Interchanging Source and Detector: Reciprocity and its Violation in Scattering Experiments

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Outline

- Introduction
  - What is reciprocity?
  - Nuclear resonant scattering of synchrotron radiation (NRS)
  - Off-specular (diffuse) reflectometry: mapping AF domains; symmetry and asymmetry of the ω-scans
- Reciprocity theorems
- Reciprocity in NRS
- Experimental demonstration of switchable reciprocity violation in NRS

What is reciprocity?

G. **Stokes**, Cambridge Dublin Math. J. **4** (1849) 89.
H. von **Helmholtz**, *Handbuch der Physiologischen Optik* (1866);
J.W. Strutt, Baron **Rayleigh**, *The theory of sound*, vol. 1, pp. 150−157, (1877):

‘*If in a space filled with air, which is partly bounded by finitely extended fixed bodies and is partly unbounded, sound waves be excited at any point A, the resulting velocity potential at a second point B is the same both in magnitude and phase, as it would have been at A, had B been the source of sound*’

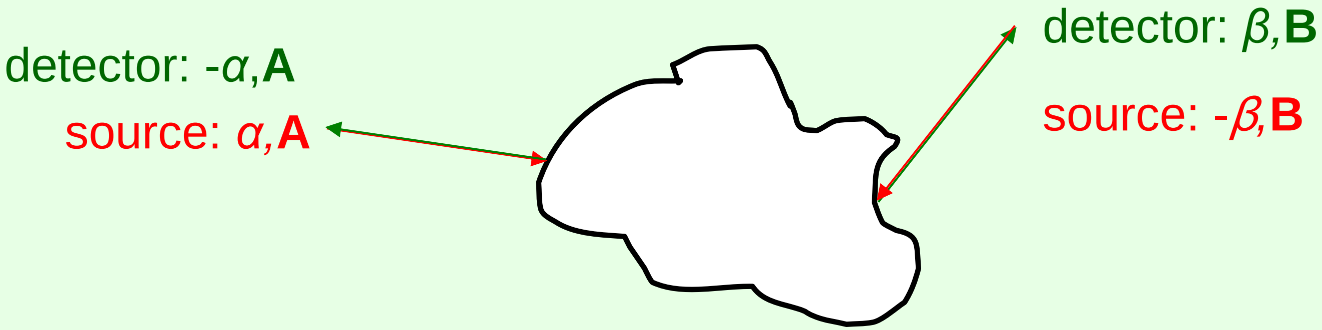
de Hoop (1959, 1987), **Raab** (2001) and **de Lange** (2003) as interpreted by **Potton** (2004):

The scattering amplitude of the process when a wave incident from the direction α having polarization **A**, scatters to the direction β and to polarization state **B** is identical with the scattering amplitude of the process of scattering from the direction $-\beta$ with polarization **B** to the direction $-\alpha$ and polarization **A**

$$f(\alpha, \beta) = f(-\beta, -\alpha)$$

What is reciprocity?

$$f(\alpha, \beta) = f(-\beta, -\alpha)$$



R.J. Potton, Rep. Prog. Phys. **67** (2004) 717.

What is reciprocity?

Reciprocity is not a basic symmetry

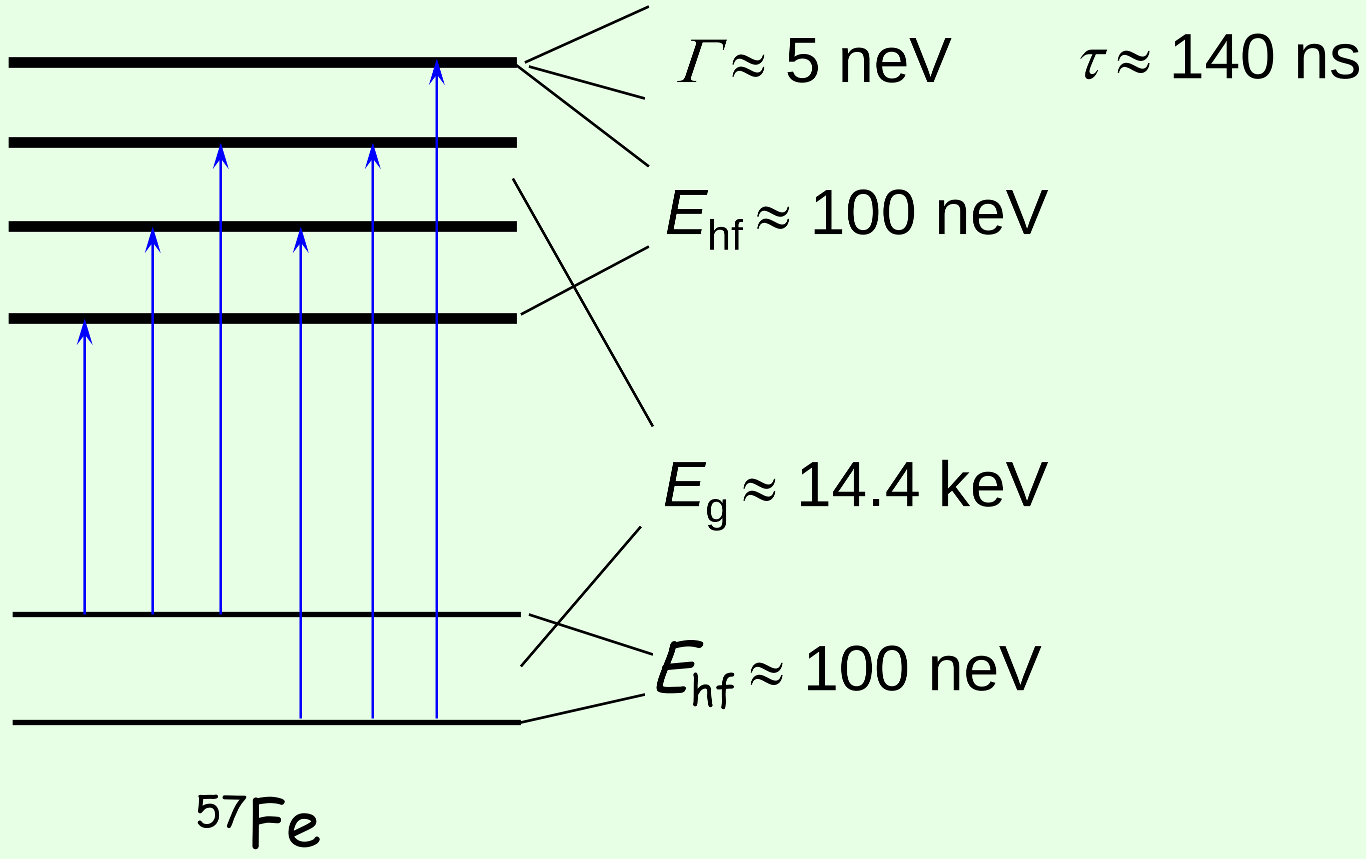
of all scattering processes;

depending on the type

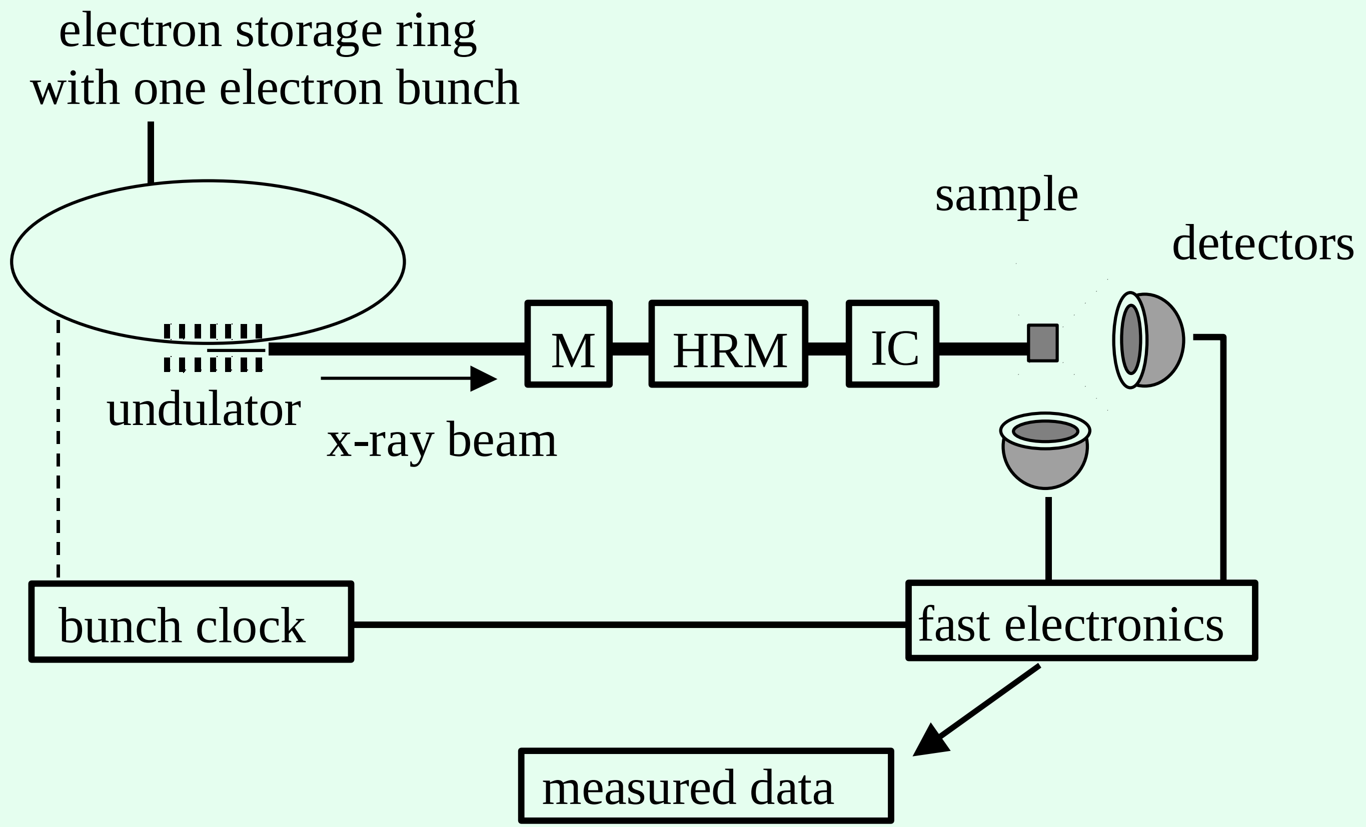
of the scattering potential, it

may or may not be fulfilled.

Hyperfine splitting of nuclear levels

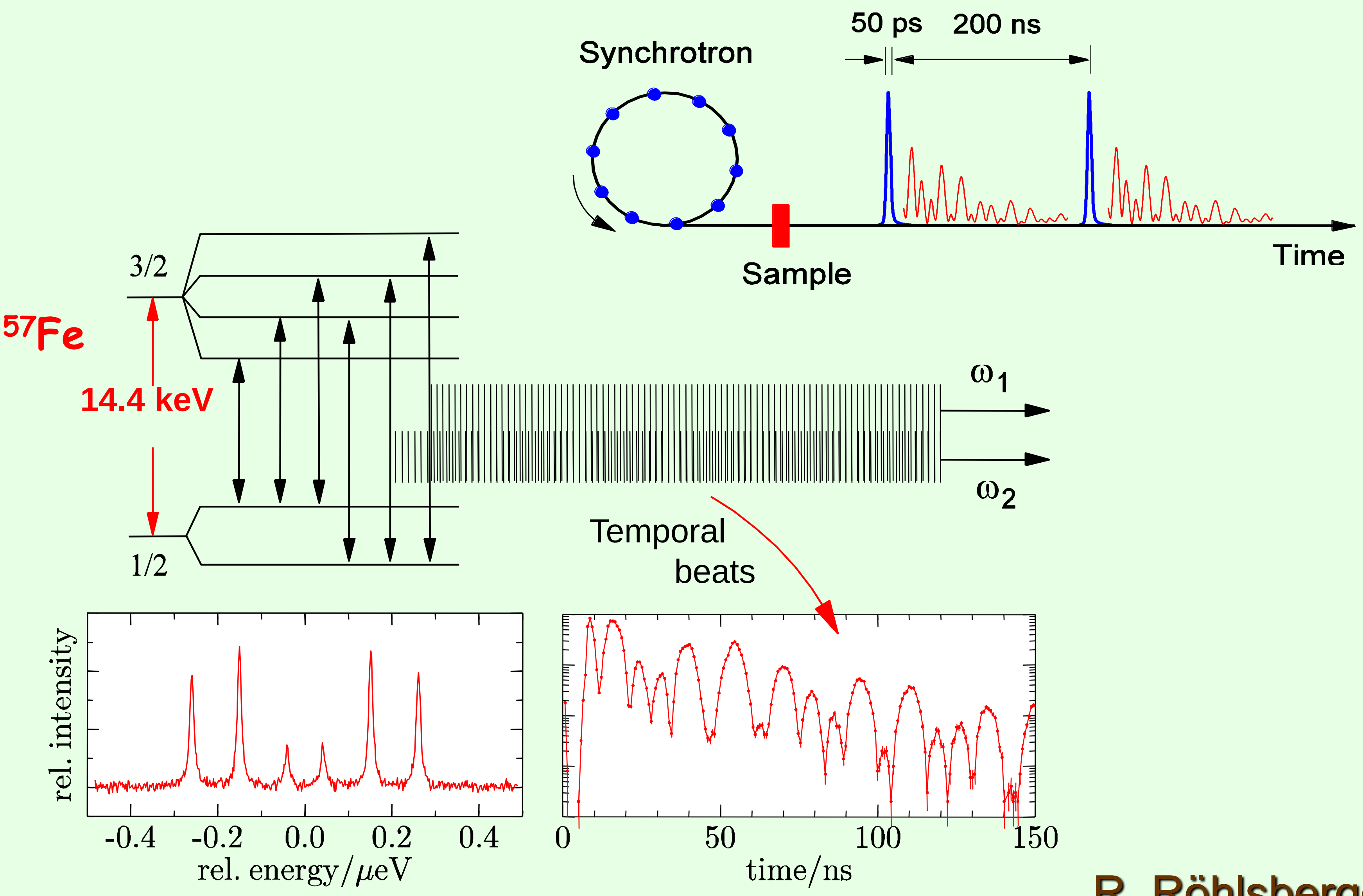


Principle of a nuclear resonant scattering experiment



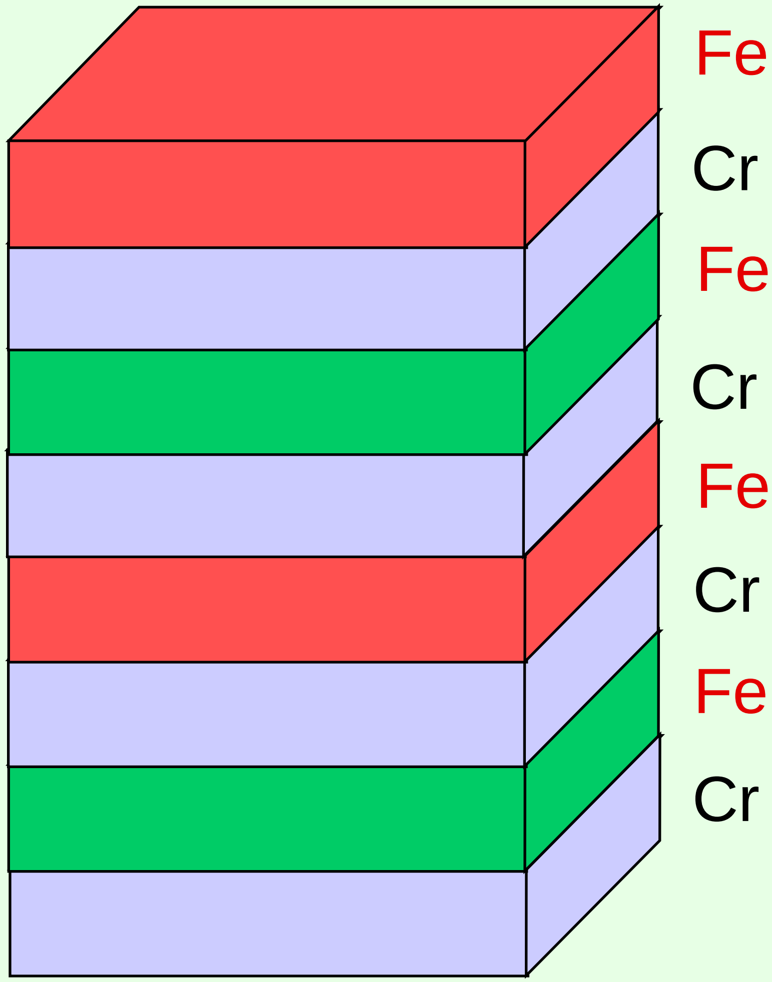
H. Grünsteudel

Energy- and time-domain Mössbauer spectra

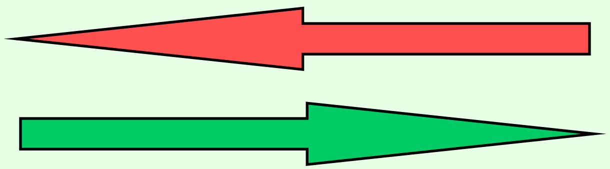


R. Röhlsberger

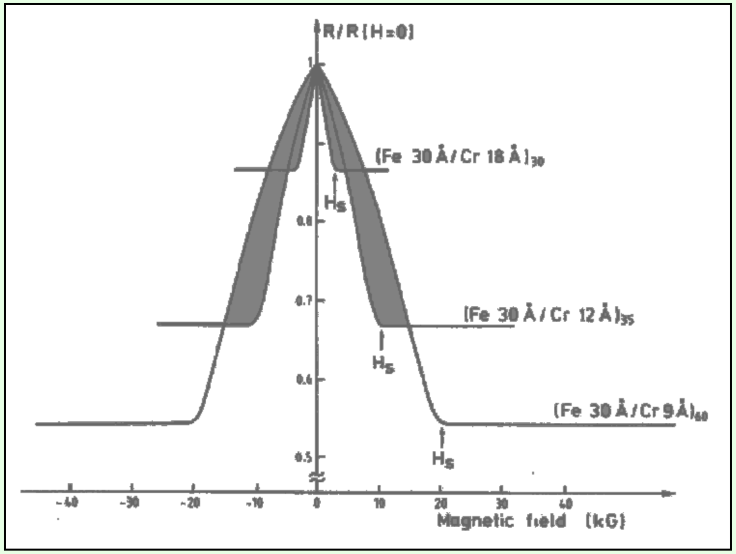
Antiferromagnetically coupled Fe/Cr multilayer



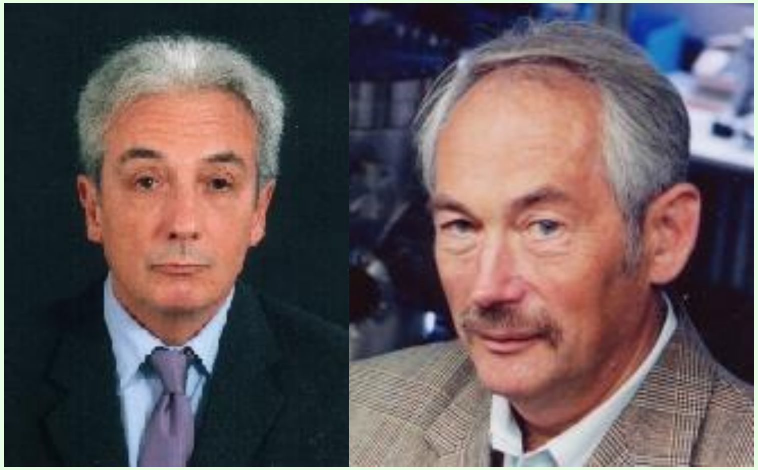
Layer magnetisations:



Giant magnetoresistance

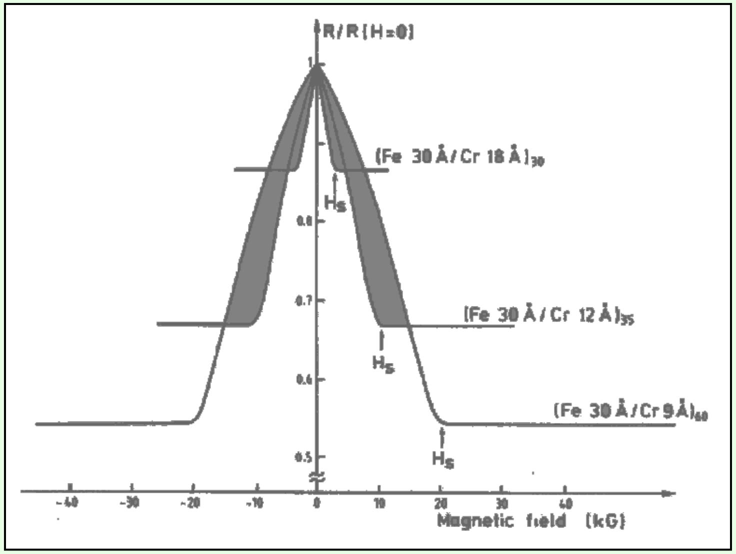


Antiferromagnetically coupled Fe/Cr multilayer

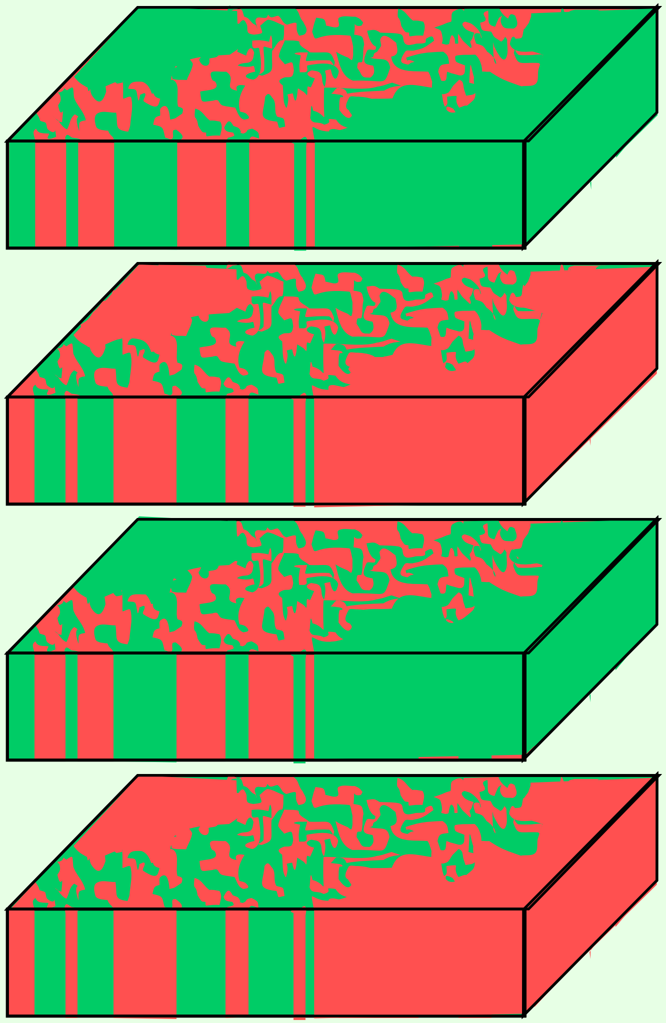


A. Fert and P. Grünberg
Nobel Prize in physics, 2007

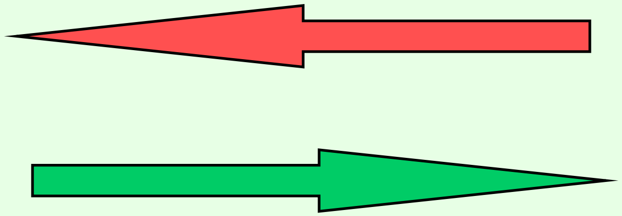
Giant magnetoresistance



Patch domains in AF-coupled multilayers

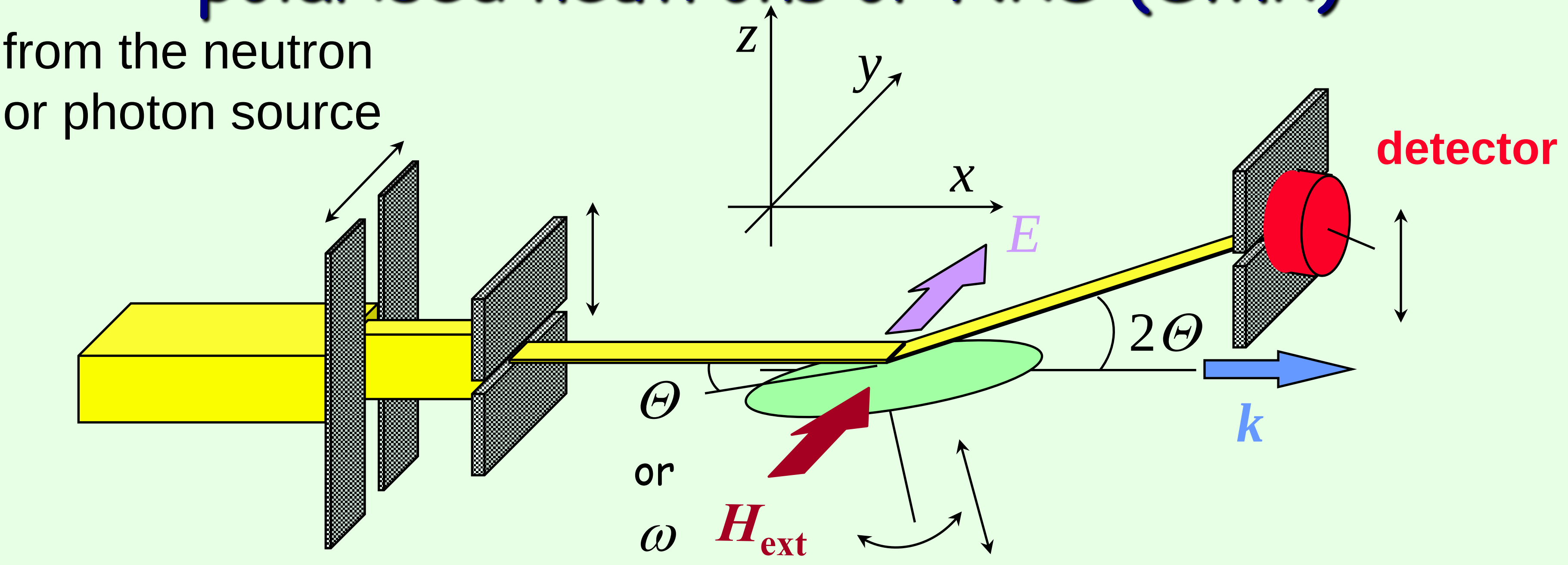


Layer magnetisations:



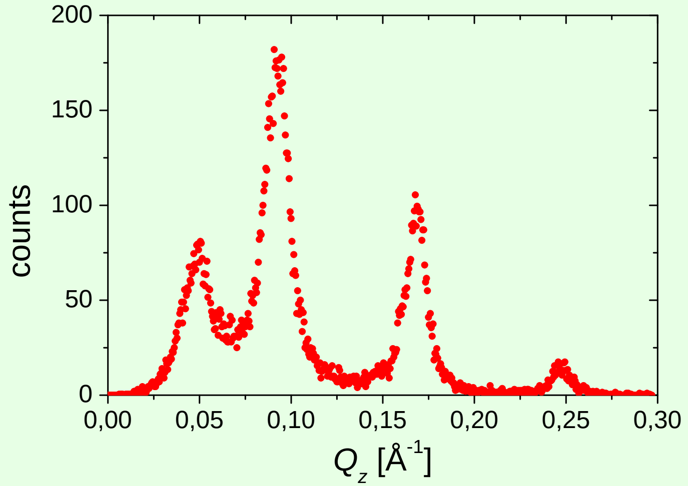
The 'magnetic field lines' are shortcut by the AF structure → the stray field is reduced → no 'ripple' but 'patch' domains are formed.

Specular and off-specular reflectometry: polarised neutrons or NRS (SMR)

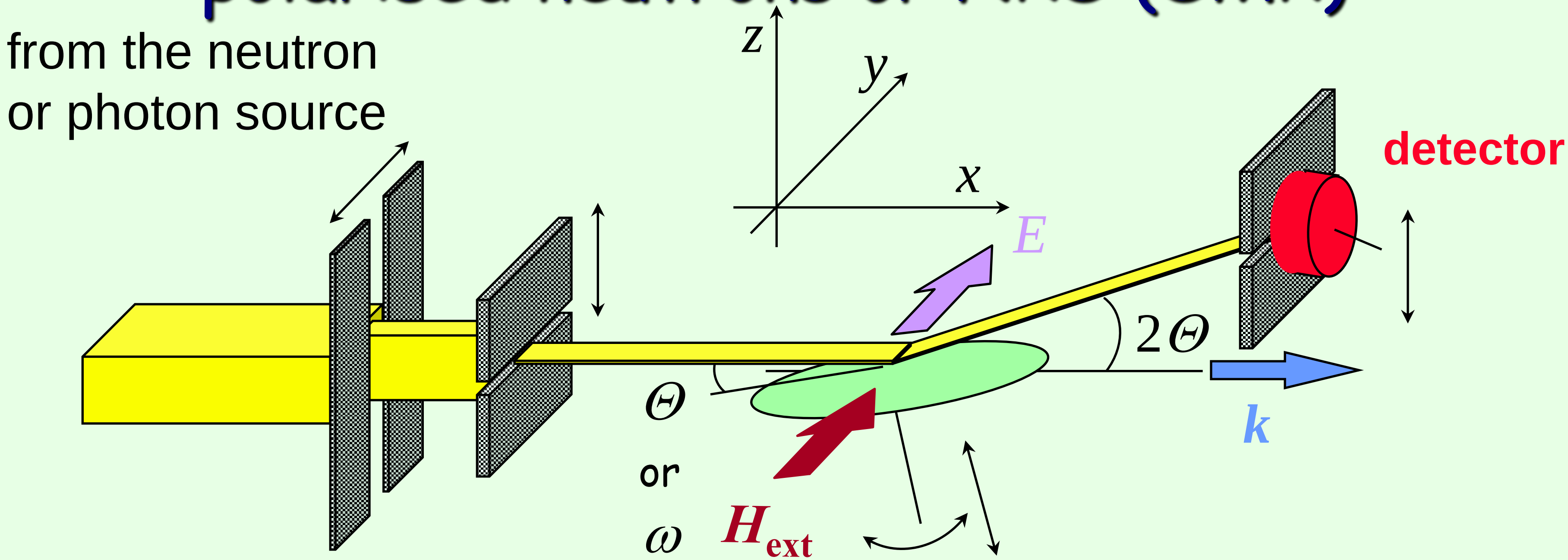


Θ/2Θ-scan: Q_z-scan

$d = 2\pi/Q_z$

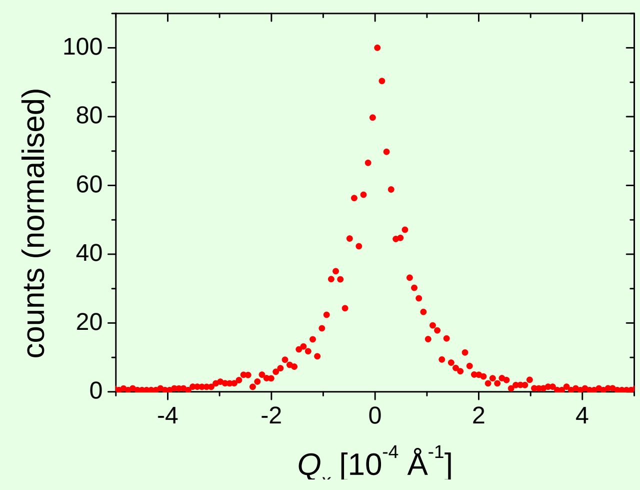


Specular and off-specular reflectometry: polarised neutrons or NRS (SMR)



ω-scan: Q_x-scan

$$\xi = 1/\Delta Q_x$$



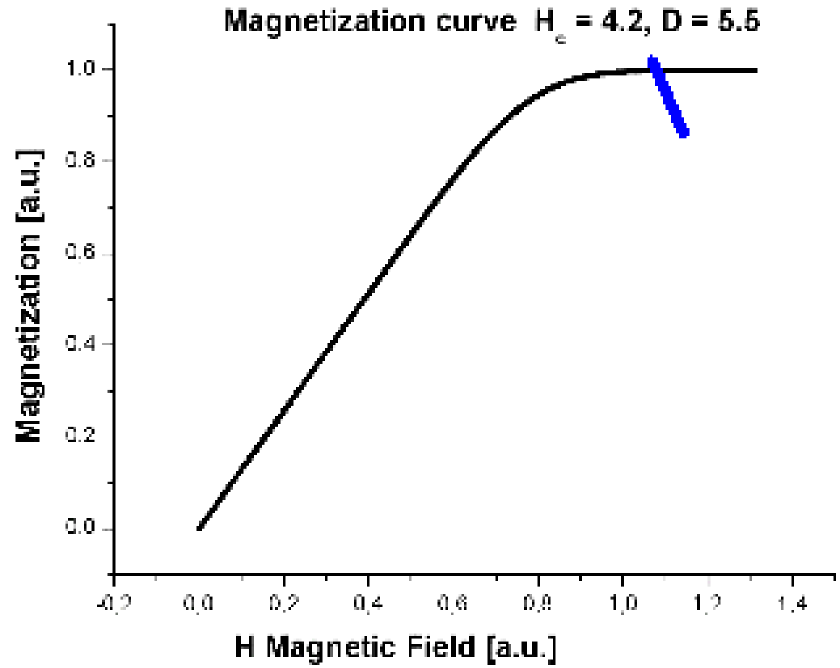
The off-specular scattering width

- The off-specular (diffuse) scattering width around an AF reflection stems only from the lateral correlation of the magnetisation (magnetic domains and magnetic roughness).
- The diffuse scattering width ΔQ_x at an AF reflection is, in first Born approximation, inversely proportional to the correlation length ξ of **M**:

$$\xi = 1/\Delta Q_x$$

At an AF reflection, ξ is the average domain size!

AF domain formation and ripening



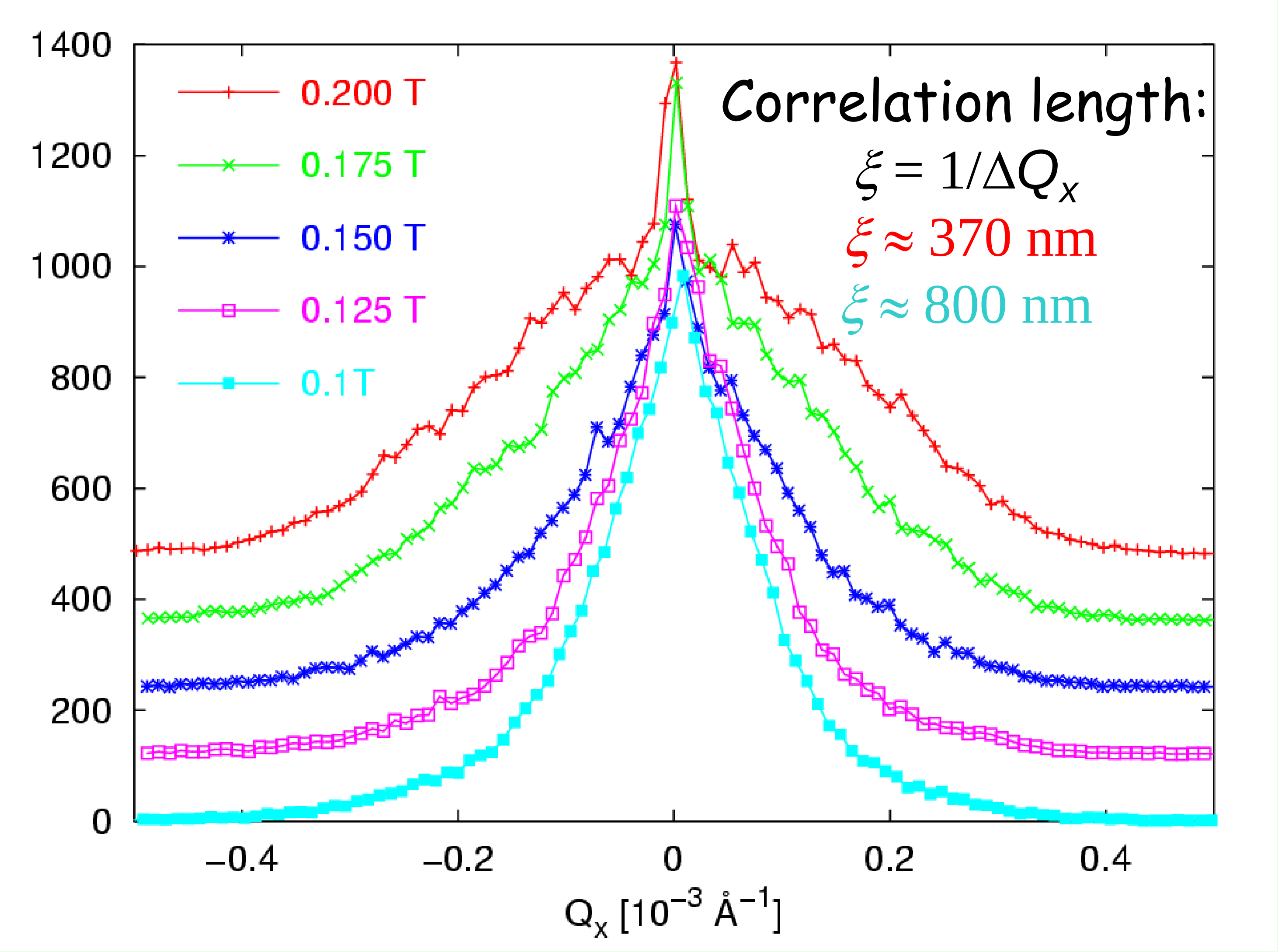
Domain pattern

Autocorrelation

Domain ripening at H_c = 4.2, D = 5.5

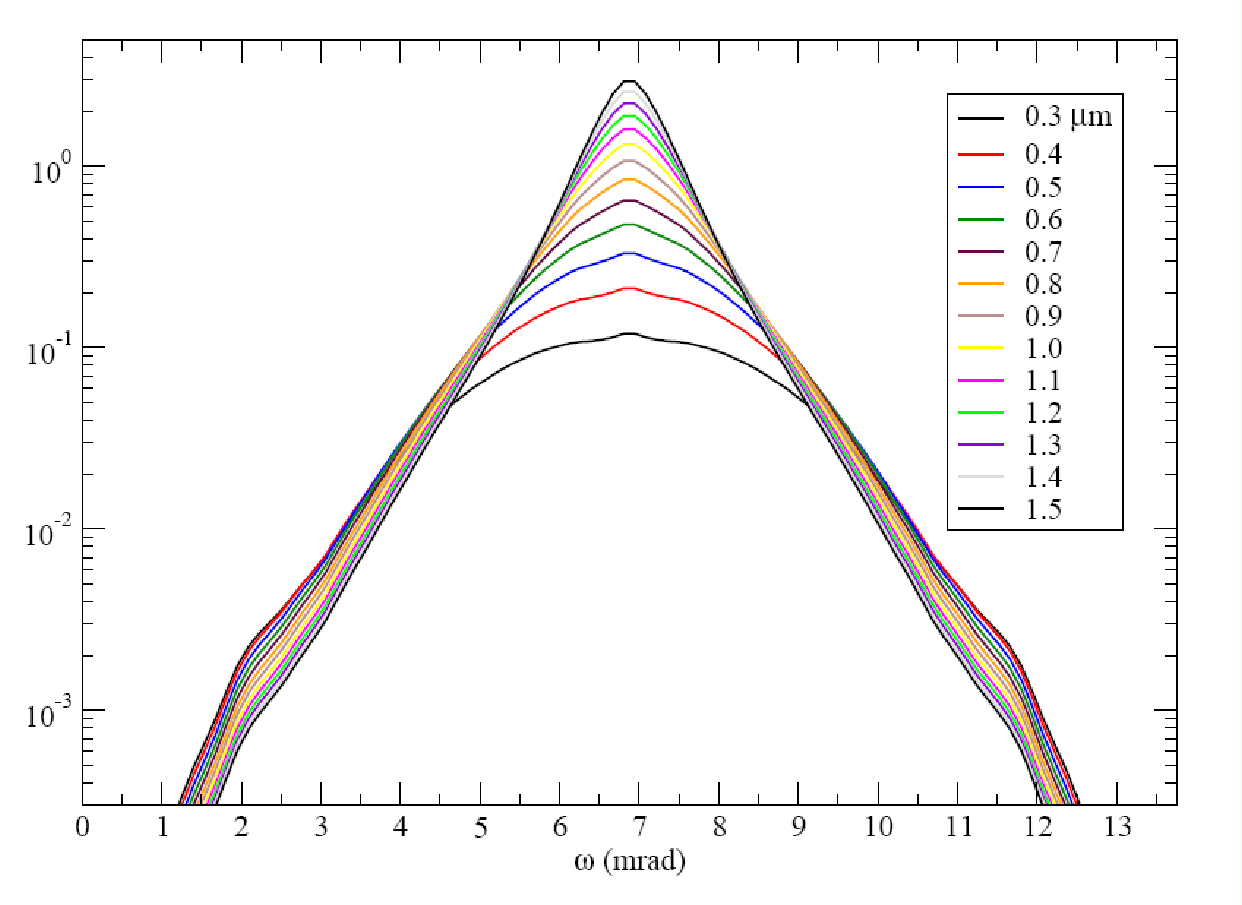
Domain ripening: off-specular SMR

MgO(001)[^{57}Fe(26Å)/Cr(13Å)]$_{20}$
2Θ @ AF reflection, hard axis



ESRF
ID18

Change of the diffuse scatter shape from Gaussian to exponential

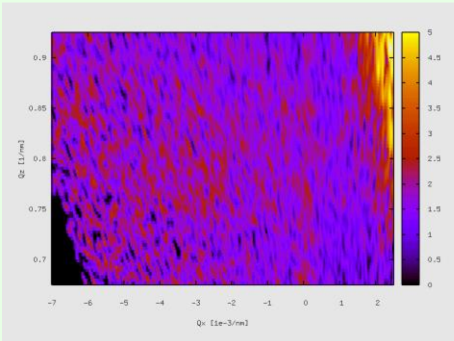


Full DWBA calculation of the SMR diffuse scatter

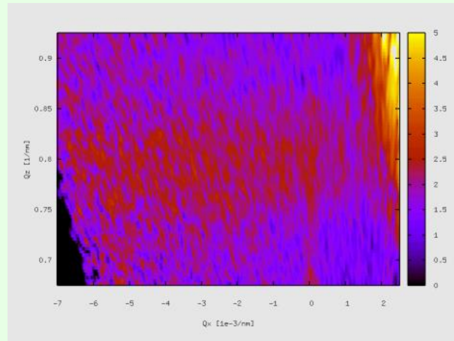
L. Deák et al., PRB **76**, 224420 (2007).

Domain ripening: off-specular PNR, easy axis

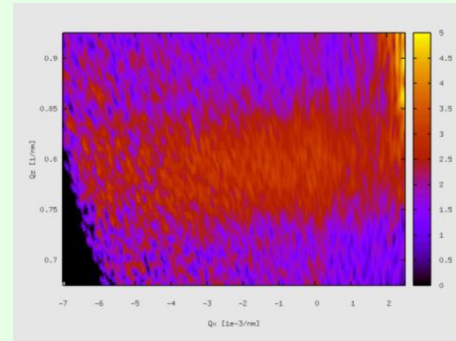
Q_z



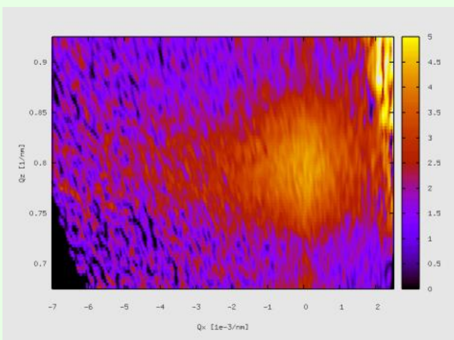
600 mT



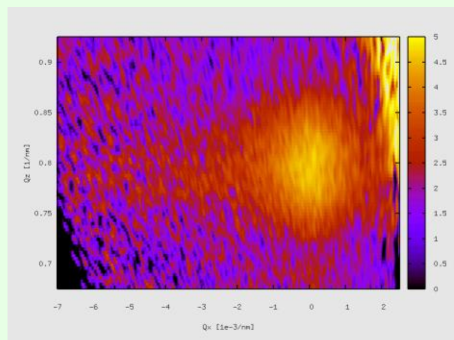
450 mT



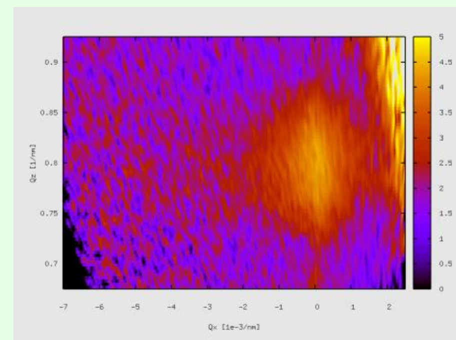
300 mT



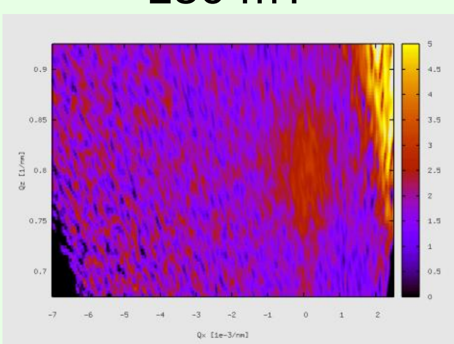
150 mT



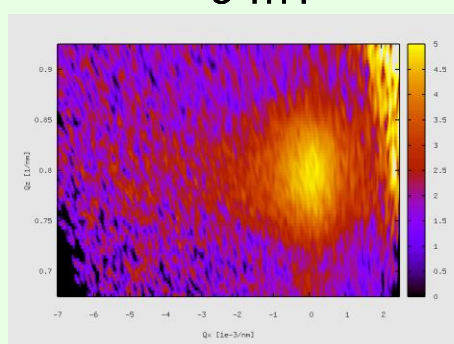
9 mT



300 mT



600 mT



9 mT

MgO(001)[^{57}Fe(26Å)/Cr(13Å)]$_{20}$

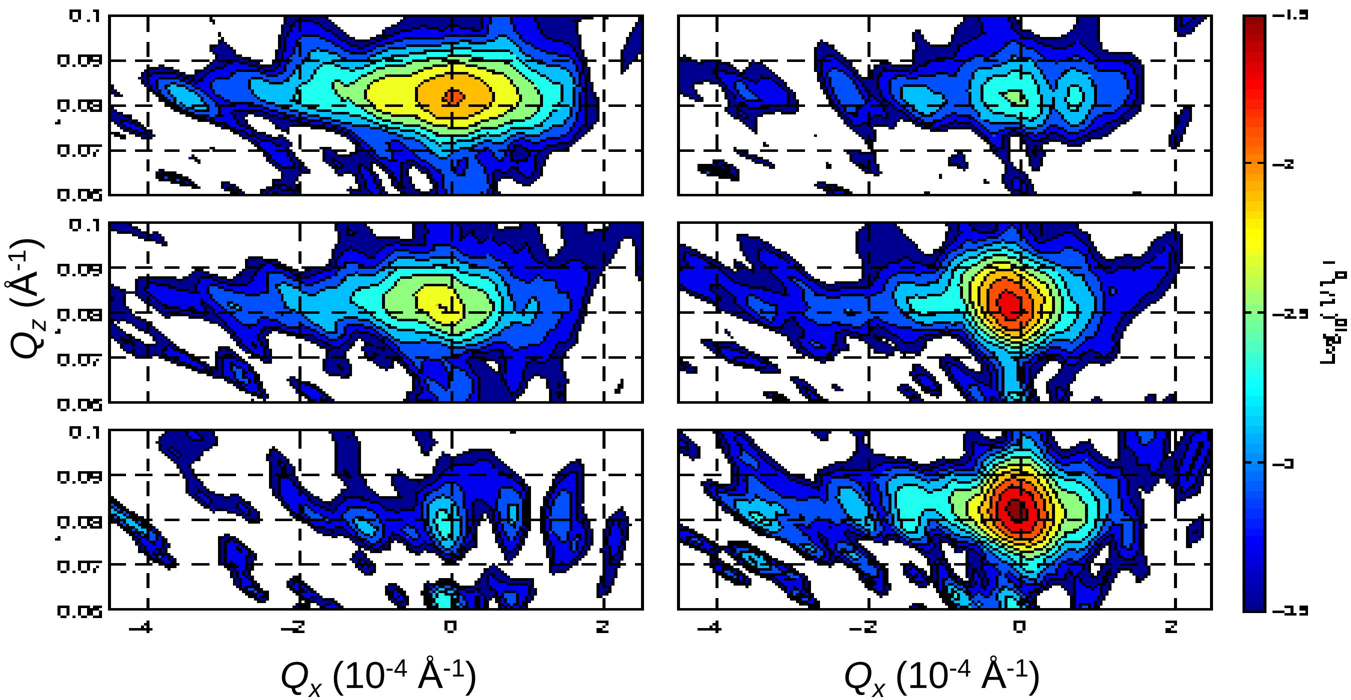
+ → - scattering

JINR Dubna, REMUR

Q_x

Spin-flop induced domain coarsening (PNR)

$MgO(001)[^{57}Fe(26Å)/Cr(13Å)]_{20}$, easy axis



7 mT

14.2 mT

35 mT

non-spin-flip scattering $\mathbf{S}_n \parallel \mathbf{M}$

spin-flip scattering $\mathbf{S}_n \perp \mathbf{M}$

D.L. Nagy et al., PRL **88**, 157202 (2002).

JINR Dubna SPN-1

Asymmetric ω-scan in X-ray reflectometry

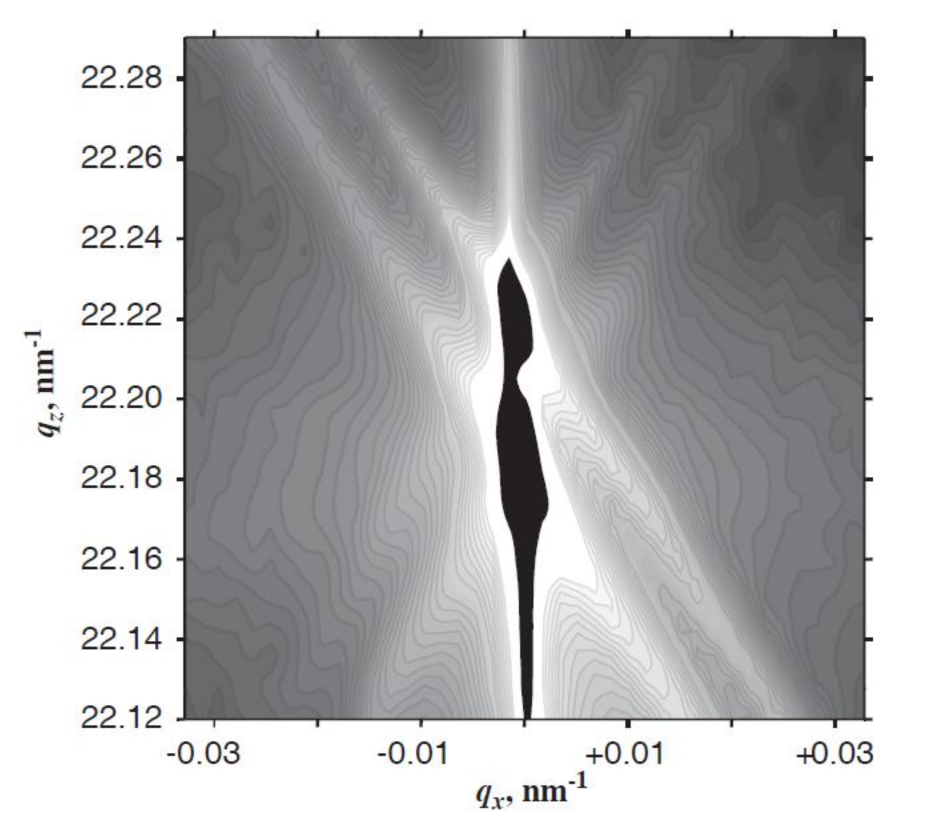
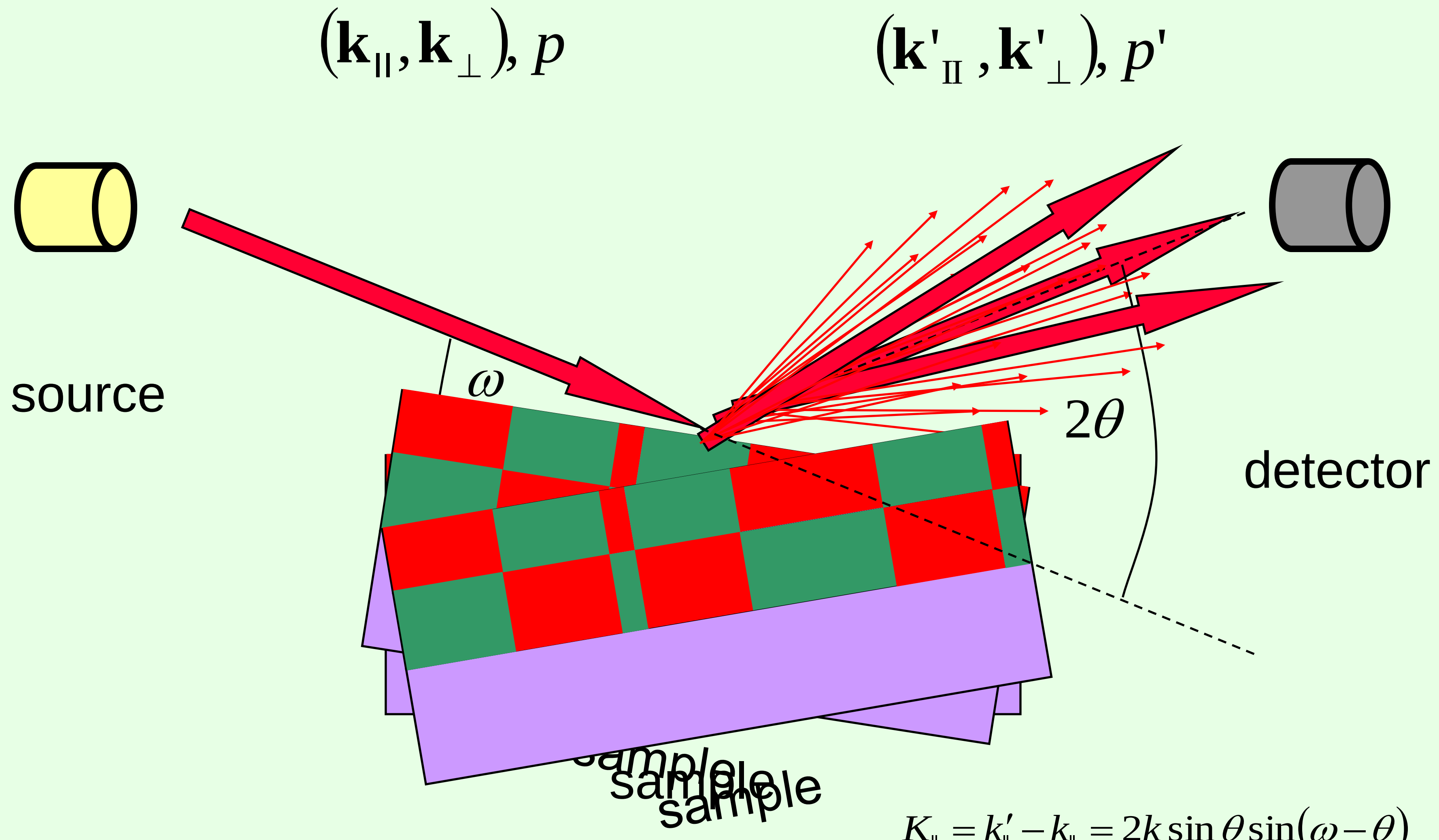


Fig. 3. Diffraction maps of the 0 0 2 reflections from the AlAs/GaAs superlattice and GaAs substrate. The in-plane projection of the momentum transfer is plotted parallel to the horizontal axis. The momentum transfer projection, normal to the lateral planes, is plotted parallel to the vertical axis.

V.A. Chernov et al, Physica B **357,** 232 (2005).

The ω-scan



$$K_{\parallel} = k'_{\parallel} - k_{\parallel} = 2k\sin\theta\sin(\omega-\theta)$$

$$k_{\perp} = k\sin\omega$$

$$k'_{\perp} = -k\sin(2\theta-\omega)$$

Symmetry and asymmetry of the ω-scan

Even if the scattering process fulfils reciprocity,

the ω-scan is only symmetric if the sample has a 180º plane-perpendicular rotational symmetry.

Reciprocity theorems

Scattering theory:

(J.R. Taylor, „Scattering Theory: The Quantum Theory on Non-relativistic Collisions” Wiley, p 354, footnote 6):

- "*Time-reversal* invariance",
- "principle of *micro-reversibility*",
- "principle of *detailed balance*" and
- "*reciprocity*" (principle, theorem or symmetry?)

"... there is a considerable confusion as to the precise meaning of these terms...".

reciprocity vs. time-reversal or 180° rotation??

Reciprocity theorems

1. Scattering theory (QM): $\langle \alpha | \mathrm{T} | \beta \rangle = \langle -\beta | \mathrm{T} | -\alpha \rangle$

(L. D. Landau and E. M. Lifshitz, Quantum Mechanics – Non-Relativistic Theory)

for real potentials (time reversal symmetry)

time reversal ≡ reciprocity → Landau's "reciprocity theorem"

2. Optics: $\langle \alpha | \mathrm{T} | \beta \rangle = \langle -\beta | \mathrm{T} | -\alpha \rangle$

(M. Born, E. Wolf: Principles of Optics)

for short range, complex scalar potentials (absorption), reciprocity is always fulfilled (no time reversal symmetry)

time reversal ≢ reciprocity

3. Nuclear resonant scattering: $\langle \alpha | \mathrm{T} | \beta \rangle \neq \langle -\beta | \mathrm{T} | -\alpha \rangle$

absorption + polarization dependence →

In general there is no proof for the (de Hoop) reciprocity!!!

Reciprocity theorems

4. L. Deák and T. Fülöp, Ann. Phys. (N.Y.) 327, 1050 (2012).

$$\langle \mathbf{k}_\beta, p_\beta | \mathrm{T} | \mathbf{k}_\alpha, p_\alpha \rangle = \langle -\mathbf{k}_\alpha, p_\alpha^* | \mathrm{T} | -\mathbf{k}_\beta, p_\beta^* \rangle + \left(\kappa_{S\beta}^{T-}, A\kappa_\alpha^+ \right) - \left(\kappa_{S-\alpha}^{T-}, A\kappa_{-\beta}^+ \right)$$

$$\hat{H} = \hat{H}_0 + \hat{V} \quad (\hat{H}_0 \text{ is self-adjoint})$$

Reciprocity-violating terms

Reciprocity appears if $\boldsymbol{V_{ij} = V_{ji}}$ Common belief: if and only if. Is that true?

a satisfactory but not necessary condition, which is also non-covariant.

J.E. Tener, J. Math. Anal. Appl. 341 (2008) 640:

Any 2 × 2 matrix is unitarily equivalent to a complex symmetric matrix.

All 2 × 2 homogeneous potentials are reciprocal!
Can inhomogeneous potentials result in reciprocity violation?

Yes, since for $V=V(\boldsymbol{r})$ a simultaneous basis would be needed to make V complex symmetric (self-transpose) for any $\boldsymbol{r}$ and, **in general, such a basis does not exist.**

Reciprocity in NRS

What is the potential V?

$$rot\ \mathbf{H} = \frac{\varepsilon}{c}\dot{\mathbf{E}} = -\frac{i\omega\varepsilon}{c}\mathbf{E}$$

$$div\ \mathbf{E} = 0$$

$$rot\ \mathbf{E} = -\frac{\mu}{c}\dot{\mathbf{H}} = \frac{i\omega\mu}{c}\mathbf{H}$$

$$div\ \mathbf{H} = 0$$

$$rot\ rot\ \mathbf{H} = -\Delta\mathbf{H} = \frac{\omega^2\varepsilon\mu}{c^2}\mathbf{H}$$

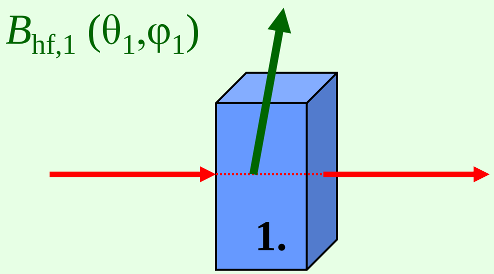
$$rot\ rot\ \mathbf{E} = -\Delta\mathbf{E} = \frac{\omega^2\varepsilon\mu}{c^2}\mathbf{E}$$

$$\Delta\Psi + k^2 n^2 \Psi = 0 \quad \text{with } k = \frac{\omega}{c} \ and\ n^2 = \mu\varepsilon$$

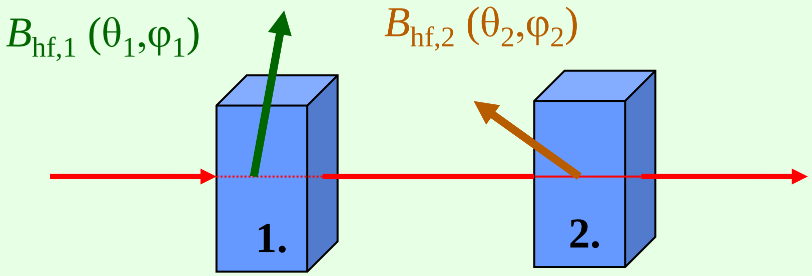
which has the form of the Schrödinger equation with

$$\Delta\Psi + k^2\Psi = -k^2\left(n^2 - 1\right)\Psi = V\,\Psi$$

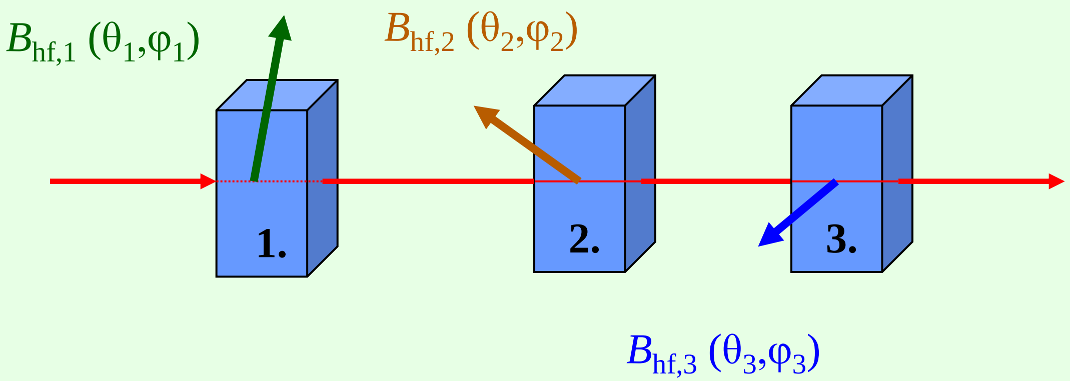
Reciprocity in NRS



- reciprocity violation may appear in phase only
- there exist, however, always such polarizations for which the system is reciprocal



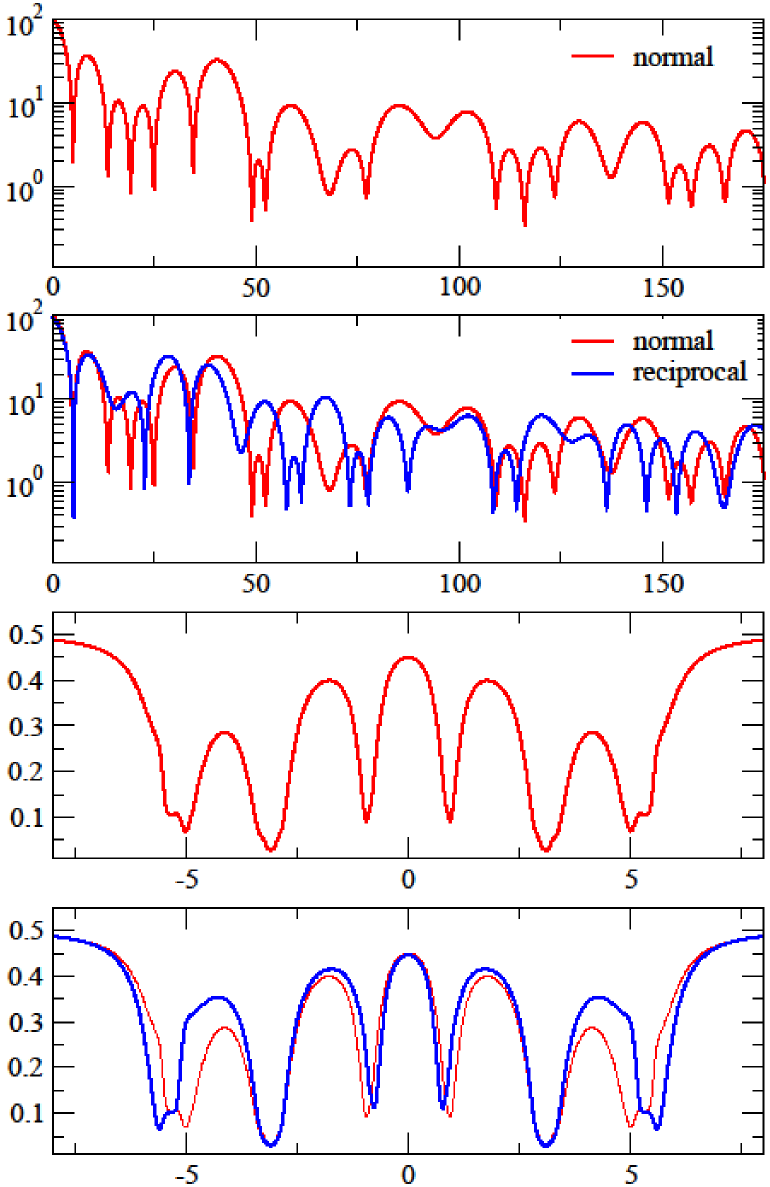
- reciprocity violation is possible both in phase and intensity
- there exists always such polarizations for which the system is reciprocal



- in general, reciprocity violation exists
- in general, no polarizations fulfil reciprocity

Reciprocity test (σ-σ scattering)

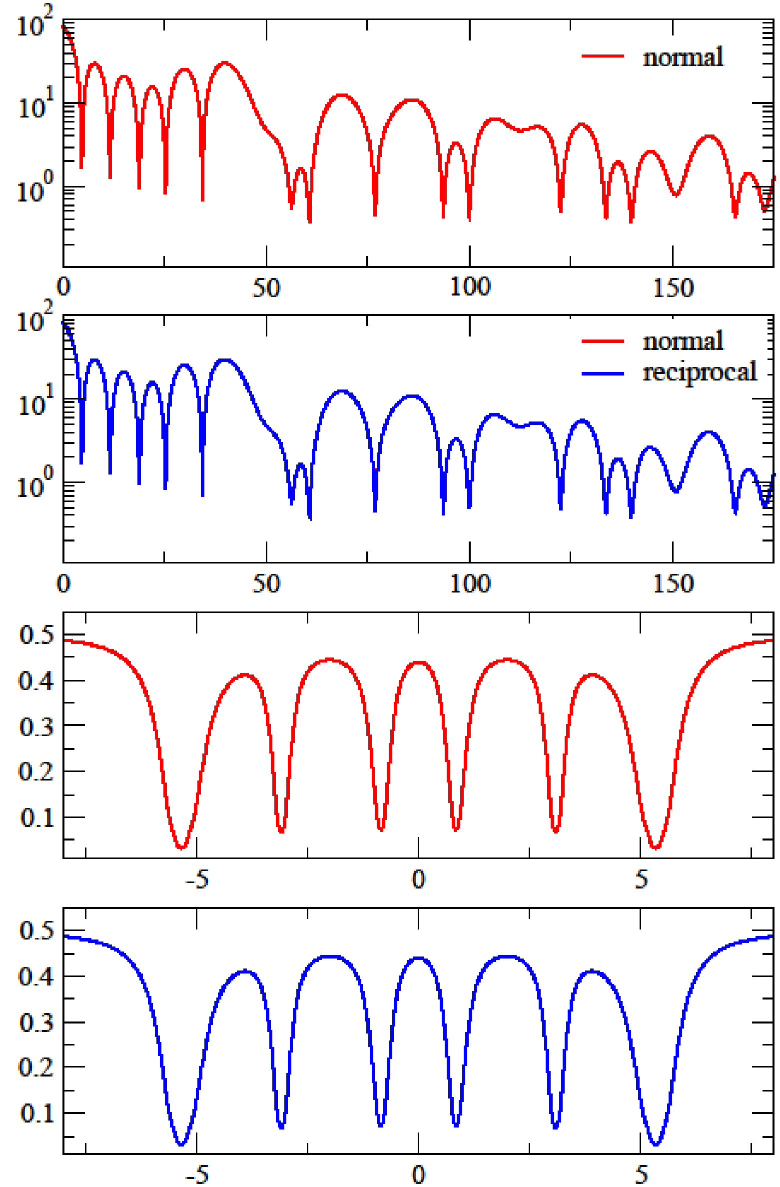
$t_{1,eff} = t_{2,eff} = 76\ (5\mu m),\ \theta_1 = 90^o,\ \phi_1 = 45^o,\ \theta_2 = 135^o,\ \phi_2 = 0^o$



Fri Sep 17 13:17:39 2010

Reciprocity test (σ-σ scattering)

$t_{1,eff} = t_{2,eff} = 76\ (5\mu m),\ \theta_1 = 90^o,\ \phi_1 = 90^o,\ \theta_2 = 135^o,\ \phi_2 = 0^o$



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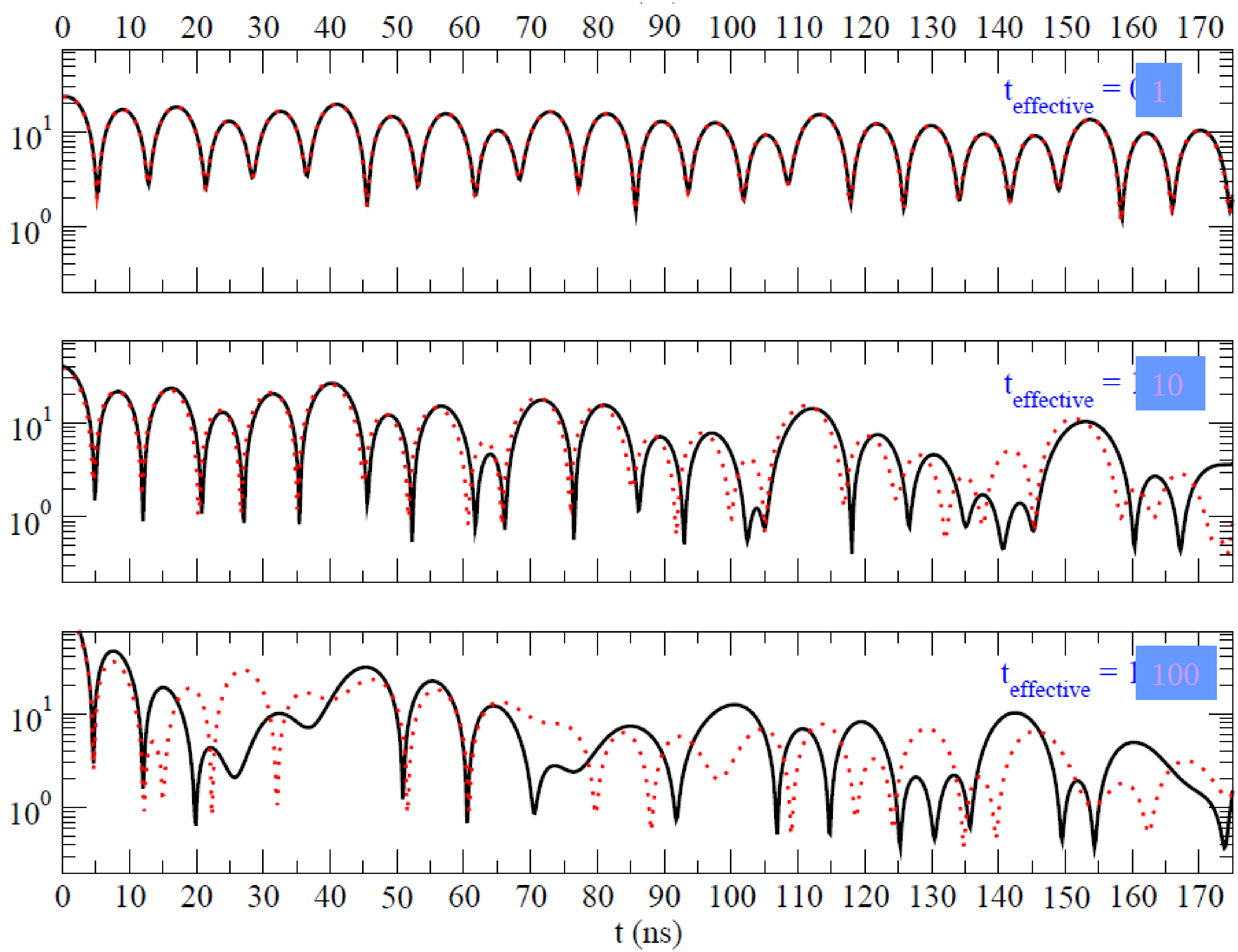
Reciprocity test ($\sigma \rightarrow \sigma$) scattering

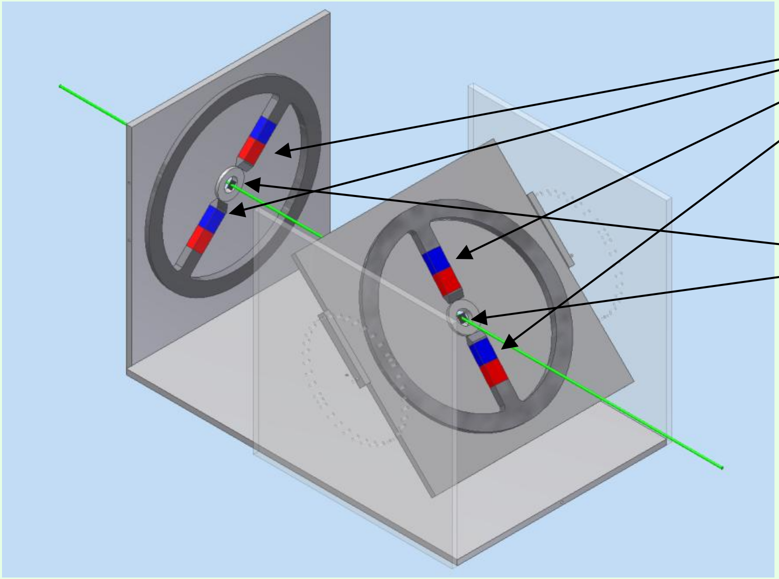
normal
reciprocal



Reciprocity violation in NRS: the experiment

Samples: two 1 cm × 1 cm rolled α-^{57}Fe foils of 6 μm thickness

Sample holder:



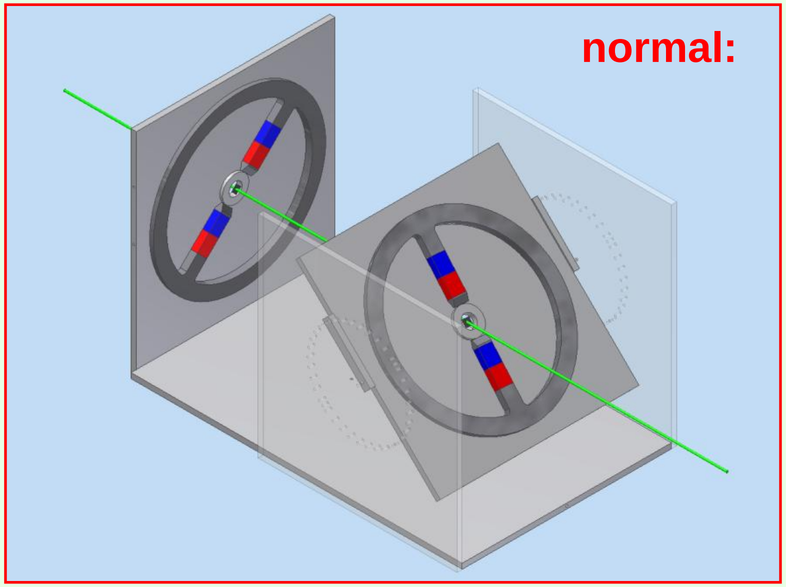
Beamline:

ID22N at the European Synchrotron Radiation Facility, Grenoble.

Experiment: SI-1794

main proposer: **L. Deák**

co-proposers: **L. Bottyán, D. L. Nagy, F. Tanczikó, Gy. Vankó, G. Kertész, H. Spiering, R. Rüffer**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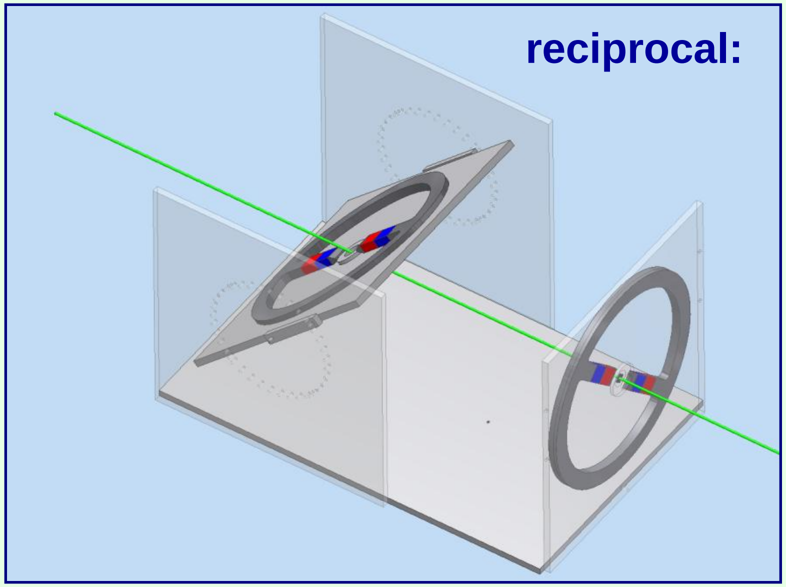
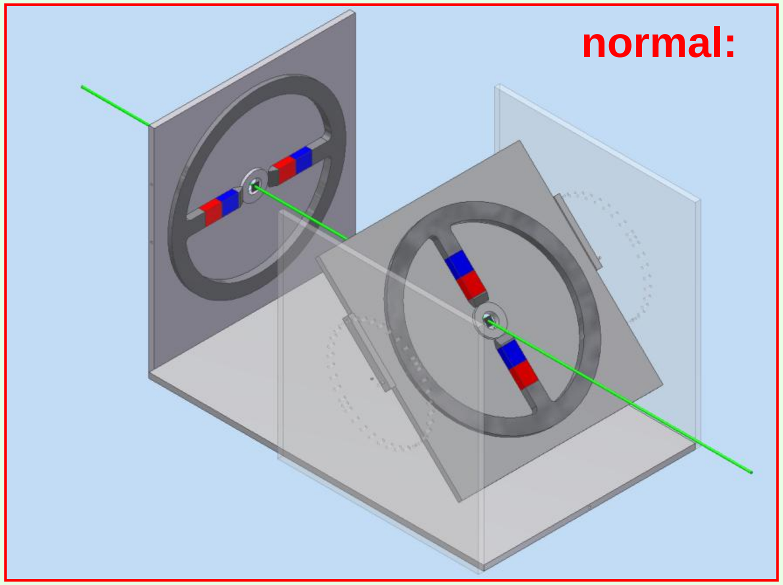
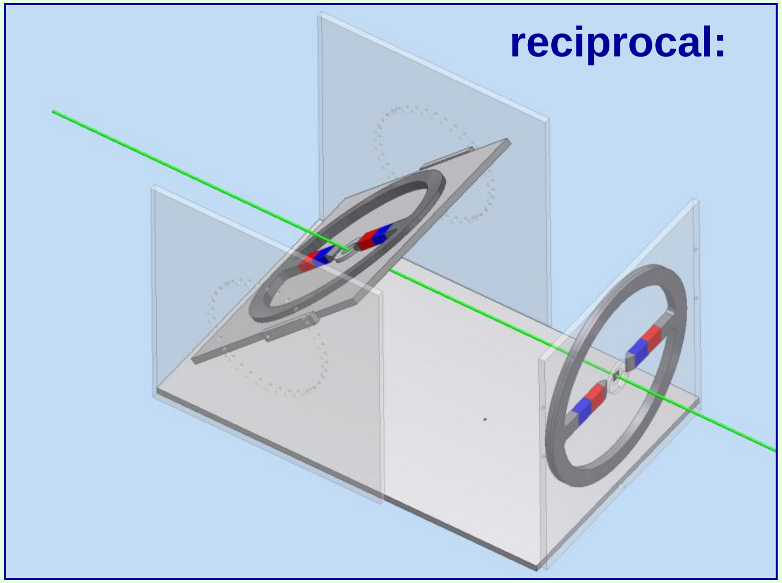
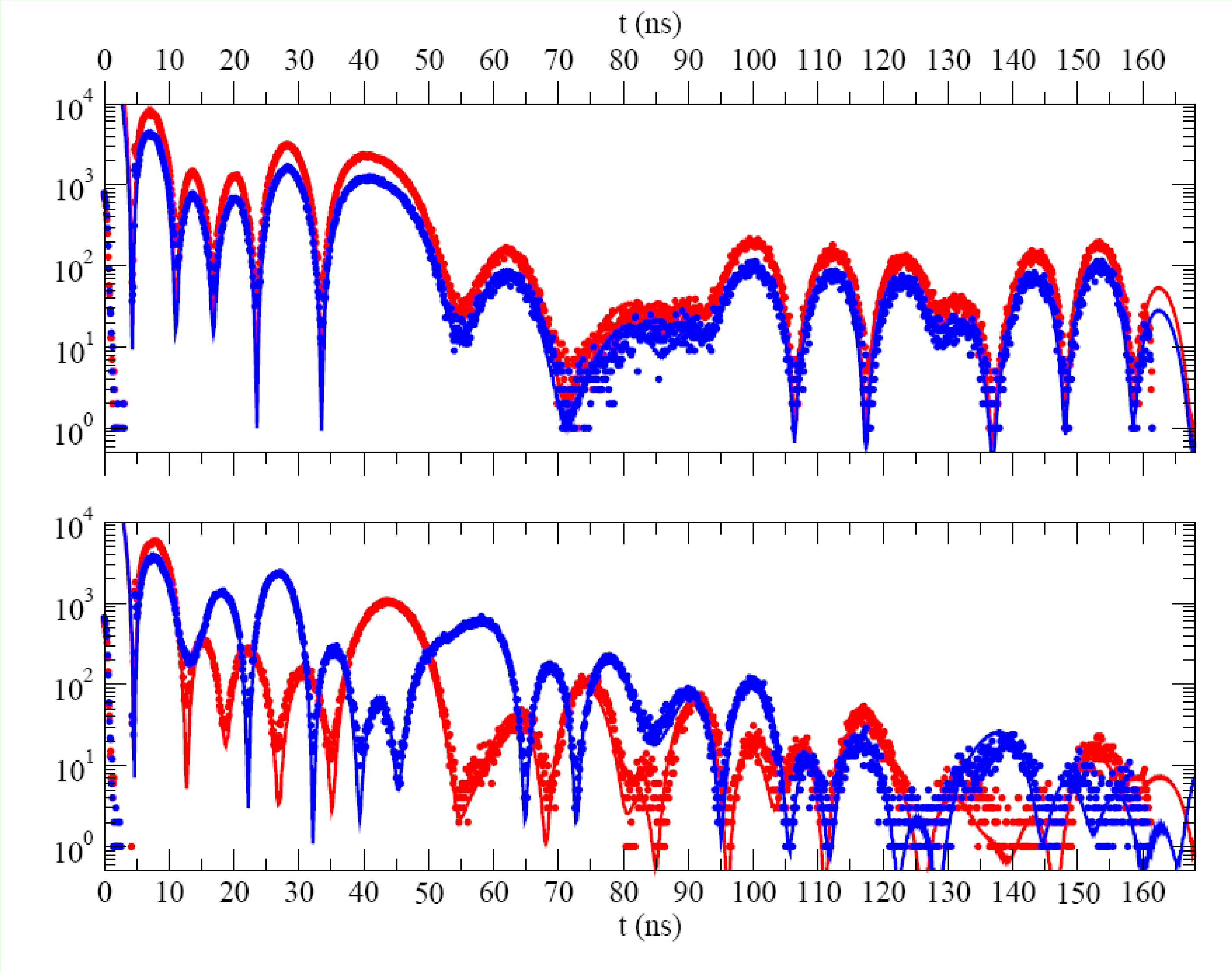
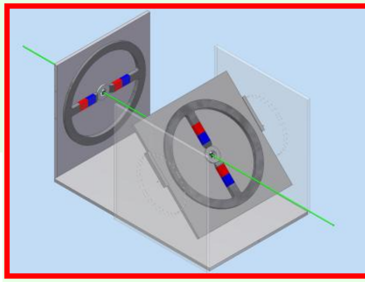
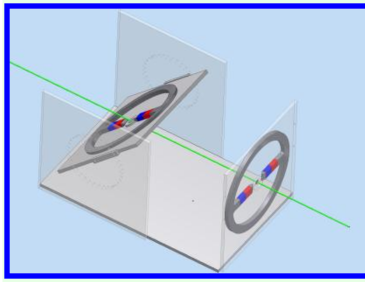
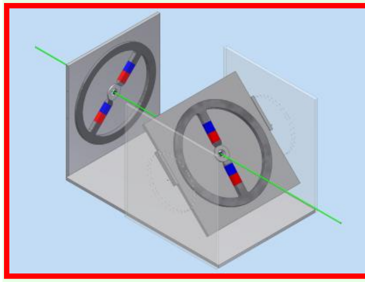
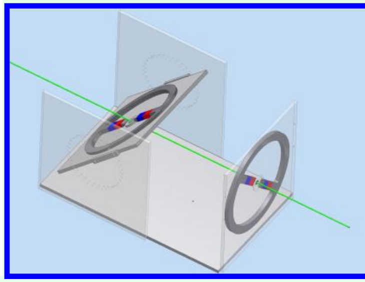

reciprocal:


normal:

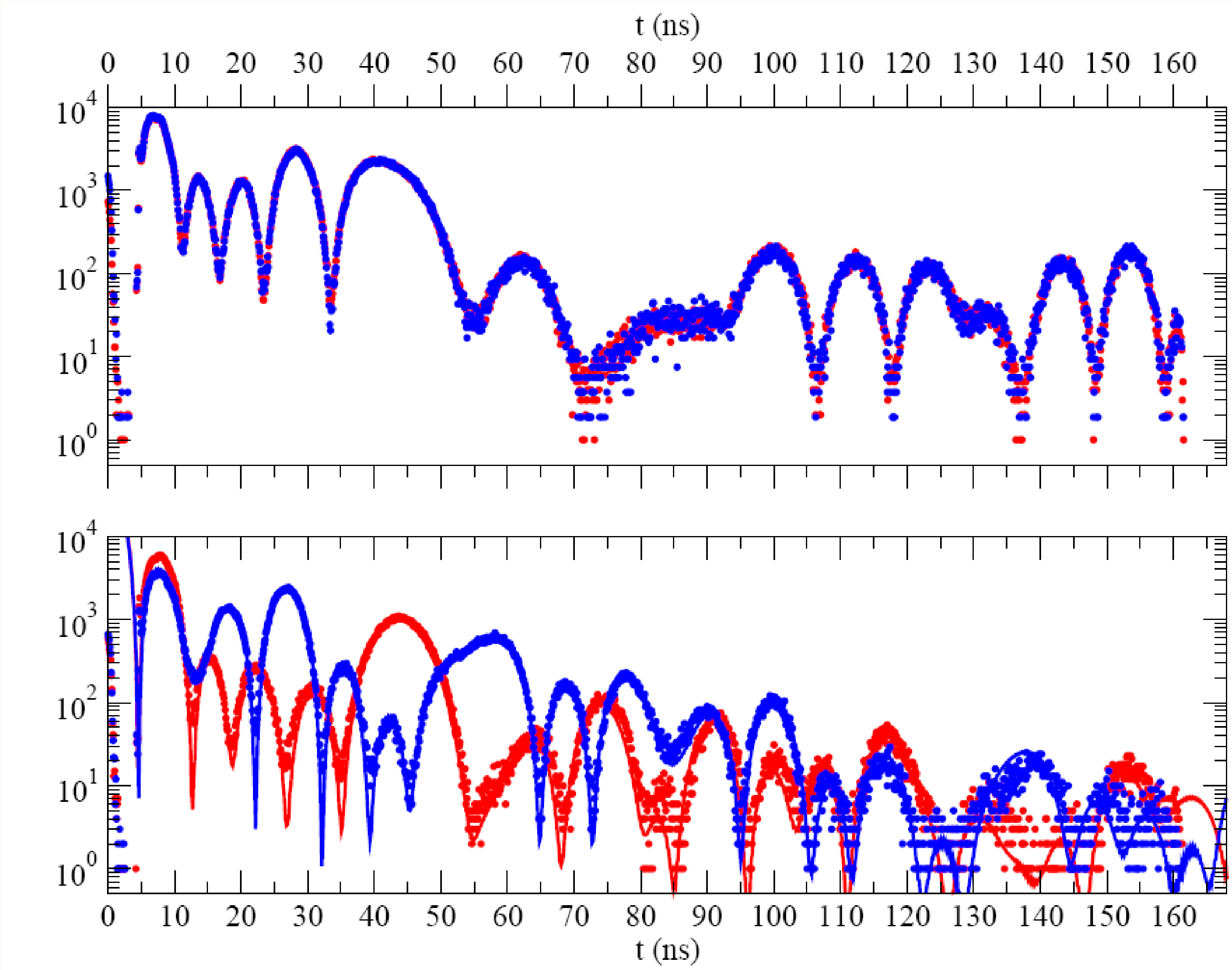

reciprocal:

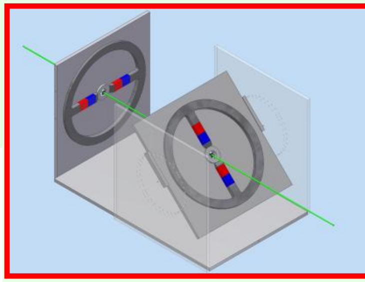
The experiment and its fit by EFFI

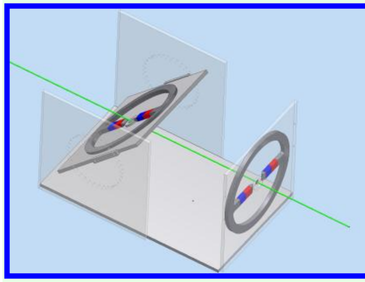


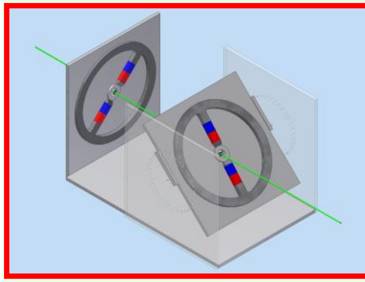





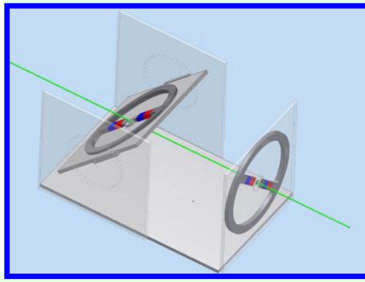
The experiment and its fit by EFFI



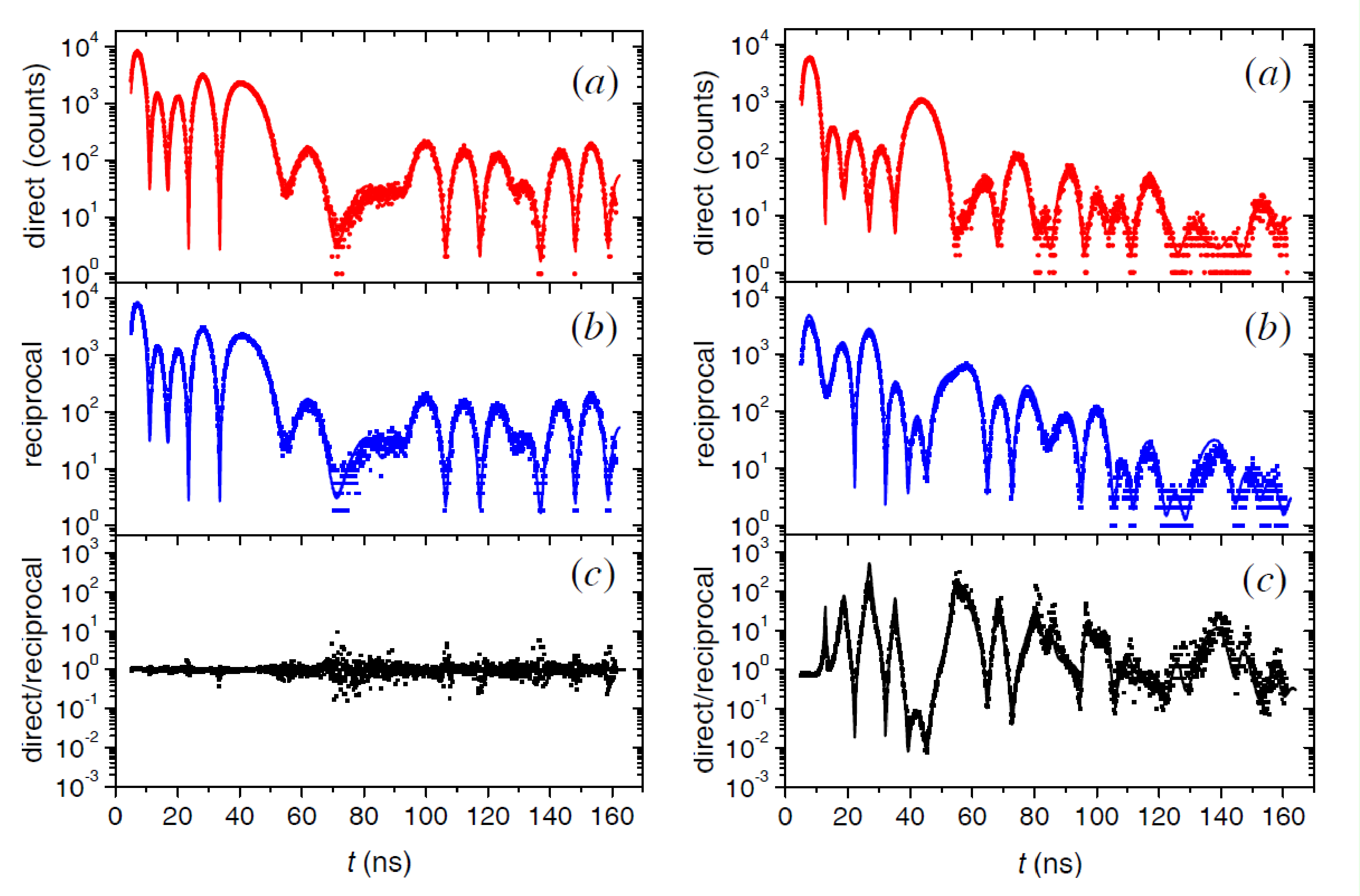








Comparison of normal and reciprocal cases



Conclusion

- The symmetry and asymmetry of the diffuse scatters (ω-scans) in reflectometry are not equivalent with fulfilling and violating reciprocity, respectively.
  The ω-scans may or may not be symmetric for both reciprocity-fulfilling and reciprocity-violating scattering.

- Simulations of a significant and switchable reciprocity violation in nuclear resonant scattering of SR were fully verified in a forward scattering experiment at the ID22N beam line of the European Synchrotron Radiation Facility, Grenoble (L. Deák et al, Phys. Rev. Lett. **109** (2012) 237402).

Dziękuję za uwagę!