Jan Vogel

Institut Néel, CNRS

Grenoble, France

European School on Magnetism 2023

Light – Matter Interaction

↓

Interaction between photons and magnetism

About me :

- Ph.D. in Physics in 1994 (University of Nijmegen, the Netherlands) : study of the magnetic properties of thin films using polarized x-rays
- Postdocs in LURE (Orsay), ESRF (Grenoble) and Laboratoire Louis Néel (Grenoble)
- CNRS researcher in LLN -> Institut Néel sinds 1998

- Magnetic imaging (PEEM, Kerr), magnetization and domain wall dynamics in magnetic thin films and microstructures

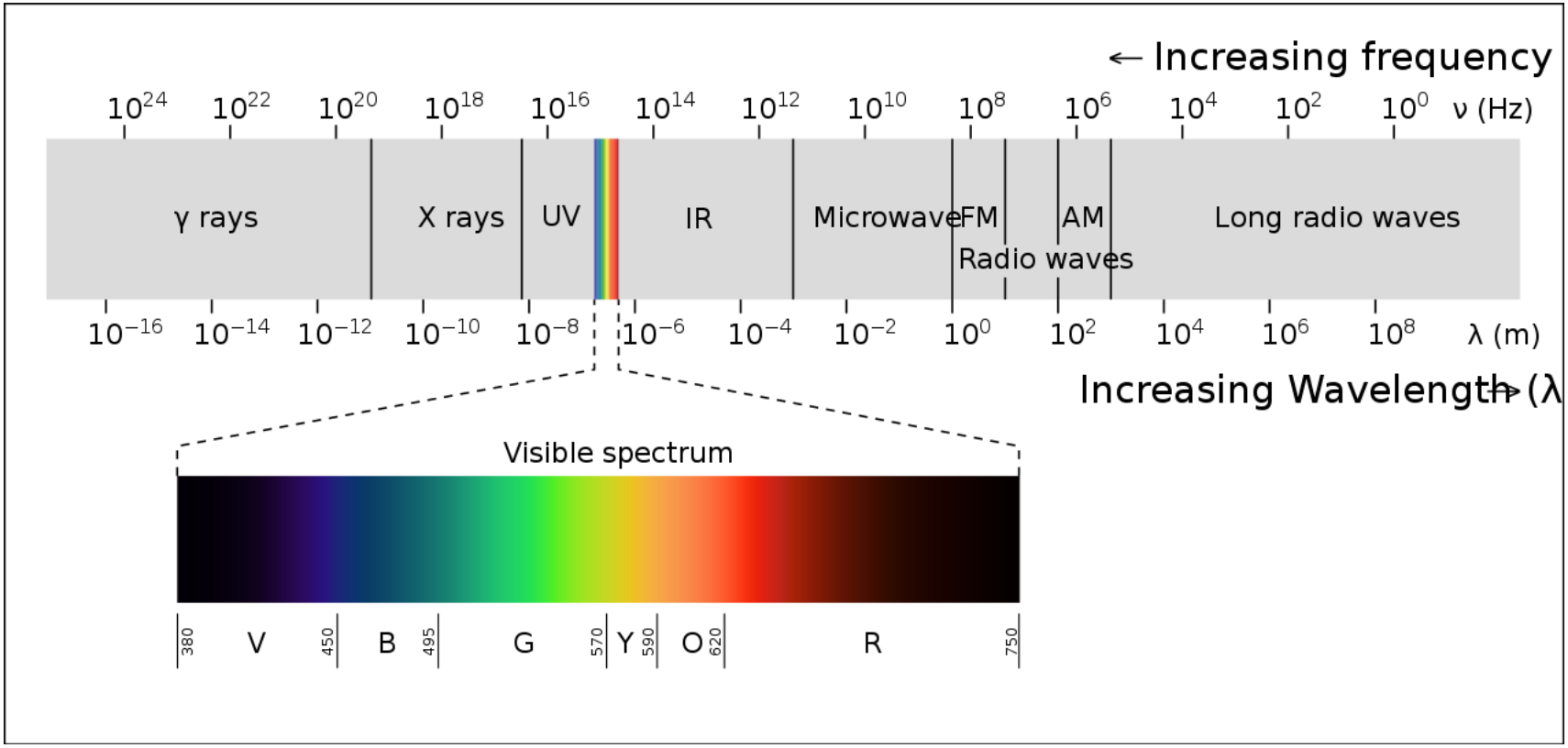
Outline of the lecture

1) **Photons to probe magnetism** :
   - X-ray magnetic circular and linear dichroism
   - Faraday and Kerr effects
   - Brillouin Light Scattering

2) **Influence of light on magnetic properties**
   - Inverse Faraday effect
   - Ultrafast demagnetization using light
   - All-optical magnetic switching

1) Photons to probe magnetism

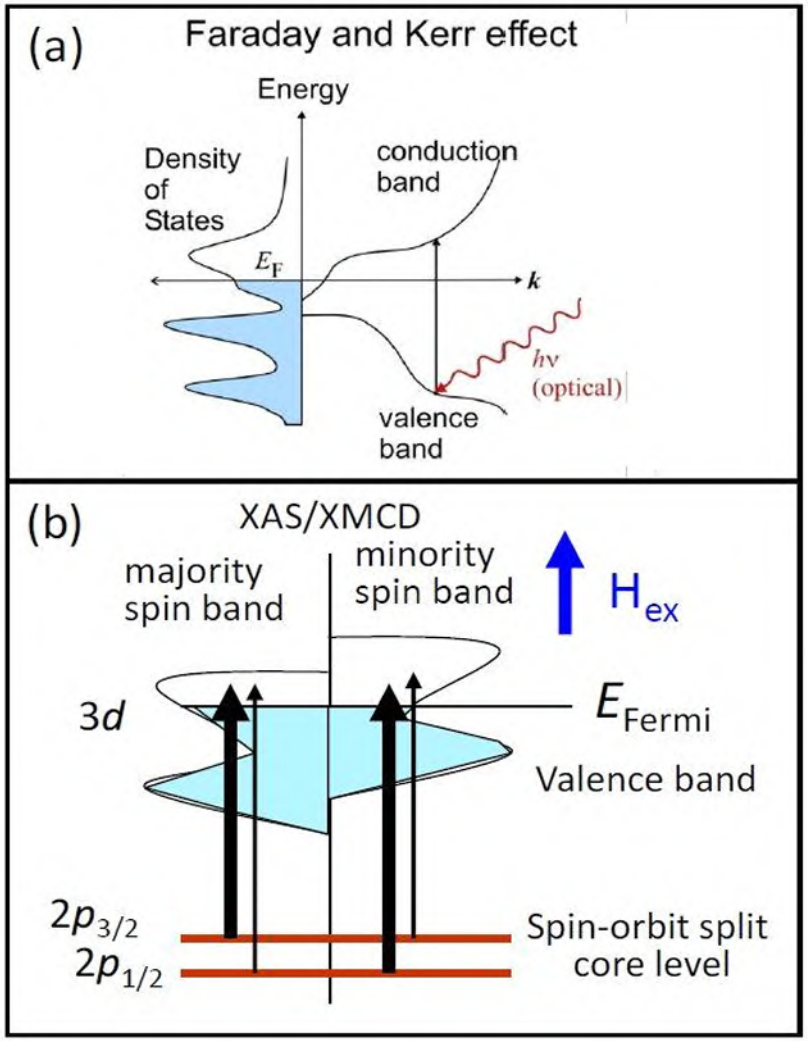
Optical photons vs X-ray photons



$E = h\nu = 1.6 – 3.3$ eV for visible light

$E = h\nu = 50$ eV – 10 keV for x-rays

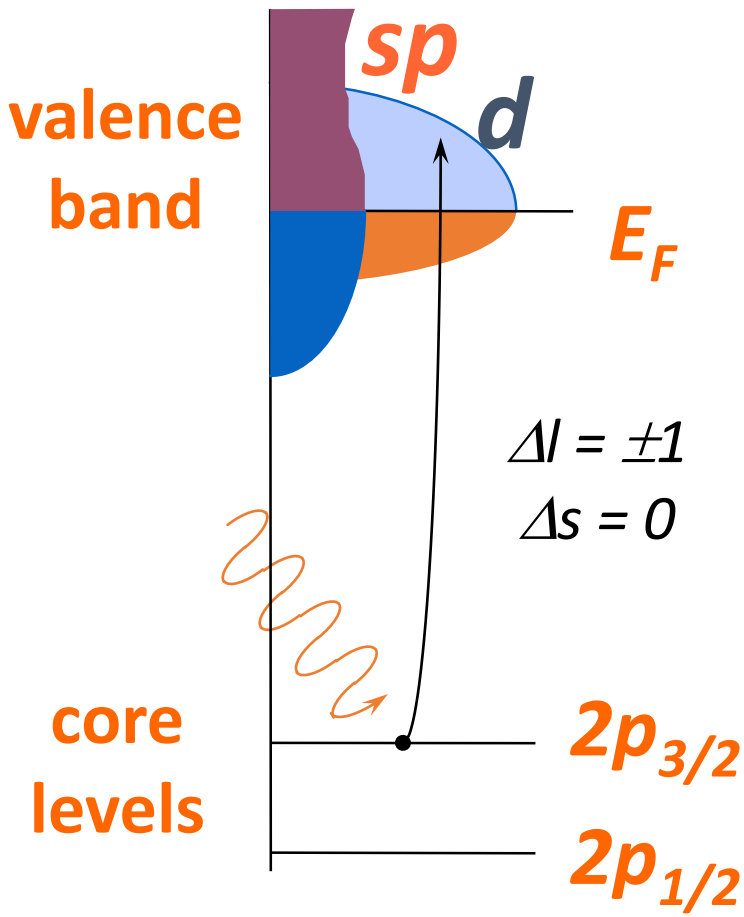
Optical photons vs X-ray photons



Optical photons : transitions between valence band and conduction band → complex initial and final states

X-ray photons : transitions between core level and conduction band → well-defined initial states

X-ray Absorption



To study magnetism :

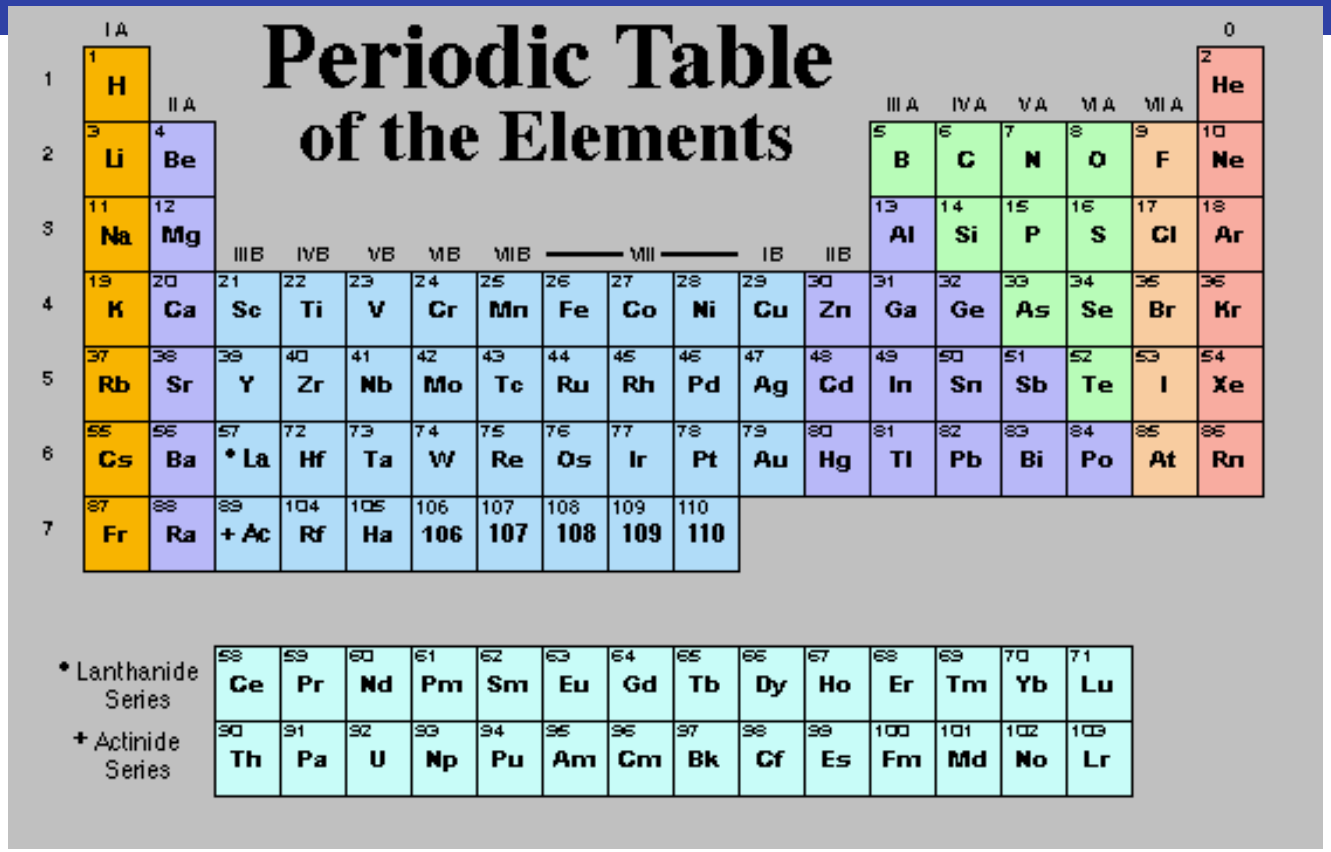
$L_{2,3}$-edges : 2p → 3d, 4d

$M_{4,5}$-edges : 3d → 4f

Absorption cross-section :

$w_{abs} = (2\pi/h) \; |< \Phi_f \,|T|\, \Phi_i >|^2 \; \rho_f(E_{h\nu} - E_i)$ Fermi 's Golden Rule

X-ray Absorption

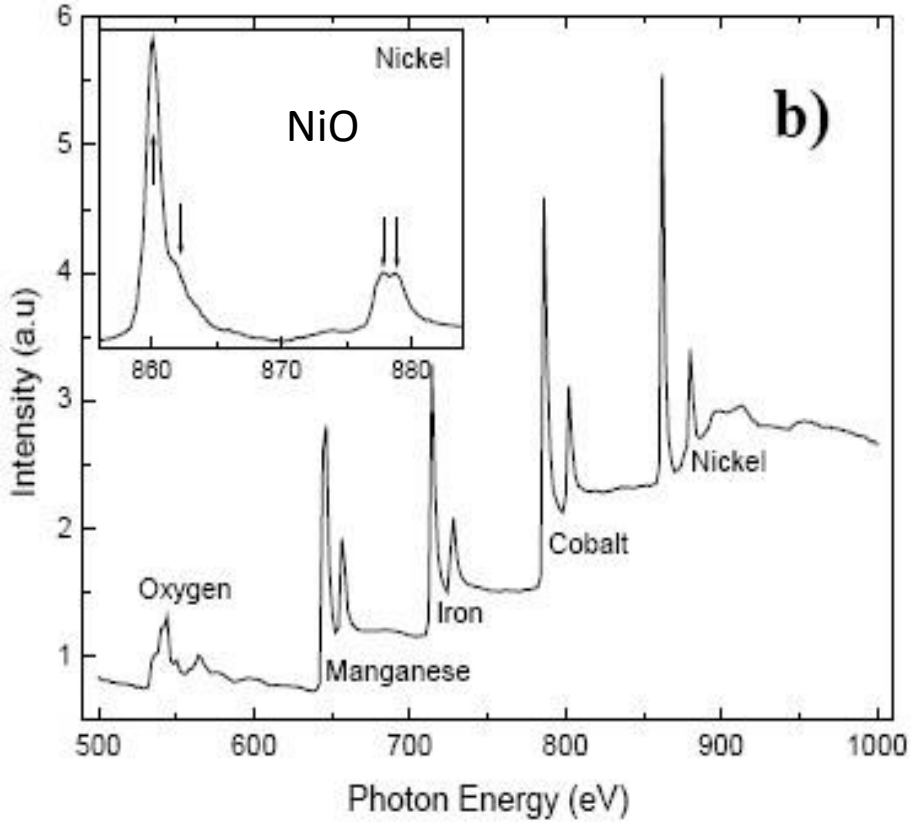


Main magnetic elements:

- Transition Metals (Mn, Fe, Co, Ni but also Pd, Pt,…) 2p → 3d (4d, 5d)
- Rare Earths (Nd, Gd, Tb, Dy, ..) 3d → 4f (also 2p → 5d)

X-ray Absorption

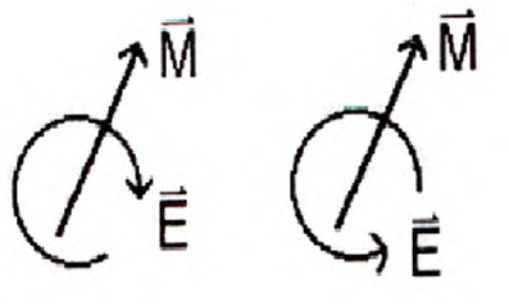
3d-transition metal $L_{2,3}$-edges



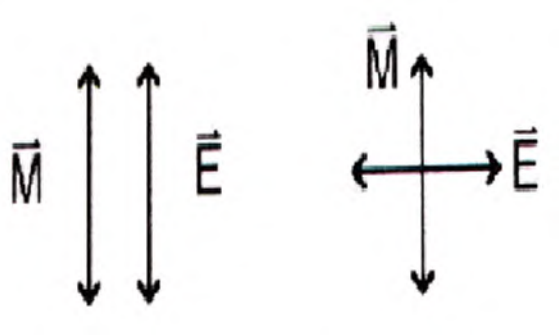
Element selective, information on electronic state

Magnetic sensitivity

X-ray Magnetic Circular Dichroism (XMCD): difference in absorption for left- and right circularly polarized light.

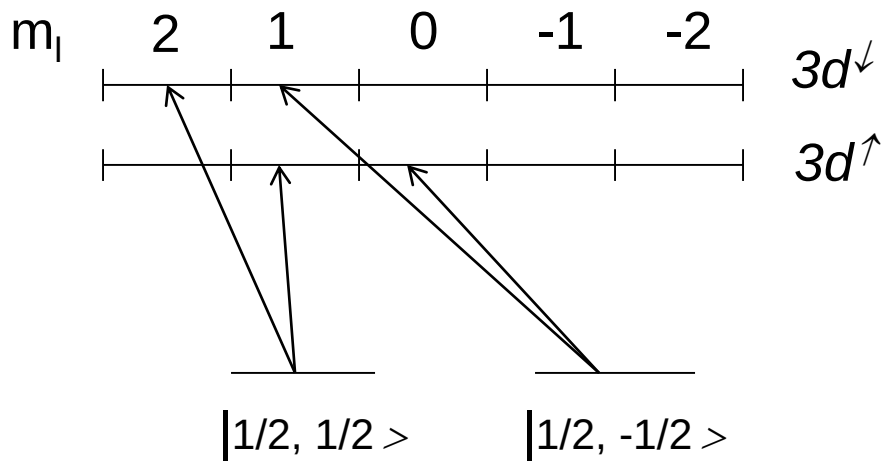


X-ray Linear Dichroism: difference in absorption for linearly polarized light ⊥ and // to quantization axis.



XMCD : origin

One electron picture: transitions from 2*p* to 3*d* band, split by exchange in $3d^{\uparrow}$ and $3d^{\downarrow}$



L_2 edge - left polarisation (Δm_l=+1)

$$I^{\uparrow} = \sum_{i,f} |\langle f|P_1|i\rangle|^2 = (1/3\,|\langle 2,1|P_1|1,0\rangle|^2 + 2/3\,|\langle 2,0|P_1|1,-1\rangle|^2)\,R^2$$

$$I^{\downarrow} = \sum_{i,f} |\langle f|P_1|i\rangle|^2 = (2/3\,|\langle 2,2|P_1|1,1\rangle|^2 + 1/3\,|\langle 2,1|P_1|1,0\rangle|^2)\,R^2$$

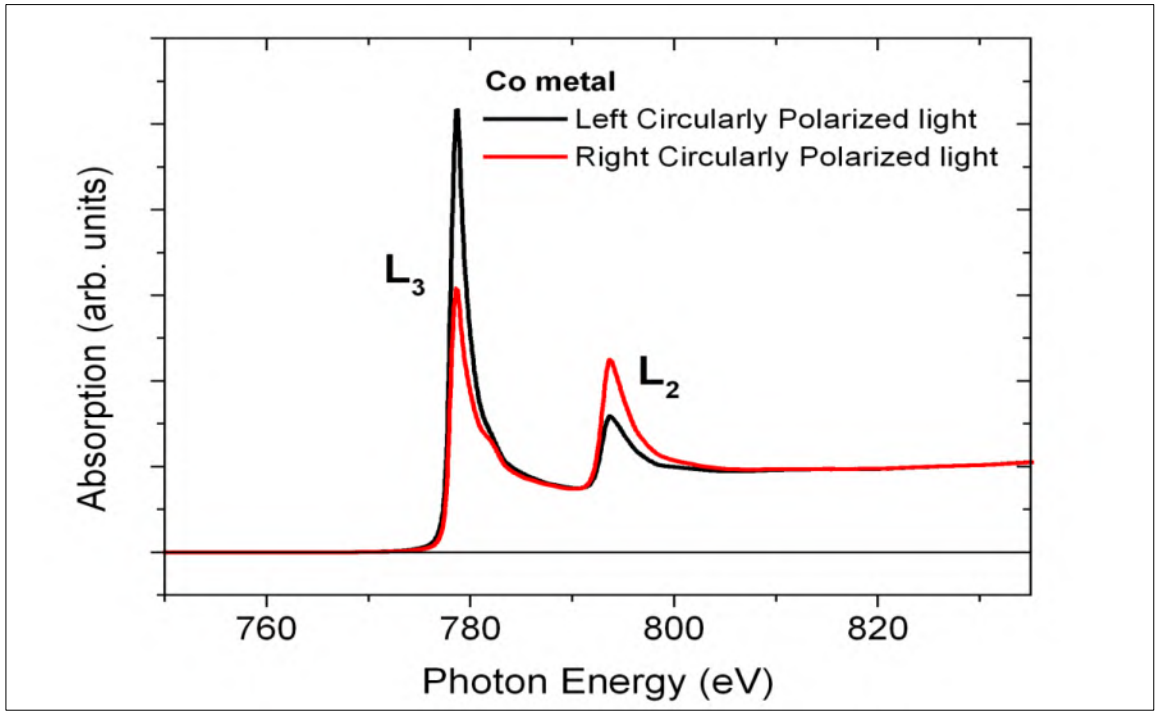
$$\mathbf{R}=\int R_{nl}^*(r)R_{n'l'}(r)\,r^3 dr$$

J.L. Erskine and E.A.Stern, Phys.Rev.B 12, 5016 (1975).

XMCD : origin

Step 1 : spin-polarised electrons emitted by the spin-orbit split 2p band
75% spin down and 25% spin up electrons at the **L_2**-edge with LCP light
37.5% spin down and 62.5% spin up electrons at the **L_3**-edge with LCP light

Step 2: the exchange split *d*-band acts as spin-detector.



Sum rules for x-ray dichroism

Sum rules relate dichroism and total absorption to the ground-state orbital and spin magnetic moment of the probed element and shell:

$L_{2,3}$-edges of Fe → Fe 3*d*-moments.

Orbital moment sum rule:

$$\langle L_Z \rangle = [2l(l+1)(4l+2-n)]/[l(l+1)+2 - c(c+1)] \bullet$$

$$[\int_{j+ + j-} d\omega\,(\mu^+ - \mu^-) / \int_{j+ + j-} d\omega\,(\mu^+ + \mu^- + \mu^0)]$$

l = orbital quantum number of the valence state
c = orbital quantum number of the core state
n = number of electrons in the valence state

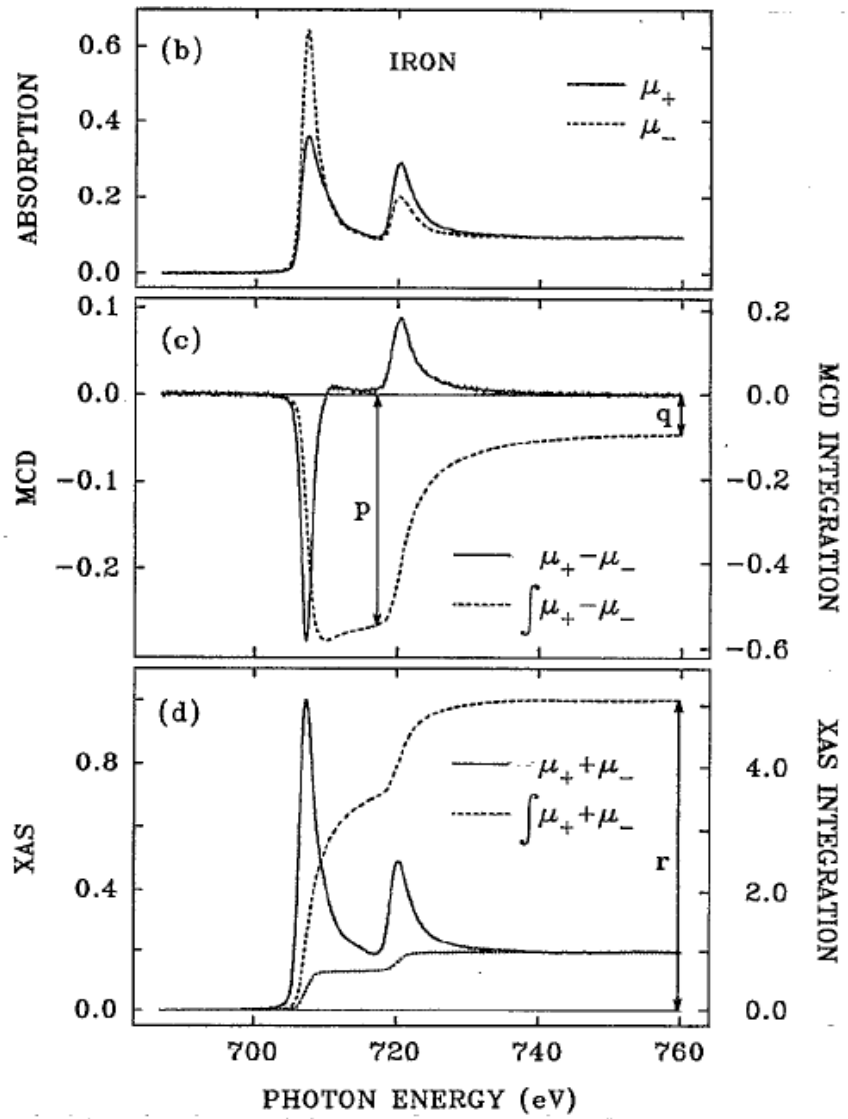
μ^+ (μ^-) = absorption spectrum for left (right) circularly polarized light.

μ^0 = absorption spectrum for linearly polarized light, with polarization parallel to quantization axis.

j^+ (j^-) = (l + 1/2) resp. (l - 1/2) absorption (ex. $2p_{3/2}$, $2p_{1/2}$)

B.T.Thole *et al.*, Phys.Rev.Lett. 68, 1943 (1992)
M.Altarelli, Phys.Rev.B 47, 597 (1993)

Sum rule for orbital moment



For $L_{2,3}$-edges $c = 1$ ($2p$), $l = 2$ (d):

$$\langle L_Z \rangle = 2(10-n) \bullet (\Delta L_3 + \Delta L_2) \Big/ \int_{L3+L2} d\omega\, (\mu^+ + \mu^- + \mu^0)]$$

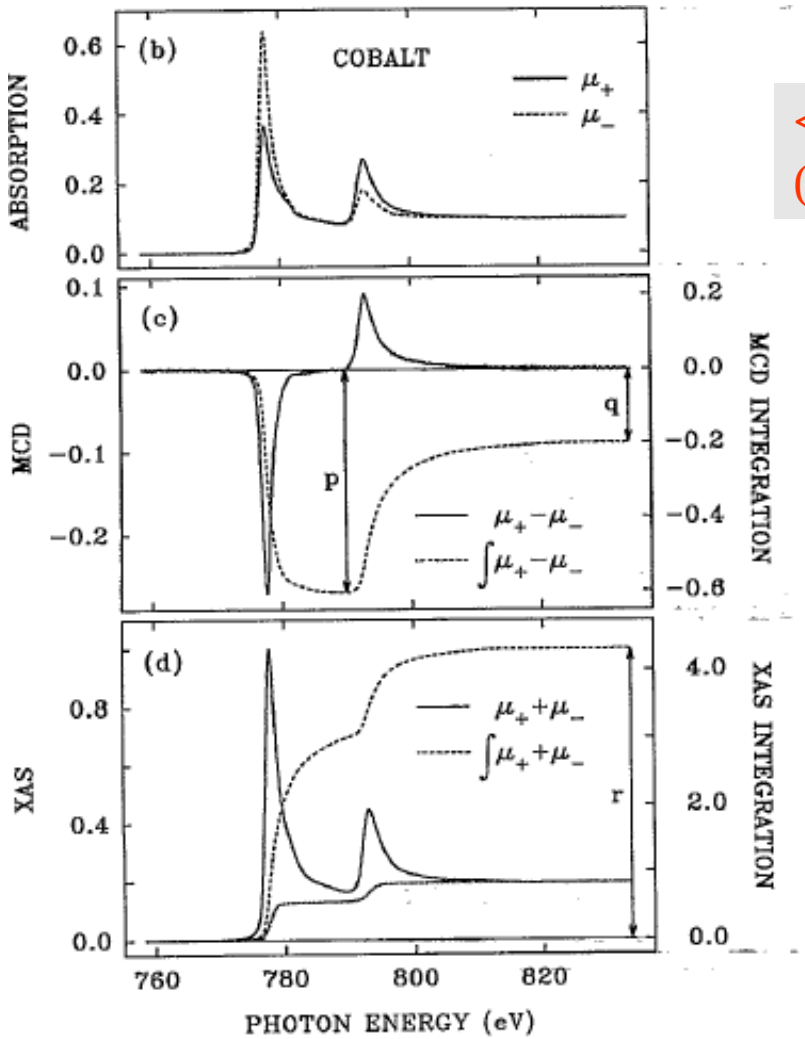
$$q = \Delta L_3 + \Delta L_2$$

$$r = \mu^+ + \mu^- = (2/3)(\mu^+ + \mu^- + \mu^0)$$

$$\langle L_Z \rangle = 4q\,(10-n) / 3r$$

C.T.Chen *et al.*, PRL 75, 152 (1995)

Sum rule for spin moment



$$\langle S_Z \rangle + (7/2) \langle T_Z \rangle = (3/2)(10-n)[(\Delta L3 - 2\Delta L2)/\int_{L3+L2} d\omega\,(\mu_+ + \mu_- + \mu_0)]$$

$$= (3/2)(10-n)(p - 2\,(q-p))/(3/2)r =$$

$$= (3p - 2q)(10-n)/r$$

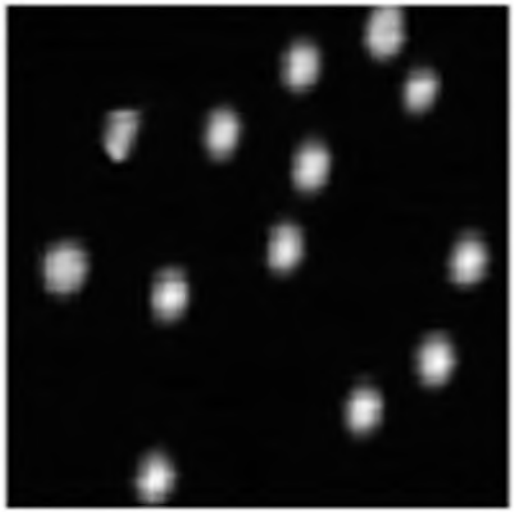
C.T.Chen *et al.*, PRL 75, 152 (1995)

XMCD sum rules : Orbital magnetic moments and anisotropy

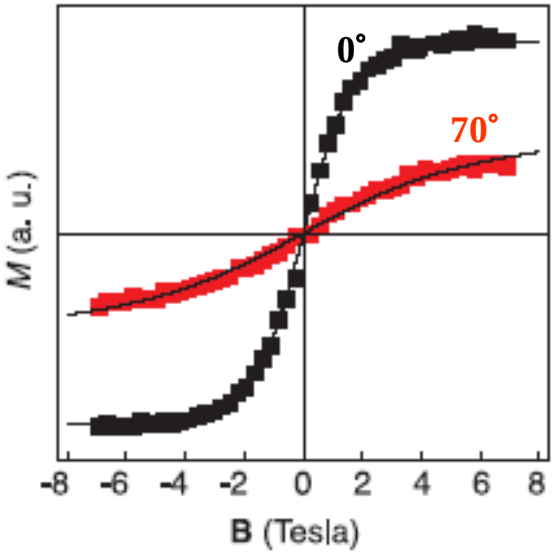
Single Co adatoms and particles MBE deposited on Pt(111) surfaces

P. Gambardella *et al.*, Science 300, 1130 (2003)

STM image of isolated Co adatoms (8.5 nm x 8.5 nm)



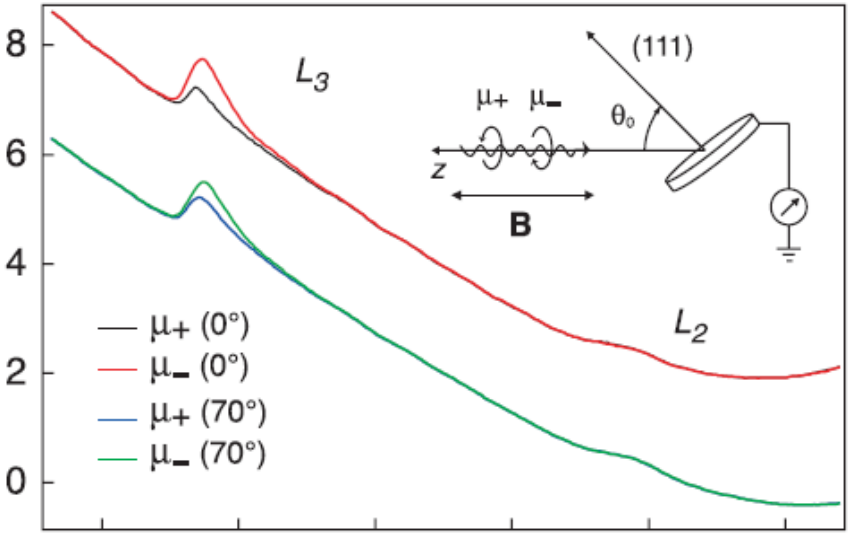
Large difference of in-plane and out-of-plane saturation field : very large Magnetic Anisotropy Energy



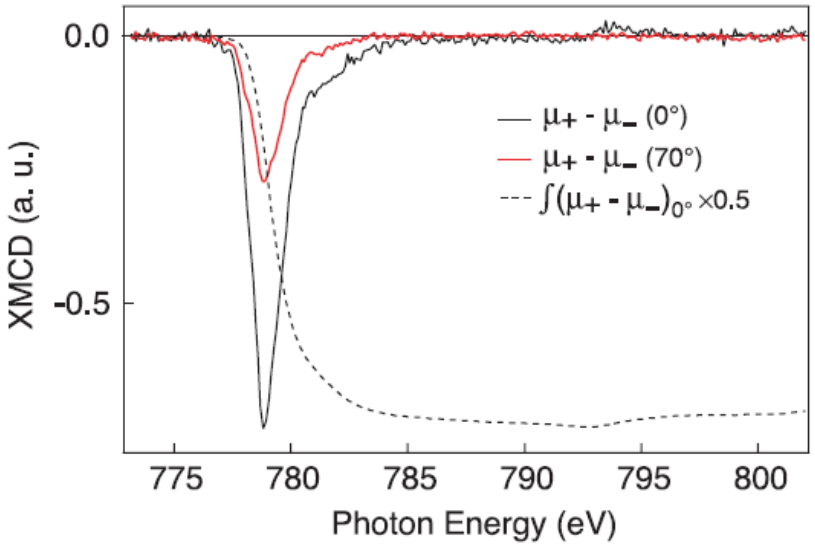
XMCD sum rules : Orbital magnetic moments and anisotropy

Single Co adatoms and particles MBE deposited on Pt(111) surfaces

P. Gambardella *et al.*, Science 300, 1130 (2003)



Co $L_{2,3}$ X-ray absorption spectra for $\theta = 0°$ and $70°$



vanishing L_2 XMCD: very large orbital magnetism

XMCD sum rules : Orbital magnetic moments and anisotropy

Single Co adatoms and particles MBE deposited on Pt(111) surfaces

P. Gambardella *et al.*, Science 300, 1130 (2003)

Sum rules : $\langle L \rangle = 1.1 \pm 0.1\ \mu_B$ for isolated Co adatoms ($L = 0.15\ \mu_B$ Co-hcp)
($L = 0.29\ \mu_B$ 1ML Co/Pt)

Reduced coordination of isolated atoms on top of a flat surface → d-electron localisation, increase of atomic character

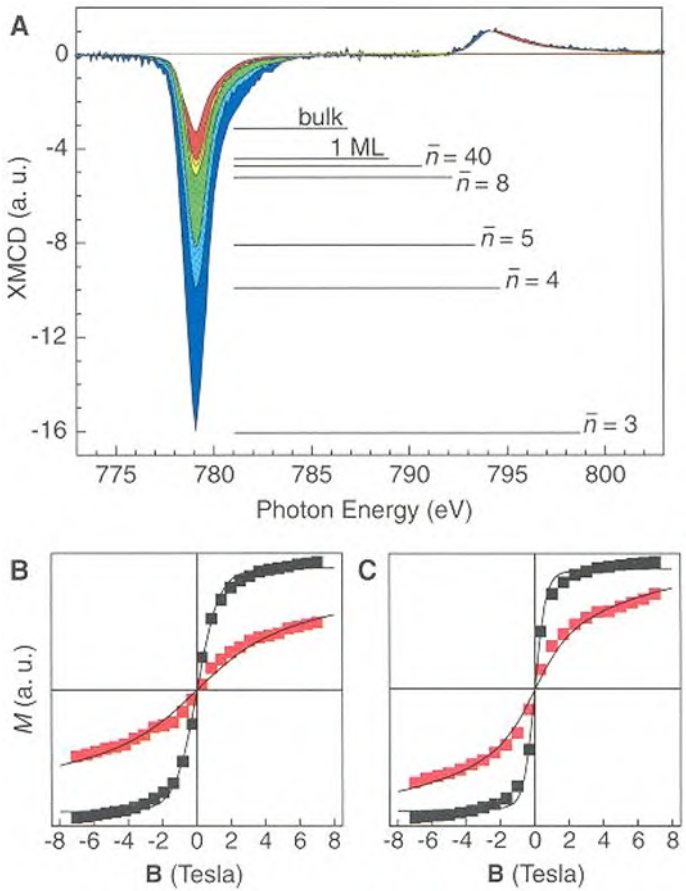
From element-selective XMCD magnetization curves (up to 7 Tesla):
very large magnetic anisotropy energy (MAE)

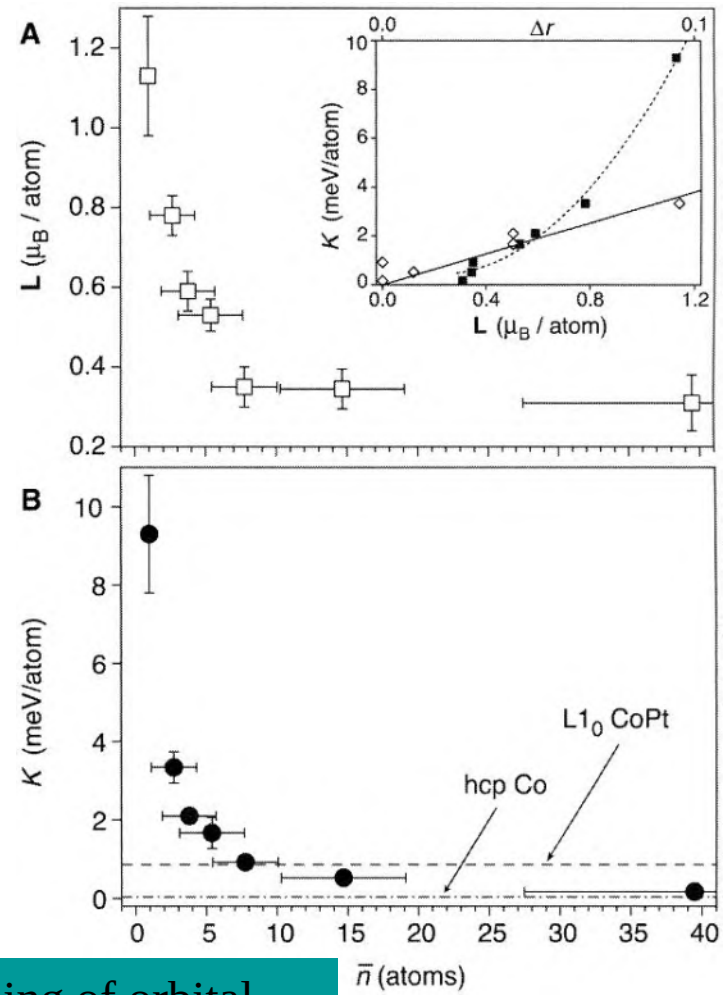
$K = 9.3 \pm 1.6$ meV/atom

($K = 1.8$ meV/Co atom in $SmCo_5$)
($K = 0.3$ meV/atom in Pt/Co multilayers)

XMCD sum rules : Orbital magnetic moments and anisotropy

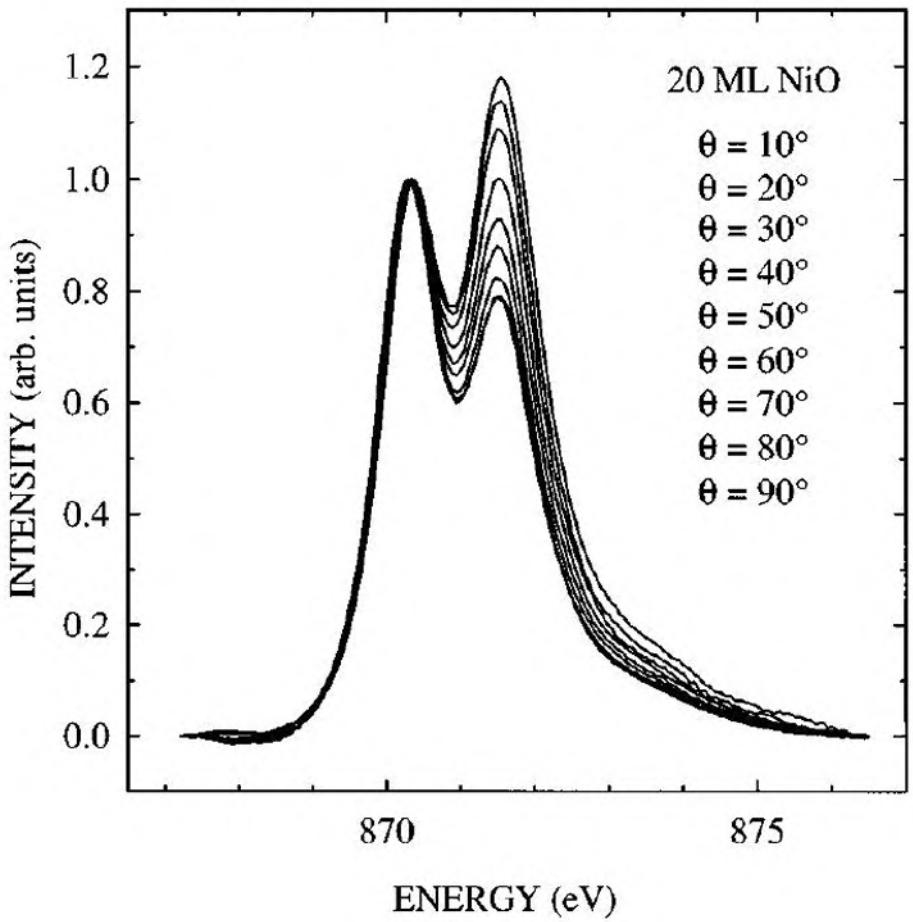
P. Gambardella *et al.*, Science 300, 1130 (2003)





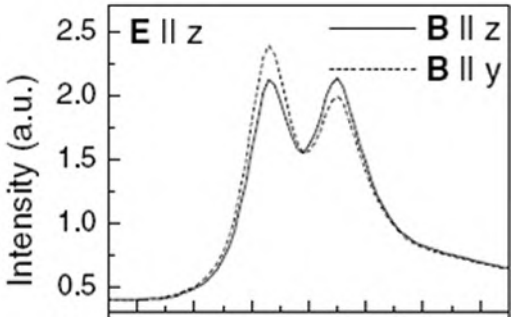
Increase of particle size : progressive quenching of orbital moment and consequent decrease of MAE

X-ray Linear Dichroism



Dependence of Ni L2 spectrum in NiO on the incidence angle of linearly polarized light
D. Alders et al., Phys. Rev. B 57, 11623 (1998).

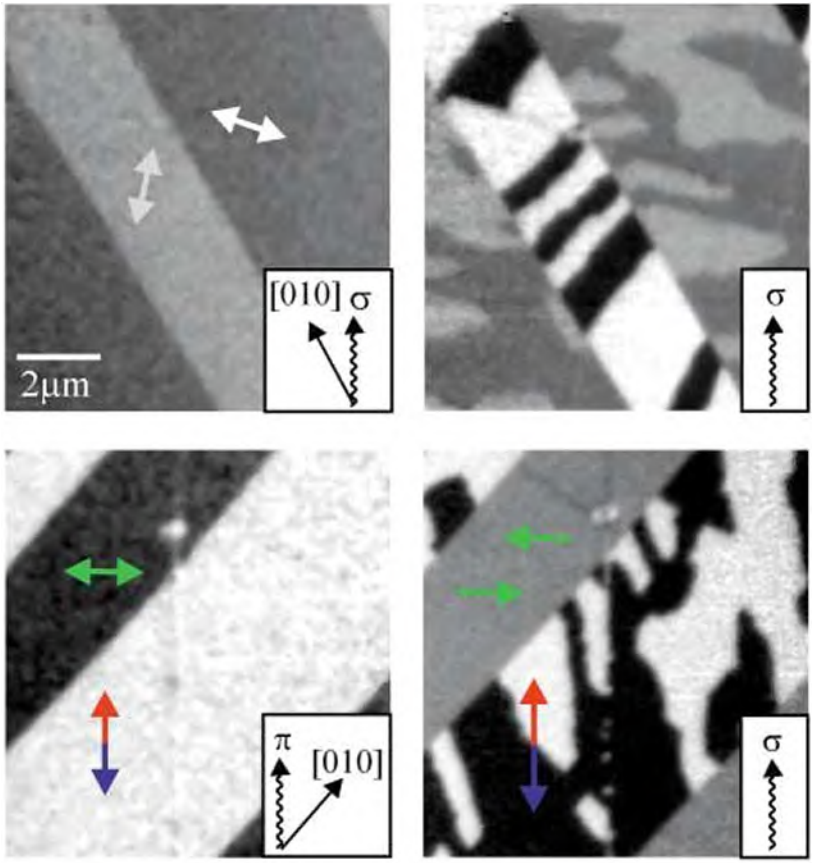
X-ray Linear Dichroism



Ni L_2-edge

Ni XMLD

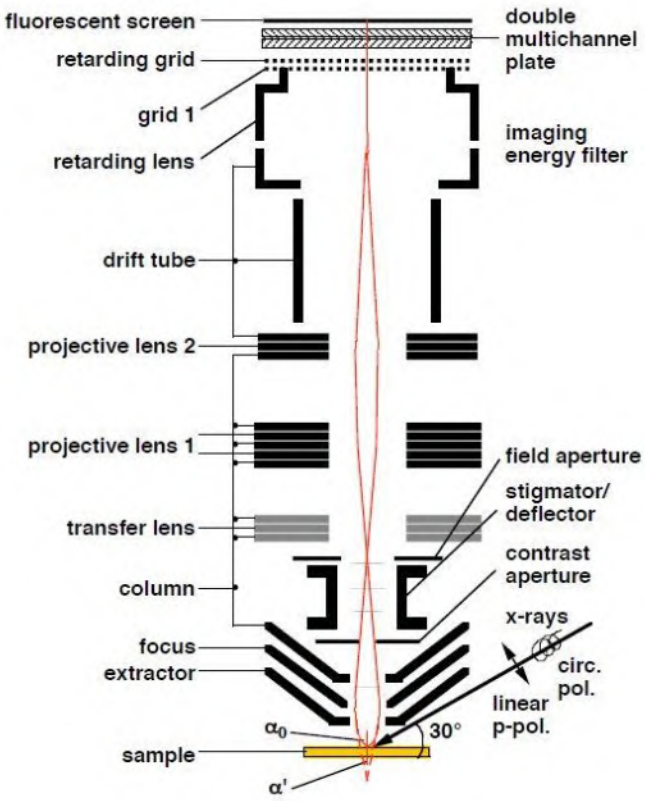
Co XMCD

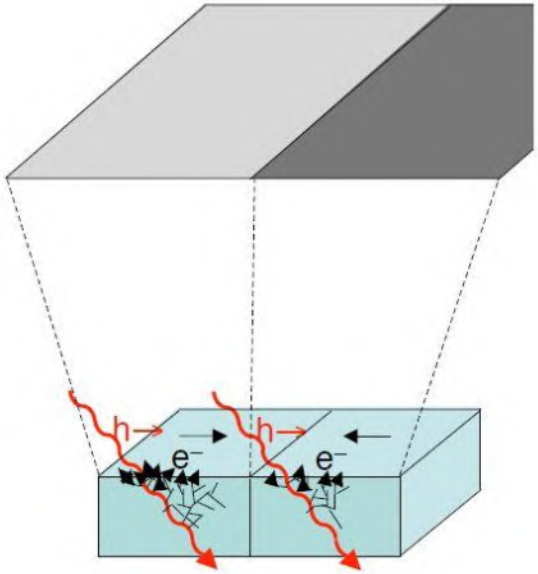


8 ML Co/NiO(001)

H. Ohldag et al., PRL 86 (2001) 2878

Magnetic Imaging using X-rays



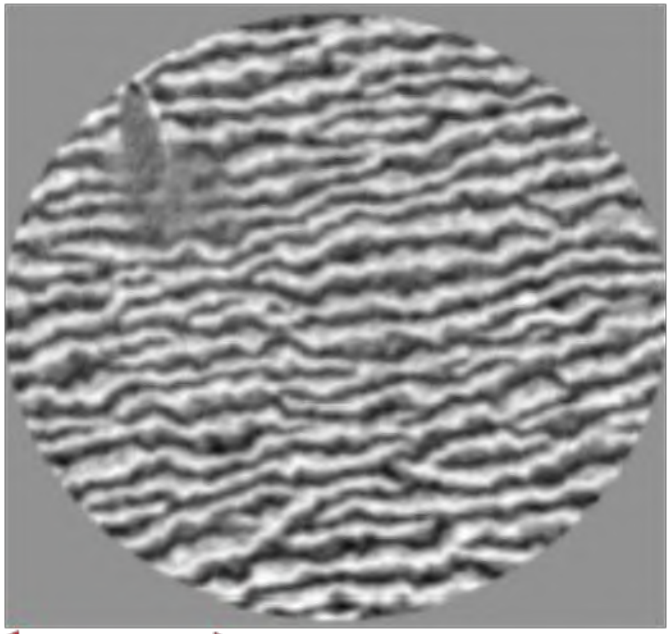


© W. Kuch 2009

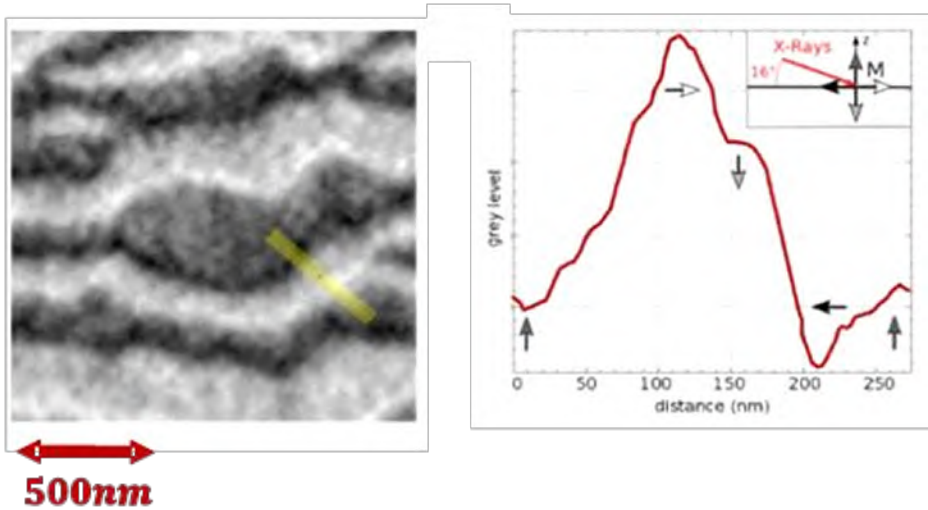
PhotoEmission Electron Microscope + XMCD

W(110)/Co 4ML (0.8nm)/$Au_{0.67}Pt_{0.33}$

XMCD-PEEM: 4 ML Co



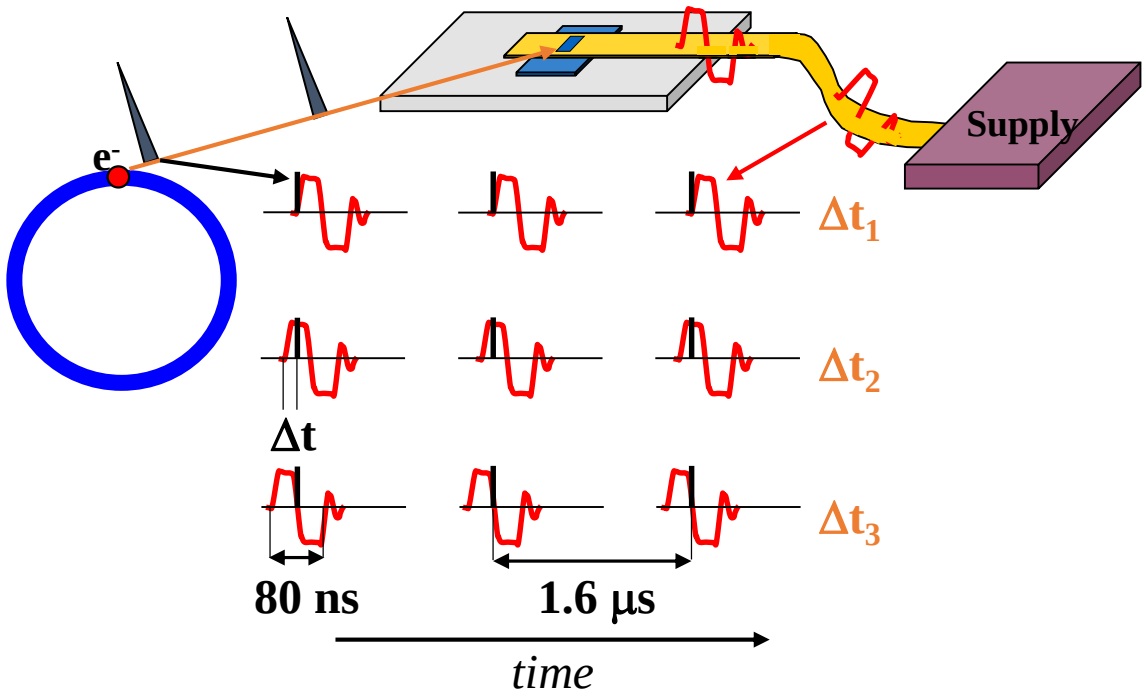
2 μm



L. Camosi, J. Peña Garcia et al., New. J. Phys. 23, 013020 (2021)

Time-resolved XMCD

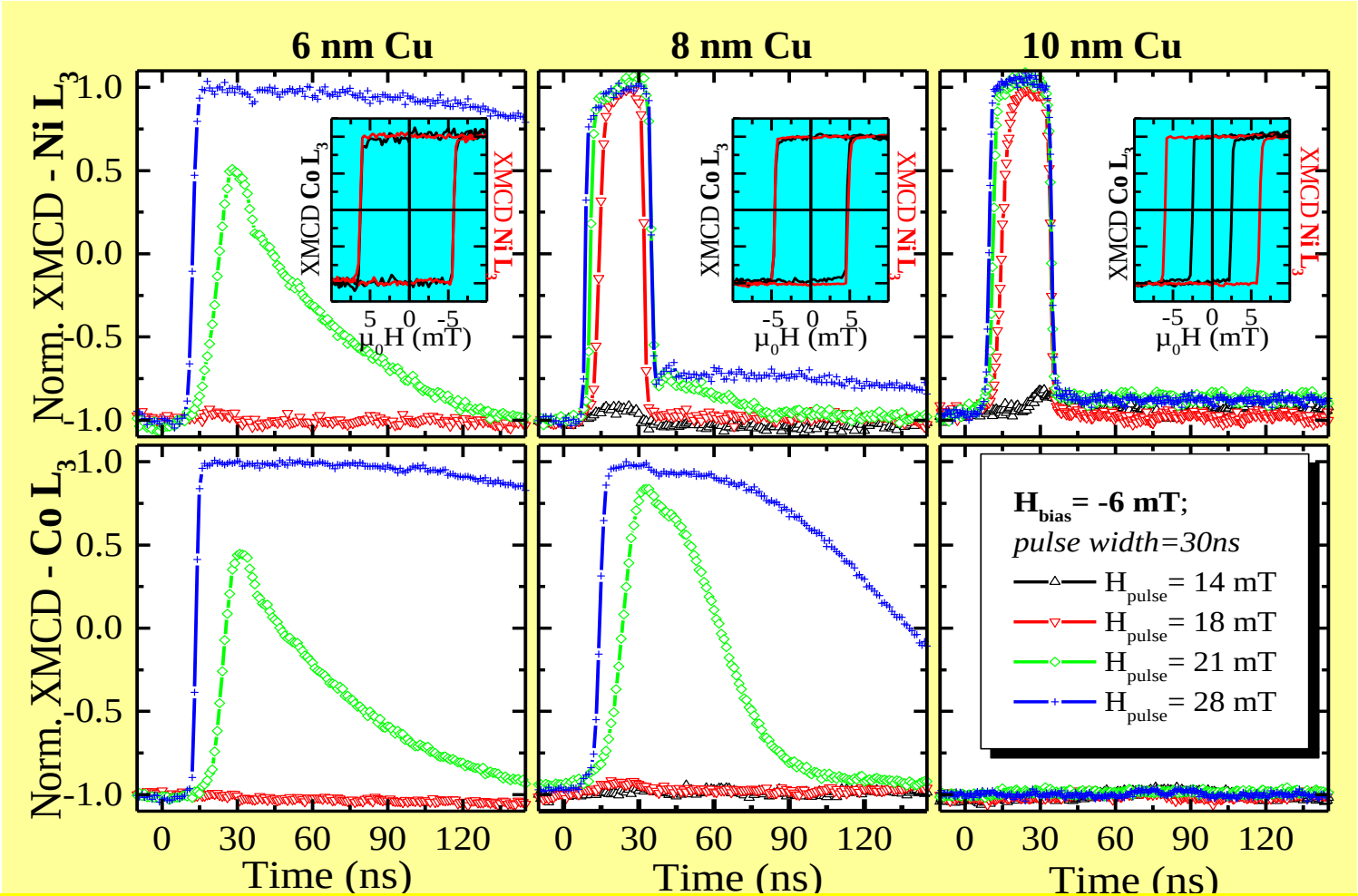
Pump-probe measurements with pulsed x-ray bunches



M. Bonfim et al., Phys. Rev. Lett. 86, 3646 (2001); J. Vogel et al., Appl. Phys. Lett. 82, 2299 (2003)

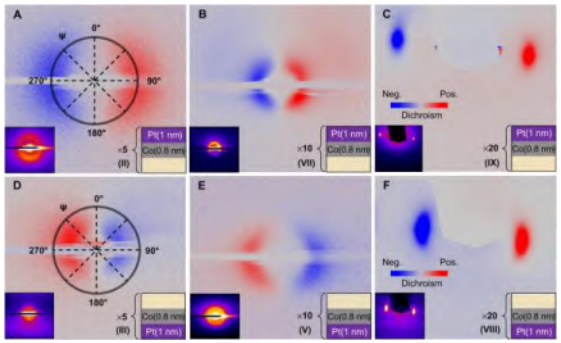
Time-resolved XMCD

FeNi/Cu/Co



The coupling between the soft and hard layers in the spin-valve systems is different in the static and dynamic regimes

Other x-ray techniques

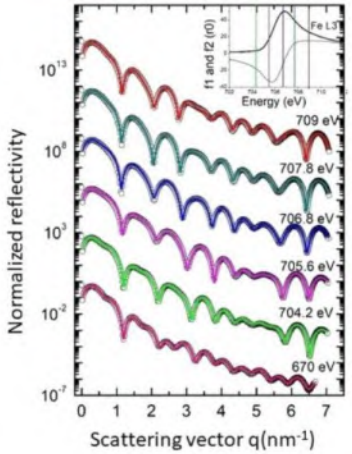


X-ray resonant magnetic scattering : periodic magnetic structures, chirality

W. Legrand et al., Sci. Adv. 2018;4:eaat041

X-ray resonant magnetic reflectivity : depth resolved magnetization profile

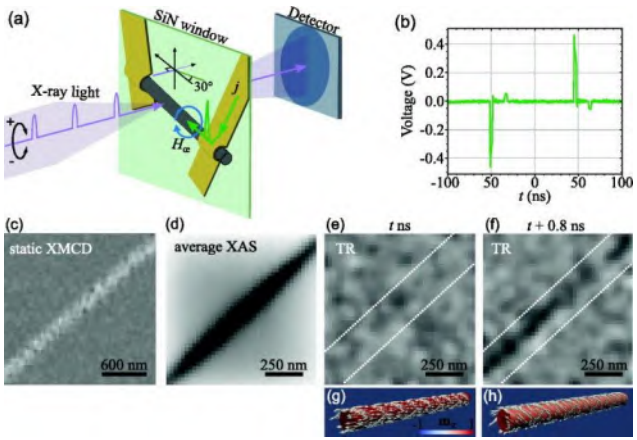
J.-M. Tonnerre et al., AIP Adv. 12, 035129 (2022).

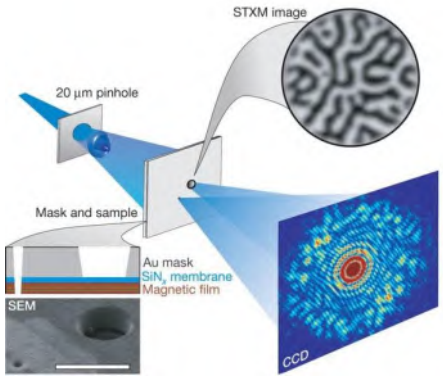


Other x-ray techniques

Scanning Transmission X-ray Microscopy

O. Fruchart et al., APL 2022



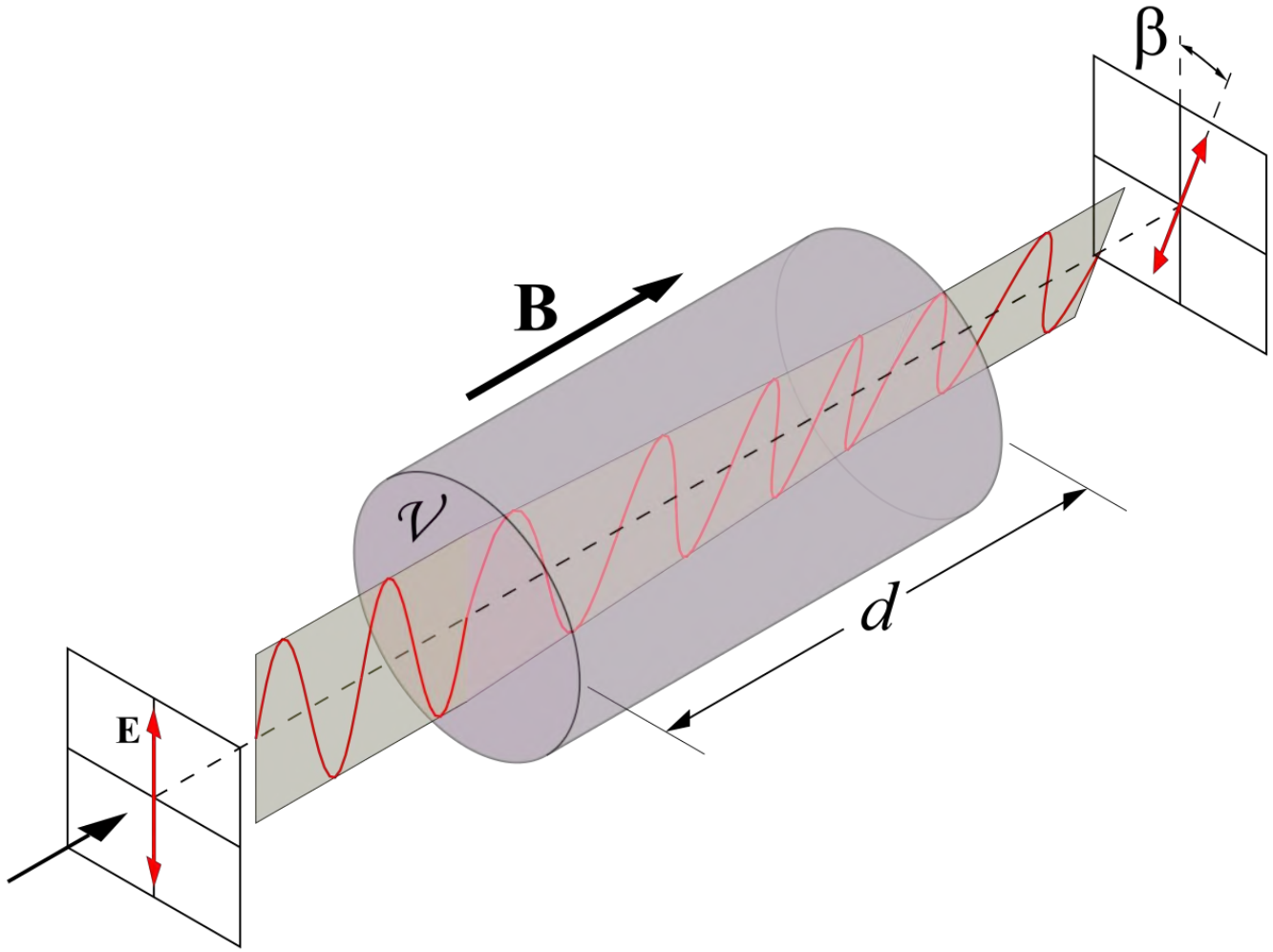


Magnetic X-ray Holography

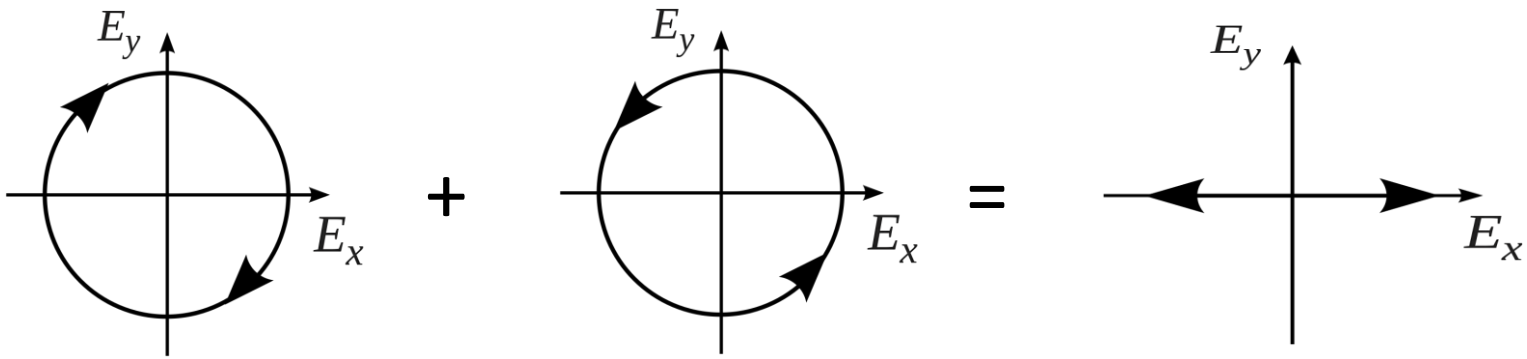
S.Eisebitt et al., Nature **432**, 885 (2004)

Magneto-optics

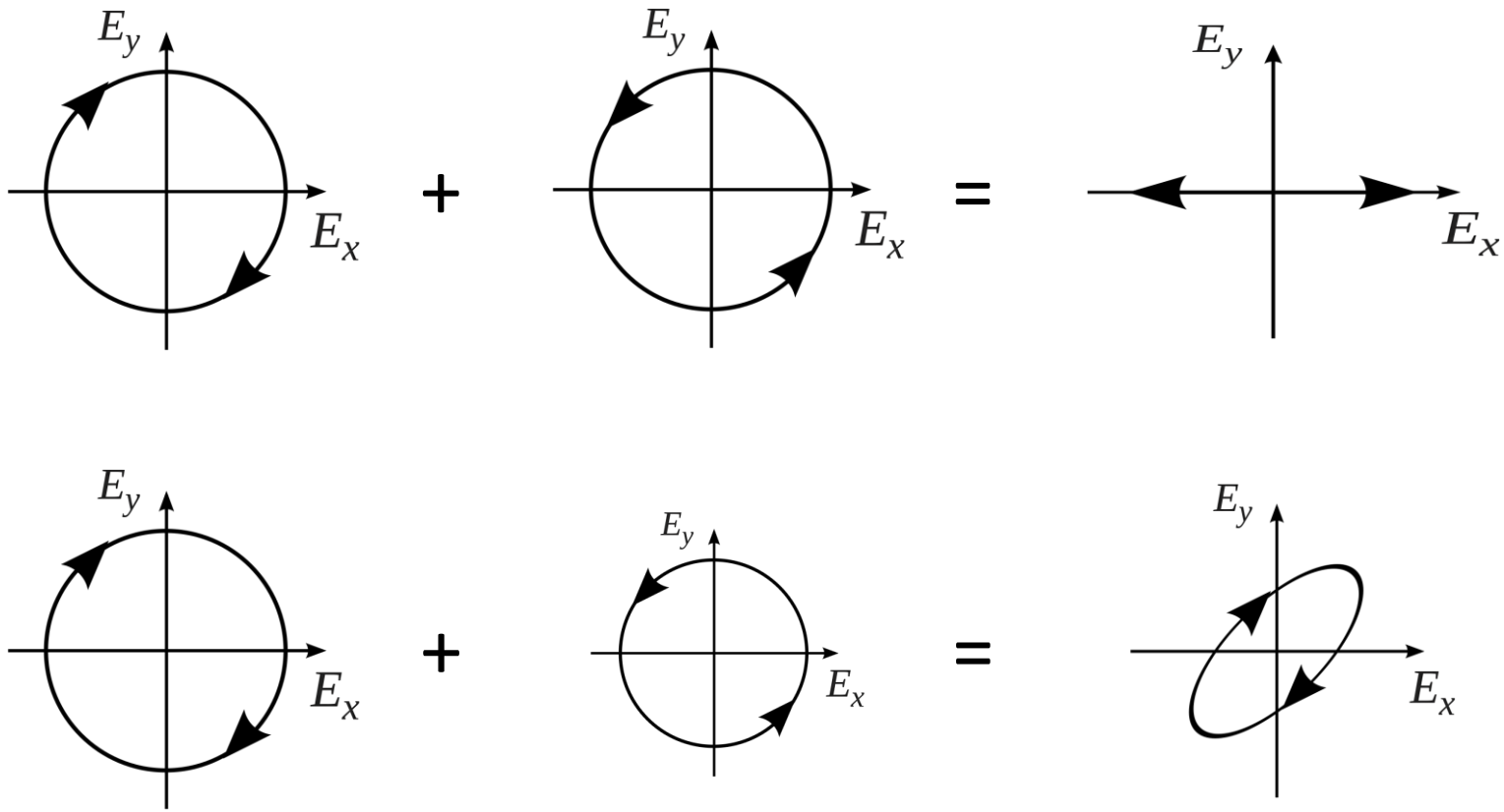
Faraday effect



Light polarization

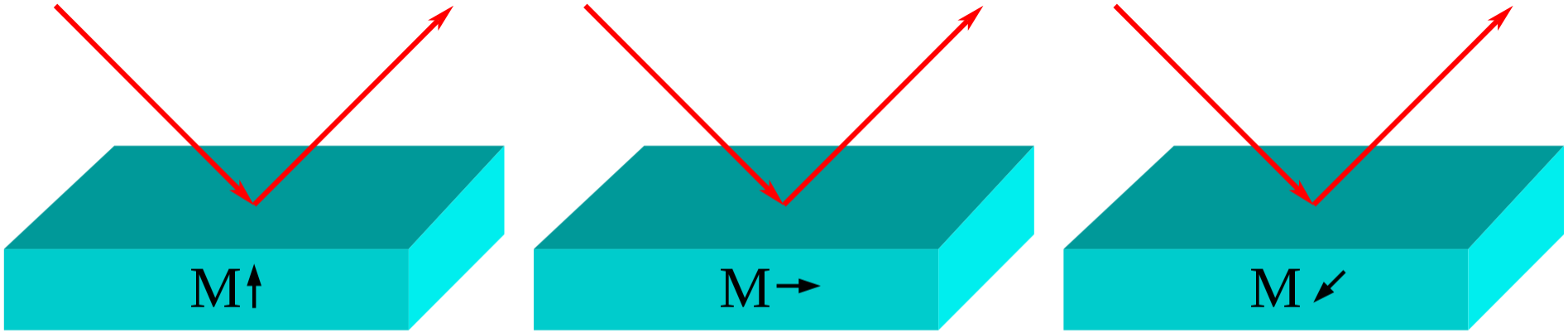


Light polarization



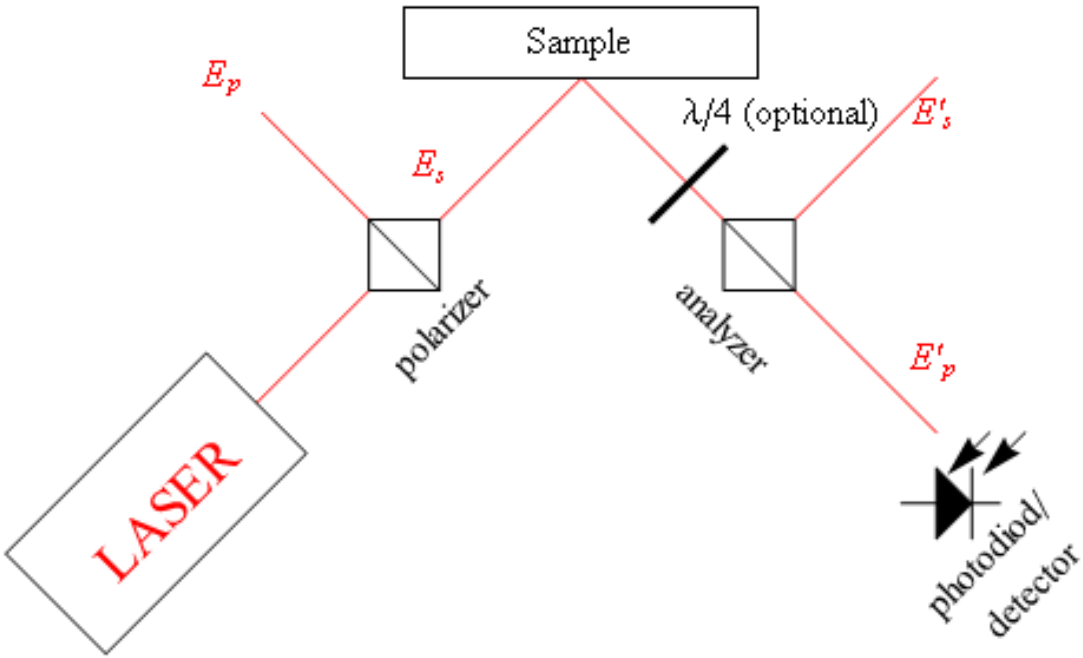
Different absorption left- and right circularly polarized light → ellipticity + rotation polarization axis if different phases

Magneto-optical Kerr effect (MOKE)

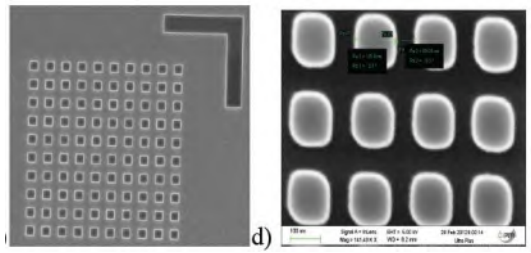


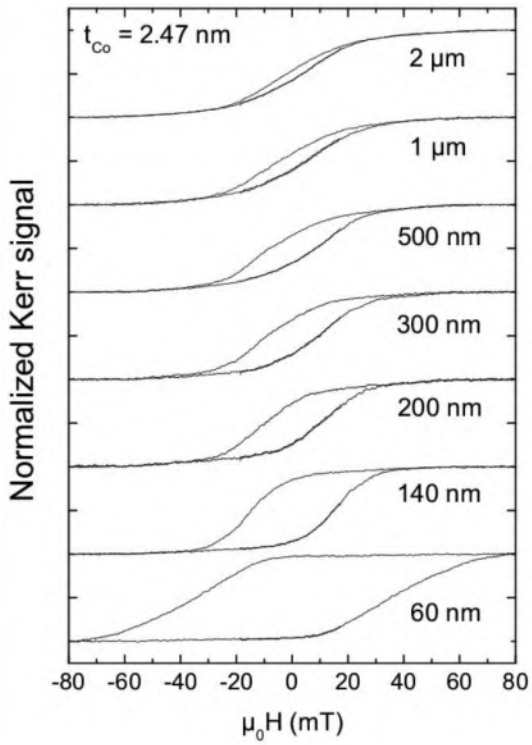
Polar | Longitudinal | Transversal MOKE

Kerr

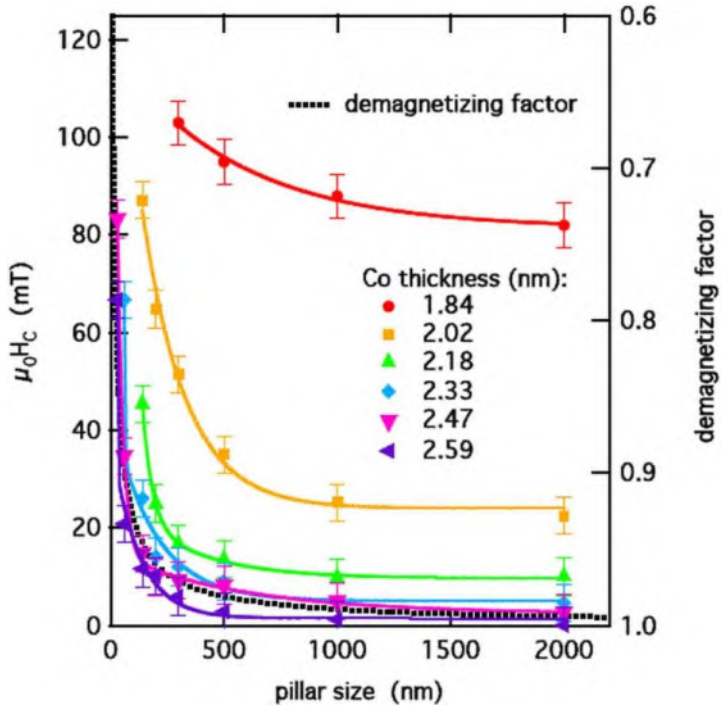

Sample
E_p
E_s
λ/4 (optional)
E'_s
E'_p
polarizer
analyzer
LASER
photodiod/
detector

Focused Kerr





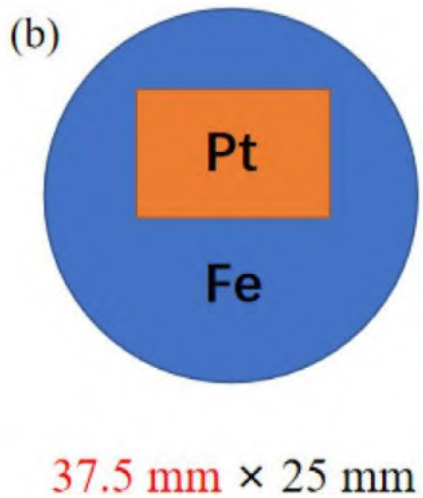
Pt/Co/MgO pillars with different size and Co thickness

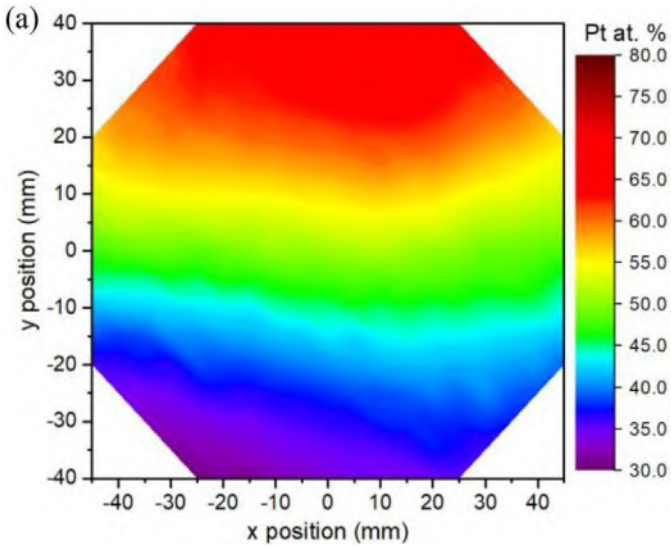


W. Stefanowicz et al., Appl. Phys. Lett. 104, 012404 (2014)

Scanning Kerr

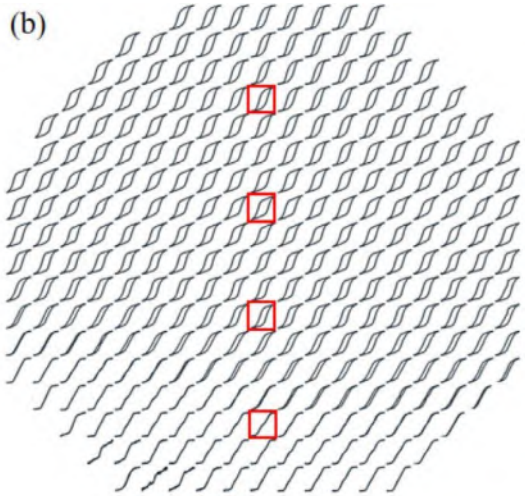
A high throughput study of compositionally graded FePt thin films using scanning MOKE



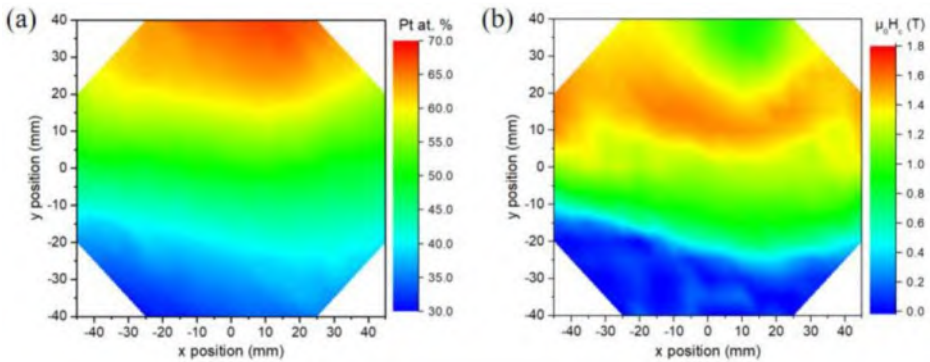


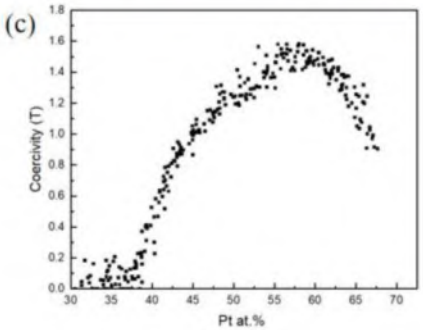
Y. Hong et al., J. Mater. Res. and Techn. 18, 1245 (2022).

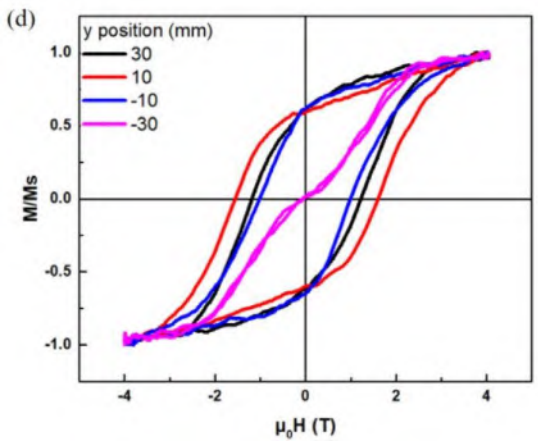
Scanning Kerr



Automatic scanning over 100mm diameter wafer with field pulses up to 3T





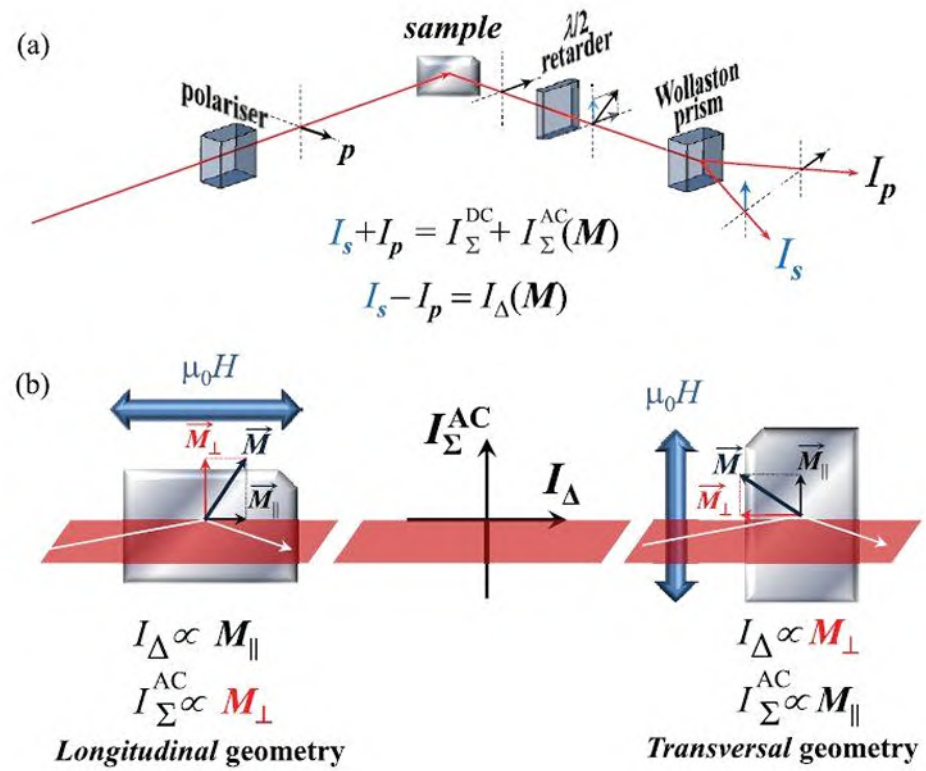


Y. Hong et al., J. Mater. Res. and Techn. 18, 1245 (2022).

Vectoriel Kerr

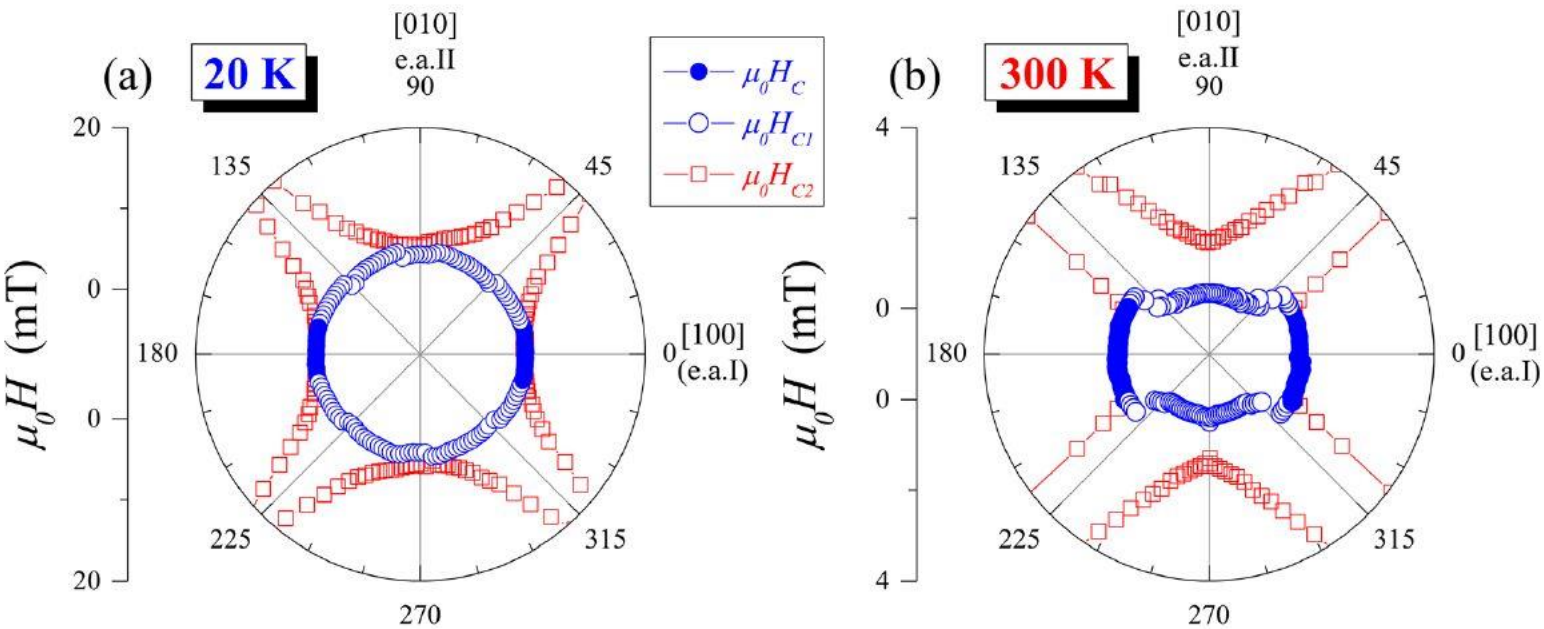
Measuring at the same time longitudinal and tranverse magneto-optical Kerr effect : vectoriel resolved MOKE magnetometry

Jose Luis F Cuñado, Javier Pedrosa, Fernando Ajejas, Paolo Perna, Rodolfo Miranda and Julio Camarero, J. Phys.: Condens. Matter **29** (2017) 405805



Vectoriel Kerr

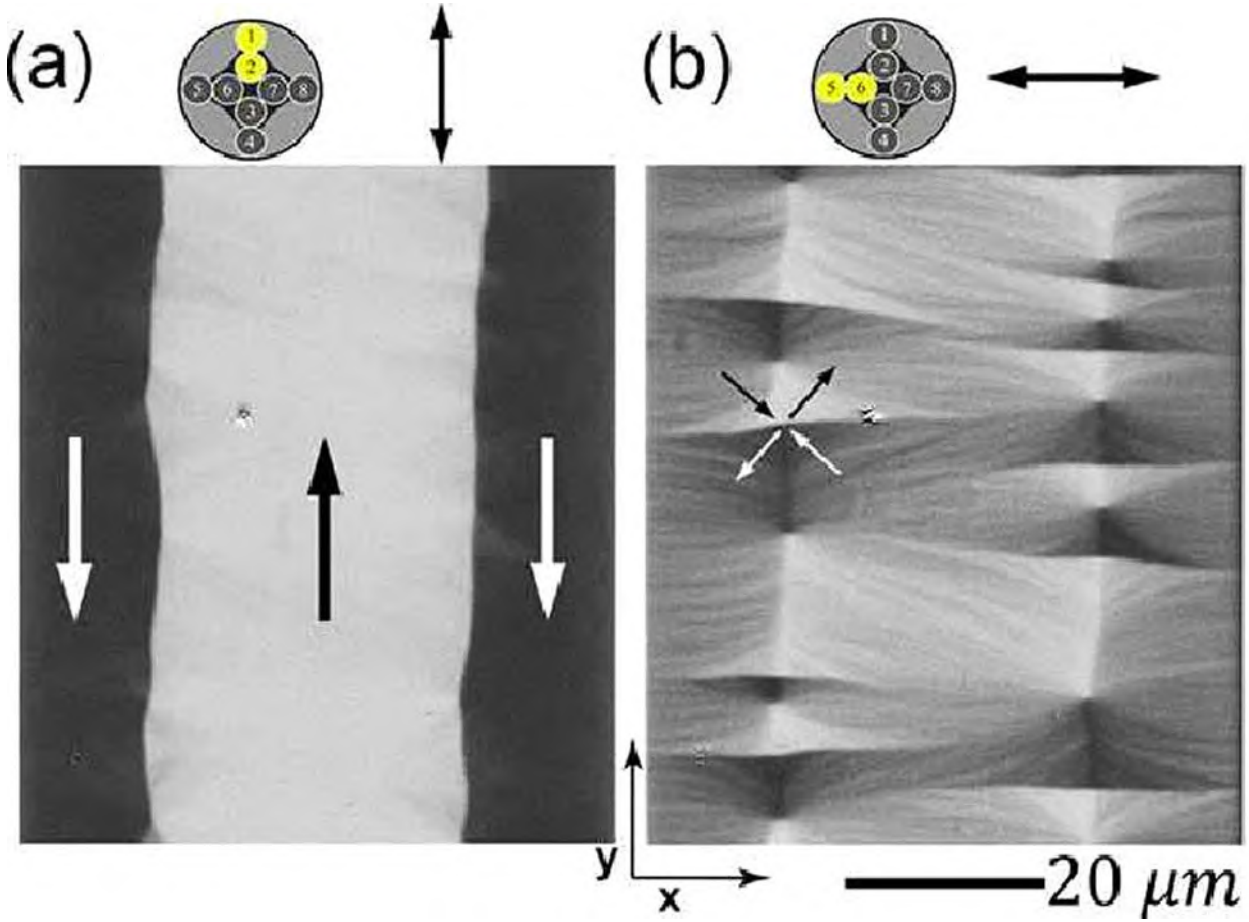
Fe(100) : fourfold in-plane symmetry



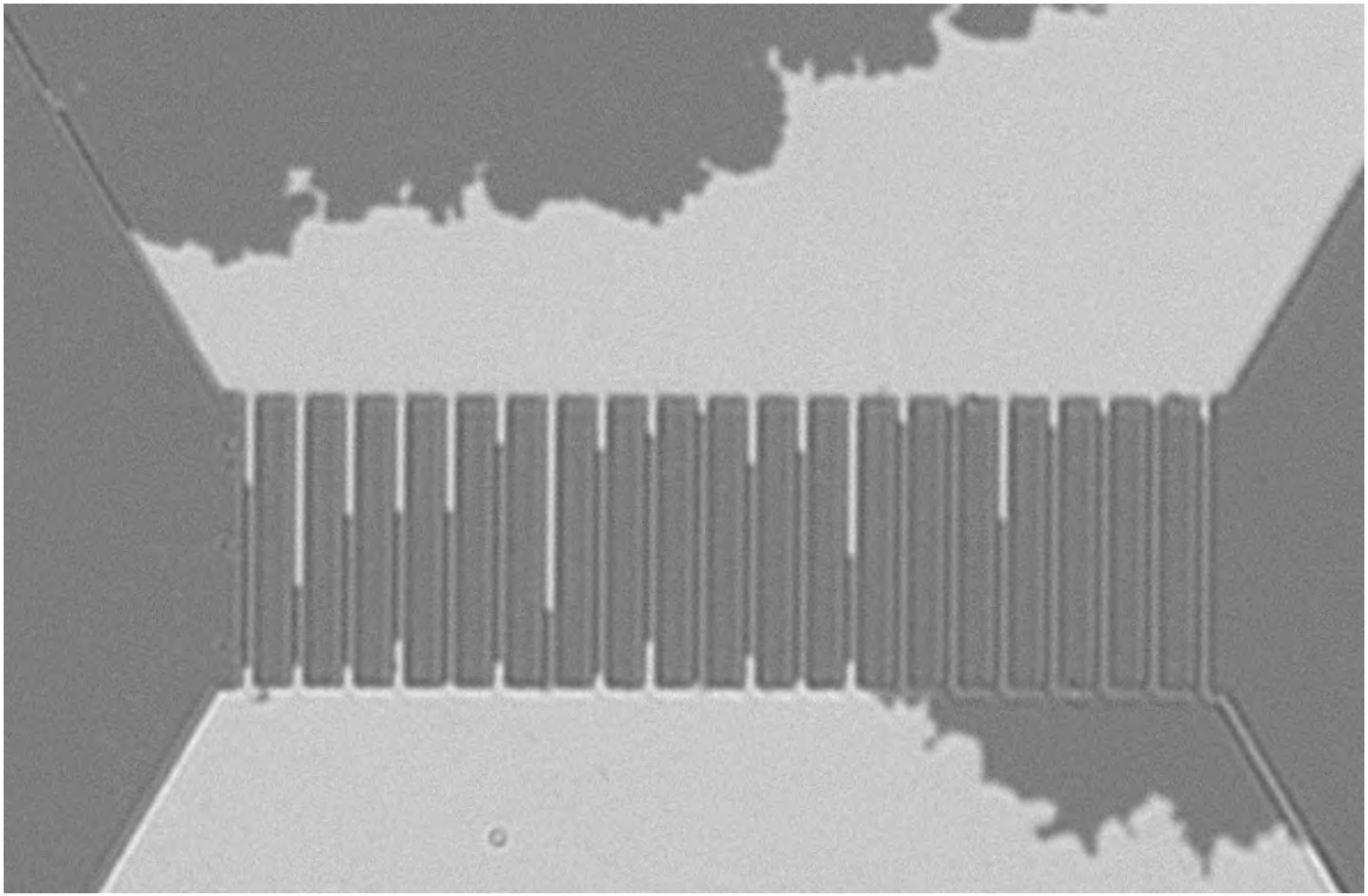
Practical by Jose Luis Coñado

Kerr microscopy

Magnetic Domains : The Analysis of Magnetic Microstructures
A. Hubert and R. Schäfer (Springer)

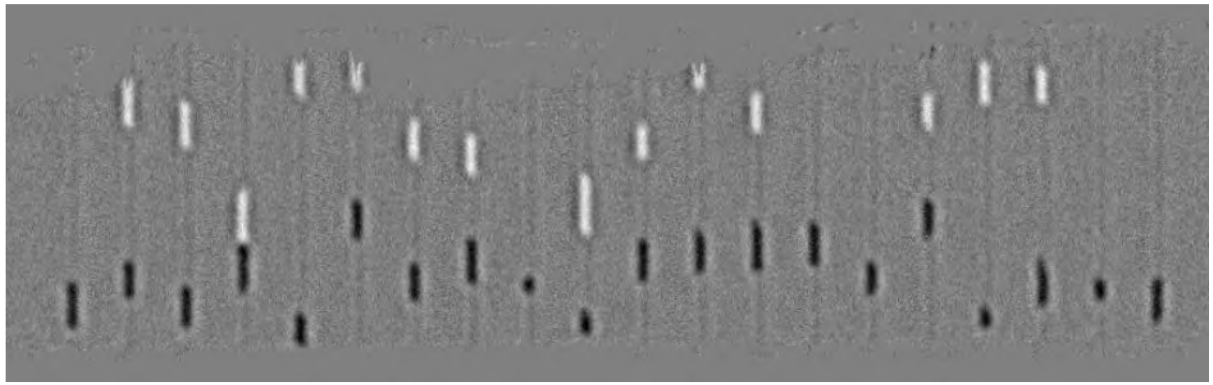


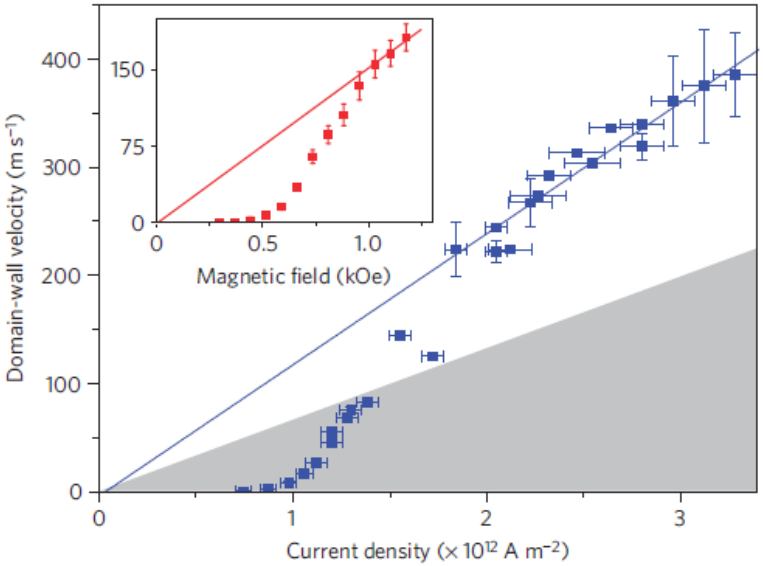
Kerr microscopy



Magnetic domain walls in Pt/Co/AlOx with perpendicular anisotropy

Kerr microscopy





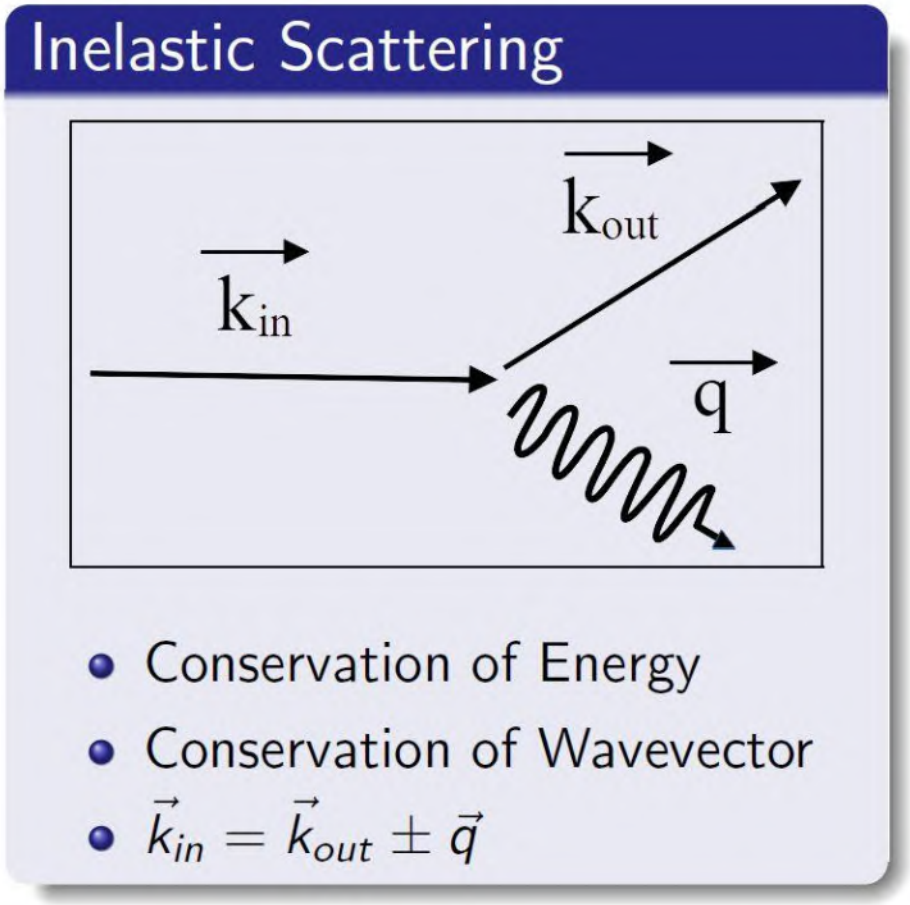
Fast current-induced domain wall motion by spin-orbit torque + Dzyaloshinskii-Moriya interaction

I.M. Miron et al., Nature Mater. **10**, 419 (2011).

Probing magnetic excitations (magnons) with light

Brillouin Light Scattering

Photon IN – Photon OUT : scattering by Phonons and Magnons



$$k = 4\frac{\pi}{\lambda}$$

Visible light (HeNe – laser) :

$\lambda = 532$ nm → $k = 23.6\ \mu m^{-1}$

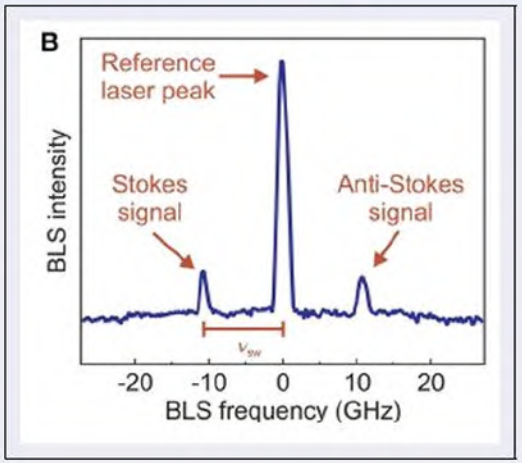
$\nu = 563$ THz

©Laurent Ranno

BLS : spectrometry of the scattered light around the elastic line

BLS

Inelastic Scattering



- Elastic mode : Rayleigh scattering
- Emission of excitation : Stokes mode
- Absorption of excitation : Anti-Stokes mode

E(q) dispersion

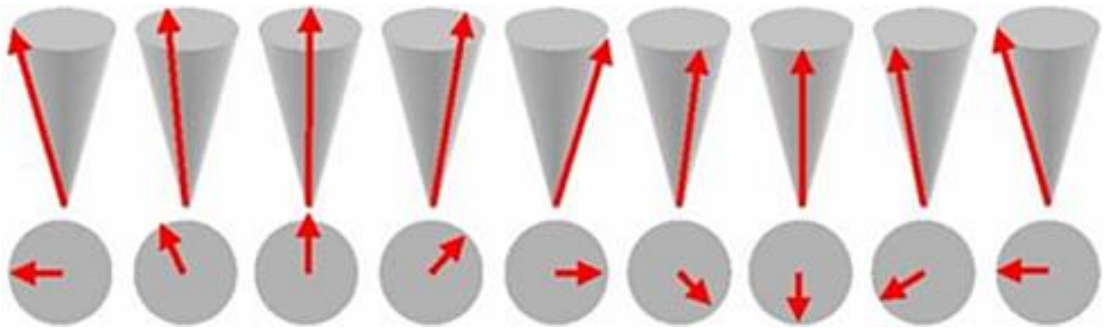
- Acoustic Phonons : $E = \hbar c q$
- Magnons : $E = Dq^2$
- Dispersion E(q) by rotating the sample
- Need for stable and high resolution spectrometer

E(q) $\Longrightarrow$ Sound velocity, Mechanical properties (contactless)
E(q) $\Longrightarrow$ A_{ex}, DMI, K_{anis}, M_s, relaxation, excitation distribution
...

©Laurent Ranno

Brillouin Light Scattering

Spin waves (magnons)



Frequency (energy) of magnons depends on M_S, H, K, $A_{ex,}$ DMI…

Lecture by Oksana Chubykalo-Fesenko on Friday

BLS

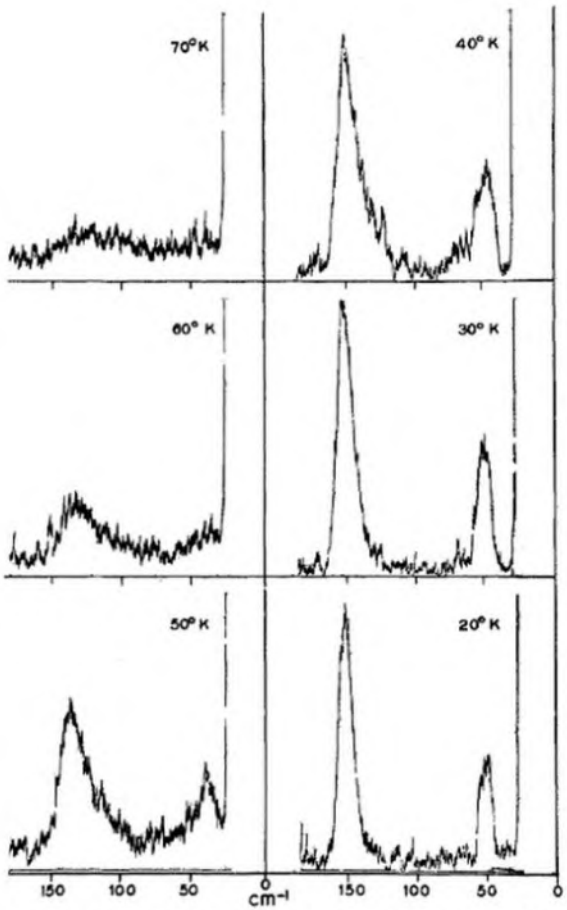


FIG. 1. Recorder traces showing frequency shifts of Stokes scattered light in the (zy) experimental geometry for various temperatures in FeF_2. The lines at ~52 and ~154 cm^{-1} are due to photons scattered by one and two magnons, respectively.

First BLS measurement on magnons

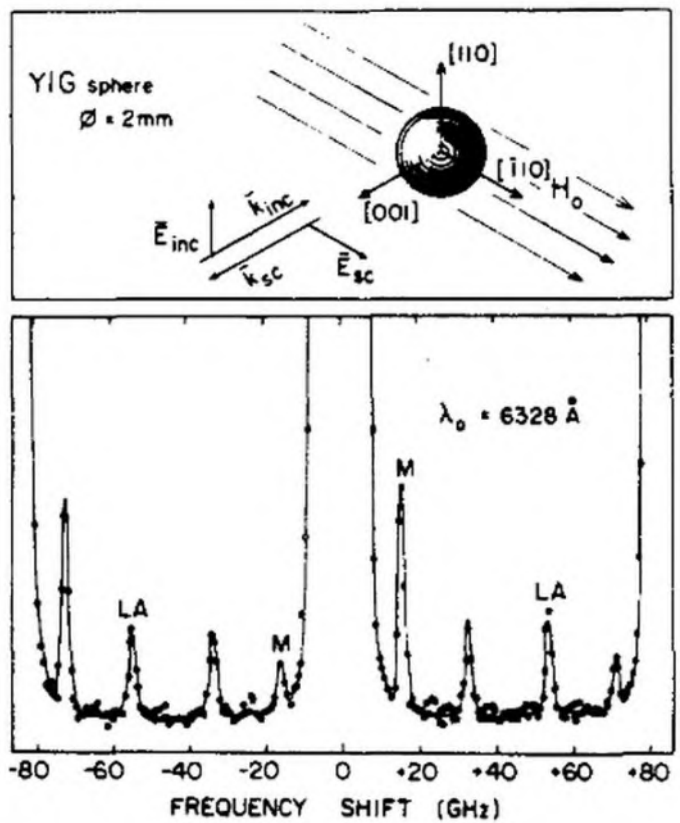
FeF_2

- Néel Temp = 78K
- Polarisation analyses
- Scattering mechanism : one-magnon process (52 cm^{-1})
- Scattering mechanism : 2-magnon process (154 cm^{-1})

P.A. Fleury et al, Phys. Rev. Lett. 18, 84 (1966)

BLS

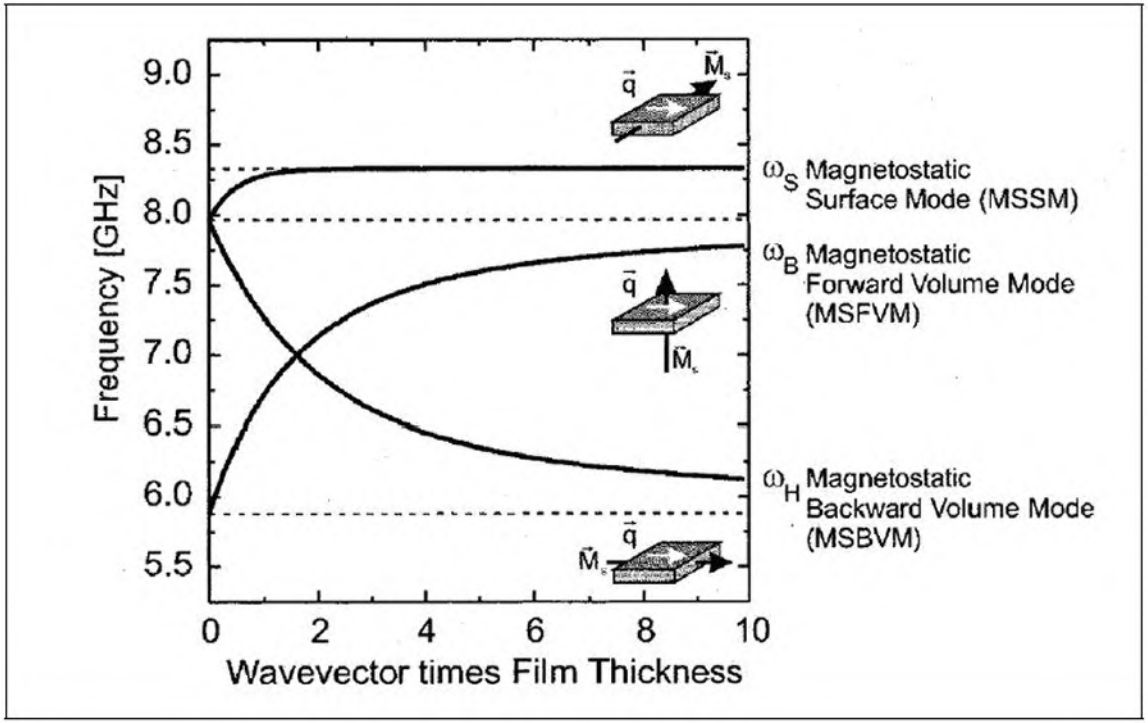
Ytrrium Iron Garnet



Very narrow scattering peaks → very small damping constant

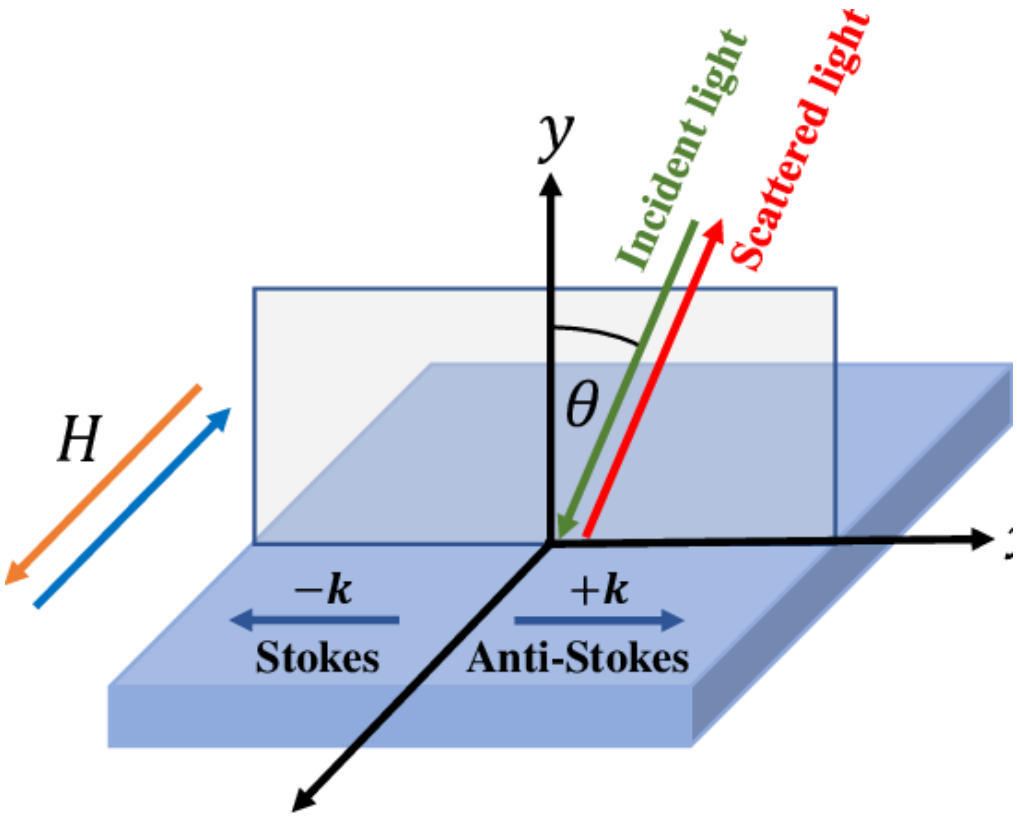
BLS

Excited spin-waves depend on measuring geometry



BLS

Most measurements on thin films in Damon-Eshbach geometry



BLS

Three informations : Line energy, line intensity, linewidth

- Scattering amplitude related to magnon population
Low temperature : Little Anti-Stokes ! (no excitation to absorb)

- Linewidth : damping mechanisms or excitation distribution (+ resolution).

BLS

Frequency of Stokes and Anti-Stokes in presence of DMI :

$$f = \frac{\gamma}{2\pi}\sqrt{B_x B_z} \pm \frac{\gamma}{\pi M_s} Dk \equiv f_0 \pm f_D$$

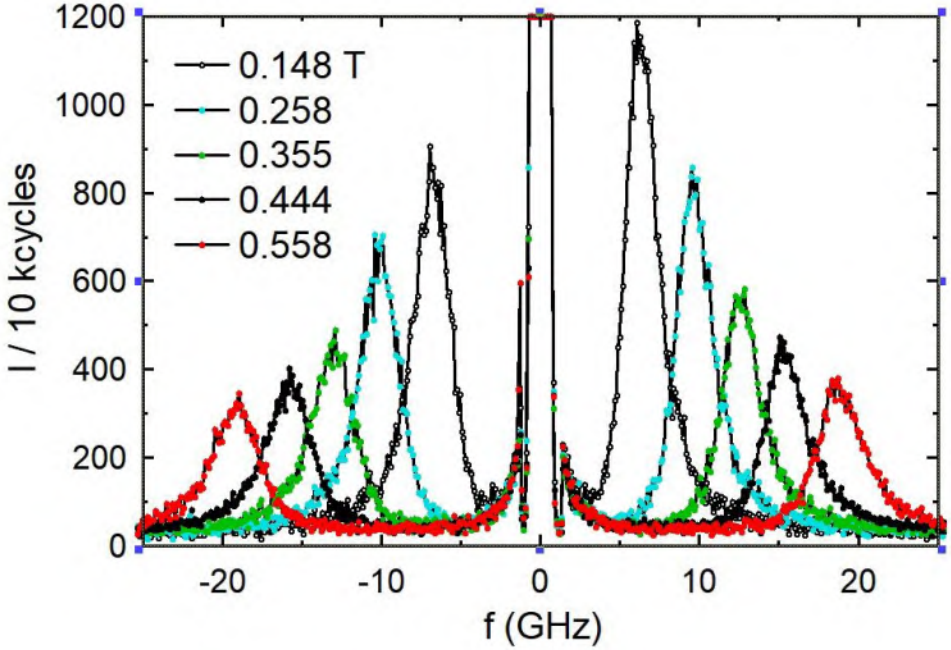
with

$$B_x = \mu_0 H + P(kt)\mu_0 M_s + \frac{2A}{M_s}k^2$$

$$B_z = \mu_0 (H - H_K) - P(kt)\mu_0 M_s + \frac{2A}{M_s}k^2$$

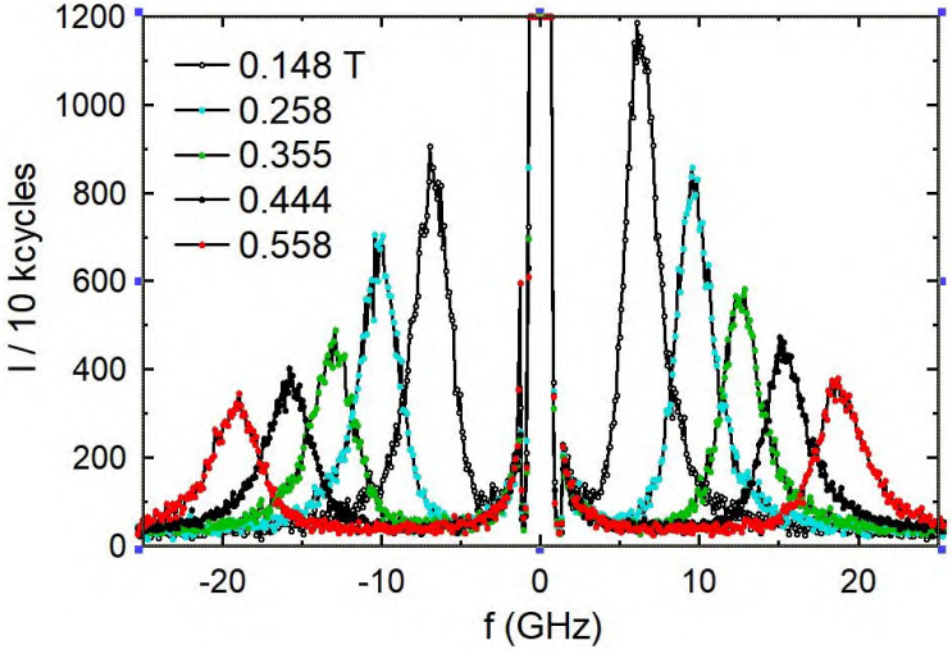
BLS to determine anisotropy field

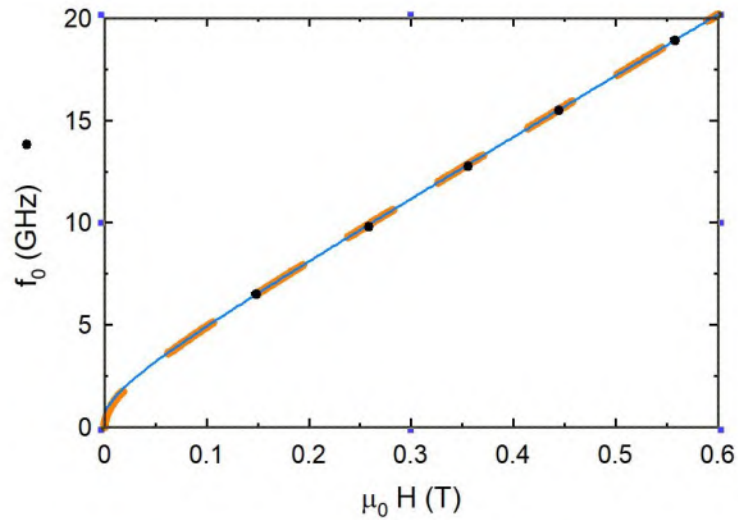
BLS spectra for Pt/Co(1.1nm)/AlOx
as a function of field, 10° incidence



BLS to determine anisotropy field

BLS spectra for Pt/Co(1.1nm)/AlOx
as a function of field, 10° incidence



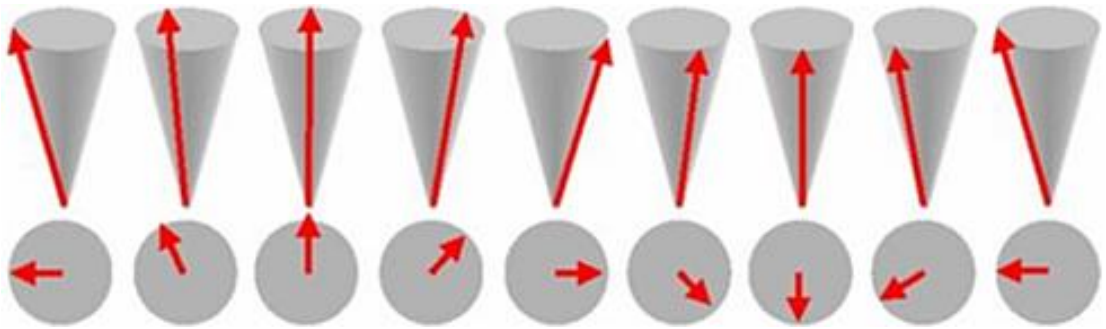


$\mu_0 H_K = 0.176 \pm 0.009$ T

Measurements by A. Thiaville
C. Balan et al., Small 19, 2302039 (2023)

BLS to determine sign and strength of DMI

→ k

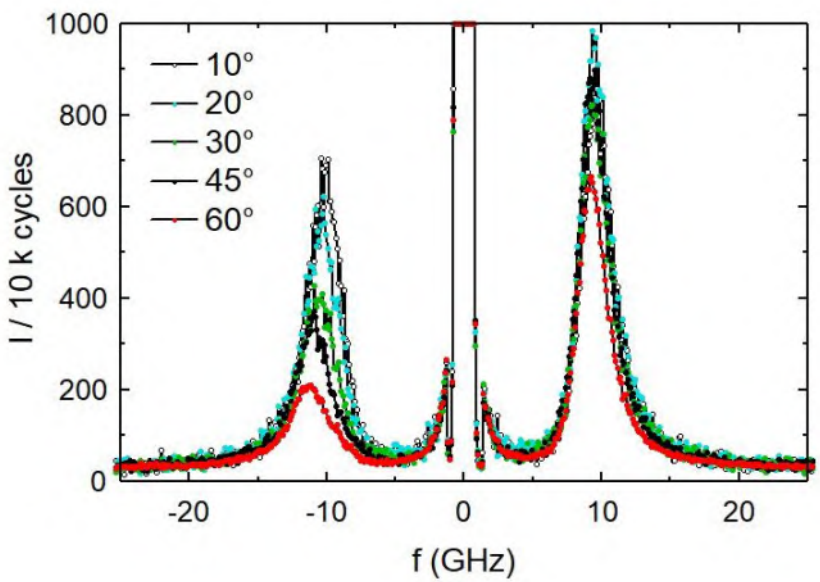


← k

In the presence of DMI, magnons with k ← and k →
do not have the same energy

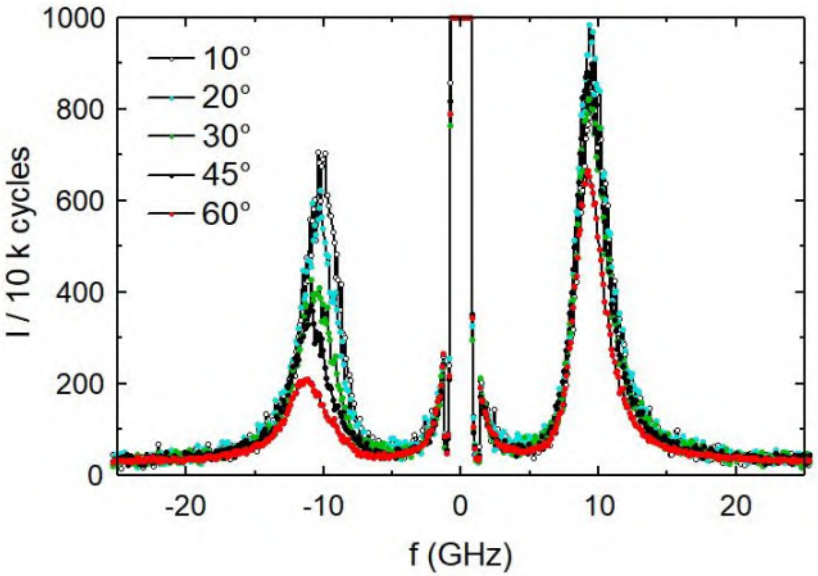
BLS to determine sign and strength of DMI

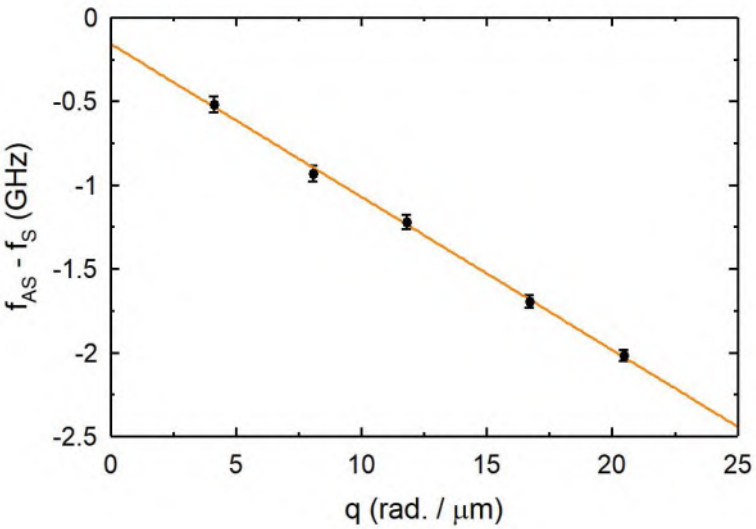
BLS spectra for Pt/Co(1.1nm)/AlOx



BLS to determine sign and strength of DMI

BLS spectra for Pt/Co(1.1nm)/AlOx





$$D = -0.827 \pm 0.017 \text{ mJ/m}^2$$

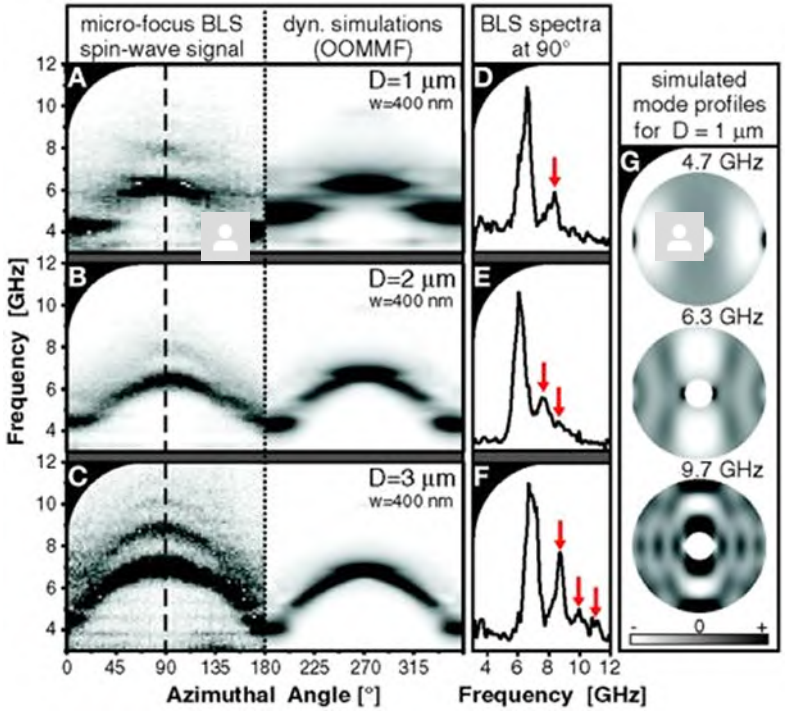
Measurements by A. Thiaville

C. Balan et al., Small 19, 2302039 (2023)

BLS

Micro-BLS can be used to study spin wave spectrum in small structures

Loss of k (divergent beam)

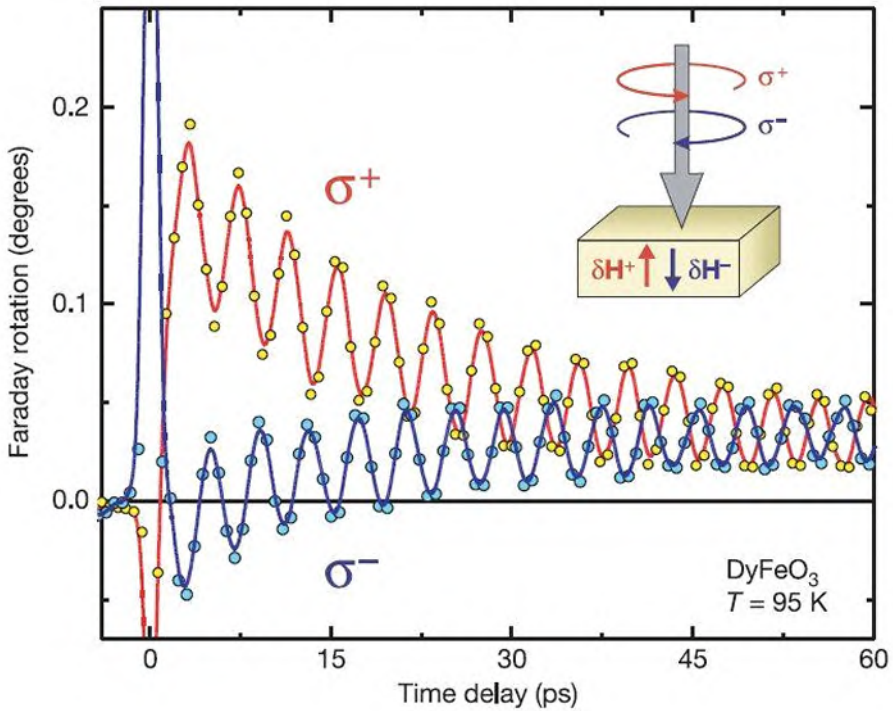


Micro-focused Brillouin light scattering: imaging spin waves at the nanoscale
T. Sebastian, K. Schultheiss, B. Obry, B. Hillebrands, H. Schultheiss
Frontiers in Physics 3 (2015)

2) Influence of light on magnetic properties

Influence of light on magnetic properties

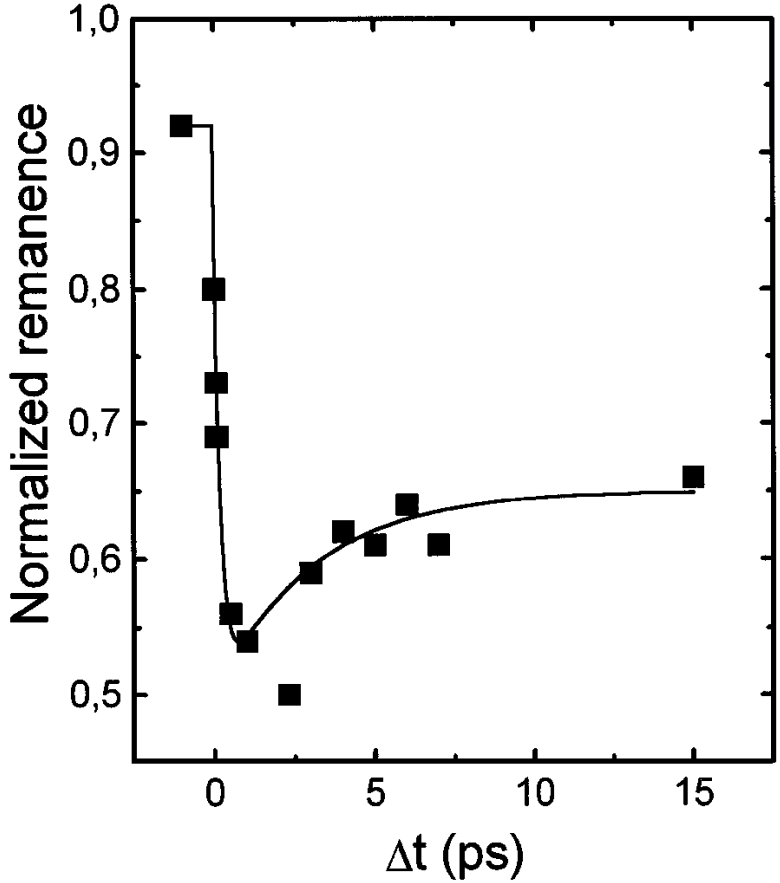
A.V. Kimel et al., Nature 435, 655 (2005)



Magnetic excitations in DyFeO3 probed by the magneto-optical Faraday effect
Left- and right-circularly polarized photons induced opposite (small) magnetic moments
→ Inverse Faraday effect

Influence of light on magnetic properties

Ultrafast demagnetization induced by fs laser pulses



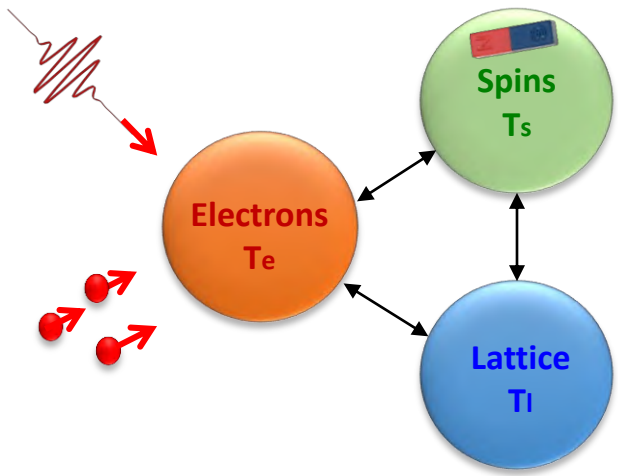
20nm Ni film
Excitation by 60fs laser pulse
620nm, 7 mJ/cm^2

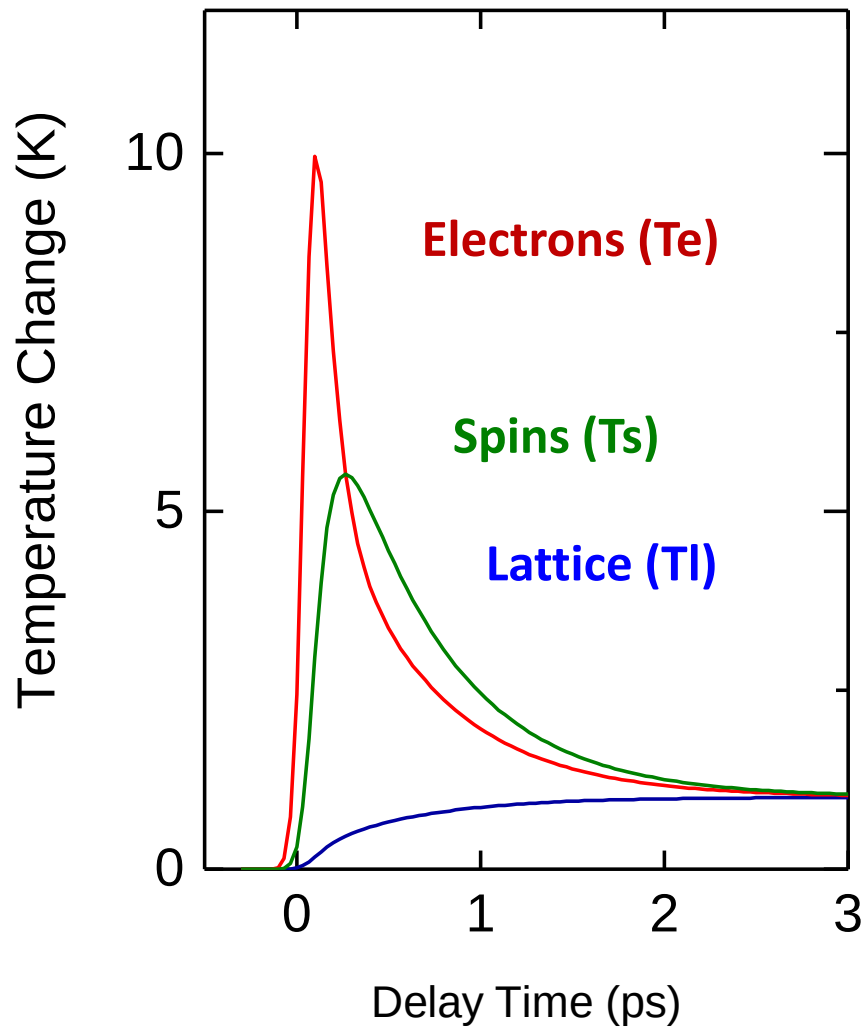
Magnetic response measured using longitudinal Kerr in pump-probe mode

Demagnetization in few hundred femto-seconds

E. Beaurepaire, J.-C. Merle, A. Daunois and J.-Y. Bigot, Phys. Rev. Lett. 76, 4250 (1997)

Ultrafast demagnetization by laser pulses





©S. Mangin

- Energy Transfer
- Angular Momentum Transfer
- Charge / Spin Current Transfer

Ultrafast demagnetization by laser pulses

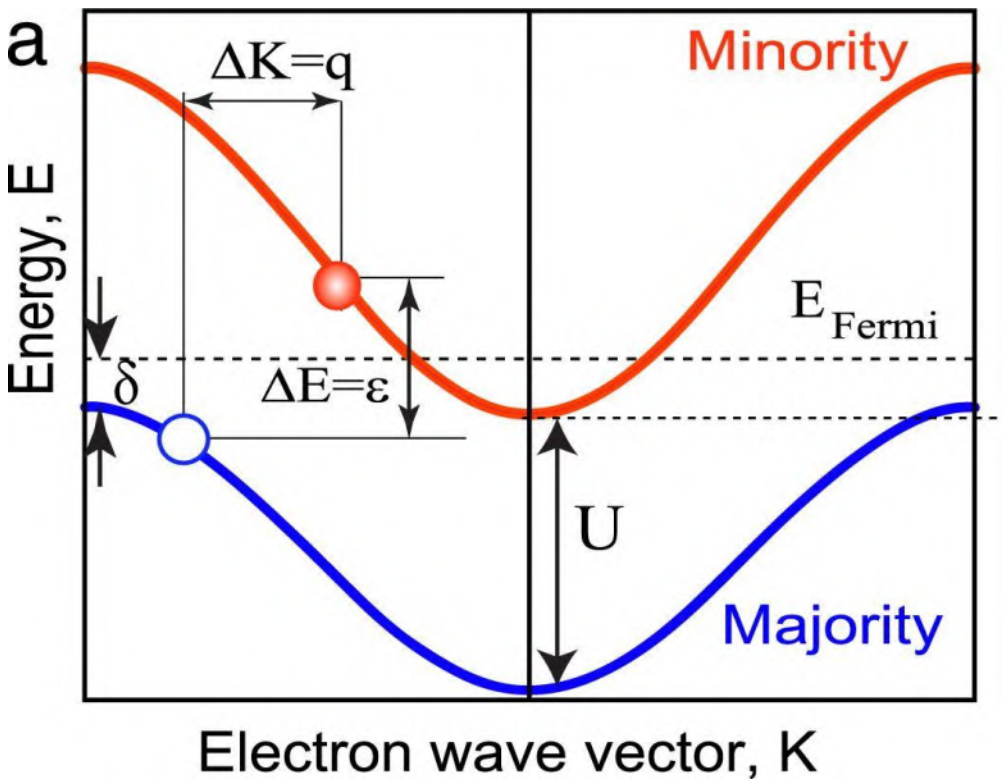
- Reduction of the length of the magnetic moment carried by each atom in the matter (creation of Stoner excitations resulting from a decrease of the exchange splitting).

- a tilt of these vectors resulting in a lower magnetization in average (creation of magnons).

- the propagation of spin-polarized hot electrons.

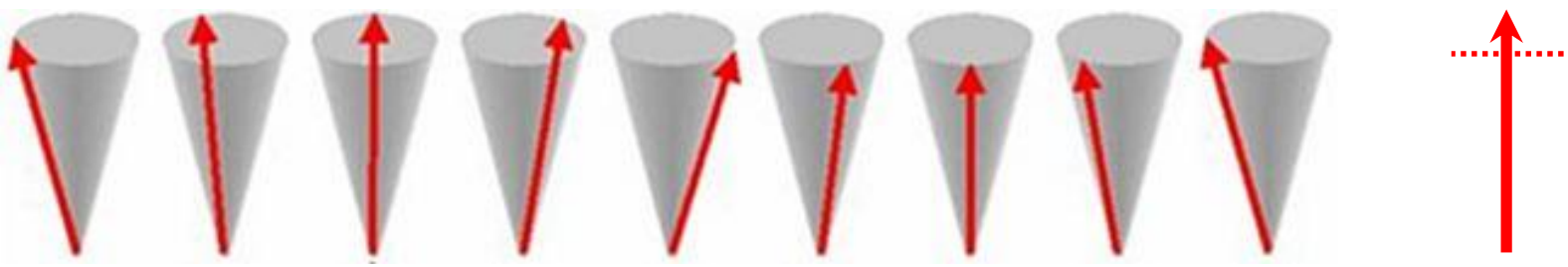
Review :
P. Scheid, Q. Remy, S. Lebègue, G. Malinowski, S. Mangin
J. Magn. Magn. Mater. 560, 169596 (2022).

Ultrafast demagnetization by laser pulses



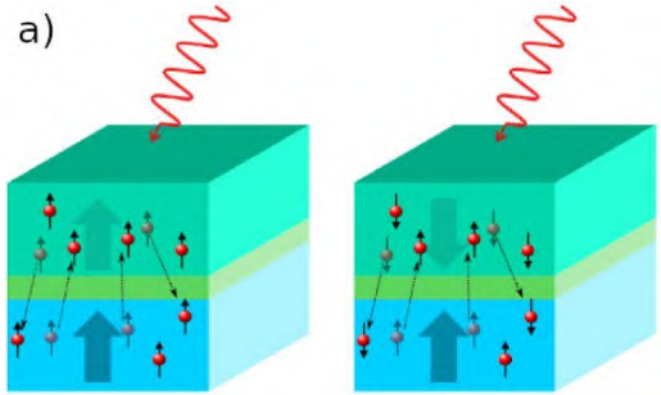
Stoner excitations : electron–hole pair excitations with electrons and holes in the bands of opposite spins

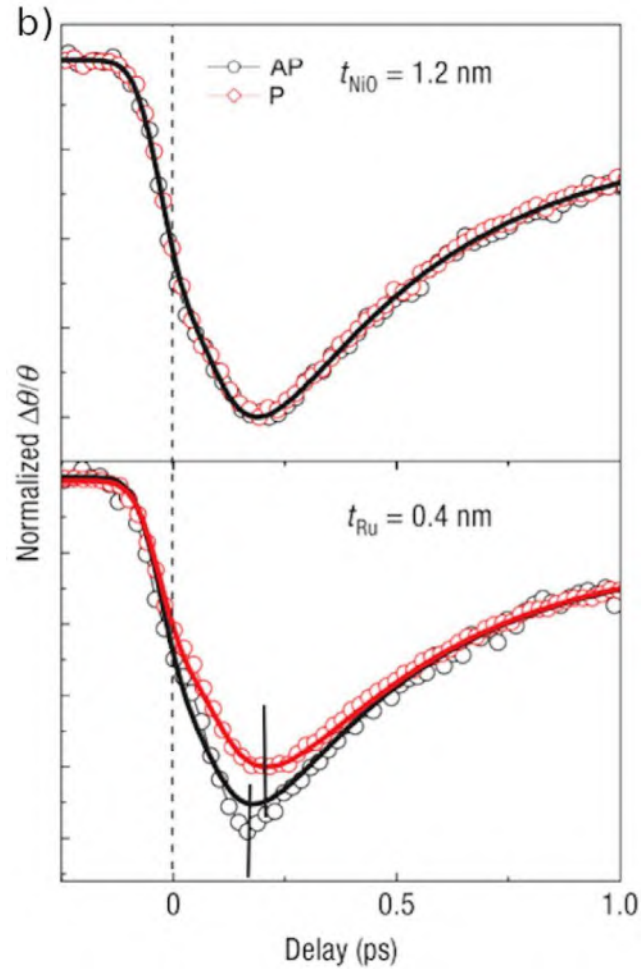
Ultrafast demagnetization by laser pulses



Spin-waves : magnetic « disorder » → magnetization decrease

Ultrafast demagnetization by laser pulses





Comparison (Co/Pt)/Ru/(Co/Pt) and (Co/Pt)/NiO/(Co/Pt) trilayers

G. Malinowski et al., Nat. Phys. 4, 855 (2008).

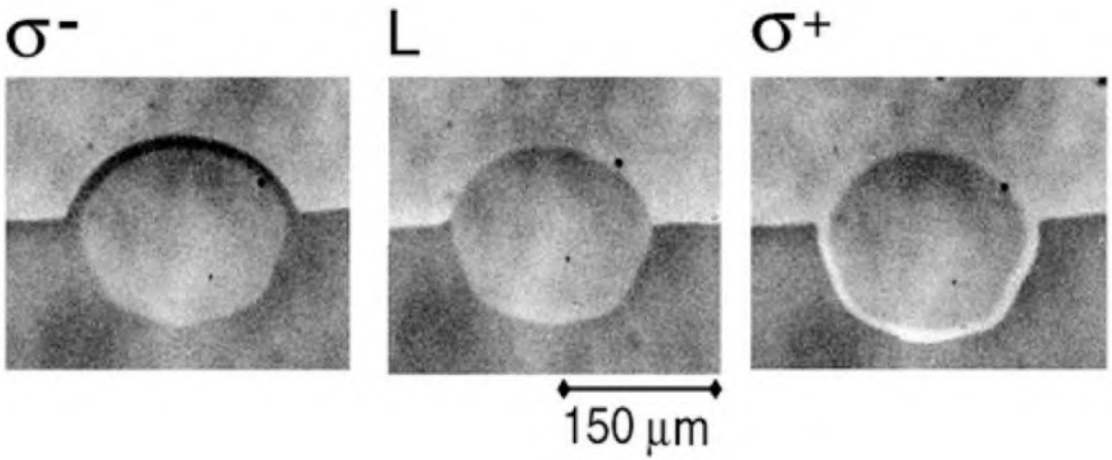
« Superdiffusive » spin currents : hot spin-polarized electrons leaving the irradiated zone

Ultrafast demagnetization by laser pulses

Where does the angular moment go ?

- Spin → orbital magnetic moment ?
- Lattice (phonons) ?
- Superdiffusive spin currents ?
- THz emission ?

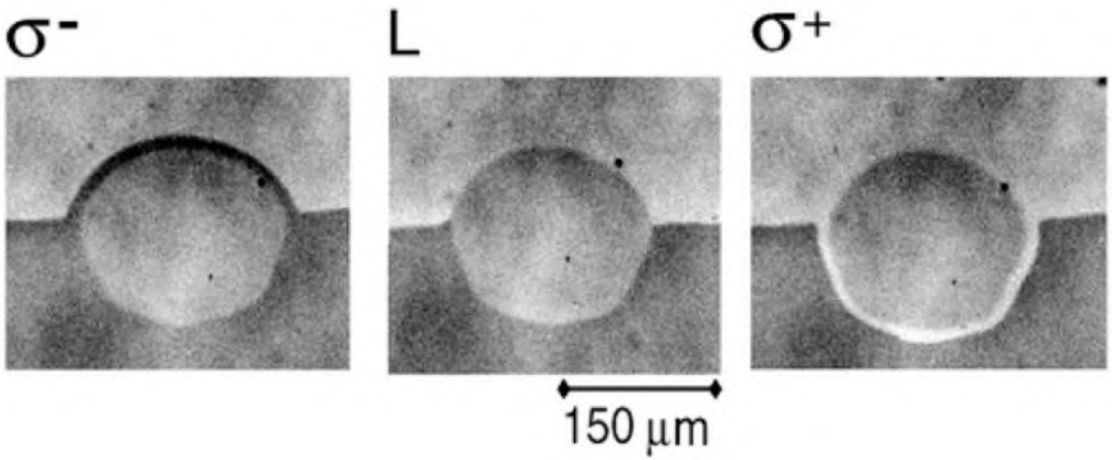
All-optical magnetic switching

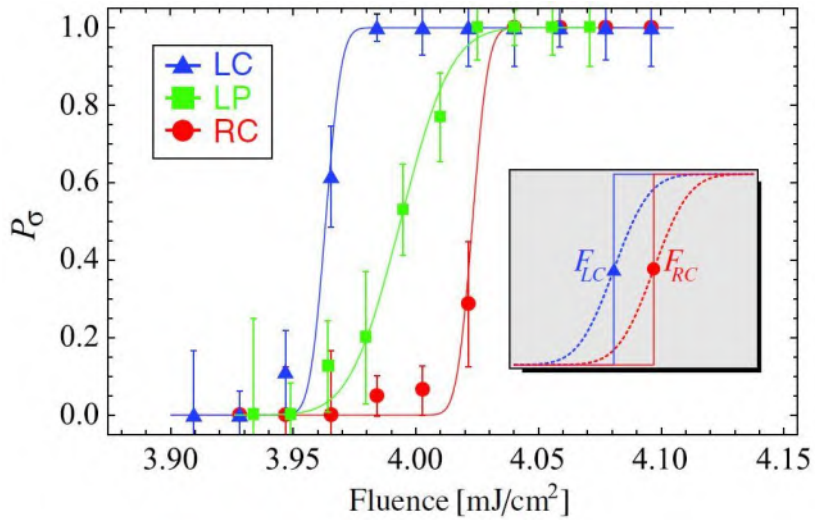


C.D. Stanciu, F. Hansteen, A.V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, T. Rasing, Phys. Rev. Lett. 99 (4) (2007) 47601

Sample : $Gd_{22}Fe_{74.6}Co_{3.4}$

All-optical magnetic switching



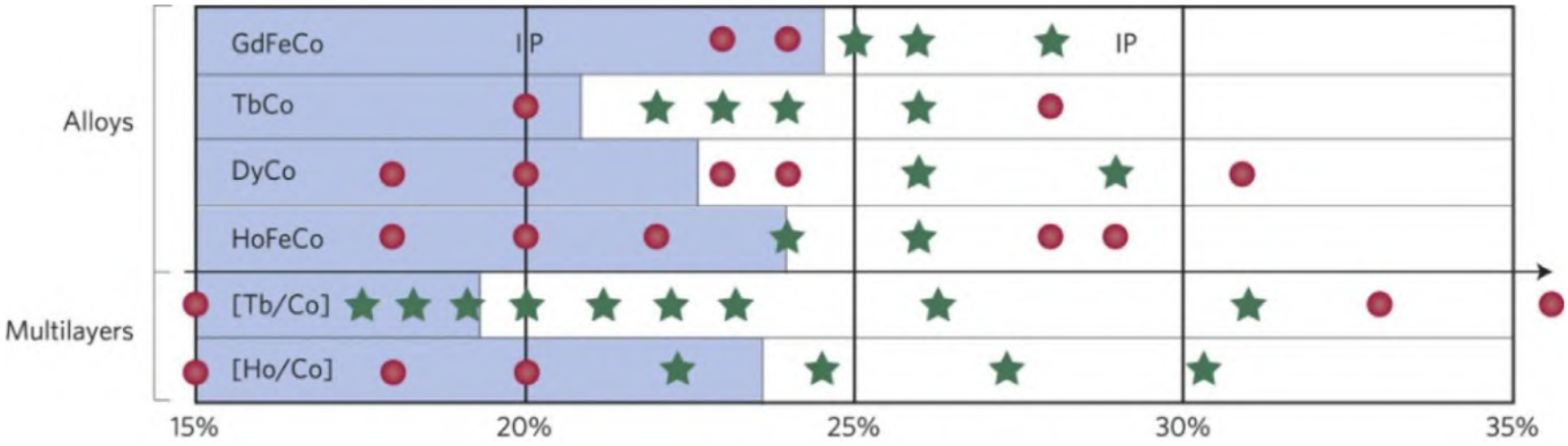


Magnetic circular dichroism in photon absorption

A.R. Khorsand, M. Savoini, A. Kirilyuk, A.V. Kimel, A. Tsukamoto, A. Itoh, T. Rasing, Phys. Rev. Lett. 108 (12) (2012)

All-optical magnetic switching

AO - HDS



AO – HDS observed also in Rare-Earth-free synthetic antiferromagnets

Review :
P. Scheid, Q. Remy, S. Lebègue, G. Malinowski, S. Mangin
J. Magn. Magn. Mater. 560, 169596 (2022).

All-optical magnetic switching

AO – HDS :

1) Magnetic circular dichroism : different absorption of left- and right-circularly polarized → different thermal effects

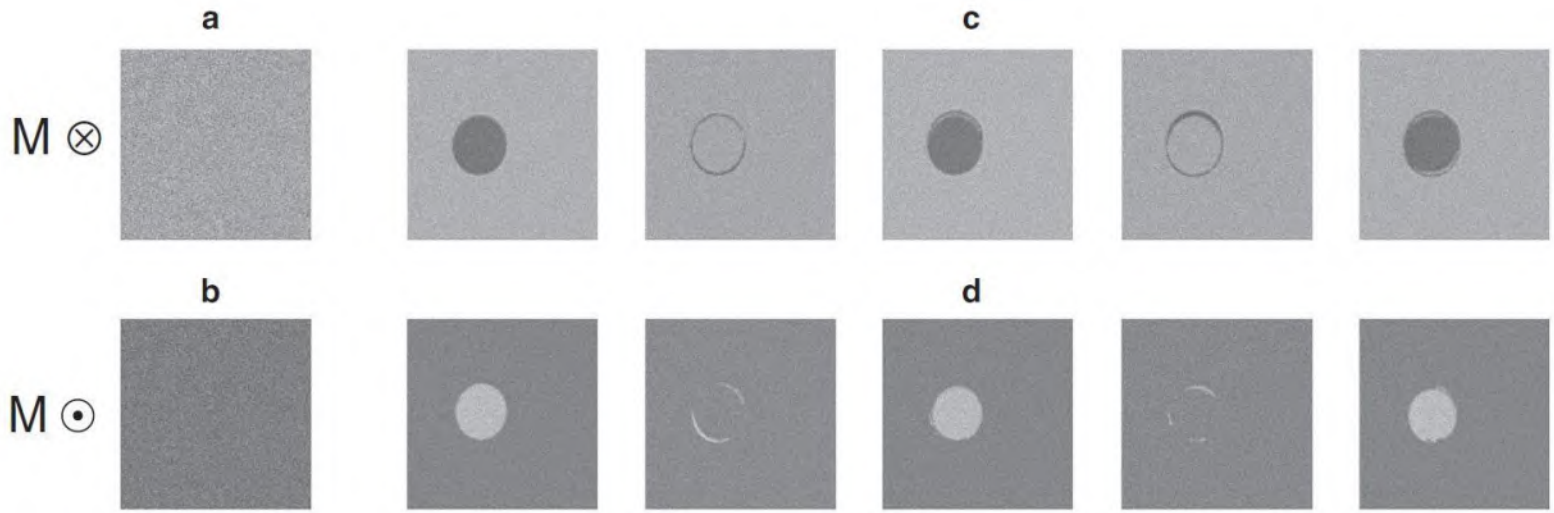
2) Inverse Faraday effect (effective magnetic field induced by the light, induced magnetization ?)

3) Demagnetization induced by optical induced transitions that, in the presence of spin-orbit coupling, do not conserve the magnetization. Magnitude depends on both the helicity of the light and the direction of the magnetization.

4) Direct spin-light coupling (in presence of SOC).

Influence of light on magnetic properties

All-optical helicity-independent switching (AO HIS)

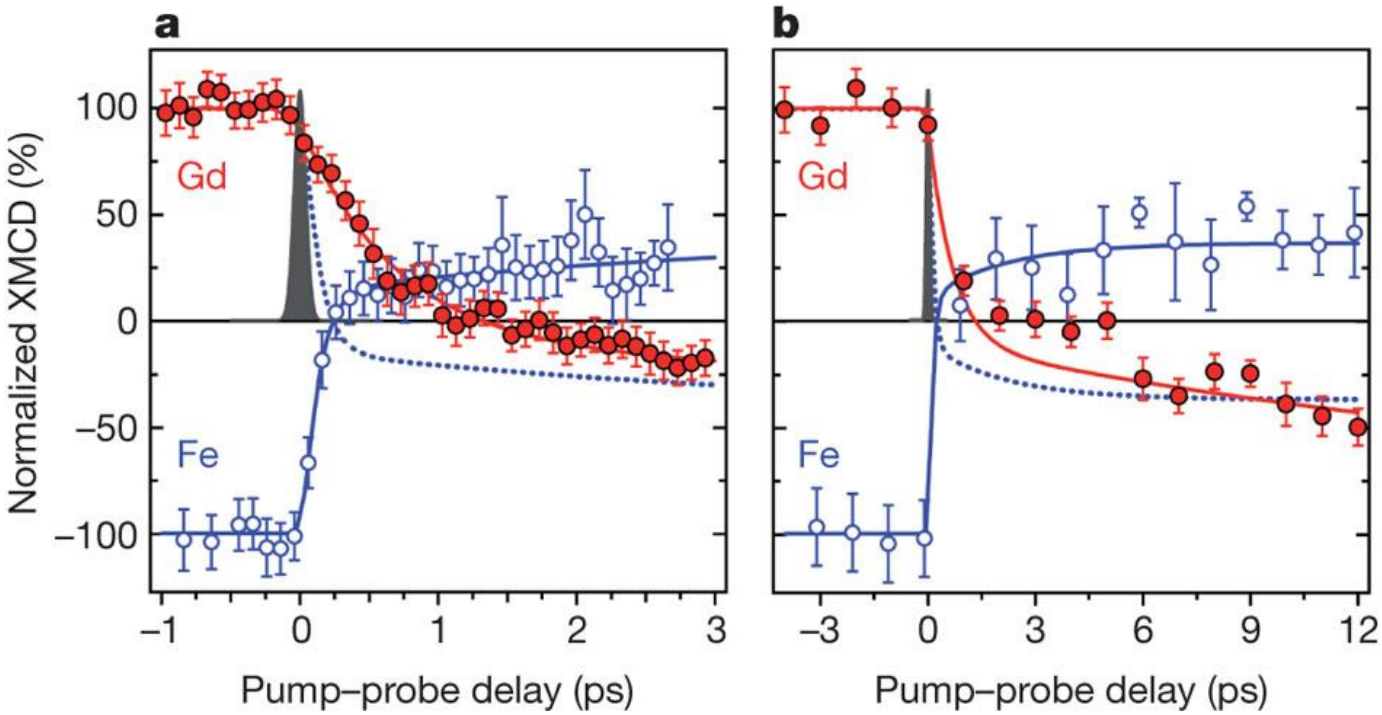


T.A. Ostler et al., Ultrafast heating as a sufficient stimulus for magnetization reversal in a ferrimagnet, Nature Commun. 3 (2012) 666

$Gd_{24}Fe_{66.5}Co_{9.5}$

Influence of light on magnetic properties

All-optical helicity-independent switching (AO HIS)



I. Radu, K. Vahaplar, C. Stamm, T. Kachel, N. Pontius, H.A. Dürr, T.A. Ostler, J. Barker, R.F.L. Evans, R.W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, T. Rasing, A.V. Kimel, Nature 472, 205 (2011).

Summary

- X-ray photons can be used to study element selective magnetic properties (spin, orbital moment) with good spatial resolution (<20nm) and temporal resolution BUT needs a synchrotron
- Optical photons can be used to study magnetic properties (hysteresis, reversal modes, imaging) with <µm spatial resolution (Kerr, Faraday)
- Brillouin Light Scattering spectroscopy is a versatile tool to study (thin film) properties like A, K, DMI

- High-intensity femtosecond laser pulses can strongly influence magnetic properties of thin films (demagnetization, all-optical switching)