Magnetic Imaging with Synchrotron Radiation and Neutrons

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Outline

- 1 – Absorption (dichroism) imaging
- 2 – Bragg diffraction imaging
    - Neutron case (mainly “exotic domains”)
    - X-ray case (mainly phase transitions)
- 3 – Microbeam-based imaging
- 4 – Perspectives/conclusion

IMAGING

What is imaging about ?

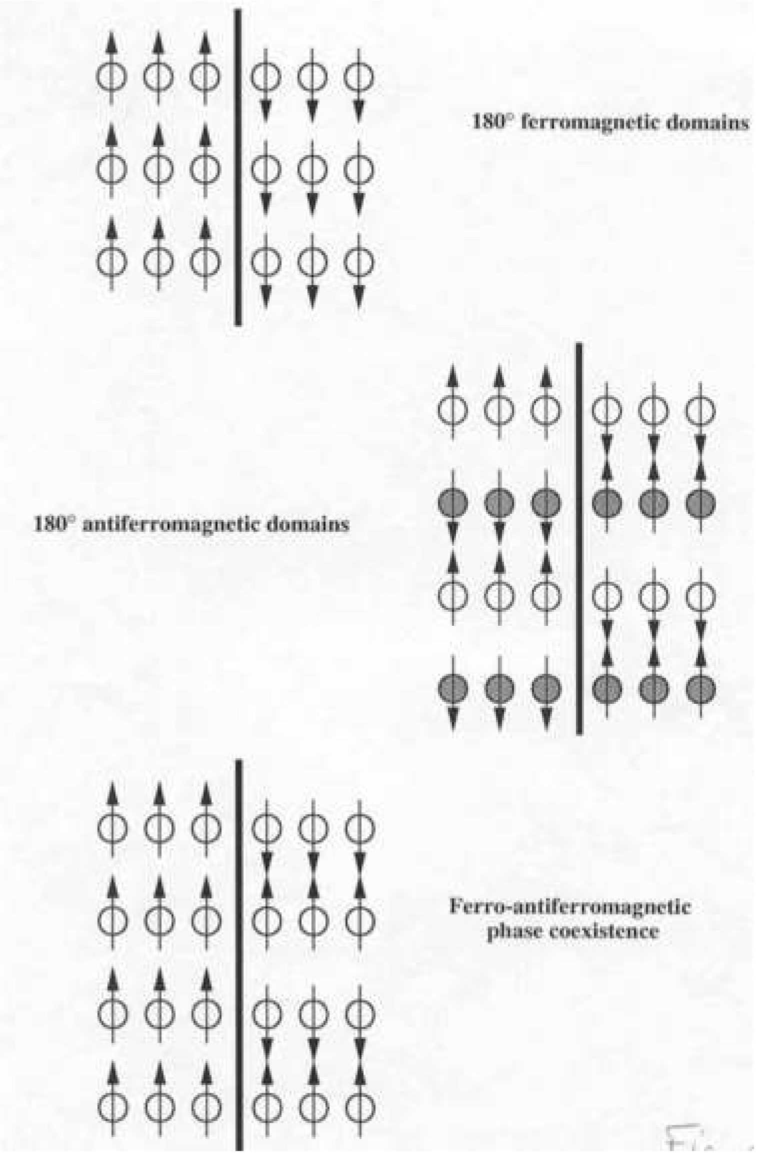
- **revealing inhomogeneities and singularities in the sample**
- **through variations in reflection, transmission, … behaviour for the probe used**

Images can be made with

light, electrons, ultrasound, nuclear magnetic resonance, **X-rays, neutrons**, …

Making images with different probes is valuable because it can yield different information

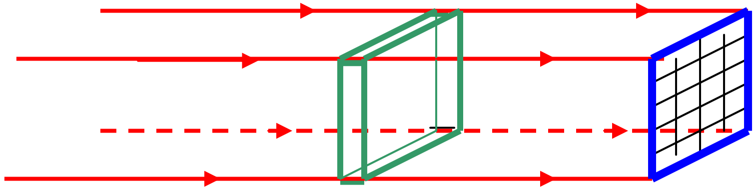
Magnetic inhomogeneities



Imaging with a parallel and extended beam

Magnetic Contrast mechanism: differences in absorption

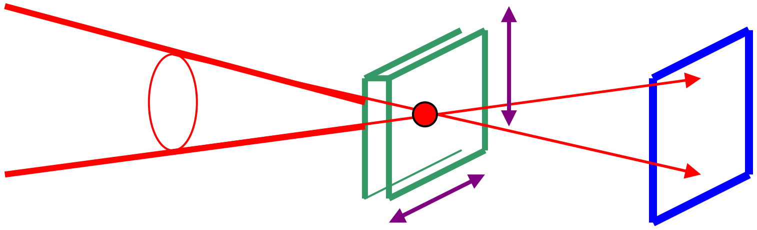
the spatial resolution is mainly a function of the detector



Imaging with a microbeam (« scanning »)

Magnetic Contrast mechanism: absorption or structure factor

the spatial resolution is mainly a function of the spot size



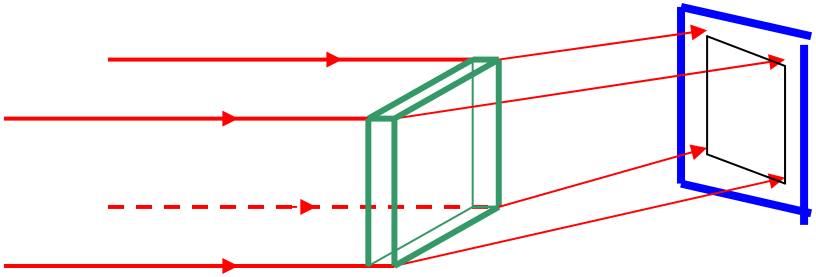
Bragg-diffraction-imaging

Magnetic Contrast mechanism: differences in

⇨structure factor

⇨associated distortion (magnetostriction)

more complex dependence of the spatial resolution



Single-crystal
sample

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Absorption-based
Magnetic Imaging Techniques

ABSORPTION IMAGING

X-ray imaging is not a new technique....

But its application to magnetism is new!

First X ray made in public. Hand of the famed anatomist, Albert von Kölliker, made during Roentgen's initial lecture before the Würzburg Physical Medical Society on January 23, 1896.

Basic idea of magnetic dichroism (1)

X-ray magnetic circular dichroism (XMCD) is the difference in the absorption of left and right circularly polarized X-rays.

⇨Circularly polarized photons are in an « eigenstate » of $J_{z;}$

(linear polarization $< J_z > = 0$)

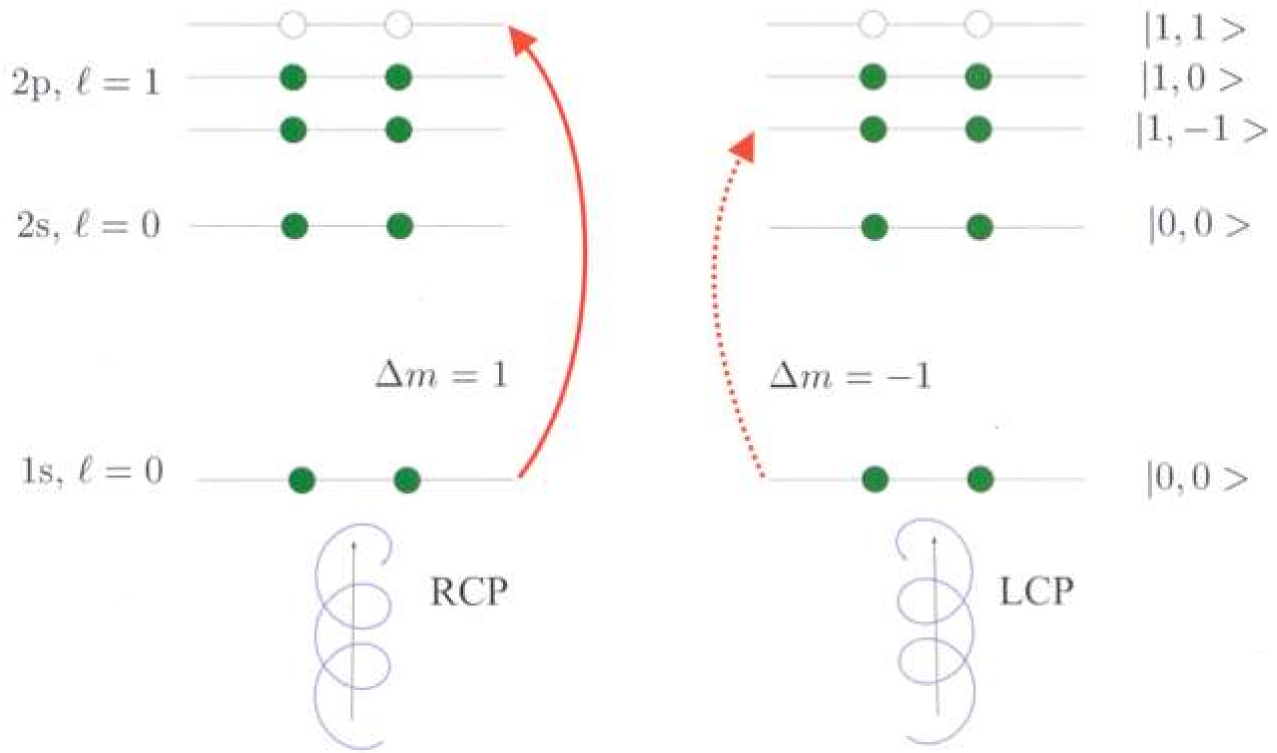
⇨Selection rules for electronic transitions: non zero probability (matrix element) only if $\Delta l = +1$ or -1

⇨Photon absorbed: its angular momentum transmitted to sample

Basic idea of magnetic dichroism (2)

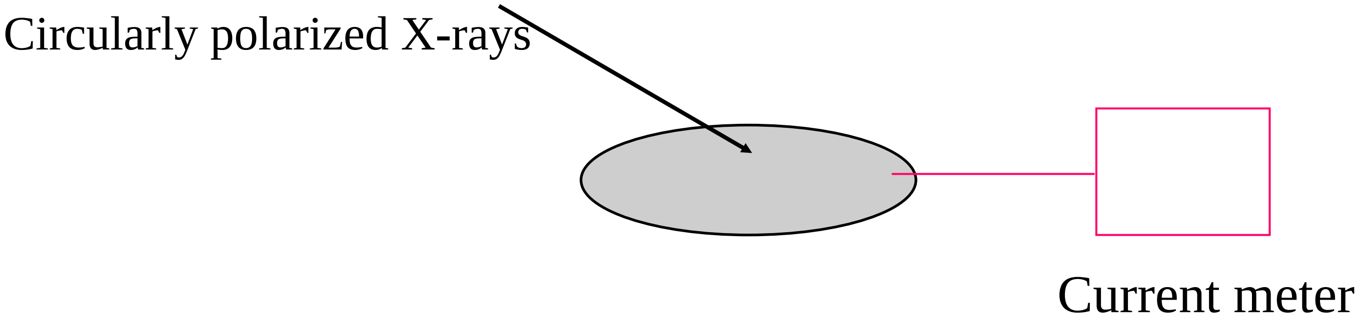
Academic example: atom with 8 electrons in a magnetic field

(a) Simplified energy level diagram



Magnetic dichroism

XMCD proportional to <M>; « big » effect near the absorption edges



For 3d metals: L edge E =0.7 keV

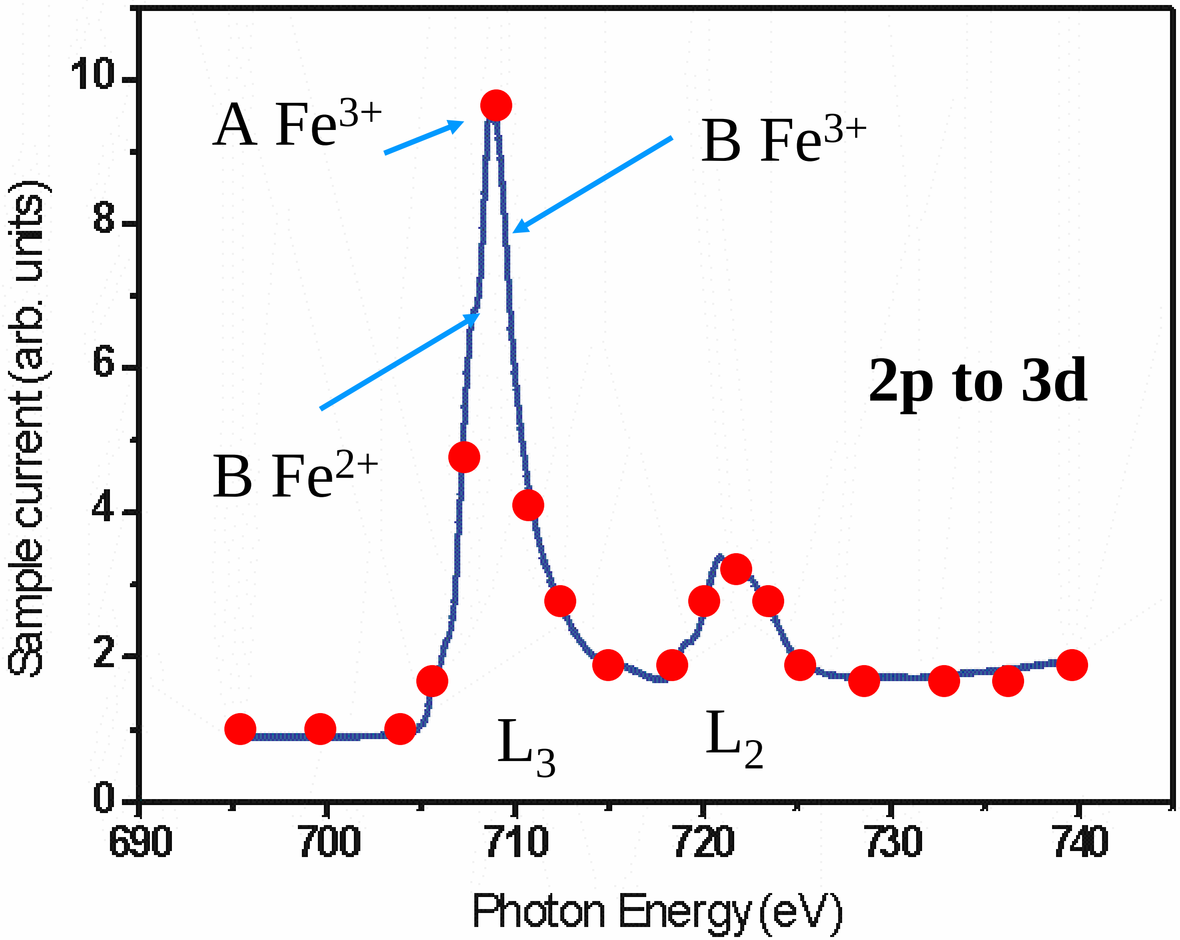
Measure: electron yield for soft X-rays (99% of the yield are electrons for E<1 keV) and 1% is fluorescence.

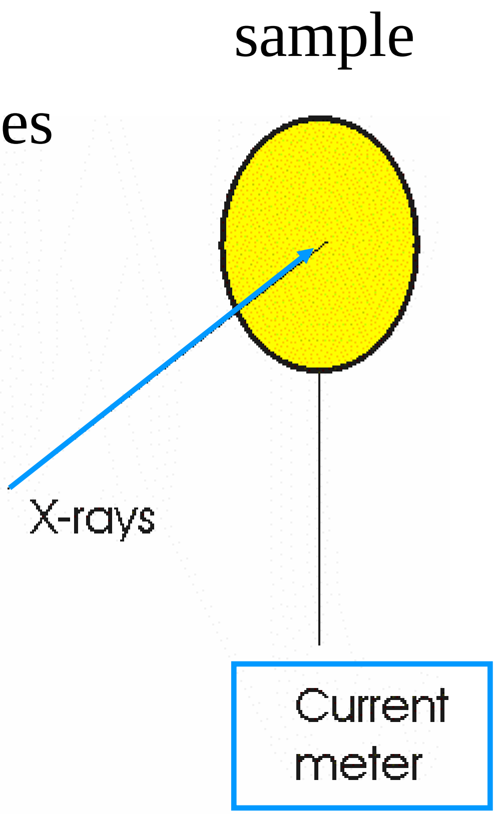
Opposite situation for hard X-rays

Review paper: J. Stöhr, J.Magn.Magn.Mat. ***200****, 470 (1999)*

X-ray absorption spectroscopy

Fe_3O_4 –ferrimagnet iron 2+, and 3+ in different sites

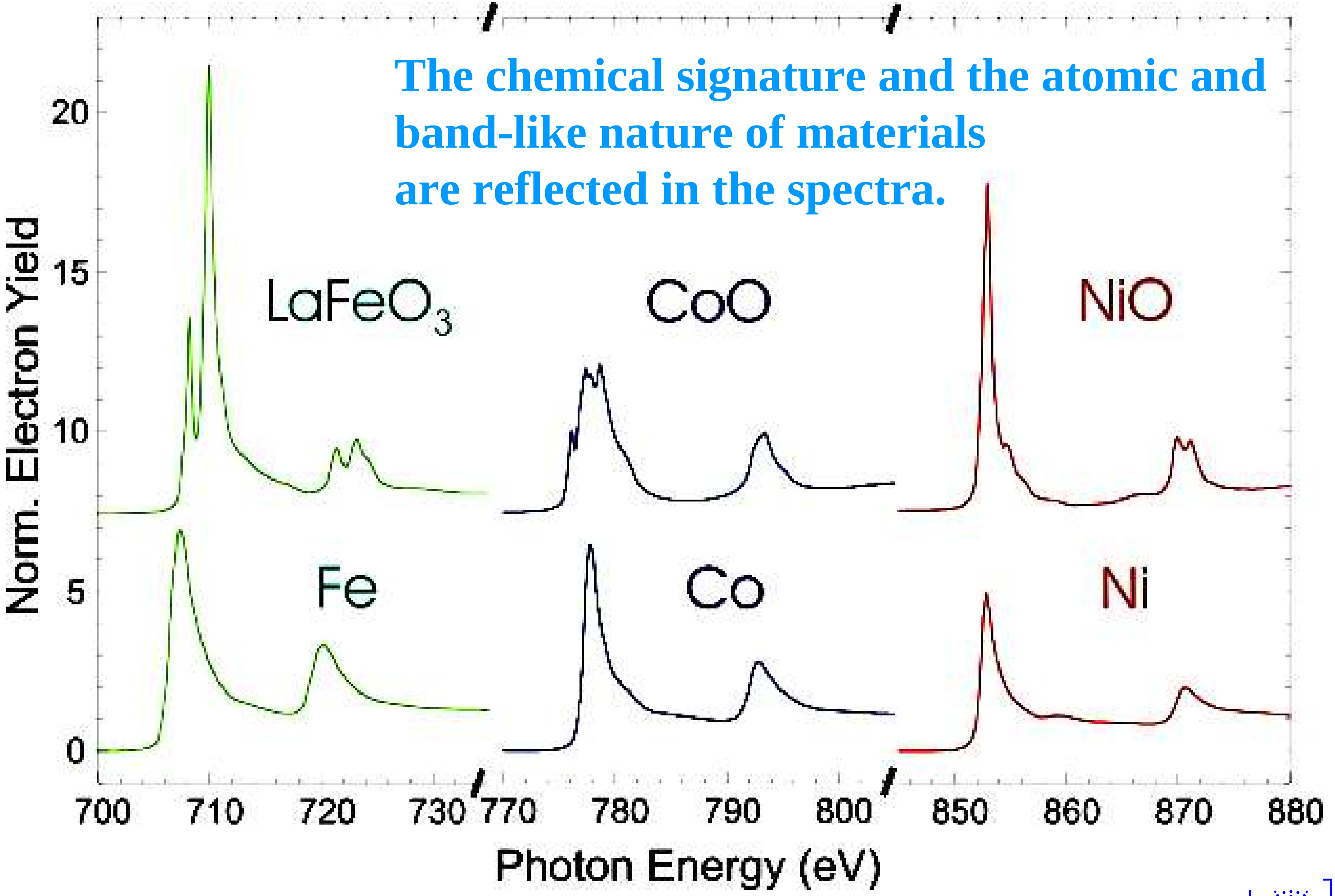




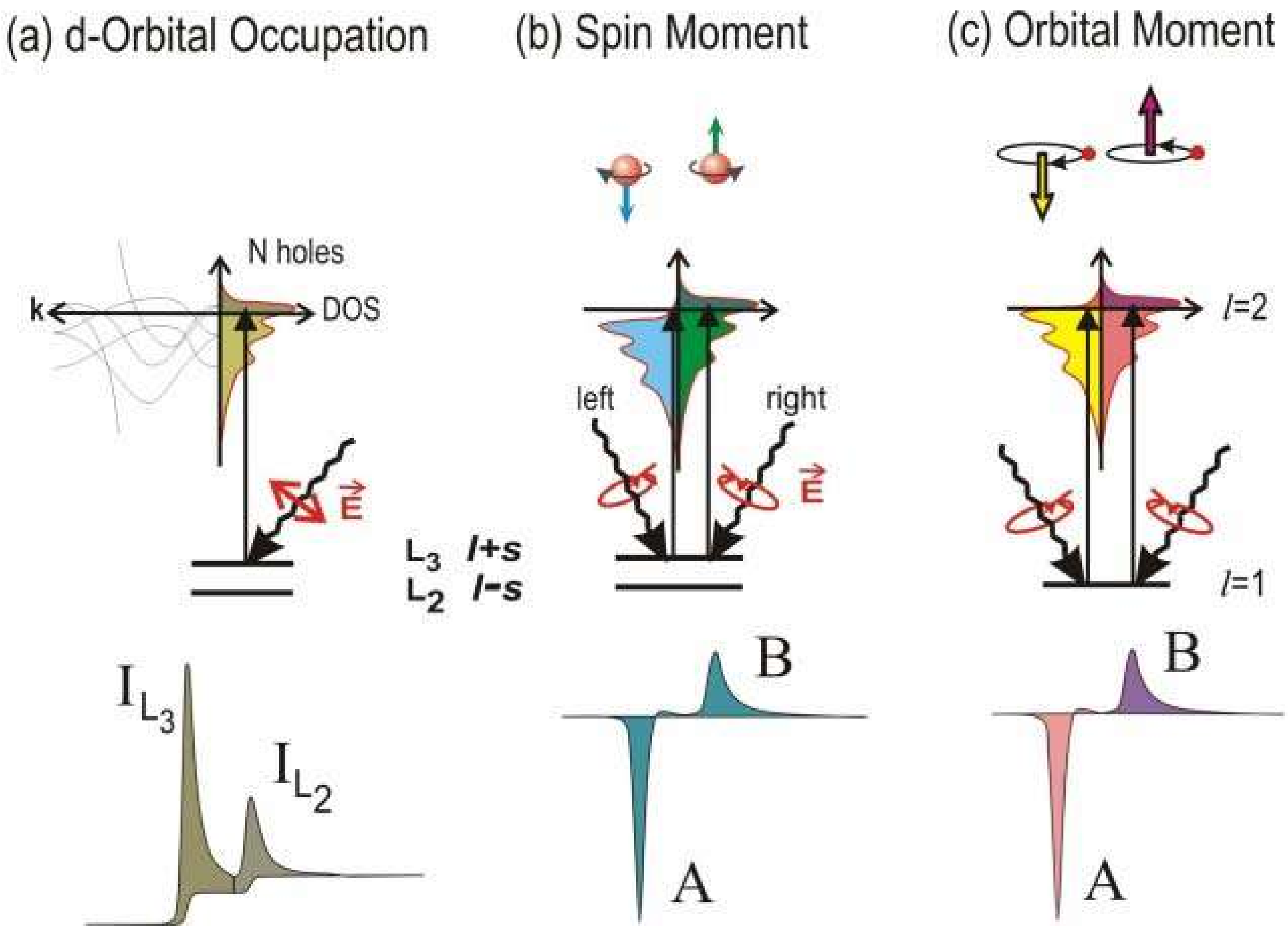
Electron yield is proportional to the absorption coefficient

Scan photon energy; measure electron yield

The chemical signature and the atomic and band-like nature of materials are reflected in the spectra.



Spin and Orbital Moments: X-Ray Magnetic Circular Dichroism



Using the example of the 2p edges of 3d transition metals

Instrumentation:

What is needed?

Circularly polarised x-rays.
Tunable energy
Magnetic field
Measure the absorption.

Circularly Polarized X-Rays

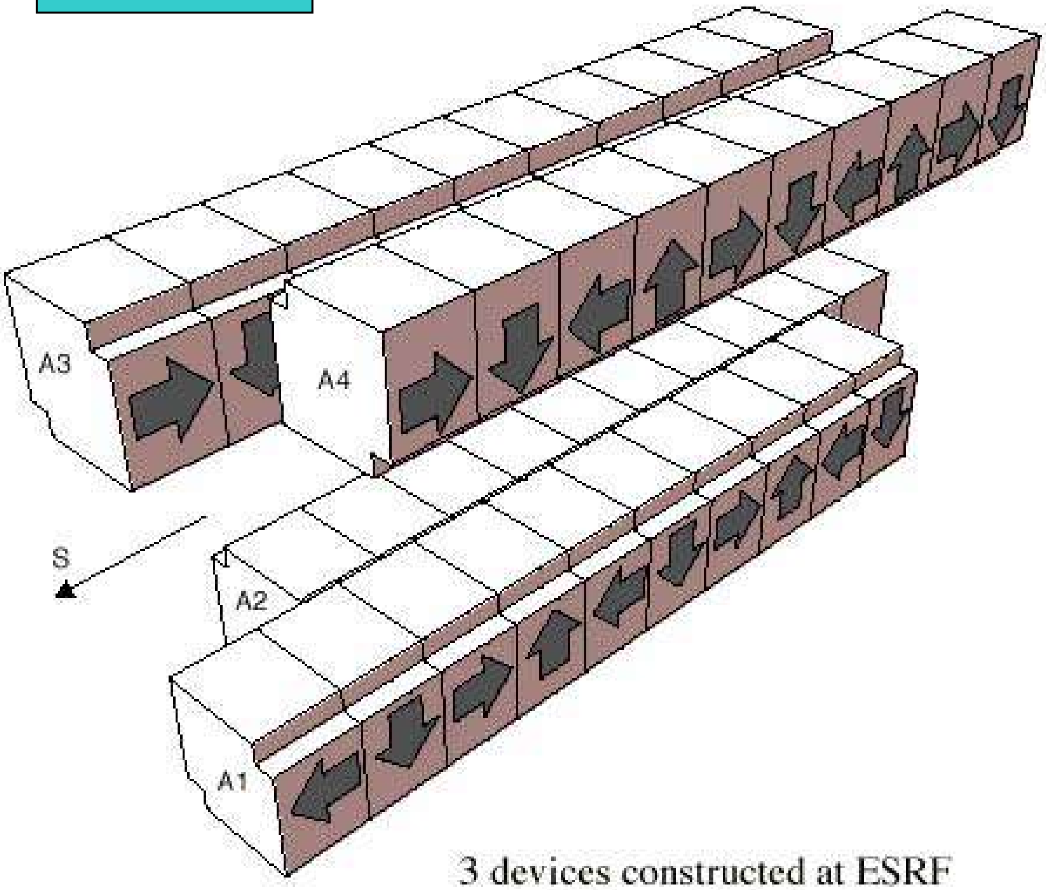
A storage ring, where the electrons turn and produce X-rays, is composed by bending magnets and straight sections with insertion devices (undulators, wigglers)

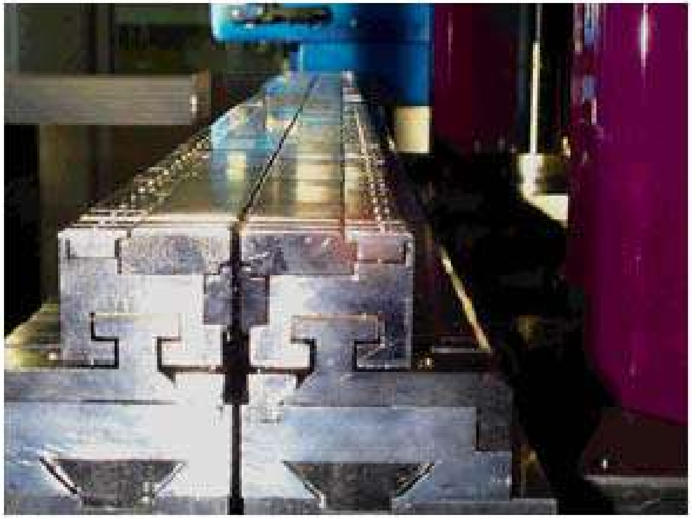
Radiation from a bending magnet: linearly polarized in the plane of the orbit, elliptically polarized (right and left) above and below the orbit plane

Usual insertion devices produce linearly polarized lights; but special inserrtion devices producing circularly polarized light have been produced

J. Chavanne
P. Elleaume

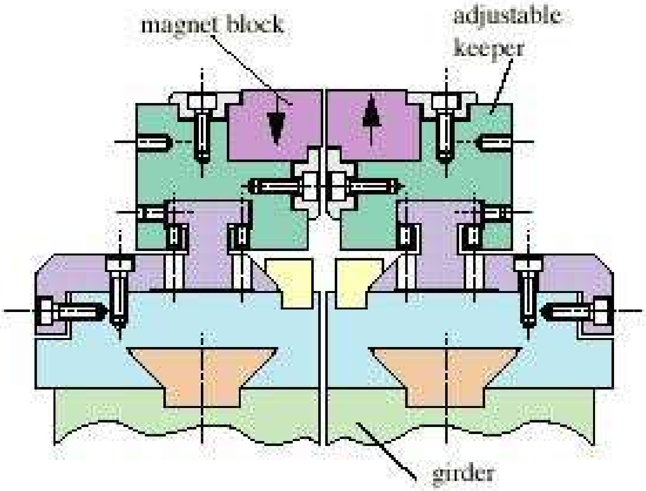
APPLE II STRUCTURE



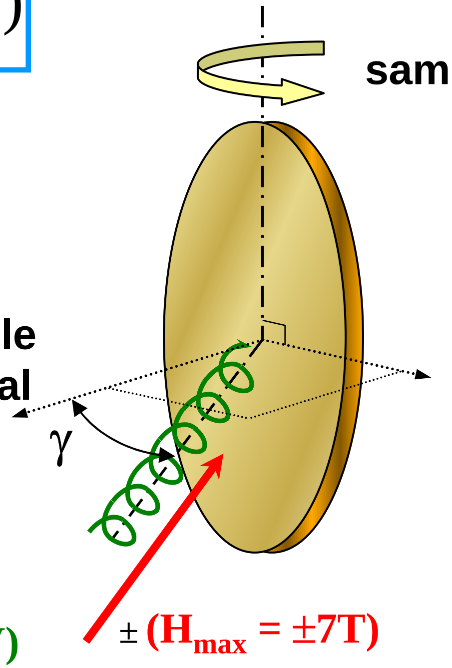


3 devices constructed at ESRF
period 88 mm (2 devices)
period 38 mm

Advanced **P**lanar **P**olarized **L**ight **E**mitter



Experimental set-up, ID08 (ESRF)



Circular polarization rate : ~100%

Electron and fluorescence yield detection,

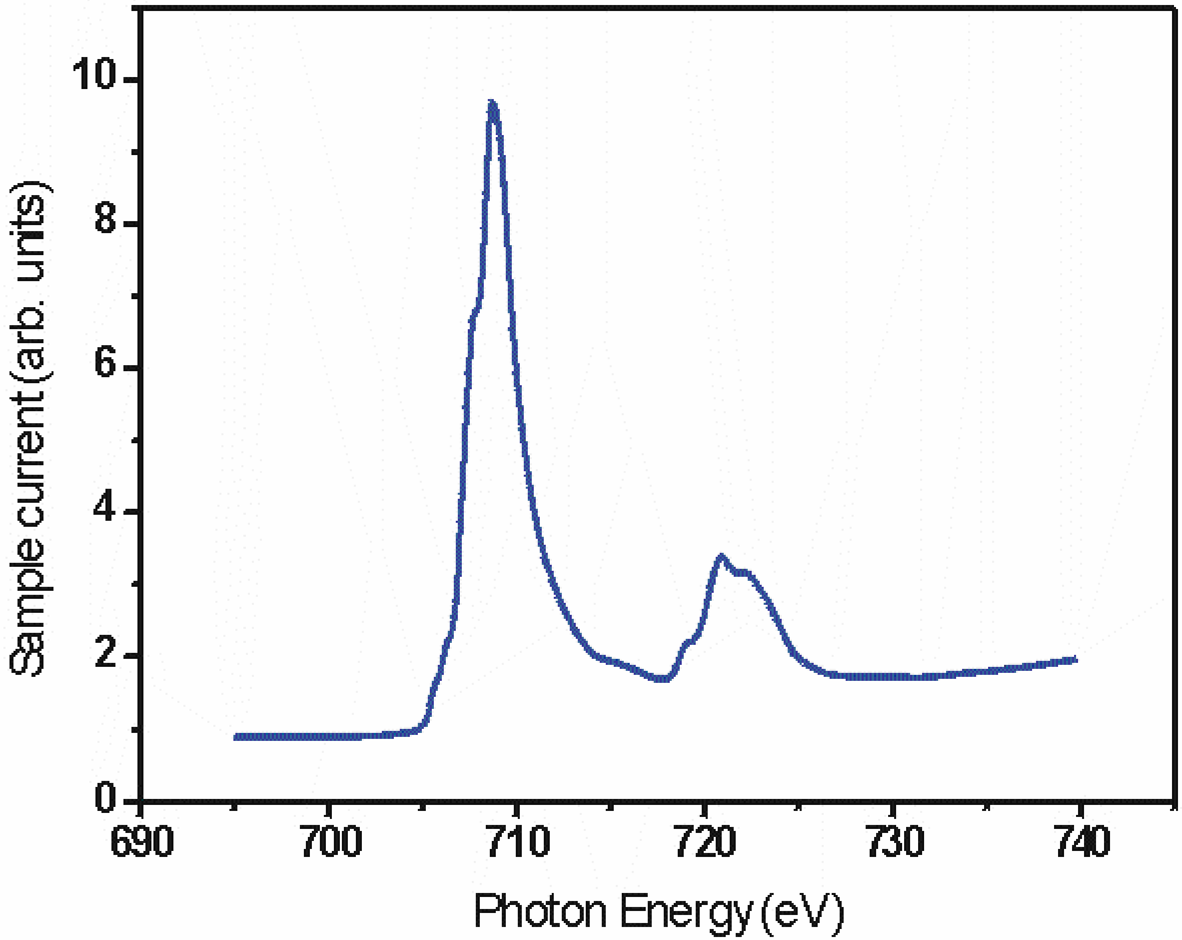
Superconducting magnet (+/- 7T),

Ultra-High Vacuum, p » 5 x 10^{-11} mbar,

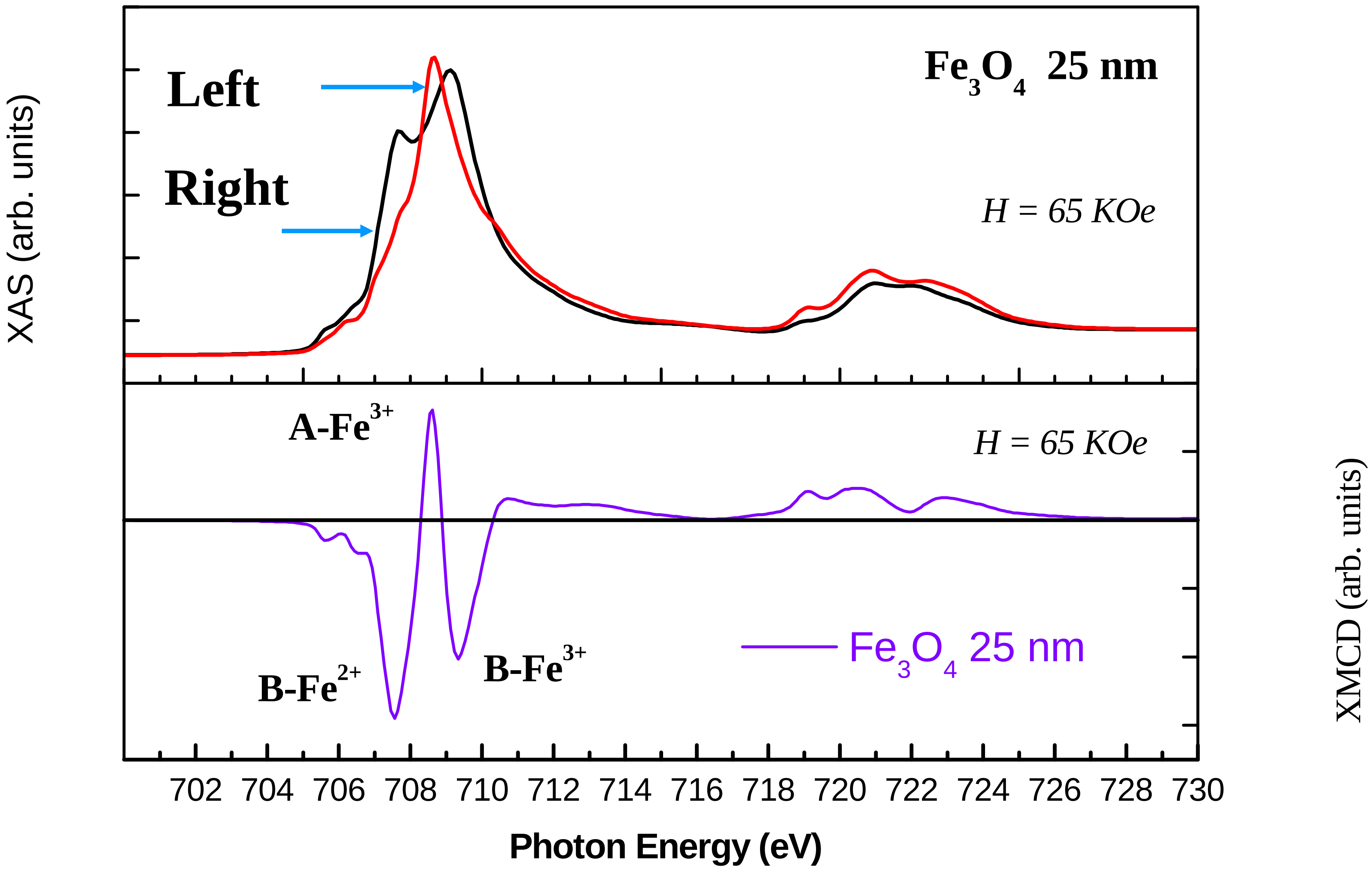
6.5K < T_{sample} < 300K,

in-situ e- beam evaporator and sample preparation facilities.

Fe_3O_4

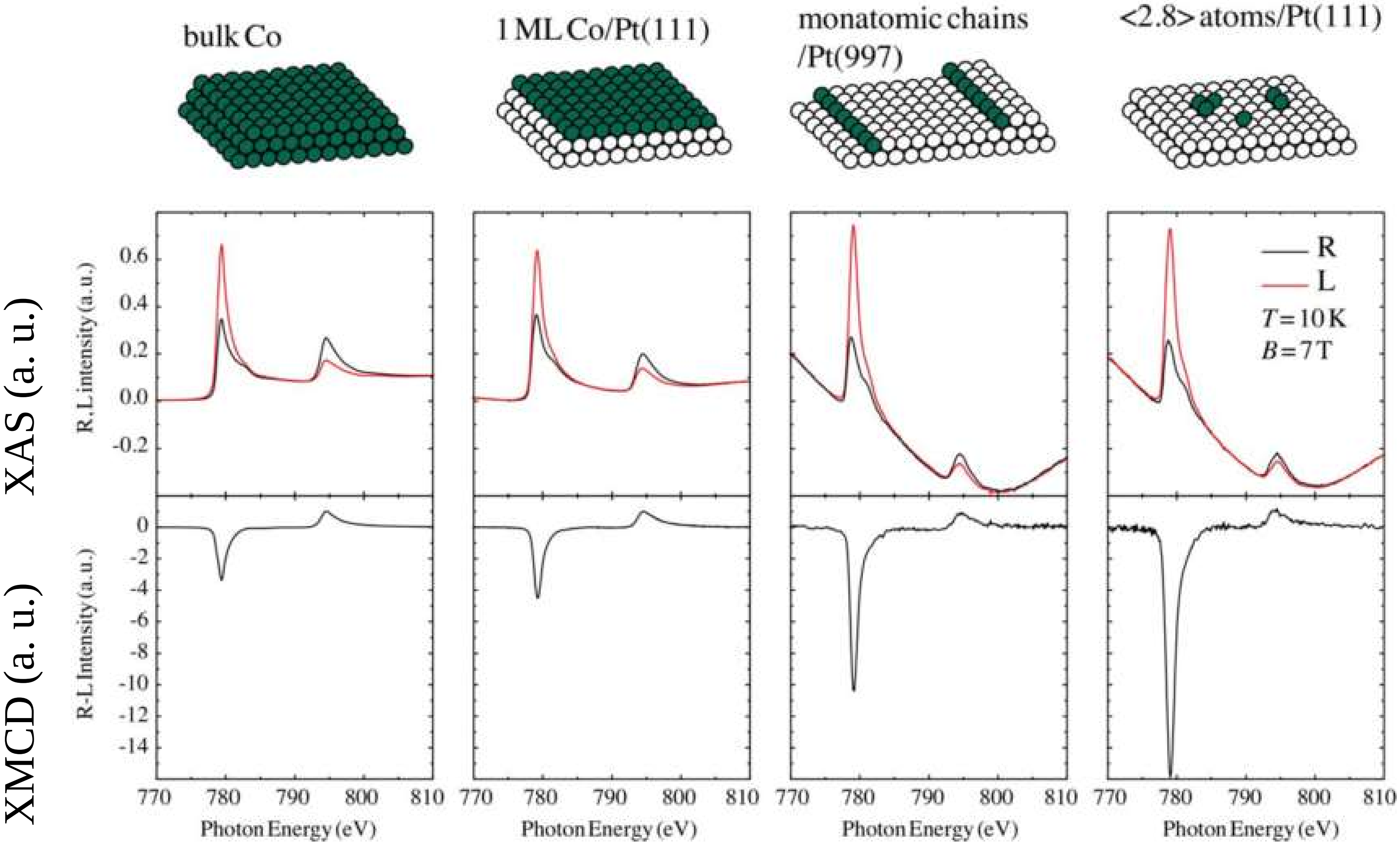

10
8
6
4
2
0
Sample current (arb. units)
690
700
710
720
730
740
Photon Energy (eV)

Fe_3O_4 Ferrimagnetic – chemical and magnetic information

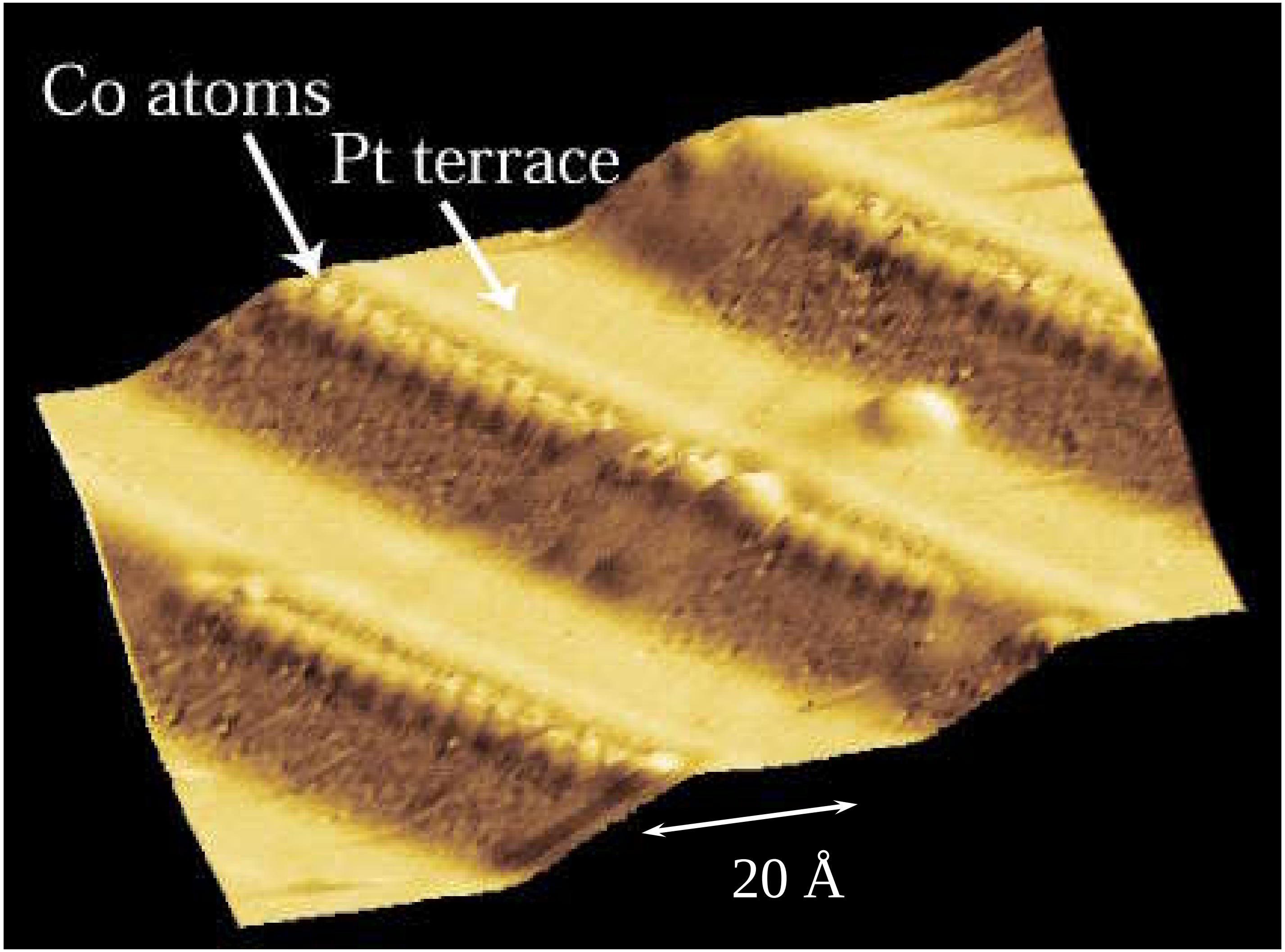


Bulk

XMCD spectra of 3, 2, 1, 0 - dimensional Co structures on Pt surfaces



m_L	**$0.14\ \mu_B$**	**$0.27\ \mu_B$**	**$0.67\ \mu_B$**	**$0.79\ \mu_B$**

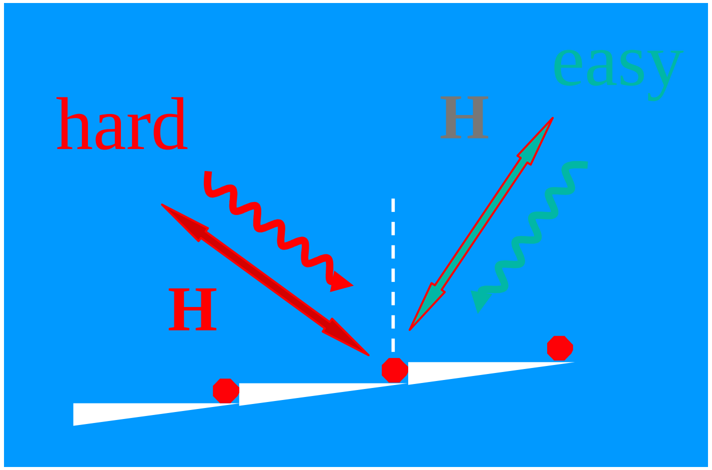


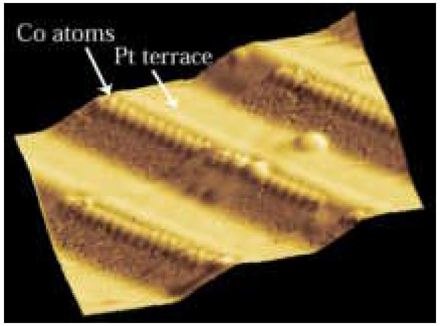
Atomic wires

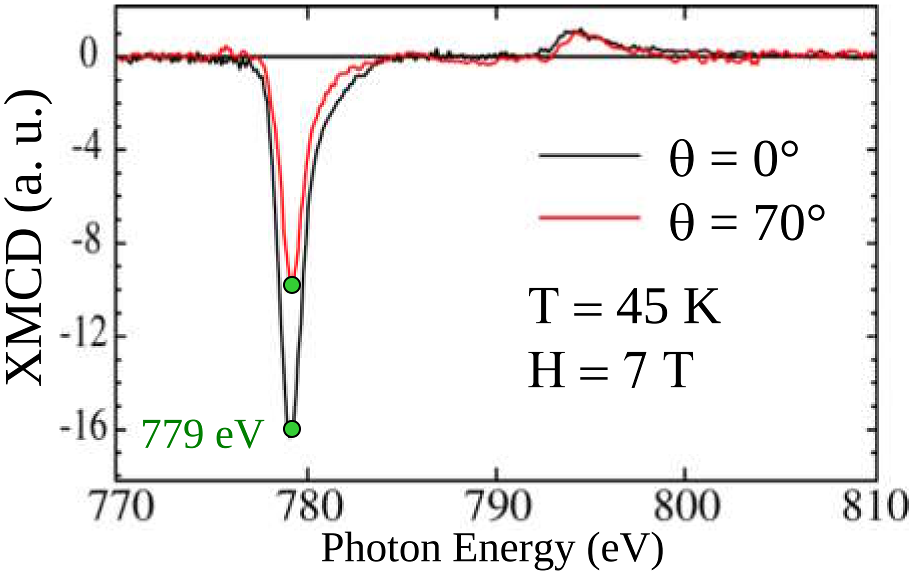
Gambardella et al. Nature **416**, 301 (2002).

Magnetization loops by XMCD

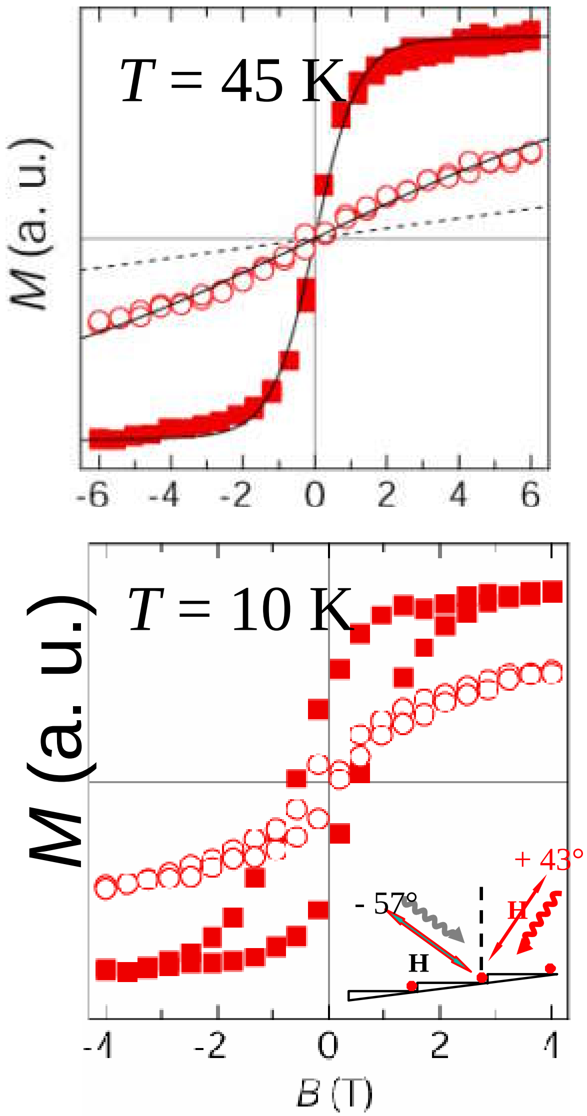
P. Gambardella et al., Nature **416**, 301 (2002)

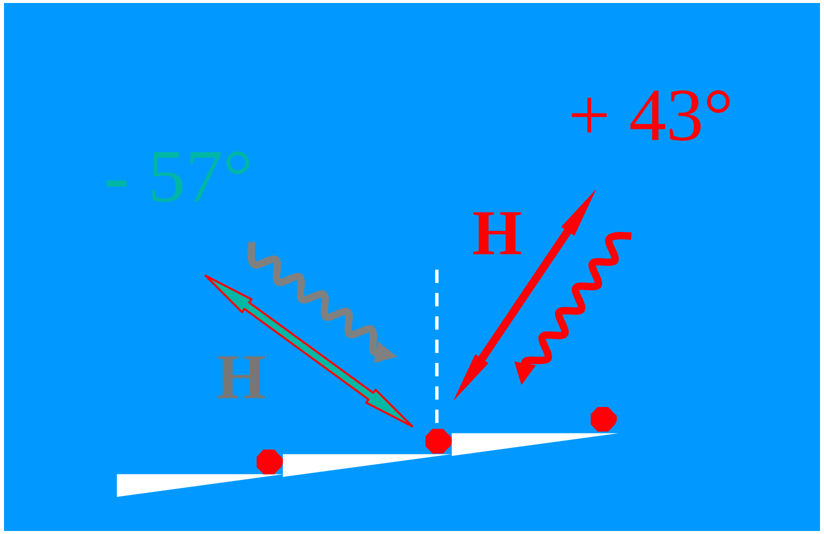






FM order in 1D





$$Energy = -N\boldsymbol{\mu}\cdot\mathbf{B} - E_a(\mathbf{easy}\cdot\boldsymbol{\mu})^2$$

P. Gambardella et al., Nature 416, 301 (2002)

Imaging use of XMCD

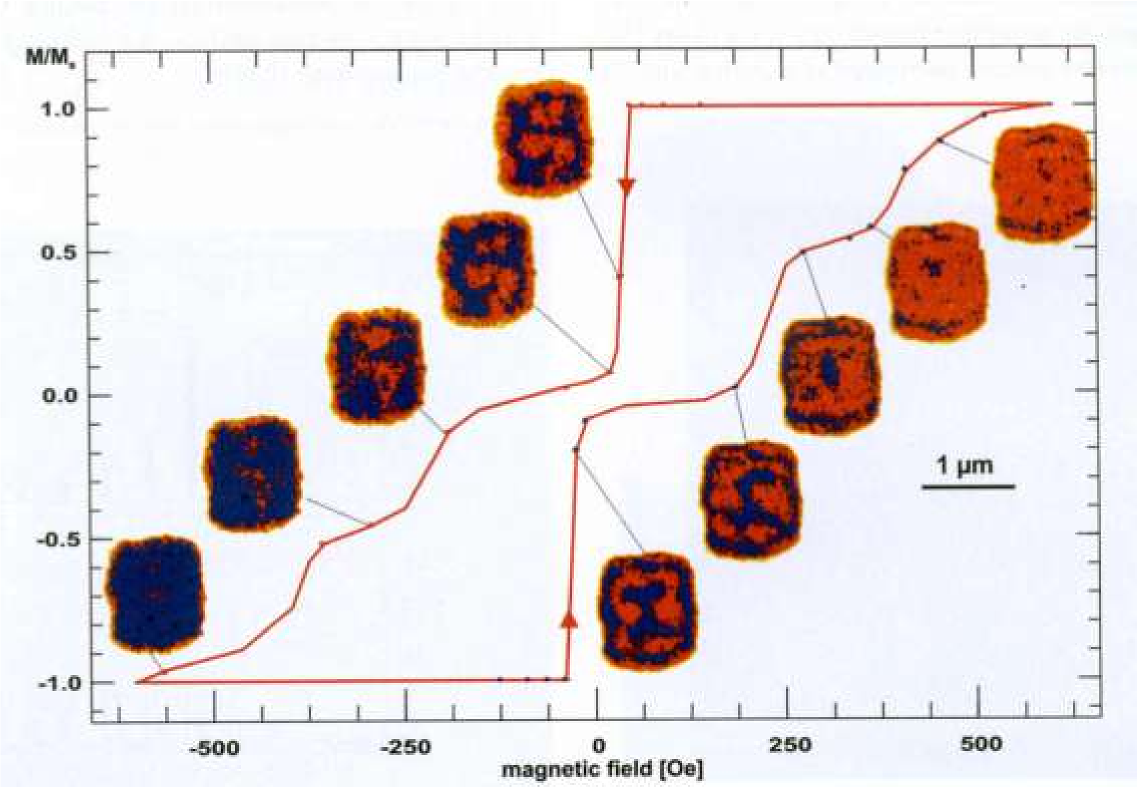
Collecting the photons:

Magnetic Transmission(soft) X-ray Microscopy (MTXM)

Collecting the electrons:

PhotoEmission Electron Microscopy (PEEM)

Magnetic Transmission X-Ray Microscopy

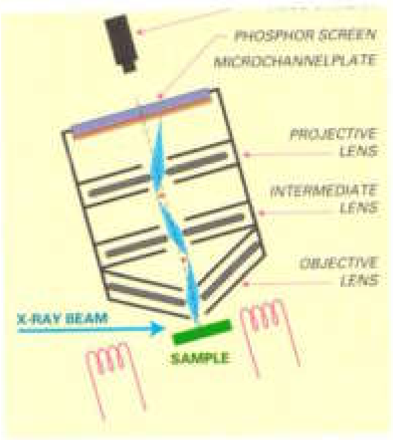


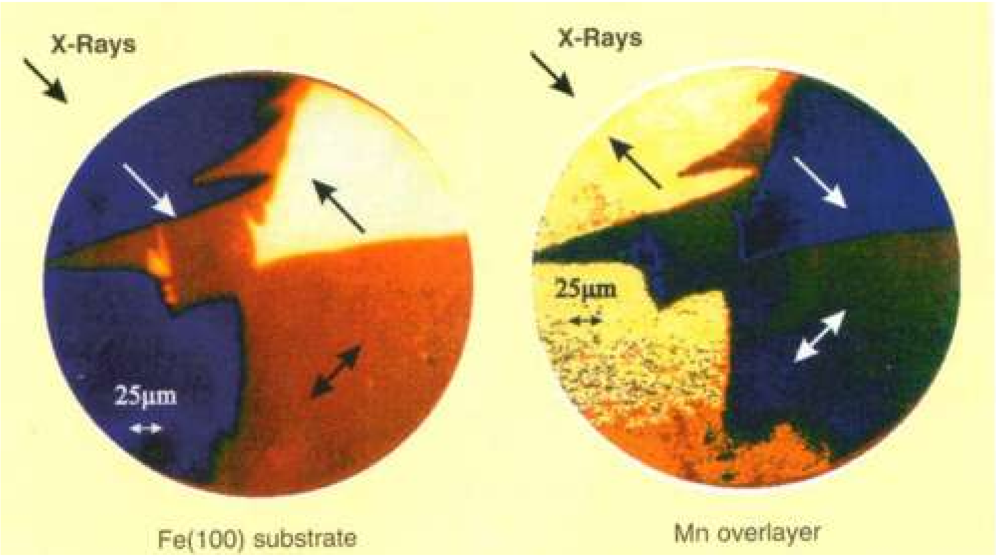
Element specific magnetization loop of an individual Fe/Gd dot, directly deduced from the presented MTXM images

T. Einmüller et al., J. Appl. Phys. ***89****, 7162 (2001)*

PEEM imaging

Photo**E**mission **E**lectron **M**icroscopy, coupled with **dichroism**, allows imaging magnetic domains in layers





0.5 monolayer of Mn on Fe substrate

Antiferromagnetic coupling between Mn and Fe

Bragg-diffraction-based Imaging Techniques

Diffraction Imaging

What is X-ray diffraction imaging (X-ray topography) ?

What can we see on a topograph ?

Order of magnitude of the distortions

What is X-ray (or neutron) diffraction imaging?

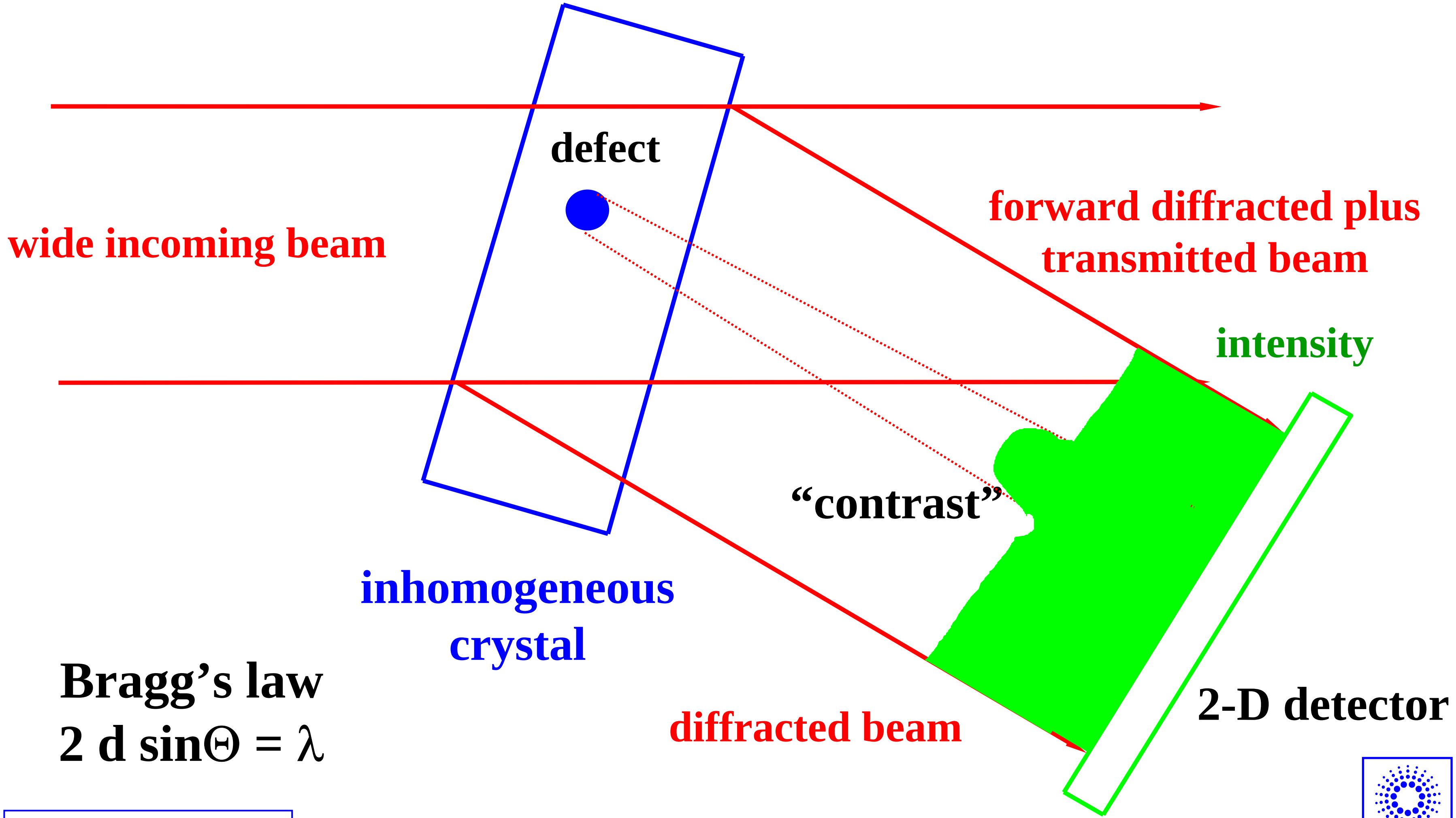
diffraction imaging

(historically called « topography »)

is an imaging technique

- **based on Bragg diffraction**
- **which applies to single crystals**
- **which shows the inhomogeneities within the crystal**

X-ray diffraction topography
basic principle for an extended, homogeneous, white or monochromatic beam



Bragg's law

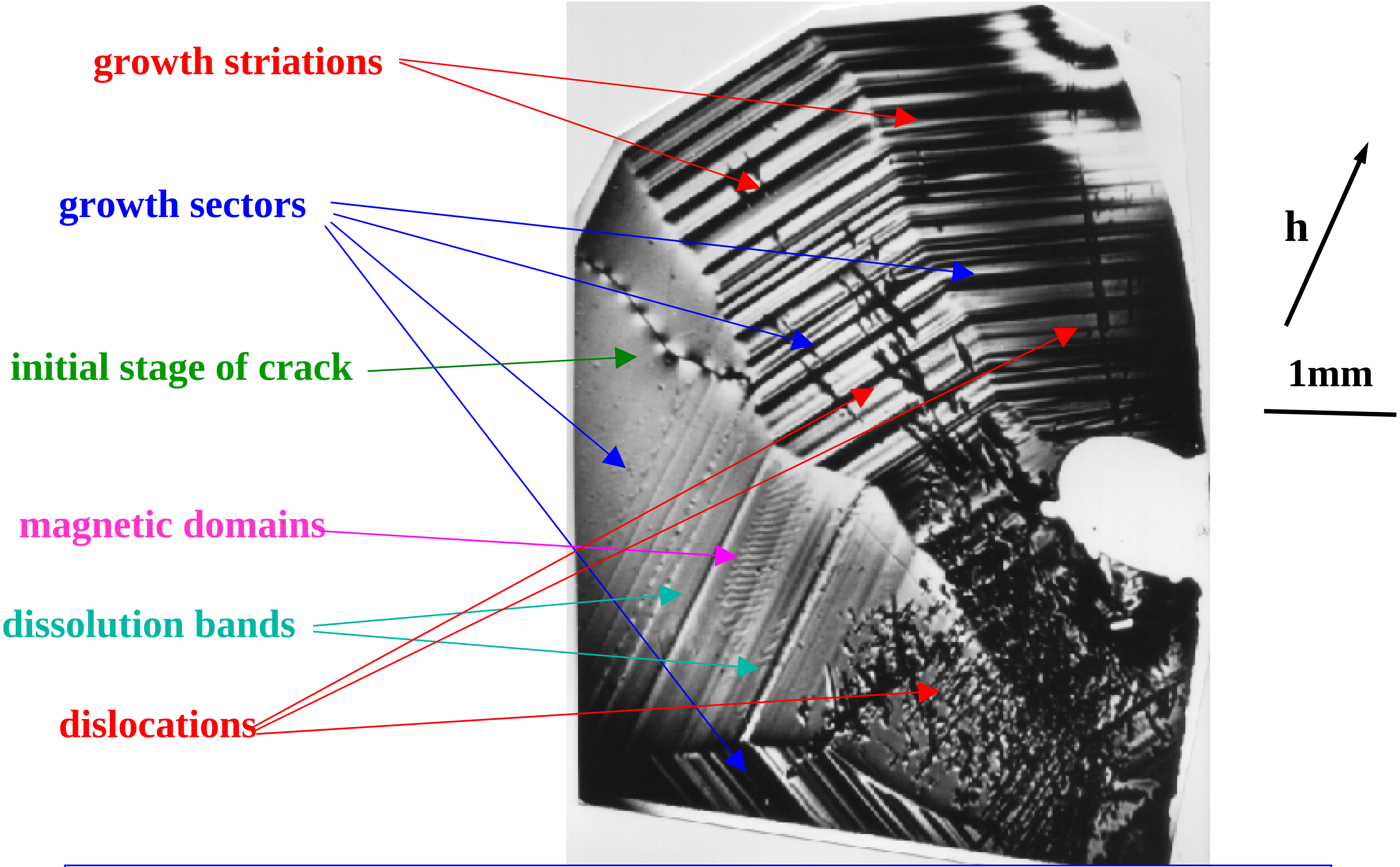
$2\,d\,\sin\Theta = \lambda$

Which inhomogeneities can be observed?

X-ray diffraction topography is used for

the visualisation of « defects » (dislocations, twins,, inclusions, impurity distribution, bending, acoustic waves, domain walls, interfaces between phases …) present within single crystal samples

How a X-ray topograph may look like:



growth striations

growth sectors

initial stage of crack

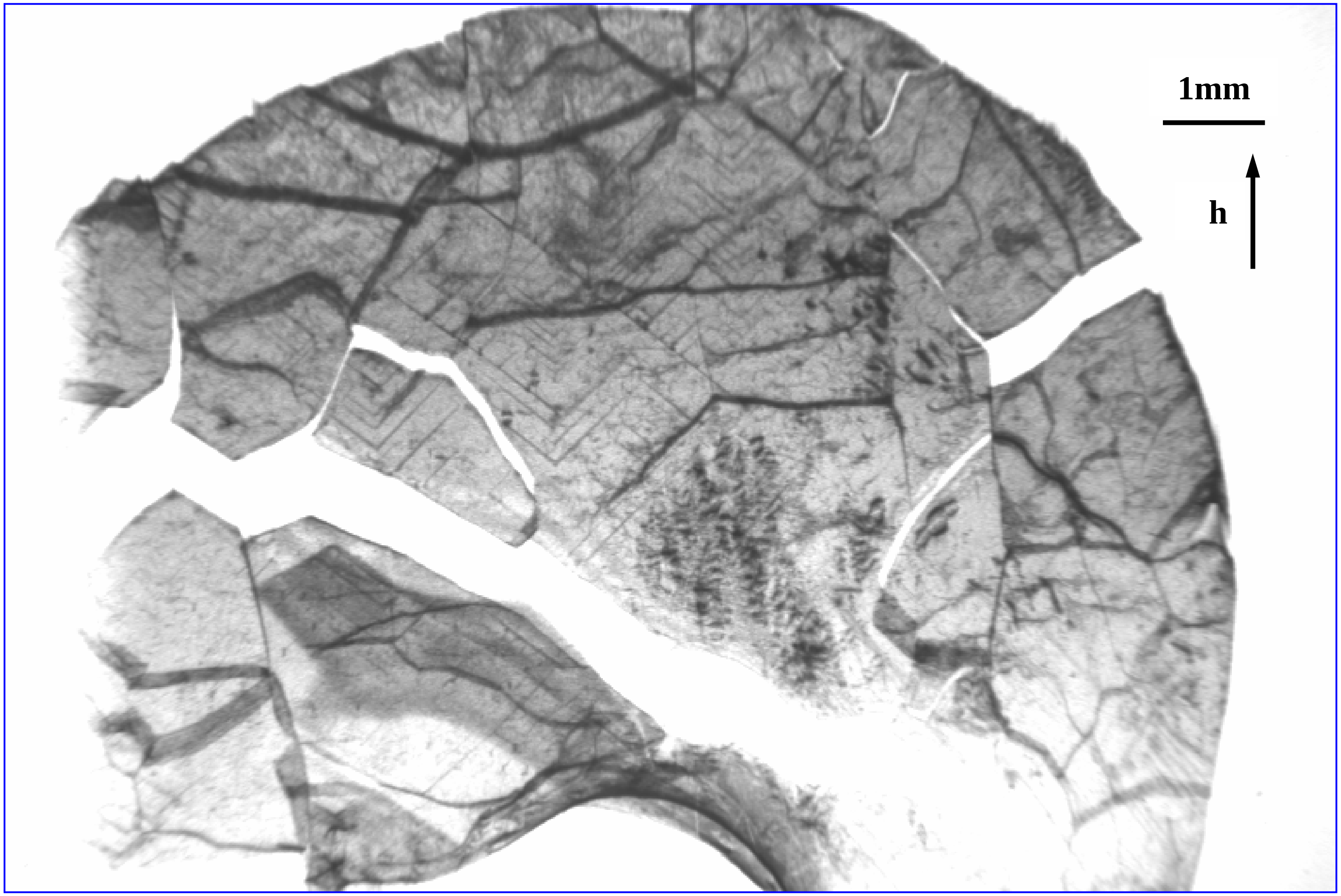
magnetic domains

dissolution bands

dislocations

Transmission X-ray topograph of a flux grown Ga-YIG ($Y_3Fe_{5-x}Ga_xO_{12}$, $x \approx 1$) crystal plate, $MoK_{\alpha 1}$-radiation ($\lambda = 0.709$ Å), 44-4 reflection

Fe-3%Si crystal



Order of magnitude of the distortions observed on the previous topographs

(either $\delta\theta$, or Δd, or combination of both)

Growth striations	#10^{-4}
magnetic domains (magnetostriction)	#10^{-6}
misorientation between subgrains	#10^{-4} -10^{-3}
region where fringes appear	<10^{-6}
dislocations	function of the distance to the core; related to the width of the image

Limits of the technique

Spatial resolution:

≈ 1µm for X-rays, ≈ 60µm for neutrons

Strains possible to detect: up to 10^{-8}

Sample dimensions:

laterally: from ~ 50µm (⇔ resolution!)
to centimeters and decimeters

thickness: ~ 1µm - 10 mm

Image formation: defects are imaged because of their different structure factor, or through their associated long-range distortion field

Basic contrast mechanisms

Inhomogeneities can show up through various mechanisms

Structure factor contrast

Orientation contrast

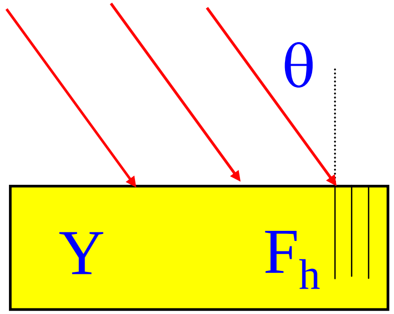
"Extinction" contrast

Basic contrast mechanisms

Effect of imperfections: contrast mechanisms (1)

Non-absorbing plate-shaped crystal illuminated by a parallel and polychromatic beam

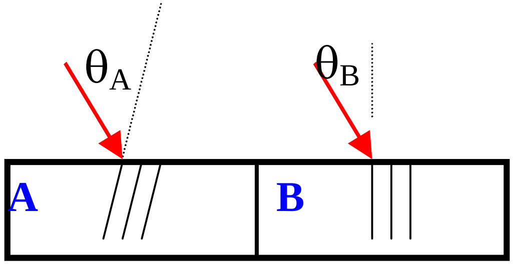
➔ the direction and intensity of the locally diffracted intensity depends upon



- ➔ θ, angle formed by the lattice planes and the beam
- ➔ F_h , structure factor
- ➔ Y, “extinction parameter” (which is intended to incorporate all the modifications introduced by the crystal inhomogeneities on the dynamical theory results)

Effect of imperfections: contrast mechanisms (2)

The beams diffracted by two neighbouring regions A and B of the sample produce contrast on the topograph if

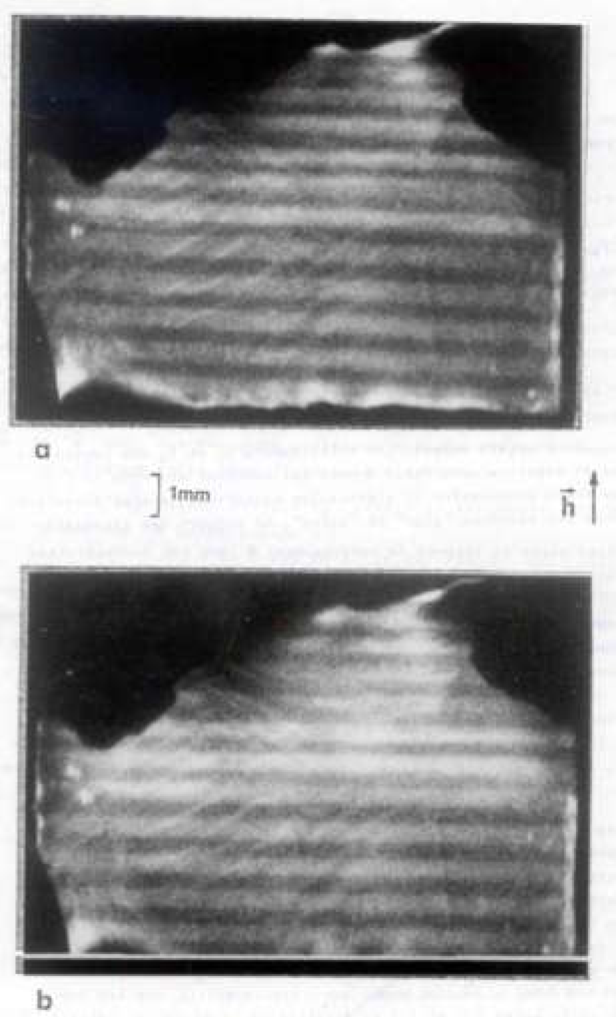


$(F_h)_A$ & $(F_h)_B$ different (modulus or phase) **structure factor contrast**

θ_A & θ_B different (regions A and B are misoriented) **orientation contrast**

Y_A & Y_B different, the two regions display **extinction contrast**

Structure factor contrast

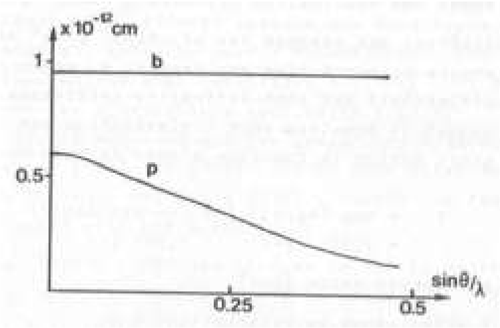


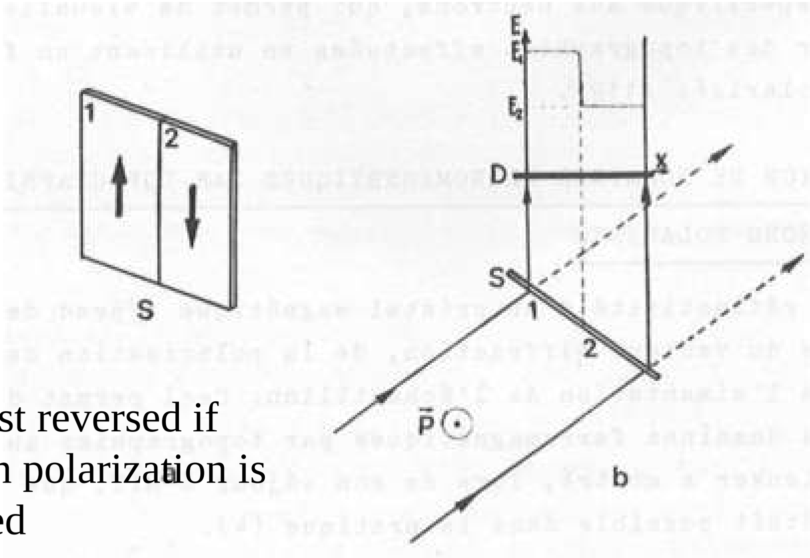
Fe-3%Si crystal

For neutrons polarized // M

F+ = b + p

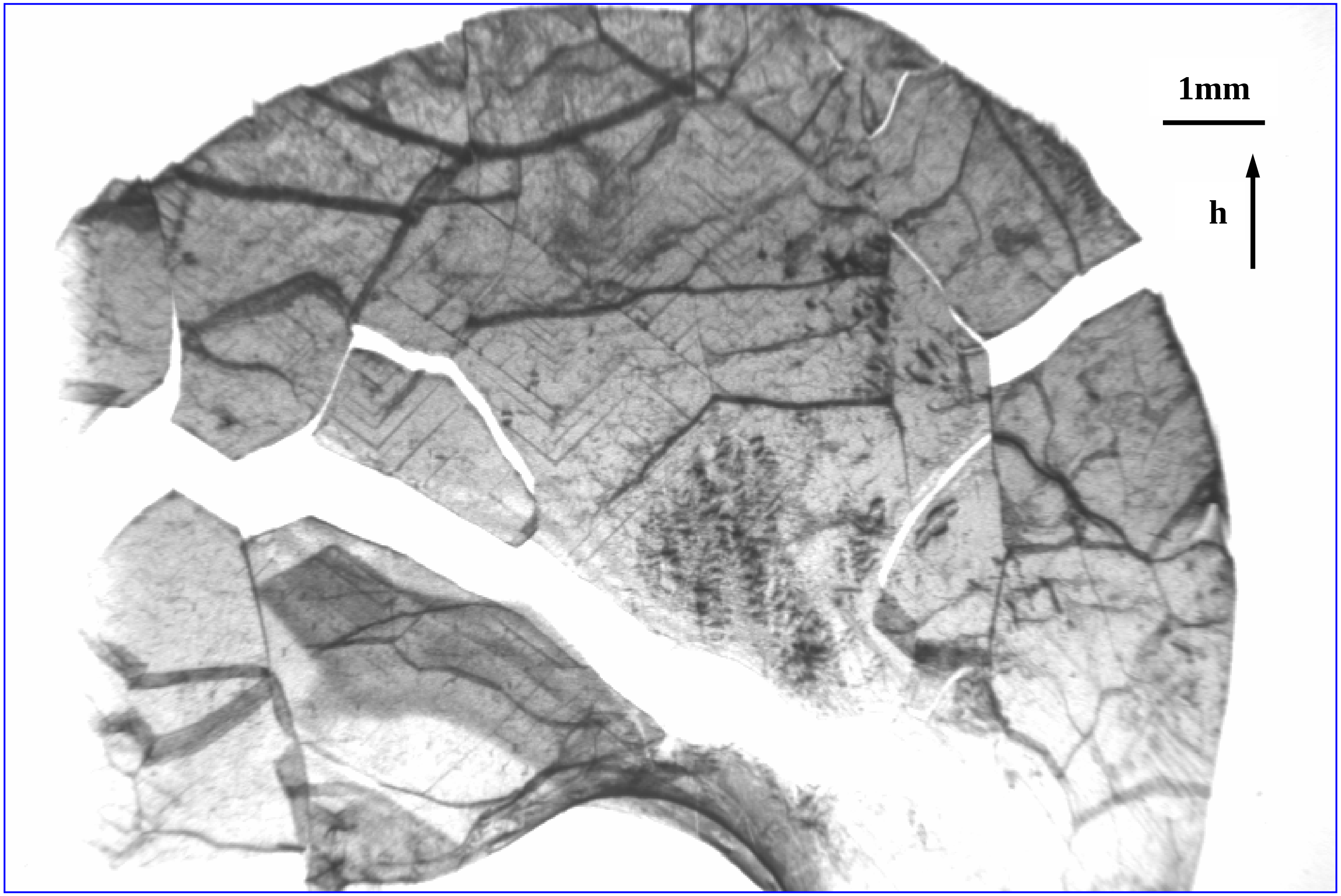
F- = b - p





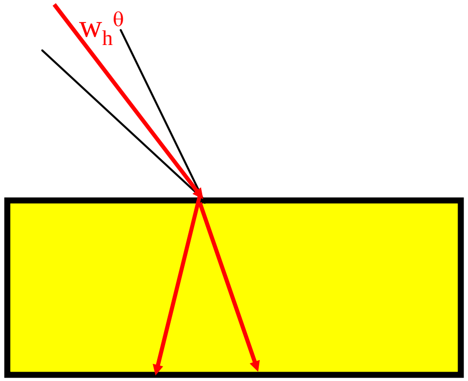
Contrast reversed if neutron polarization is reversed

Orientation contrast



Perfect crystal: Darwin width

An incident monochromatic plane wave can produce a diffracted beam within the crystal if the direction of the incident beam propagation vector lies within an angle w_h^{θ}
around the « exact Bragg position »



$$w_h^{\theta} = (2\lambda^2 C_p |F_h| r_0) / (\pi V_C \sin 2\theta_B)$$

w_h^{θ} intrinsic width of the diffraction curve (Darwin width) (# 10^{-5}-10^{-6})

C_p: polarization factor (often = 1)

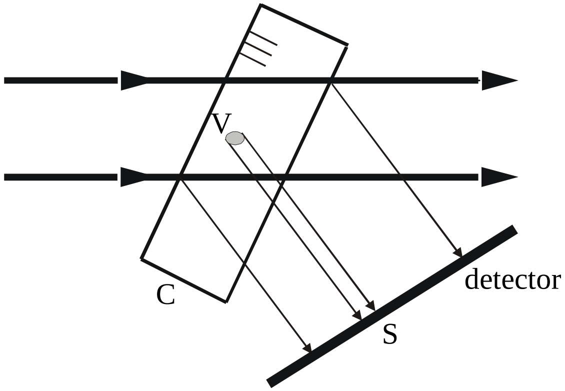
V_C : unit cell volume

F_h : structure factor corresponding to the Bragg reflection used.

Direct image mechanism

- ➔ polychromatic, parallel, incident beam

➔ V contains a defect (inclusion, dislocation,...) ➔ distortion field, which decreases with growing distance from the defect core



➔ $\Delta\lambda/\lambda = w_h^{\theta} / (\mathrm{tg}\ \theta_B)$ ($\sim 10^{-4}$) participates to diffraction by "perfect" crystal (where w_h^{θ} is the Darwin width and θ_B is the Bragg angle)

- ➔ regions around the defect ➔ Bragg position for components of the incoming beam which are outside this spectral range
- ➔ the defect thus leads to **additional diffracted intensity** on the detector

Effective misorientation

- the image of a given defect is produced by regions which are distant (μm range) from the core of the defect
- this distance depends not only on the nature of the defect, but also on the diffraction process itself
- the lattice distortion acts on diffraction through an angle, the effective misorientation $\delta\theta$, which reflects the change in the departure from Bragg angle that is associated with the existence of the defect.

$$\delta\theta = - (\lambda / \sin 2\theta_B)\, \partial(\mathbf{h}\cdot\mathbf{u})/\partial s_h$$

h : undistorted reciprocal lattice vector

u : displacement vector

$\partial/\partial s_h$:differentiation along reflected beam direction

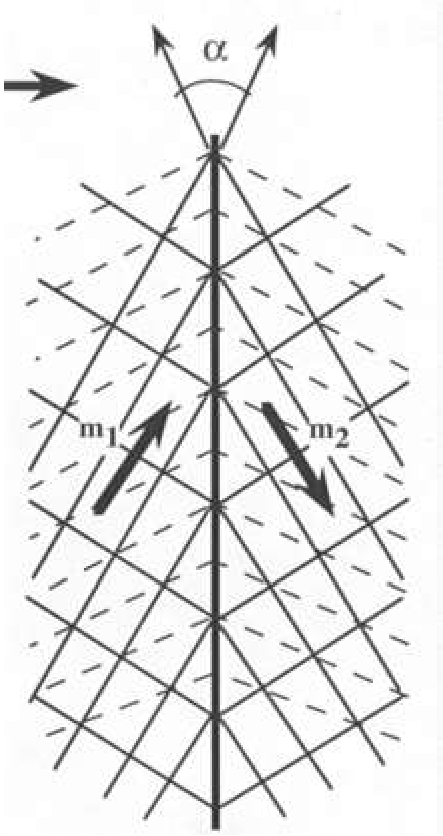
Effective misorientation in the magnetic case

Effective misorientation between two regions (domains, phases, subgrains, twins, …) which exhibit a variation in rotation $\delta\varphi$ of the corresponding planes and/or a relative variation in lattice parameter $\Delta d/d$

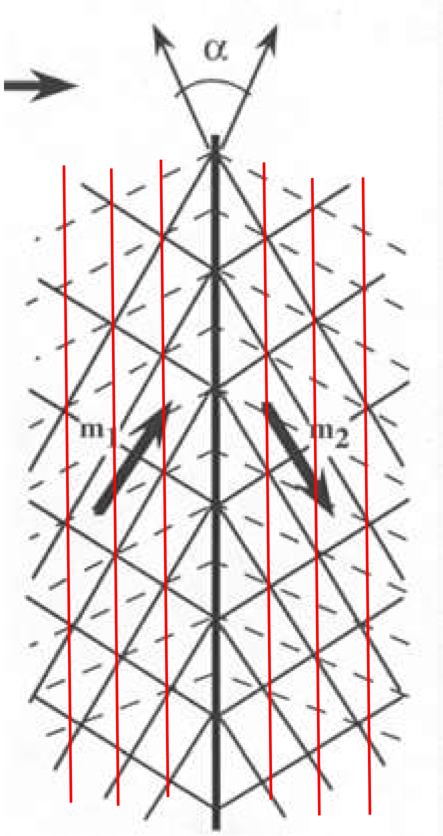
$$\delta\theta = -(\Delta d/d)\ \mathrm{tg}\ \theta_B \pm \delta\varphi$$

90° magnetic domains:

rotation α associated with the magnetostriction



Visibility (or not) of a given magnetic wall



90° magnetic domains:

rotation α associated with the magnetostriction

The effective misorientation between two domains depends on the set of reflecting planes used

$\delta\theta = 0$ for "red" planes,
(unaffected by the presence of the wall)
$\delta\theta$ not 0 for "black" planes

The specific contribution of neutrons

Neutron diffraction topography display the same advantages –and drawbacks- than neutron diffraction

Advantages

- **weak absorption by most materials**

  thick or heavy crystals

- **magnetic interaction**

Drawbacks

- **neutrons are few and expensive**

  poor spatial resolution (70-100 µm)

  long exposure times

A few examples of magnetic neutron diffraction imaging

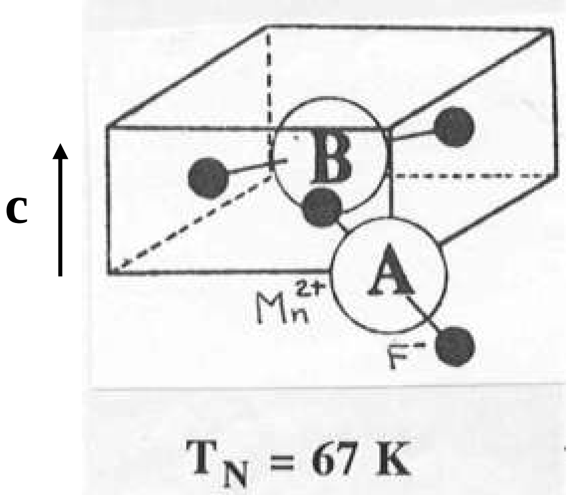
Antiferromagnetic domains in MnF_2

Chirality domains in Tb and MnP

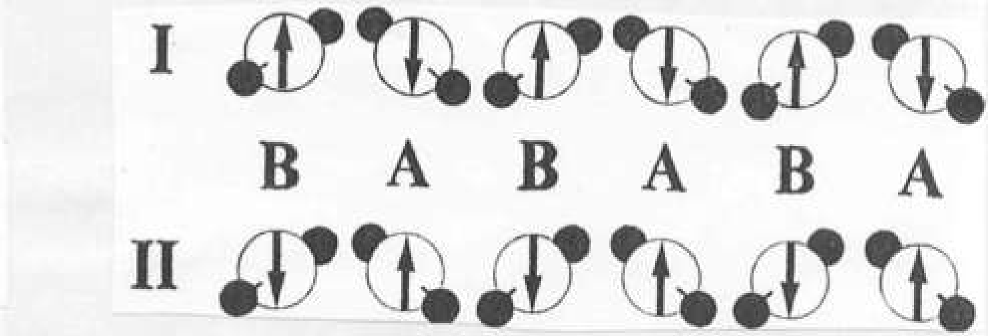
Phase coexistence in MnP

180° antiferromagnetic domains in MnF_2

The two Mn^{2+} sites A and B are not equivalent



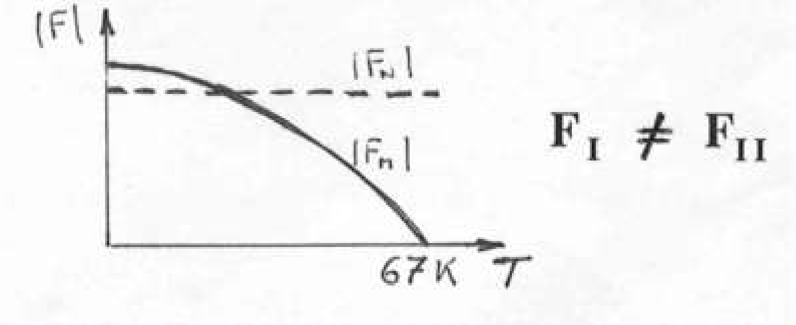
$T_N = 67$ K



two types of 180° antiferromagnetic domains

For neutrons polarized // **c**

$F_I = F_N + F_M$ and $F_{II} = F_N - F_M$



$F_I \neq F_{II}$

180° antiferromagnetic domains in MnF_2

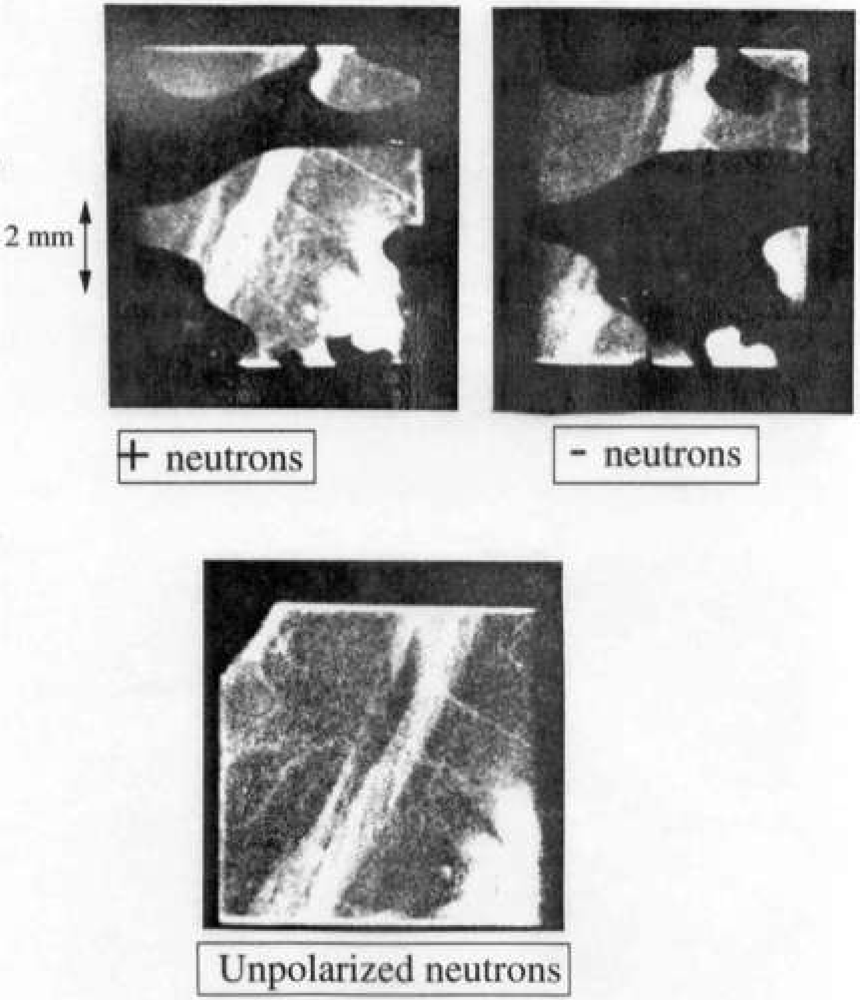
T = 20 K

Neutrons ⊕

⇨ $F_I = 0$ ⇨ $F_{II} = F_N + F_M$

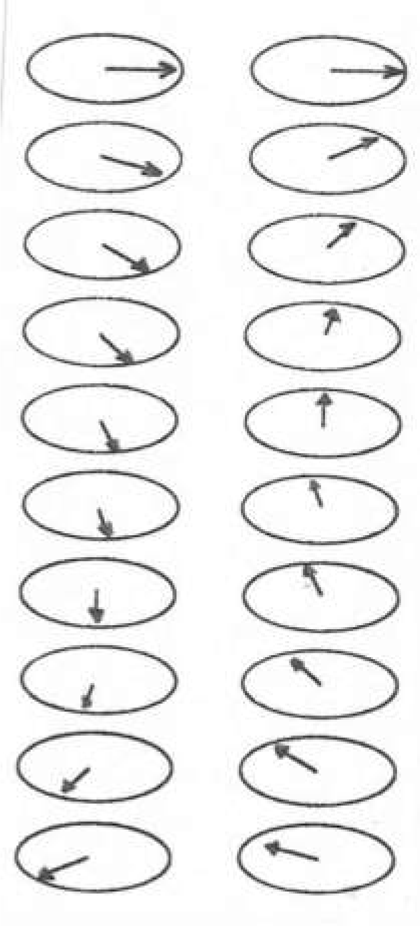
Neutrons ⊖

⇨ $F_{II} = 0$ ⇨ ,$F_I = F_N + F_M$

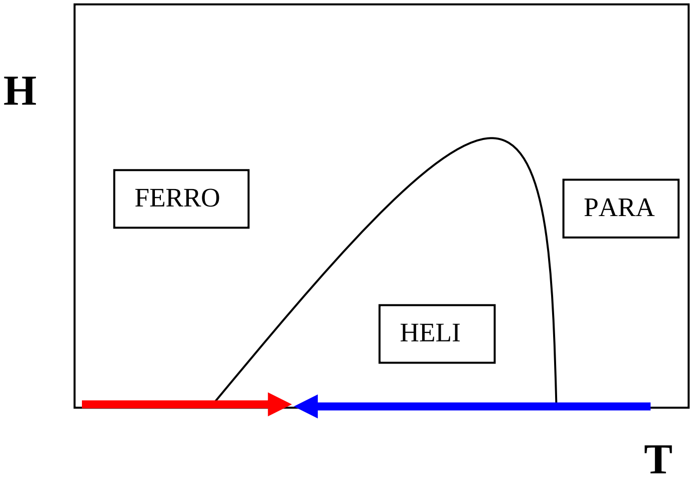


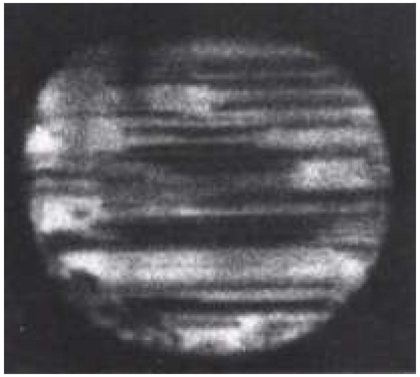
CHIRALITY DOMAINS IN TERBIUM

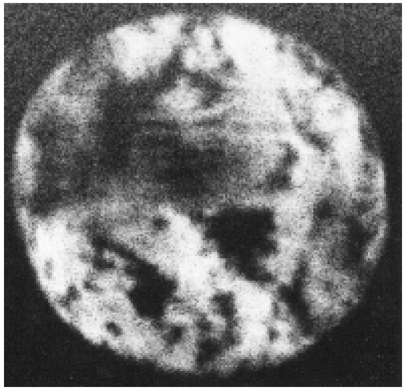
(Polarized neutrons, satellite reflections)



(H, T)
Phase
Diagram



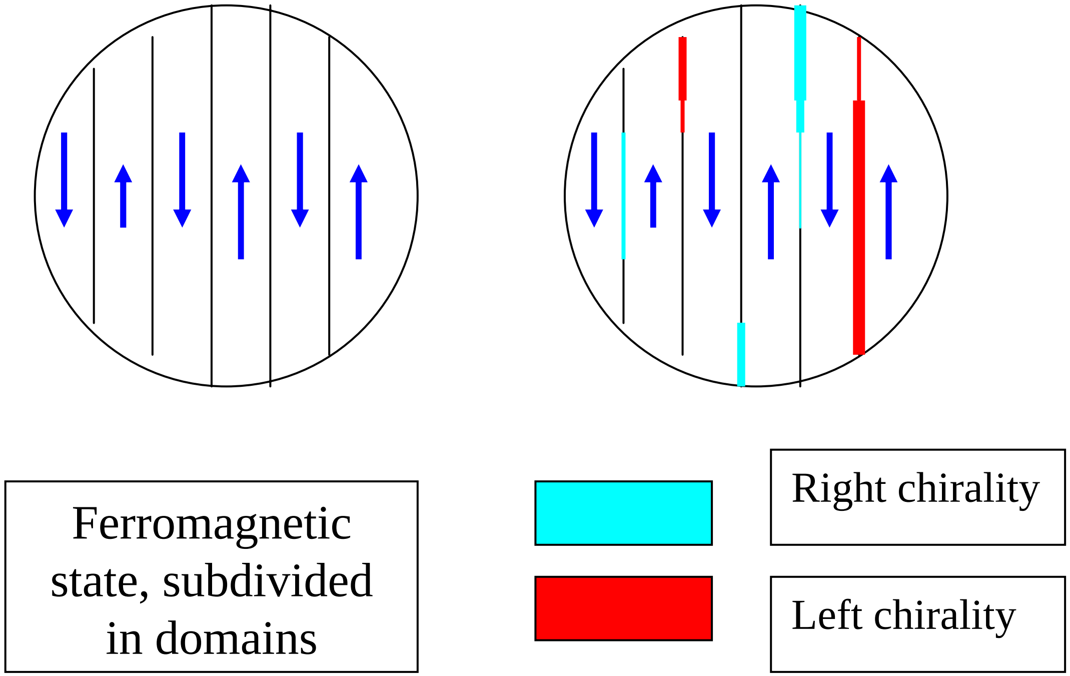




Coming from the ferromagnetic phase

Coming from the paramagnetic phase

CHIRALITY DOMAINS IN TERBIUM:
WHY THESE DIFFERENT SHAPES ?



Ferromagnetic state, subdivided in domains

🟦 Right chirality

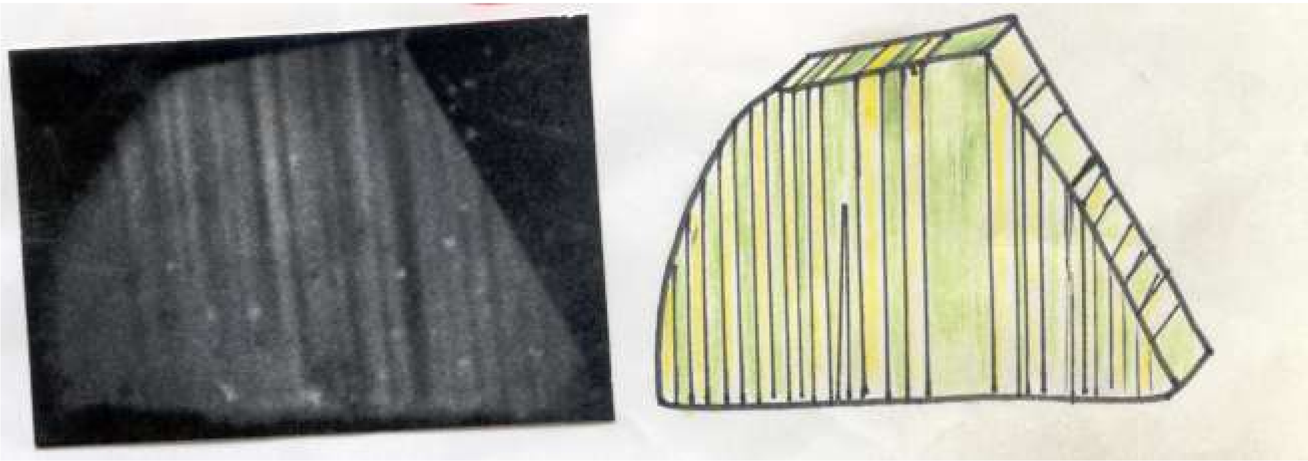
🟥 Left chirality

MEMORY EFFECTS: a given region of the sample always turn left, or turn right !!!!

Chirality domains in MnP

MnP is helimagnetic for T< 47K, ferromagnetic above 47K

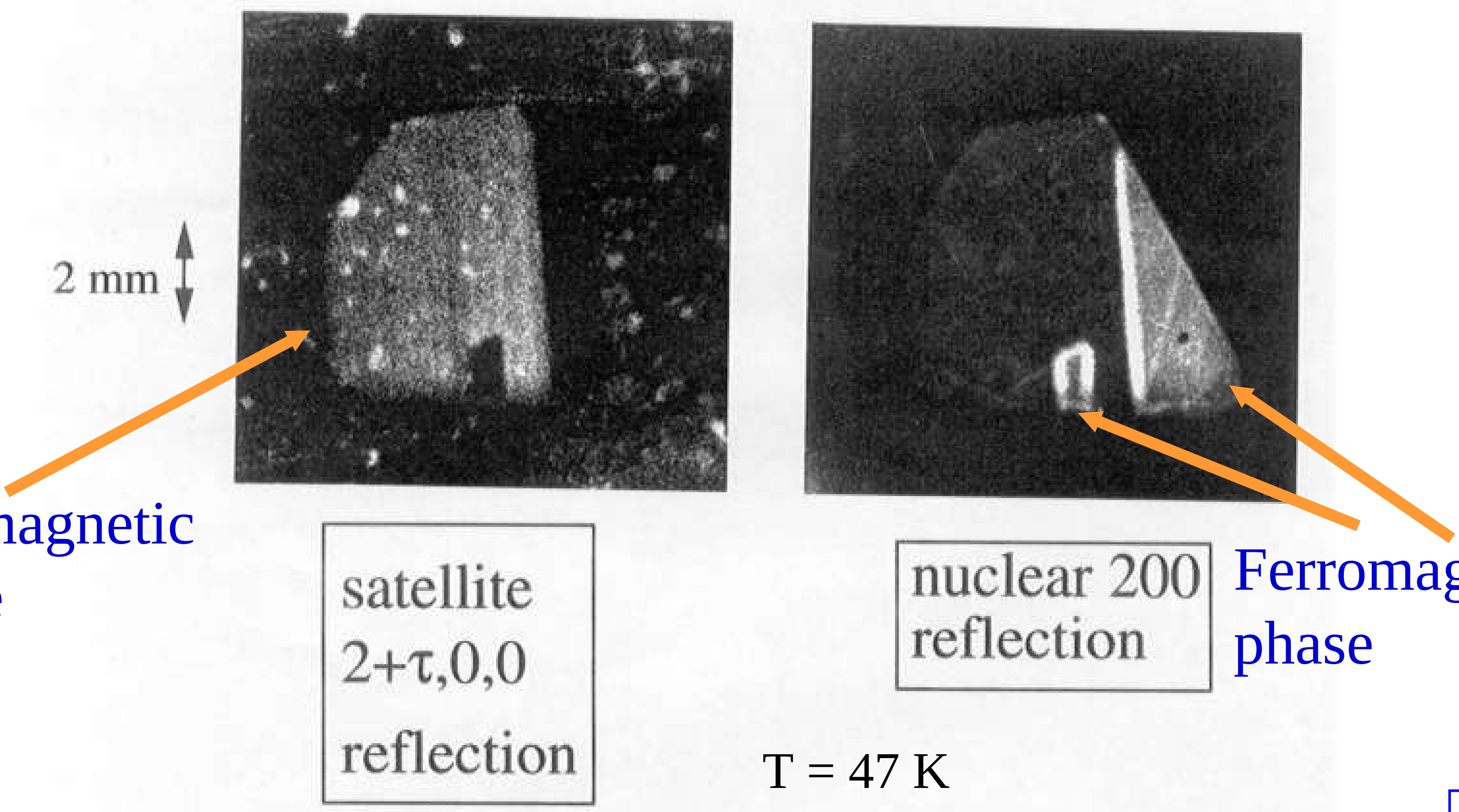
The helimagnetic phase is therefore reached, when cooling the sample, from the ferromagnetic one ⇨ stripe chirality domains



Heli-ferromagnetic phase coexistence in MnP

200 nuclear reflection F(ferro) = F_N+F_M, F(heli)= F_N

2+t,00 satellite magnetic reflection F(ferro) = 0, F(heli)= F_M



Helimagnetic phase

Ferromagnetic phase

T = 47 K

The new aspects of Bragg diffraction synchrotron radiation imaging

SR diffraction topography on magnetic materials is being used for in-situ real time experiments (for instance phase transitions) or to visualize weak effects

Mostly based on the visualization of the distortion associated with the magnetic order

Some examples of use of the (very weak) magnetic interaction of the X-rays with matter

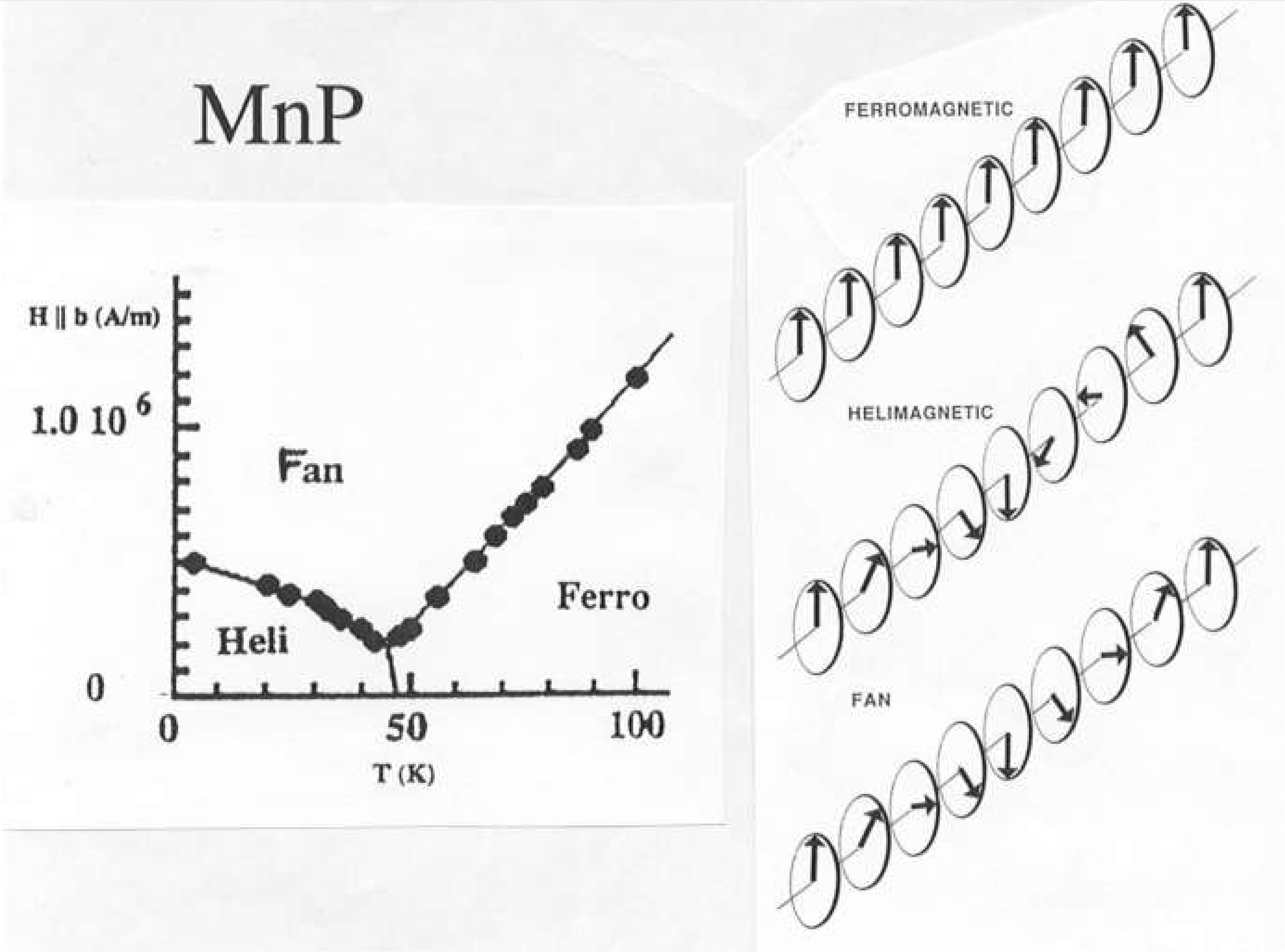
A few examples of magnetic synchrotron radiation diffraction imaging

Triple point phase coexistence in MnP

Magnetization process of hematite α-Fe_2O_3

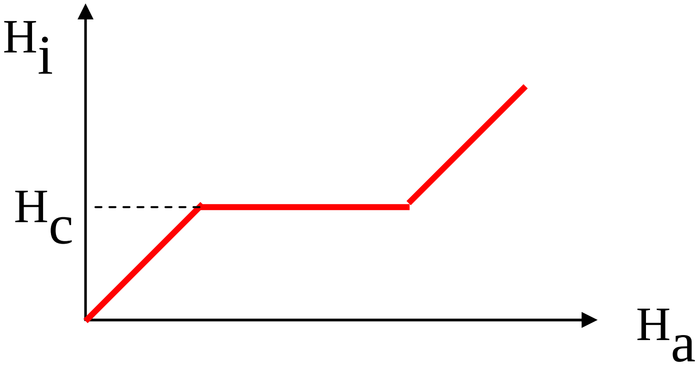
Piezomagnetic time reversal domains in CoF_2

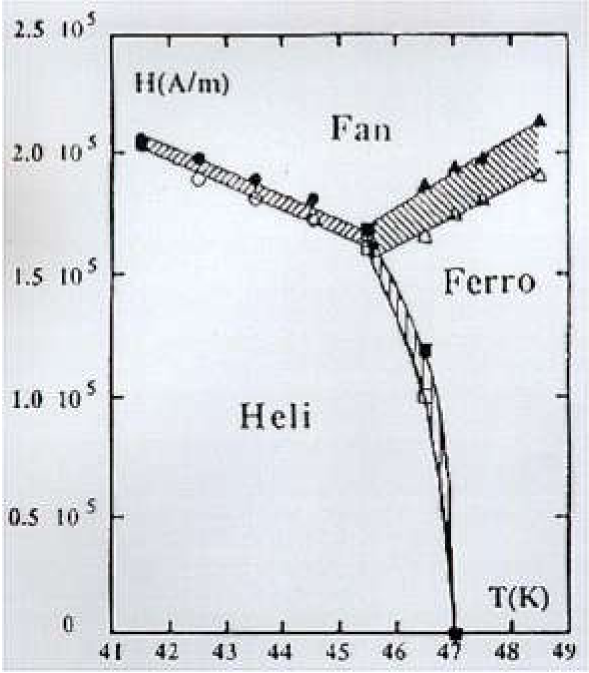
Nucleation of the fan phase at the magnetic triple point in MnP (**H**//**b**)



When considering the demagnetizing field, the **internal** field $\mathbf{H_i}$ transition lines transform into transition regions (corresponding to a constant internal field $\mathbf{H_c}$) as a function of the **applied** field $\mathbf{H_a}$

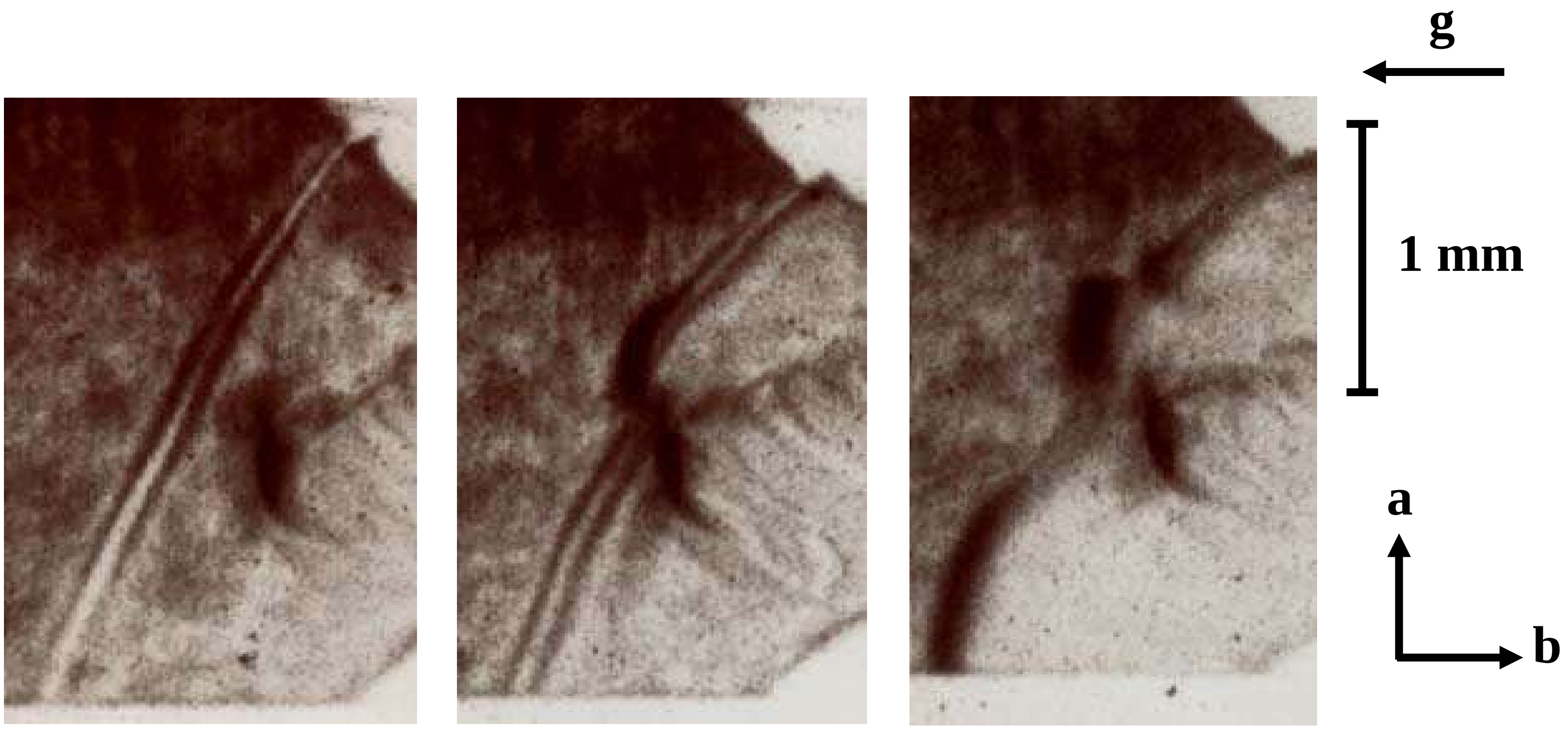
⇨ the triple point actually is a coexistence region





Phase nucleation at a triple point

Nucleation of fan phase in presence of the helimagnetic and ferro ones occurs at the heli-ferromagnetic interface



a b c

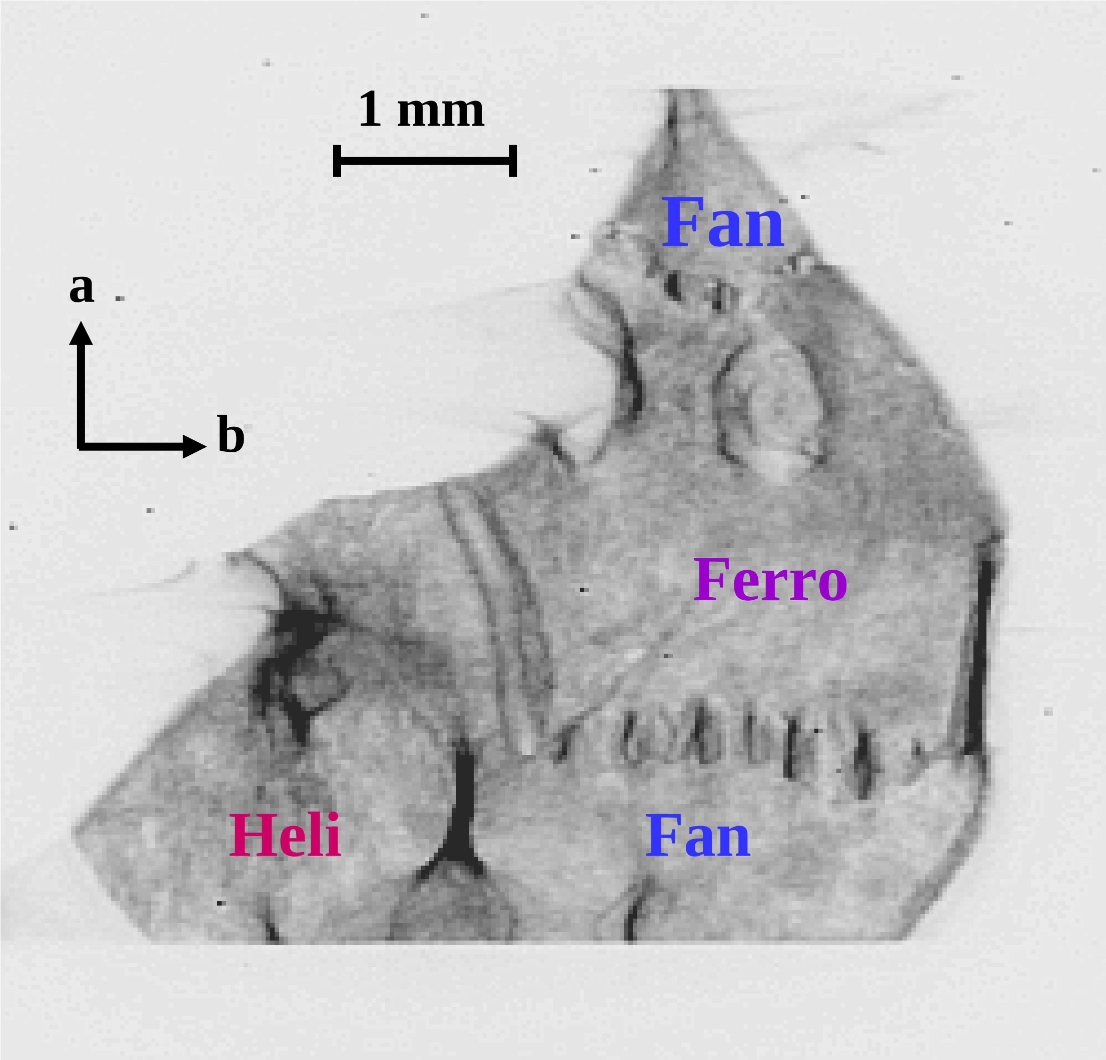
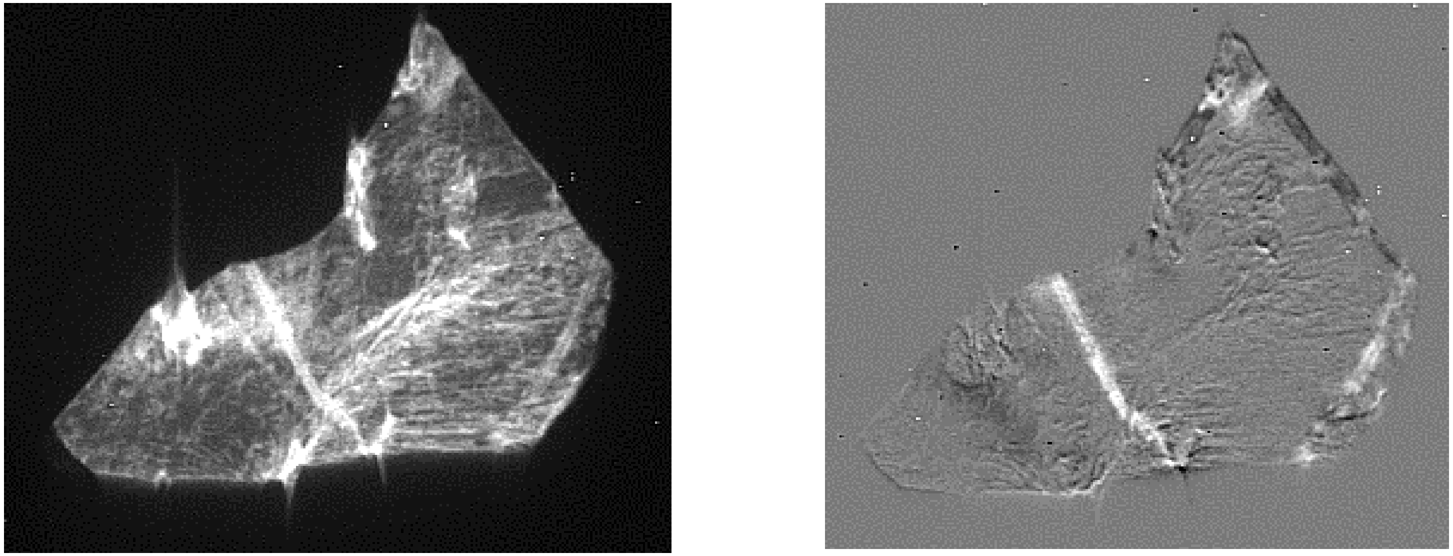


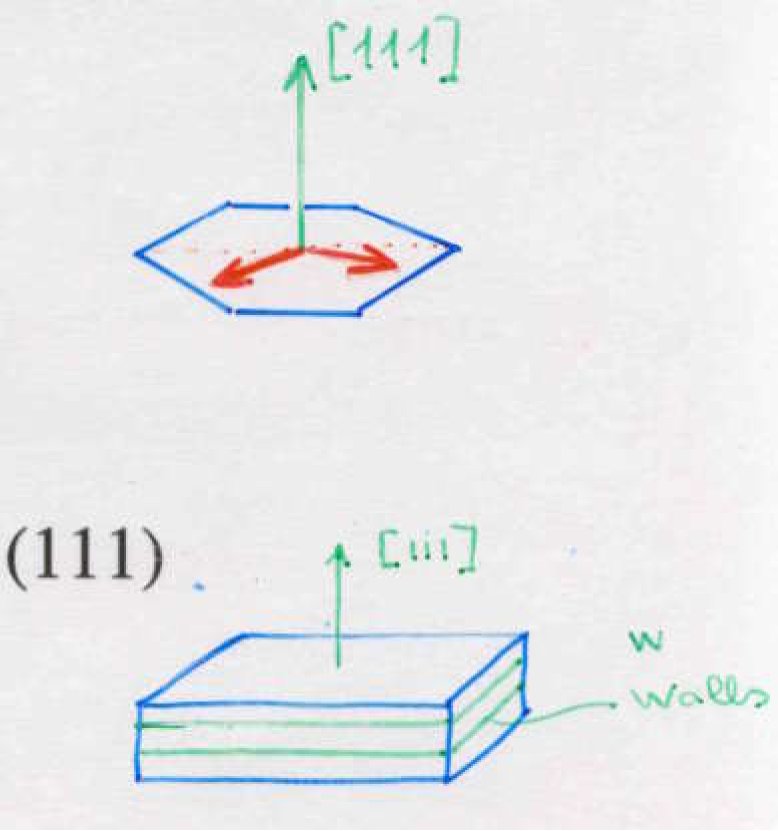
Image collected on a CCD camera, showing the three phases present in the crystal



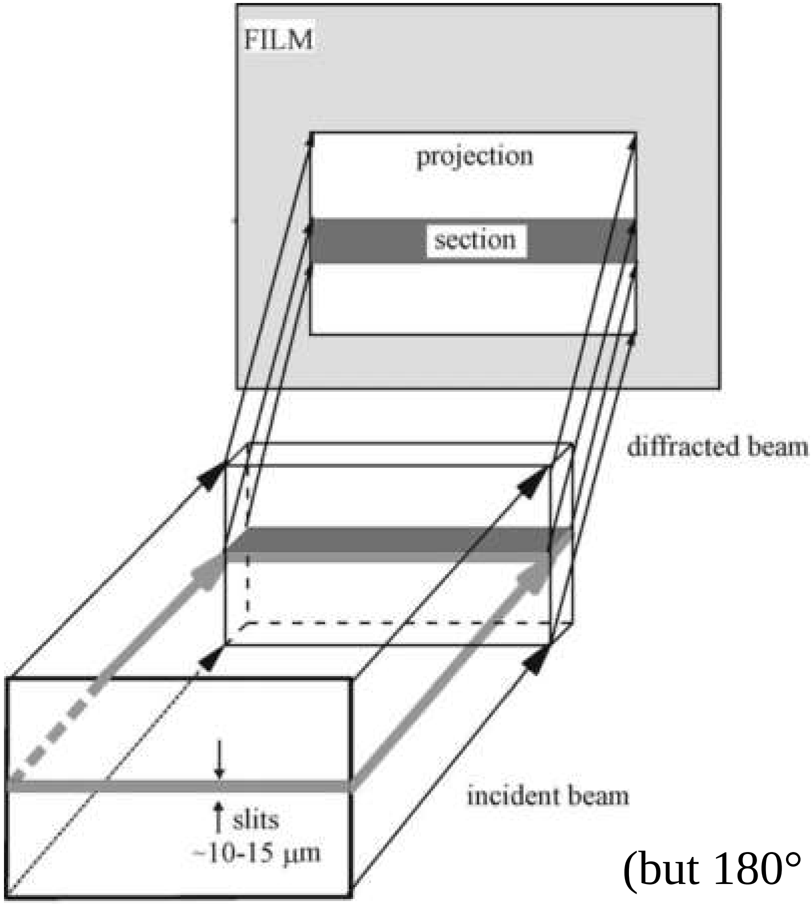
Left: image during the transition, Right: image difference of the first one and the image collected far from the phase transition

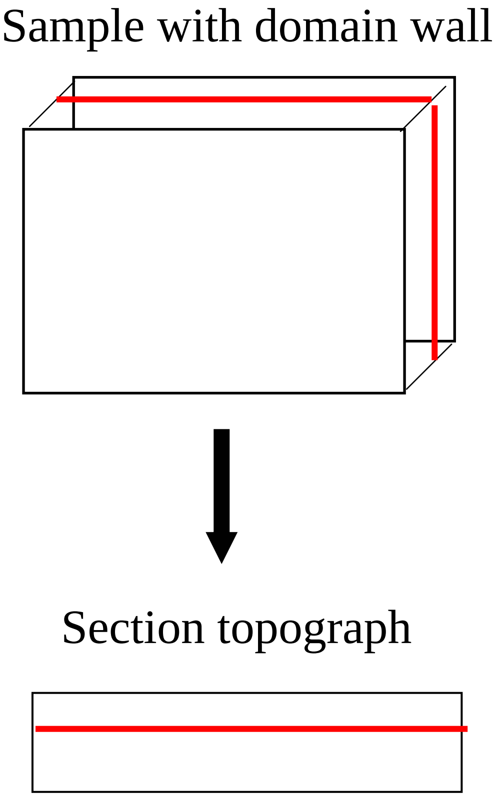
Magnetization process of hematite (α-Fe_2O_3)

- ➩Rhombohedral weak ferromagnet
- ➩(111) easy plane
- ➩Vanishing in-plane anisotropy
- ➩Domain walls almost // (111)
- ➩Growth: (111) platelets



Using « section » topographs to observe the walls

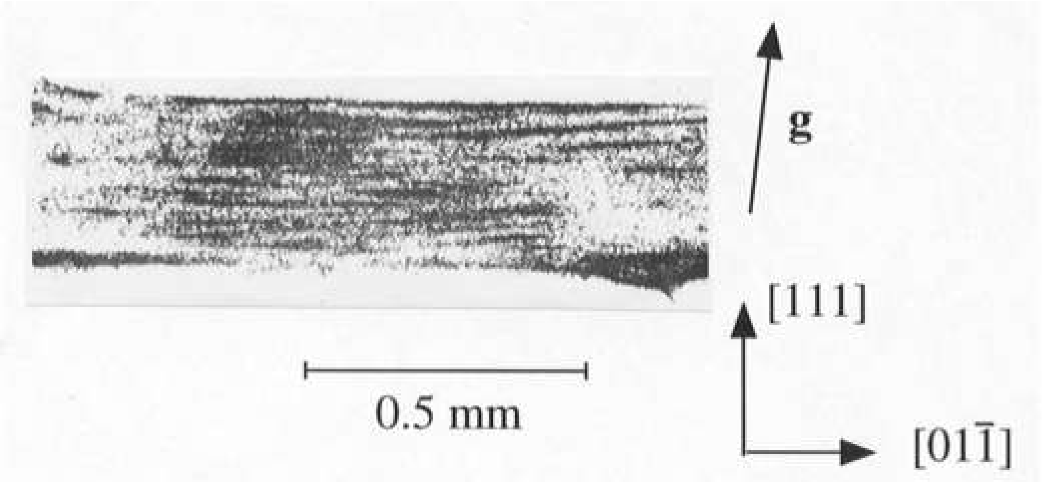



Sample with domain wall

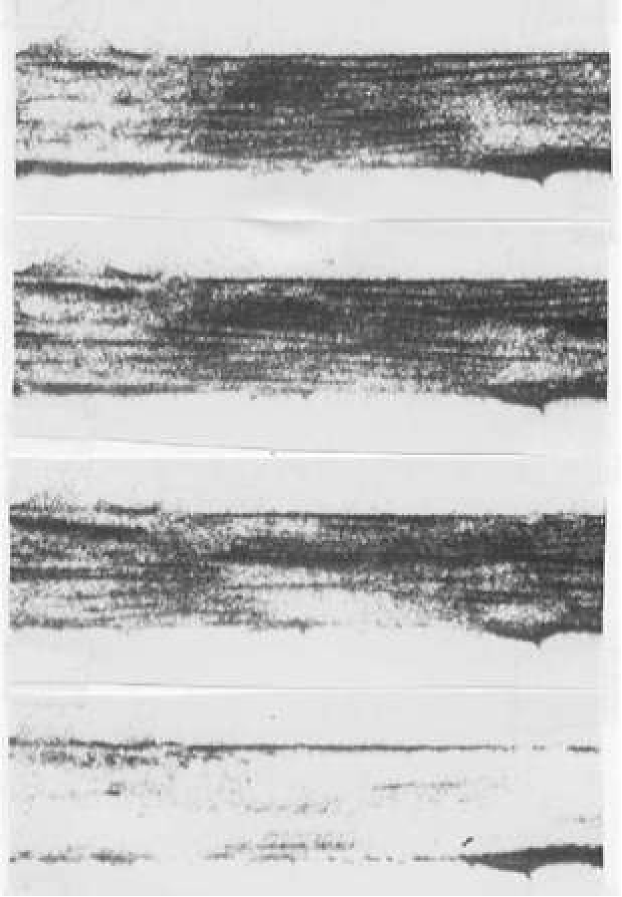
Section topograph

(but 180° domains induce no difference in magnetostriction, and should therefore not be visible)

Domains are visible on the section topographs



Evolution as a function of the applied field

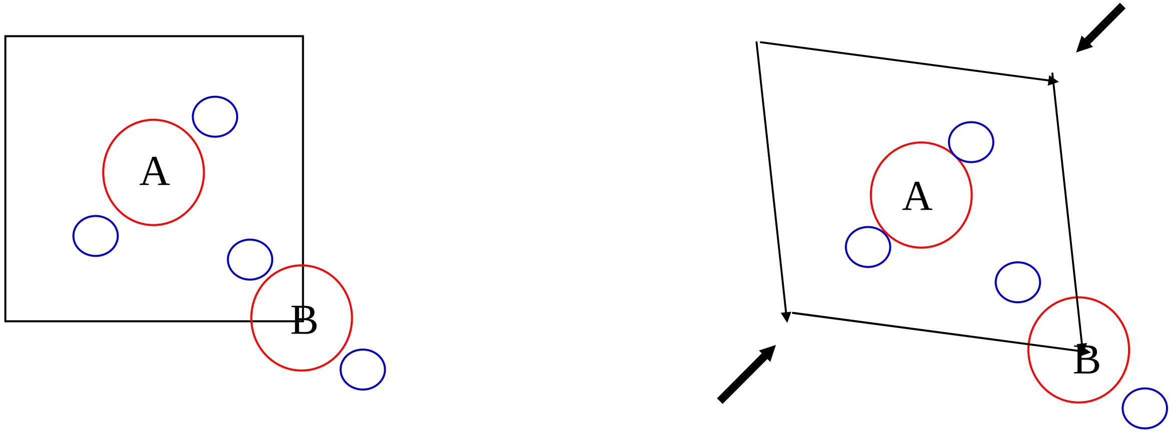


- ➪ wall parts are visible on a given image
- ➪ The visible parts evolve with the field
- ➪ No walls are visible at the saturation field

Interpretation

anisotropy very small ➪ magnetization process do not occur through wall movements but by a rotation of the magnetization within the domains (which are not anymore 180° domains) up to the complete alignment of the magnetization along the field

Piezomagnetic domains in CoF_2



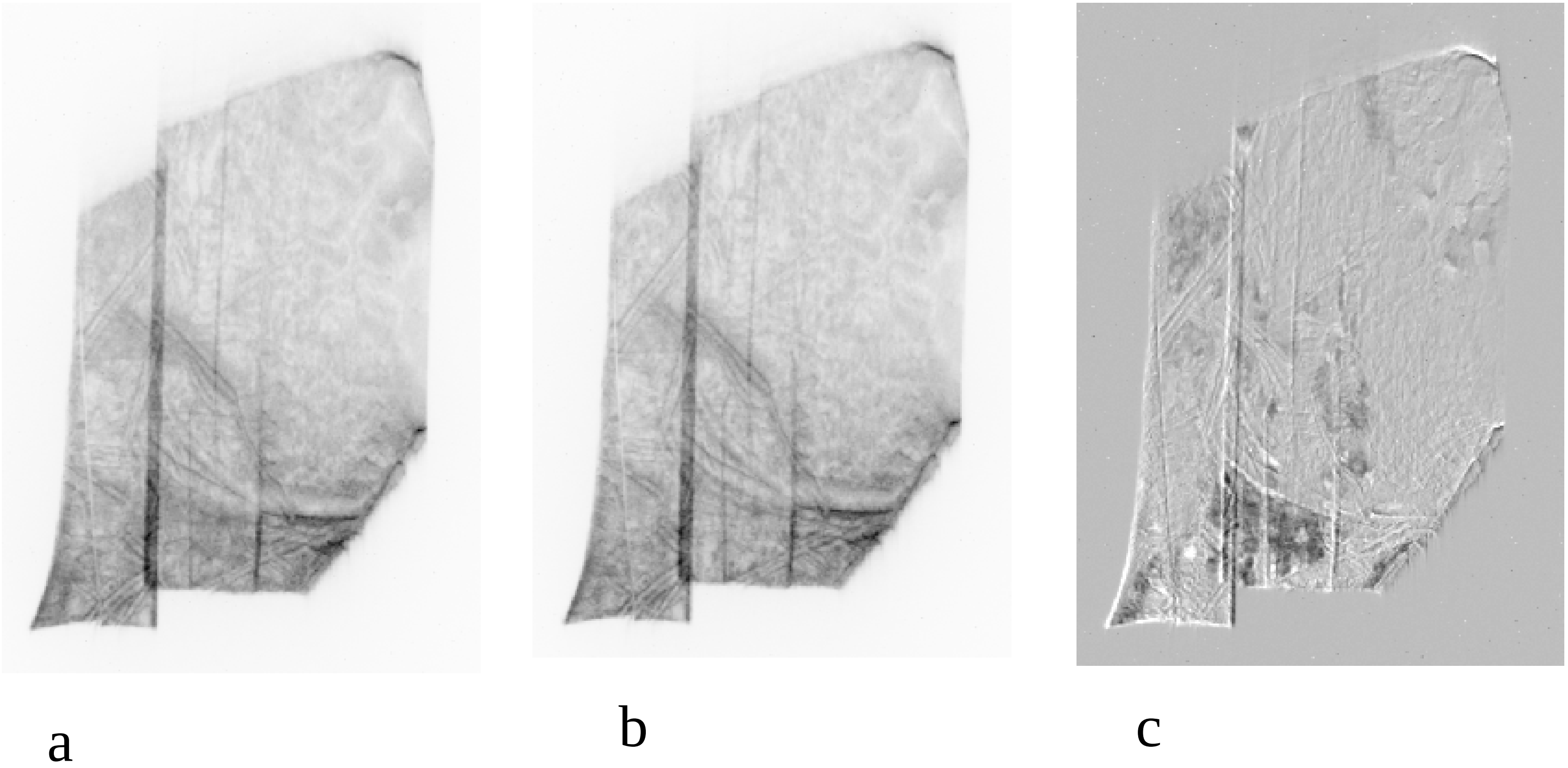
Environment of the sublattices A and B is changed by the applied stress

⇨ $M_k = P_{ijk} \sigma_{ij}$

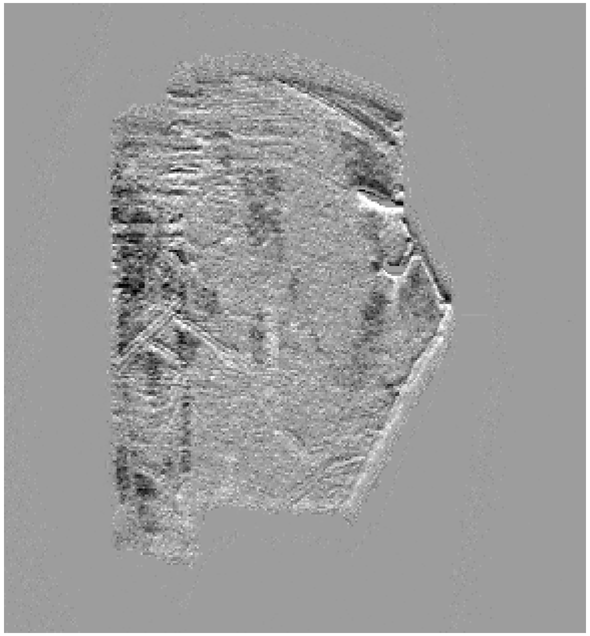
Inverse effect: field ⇨ magnetization ⇨ distortion of the sample

The sign of the magnetization (and of the distortion) is opposite for opposite domains ⇨ possibility of visualizing them

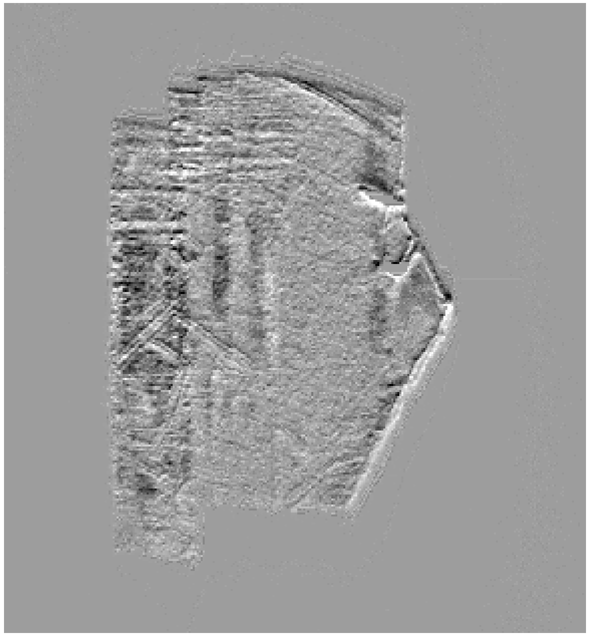
Images of a CoF_2 crystal without and under field, and difference image showing the piezomagnetic domains



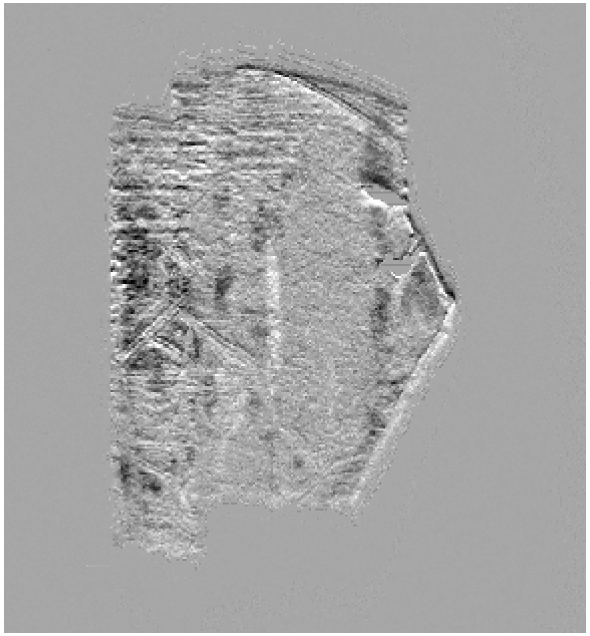
Memory effect



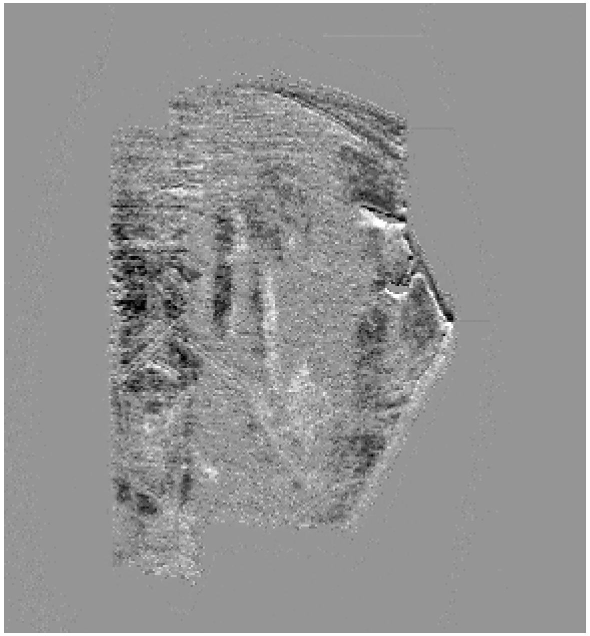
As cooled to 10 K



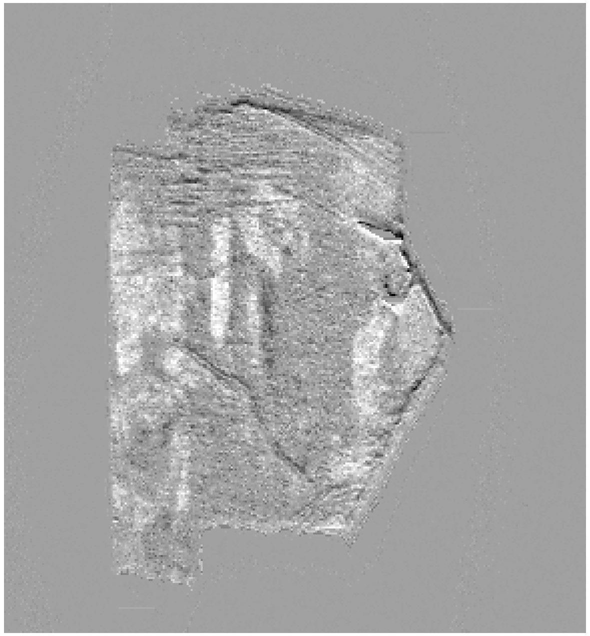
heated to 47 K and cooled to 10 K



heated to 67 K and cooled to 10 K



Field cooled +
Applied field +



Field cooled +
Applied field -

The piezomagnetic distortion is reversed when the applied field direction is reversed

⇨ this leads to the reversal of the contrast of the images

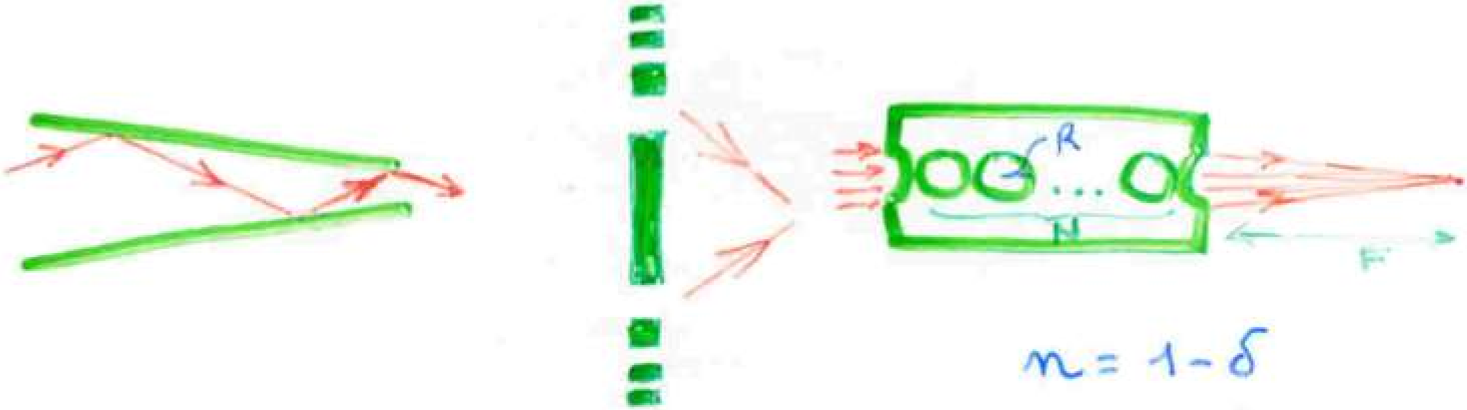
Microbeam-based Imaging Techniques

focalisation of hard X-rays
3 keV < E < 100 keV

Conical capillaries

Fresnel lenses

Refractive lenses



0.2 to 2 µm

0.1 to few µm

$n = 1 - \delta$

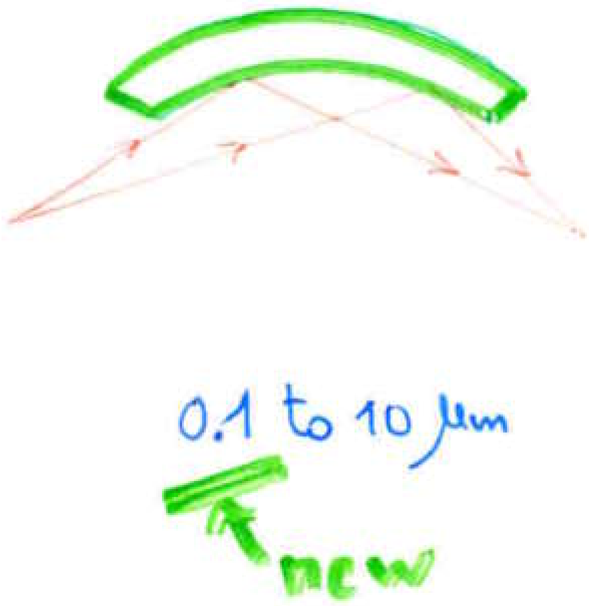
$F = \frac{R}{2 . N \delta}$

1 to some µm

Kirkpatrick-Baez arrangement:

two bent devices in perpendicular planes

Mirors
multilayers
bent crystals



Domains in Cr (Evans et al.)

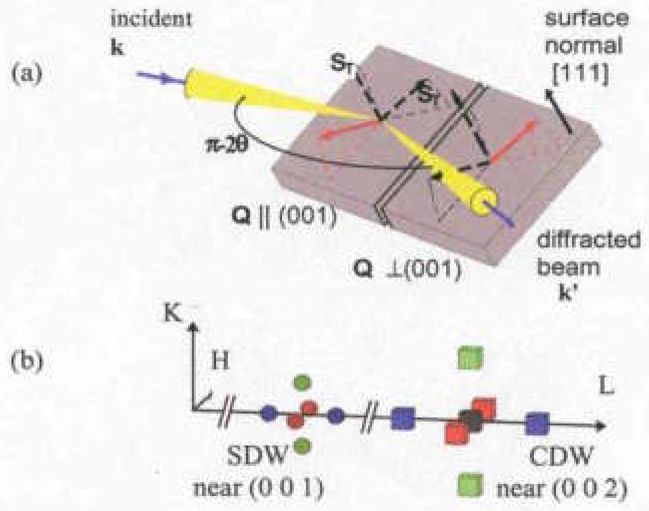


FIG. 1. (a) The (111)-oriented Cr sample was mounted so that the [001] axis was in the plane defined by incident and diffracted beams **k** *and* **k'**. *The normal to the diffraction plane was along the sample's [110] axis. (b) SDW reflections (spheres) appear in reciprocal space near (001) and can be divided into pairs corresponding to domains of* **Q** *along [100] (red), [010] (green), and [001] (blue). Corresponding reflections arising from the charge density wave (CDW) and strain wave appear near (002) (cubes).*

Scanning images of domains in Cr

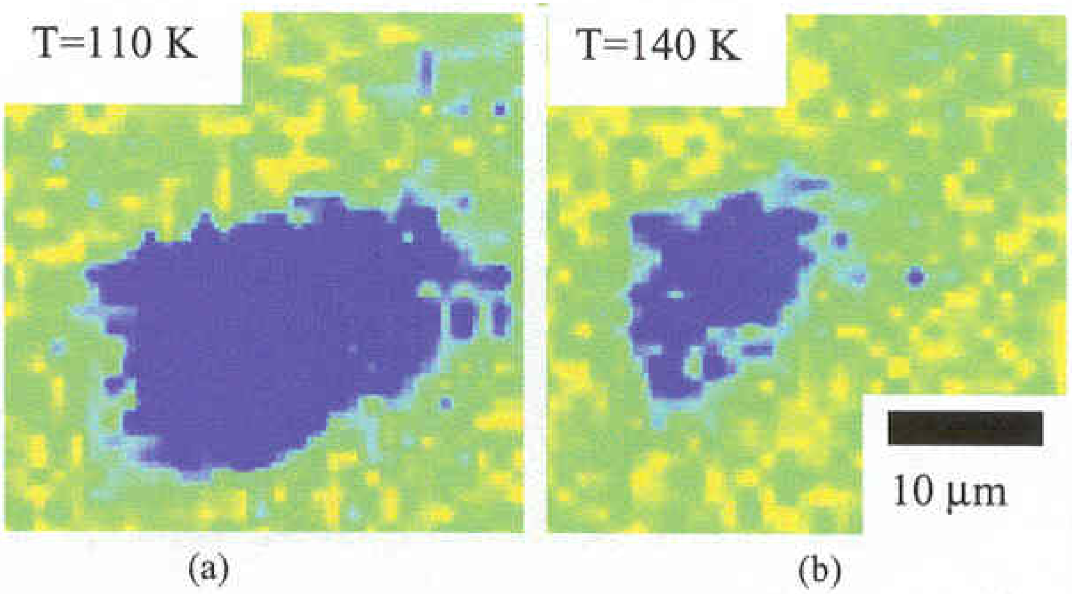


*FIG. 3. Images at (a) 110K and (b) 140K of the intensity of the (δ, 0, 1) reflection as a function of the position of the sample. At 110K, the entire **Q** domain is visible, but at 140K, only the transverse SDW domain with S along [010] is visible.*

Perspectives

XMCD is a unique technique to investigate domains in thin films and multilayers; very important for applications

Diffraction imaging (neutrons, SR) allows in situ, real time experiments and the possibility of imaging very weak peaks

⇨all kind of exotic domains, restricted to (bulk) single crystals

Conclusion

« Magnetic X-ray Imaging » covers a series of evolving techniques (beam, detectors, computers, …) which apply to a wide number of topics…

…maybe yours!!