Workshop on Nuclear Resonant Scattering and Data Analysis
November 11 – 13, Advanced Photon Source

# Studies of Thin-Film Magnetism Using Nuclear Resonant Scattering

**Ralf Röhlsberger**

Deutsches Elektronen-Synchrotron DESY, Hamburg

# Outline

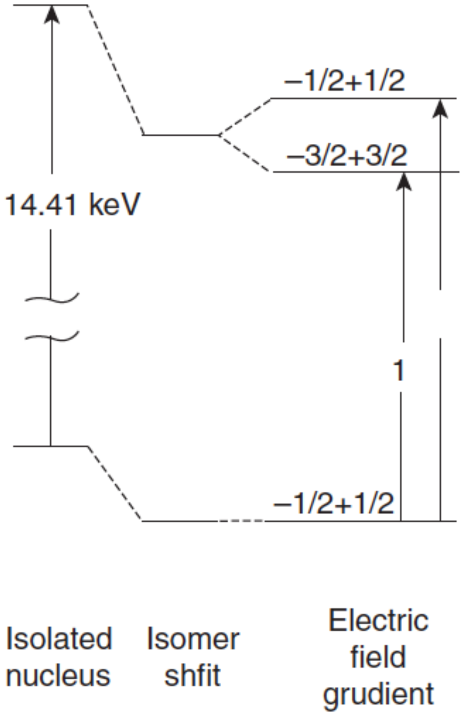
1. Hyperfine Interactions:
   Mössbauer nuclei as spectators of solid-state properties

2. Magnetic structure of thin films, multilayers and nanostripes
   a. Spin structure of exchange-spring magnets
   b. Domain-wall compression at magnetic interfaces
   c. Twisted magnetic structures in multilayers
   d. Magnetic order in nanostripe arrays

3. Magnetic dynamics in thin films and nanostructures
   a. Superparamagnetic relaxation in nanoparticles
   b. Spin precession at ferromagnetic resonance

# Hyperfine Interactions

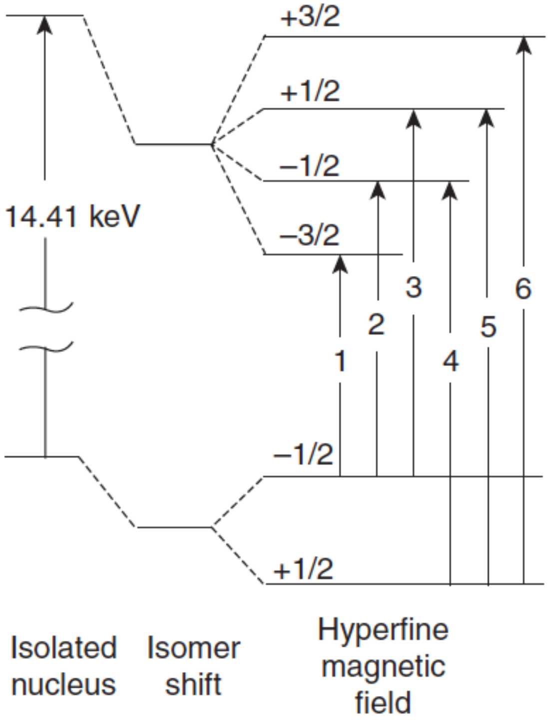
## Electric hyperfine interaction



$$\Delta E_q = \frac{\pm 1}{4} eQV_{zz}\left(1 + \frac{\eta^2}{3}\right)^{1/2}$$

$$\eta \equiv (V_{xx} - V_{yy})/V_{zz}.$$

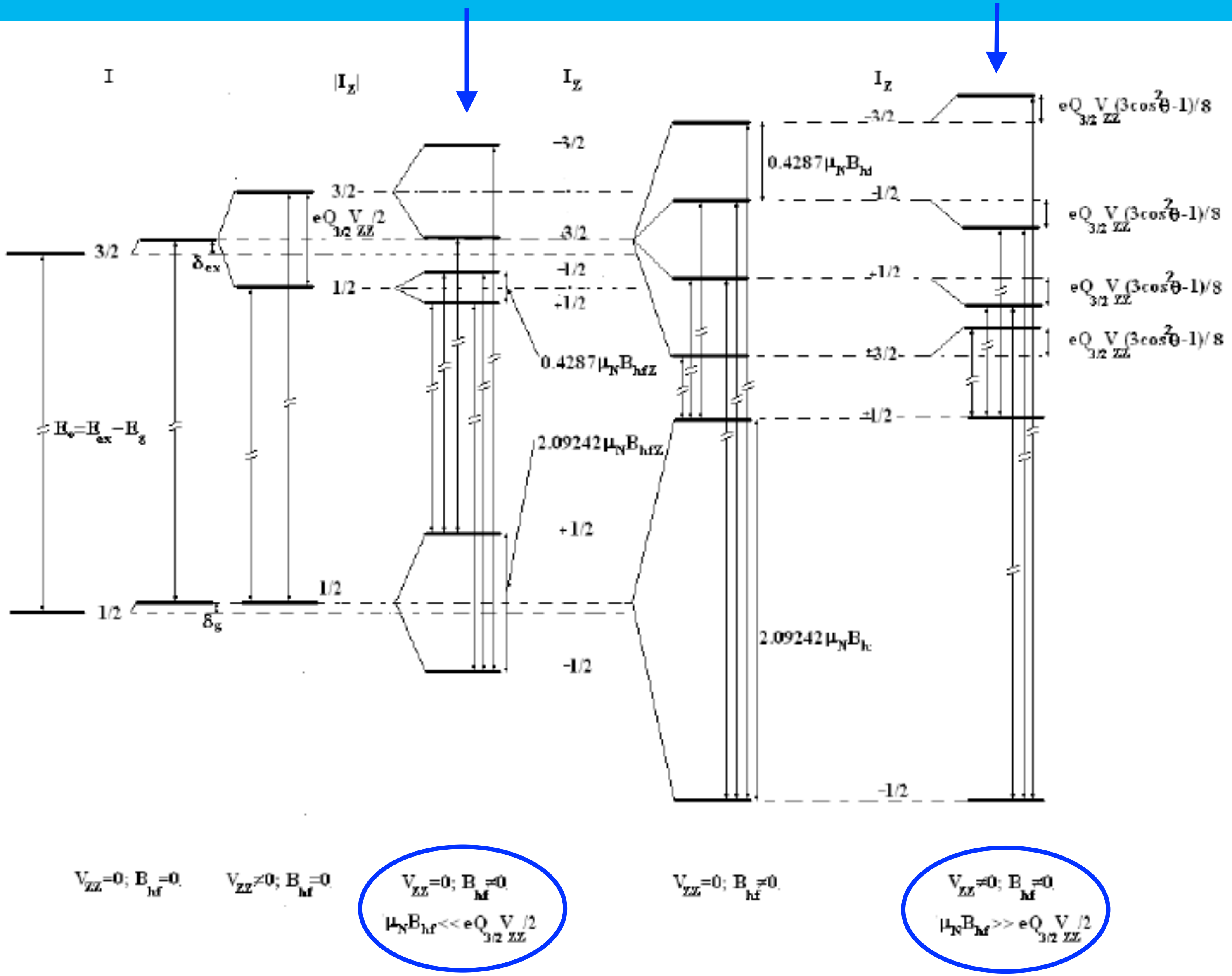
## Magnetic hyperfine interaction



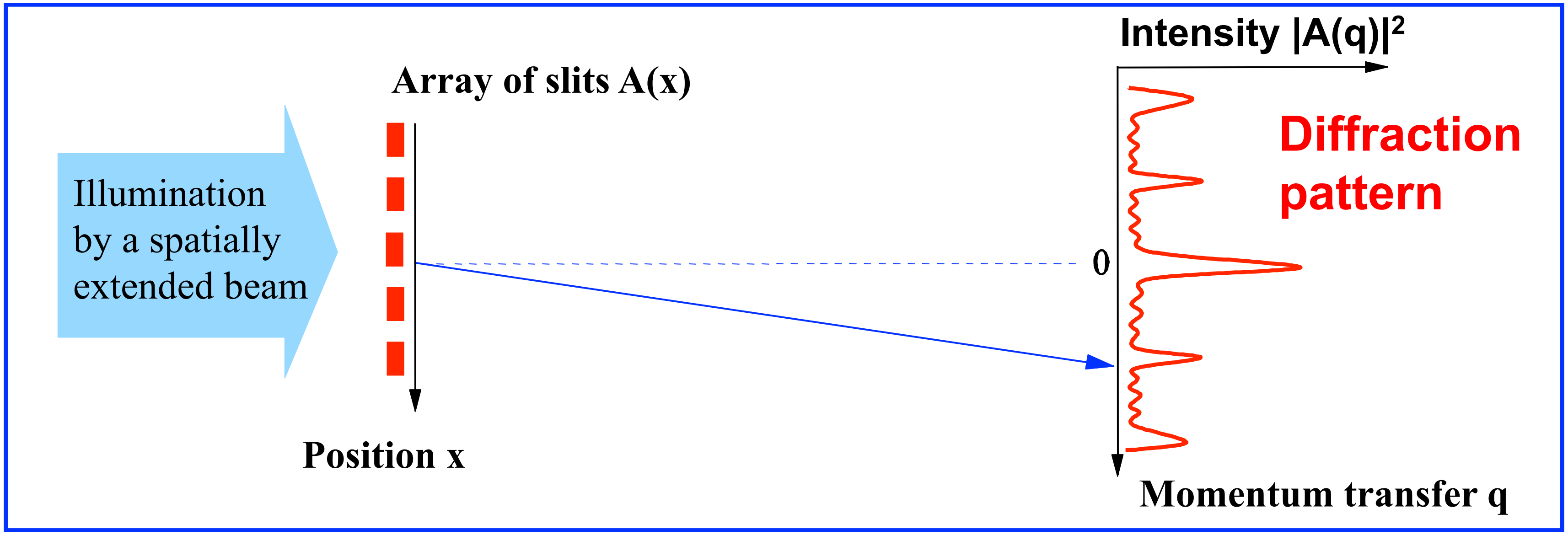
$$\Delta E_m = (m_e g_e - m_g g_g)\, B$$

From: Brent Fultz, in: ‘Characterization of Materials’, Wiley 2012

# Combined Hyperfine Interaction: Electric + Magnetic



# Diffraction as Method of Structure Determination



Illumination by a spatially extended beam

Array of slits $A(x)$

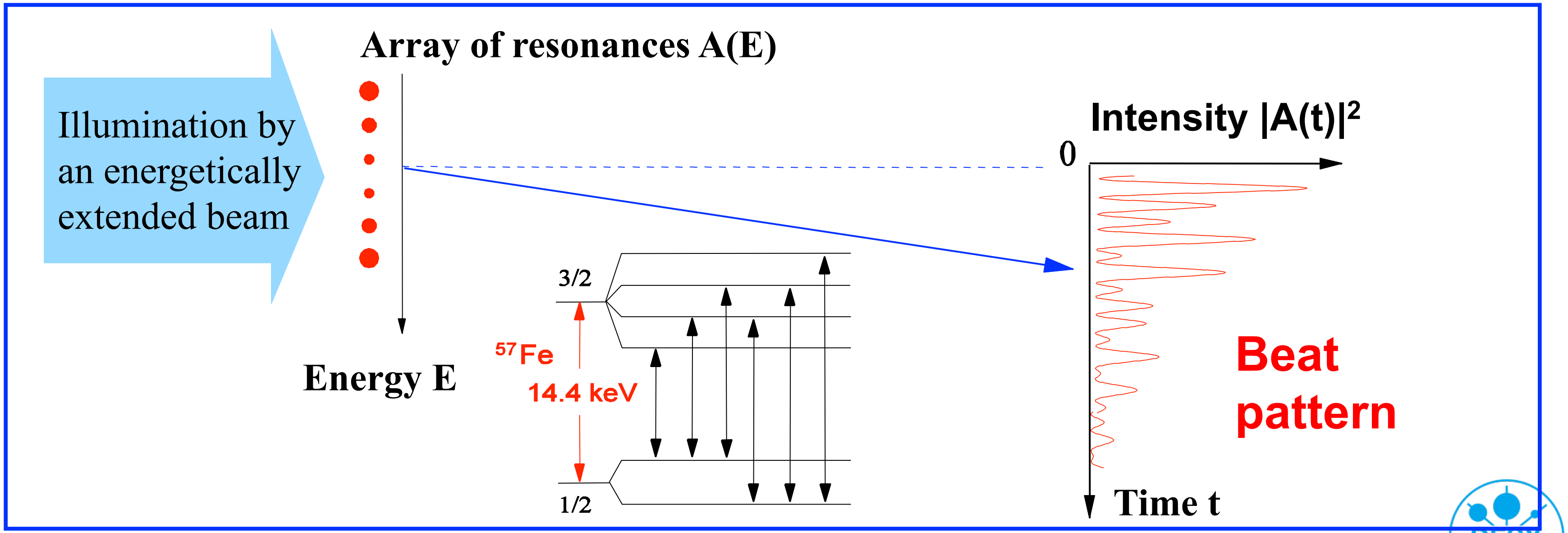
Position x

Intensity $|A(q)|^2$

0

**Diffraction pattern**

Momentum transfer q



Illumination by an energetically extended beam

Array of resonances $A(E)$

Energy E

Intensity $|A(t)|^2$

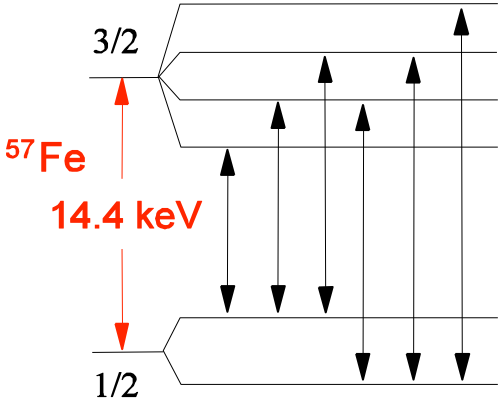
0

**Beat pattern**

Time t

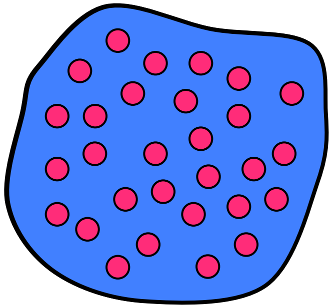
# Nuclear Resonant Forward Scattering of Synchrotron Radiation

Pulsed broadband excitation of hyperfine-split nuclear levels

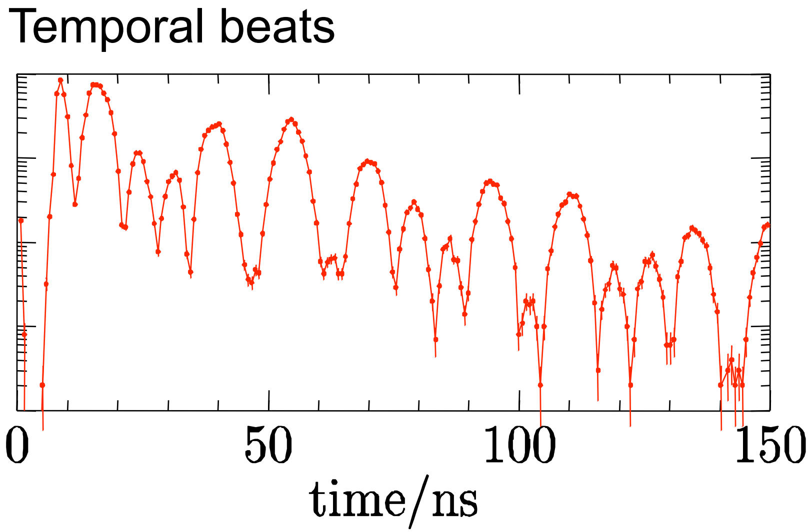


The 14.4 keV nuclear resonance of $^{57}$Fe

$\tau_0$ = 141 ns, $\Gamma_0$ = 4.7 neV

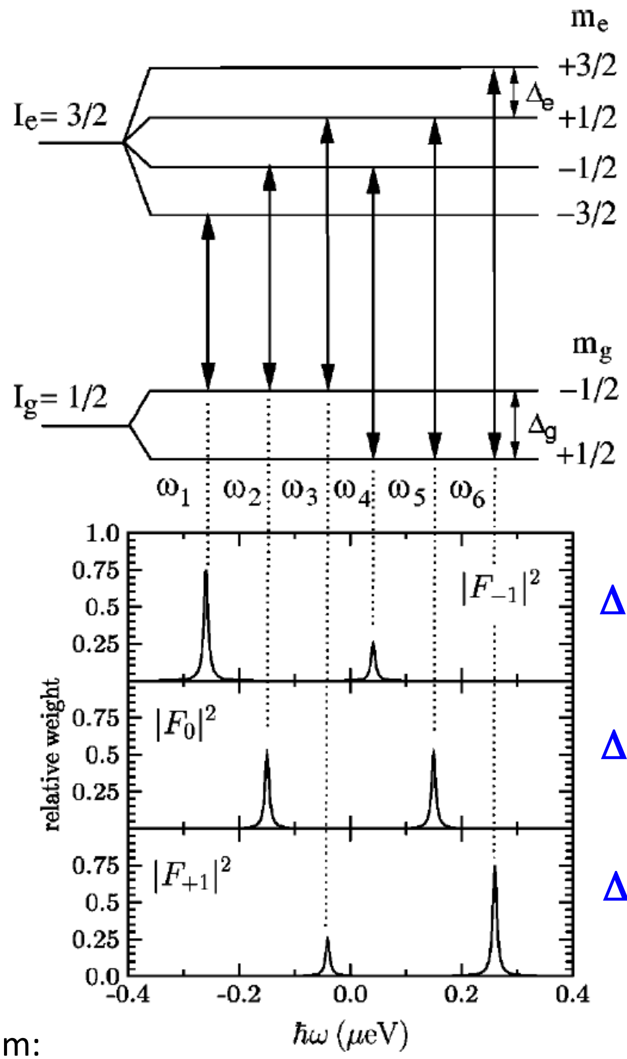


The beat pattern is a fingerprint of the magnetic structure of the sample:


Temporal beats

# Magnetic Hyperfine Interaction

## Energy dependence



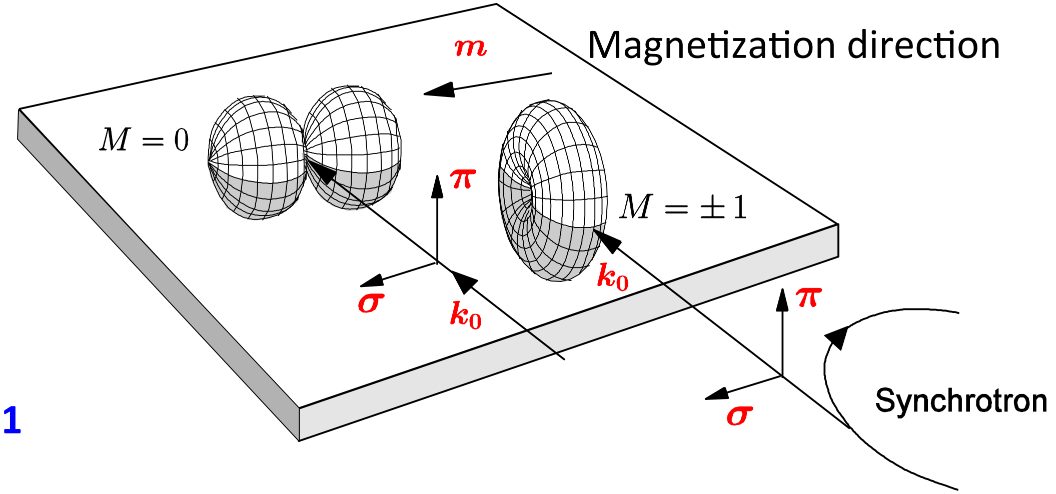
**$\Delta$m = -1**

**$\Delta$m = 0**

**$\Delta$m = +1**

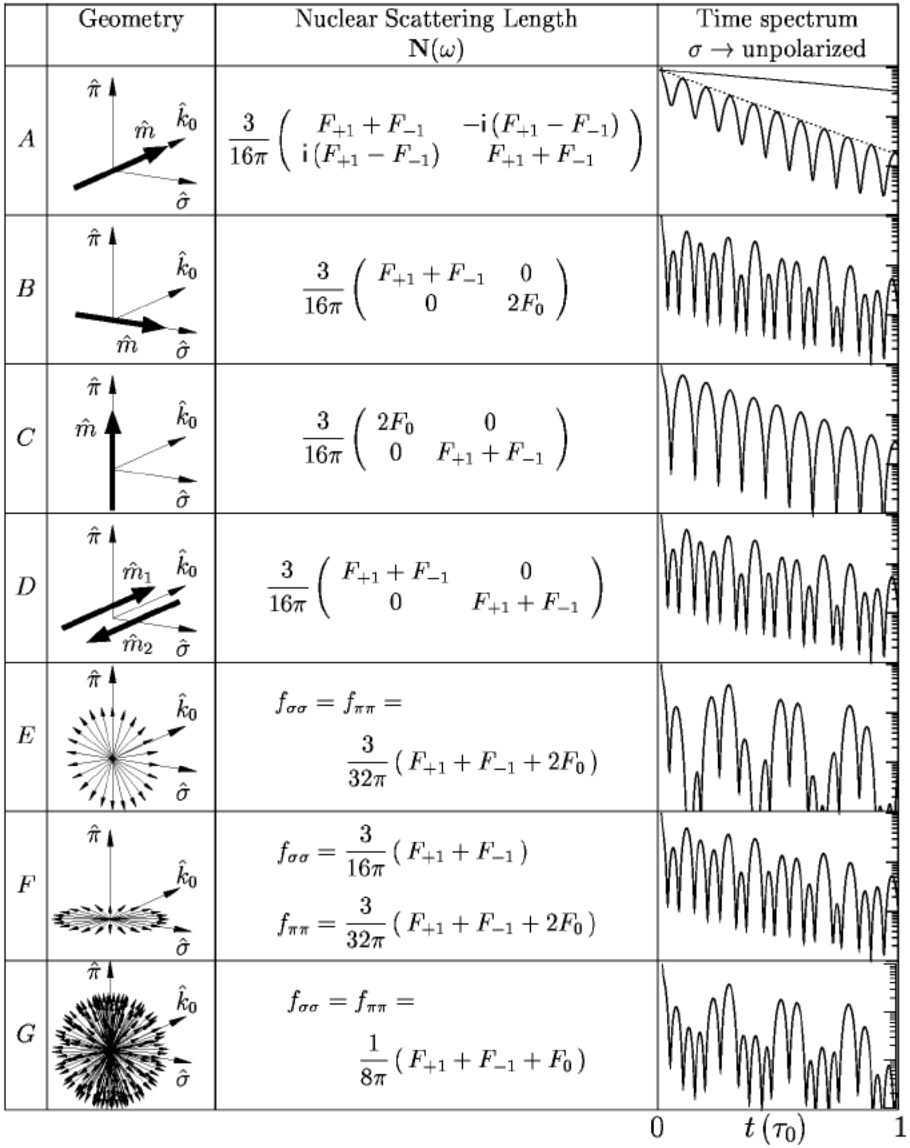
From:
Phys. Rev. B 67, 245412 (2003)

## Directional dependence
## → Dipole emission characteristics



# Magnetic Hyperfine Interaction

## Directional dependence

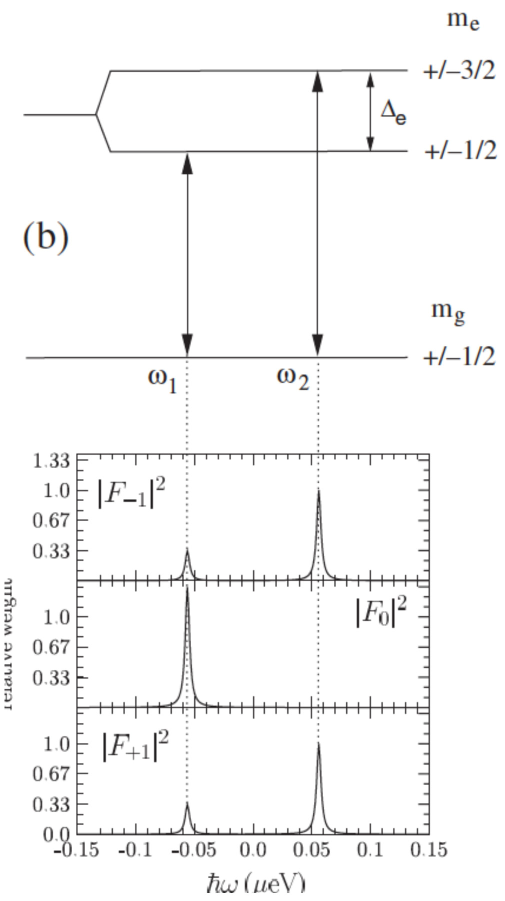


| | Geometry | Nuclear Scattering Length $\mathbf{N}(\omega)$ | Time spectrum $\sigma \rightarrow$ unpolarized |
|---|---|---|---|
| $A$ | $\hat{\pi}$ $\hat{m}$ $\hat{k}_0$ $\hat{\sigma}$ | $\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & -\mathrm{i}\,(F_{+1}-F_{-1}) \\ \mathrm{i}\,(F_{+1}-F_{-1}) & F_{+1}+F_{-1} \end{pmatrix}$ | |
| $B$ | $\hat{\pi}$ $\hat{k}_0$ $\hat{m}$ $\hat{\sigma}$ | $\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & 0 \\ 0 & 2F_0 \end{pmatrix}$ | |
| $C$ | $\hat{\pi}$ $\hat{m}$ $\hat{k}_0$ $\hat{\sigma}$ | $\frac{3}{16\pi}\begin{pmatrix} 2F_0 & 0 \\ 0 & F_{+1}+F_{-1} \end{pmatrix}$ | |
| $D$ | $\hat{\pi}$ $\hat{m}_1$ $\hat{k}_0$ $\hat{m}_2$ $\hat{\sigma}$ | $\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & 0 \\ 0 & F_{+1}+F_{-1} \end{pmatrix}$ | |
| $E$ | $\hat{\pi}$ $\hat{k}_0$ $\hat{\sigma}$ | $f_{\sigma\sigma} = f_{\pi\pi} = \frac{3}{32\pi}\,(F_{+1}+F_{-1}+2F_0)$ | |
| $F$ | $\hat{\pi}$ $\hat{k}_0$ $\hat{\sigma}$ | $f_{\sigma\sigma} = \frac{3}{16\pi}\,(F_{+1}+F_{-1})$<br>$f_{\pi\pi} = \frac{3}{32\pi}\,(F_{+1}+F_{-1}+2F_0)$ | |
| $G$ | $\hat{\pi}$ $\hat{k}_0$ $\hat{\sigma}$ | $f_{\sigma\sigma} = f_{\pi\pi} = \frac{1}{8\pi}\,(F_{+1}+F_{-1}+F_0)$ | |

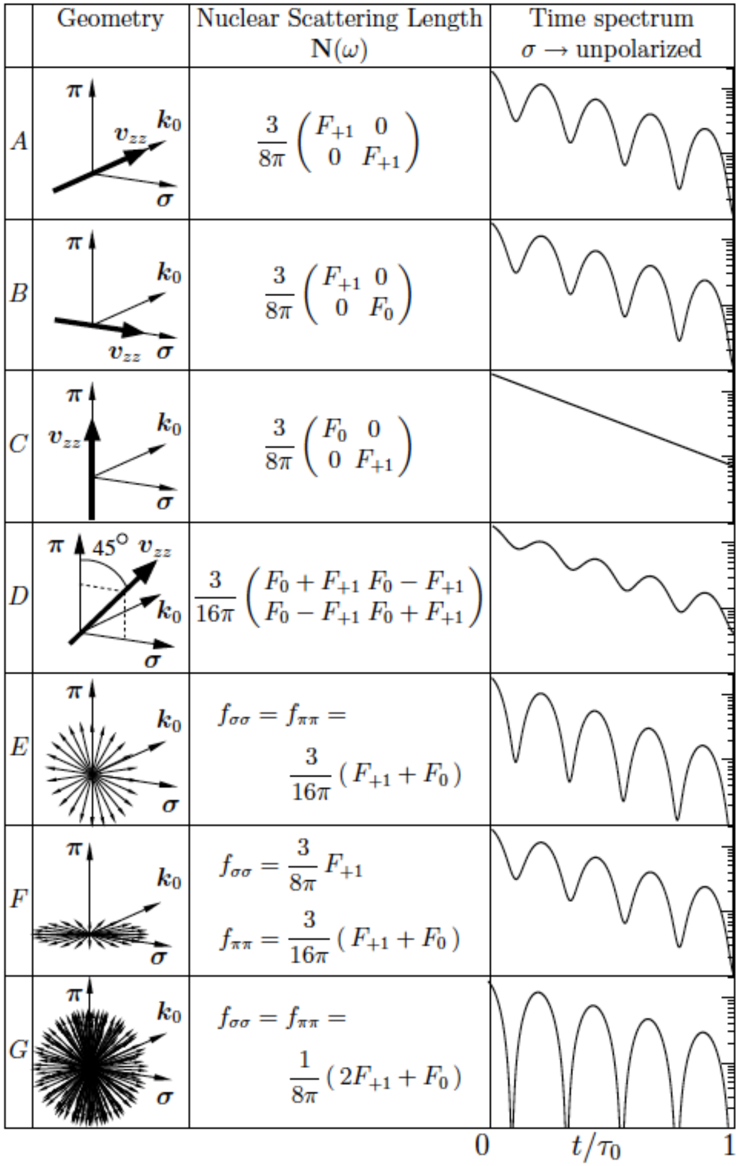
From:
Phys. Rev. B 67, 245412 (2003)

# Electric Hyperfine Interaction

## Directional dependence



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



| | Geometry | Nuclear Scattering Length N($\omega$) | Time spectrum $\sigma \rightarrow$ unpolarized |
|---|---|---|---|
| A | | $\frac{3}{8\pi}\begin{pmatrix}F_{+1} & 0\\ 0 & F_{+1}\end{pmatrix}$ | |
| B | | $\frac{3}{8\pi}\begin{pmatrix}F_{+1} & 0\\ 0 & F_0\end{pmatrix}$ | |
| C | | $\frac{3}{8\pi}\begin{pmatrix}F_0 & 0\\ 0 & F_{+1}\end{pmatrix}$ | |
| D | | $\frac{3}{16\pi}\begin{pmatrix}F_0+F_{+1} & F_0-F_{+1}\\ F_0-F_{+1} & F_0+F_{+1}\end{pmatrix}$ | |
| E | | $f_{\sigma\sigma} = f_{\pi\pi} = \frac{3}{16\pi}(F_{+1}+F_0)$ | |
| F | | $f_{\sigma\sigma} = \frac{3}{8\pi}F_{+1}$<br>$f_{\pi\pi} = \frac{3}{16\pi}(F_{+1}+F_0)$ | |
| G | | $f_{\sigma\sigma} = f_{\pi\pi} = \frac{1}{8\pi}(2F_{+1}+F_0)$ | |

# Temporal beats and magnetization direction

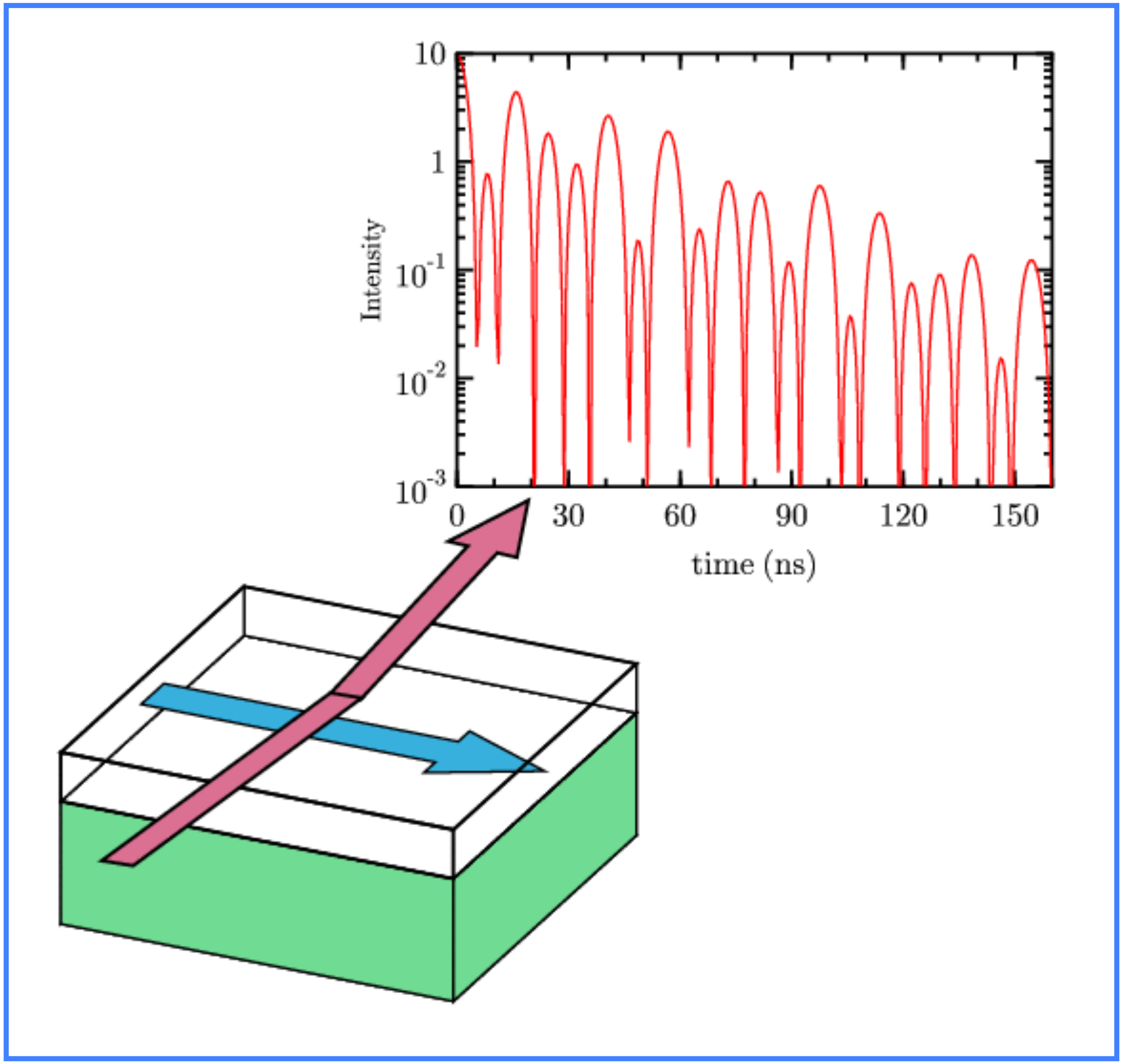
**The temporal beat pattern sensitively depends on the orientation of the**

**Magnetization M**

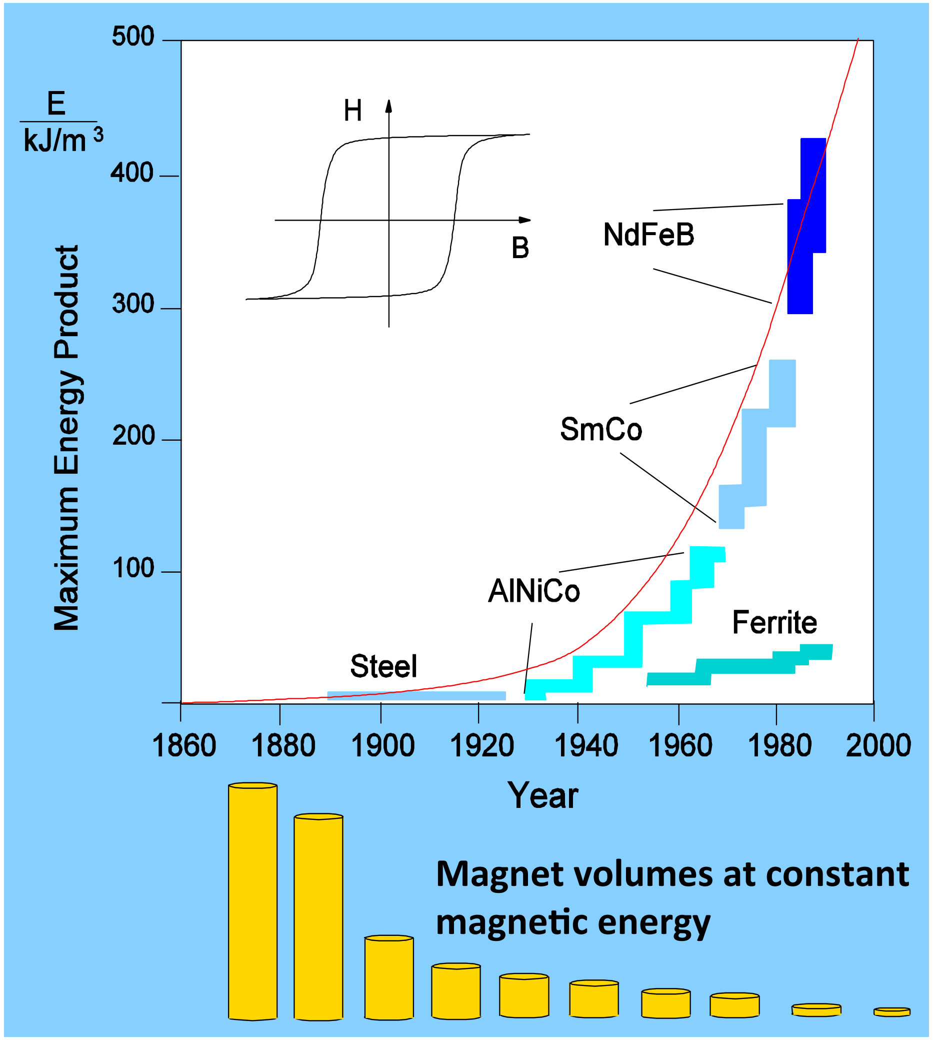
**relative to the**

**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

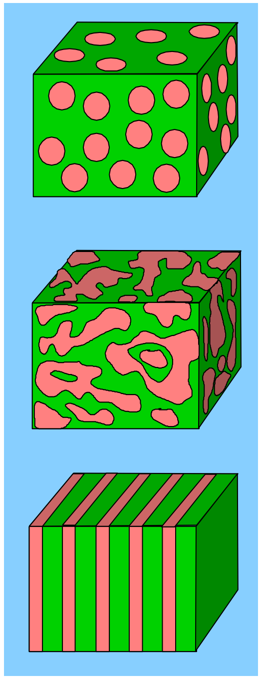


**Magnet volumes at constant magnetic energy**

**Exchange hardening in nanostructured two-phase systems:**

**Hard phase with high coercivity**

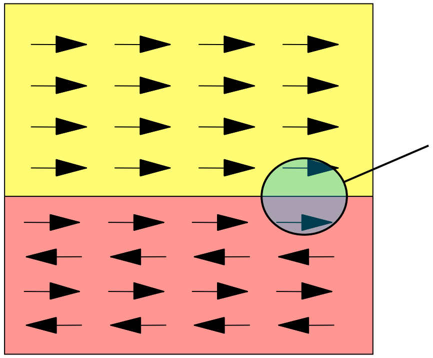
**Soft phase with high magnetization**



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

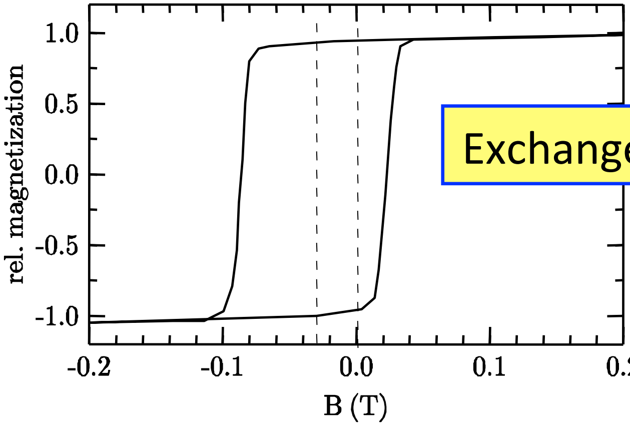
# Exchange-Coupled Bilayers

## Ferromagnet/Antiferromagnet



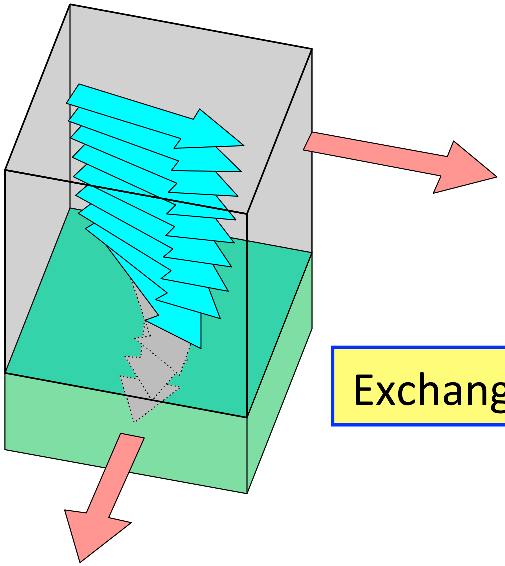
What happens at the interface ?

### Magnetic hysteresis



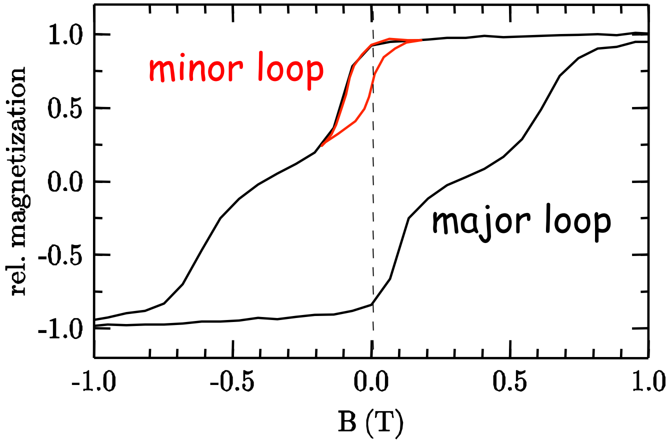
Exchange bias

## Hard magnet/Soft magnet



Exchange spring

### Magnetic hysteresis



# The Spin Structure of Magnetic Bilayers

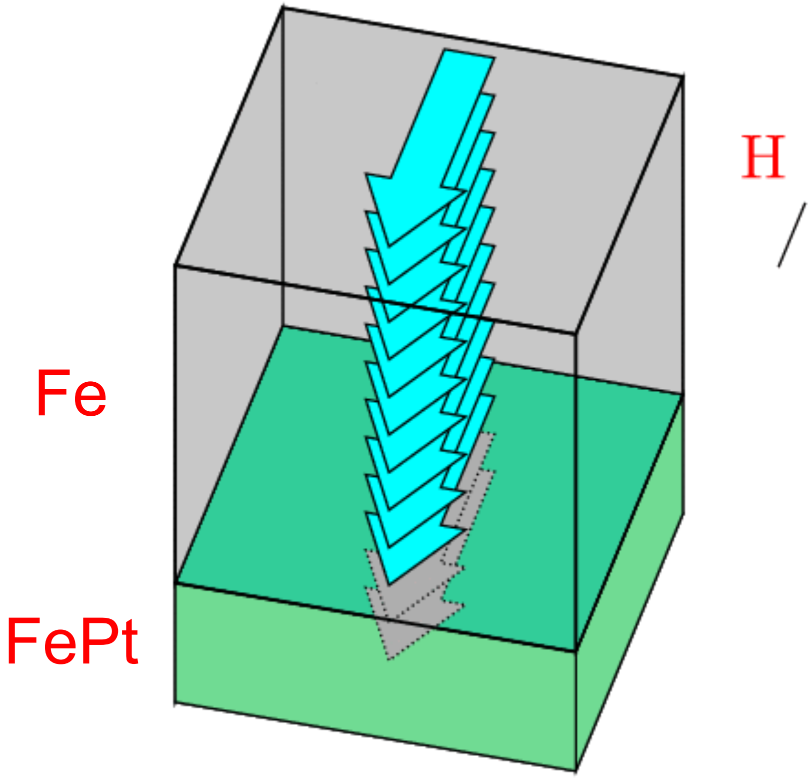
**Fe on FePt**

Soft – magnetic Fe

Hard – magnetic FePt
with uniaxial anisotropy

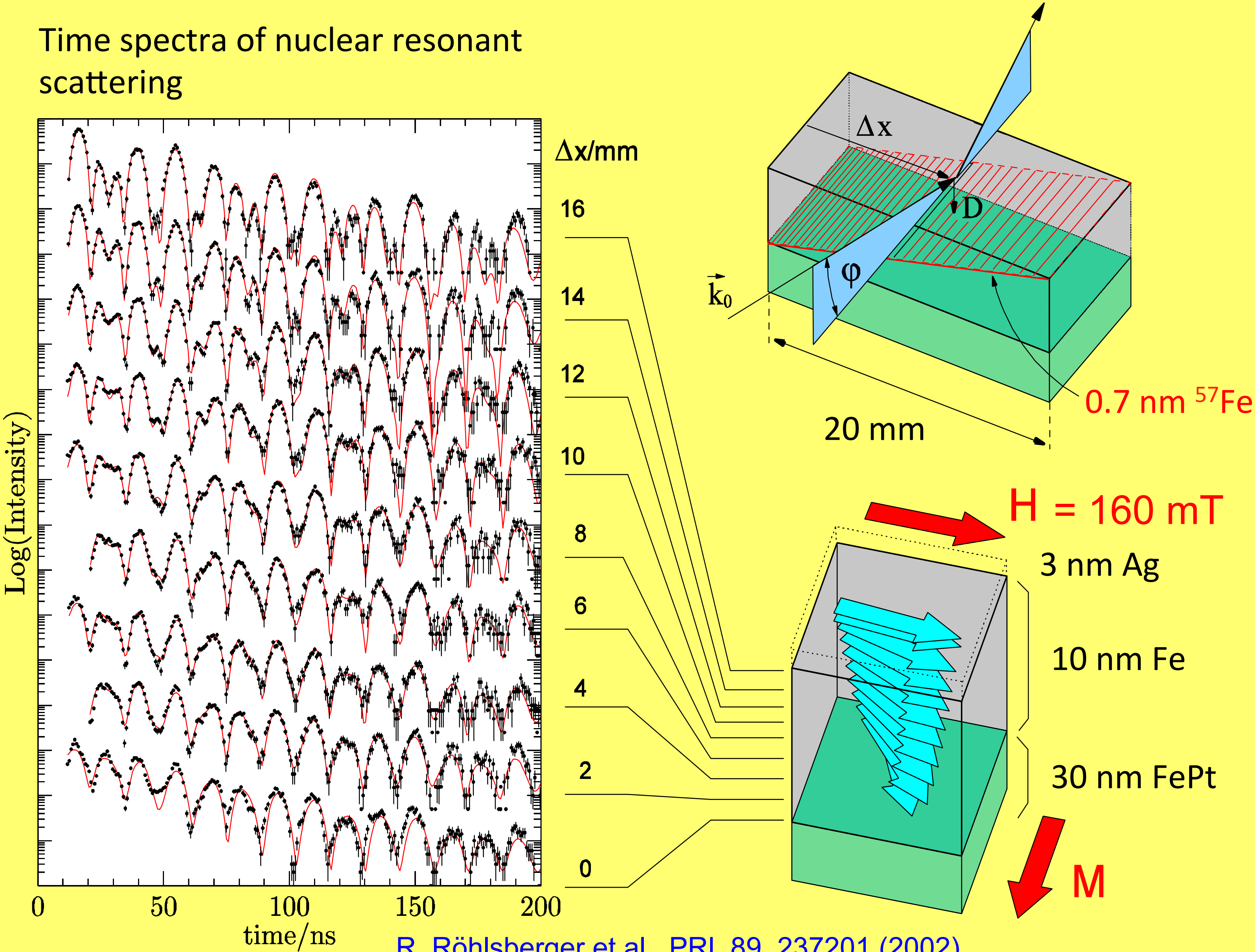
- Exchange coupling at the interface: Parallel alignment of Fe and FePt moments
- With increasing distance from the interface: Coupling becomes weaker
- External field H induces spiral magnetization
- Return to parallel alignment for H = 0

**Exchange-Spring magnets**



# Imaging the Internal Spin Structure of Exchange-Spring Magnets

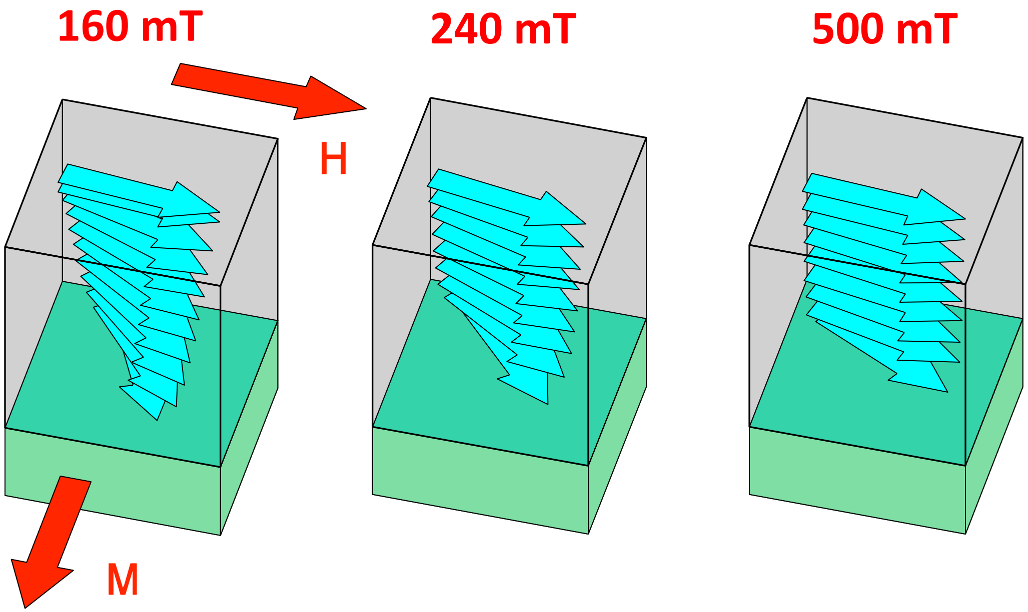
Time spectra of nuclear resonant scattering

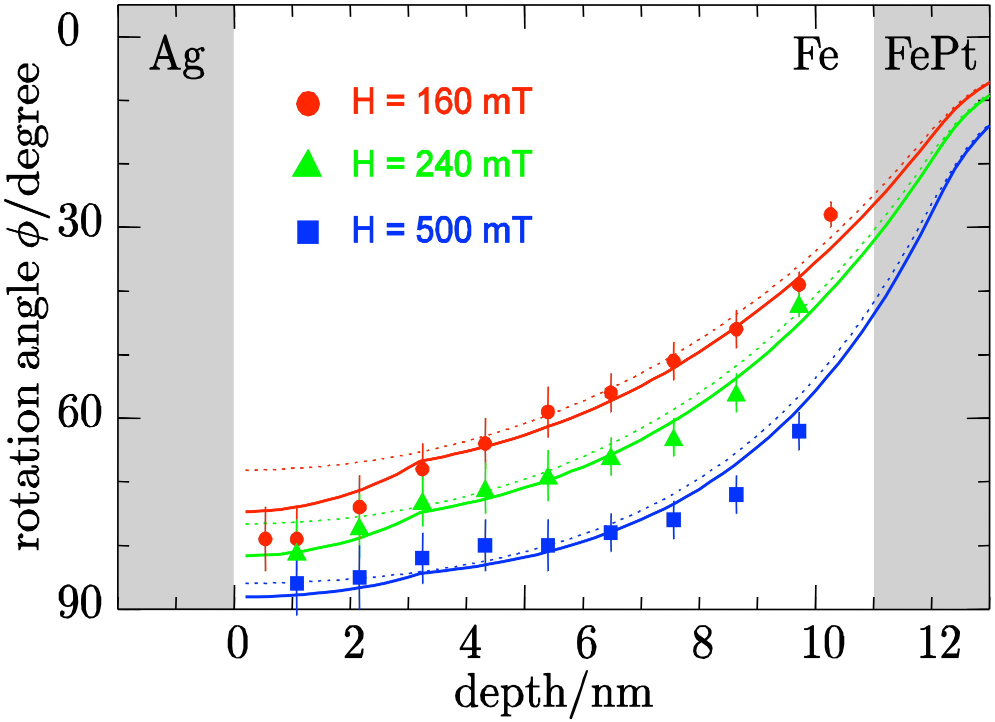


R. Röhlsberger et al., PRL 89, 237201 (2002)

**Domain wall compression with increasing external field**

**Fe on FePt**





# Simulation of Exchange-Spring Layer Systems

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

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

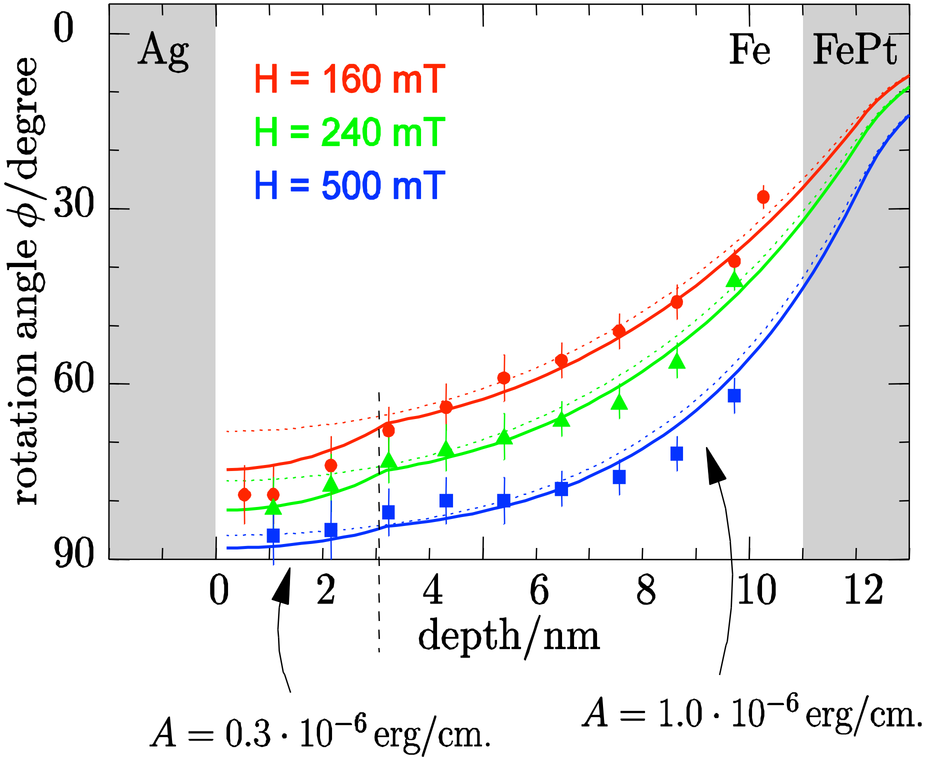
**Exchange**

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

**Anisotropy**

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

**Dipolar energy**

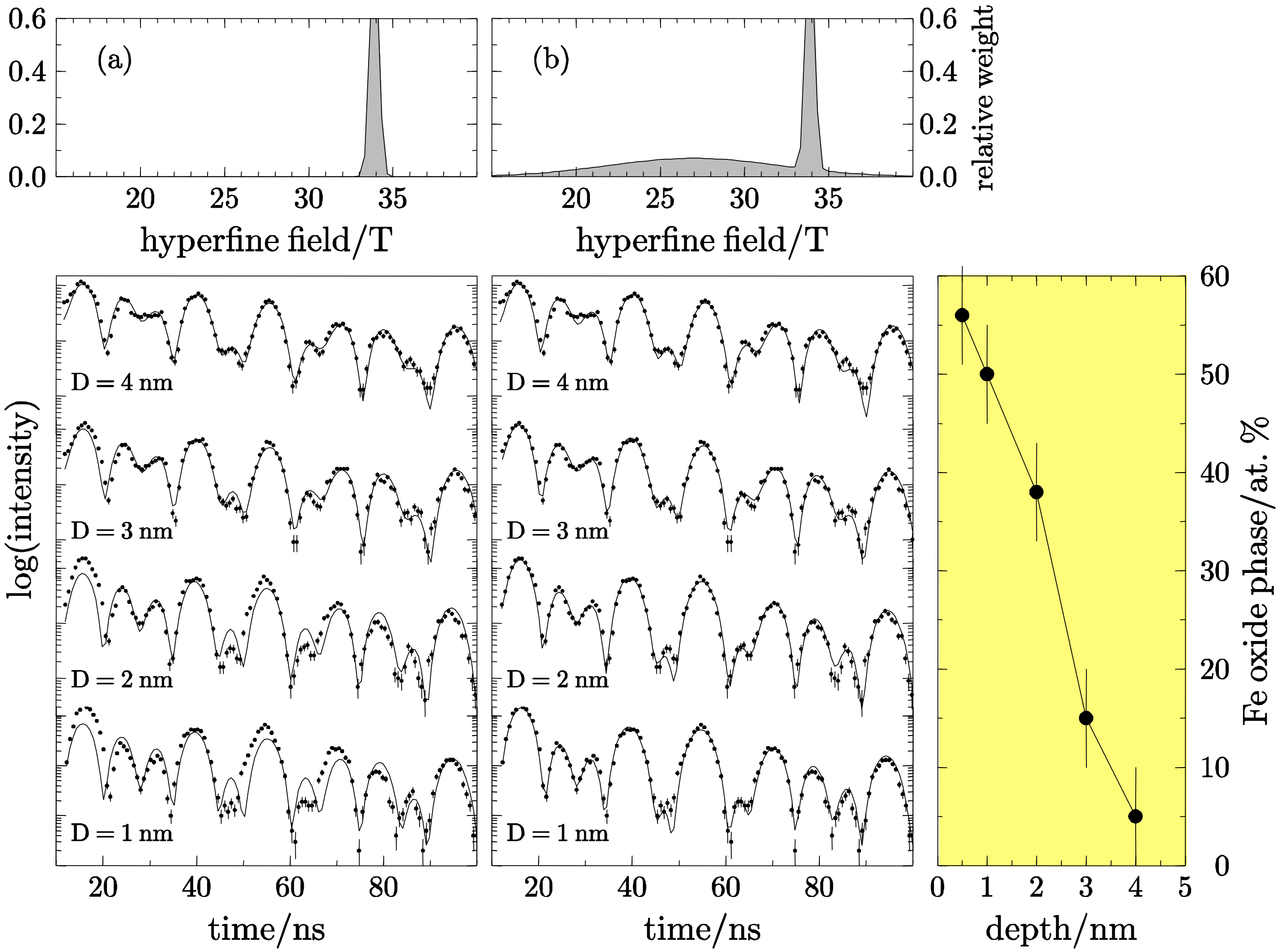


**Divide the layer system into N sublayers of thickness d**

**Minimize the magnetic free energy** $\frac{\partial E}{\partial \varphi_i} = 0$

**Depth profiling of magnetic properties**

# Depth Dependence of the Oxide Phase

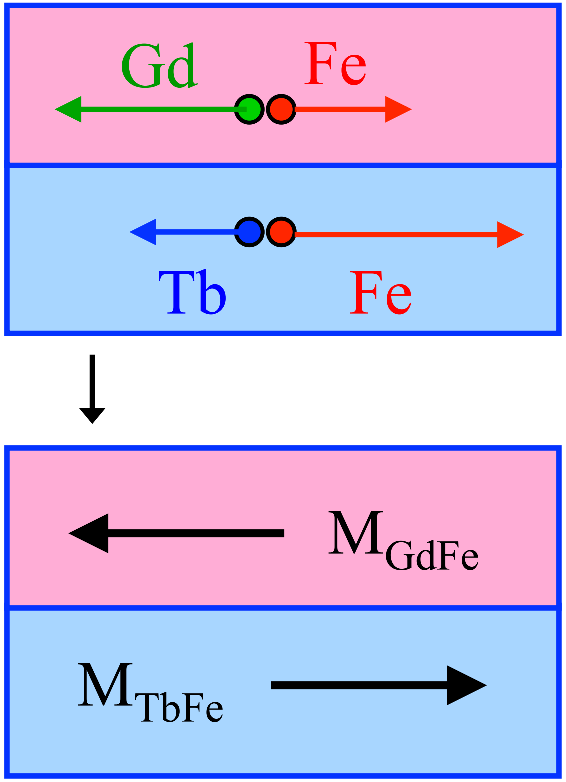


RR, H. Thomas, K. Schlage, T. Klein, J. Magn. Magn. Mater. 282, 329 (2004)

# Exchange-Coupling of TbFe/GdFe Bilayers

TbFe, GdFe → Amorphous alloys, produced by co-evaporation of the constituents at T = 77 K in an external field

Exchange constants: $|J_{FeFe}| > |J_{FeGd}| > |J_{FeTb}| > |J_{GdTb}|$



**$Gd_{40}Fe_{60}$** Soft ferrimagnetic alloys

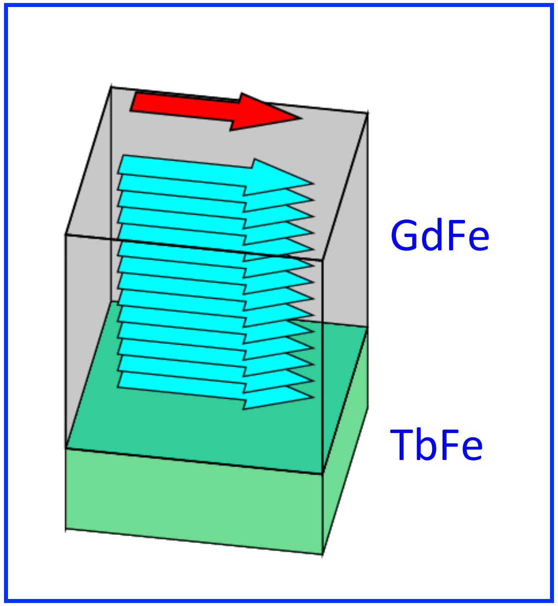
**$Tb_{12}Fe_{88}$** Soft-magnetic at room temperature, **hard-magnetic at low temperature**

Adjusting the type of coupling via composition of the alloys

In collaboration with

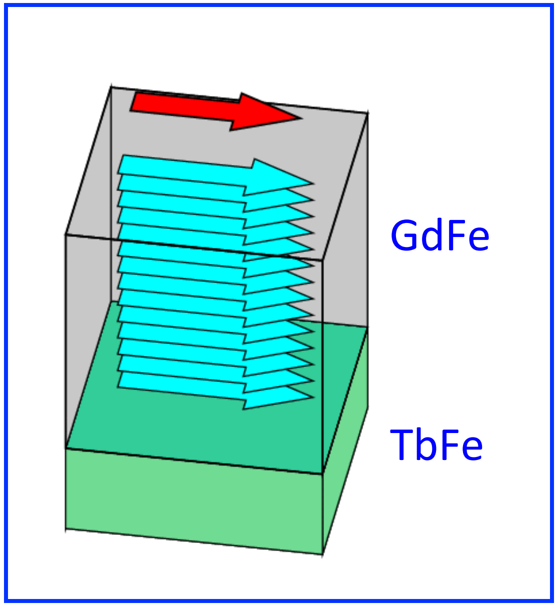
S. Mangin, F. Montaigne, T. Hauet (Nancy)
J. Juraszek, J. Teillet (Rouen)

# Domain Wall Compression in FeGd/FeTb



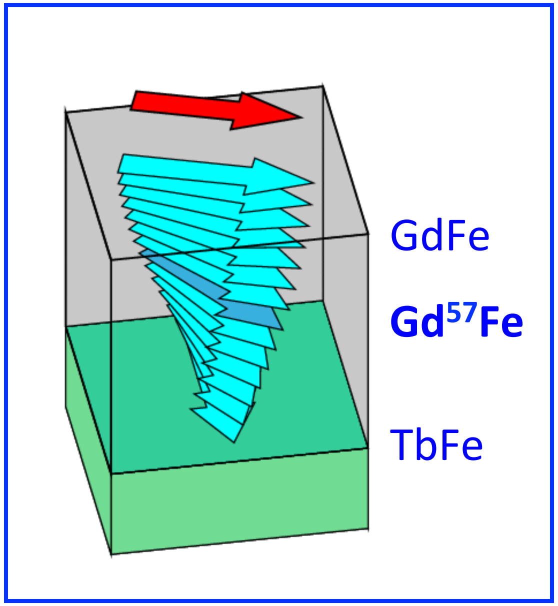
Cooling the sample in an external field below $T_B$ freezes the spin configuration in the TbFe layer

# Domain Wall Compression in FeGd/FeTb



Sample rotation in the field induces a twisted magnetic state (planar domain wall)

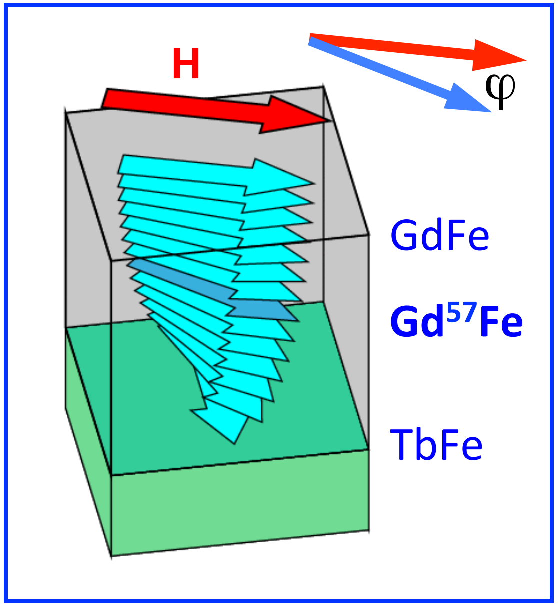
# Domain Wall Compression in FeGd/FeTb

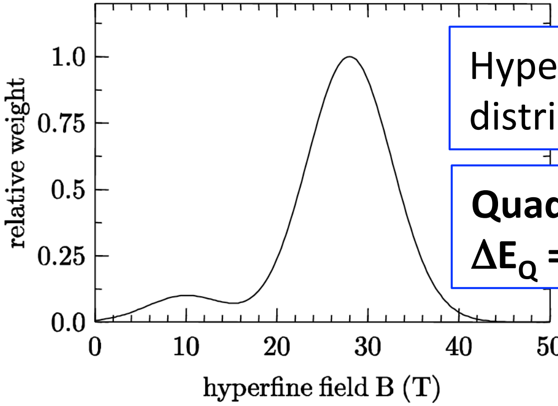


Increase of the external field leads to domain wall compression

Insert Gd$^{57}$Fe probe layer to observe the compression **(dark blue arrow)**

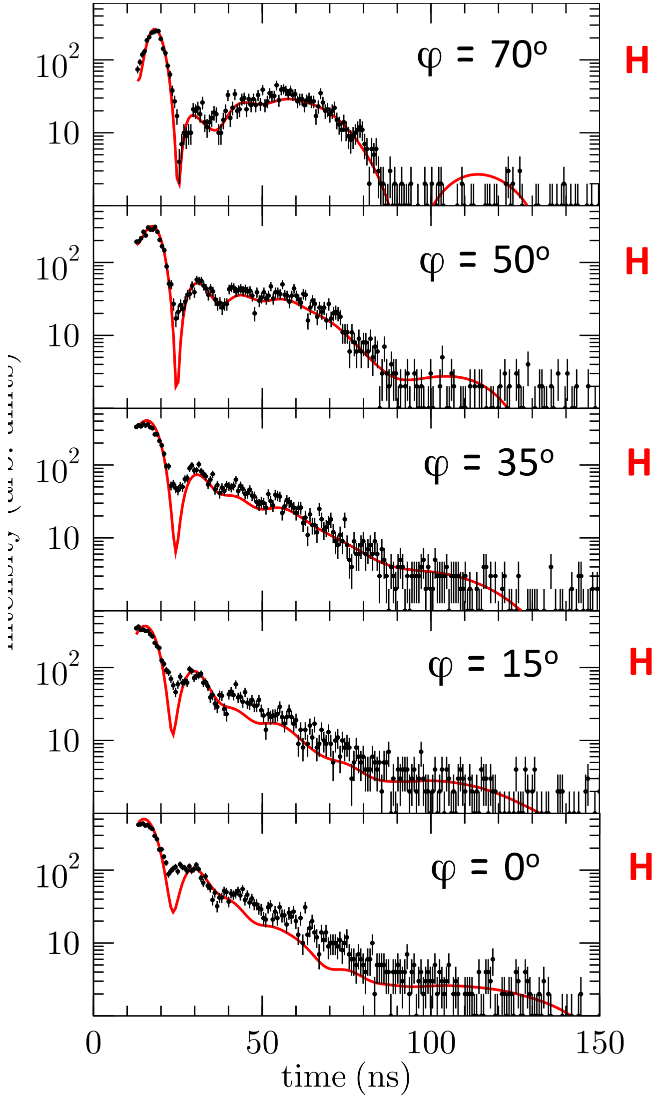
# Domain Wall Compression in FeGd/FeTb





Hyperfine field distribution

**Quad. splitting $\Delta E_Q$ = 0.5 mm/s**



**H = 30 mT**

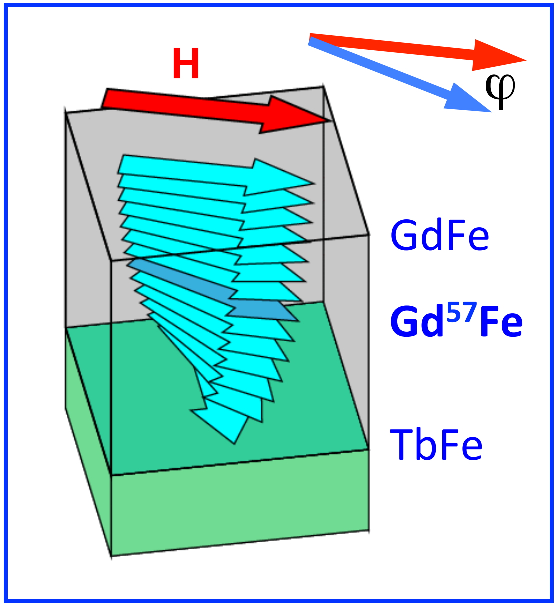
**H = 40 mT**

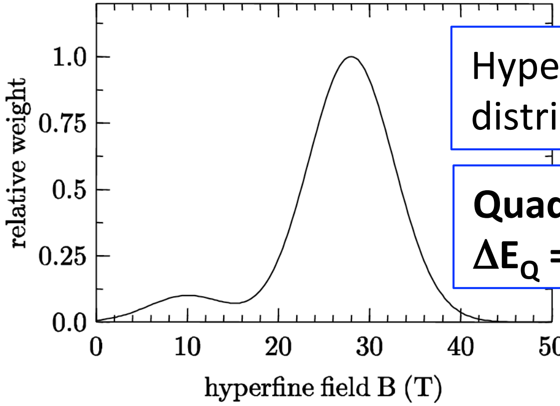
**H = 50 mT**

**H = 60 mT**

**H = 100 mT**

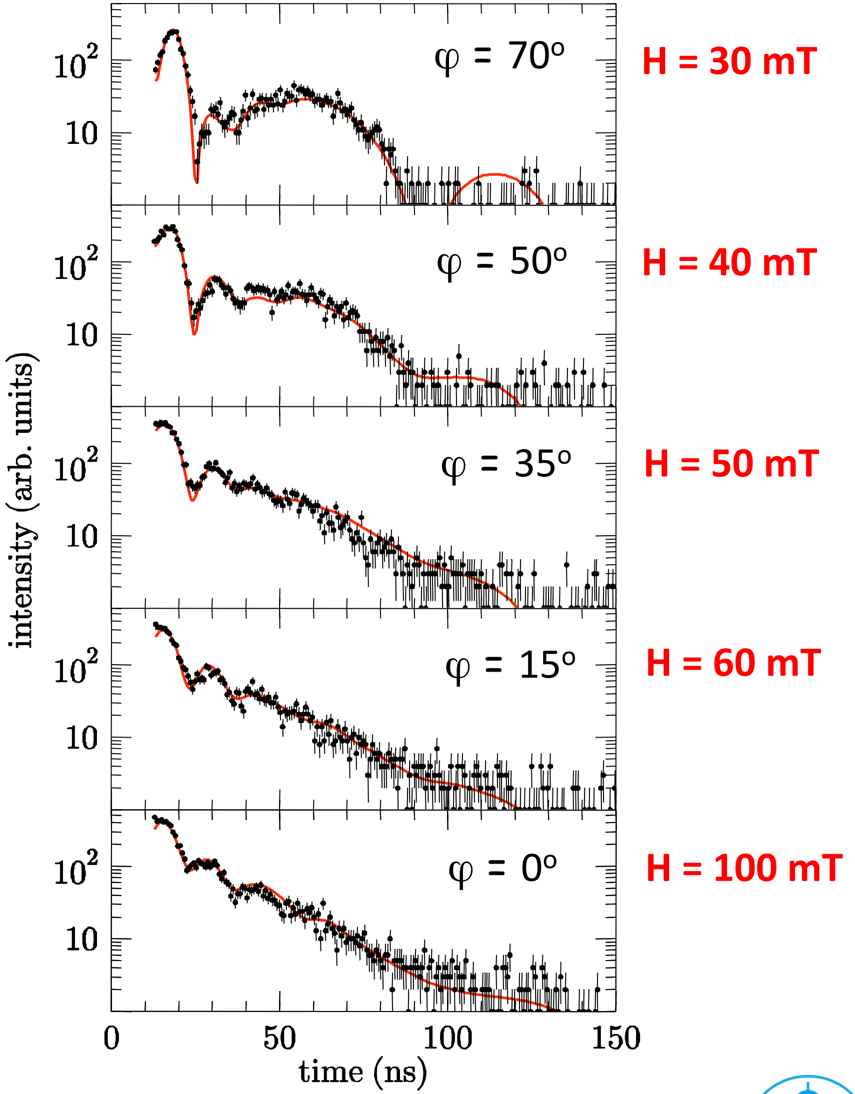
# Domain Wall Compression in FeGd/FeTb





Hyperfine field distribution

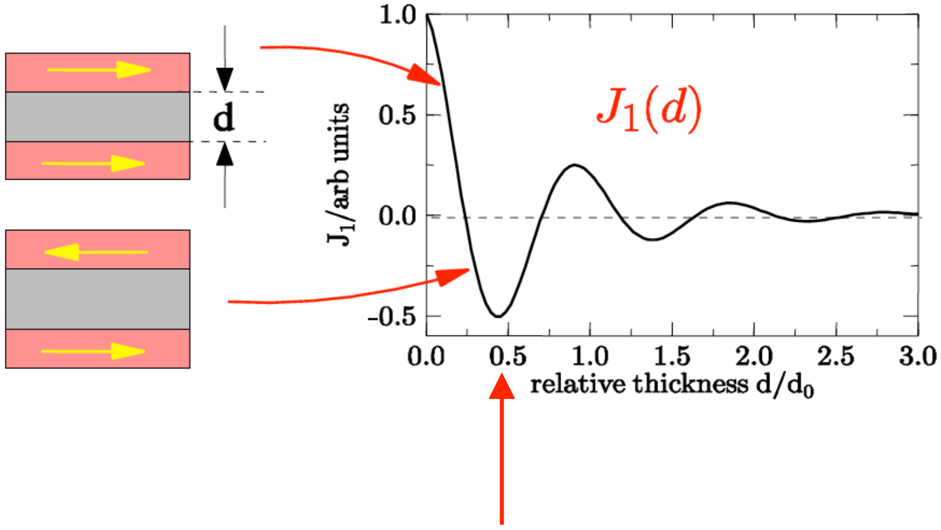
**Quad. splitting $\Delta E_Q$ = - 0.5 mm/s**



Spin structure determination is feasible in the presence of a broad field distribution

# Magnetic Order in Fe/Cr multilayers

**RKKY- oscillatory interlayer coupling**

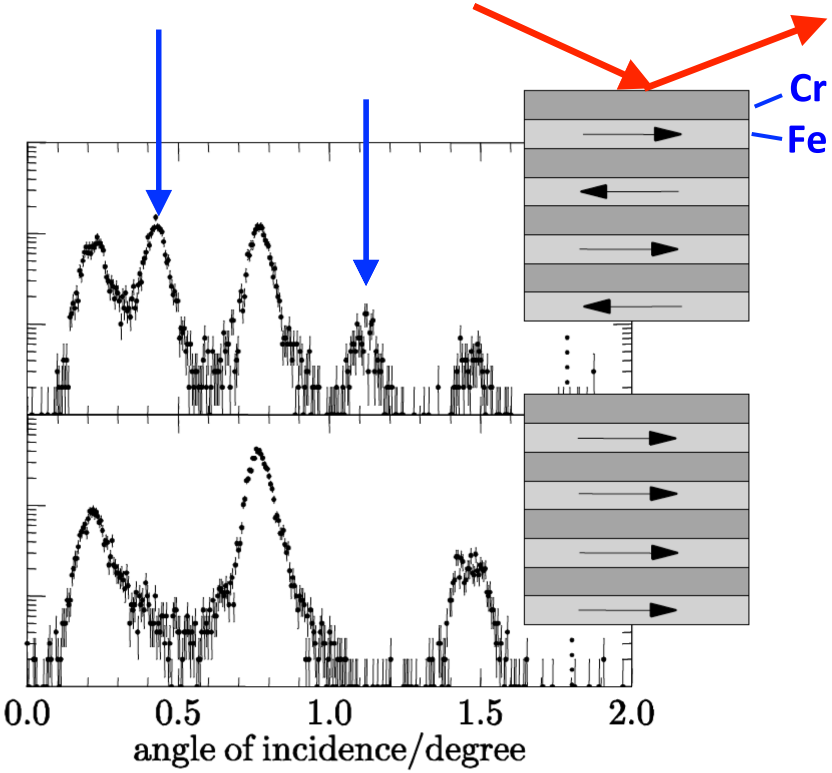


Cr : $d_{AFM}$ = 1.4 nm

**"There is no home like Iron-Chrome"**

**detected via nuclear resonant scattering of synchrotron radiation :**

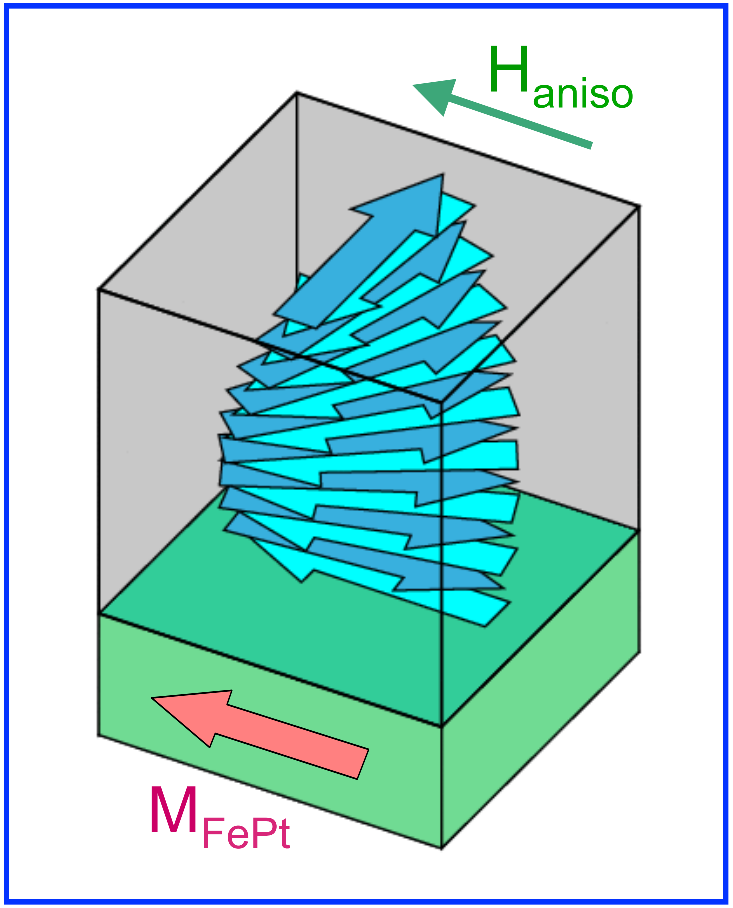
**Half-order Bragg peaks due to magnetic superstructure**



T. Toellner et al., PRL 74, 3475 (1995)

# Twisted States in Exchange-Coupled Antiferromagnetic Multilayers

**Fe/Cr multilayer exchange coupled to hard-magnetic FePt with unidirectional magnetic anisotropy**

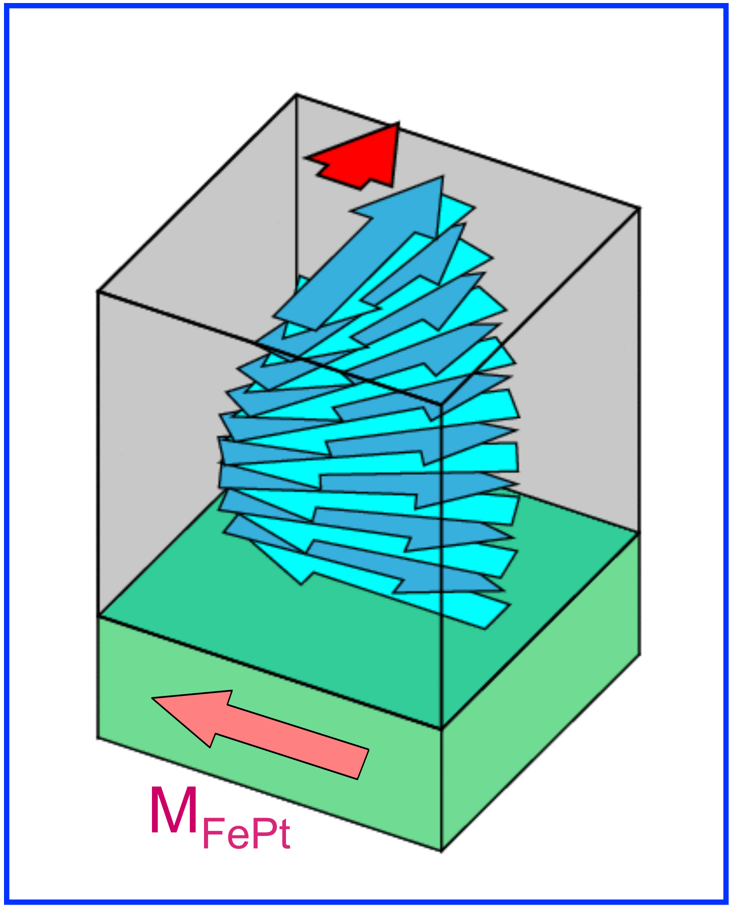


Twisted state stabilized by

a) External field

b) Intrinsic anisotropies, acting as an effective external field

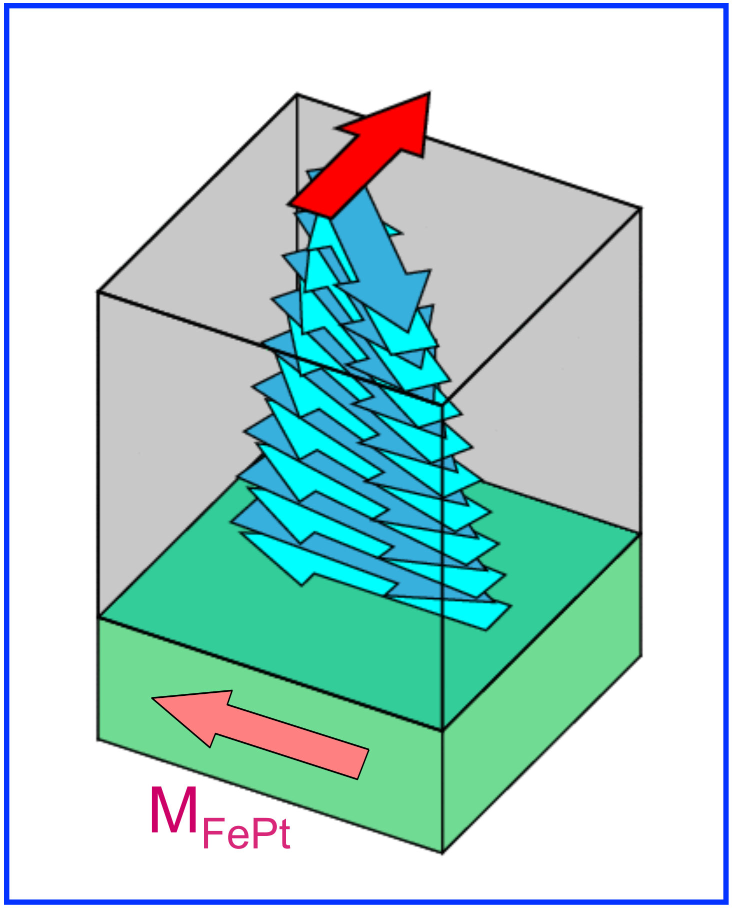
# Twisted States in Exchange-Coupled Antiferromagnetic Multilayers



**External field increases the twist angle**

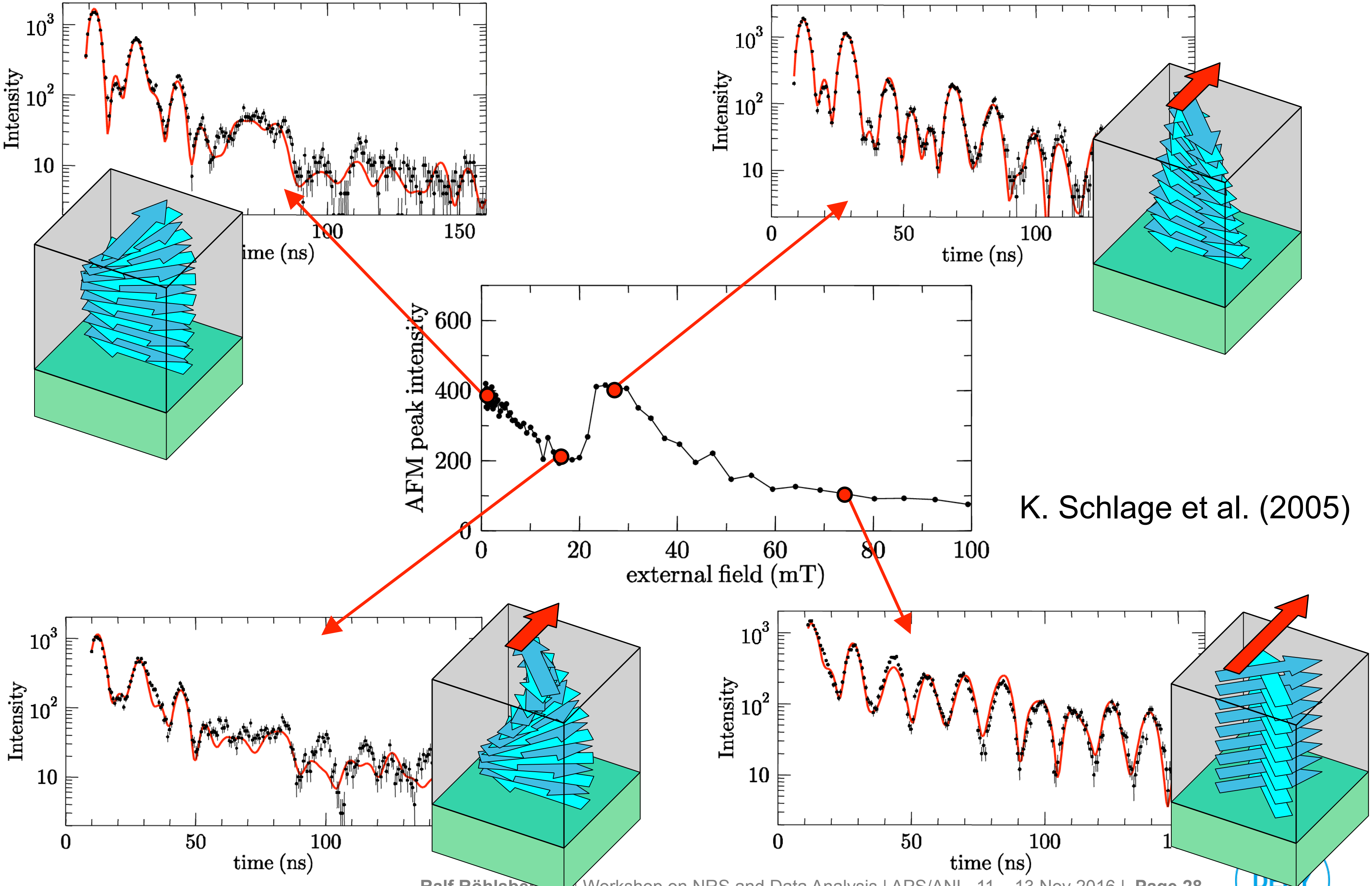
**The potential energy of the magnetic spring increases**

# Twisted States in Exchange-Coupled Antiferromagnetic Multilayers



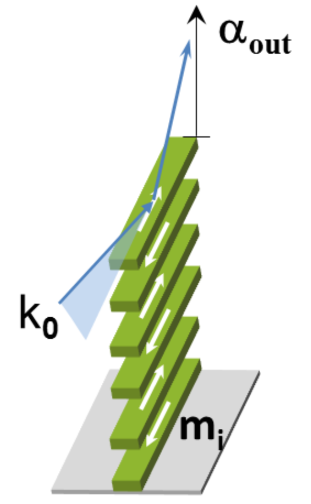
**System moves into state of lower energy by changing its chirality**

# Observation of Chirality Reversal

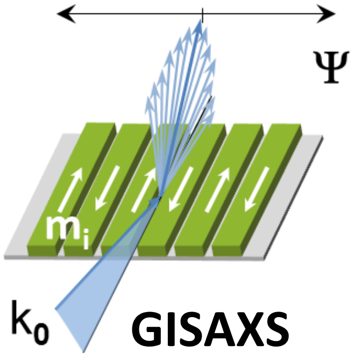


K. Schlage et al. (2005)

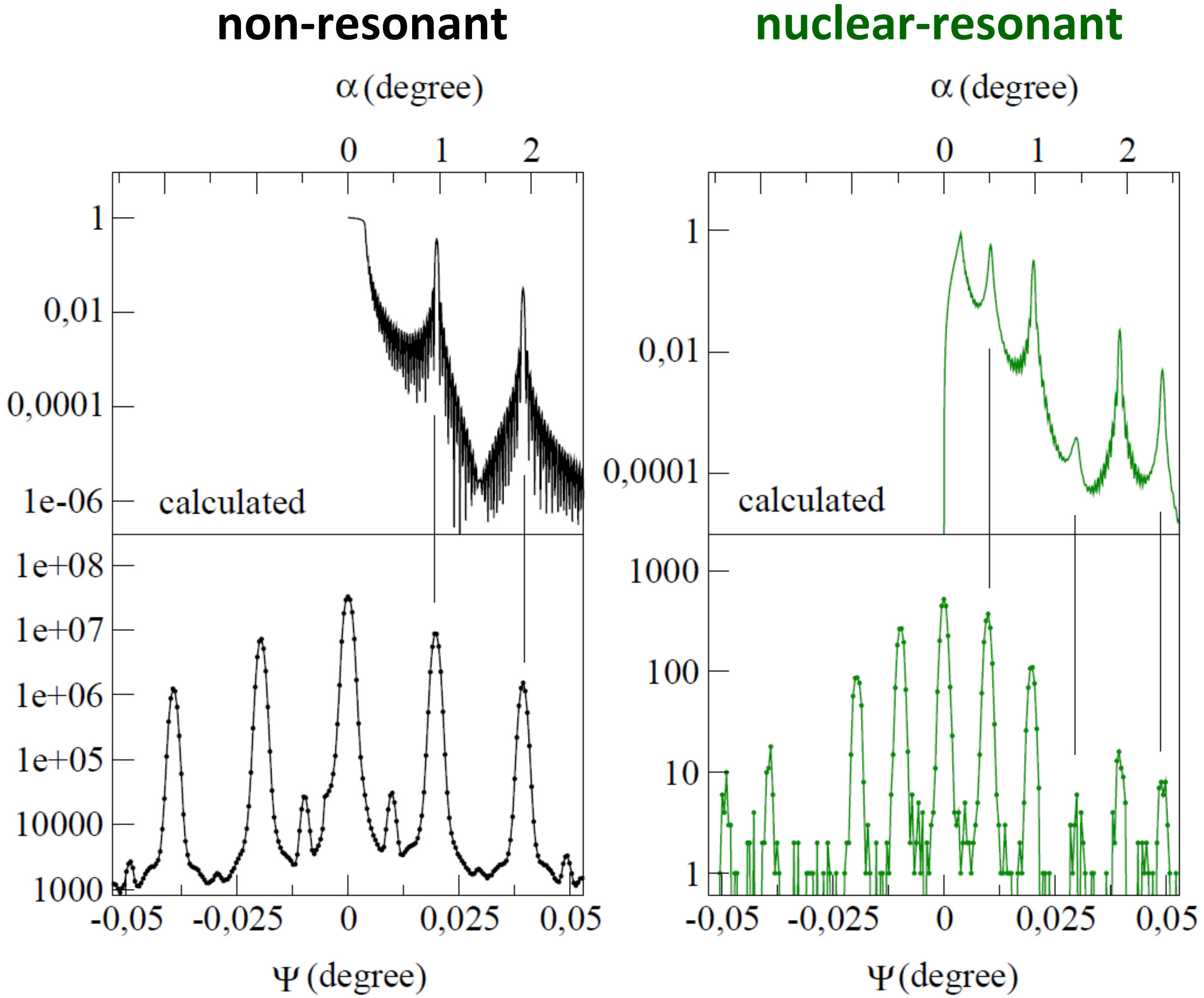
# From vertical to horizontal (lateral) magnetic structures



**Reflectometry**

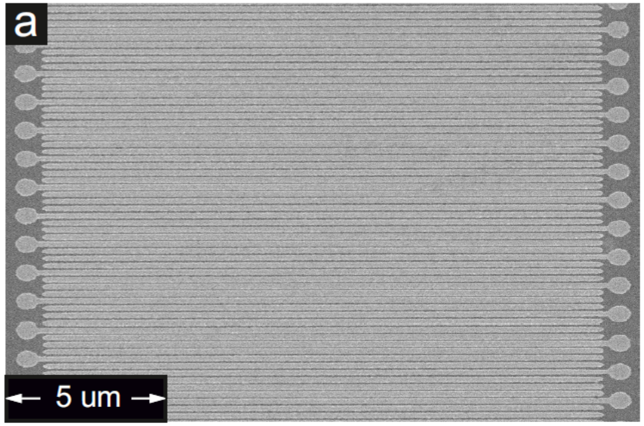


**GISAXS**



# Magnetic Superstructure in Arrays of Nano Wires

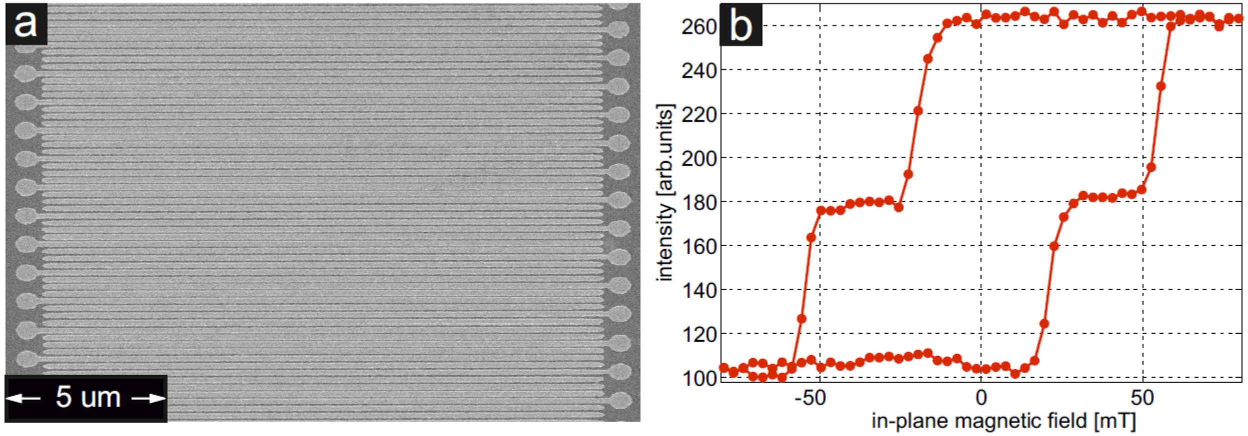
30 nm thick Permalloy ($Ni_{80}{}^{57}Fe_{20}$) nano stripes with pads prepared by electron beam lithography and lift off technique at the IAP (Liudmila Dzemiantsova, Lars Bocklage)



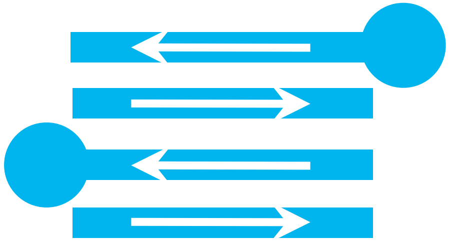
soft magnetic
hard magnetic
soft magnetic
hard magnetic

# Magnetic Superstructure in Arrays of Nano Wires

30 nm thick Permalloy ($Ni_{80}{}^{57}Fe_{20}$) nano stripes with pads prepared by electron beam lithography and lift off technique at the IAP (Liudmila Dzemiantsova, Lars Bocklage)



ferromagnetic state



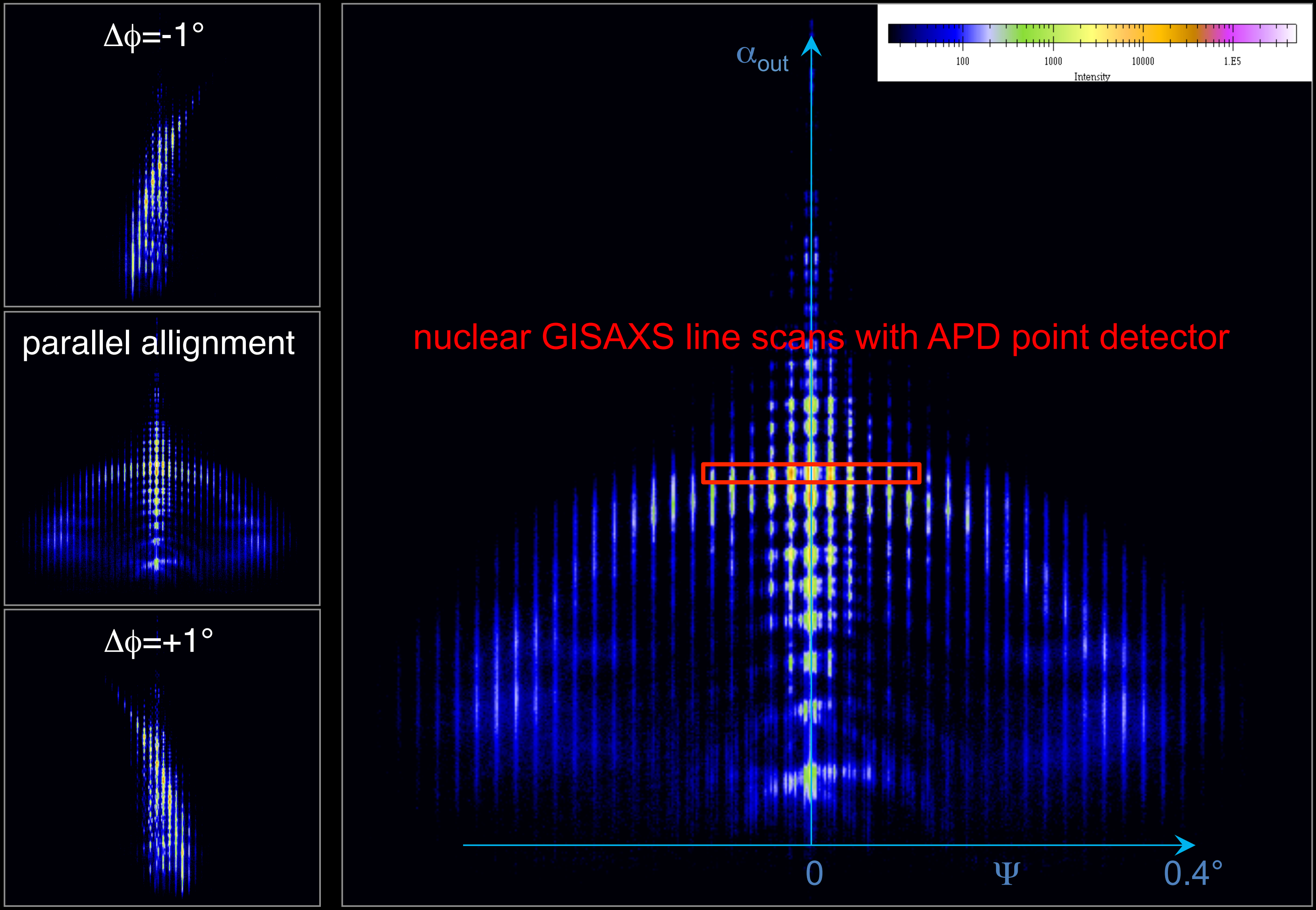
antiferromagnetic state

ferromagnetic state

+ successful preparation of perfectly antiparallel aligned nano stripes?!

- relatively large spacing of 500 nm in magnetic lattice, small sample

# GISAXS on Permalloy Nano Stripes



nuclear GISAXS line scans with APD point detector

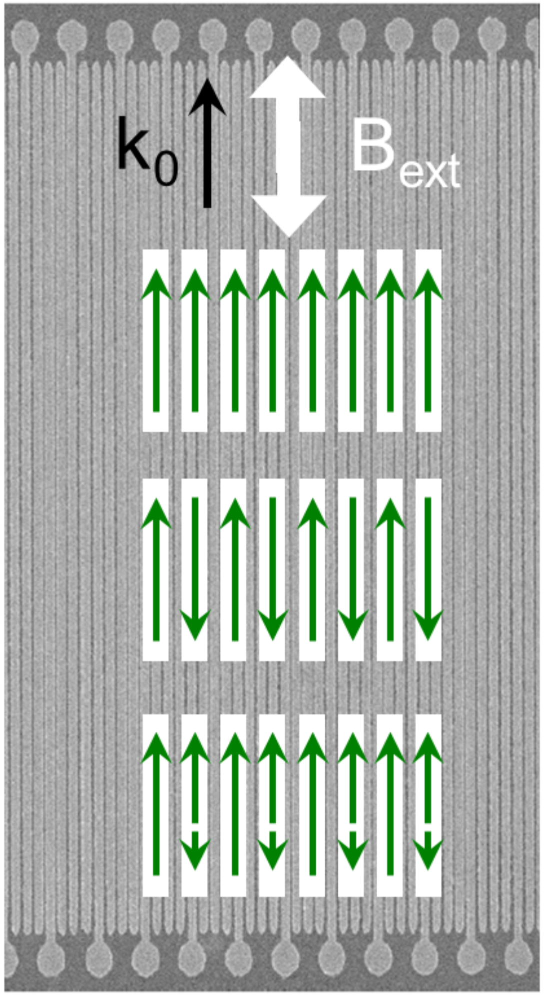
# Resonant GISAXS on Permalloy Nano Stripes

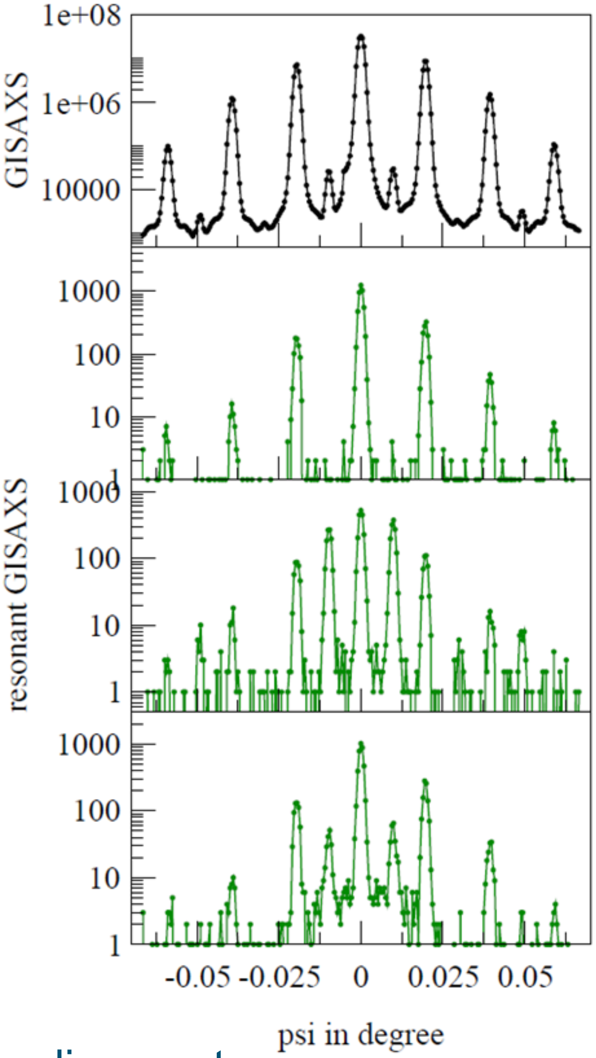
**magnetic states of nano stripes:**

**ferromagnetic**

**antiferromagnetic**

**ferrimagnetic**





Superstructure peaks indicate antiferromagnetic alignment.

Applicable to other periodic sample systems.

# Determination of magnetic dynamics via nuclear resonant scattering

# Probing Magnetic Relaxation: $Fe_2O_3$-oxide clusters in Cu

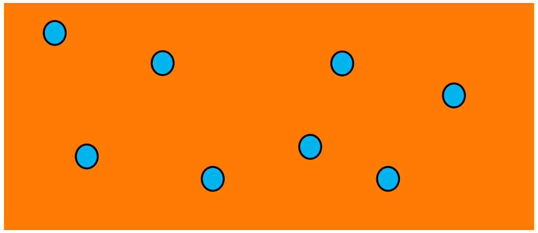
Magnetic moments are fluctuating with a rate much faster than $1/\tau_0$

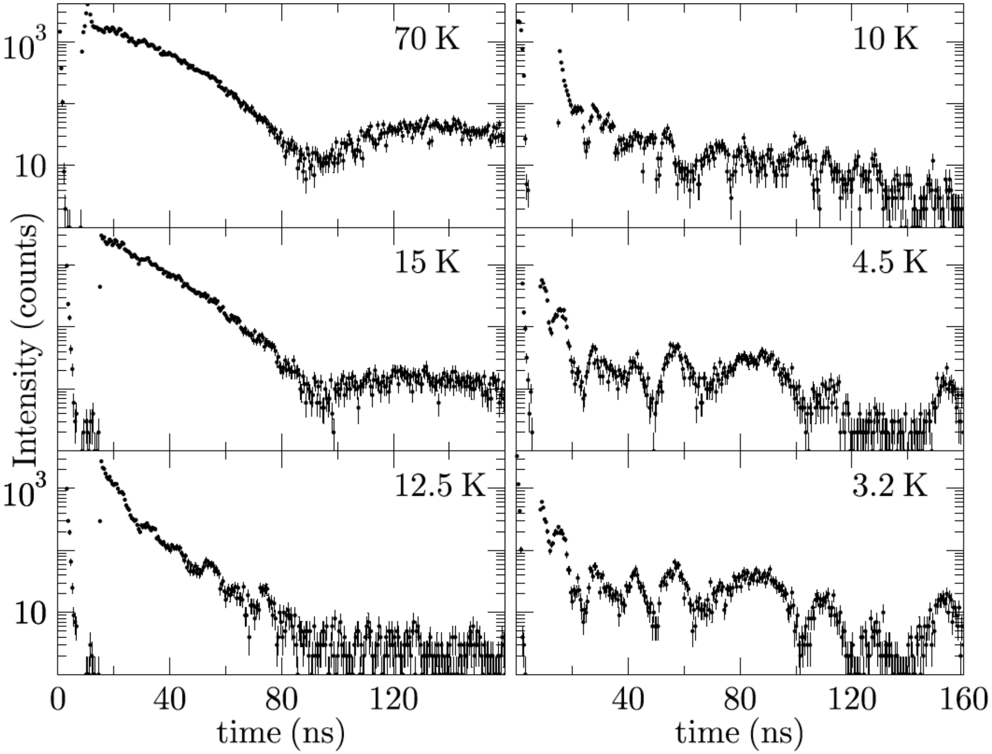
**sample appears nonmagnetic**

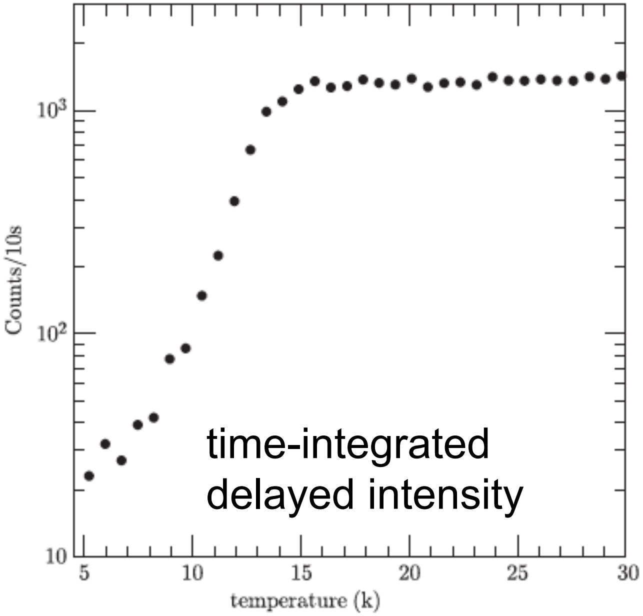
$$\tau = \tau_0 \exp\left(\frac{KV}{k_B T}\right)$$

V = particle volume

K = anisotropy constant







time-integrated delayed intensity

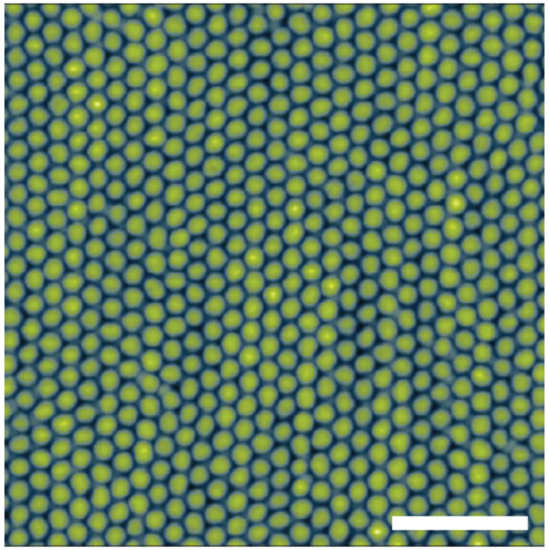
Magnetic moments are 'frozen'

→ **sample appears magnetic**

# Magnetic Relaxation in Nanodots

## Nuclear resonant scattering is very sensitive to magnetic dynamics

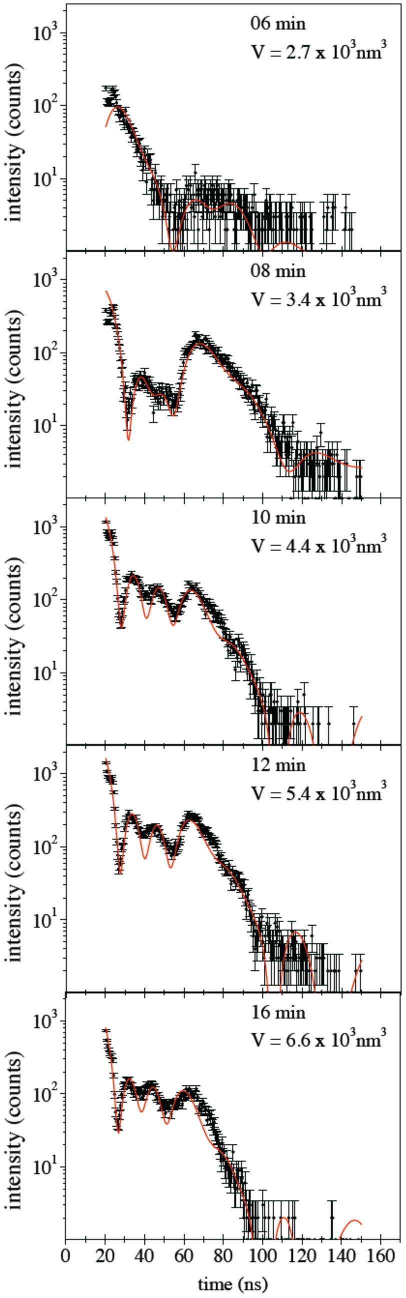
**Fe nanoparticles on patterned diblock-copolymer films**
(Ph.D. thesis of Denise Erb (2015))



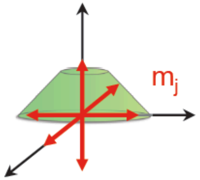
**Relaxation rate**

$$f = f_0 \exp\left(-\frac{KV}{k_B T}\right)$$

V = particle volume
K = anisotropy constant

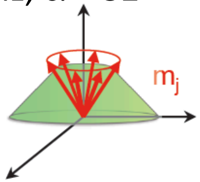


f = 7.1 MHz, isotropic



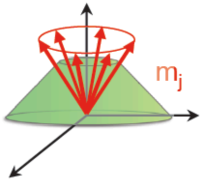
f = 6.7 MHz, $\alpha$ = 32°

f = 6.4 MHz, $\alpha$ = 32°



f = 6.4 MHz, $\alpha$ = 32°

f = 6.4 MHz, $\alpha$ = 28°



# Nuclear Resonant Scattering for Magnonics

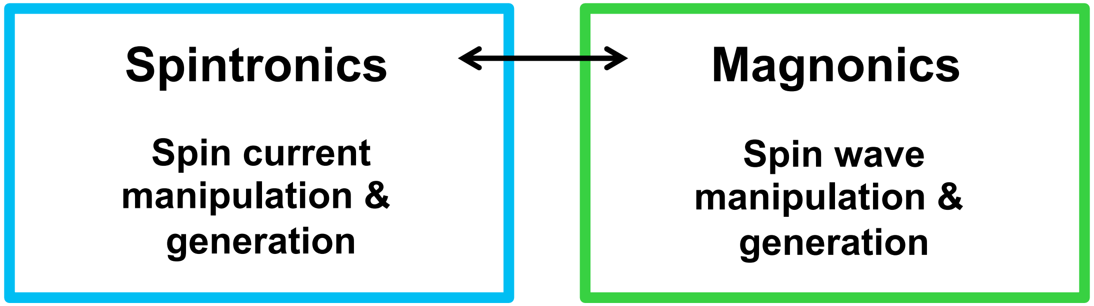
**Magnetic resonances**

magnetic storage → switching times, energy losses

www.hitachigst.com

**Spin manipulation**

functional spin devices



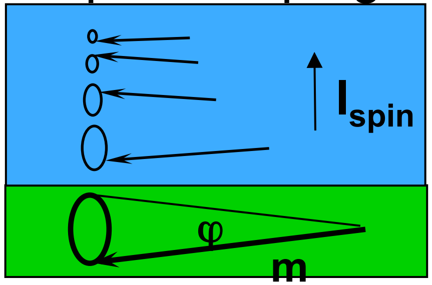
**Spintronics**

**Spin current manipulation & generation**

**Magnonics**

**Spin wave manipulation & generation**

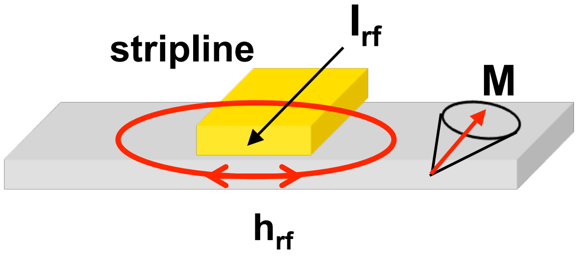
Spin Pumping



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

# Spin excitations in magnetic microstructures

Thin film system

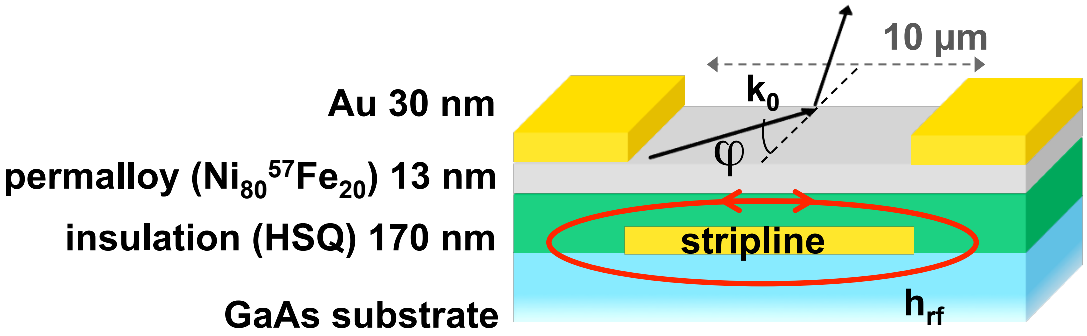


**permalloy ($Ni_{80}{}^{57}Fe_{20}$)**

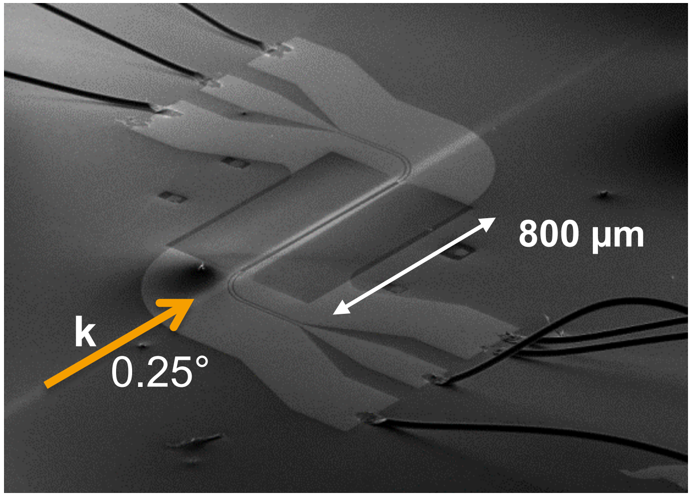
**Goal: Excitation of uniform spin precession**
**→ Kittel mode at ferromagnetic resonance**

# Experimental Setup (CUI Project C3.2)

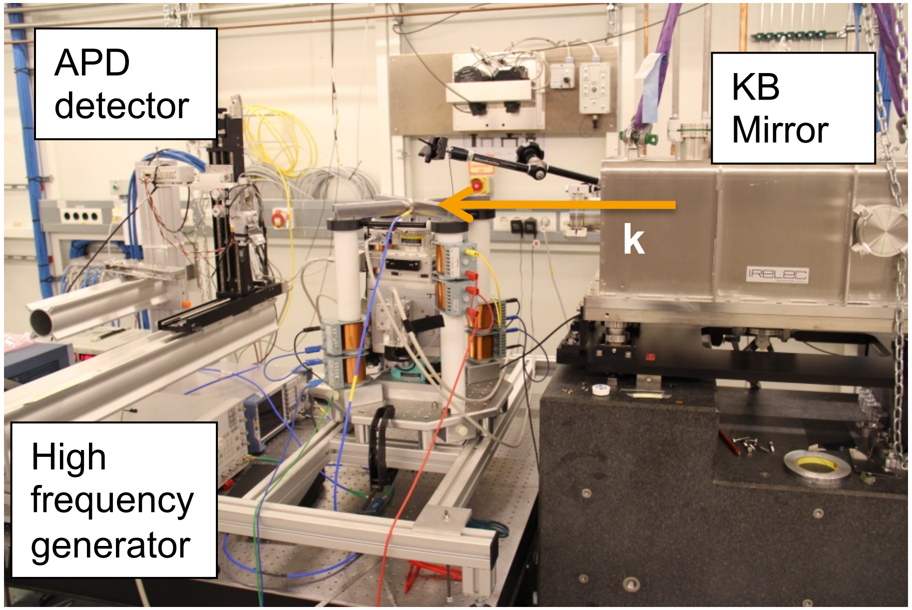
Thin film system



grazing incidence

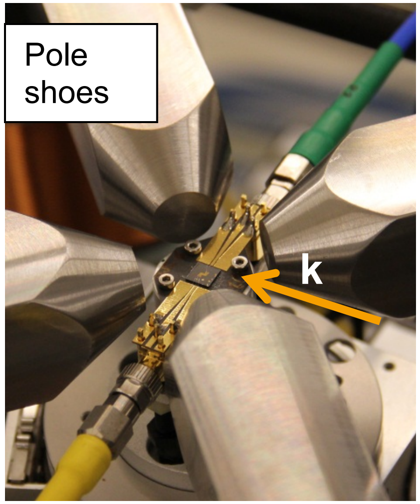


Setup at beamline P01 of PETRA III

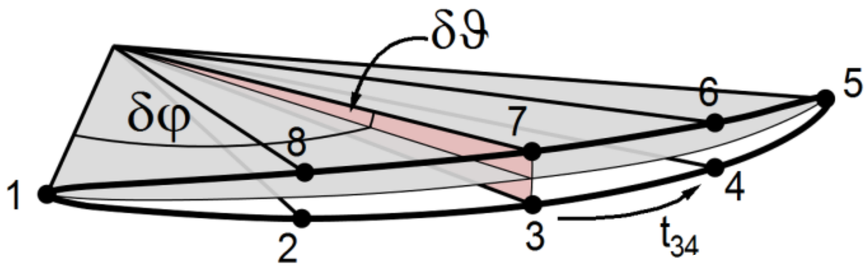


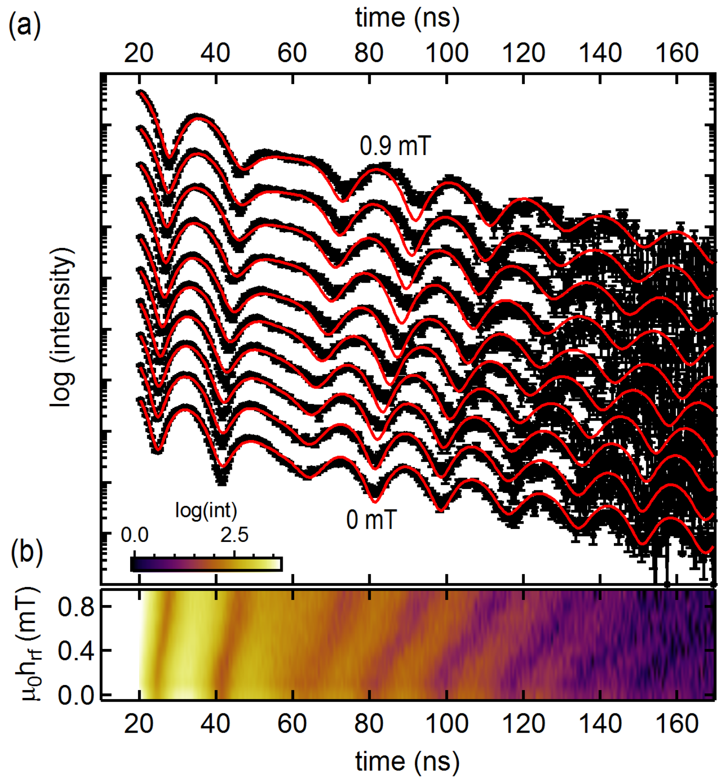
KB mirror focus
10 x 10 µm²

matches typical sample size



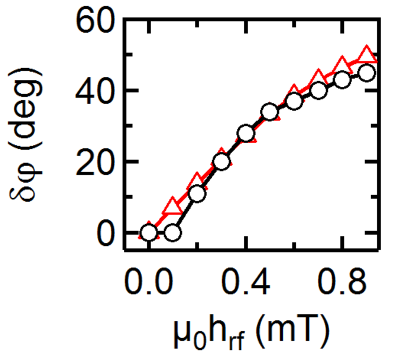
# Spin trajectory determination



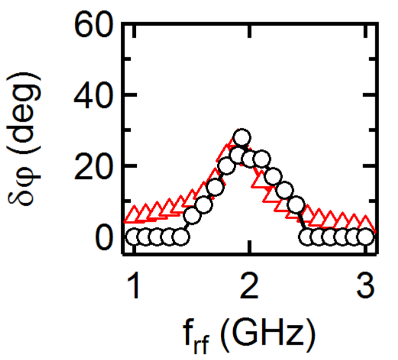


## Determination of the opening angle δφ

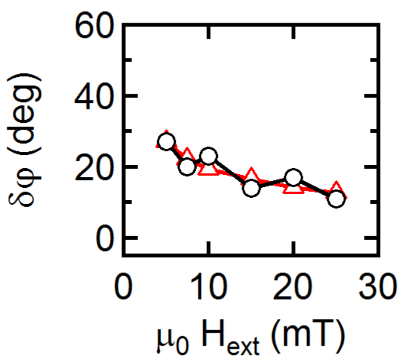
**Power sweep**



**Frequency sweep**



**Field sweep**

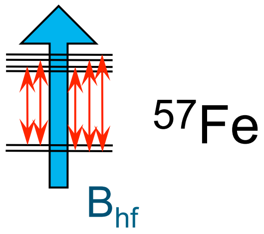


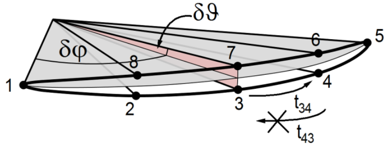
micromagnetic simulations with MicroMagnum software

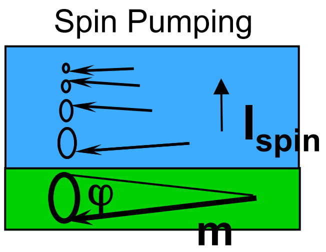
**L. Bocklage et al., Phys. Rev. Lett. 114, 147601 (2015)**

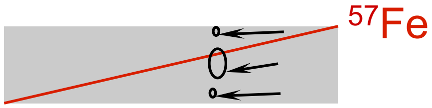
# Summary

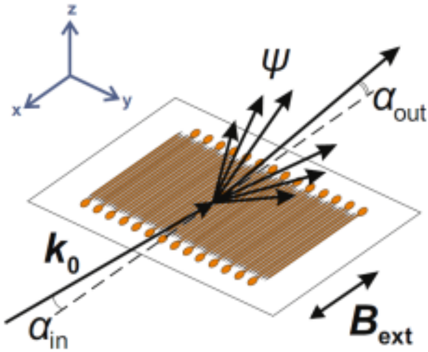
- Nuclear resonant scattering to study to spin structure and spin dynamics
- Determination of magnetic relaxation rates and precession trajectories
- New approach to study magnetization dynamics
  - Depth profile of spin waves via thin isotopic probe layers
  - Non-equilibrium spin dynamics in strongly correlated systems

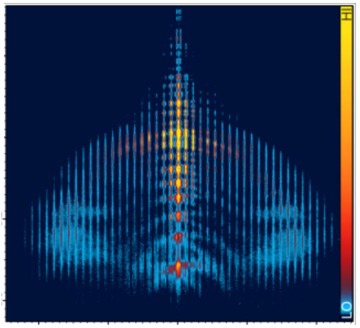




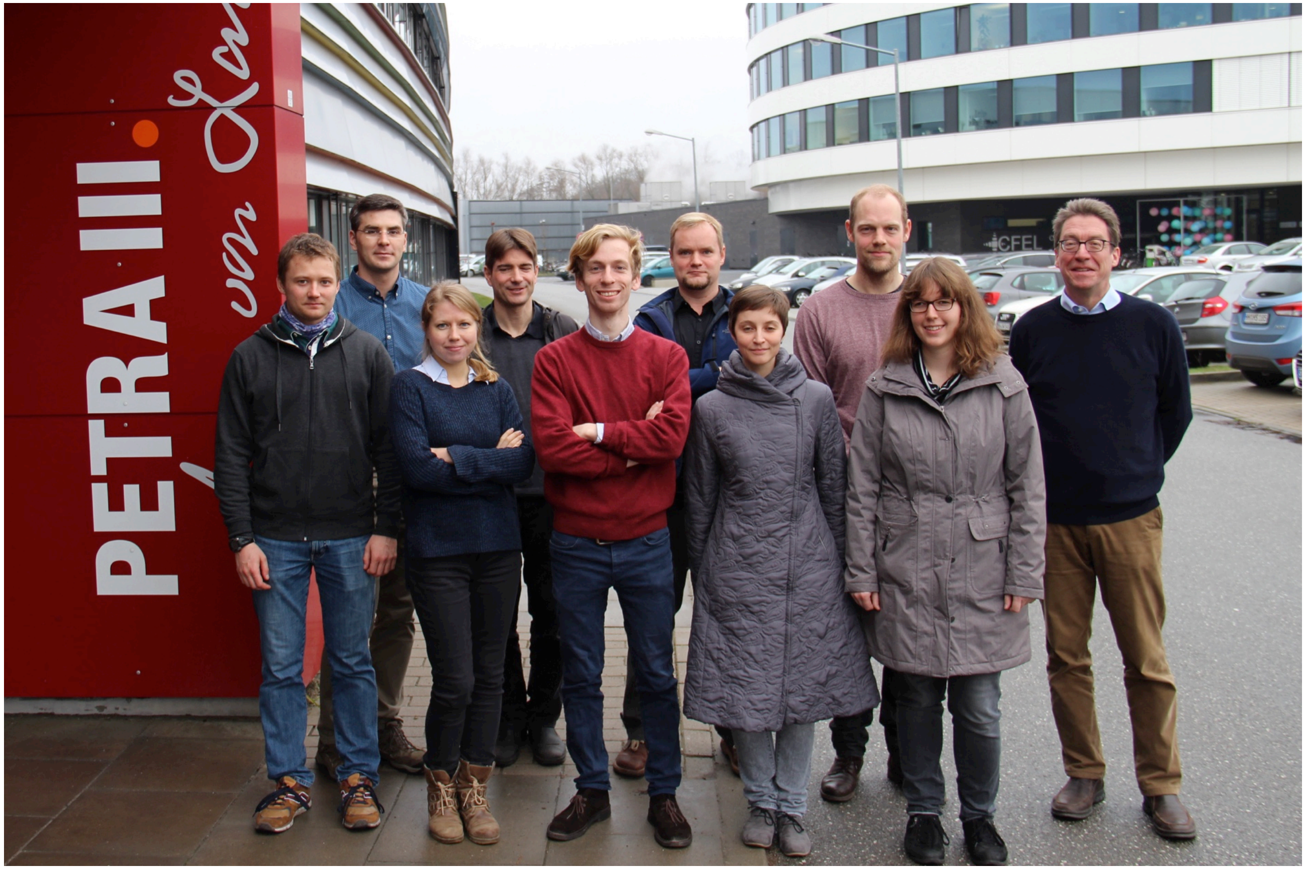








# The Group ‘Magnetism and Coherent Phenomena’



Andrey Siemens

Cornelius Strohm

Kai Schlage

Lars Bocklage

Ralf Röhlsberger

Pavel Alexeev

Liudmila Dzemiantsova

Johann Haber

Denise Erb

Svenja Willing

not on picture: Jakob Gollwitzer