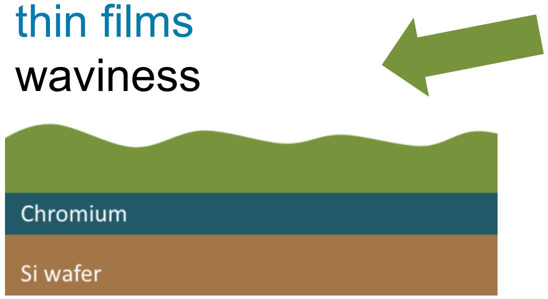
easy axis



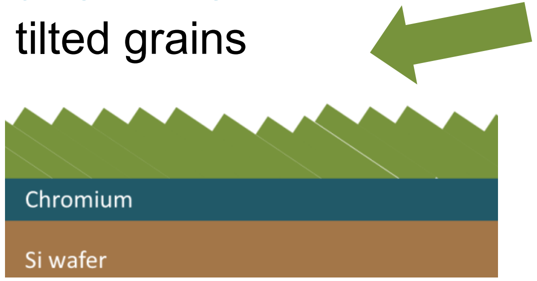
hard axis

Origin

thin films
waviness



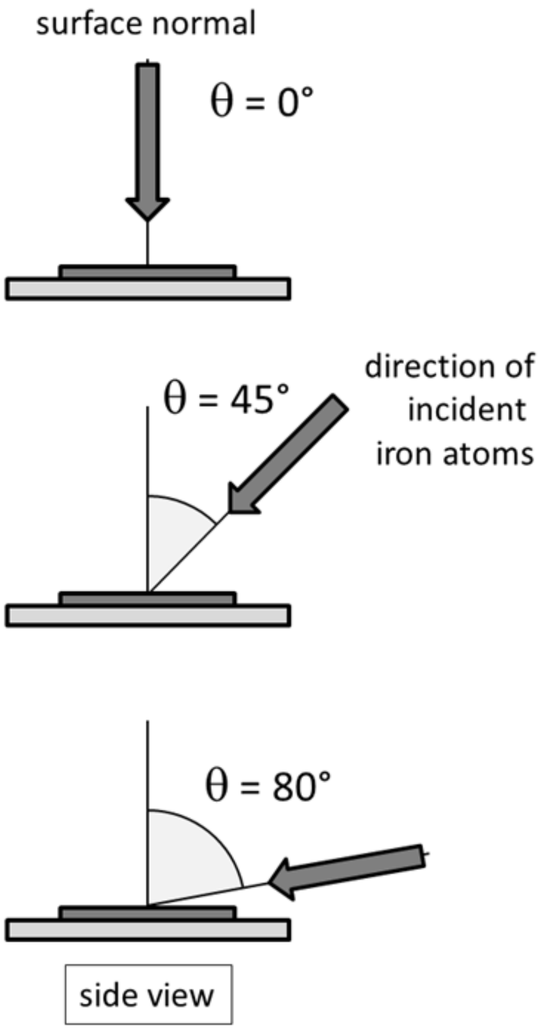
thick films
tilted grains

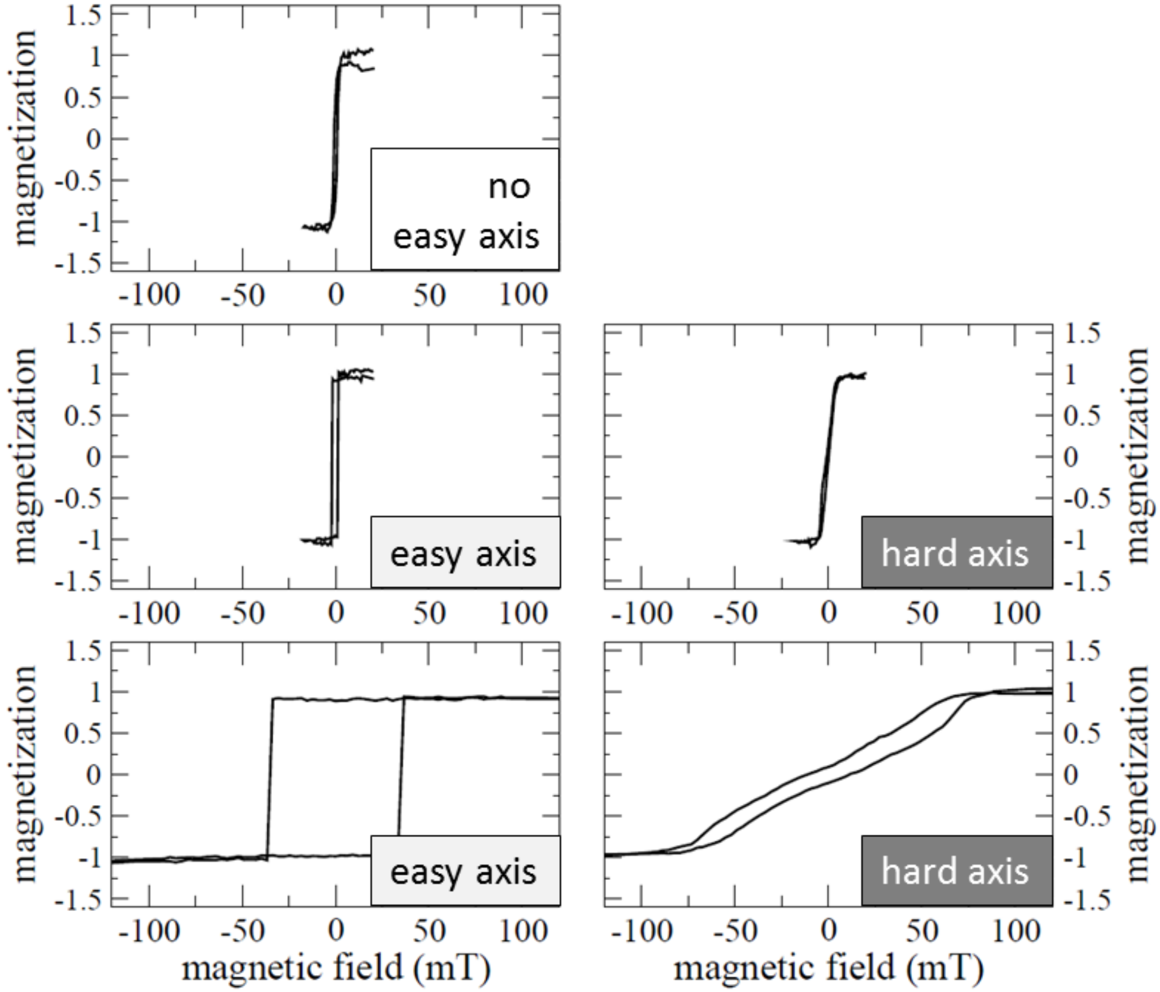


Oblique incidence deposition allows to induce a strong and adjustable magnetic anisotropy in ultrathin magnetic films.

Spin-structured multilayers: A new class of materials for precision spintronics

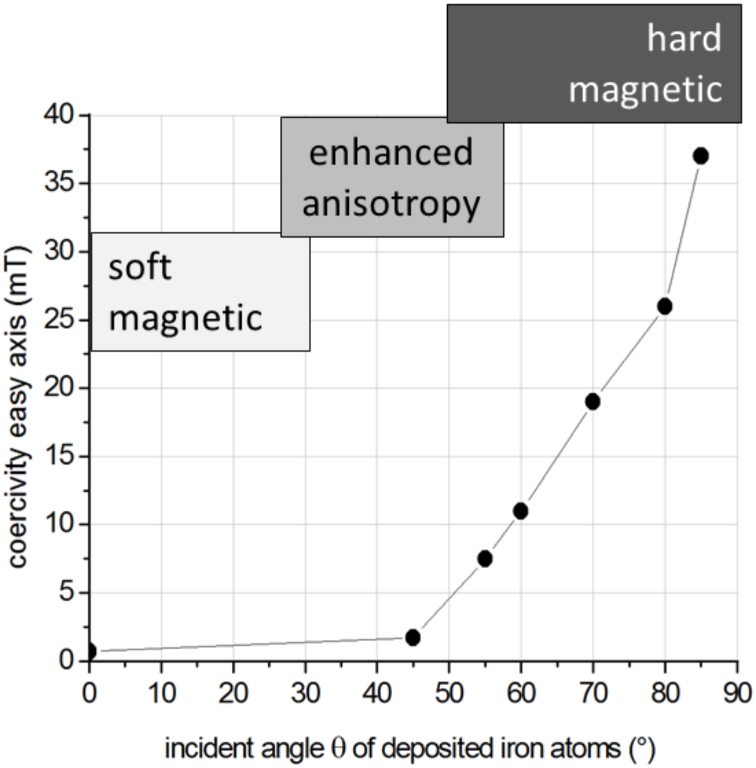
Magnetic Hardening of a Single Iron Layer

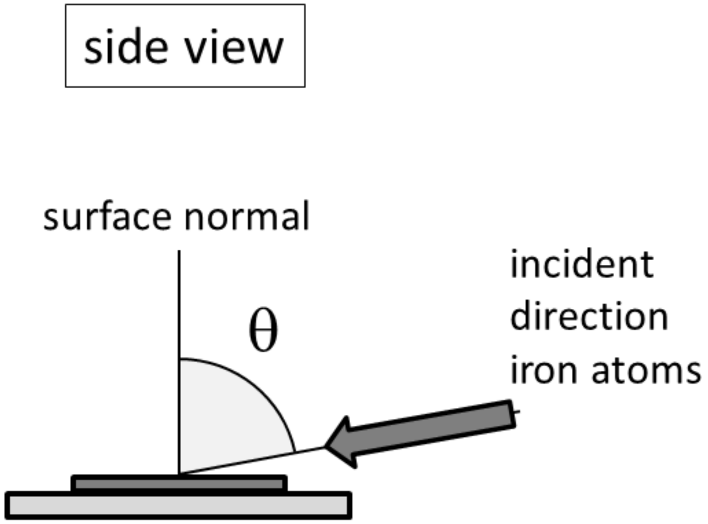




Spin-structured multilayers: A new class of materials for precision spintronics

Magnetic Hardening of a Single Iron Layer

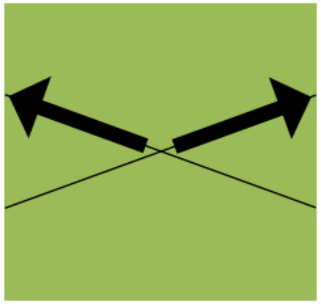




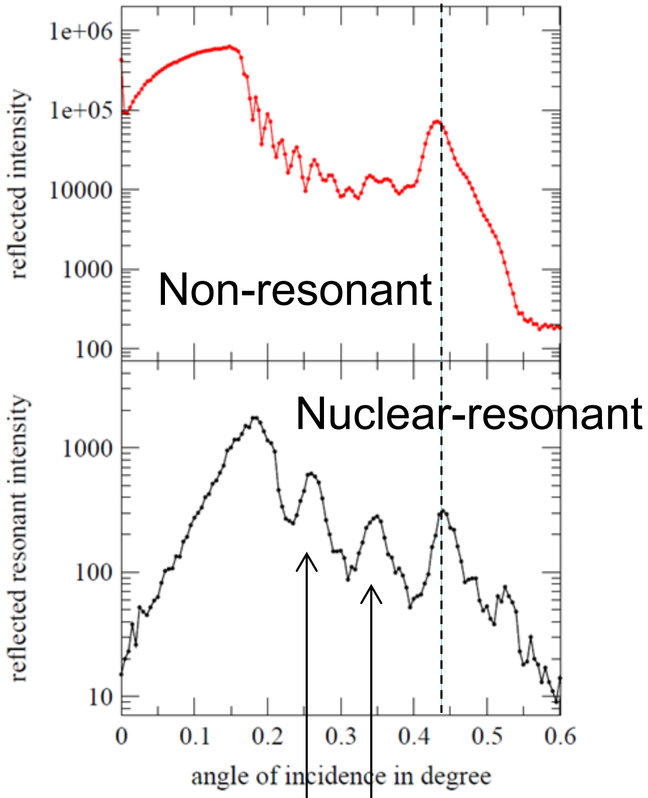
Applicable also to multilayer systems with crossed easy axis?

Probing Spin Structures in Multilayers via Nuclear Resonant Diffraction

Two magnetic sublattices

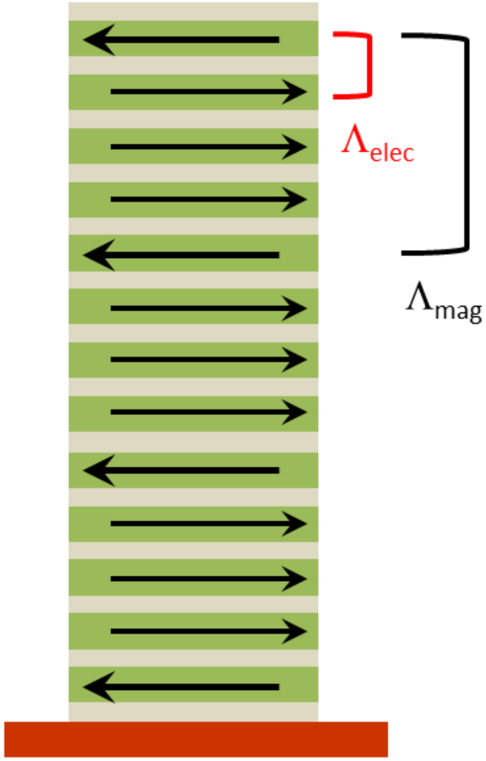


Reflectivity



Superstructure Bragg-peaks

$[^{57}Fe/C]_{13}$



Arbitrary in-plane spin structures via oblique-incidence deposition

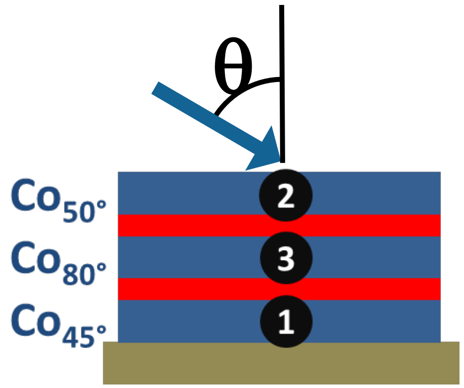
→ Magnetic order does not rely on interlayer coupling

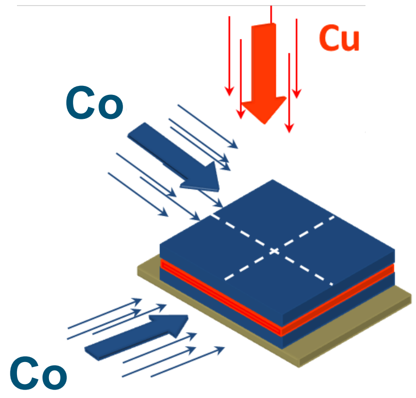
→ No limitation to prepare vertical stacking profile.

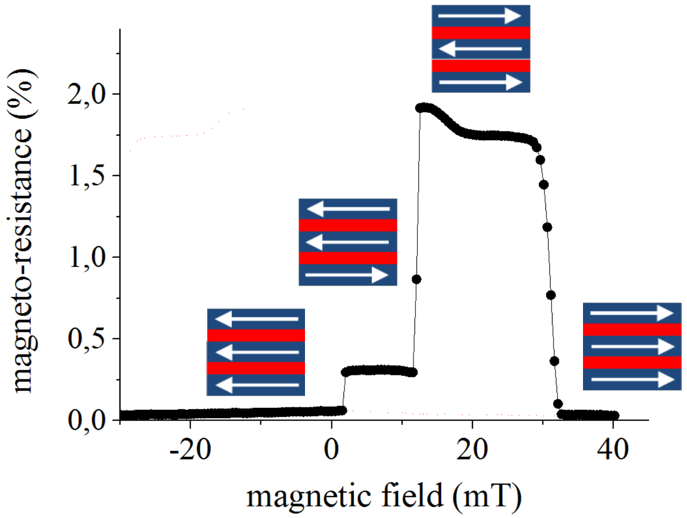
Tuning of MR via Oblique Incidence Deposition

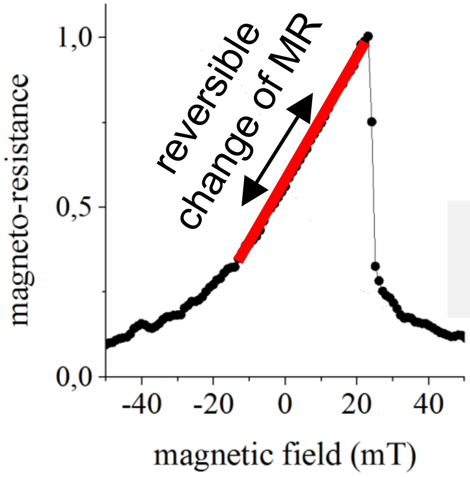
Tailored Sequential Layer Switching
via polar deposition angle

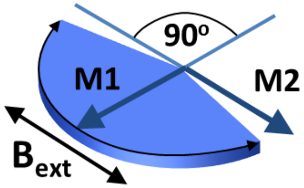
Crossed Magnetization Axes
via azimuthal deposition angle







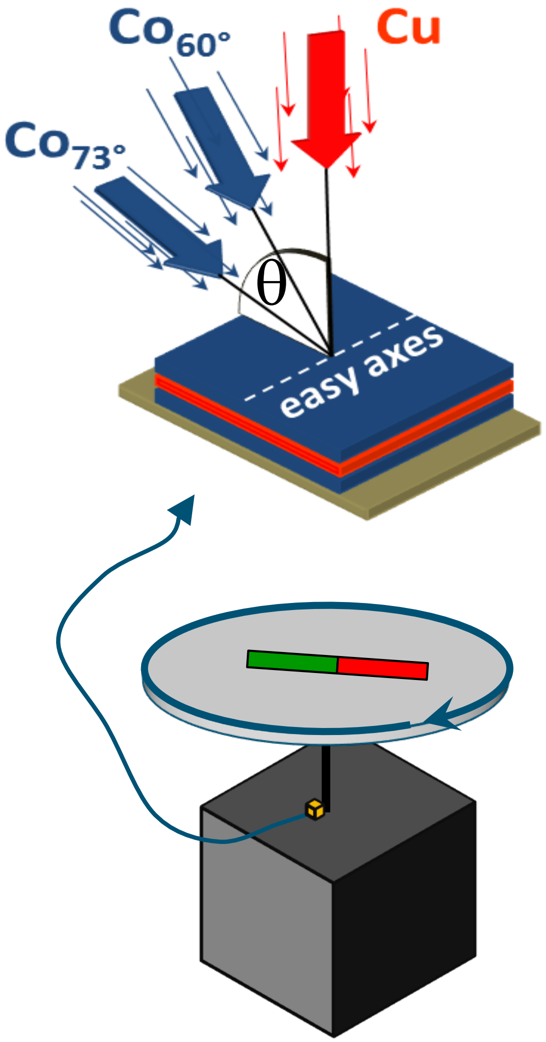


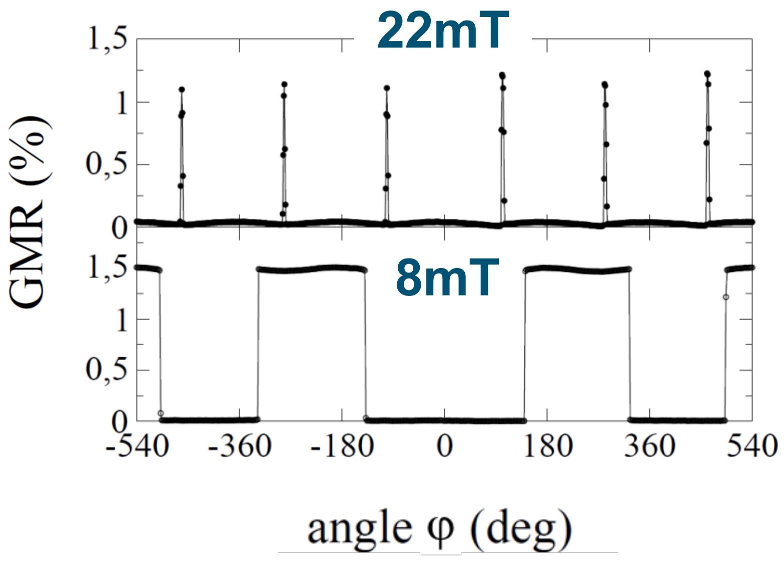


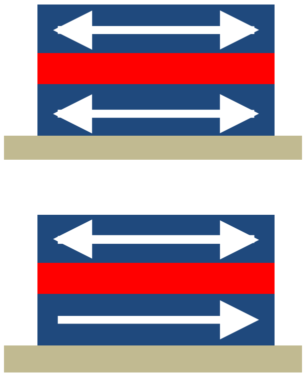
sensitive to field strength and sign!

K. Schlage et al., Adv. Funct. Mater. 26, 7423 (2016)

Application: Precision Rotational Sensing







rotary fields

- extremely **sharp switching**
- field-dependent switching characteristics
  signal shape: **peak or plateau**
- with crossed magnetization axes:
  sensitive to **frequency and sense** of rotation

European Patent EP 2 846 334 A1 (2015), pending

Determination of magnetic dynamics via nuclear resonant scattering

Nuclear Resonant Scattering for Magnonics

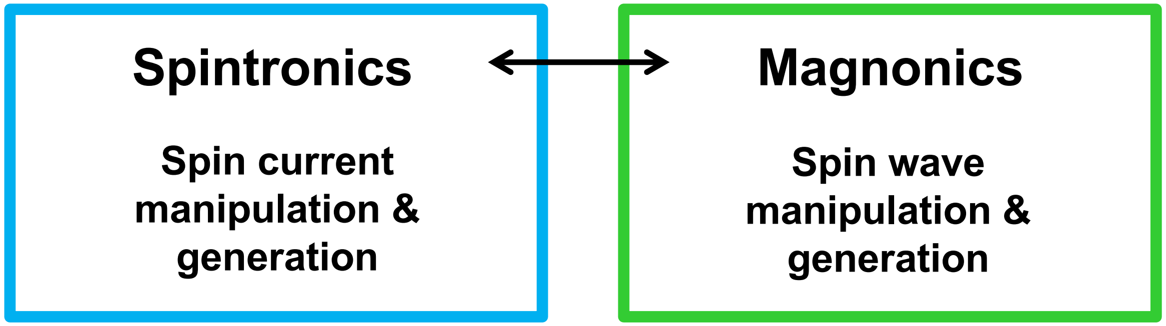
www.hitachigst.com

Magnetic resonances

magnetic storage → switching times, energy losses

Spin manipulation

functional spin devices



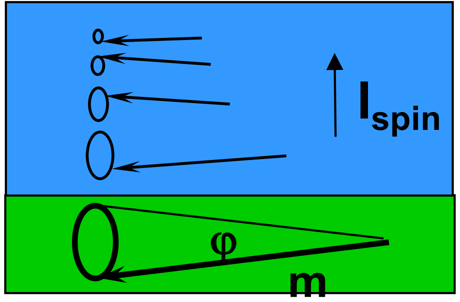
Spintronics

Spin current manipulation & generation

Magnonics

Spin wave manipulation & generation

Spin Pumping

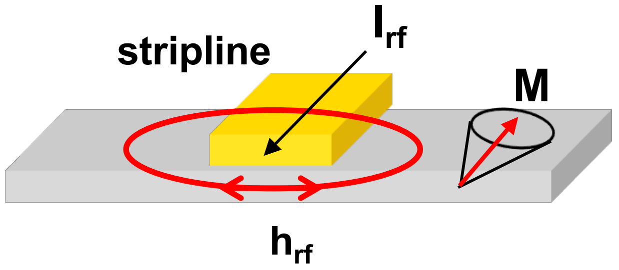


Y. Tserkovnyak et al. , Phys. Rev. Lett. 88, 117601 (2002)

Spin excitations in magnetic microstructures

Thin film system

permalloy ($Ni_{80}{}^{57}Fe_{20}$)

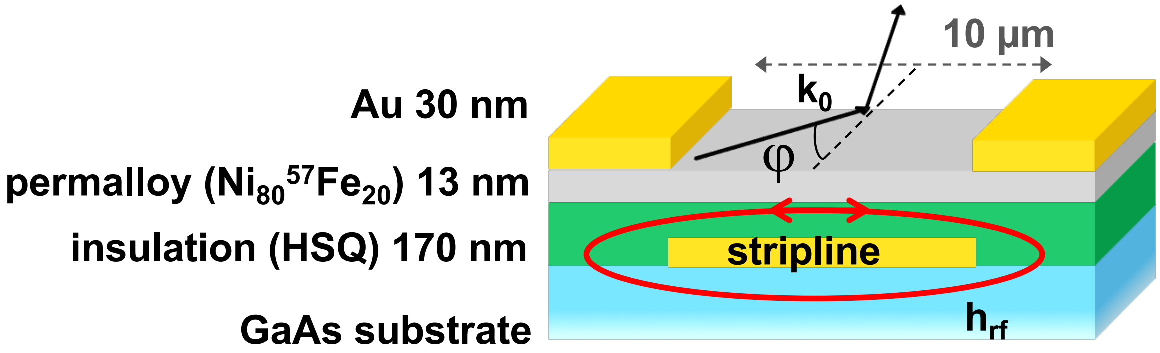


Excitation of uniform spin precession

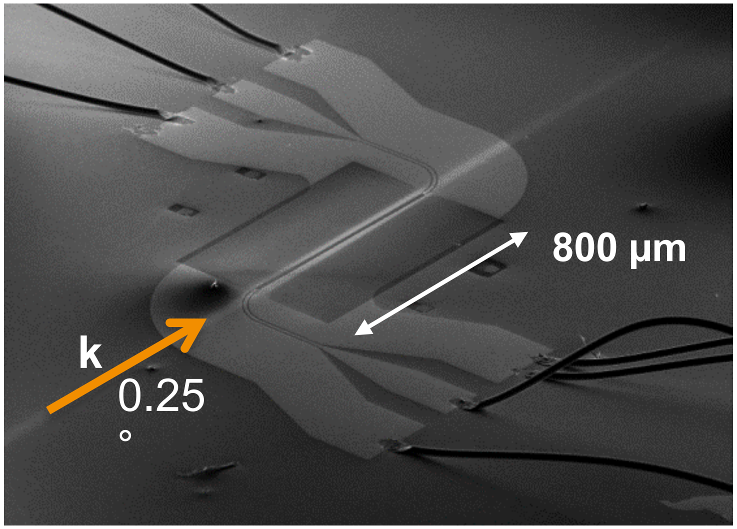
→ Kittel mode at ferromagnetic resonance

Experimental Setup

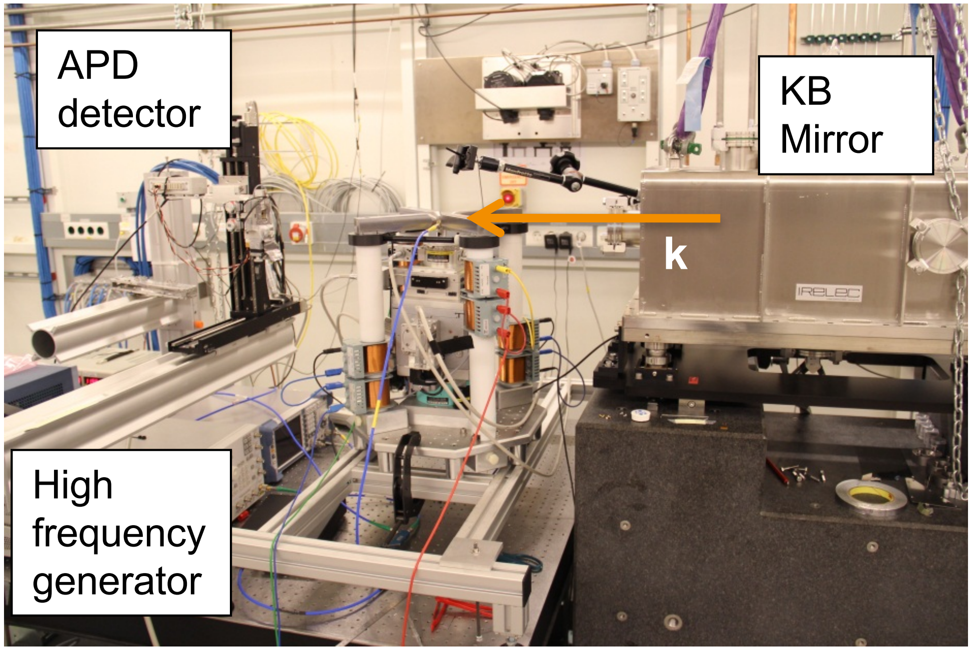
Thin film system



grazing incidence

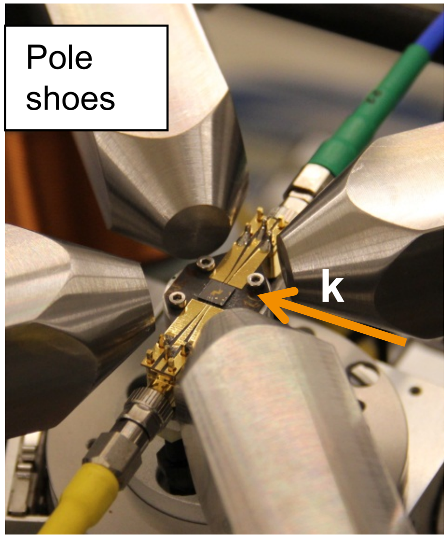


Setup at beamline P01 of PETRA III

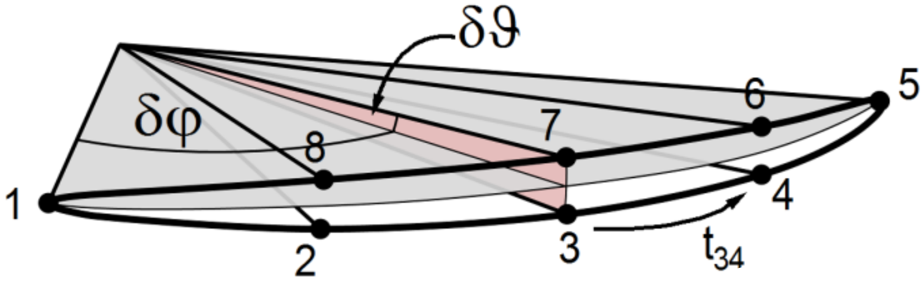


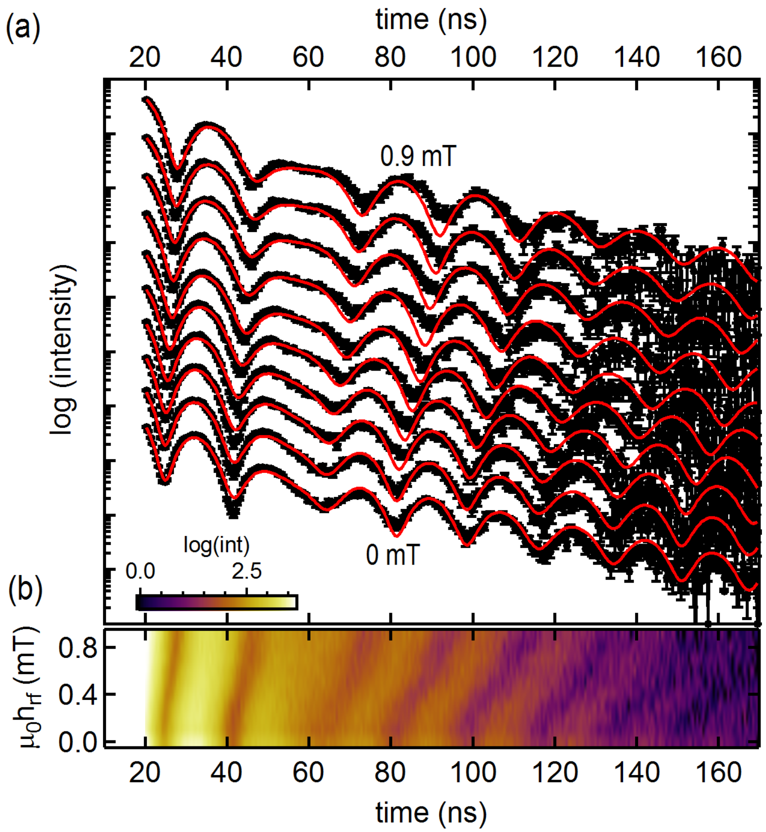
KB mirror focus 10 x 10 µm²

matches typical sample size



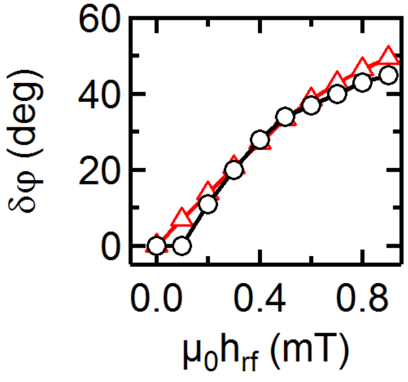
Spin trajectory determination



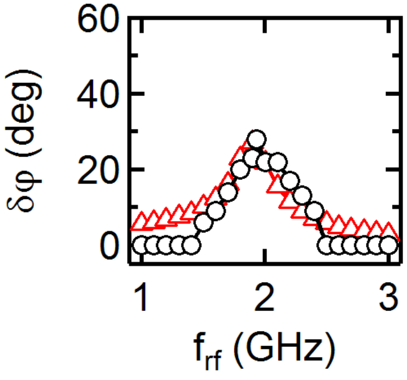


Determination of the opening angle δφ

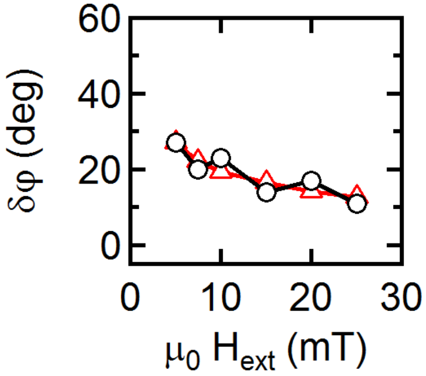
Power sweep



Frequency sweep



Field sweep



micromagnetic simulations with MicroMagnum software

L. Bocklage et al., Phys. Rev. Lett. 114, 147601 (2015)

Summary

(1) Spin structure of exchange-spring magnets

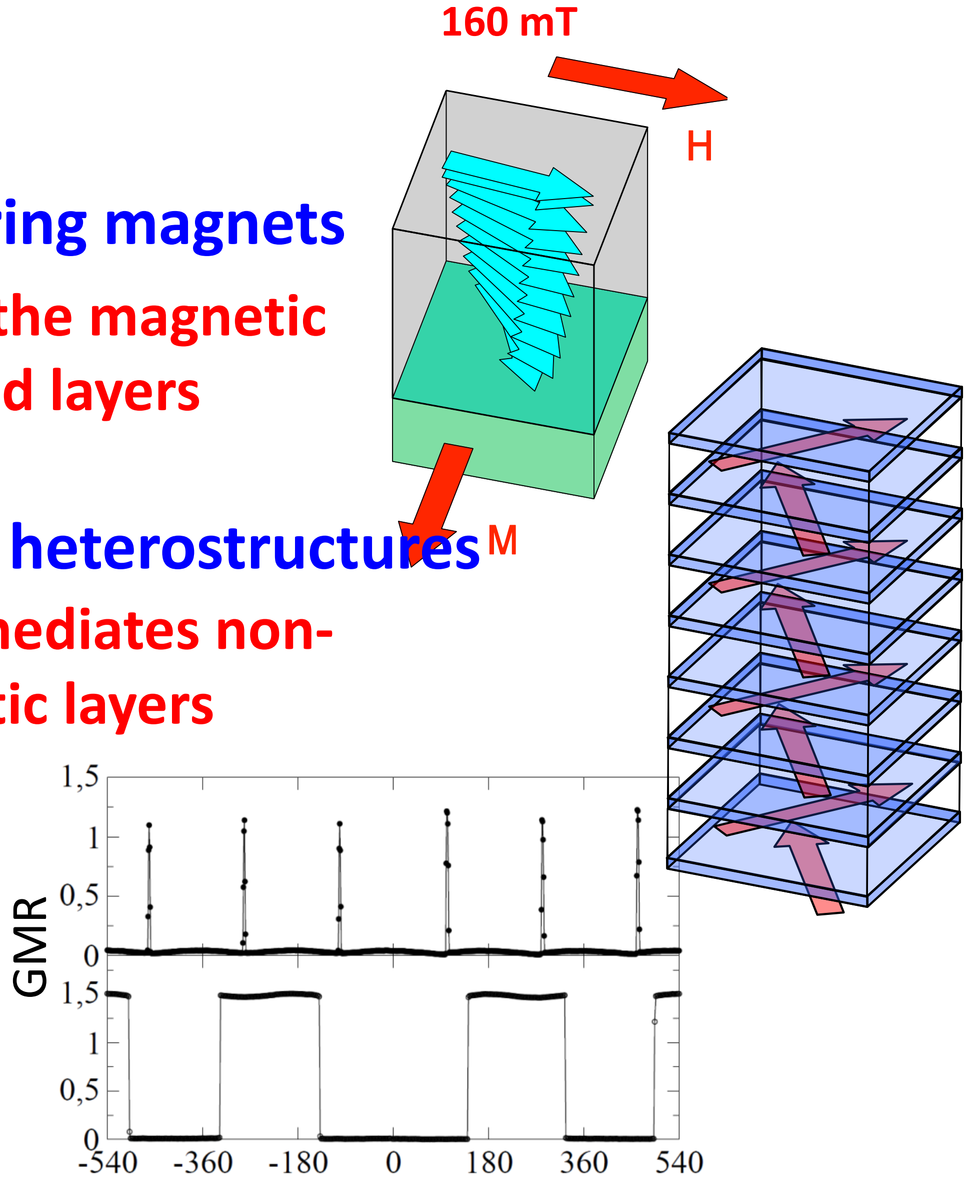
^{57}Fe isotopic probe layers reveal the magnetic depth profile of exchange-coupled layers

(2) Magnetic order in Fe/Fe-oxide heterostructures

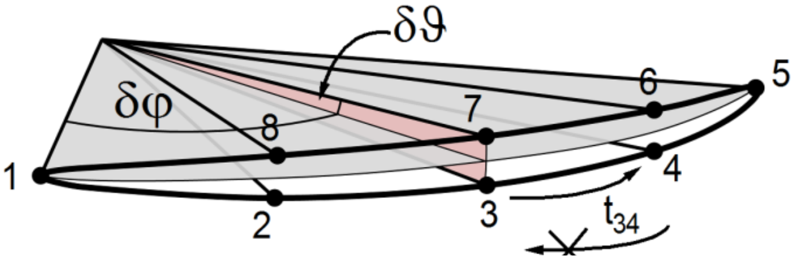
Antiferromagnetic native oxide mediates non-collinear coupling of ferromagnetic layers

(3) Spin-structured multilayers:
A new class of materials
for precision spintronics

Tuning of magnetoresistance via oblique-angle deposition



(4) Spin-trajectories in thin films under FMR conditions



The DESY research group
Magnetism and Coherent Phenomena (MCP)

May 2017

Not on picture: Sven Velten