Polarised neutron reflectivity

Frédéric OTT

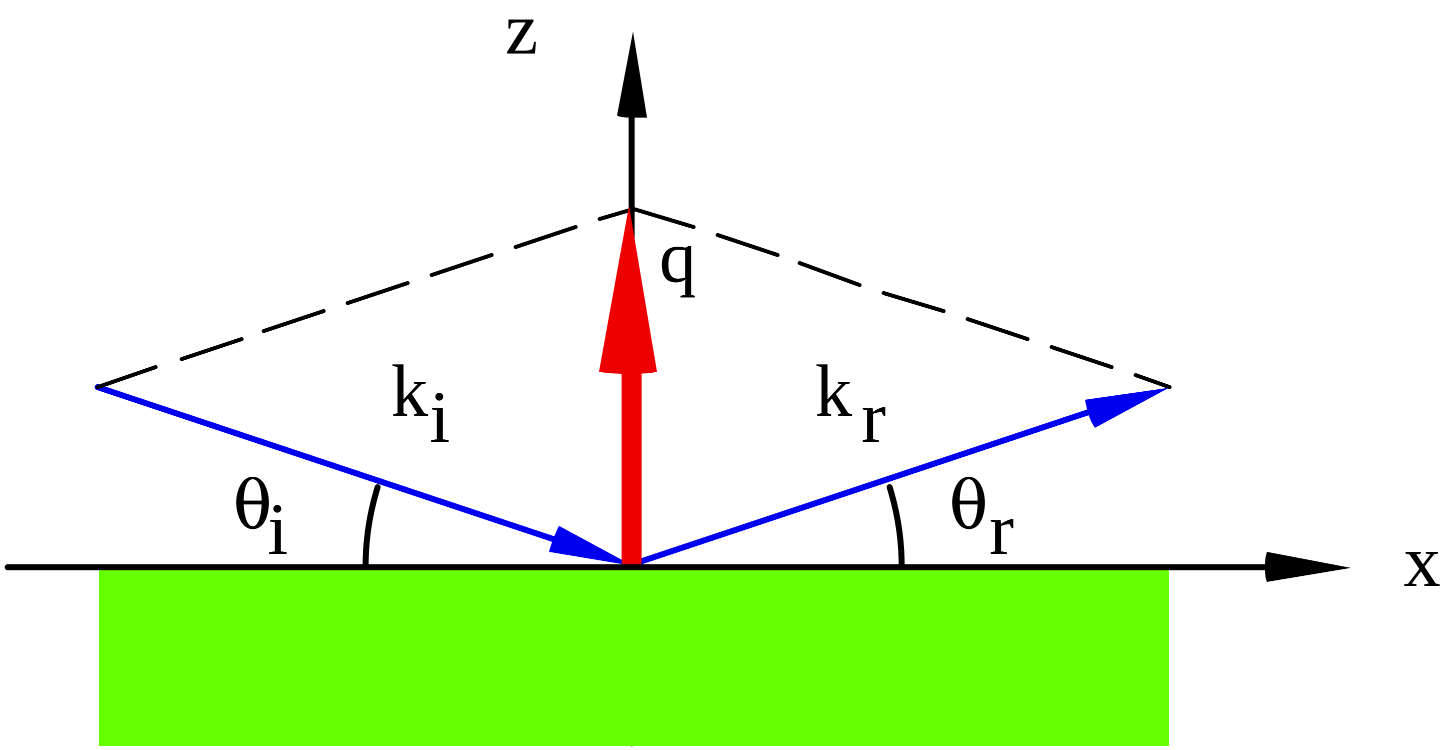
Laboratoire Léon Brillouin

fott@cea.fr

Outline

- Reflectivity
  - General principles
  - Polarised neutrons
- Instrumentation
- Examples
- Superlattices
  - Non colinear magnetism
  - GMR
  - Interface magnetism
- Resources

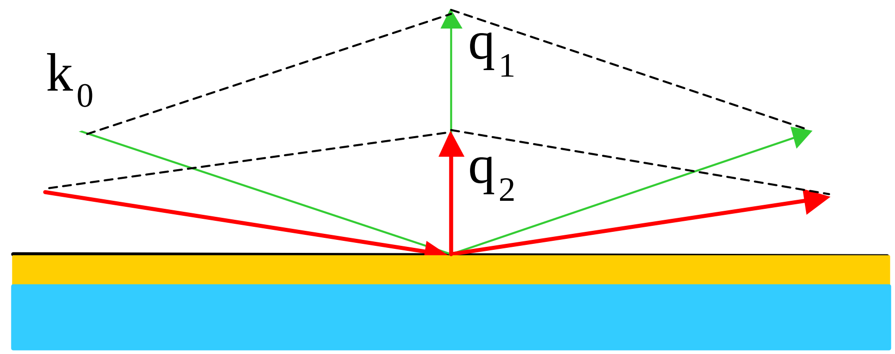
Specular reflectivity geometry



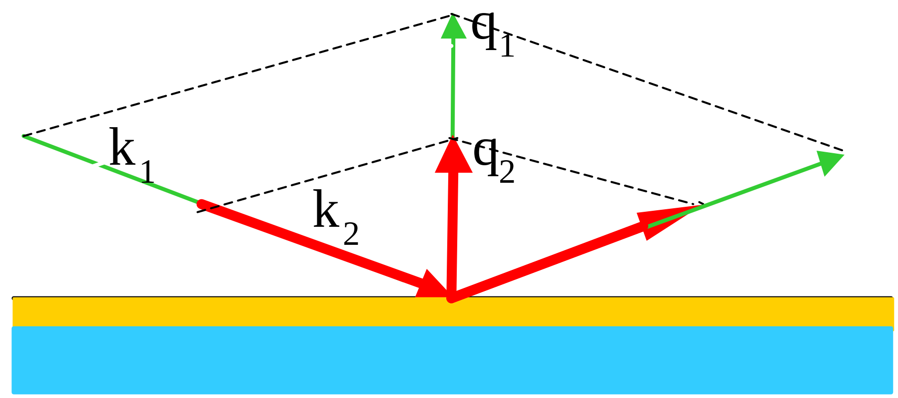
$$q = \frac{4p}{l} \sin q$$

Reflectivity measurements

- 2 ways of varying the scattering wave-vector
  - Angular scan θ – 2θ



  - Time-of-flight



Neutron-matter interaction Optical approximation.

- **Interaction neutron-nucleus**
  - Isotropic and ponctual

$$V(\mathbf{r}) = \frac{2p\hbar^2}{m} b_n d(\mathbf{r})$$

- **Zeeman interaction**
  - Neutron spin – magnetic field

$$V(\mathbf{r}) = -\vec{m} \cdot \vec{B}(\mathbf{r})$$

Neutron-nucleus

Neutron-magnetisation

Neutron-magnetic field

$$\hat{V} = \frac{2p\hbar^2}{m} rb - m_0 g_n m_n \mathrm{s} . \mathrm{M}_{//} - g_n m_n \mathrm{s} . \mathrm{B}_0$$

Limitation : planar magnetisation $M_{//}$

- In the Born approximation :
  - It can be shown that the magnetic interaction is sensitive only to the component of the magnetisation perpendicular to the scattering wave-vector

$$q.M_{\perp}$$

- Other approach
  - The neutron spin interacts with B : $B = \mu_0 (H + M - DM)$
  - For continuous thin films :

$$D = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

$$(1-D)\vec{M} = \vec{M} - \vec{M}_z = \vec{M}_{//}$$

$$B = \mu_0 (H_0 + M_{//})$$

Derivation of the reflectivity

Eigenstates

$$|+\rangle \quad et \quad |-\rangle$$

Interaction

$$V = \frac{2p\hbar^2}{m} r b - g_n m_n \vec{s}.(\vec{B}_0 + m_0 \vec{M}_{//})$$

Propagation eq. Schrödinger

$$\frac{-\hbar^2}{2m}\Delta j + V j = E j$$

Helmoltz eq. $\Delta \mathbf{U} + k^2 \mathbf{U} = 0$

$$k^2 = \frac{2m}{\hbar^2}(E - V)$$

Continuity conditions

$$y \quad et \quad \nabla y$$

Matrix formalism

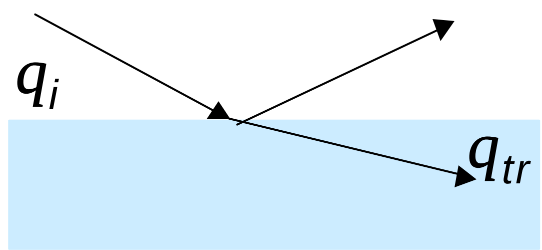
Optical index

$$\frac{d^2y}{dr^2} + k^2 y = 0 \qquad k^2 = \frac{2m}{\hbar^2}(E - V)$$

- The optical index is defined as $n^2 = \frac{k^2}{k_0^2}$

$$n^2 = 1 - \frac{V}{E} = 1 - \frac{l^2}{p} rb \qquad n \approx 1 - \frac{l^2}{2p} rb$$

- Snell's law : $\cos q_i = n \cos q_{tr}$ $\qquad n = \cos q_c$

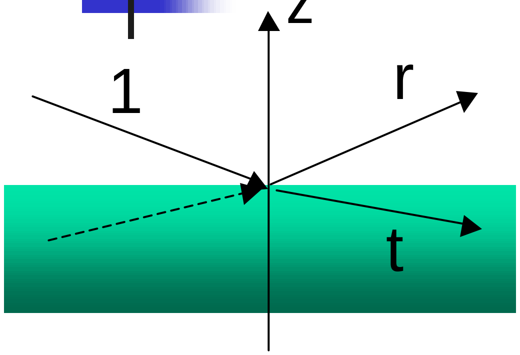


$$q_c = \sqrt{\frac{rb}{p}}\, l$$

Some values

Matérial	b_n (fm)	r (10^{28} m^{-3})	rb (10^{13} m^{-2})	d (10^{-6})	q_c (nm^{-1})	Remarks
H	-3.73					hydrogen
D (^{2}H)	6.67					deuterium
C	6.64	11.3	75	19.1	0.19	graphite
C	6.64	17.6	117	29.8	0.24	diamond
O	5.80					
Si	4.15	5.00	20.8	5.28	0.10	
Ti	-3.44	5.66	-19.5	-5.0	-	
Fe	9.45	8.50	80.3	20.45	0.20	
Co	2.50	8.97	32.6	8.29	0.13	
Ni	10.3	9.14	94.1	24.0	0.22	
Cu	7.72	8.45	65.2	16.6	0.18	
Ag	5.92	5.85	34.6	8.82	0.13	
Au	7.63	5.90	45	11.5	0.15	
H_2O	-1.68	3.35	-5.63	-1.43	-	
D_2O	19.1	3.34	63.8	16.2	0.18	
SiO_2	15.8	2.51	39.7	10.1	0.14	
GaAs	13.9	2.21	30.7	7.82	0.12	
Al_2O_3	24.3	2.34	56.9	14.5	0.17	saphir
pyrex			42	10.7	0.14	
C_8H_8	23.2	0.61	14.2	3.6	0.084	polystyrène
C_8D_8	106.5	0.61	65	16.5	0.18	

Reflection on a substrate



Vacuum « 0 »

$$\psi_0(z) = A_0 \exp(iq_0 z) + B_0 \exp(-iq_0 z)$$

Z = 0

Substrate « s »

$$\psi_s(z) = A_s \exp(iq_s z) + B_s \exp(-iq_s z)$$

$$\psi_0(z) = 1.\exp(iq_0 z) + r.\exp(-iq_0 z)$$
$$\psi_s(z) = t.\exp(iq_s z)$$

Continuity conditions

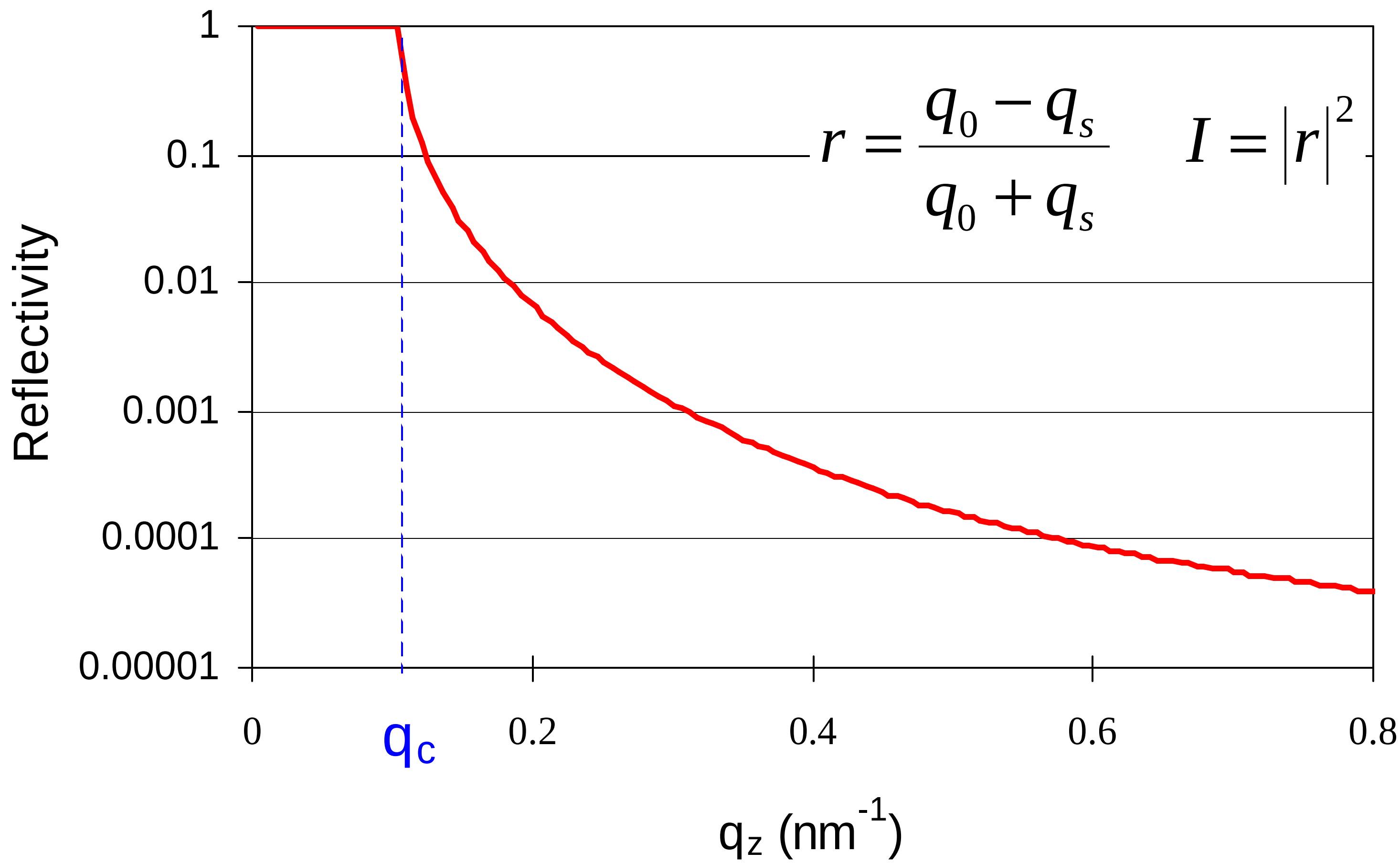
$$\begin{cases} \psi_0(z=0) = \psi_s(z=0) \\ \psi_0'(z=0) = \psi_s'(z=0) \end{cases} \qquad \begin{cases} 1 + r = t \\ q_0(1-r) = q_s t \end{cases}$$

$$r = \frac{q_0 - q_s}{q_0 + q_s} \quad and \quad t = \frac{2q_0}{q_0 + q_s}$$

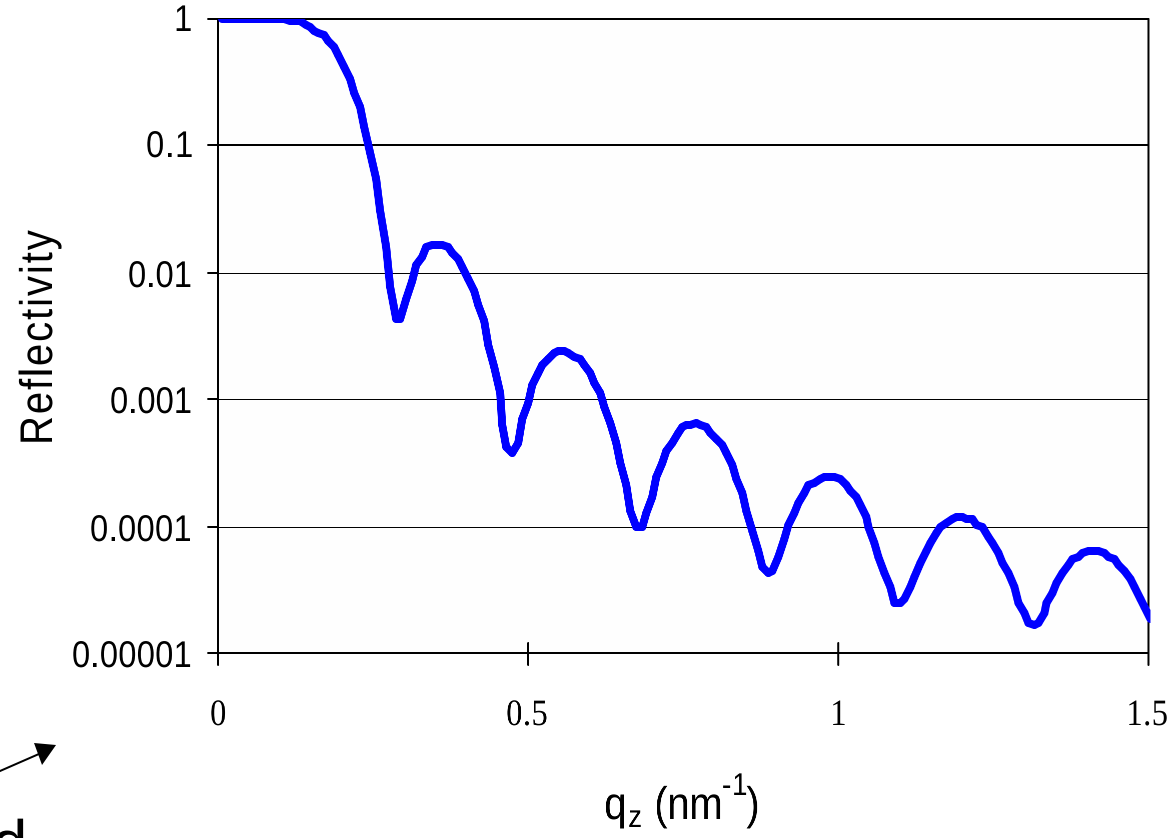
$$R = |r|^2 \quad et \quad T = |t|^2$$

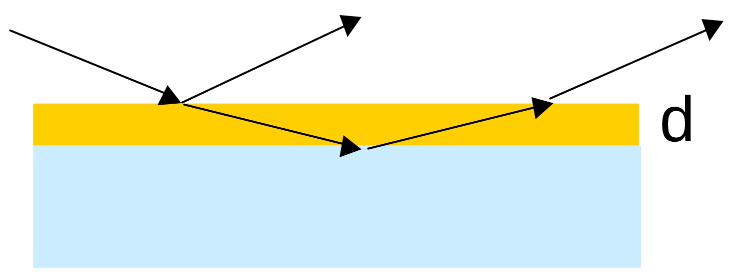
$$R = \left| \frac{q_0 - q_s}{q_0 + q_s} \right|^2$$

Case of a non magnetic substrate



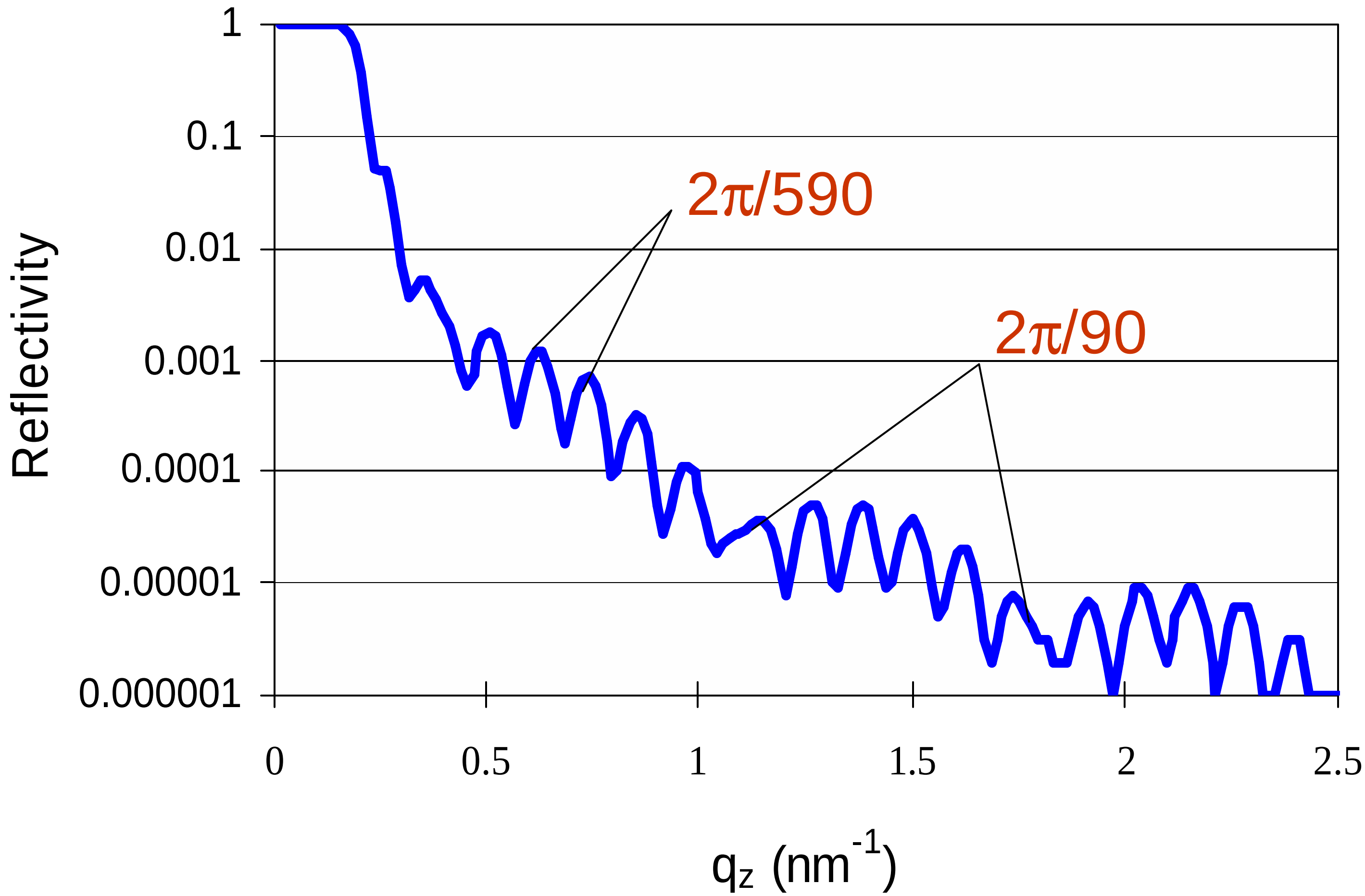
Reflection on a thin film deposited on a substrate (non magnetic case)





$$d = \frac{2p}{\Delta q}$$

Example Cu(500 Å)/Cr(90 Å) on silicon

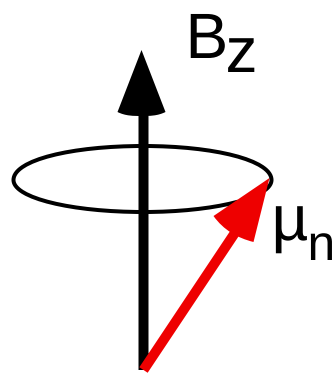


The neutron

- spin 1/2 particle ($\rightarrow$ associated magnetic moment μ_n)

$$|+\rangle \textit{ and } |-\rangle \textit{ are the eigenstate s}$$

- Interacts with the magnetic fields **B** (aligned along z):
  - *Neutron in an eigenstate* ($|+\rangle$ *ou* $|-\rangle$) : stays in this state
  - *Quantified neutron along (Ox)* ($\frac{1}{\sqrt{2}}(|+\rangle + |-\rangle)$) *:* precession around B_z



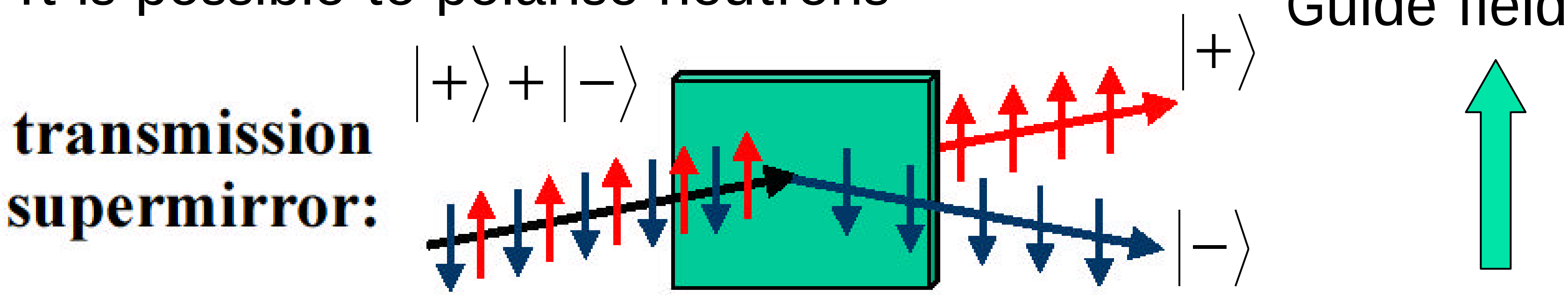
Comparison nuclear and magnetic neutron scattering lengths.

$$\hat{V} = \frac{2\pi\hbar^2}{m}\rho\left(b_N + b_M\right) \qquad b_M = \frac{m}{2\pi\hbar^2}\frac{g_n \mu_n \mu_0 M_{//}}{\rho}$$

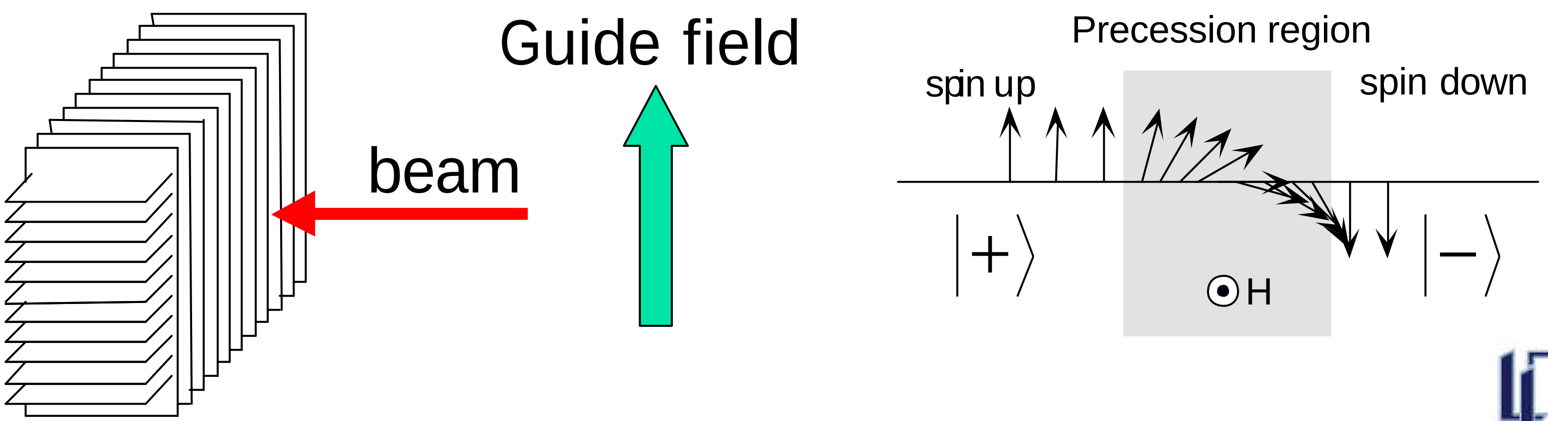
éléments	b_n (fm)	b_M (fm)	σ_a (barn)
Fe	9.45	5.4	2.56
Co	2.49	4.5	37.2
Ni	10.3	1.6	4.49
Gd	6.5 - i 13.82	18.83	49700
Si	4.15	-	0.17
Ti	-3.44	-	6.1

Polarised neutron reflectivity

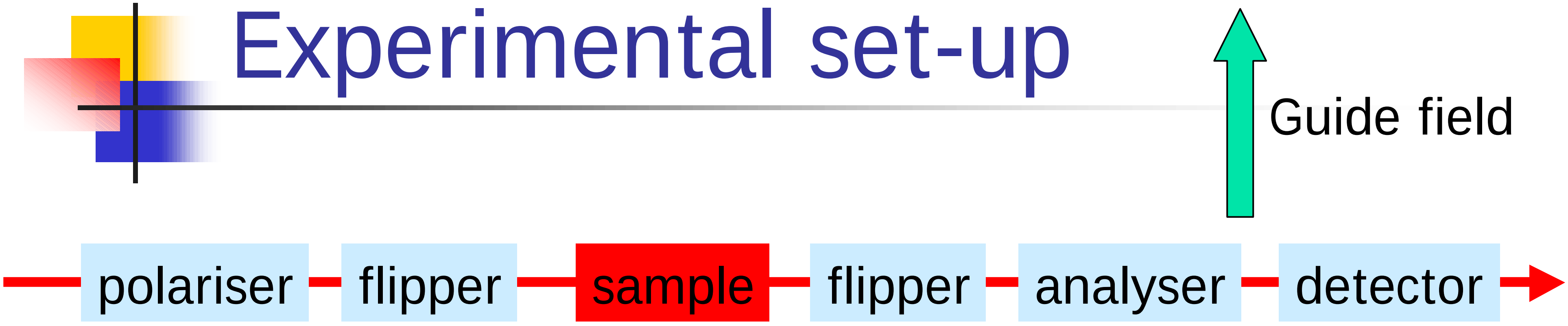
- It is possible to polarise neutrons



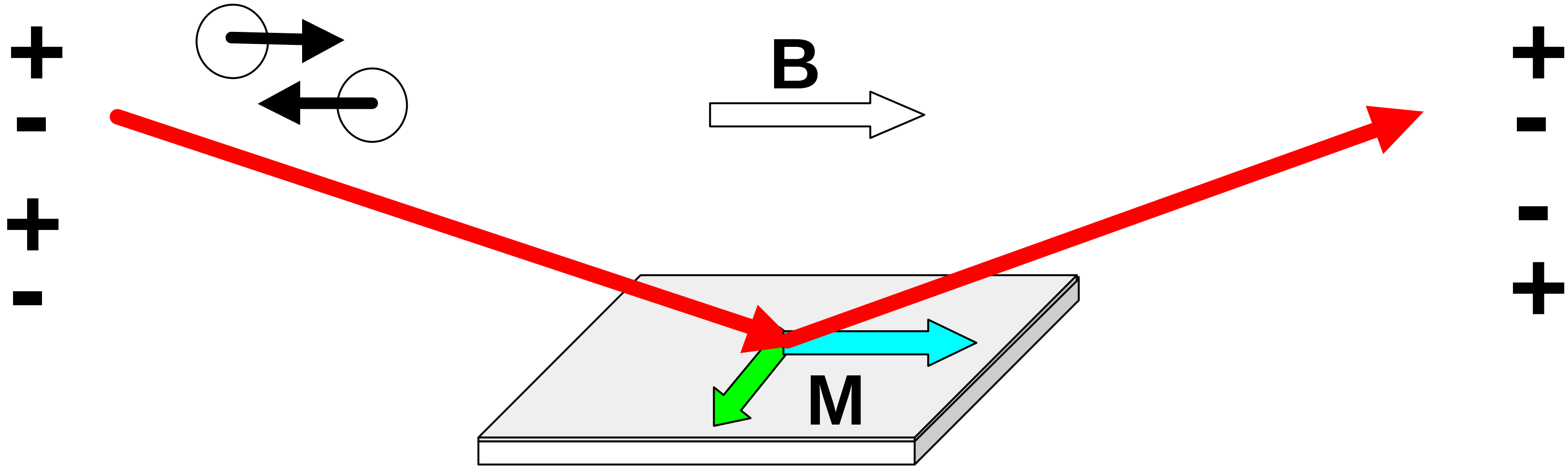
- Manipulate the polarisation : neutron « flipper »



Experimental set-up



- 4 cross-sections $R^{++}, R^{--}, R^{+-}, R^{-+}$

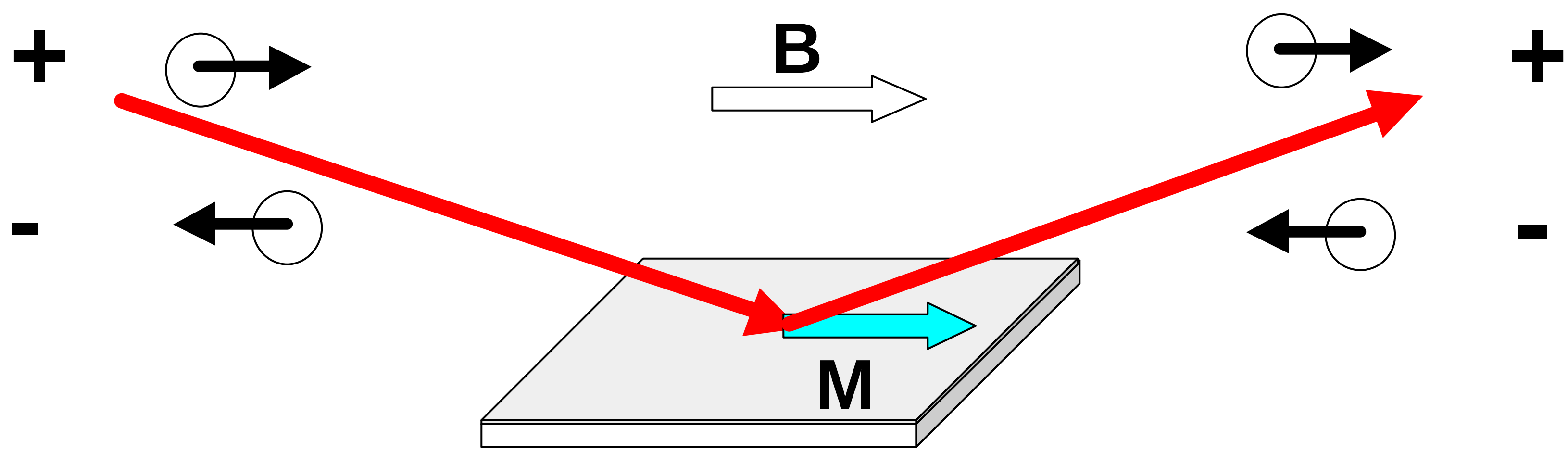


M // B

- Sample transfert matrix $\begin{pmatrix} R^+ & 0 \\ 0 & R^- \end{pmatrix}$

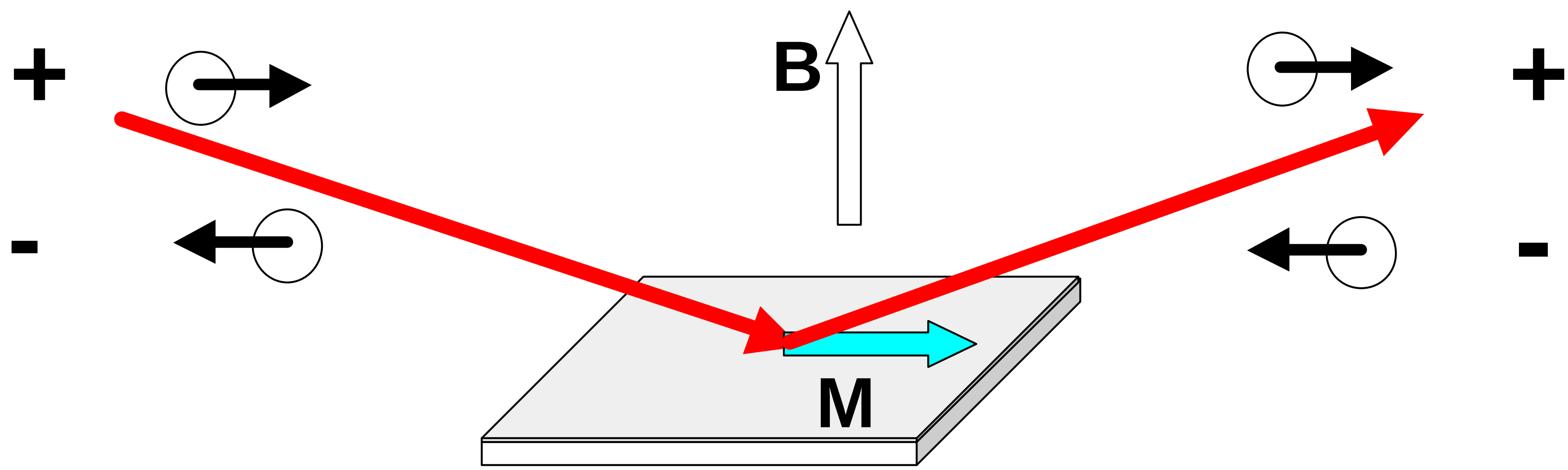
$$\begin{bmatrix} R^+ \\ 0 \end{bmatrix} = \begin{pmatrix} R^+ & 0 \\ 0 & R^- \end{pmatrix} \begin{bmatrix} 1 \\ 0 \end{bmatrix} \quad \begin{matrix} |+\rangle \\ |-\rangle \end{matrix}$$

$$\begin{bmatrix} 0 \\ R^+ \end{bmatrix} = \begin{pmatrix} R^+ & 0 \\ 0 & R^- \end{pmatrix} \begin{bmatrix} 0 \\ 1 \end{bmatrix} \quad \begin{matrix} |+\rangle \\ |-\rangle \end{matrix}$$



B perpendicular to the layer

- No magnetic contrast

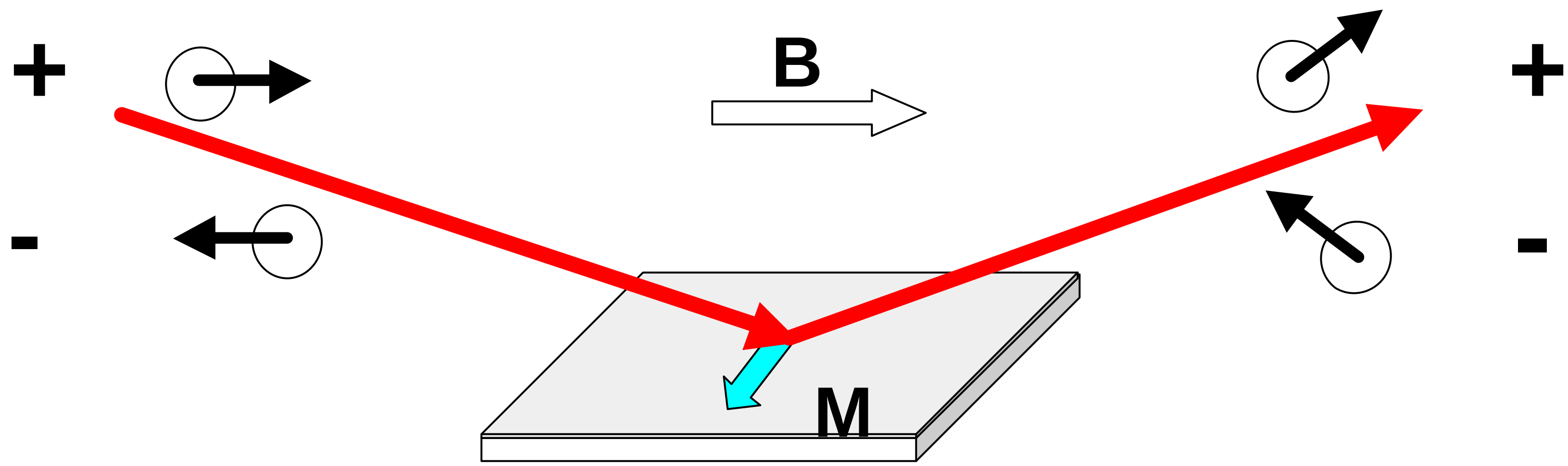


M makes an angle with B

- Sample transfert matrix $\begin{pmatrix} R^{++} & R^{+-} \\ R^{-+} & R^{--} \end{pmatrix}$

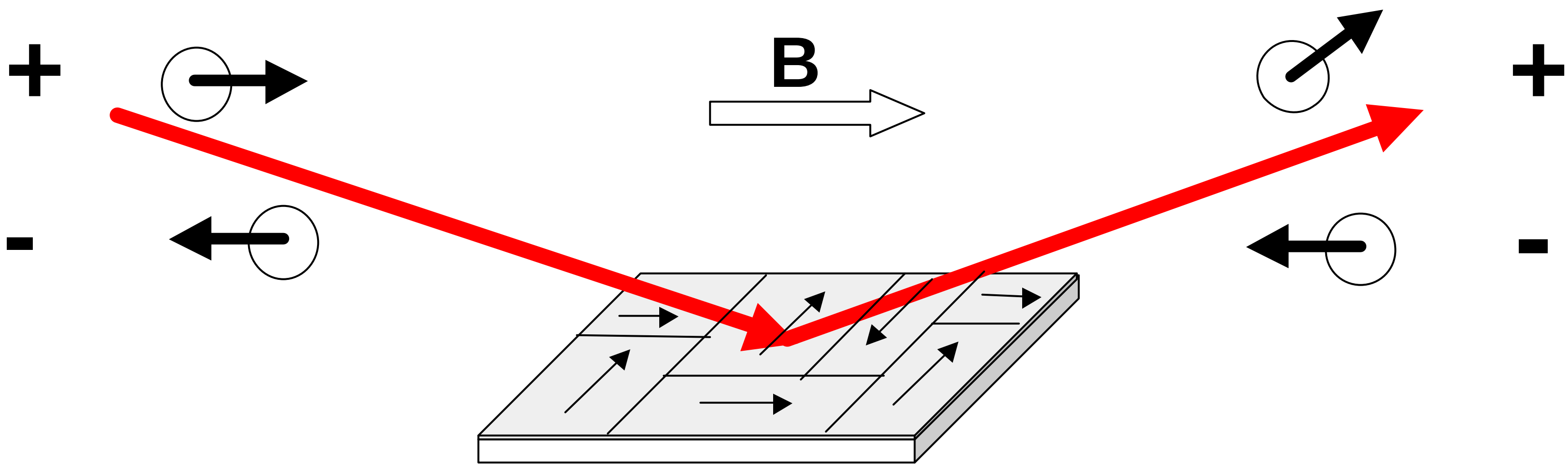
$$\begin{bmatrix} R^{++} \\ R^{+-} \end{bmatrix} = \begin{pmatrix} R^{++} & R^{+-} \\ R^{-+} & R^{--} \end{pmatrix} \begin{bmatrix} 1 \\ 0 \end{bmatrix} \quad \begin{matrix} |+\rangle \\ |-\rangle \end{matrix}$$

$$\begin{bmatrix} R^{-+} \\ R^{--} \end{bmatrix} = \begin{pmatrix} R^{++} & R^{+-} \\ R^{-+} & R^{--} \end{pmatrix} \begin{bmatrix} 0 \\ 1 \end{bmatrix} \quad \begin{matrix} |+\rangle \\ |-\rangle \end{matrix}$$



Magnetic domains

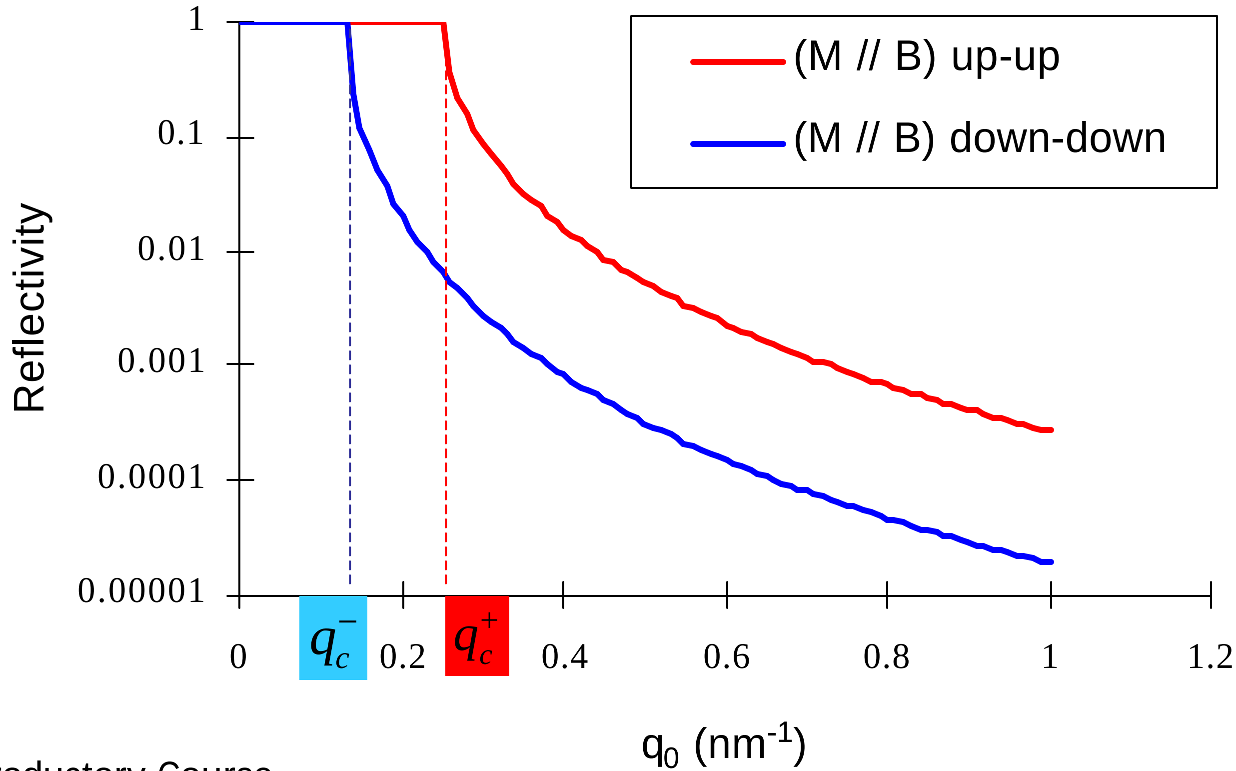
- Neutron coherent illumination ξ_N vs magnetic domains sizes ξ_M
  - If $\xi_N < \xi_M$ then $(R^+ + R^-)$
  - If $\xi_N > \xi_M$ then no magnetic contrast



Field B parallel to the magnetisation

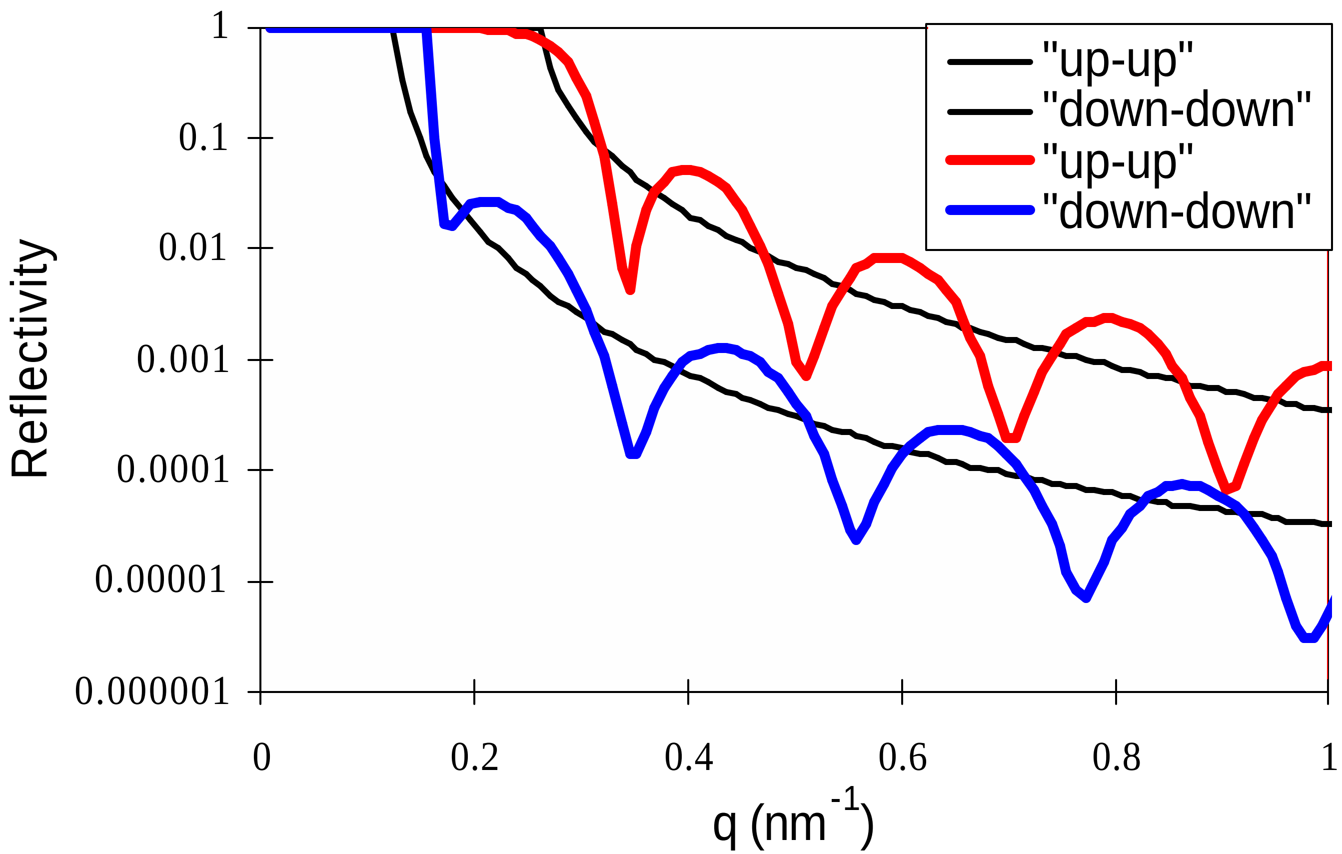
$$r^{++} = \frac{q_0 - q_s^+}{q_0 + q_s^+}$$

$$r^{--} = \frac{q_0 - q_s^-}{q_0 + q_s^-}$$



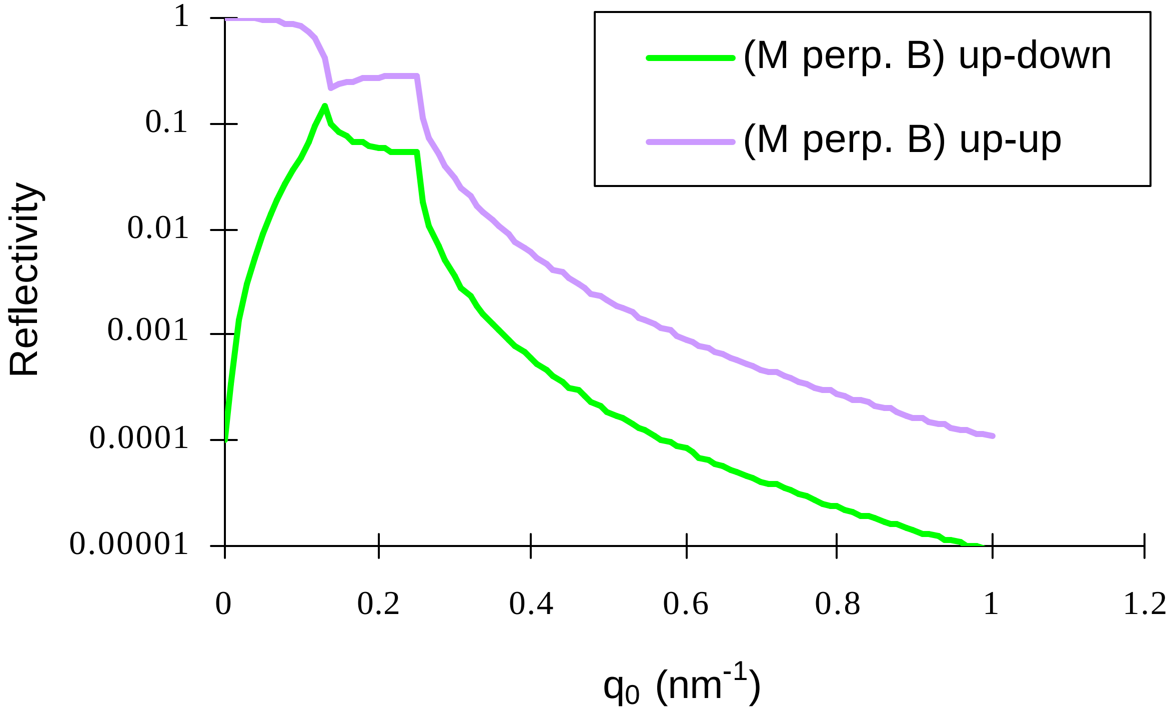
Fe thin film (30 nm) on a saphire substrate

- M // B



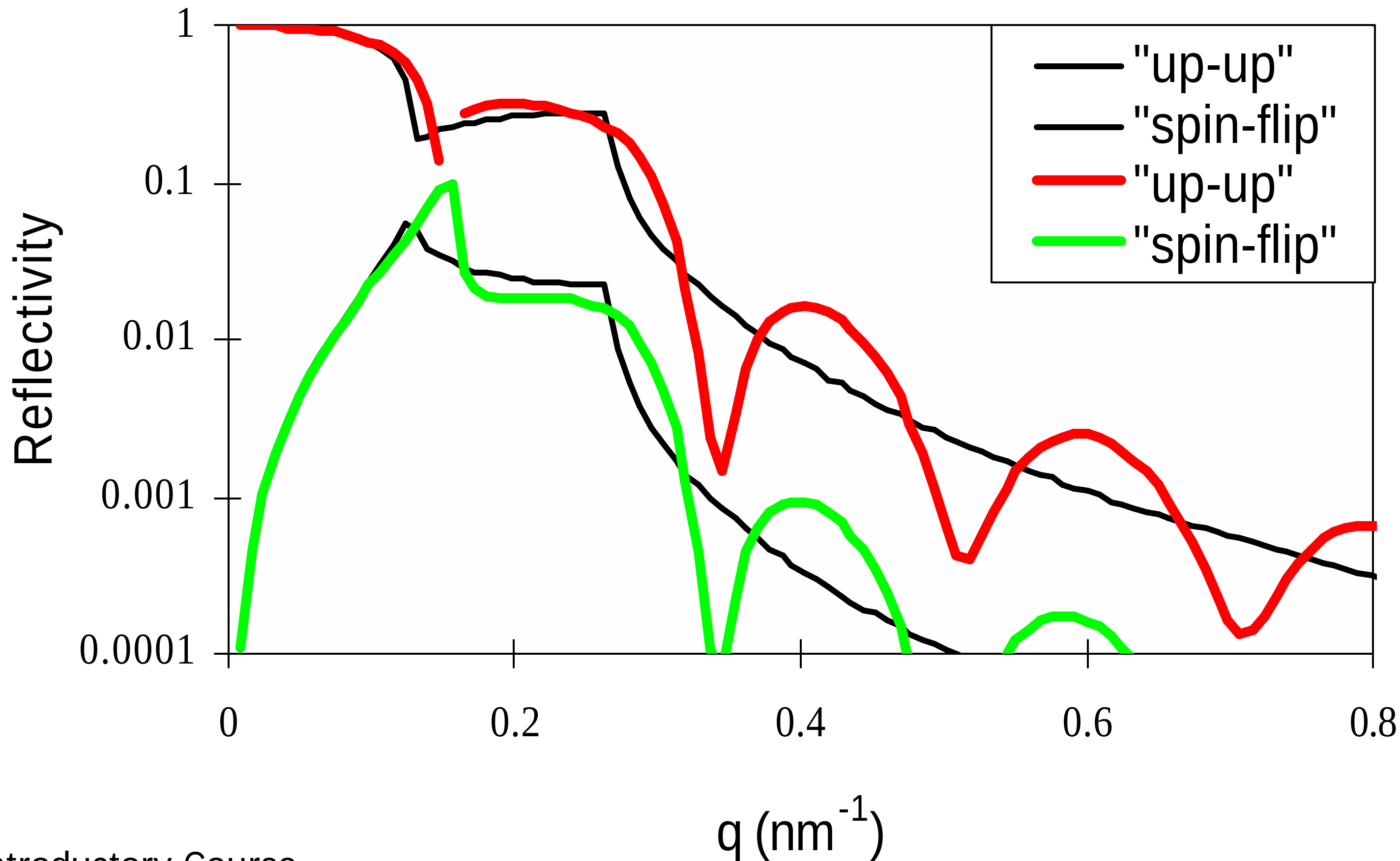
Spin-flip signal (M perp. B)

$$I^{+-} = I^{-+} = \left|r^{+-}\right|^2 = \frac{1}{4}\left|\frac{q_0\left(q_s^+ - q_s^-\right)}{\left(q_0 + q_s^+\right)\left(q_0 + q_s^-\right)}\right|^2$$

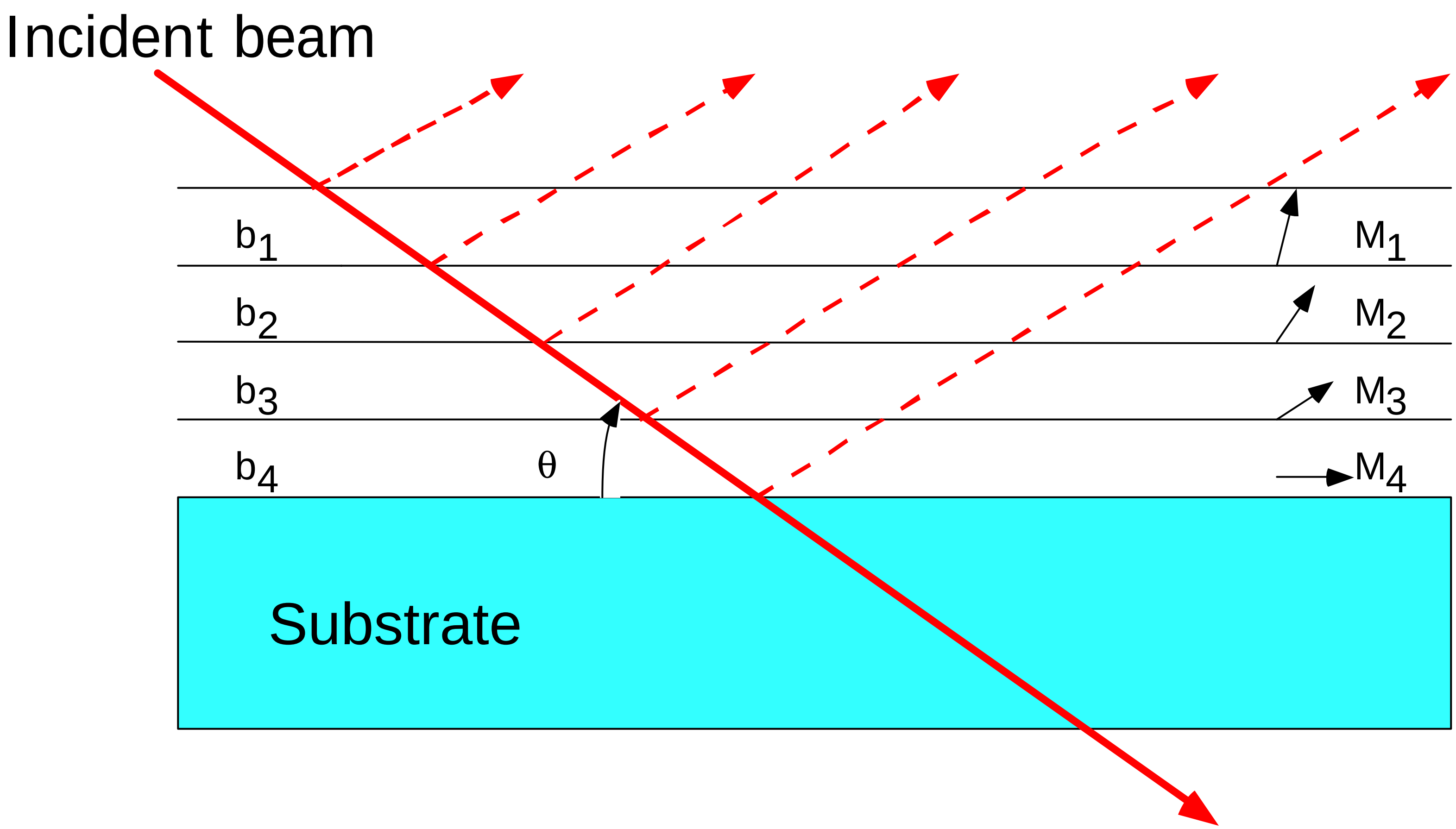


Fe thin film (30 nm) on a saphire substrate

- M perpendicular to B

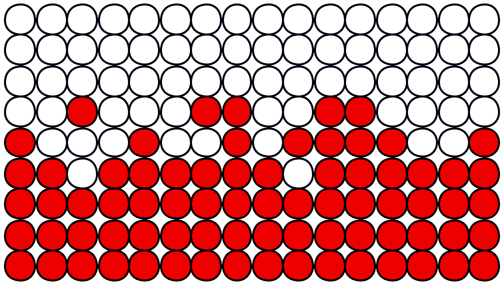


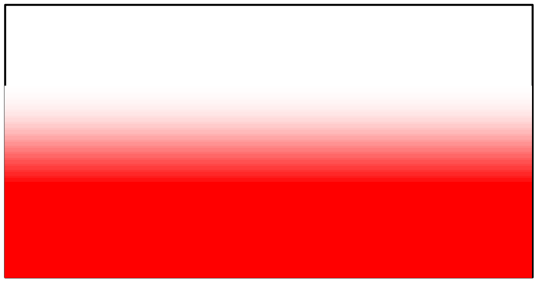
Reflectivity geometry



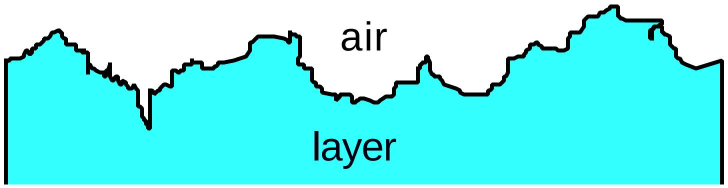
Roughness effects

- ***Roughness at the atomic level :*** interdiffusion between the thin films, ξ < 100 nm.

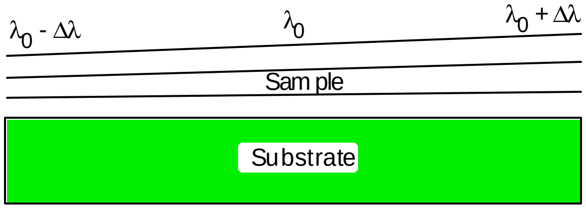




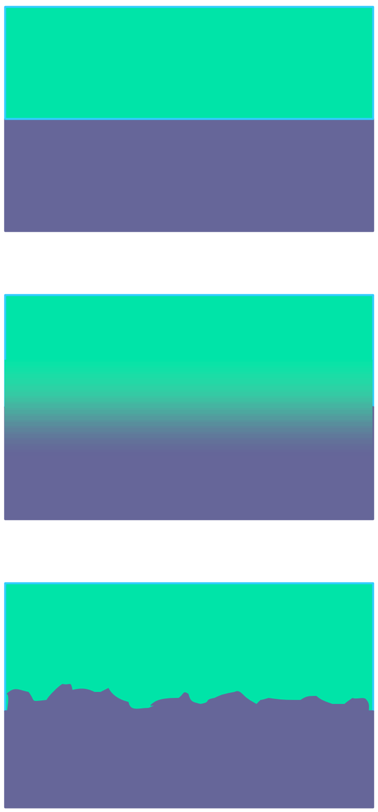
- ***Intermediate roughness*** (ξ de 0.1 µm à 50 µm).

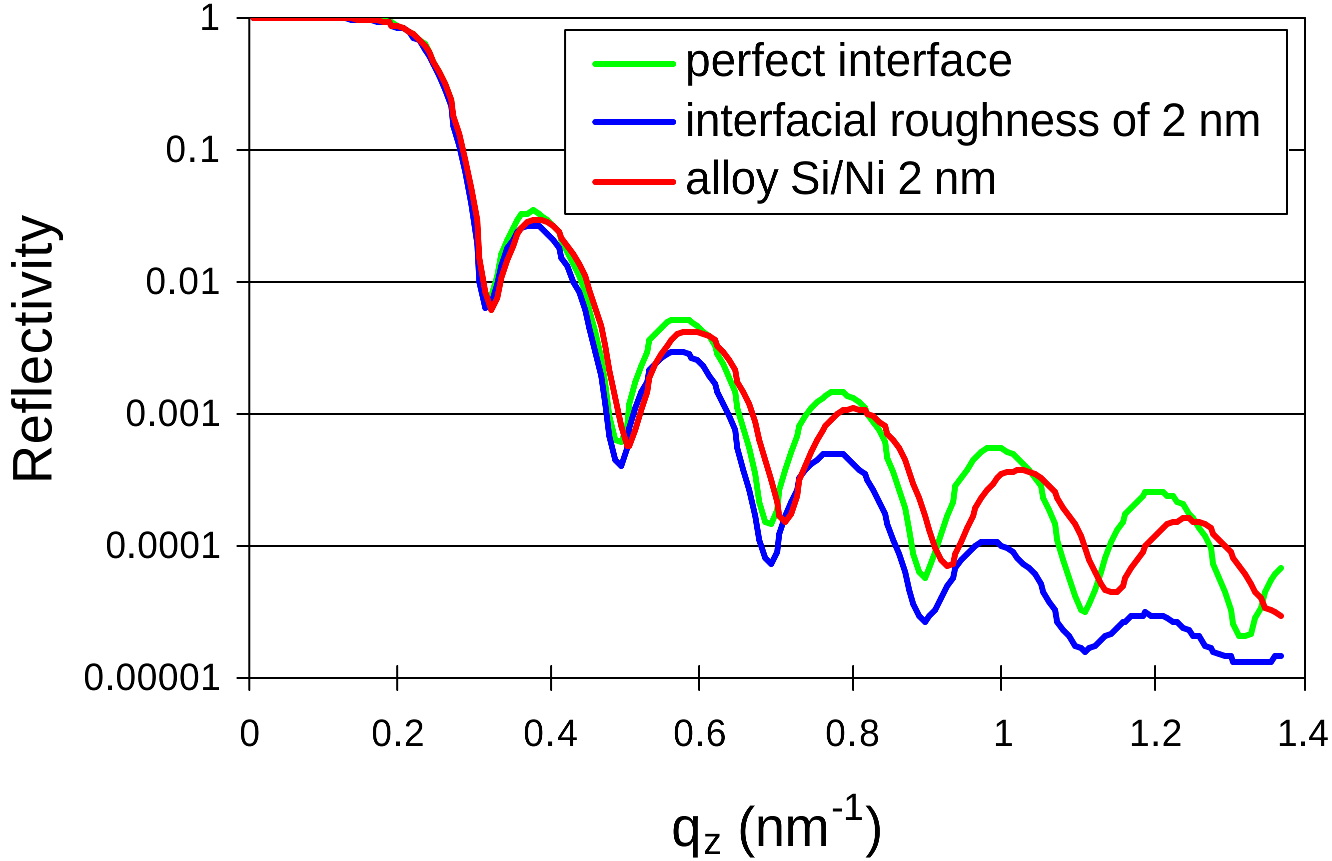


- ***A large scale roughness*** (ξ > 50 µm).



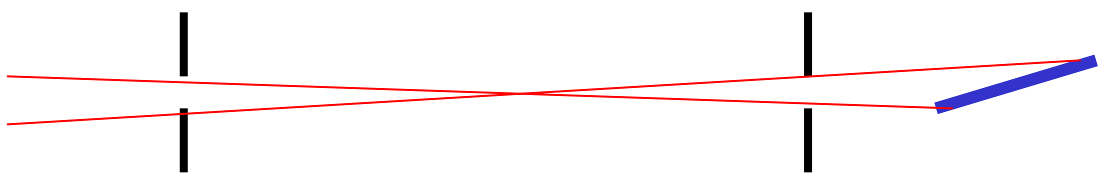
Roughness effects



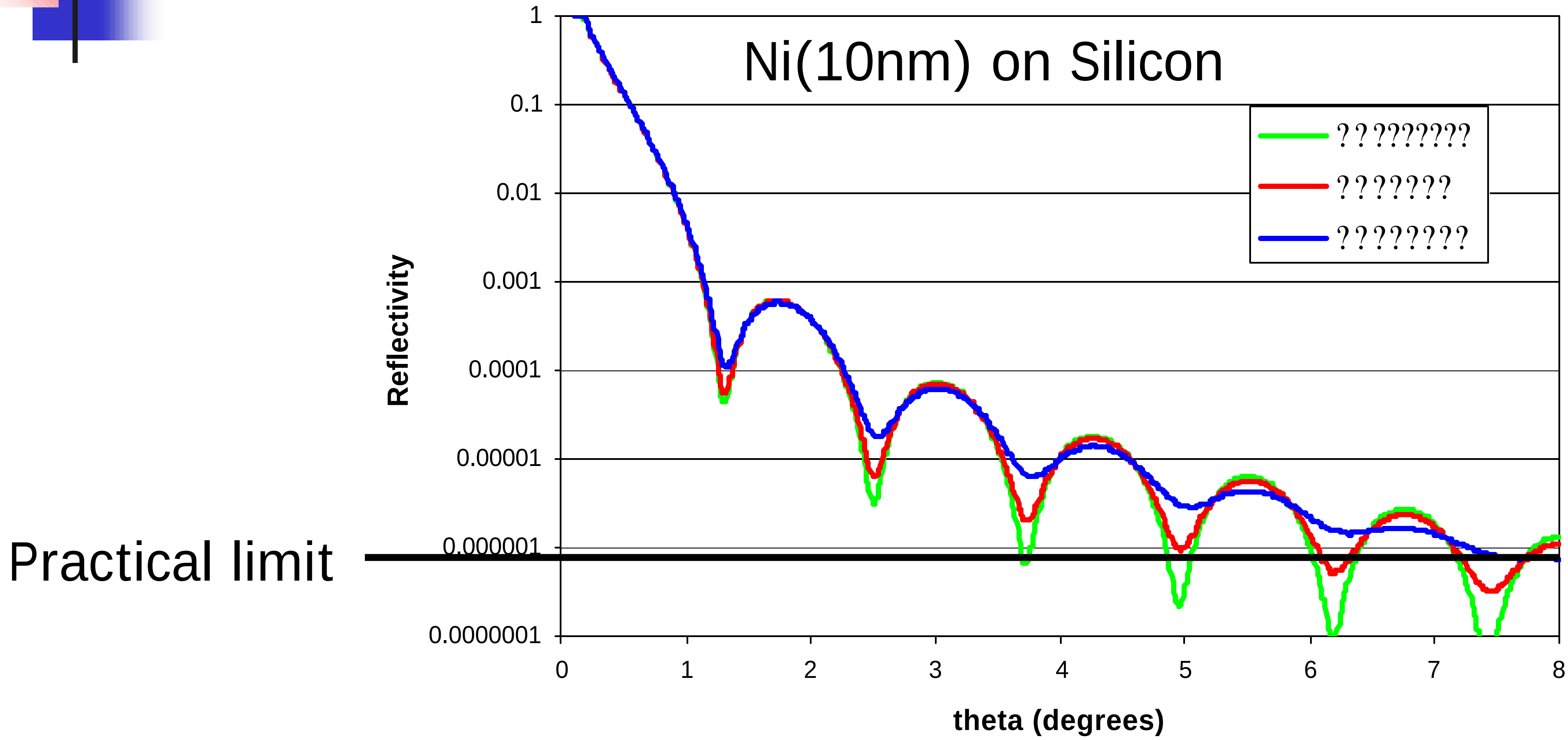


Resolution effects

- Wavelength resolution
  - Graphite monochromator : $\Delta l / l \sim 1\%$
  - Multilayer monochromator : $5\% < \Delta l / l < 20\%$
    (not adjustable)
  - Chopper (ToF) : adjustable $1\% < \Delta l / l < 20\%$
- Angular resolution
  - Defined by the slits sizes $dq = 2W / L$

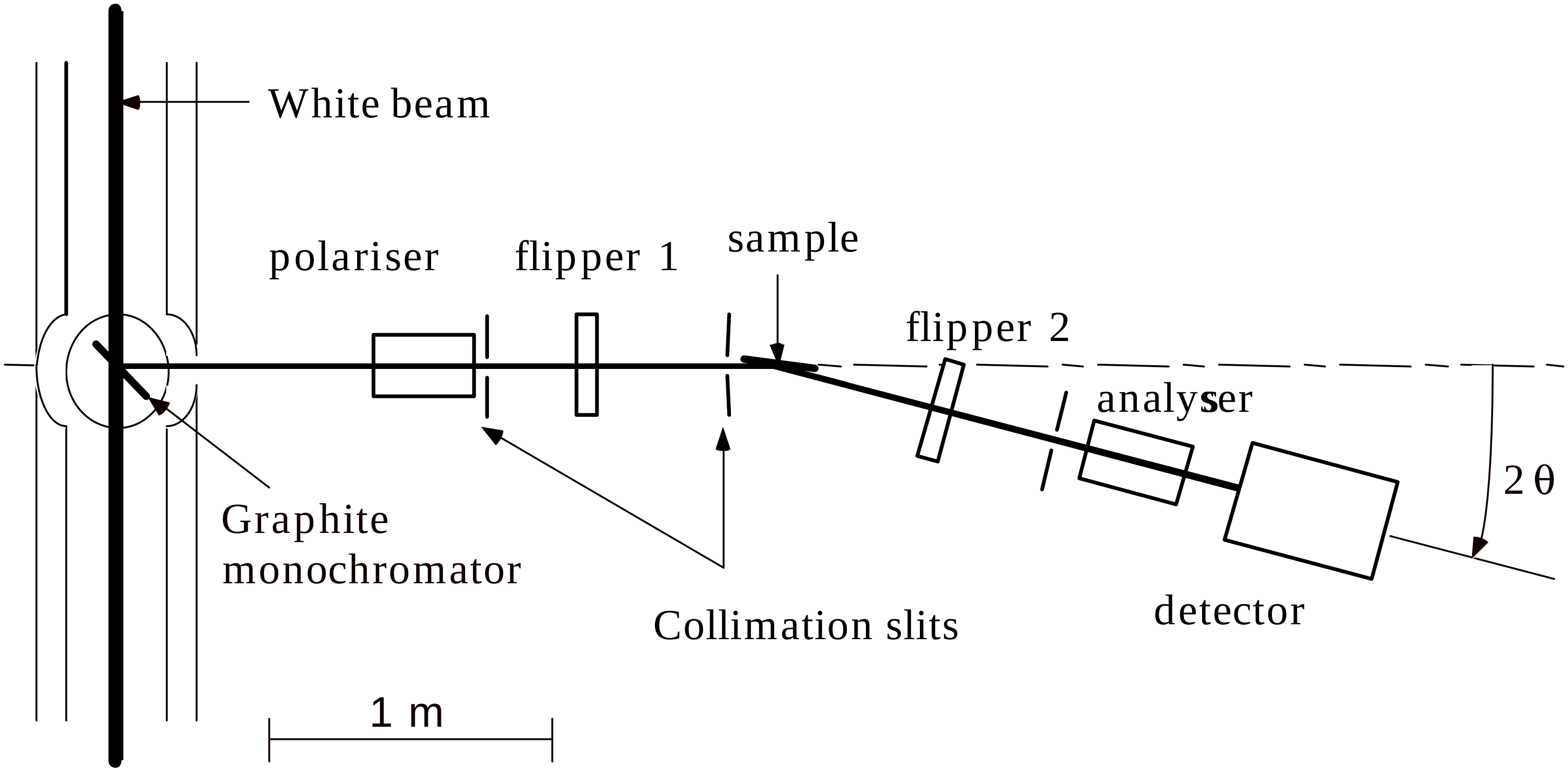


Resolution effects

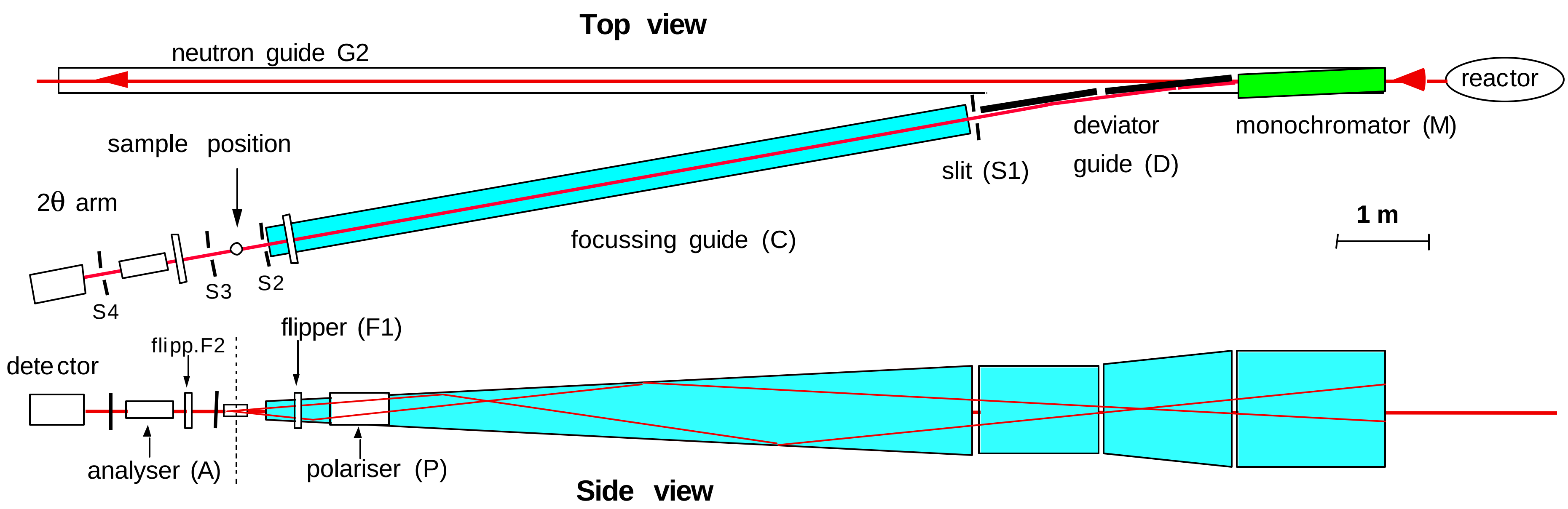


- The resolution must be adjusted to be compatible with the studied sample

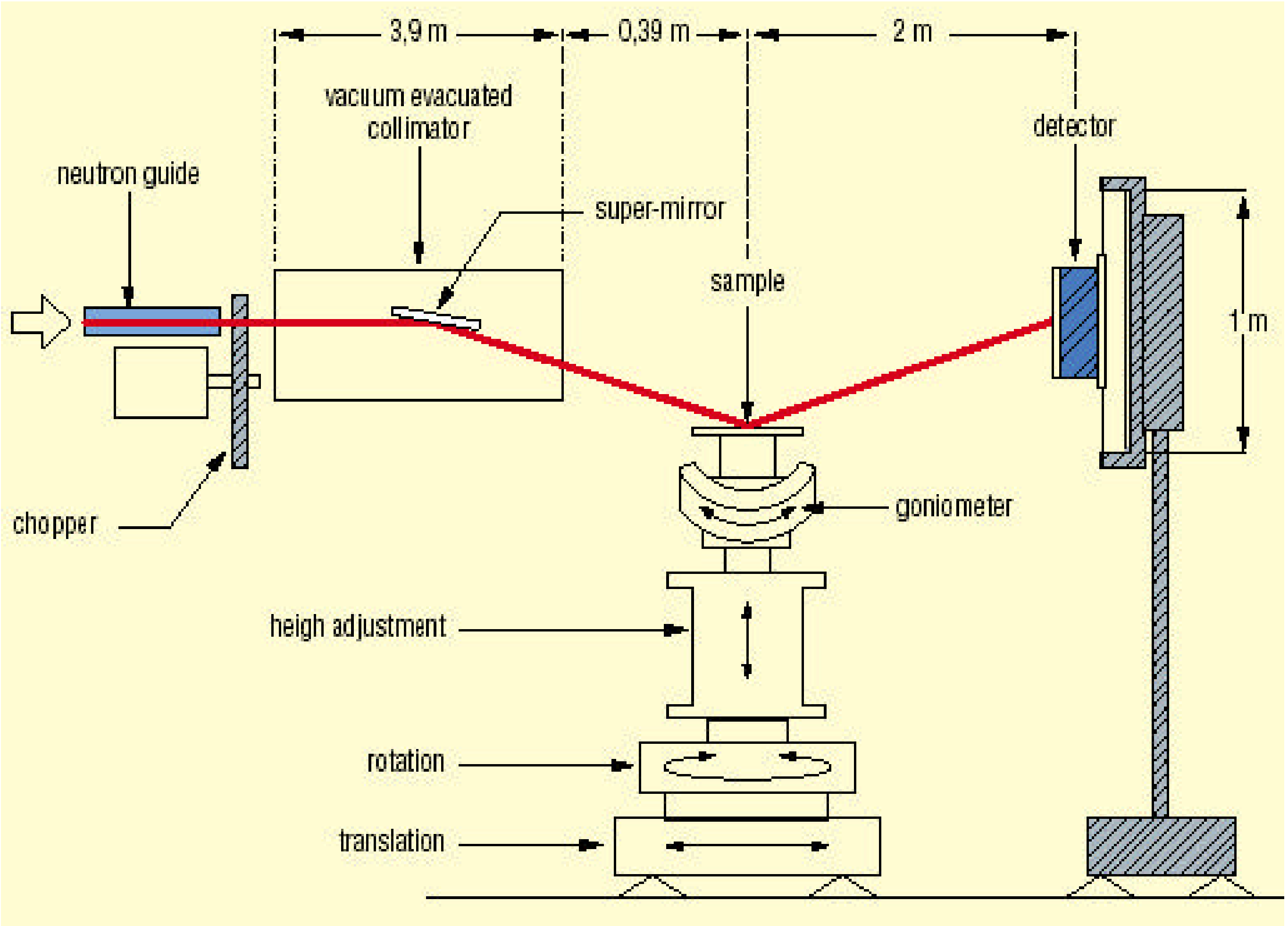
2-axis spectrometer



Upgraded 2-axis spectrometer



ToF reflectometer : EROS



PNR range of studies

- Multilayers
- Superconductors
- Non colinear magnetism
- Interface magnetism

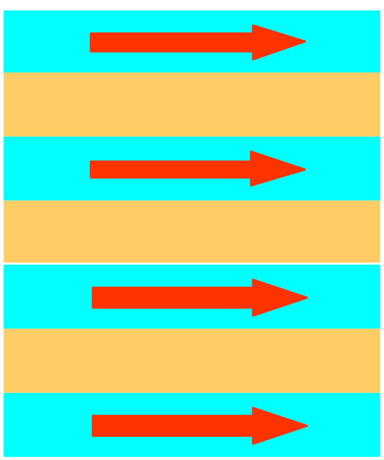
Polarised Neutron Reflectivity

- Allows the study of the magnetic configuration of a multilayer system:
  access to the magnetisation amplitude and direction in each layer.
  - Determination of in-depth magnetic profiles
  - Absolute measurement of the magnetic moment in μ_B per f.u. (sum of the spin and orbital moment)
  - But sensitivity only to the in-plane moment.
  - Resolution of the order of 0.1μB (better on simple systems)
  - No sensitivity to the substrate para/dia-magnetism.
  - No absorption, no phenomenological parameter, absolute normalisation.

Magnetic coupling

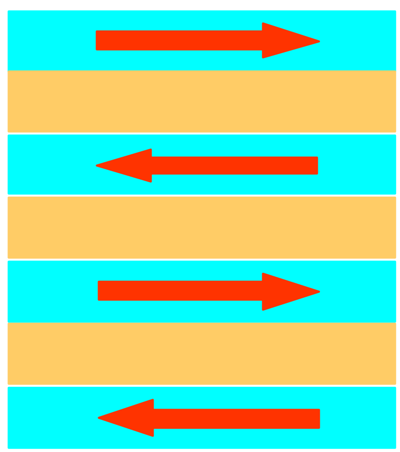
$$E_{couplage} = -J_1\, \vec{S}_1 \cdot \vec{S}_2 - J_2 \left(\vec{S}_1 \cdot \vec{S}_2\right)^2$$

FERRO



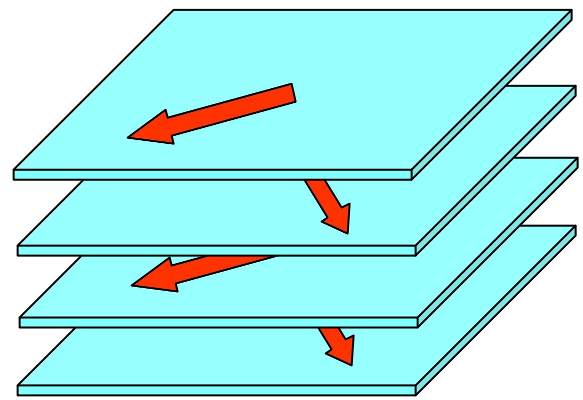
$J_1 > 0$

ANTI - FERRO



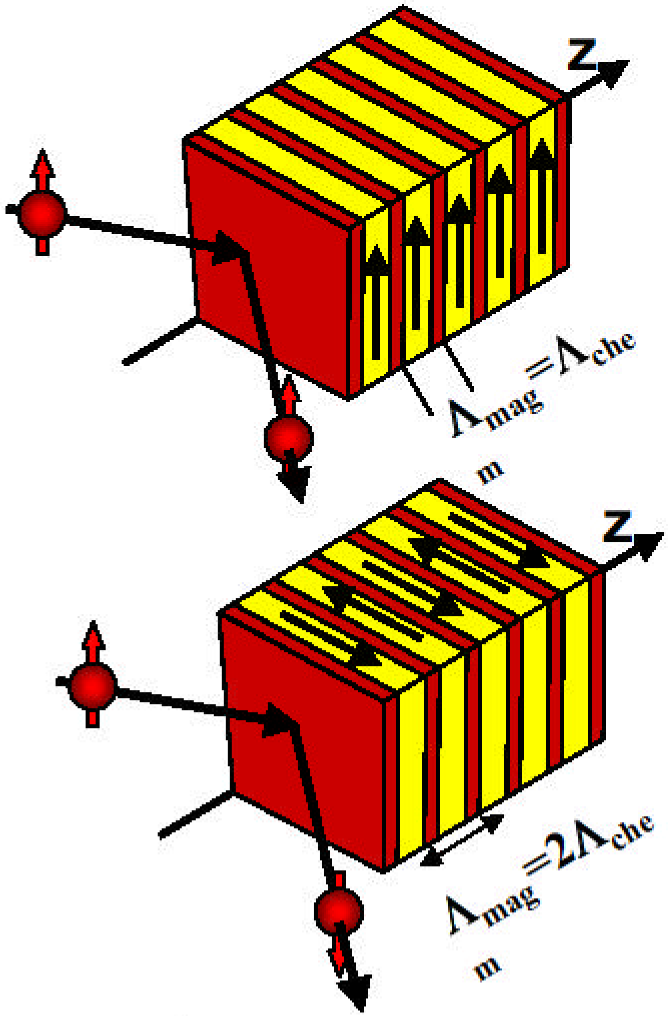
$J_1 < 0$

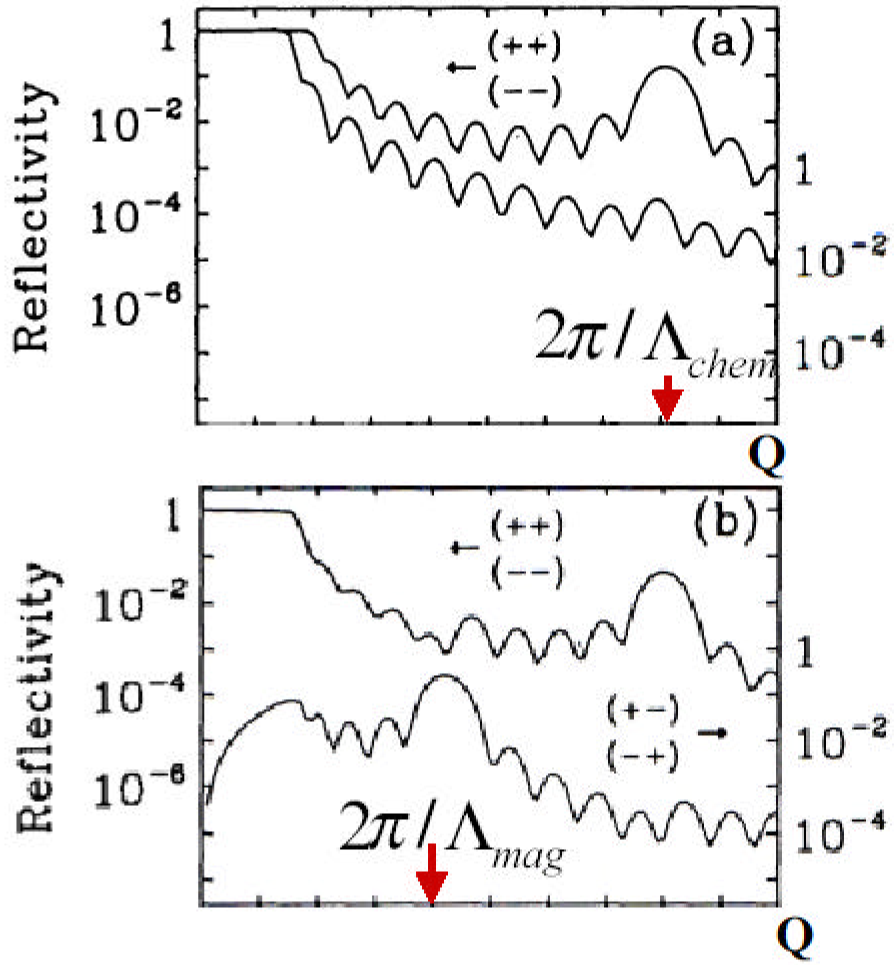
Non colinéaires



$J_1 > 0$ et $J_2 < 0$

PNR on super-lattices

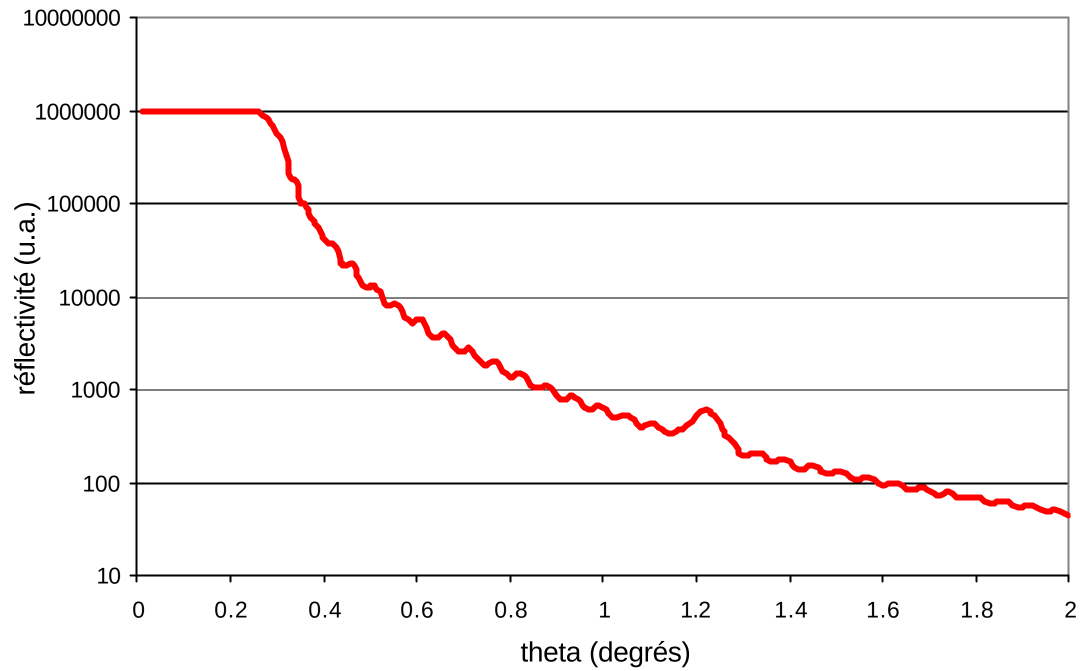




Adapted from H. Zabel

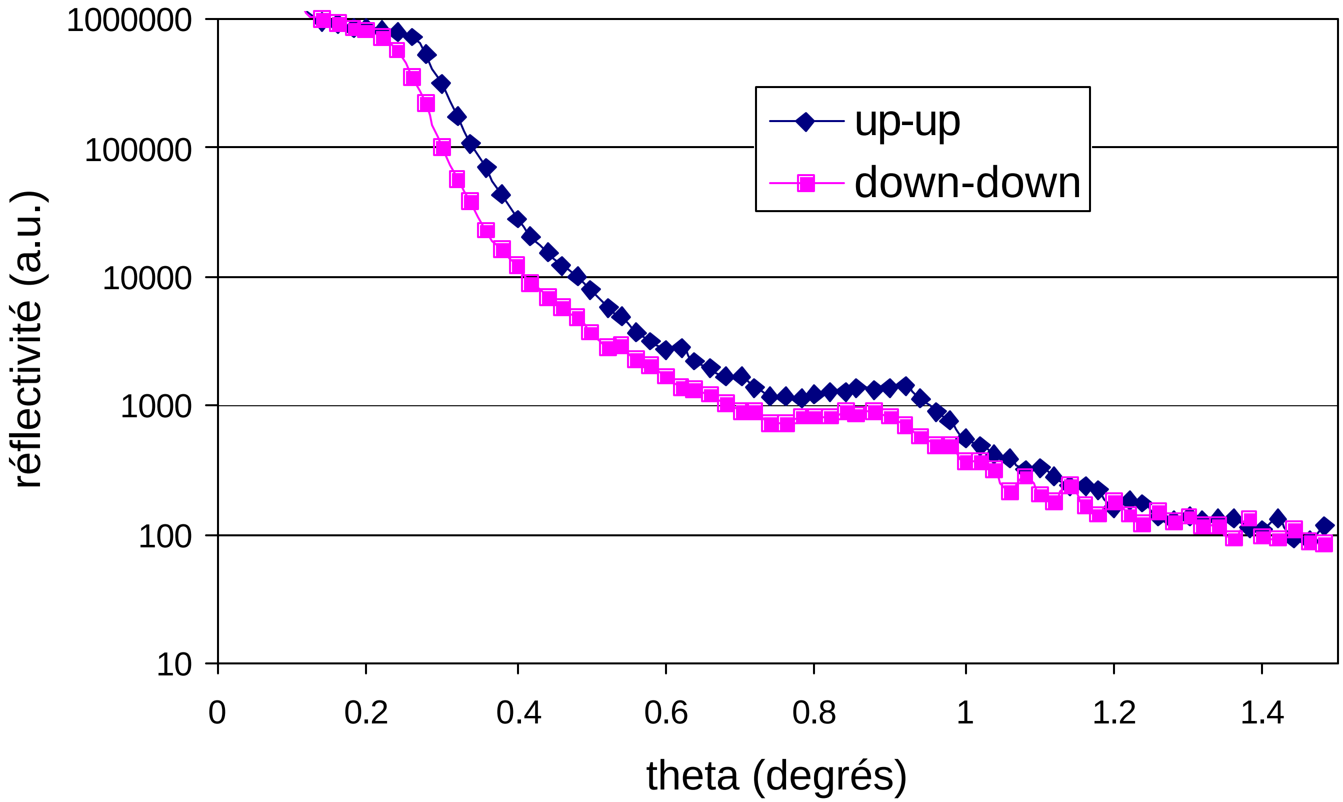
Modulated structures

- Example of a modulation of period 10 nm in a layer of thickness 200nm in YBCO/STO
- Index variation of 2% only :
  - Difference of density
  - stœchiométrie variation
  - Magnetisation modulation (l'aimantation
- YBCO
  - Y: 10%
    Ba: 12%
    Cu: 30%
    O: 49%



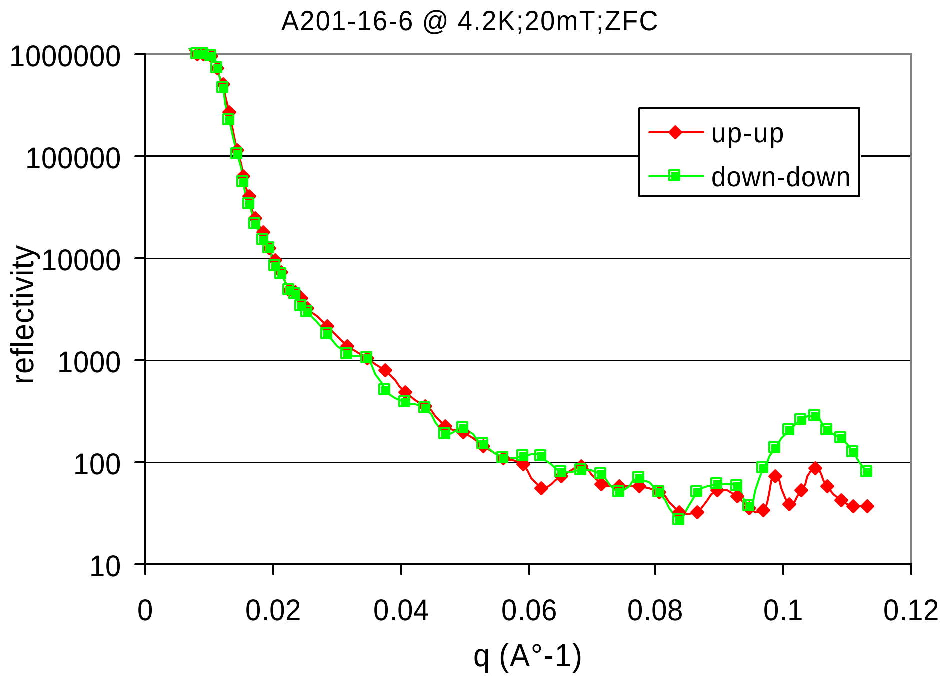
Example: magnetisation modulation

- $La_{0.3}Sr_{0.7}MnO_3$ film



Exchange coupling in super-lattices [GaMnAs/GaAs]

- Super-lattice $(GaMnAs)_m/(GaAs)_n$ where 8<m<16 et 4<n<8
  Mn doping 6-7%

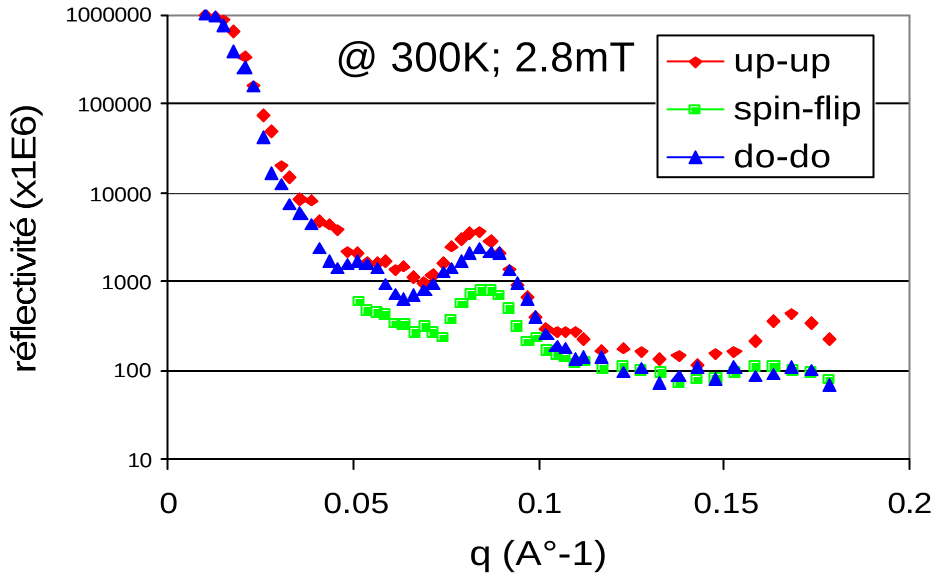


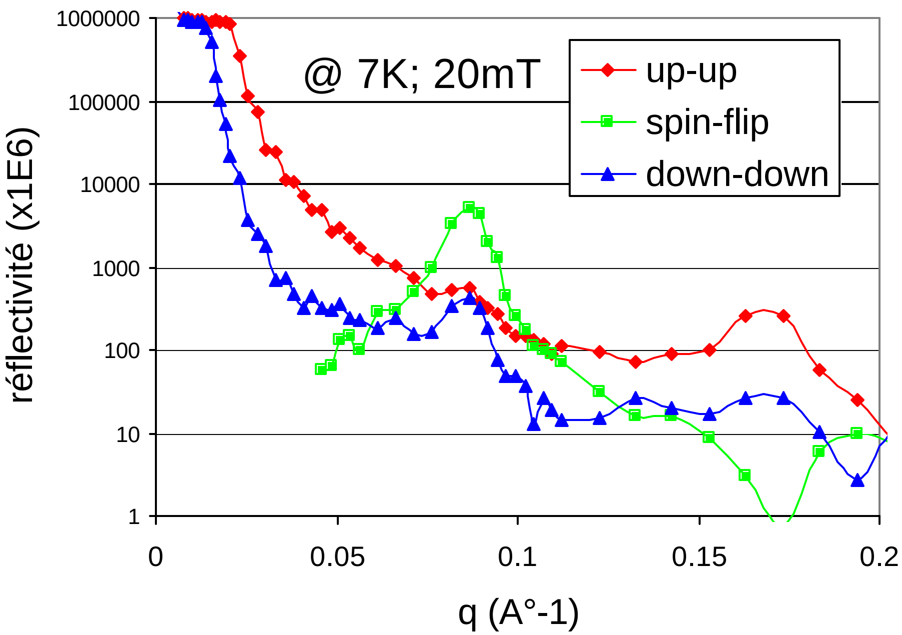
Magnetisation of 0.03 T (27kA/m)

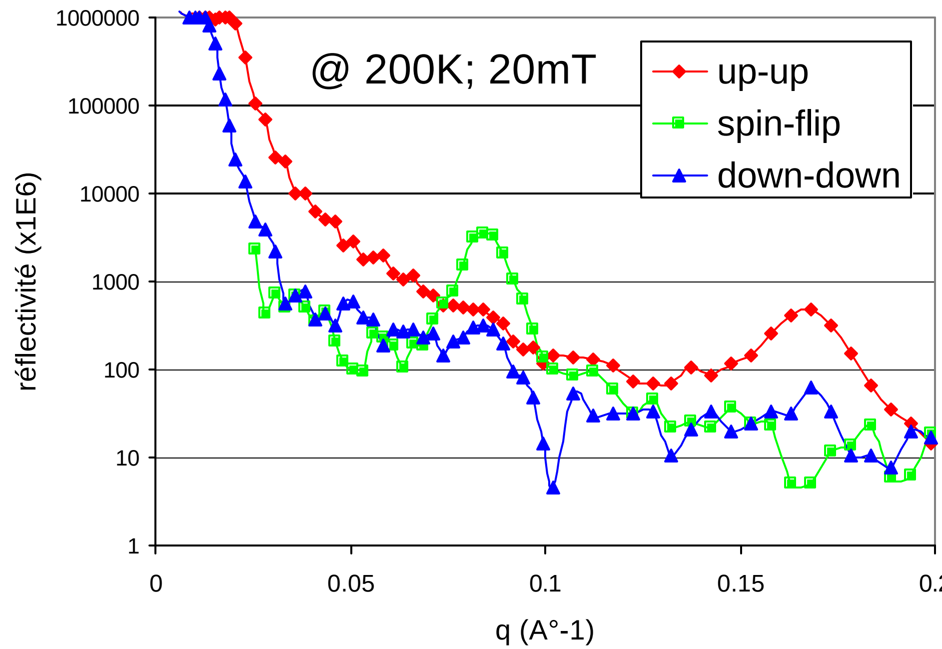
No antiferromagnetic coupling is observed.

Magnetic ordering in multilayers [Fe/Si]$_n$

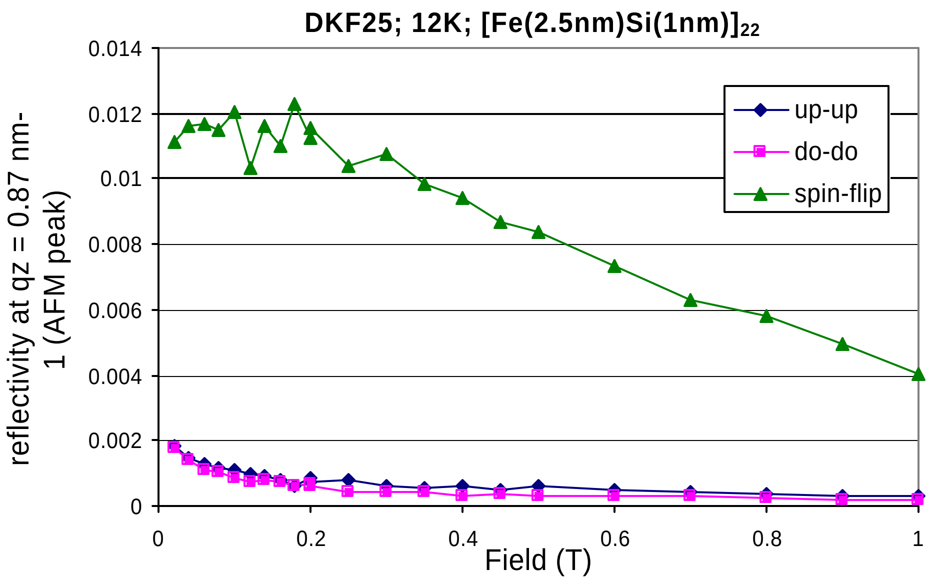
- K. Fronc (Polish. Acad. Sc.)
- GaAs//[Fe(2.4nm)Si(1.2nm)]$_n$
- magnetic AF order at 300K
- non collinear coupling at 200K

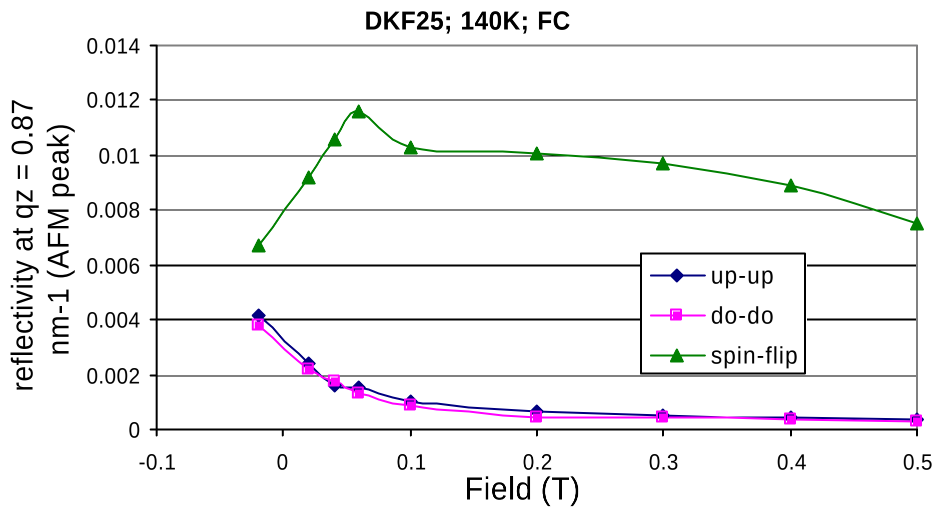






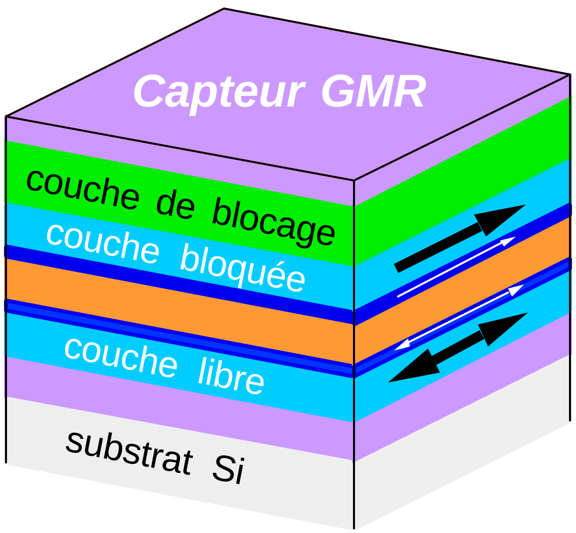
Evolution of the magnetic coupling as a function of the magnetic field



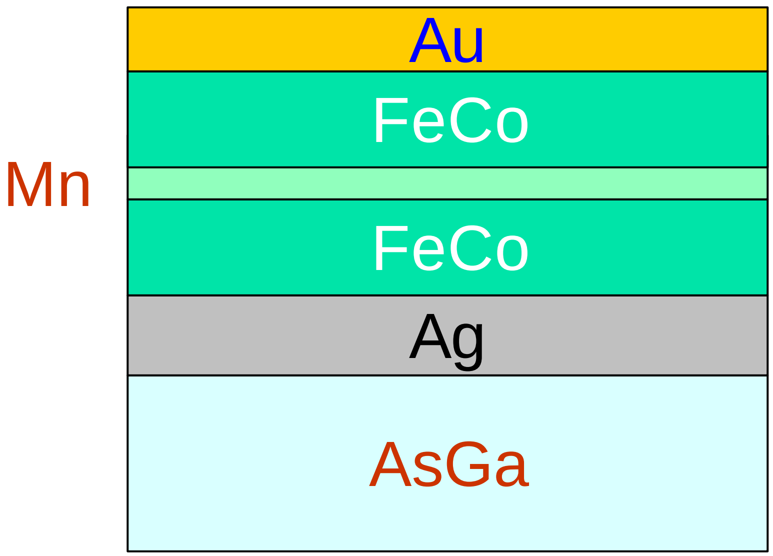


- AFM component disappears with the applied field @1.5 T (for 10K)

Spin-valves structures



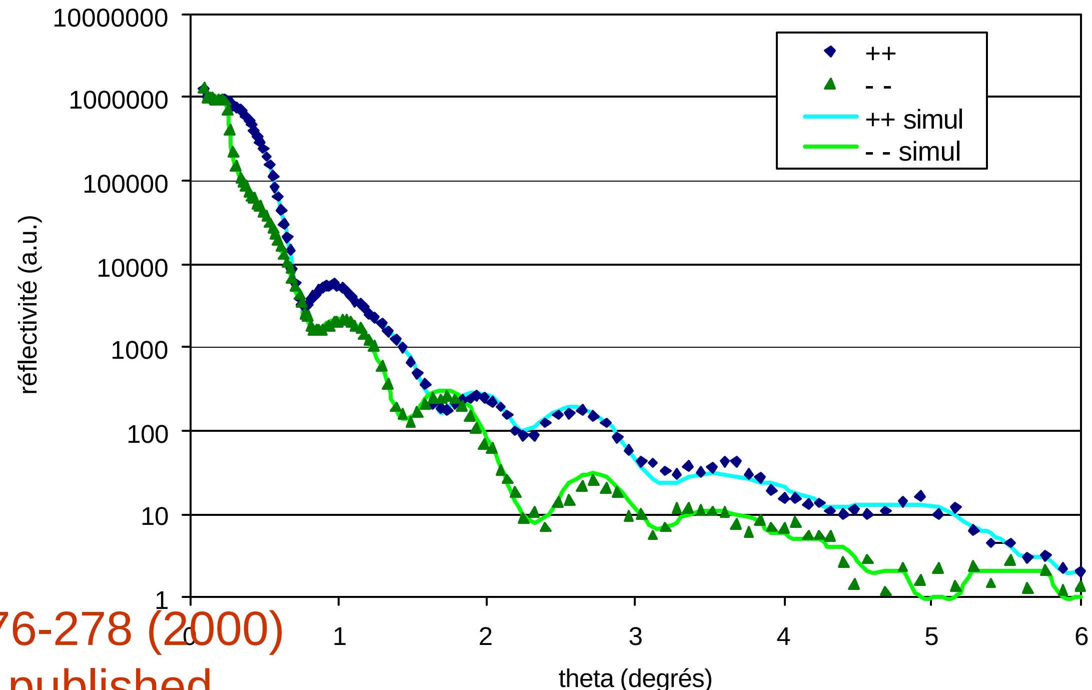
More academic structure



E. Kentzinger, S. Nerger, U. Rücker,
J. Voigt, O.H. Seeck, Th. Brückel
(Forschungszentrum Jülich, Germany)

In a saturating field of 0.5T

- $Fe_{0.5}Co_{0.5}$/Mn (8A°)/ $Fe_{0.5}Co_{0.5}$
- Moment of 2.4 µB/atom in $Fe_{0.5}Co_{0.5}$
- Moment in manganes
- Theoretically predicted in FeCo alloys (not observed in Fe alone)

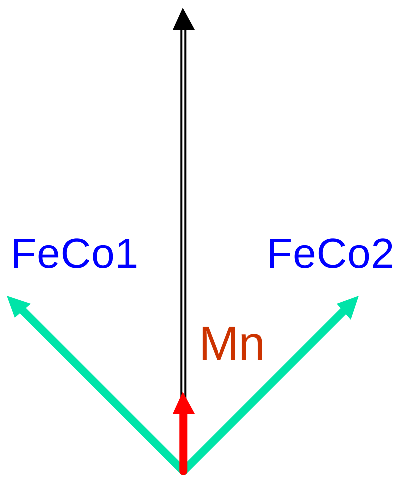


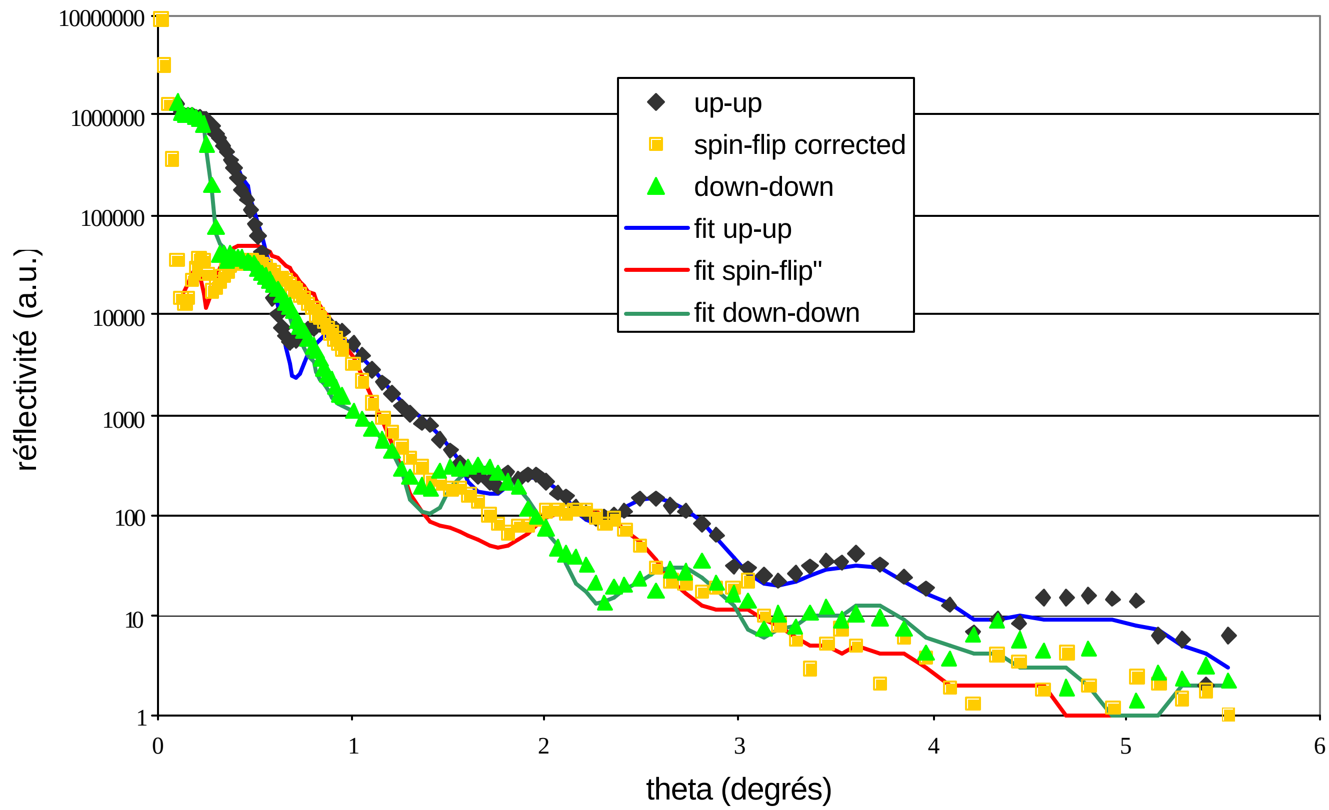
E. Kentzinger et al., Physica B 276-278 (2000)
S. Nerger et al., Physica B, to be published.

Measurement in low field

Quadratic coupling

B = 1.2mT

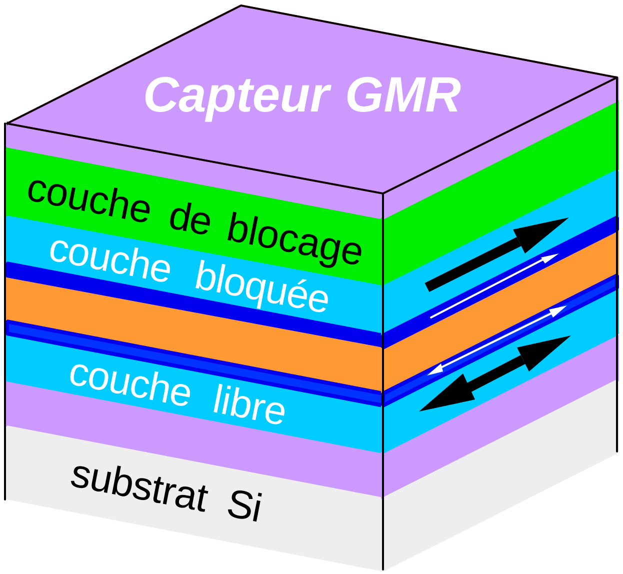




GMR optimisation

Typical GMR structure

SiO_2// Ta/ NiFe/ CoFe/ Cu/ CoFe/ MnPt/ Ta

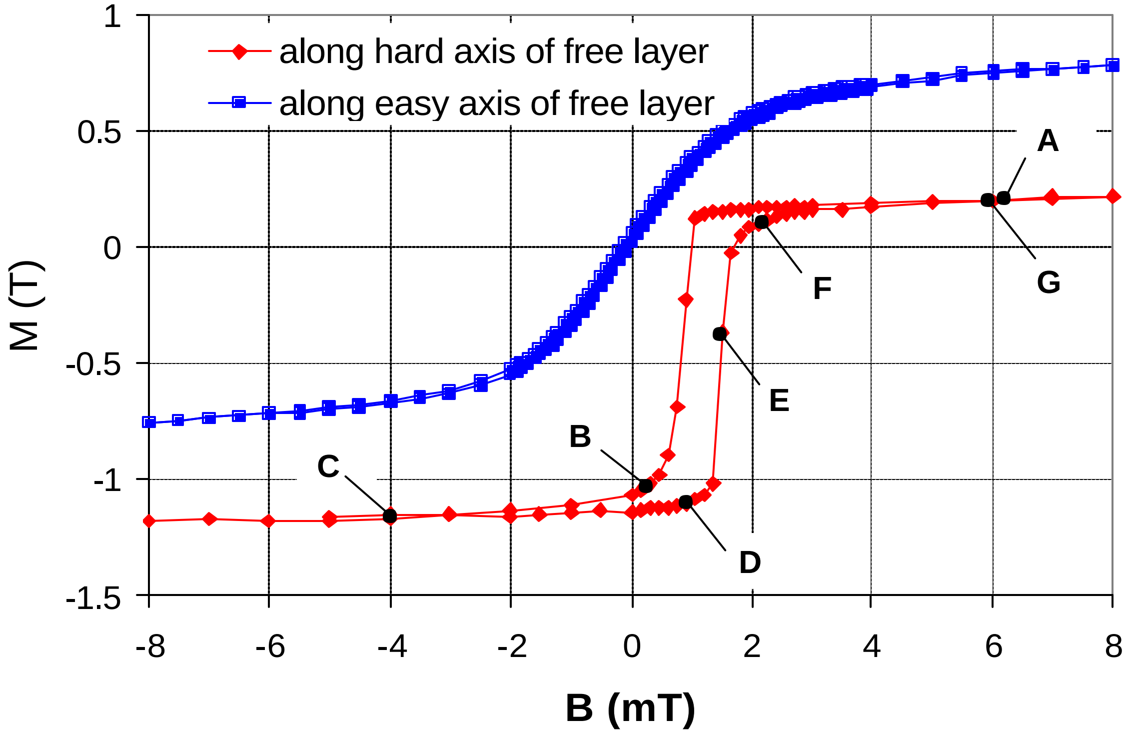


- Aim to to optimize GMR sensors used in high density tape recording

PNR magnetometry

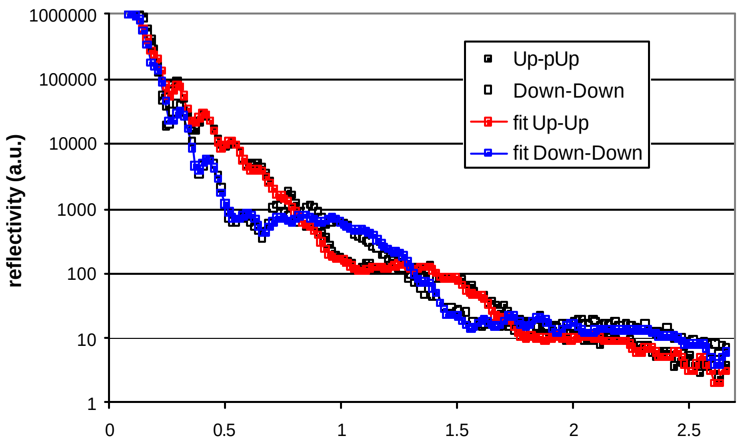
- characterize the system magnetically in a saturating field (thickness and amplitudes of the magnetic moments).
- sweep the field H or the temperature T but restrict the measurement to a few points of the reflectivity curve
- Adjust these points by letting vary only the amplitude and direction of the magnetic moments in the multilayer model.

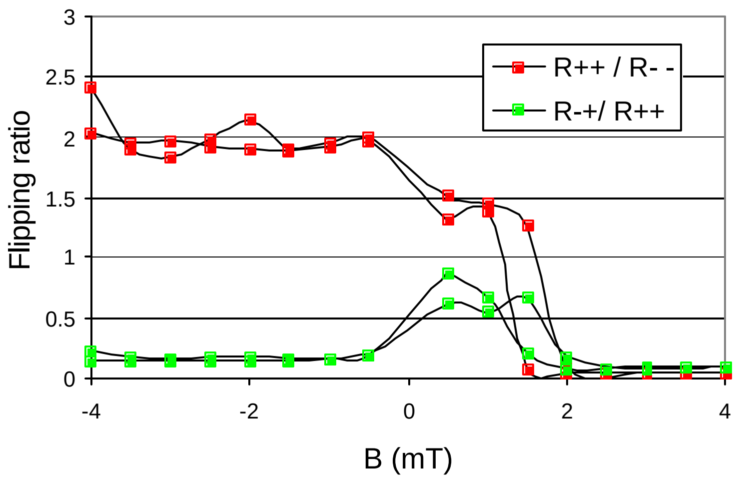
Hysteresis cycle



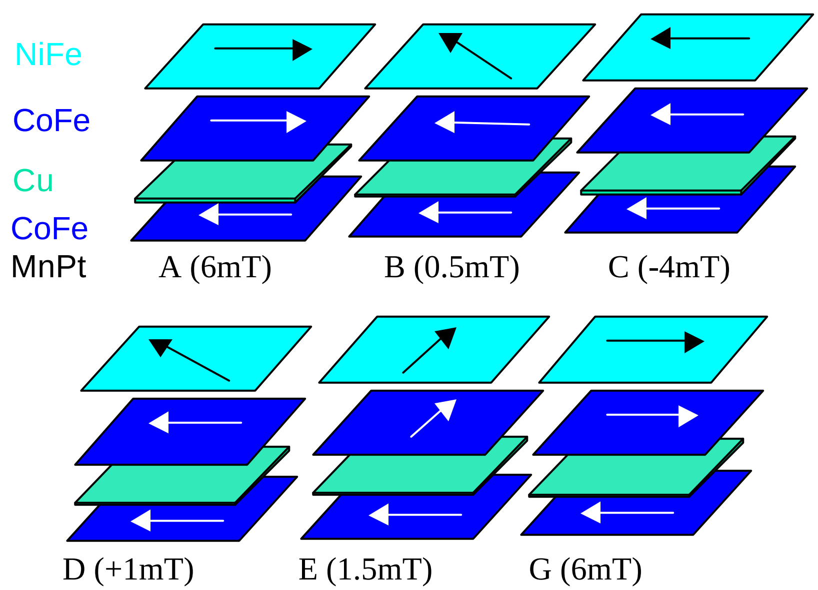
The easy axis of the AF layer is perpendicular to the easy axis of the free layer

PNR magnetometry





- Magnetisation of the different layers as a function of the applied field



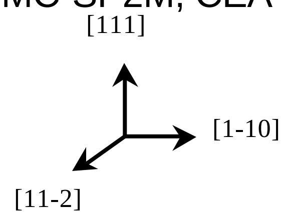
Spin injection materials

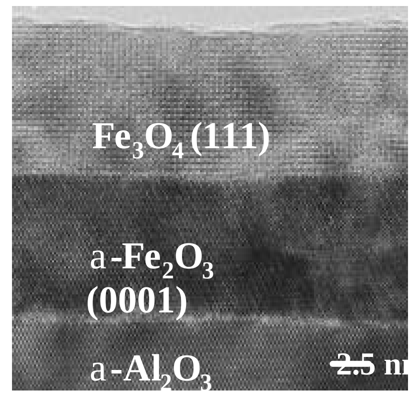
Magnetite (DRECAM/SPCSI, J.B. Moussy et al)

- Fabrication of all oxide magnetic junctions
- Combination of Al_2O_3, Fe_2O_3, Fe_3O_4 layers
- Magnetite Fe_3O_4 is a potential candidate as spin-injector material
- Typical structure : A_2O_3//Fe_2O_3/Fe_3O_4/Al_2O_3/Fe_3O_4

HRTEM

Collaboration : P. Bayle-Guillemaud, P. Warin, DRFMC-SP2M, CEA-Grenoble

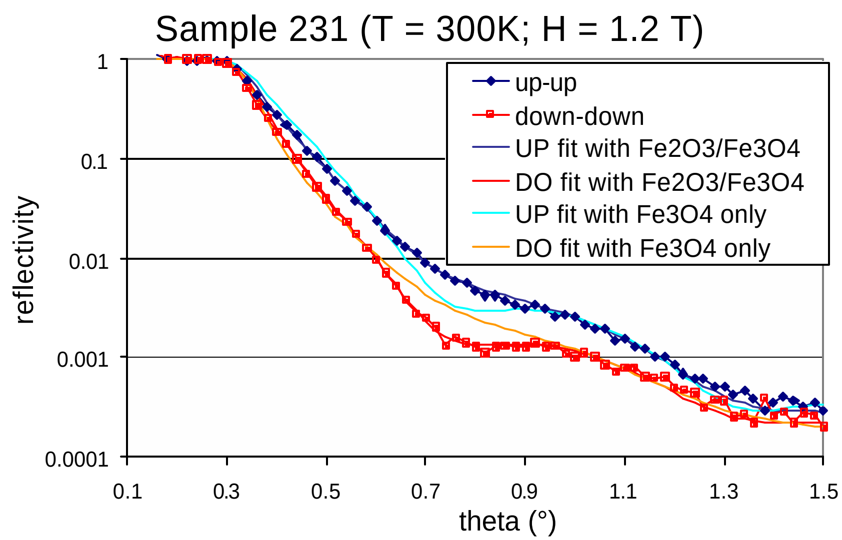




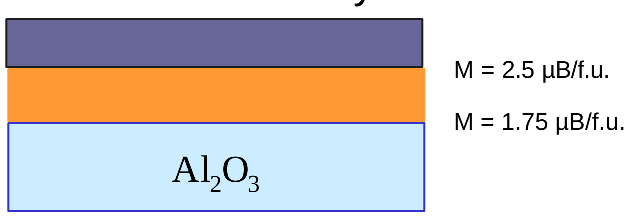
- BUT often a partial or total transformation of the Fe_2O_3 into Fe_3O_4 occurs
(not visible during the deposition process using XPS or RHEED)

PNR characterisation

- Neutron reflectivity allows to very quickly check the presence or absence of Fe_2O_3 layer (by using the magnetic contrast)



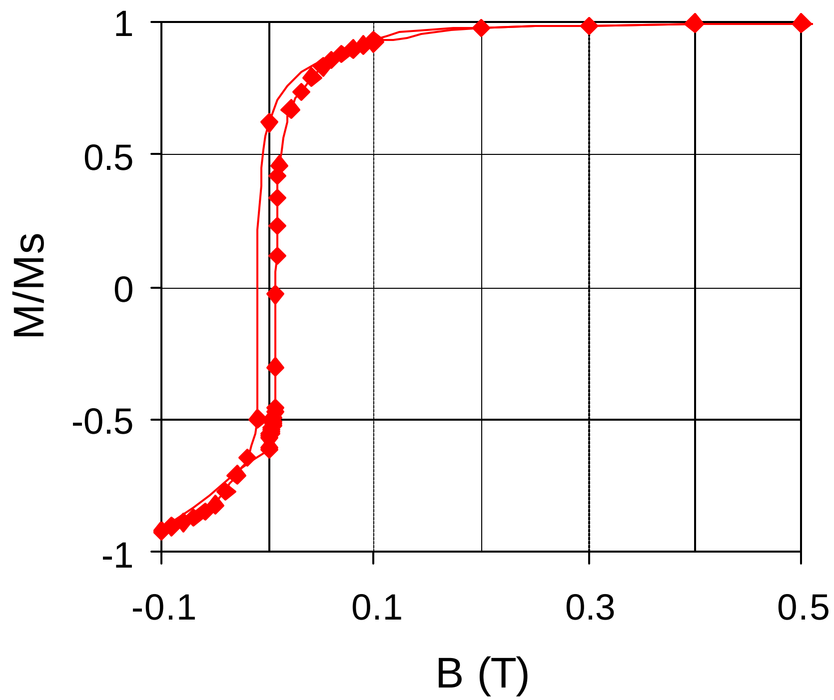
- Information on the magnetic moments :
  - the transformed layer has a reduced magnetic moment

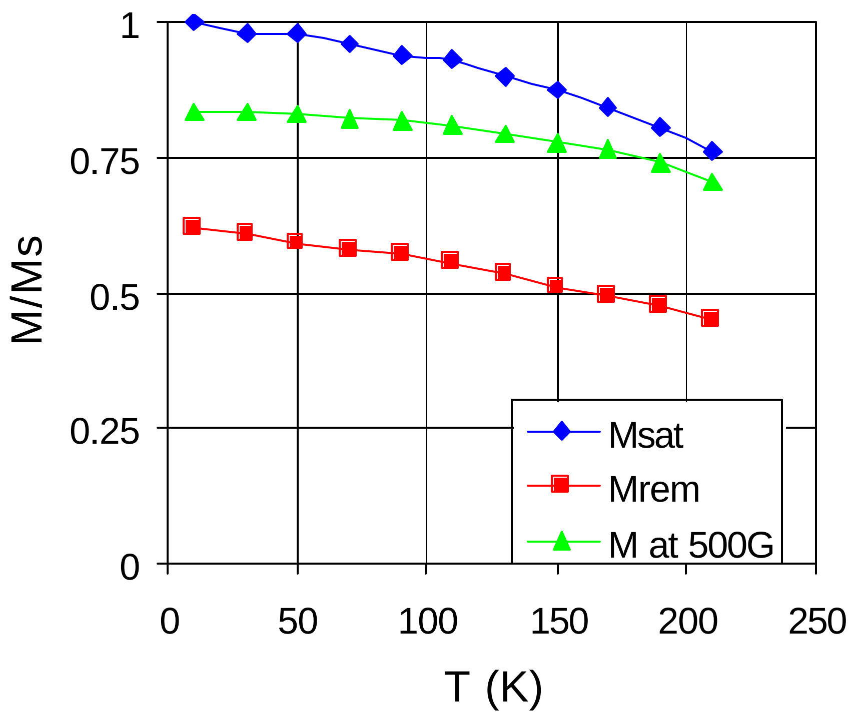


The transformation process is not yet understood

$La_{0.7}Sr_{0.3}MnO_3$: Hysteresis cycle

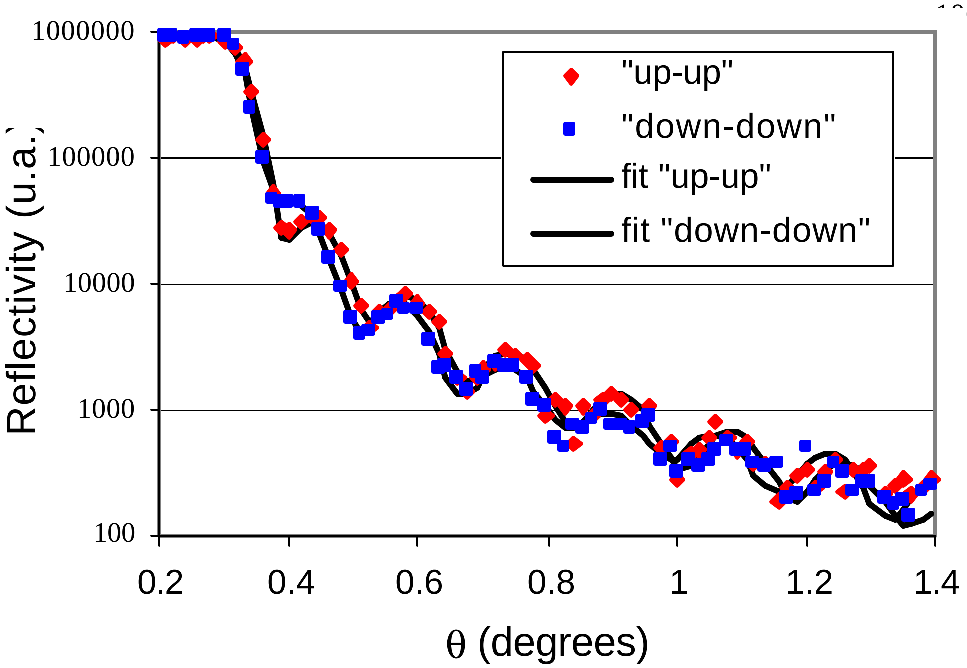
LSMO film (40 nm) deposited on a $SrTiO_3$ substrate

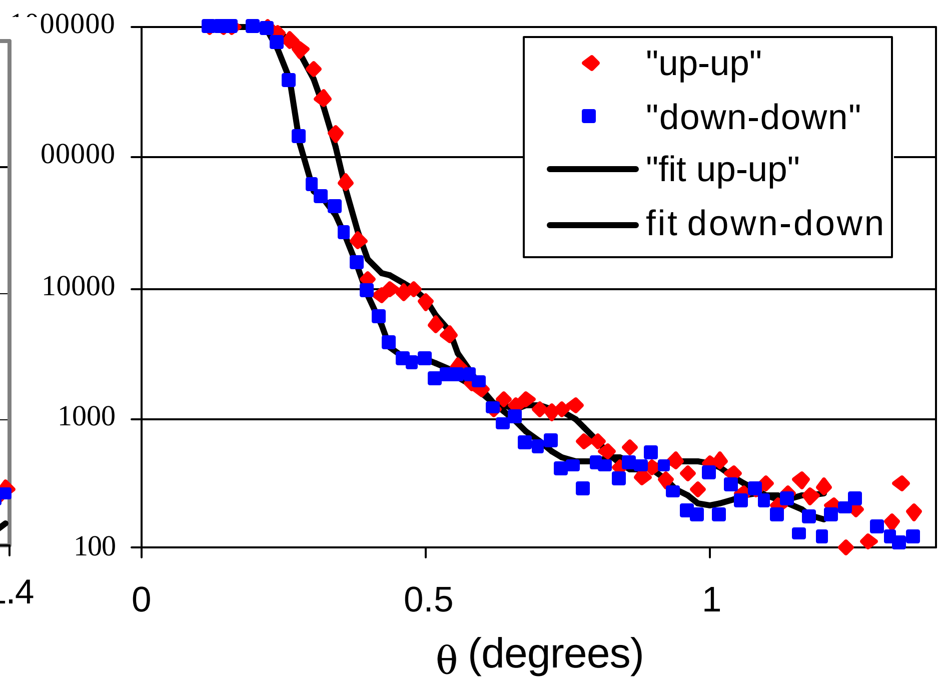




Reflectivity measurements.

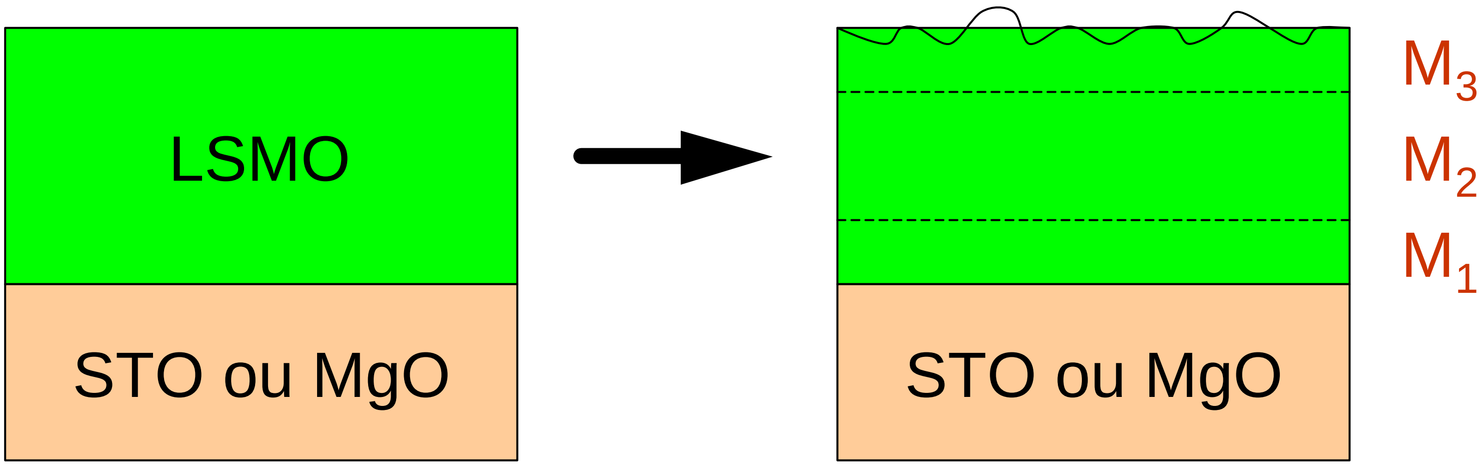
- LSMO on MgO (68nm) LSMO on STO (56nm)



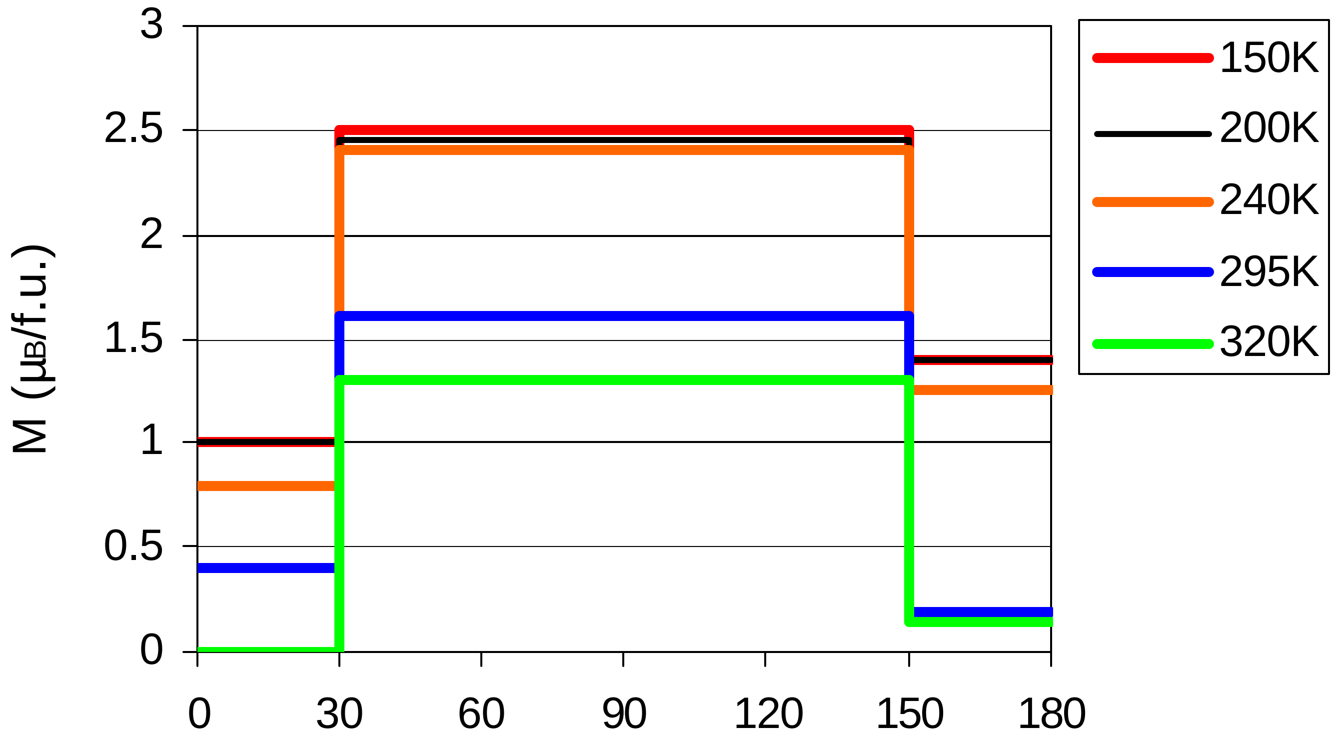


Fitting procedure

- Perfect system
- More realistic model



Magnetic profile in a LSMO on STO

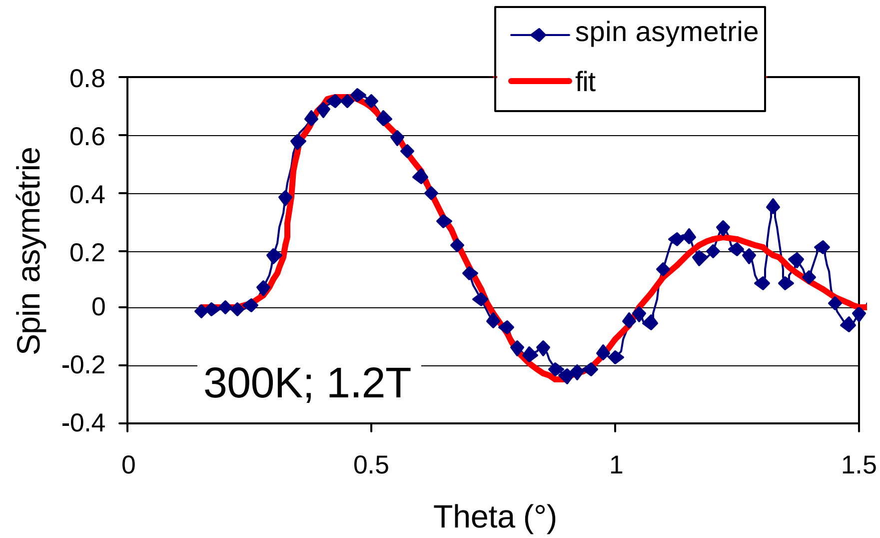


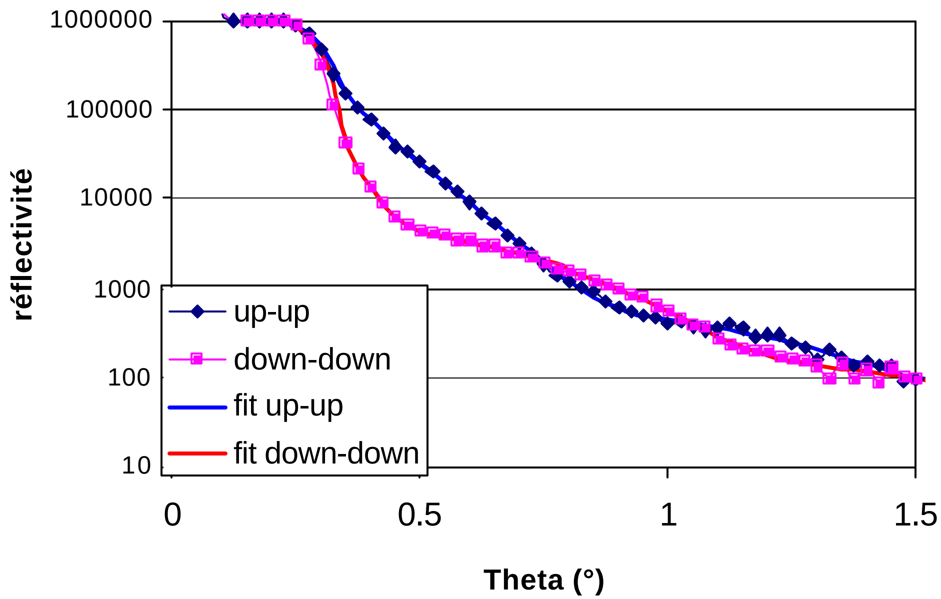
$SrTiO_3$ vacuum

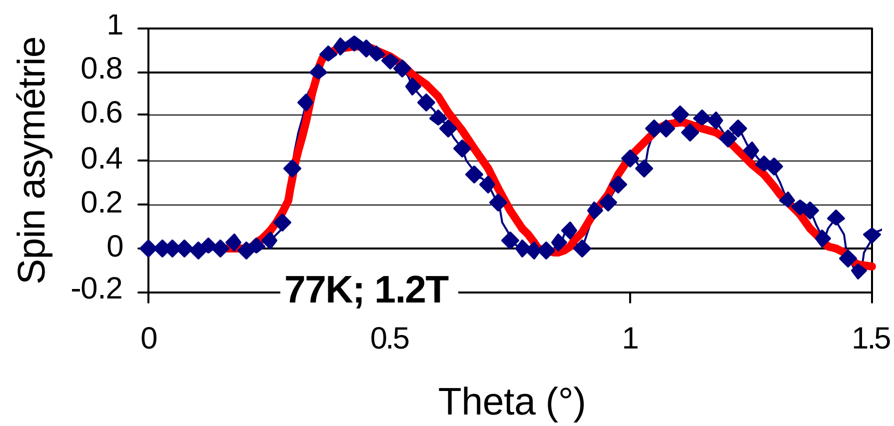
LSMO film (16nm) sur STO

- Spin asymmetry

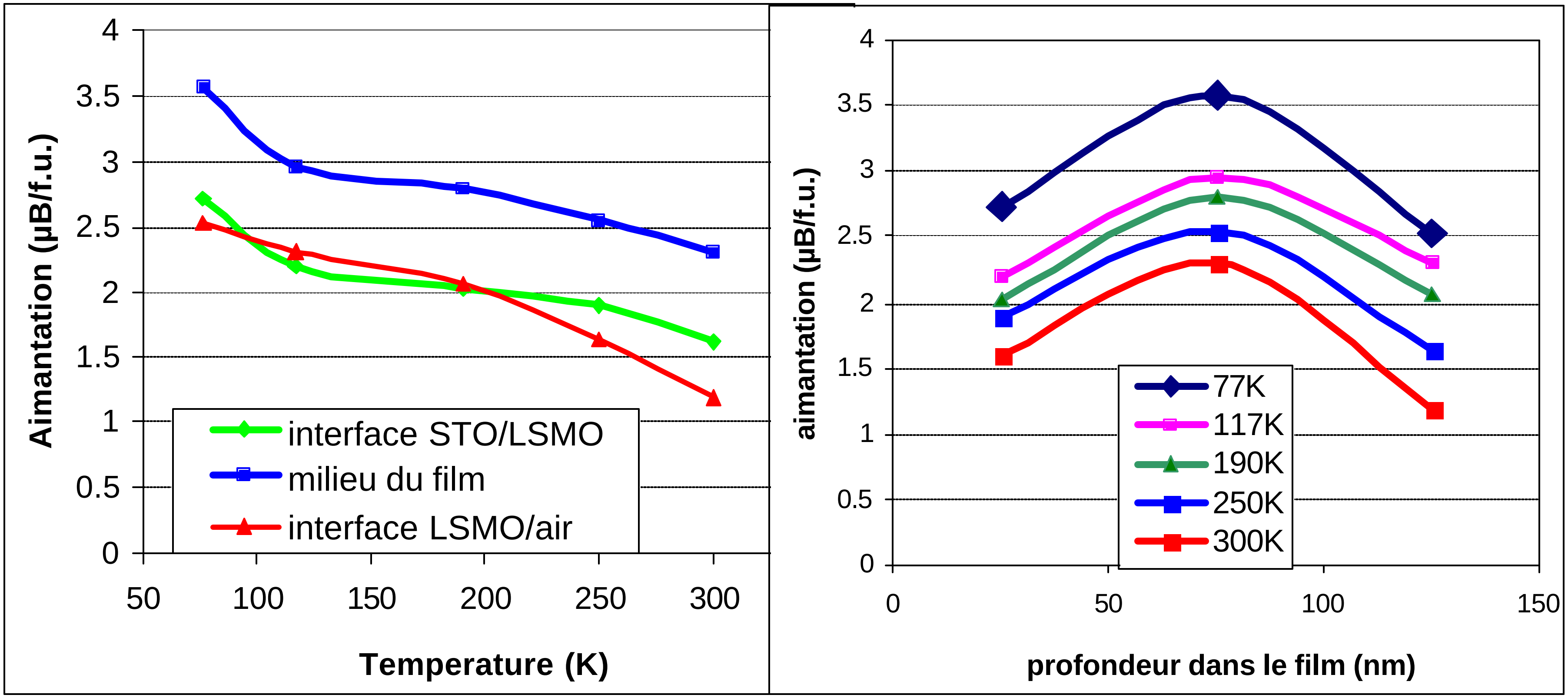
$$\frac{R^{++} - R^{--}}{R^{++} + R^{--}}$$







Magnetisation variations (LSMO(16nm)/ STO)



Conclusion

- Applications
  - Multilayers
  - Non colinear magnetism
  - Interface magnetism
- Determination of magnetic profiles with a depth resolution: access to the magnetisation amplitude and direction in each layer.
  - Determination of in-depth magnetic profiles
  - Absolute measurement of the magnetic moment in μ_B per f.u. (sum of the spin S and orbital moment L)
  - But sensitivity only to the in-plane moment.
  - Resolution of the order of 0.1µB (better on simple systems)
  - No sensitivity to the substrate para/dia-magnetism.
  - No absorption, no phenomenological parameter, absolute normalisation.
  - “low” flux.

Bibliography

- A few recent general references

  - H. Zabel et al, Physica B 276-278 (2000) 17-21.
    « Neutron Reflectometry on magnetic thin films »
  - H. Zabel et al, J. Phys.: Condens. Matter 15 (2003) S505-S517.
    Polarized neutron reflectivity and scattering studies of magnetic heterostructures.
  - G.P. Felcher, J. Applied Physics 87 (2000) 5431
    Neutron reflectometry as a tool to study magnetism
  - G. Fragneto-Cusani, J. Phys. : Condens. Matter 13 (2001) 4973-4989

- Other ressources
  - www-llb.cea.fr/prism/PRISM.html
  - http://www.neutron.anl.gov/software.html
  - All existing reflectometers :
    http://www.studsvik.uu.se/research/NR/reflect.htm