



XVIII School on Synchrotron Radiation
"Gilberto Vlaic":
Fundamentals, Methods and Applications
Muggia (Trieste), Italy
21 September – 1 October September 2026

CONDENSED MATTER STUDIES WITH POLARIZED X-RAY

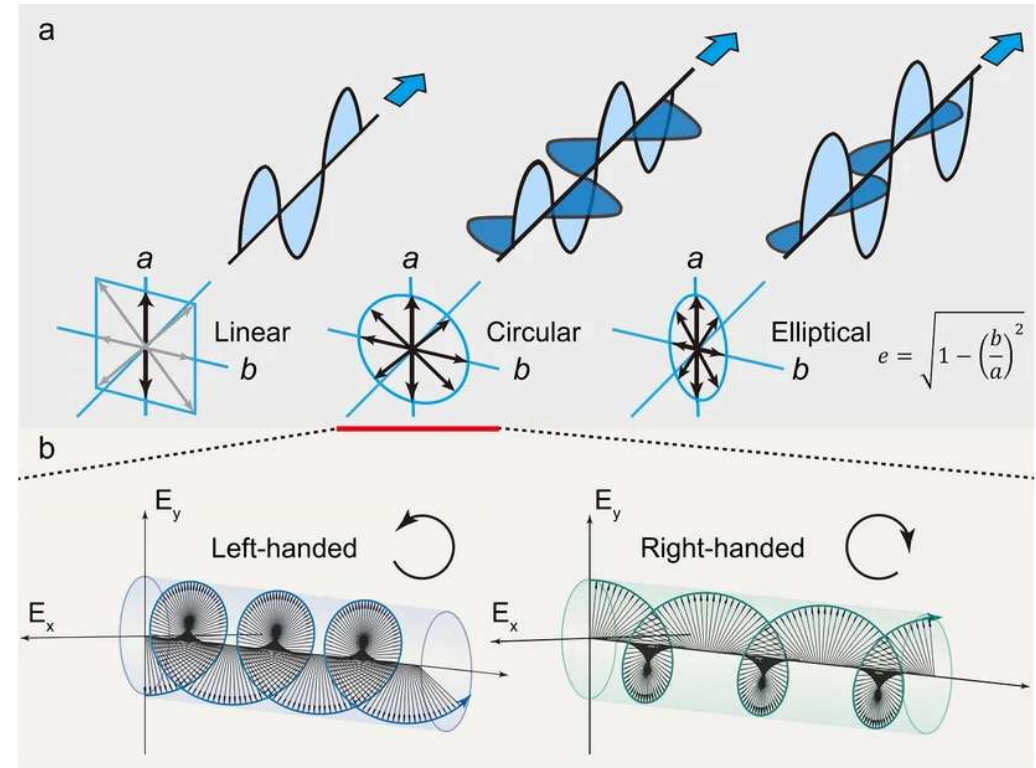
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MAIN FEATURES OF SYNCHROTRON RADIATION

- ✓ Tunability in a very large energy spectrum
- ✓ Very high brightness
- ✓ Strong collimation
- ✓ Time structure
- ✓ High polarization



Qiu *et al.*, Small **22**, e12675 (2026)

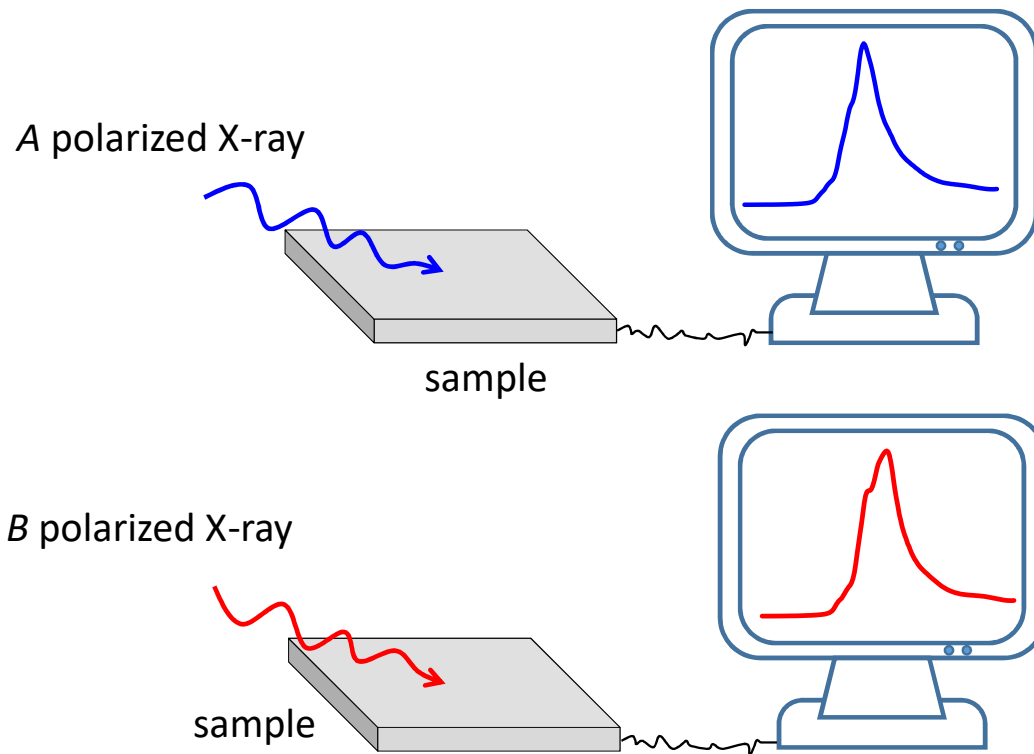
In this lesson, we will not cover:

- Definition and description of what is polarization of electromagnetic radiation
- How to obtain polarized synchrotron radiation X-ray
- Physical mechanism describing the interaction between matter and polarized X-ray

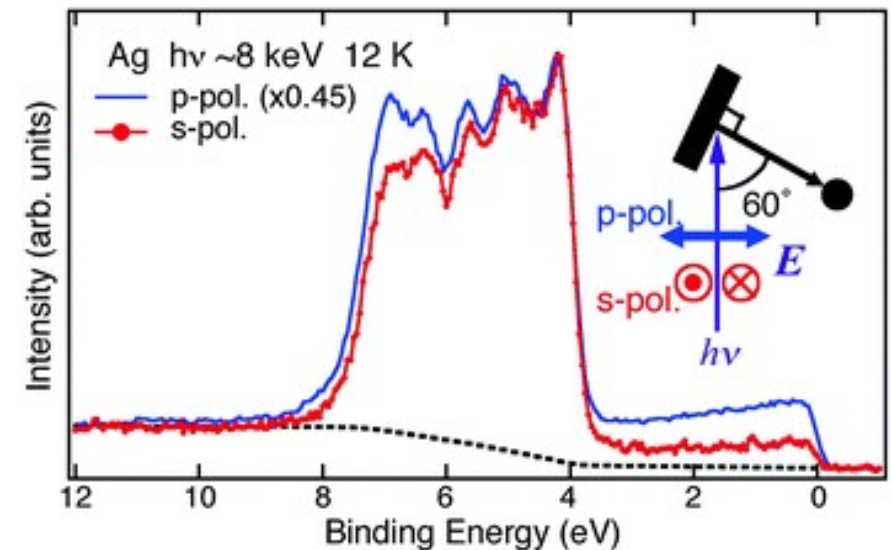
In this lesson, we will discuss examples on how to characterize materials by exploiting the spectral variations that occur when polarized X-rays interact with matter

X-ray↔matter interaction depend on polarization because the oscillating electric field of a polarized X-ray exerts a directional force on bound or free electrons: This force drives electron motion preferentially along the electric field vector, which changes how X-rays scatter, absorb, or transfer momentum during the interaction.

The general approach involve measurement of some signal (electron or photon intensity) and describe why such signal is different upon absorption of circular right/left or linear vertical/horizontal polarized light.



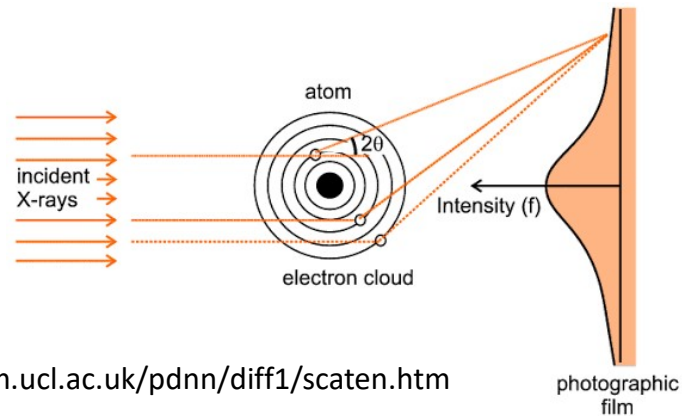
An example from photoemission spectroscopy: Au valence band is different when measured with X-ray polarization along the surface plane or vertical to it, due to the symmetry of the involved valence band orbital



A. Sekiyama, in *Compendium of Surface and Interface Analysis* **38**, 229 (2018)

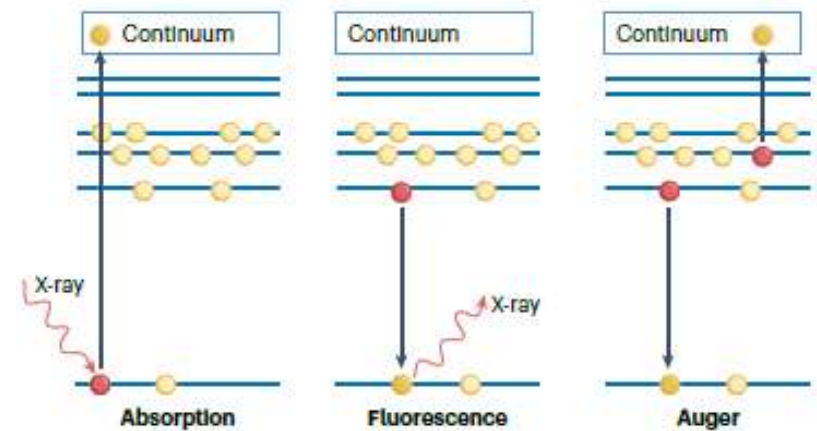
TECHNIQUES

- **X-ray absorption spectroscopy (XAS)**
measured either in transmission or by electron and fluorescence yield
- **X-ray photoelectron spectroscopy (PES)**
- **X-ray elastic and inelastic scattering**
- **X-ray diffraction**

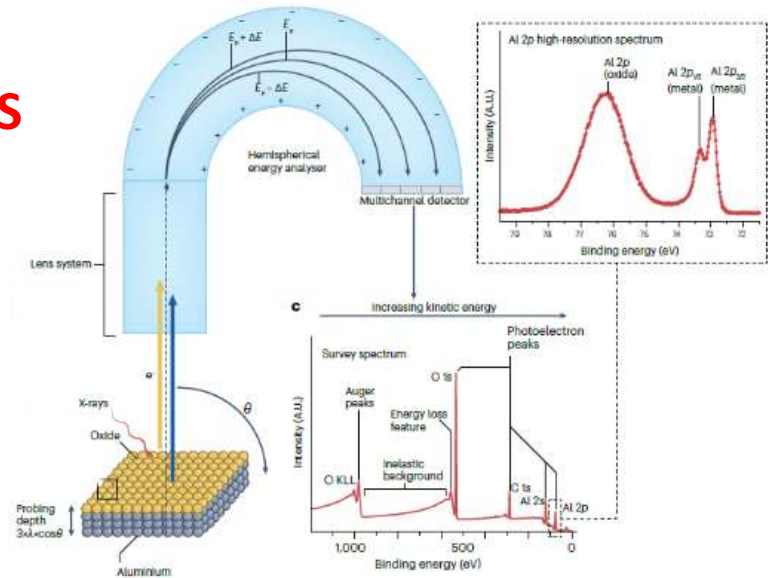


<http://pd.chem.ucl.ac.uk/pdnn/diff1/scaten.htm>

XAS



PES



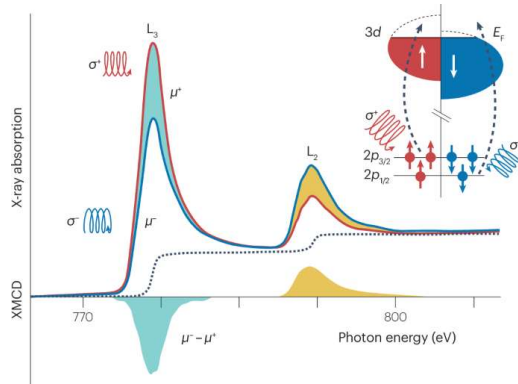
G. Greczynski *et al.*, Nat. Rev. Methods Primers **3**, 40 (2023)

C. T. Chantler *et al.*, Nat. Rev. Methods Primers **4**, 89 (2024)

WHAT CAN BE INVESTIGATED

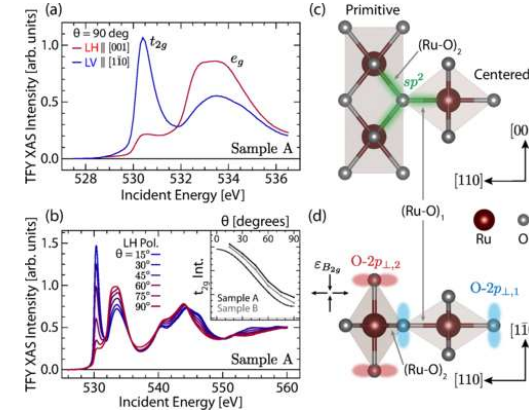
Magnetic and ferroelectric properties of materials

XAS
PES



Vaz *et al.*, Nat. Rev. Methods Primers **5**, 27 (2025)

Structure and orientation

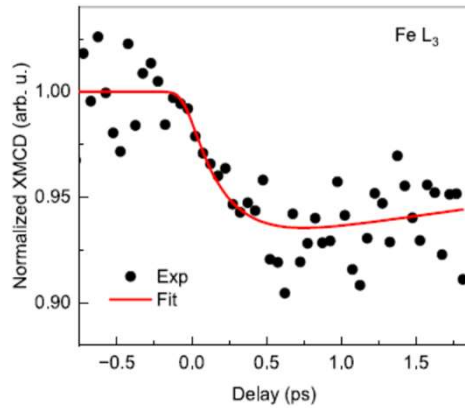


XANES
EXAFS

Occhialini *et al.*, Phys. Rev. Mater. **6**, 084802 (2022)

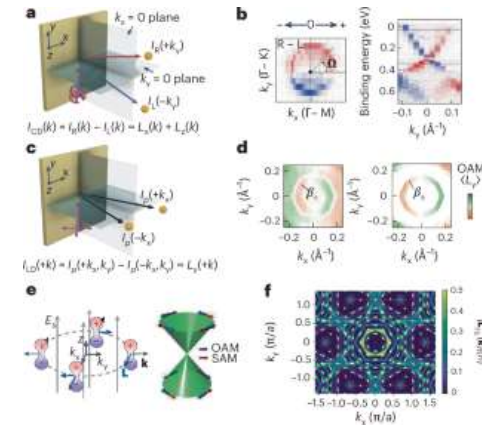
Electron and magnetic dynamics

XAS at a free
electron laser



Marotzke *et al.*, Struct. Dyn. **12**, 034301 (2025)

Dynamic of orbital angular momentum



ARPES

Oh *et al.*, Nature Physics **22**, 1006 (2026)

In this lecture, I will focus on how polarization-dependent X-ray absorption spectroscopy (XAS) can be used to characterize ferromagnetic and ferroelectric materials and heterostructures, through the analysis of three examples:

- **Investigating Ferromagnetic Materials Exploiting Circular Polarized Radiation**
- **Investigating Ferroelectric Materials Exploiting Linear Polarized Radiation**
- **Microscopy Exploiting Polarized Radiation: Imaging Magnetic and Ferroelectric Domains**

Before moving on to examples, a brief overview is needed on what is meant by dichroism.

DICHROISM

Dichroism $\Rightarrow$ The absorption of radiation depends on the polarization of the radiation itself

In general, the response of an electric charge distribution to electromagnetic excitation can be anisotropic, for instance, due to the directionality of chemical bonds, the crystal field, and molecular orientation. Depending on a material's local symmetry, this can result in the radiation absorption coefficient depending on the polarization state (linear or circular) of the radiation itself.

Magnetic dichroism, in particular, is an effect in which radiation absorption (and photoemission) intensity depend on the symmetry of the magnetization distribution. From a conceptual standpoint, the phenomenon of magnetic dichroism is relatively easy to understand if one begins by describing it in the context of circularly polarized X-ray absorption, referred to in the literature as **X-ray magnetic circular dichroism (XMCD)**

Magnetic dichroism is essentially a phenomenon in which a photon transfers its angular momentum to the material (atom) with which it interacts. The fundamental principle is therefore the conservation of angular momentum.

A note: X-ray absorption spectroscopy can be measured by different means, namely:

- *Transmission Mode*: Measures incident versus transmitted X-ray intensity through the sample
- *Fluorescence Yield Mode*: Detects secondary X-rays emitted as core holes are filled
- *Electron Yield Mode*: Detects electron cascade emitted after core-hole relaxation

The measurement strategy affects the information depth of the processes (larger when photons are detected, smaller when electrons are detected). Examples in this lecture refer to XAS measured in total electron yield mode.

Let us consider the case of XMCD in a magnetic material belonging to the 3d transition metals (iron, cobalt, and nickel) and employ a **two-step model**:

1) First step: photon absorption and the generation of a spin-polarized photoelectron.

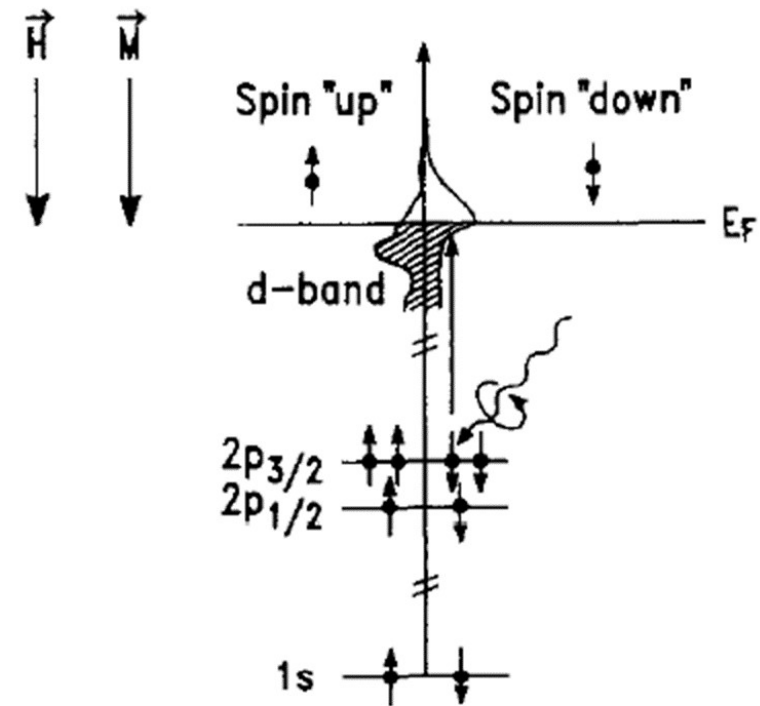
Circularly polarized light transfers its angular momentum to the excited electron. The quantization axis is defined by the direction of photon propagation.

2) Second step: the valence band, characterized by spin splitting, acts as a spin detector. The quantization axis is defined by the magnetization direction.

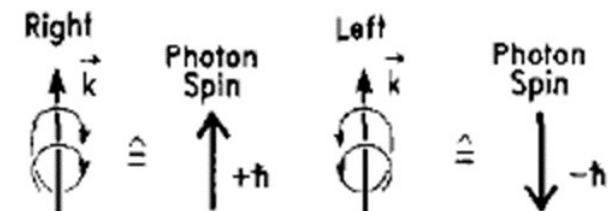
First step: the absorption of a photon and the production of a spin-polarized photoelectron.

Excitation occurs at a $2p$ level characterized by spin-orbit splitting; the electron's magnetic moment arises from both orbital and spin degrees of freedom, meaning the photon's angular momentum is transferred to both the electron's spin and its orbital motion.

Thanks to spin-orbit coupling, a significant portion of the photon's angular momentum can be imparted to the spin, **resulting in spin-polarized photoelectrons.**

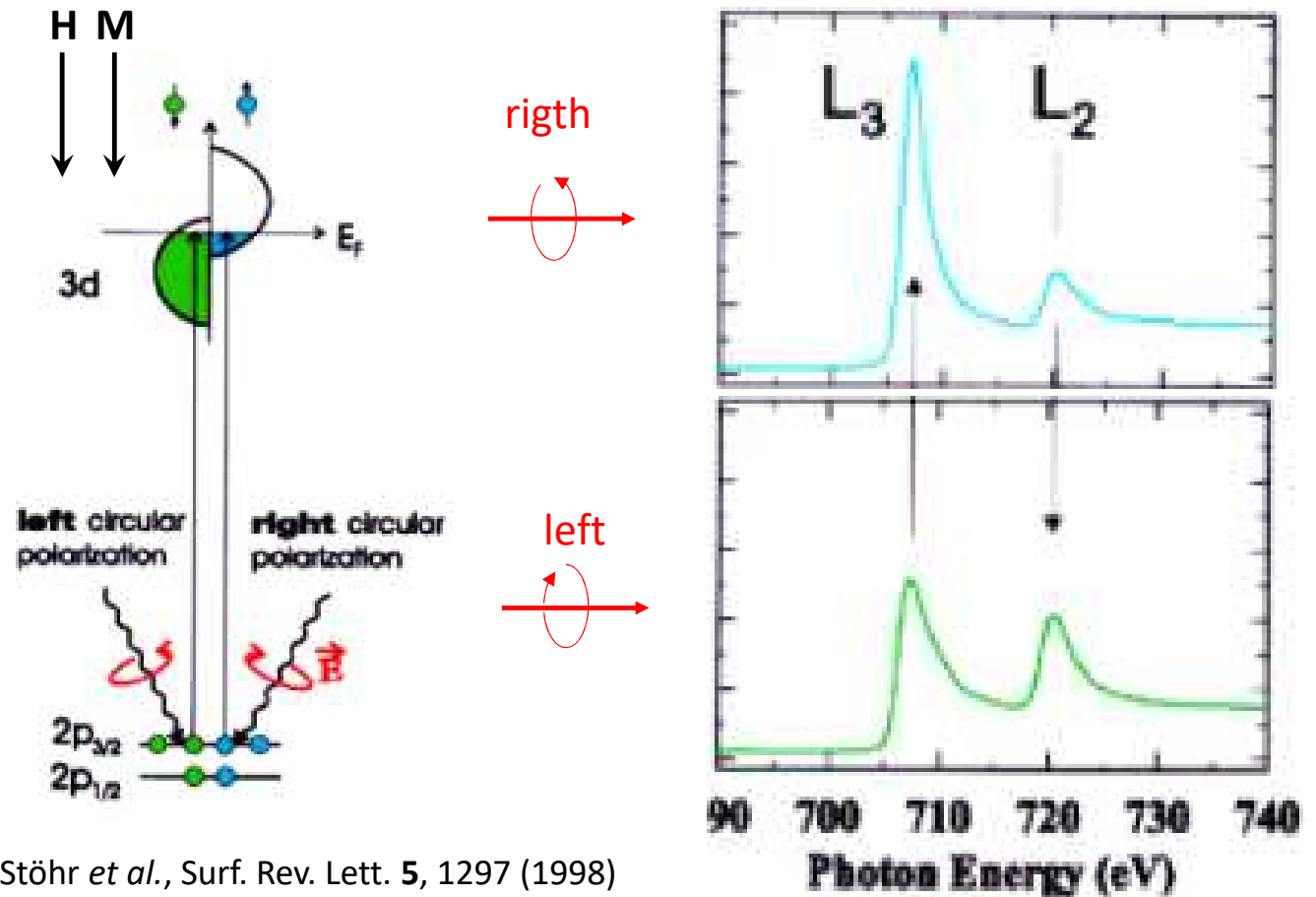


Polarization and Photon Spin



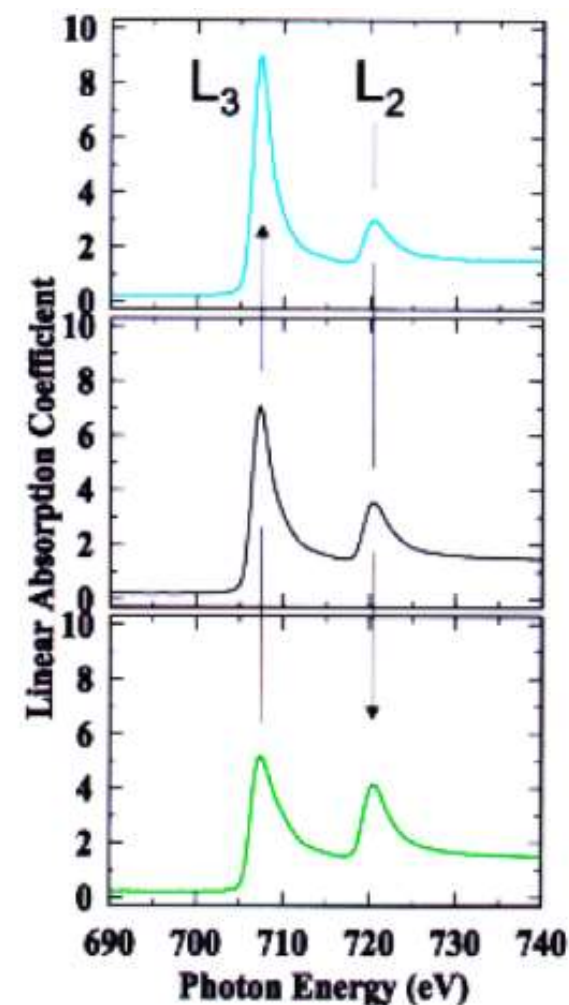
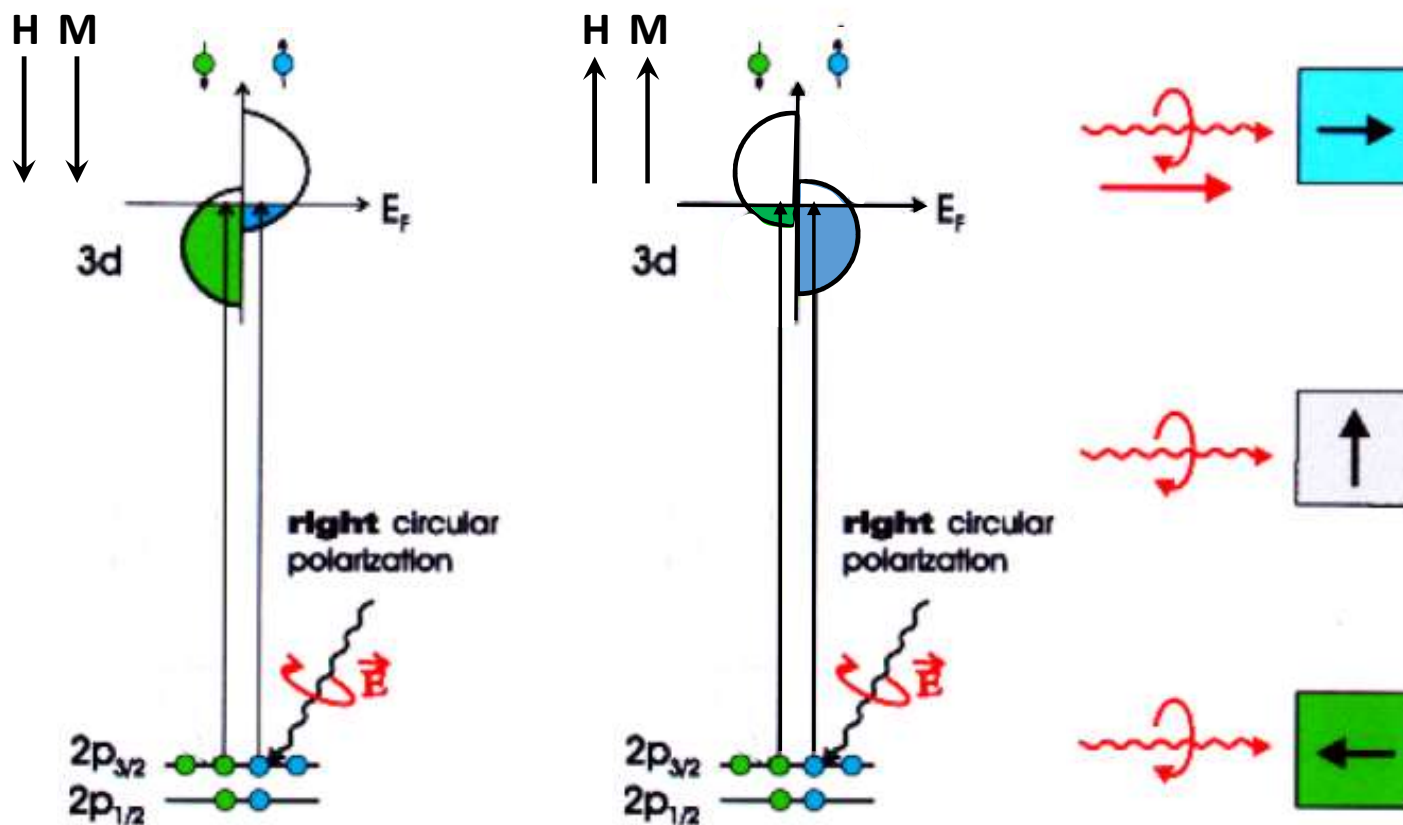
The second step arises from the spin-filtering effect of the valence band.

In this step, the magnetic properties of the sample come into play. The excited electrons undergo an energy transition to the valence band. Given the material's magnetization, this band acts as a "detector" for the polarization state of the excited electrons. The quantization axis of this detector is defined by the sample's magnetization, which must be aligned with the direction of the photon's spin to maximize the dichroic effect.



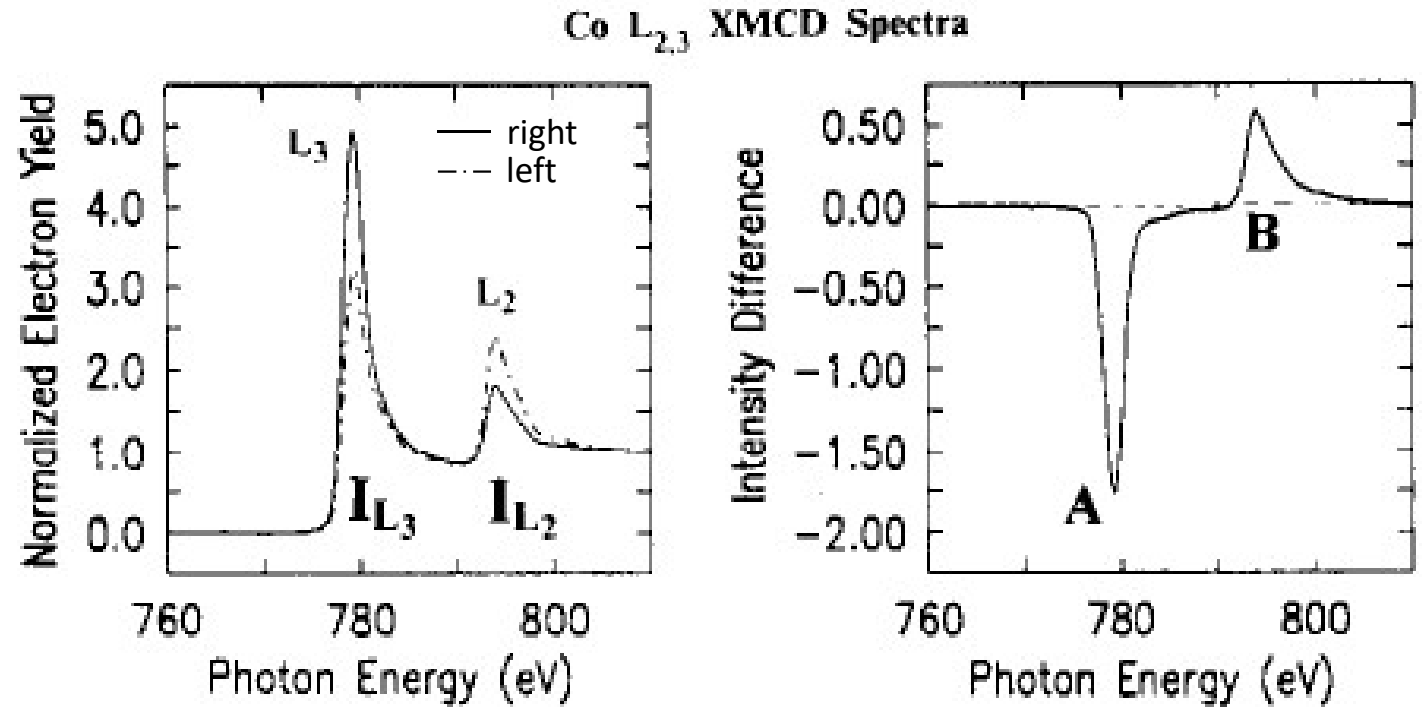
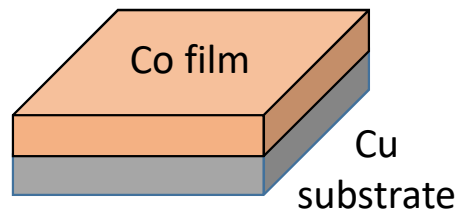
In the case under consideration, there are far more available empty spin-up states than empty spin-down states close to the Fermi level. Consequently, electrons excited from the $2p_{3/2}$ level by right-circularly polarized light (predominantly spin-up) will encounter several available states at the Fermi level, resulting in a high transition probability. Conversely, there is a high transition probability for excitation from the $2p_{1/2}$ level using left-circularly polarized light, given the predominantly spin-up character of the resulting electrons.

Analogous result is obtained by using one of the two circularly polarized direction and measuring two spectra after reverting magnetization direction



The maximum XMCD effect is obtained when the direction of light propagation is parallel to the direction of magnetization ($\text{XMCD} \propto \cos\theta$)

Example: XAS spectra, measured by total electron yield, obtained from a ferromagnetic cobalt thin film with two opposite direction of circular polarized radiation



J. Stöhr, JES Relat. Phenom. **75**, 253-272 (1995)

The difference in absorption intensity between the two cases is significant at the two absorption edges and is opposite in sign, as expected from the previous description. This effect, arising from magnetic dichroism, is quantified by calculating the difference between the two curves, thereby yielding the magnetic dichroism spectrum. The XMCD effect is extremely useful for the magnetic characterization of ferromagnetic thin films

Estimation of the magnetic moment from XMCD sum rules

C. T. Chen *et al.*, Phys. Rev. Lett. **75**, 152 (1995)

$$m_{\text{orb}} = -\frac{4 \int_{L_3+L_2} (\mu_+ - \mu_-) d\omega}{3 \int_{L_3+L_2} (\mu_+ + \mu_-) d\omega} (10 - n_{3d}), \quad (1)$$

$$m_{\text{spin}} = -\frac{6 \int_{L_3} (\mu_+ - \mu_-) d\omega - 4 \int_{L_3+L_2} (\mu_+ - \mu_-) d\omega}{\int_{L_3+L_2} (\mu_+ + \mu_-) d\omega} \times (10 - n_{3d}) \left(1 + \frac{7\langle T_z \rangle}{2\langle S_z \rangle} \right)^{-1}, \quad (2)$$

~ 0

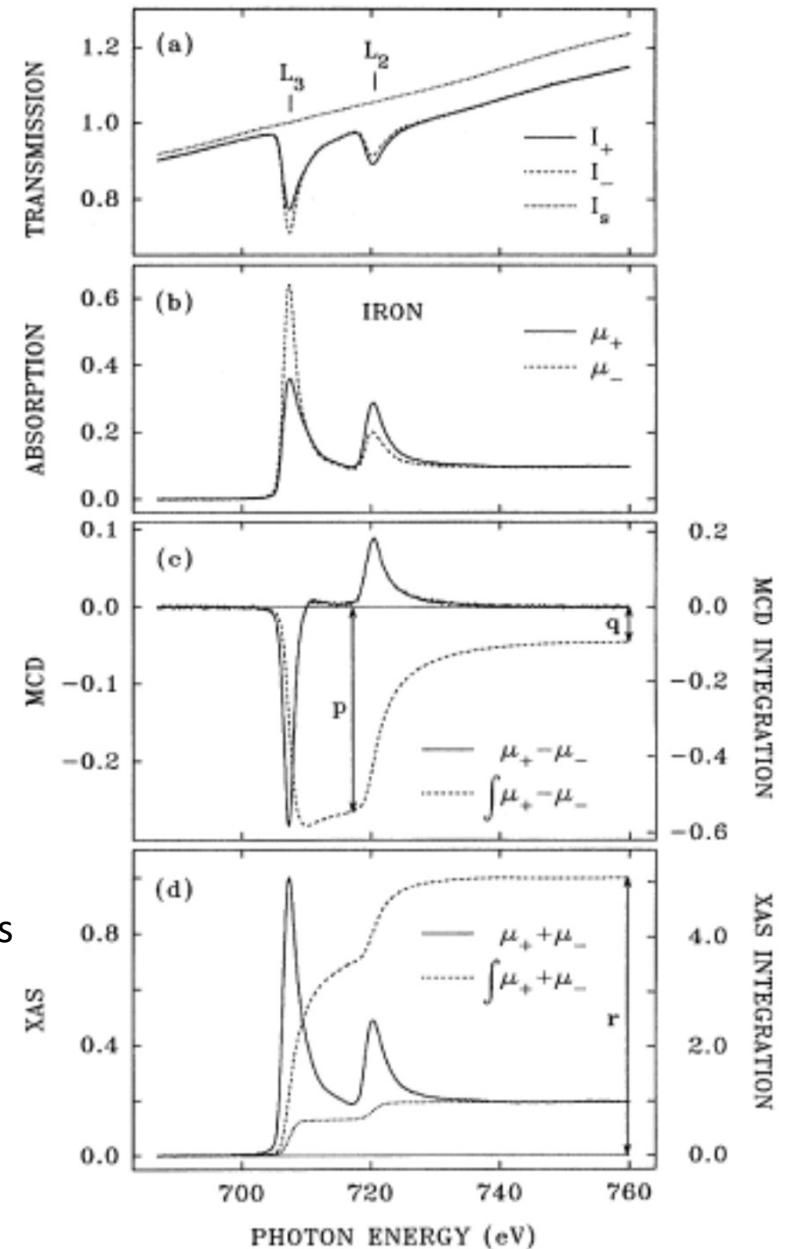
$$\text{Orbital moment} = m_{\text{orb}} = -\frac{4}{3} \frac{q}{r} N_d^h$$

n_{3d} = number of d electrons

$$\text{Spin moment} = m_{\text{spin}} = -\frac{6p-4q}{r} N_d^h$$

N_d^h = number of d holes

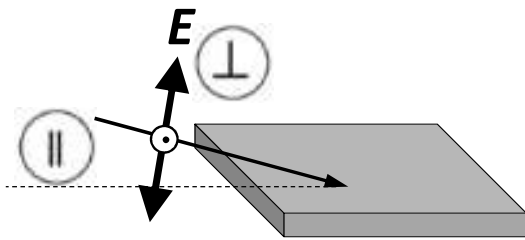
...with caution



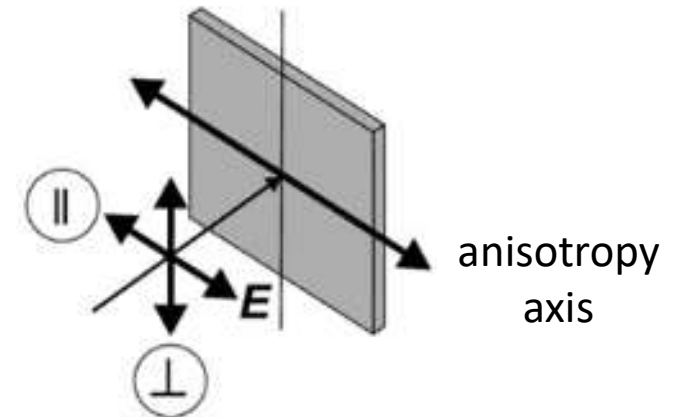
X-RAY LINEAR DICHROISM (XLD)

X-Ray linear dichroism exploits linearly polarized X-rays, with mutually perpendicular polarizations and spectrum is obtained as the difference between XAS intensity data obtained with two orthogonal polarizations.

polarizations that are (almost) orthogonal ($\perp$) and parallel ($\parallel$) with respect to the sample surface



polarizations that are perpendicular ($\perp$) and parallel ($\parallel$) to some anisotropic direction in the material



The linearly polarized photons carry no angular momentum, and XLD intensity arises due to **anisotropic distributions of charge**, for example a charge anisotropy associated with chemical bonding (*e.g.* in ferroelectrics) or a charge anisotropy associated with magnetism via spin-orbit coupling (*e.g.* in ferromagnets and antiferromagnets). The XLD intensities evaluated in this way are sensitive to the presence of an **anisotropy axis** and proportional to the square of the cosine of the angle between the single X-ray polarization and this symmetry axis.

Consider the transition from an L -edge ($2p \rightarrow 3d$) taking place from spherically symmetric filled (core) states to available (valence) states. The fully occupied degenerate core states from which electrons are excited yield spherically symmetric charge distributions (yellow discs) in both cubic and lower symmetries.

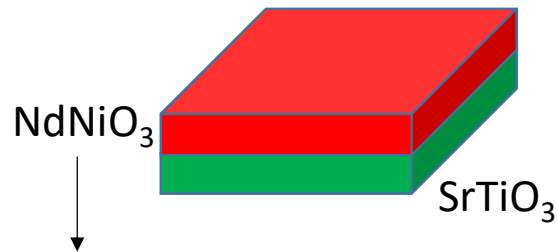
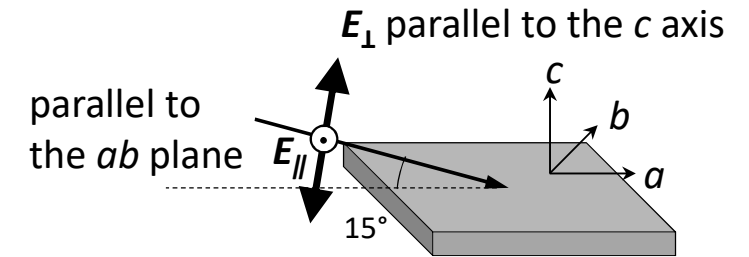
The degenerate valence states to which electrons are excited yield:

- spherically symmetric charge distributions (yellow discs) in cubic symmetry in which the density of available states does not depend on orientation: XAS intensity is independent of the X-ray polarization direction.
- anisotropic charge distributions (yellow ellipses) in lower symmetry, in which the density of available states depends on orientation: XAS intensity depends on the X-ray polarization direction.

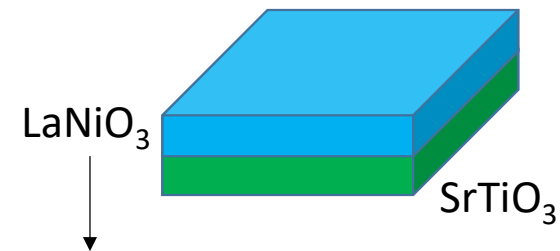
A search light effect!

	Core states	Valence states	
	Cubic or lower symmetry crystals	Cubic crystals	Lower-symmetry crystals
L -edge $2p \rightarrow 3d$	$2p$ 	$3d$ e_g  t_{2g} 	

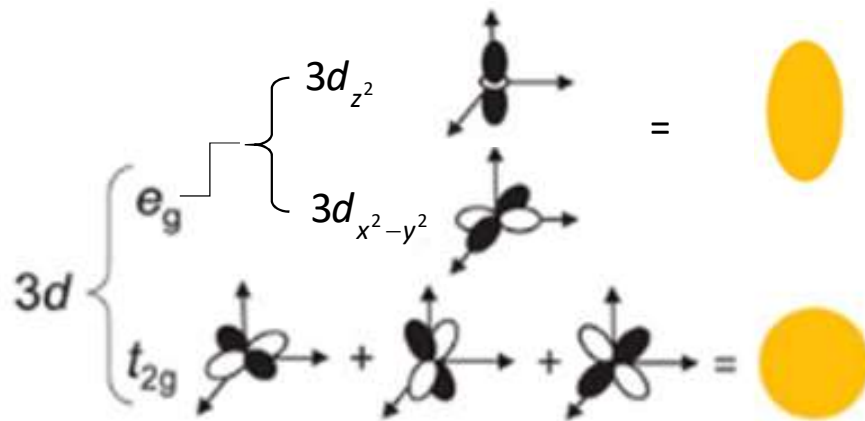
Example: XAS measured in fluorescence yield around the Ni L edge ($2p \rightarrow 3d$) transition from two nickelate films (10 – 15 unit cells) both grown on cubic SrTiO_3 . Valence is Ni^{3+} in both compounds (7 electrons in d shell)



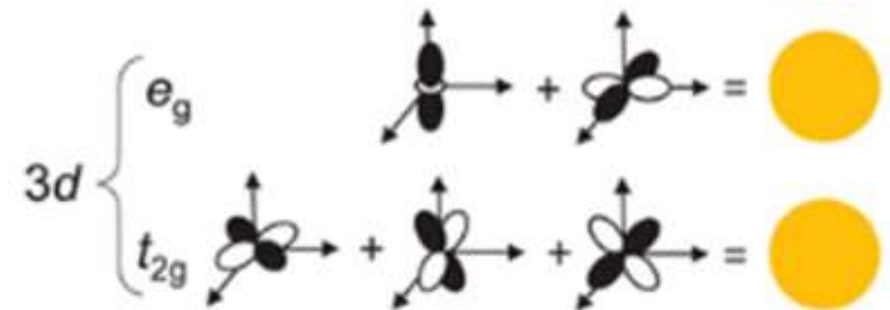
Out of plane contraction



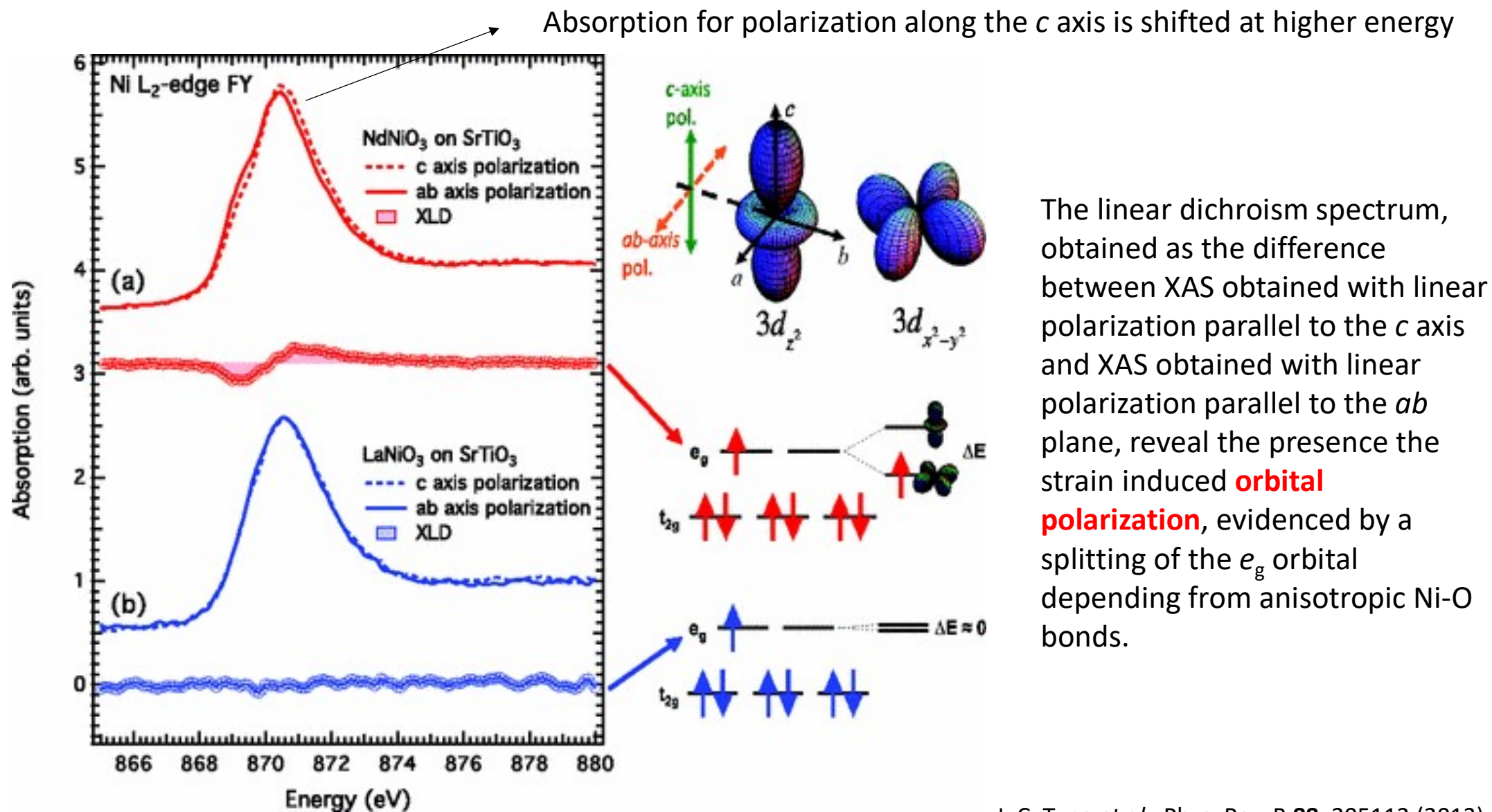
No out of plane contraction



Linear dichroism expected



No linear dichroism expected

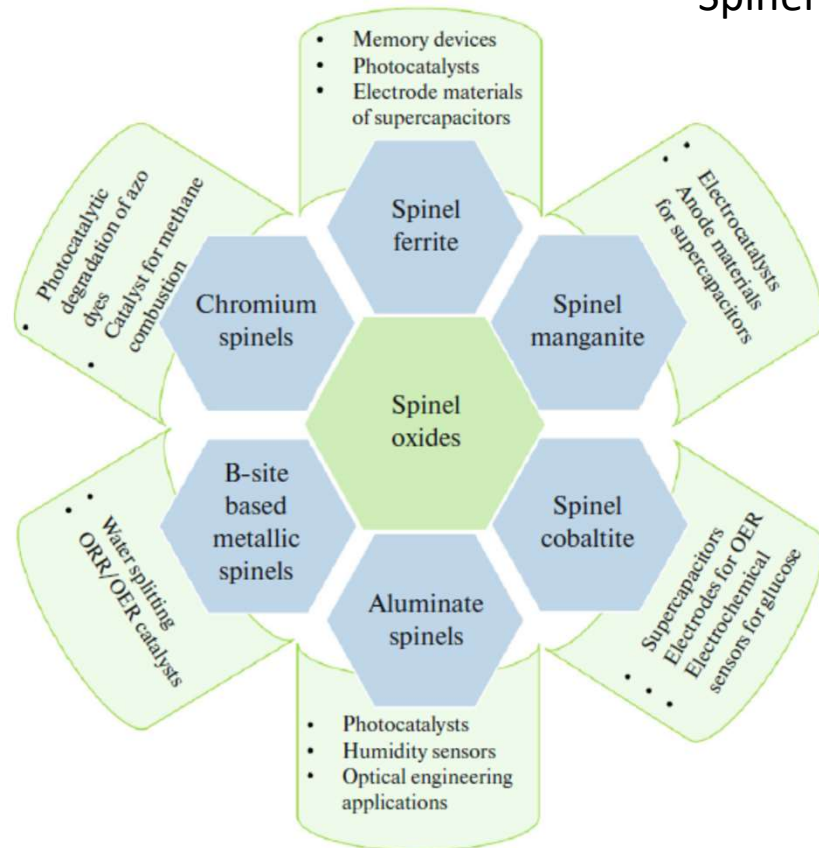


I. C. Tung *et al.*, Phys. Rev. B **88**, 205112 (2013)

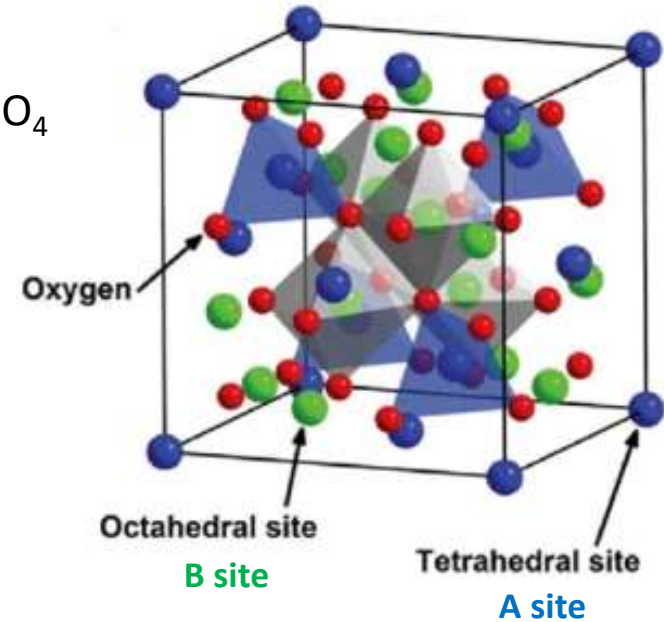
INVESTIGATING FERROMAGNETIC MATERIALS EXPLOITING CIRCULAR POLARIZED RADIATION

SPINEL OXIDES HETEROSTRUCTURES

L. Zhang *et al.*, Electrochem. Energ. Rev. **2**, 518 (2019)



Spinel is the mineral MgAl_2O_4



- Spinel group has the typical structure $A^{2+}B_2^{3+}O_4^{2-}$
- Interesting for several technological applications
- Necessity to characterize the physical properties in nanostructured systems

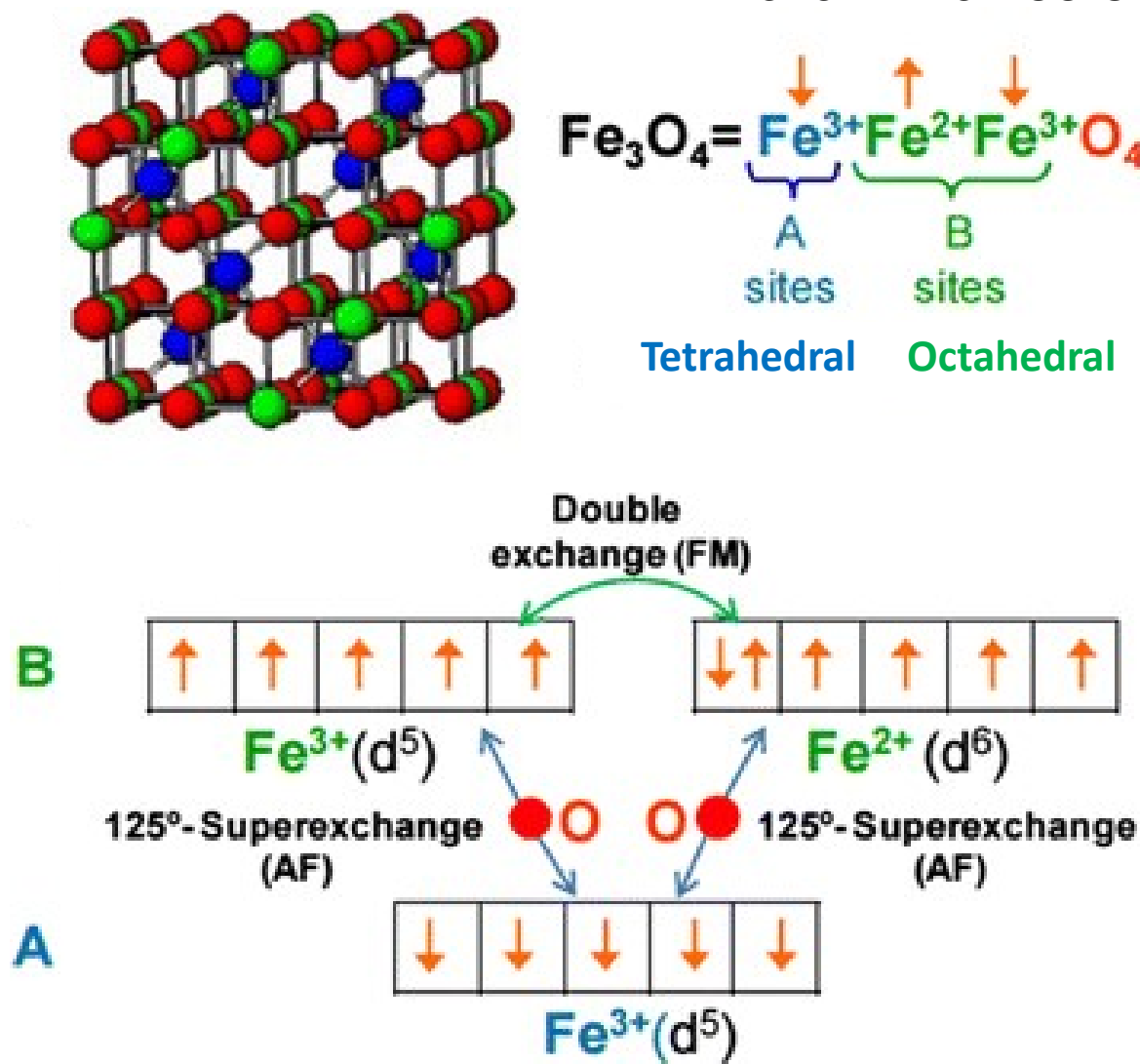
D. Srikala *et al.*, Phys. Solid State **66**, 327 (2024)

INVERSE SPINEL STRUCTURE

Fe_3O_4 (magnetite)

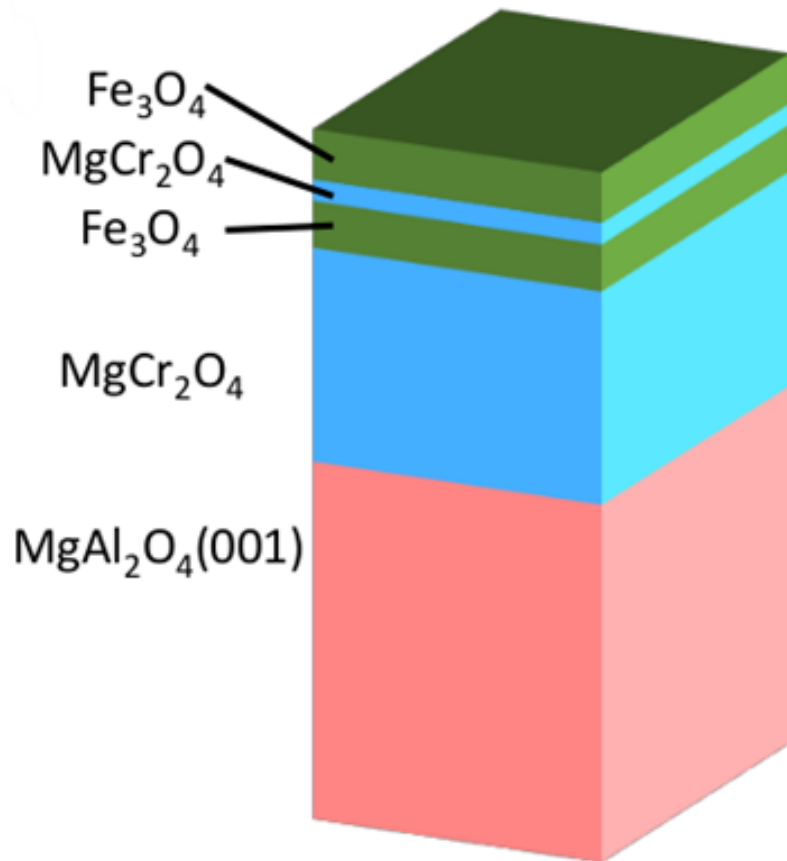
Mixed valence ferrimagnetic oxide with three different Fe ions in different crystallographic position and a complicated magnetic configuration.

Half-metal ferrimagnetic material with a high Curie temperature $T_C \sim 850$ K.



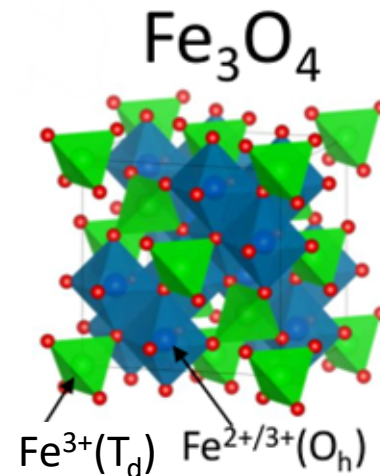
MAGNETIC INTERACTION AT THE $\text{Fe}_3\text{O}_4/\text{MgCr}_2\text{O}_4$ INTERFACE

- ✓ Investigate interface induced phenomena in Fe_3O_4 multilayers as a function of the spacer thickness
- ✓ Use all spinel oxides heterostructures grown by pulsed laser deposition with good lattice matching to improve epitaxial growth and limit the influence of antiphase boundaries (APBs)



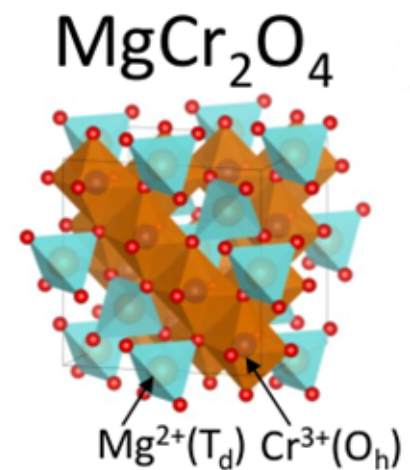
Lattice mismatch less than 1%

**ferrimagnetic at
room temperature**



$a = 8.39 \text{ \AA}$

**paramagnetic at
room temperature**



$a = 8.33 \text{ \AA}$

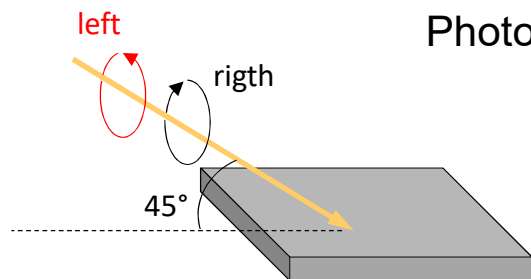
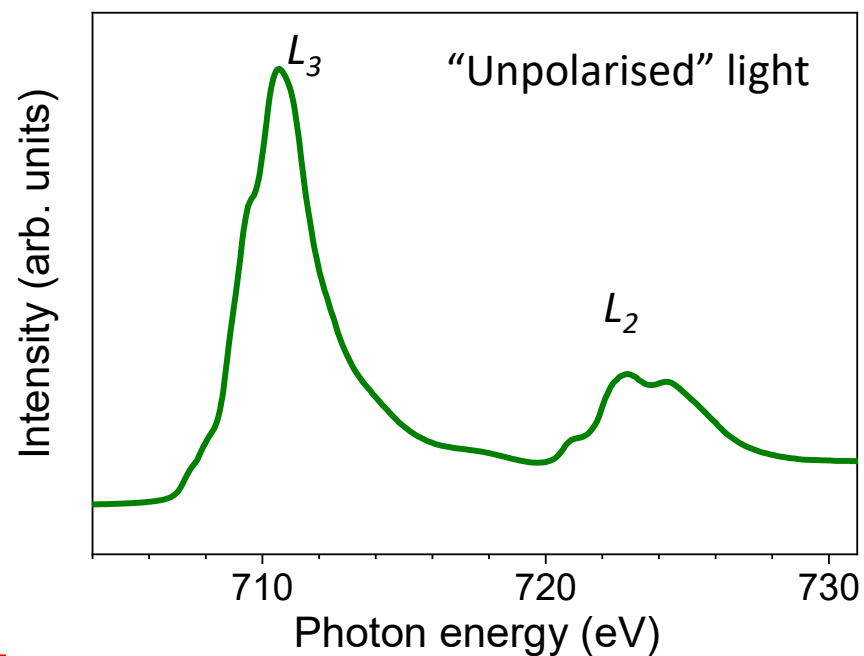
XAS AND XMCD IN Fe

Thick Fe_3O_4 film

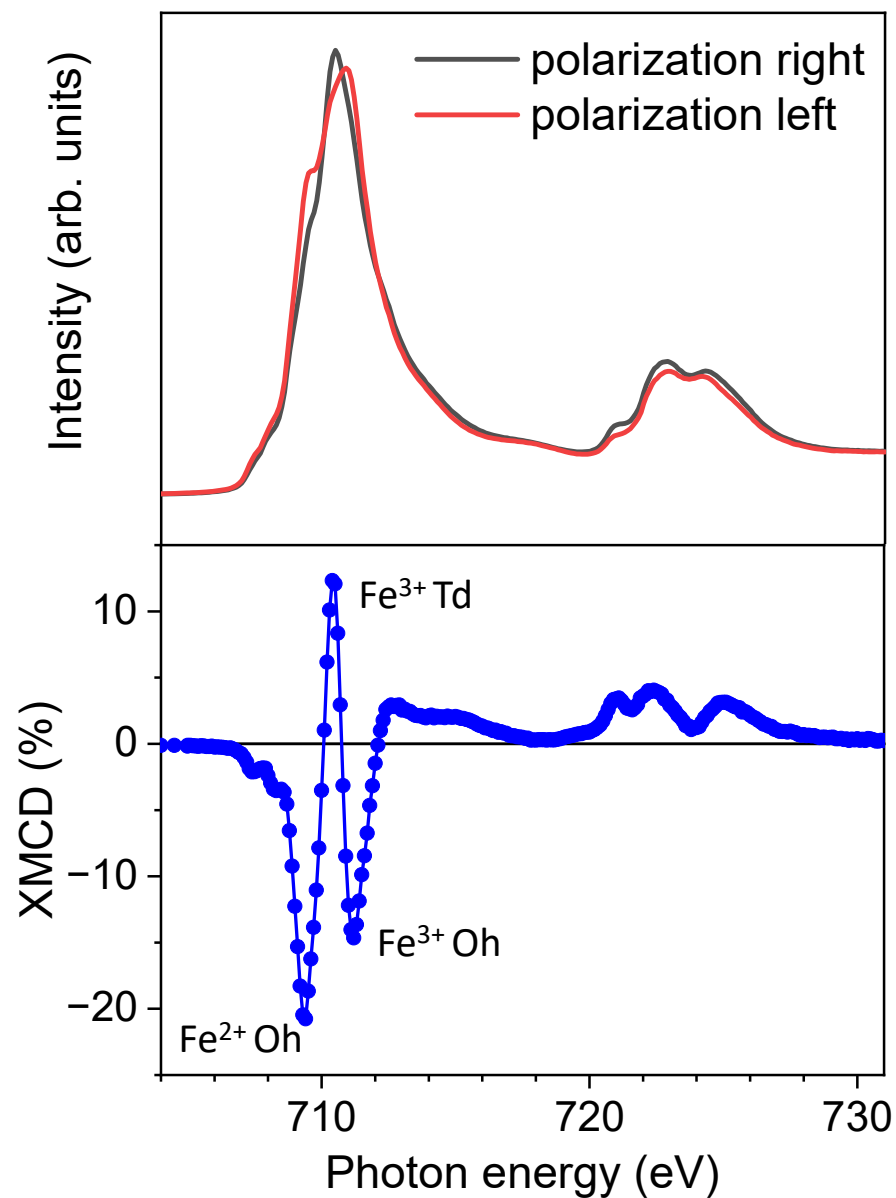
Fe_3O_4
(60 nm)

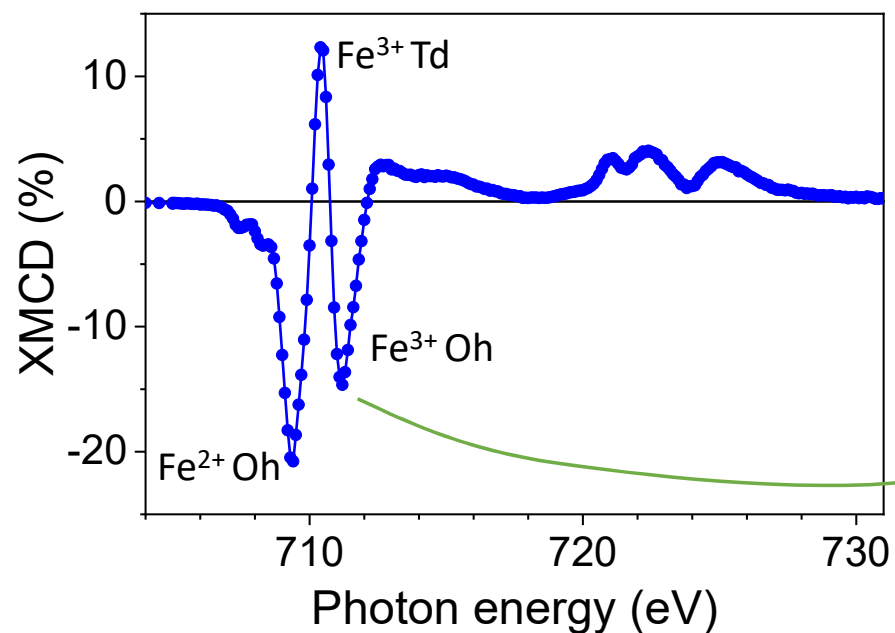


MgCr_2O_4
(20 nm)

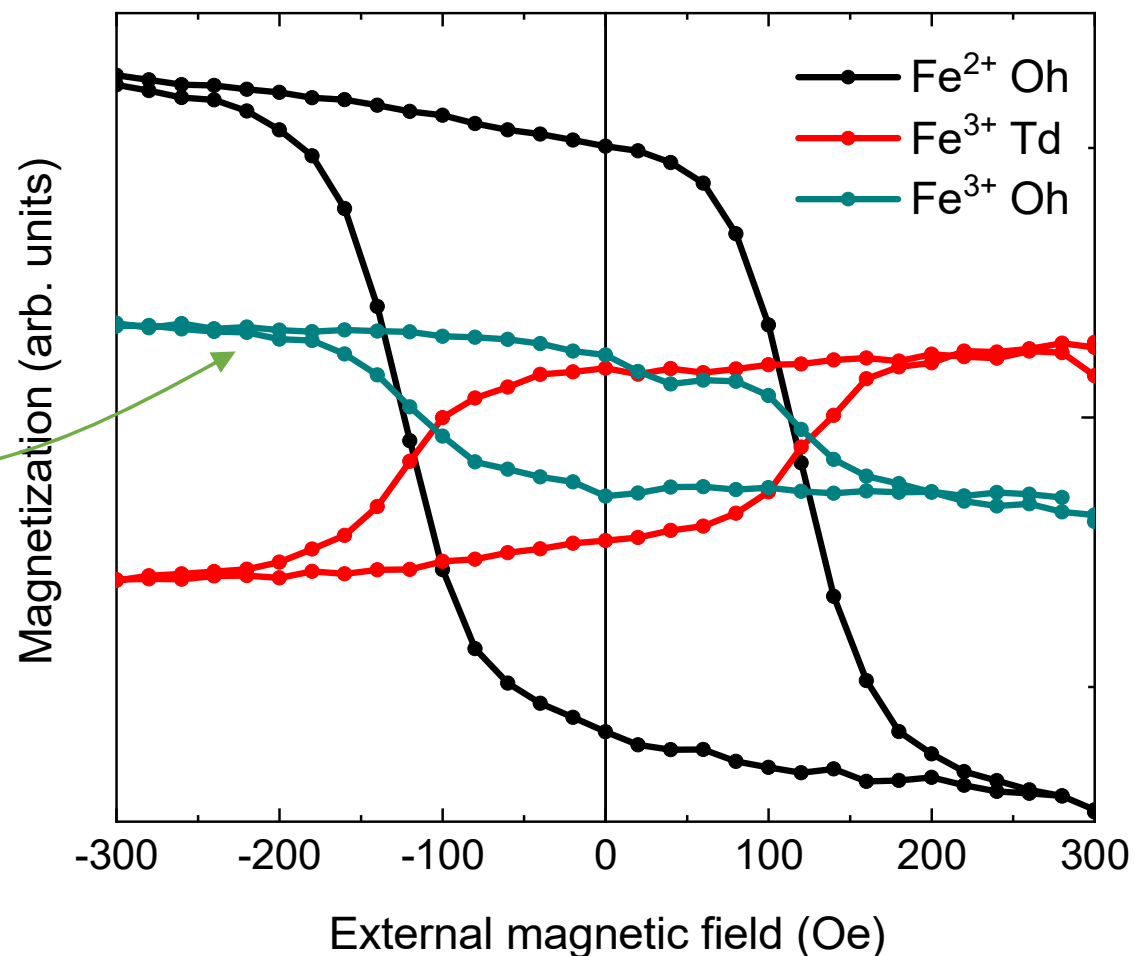


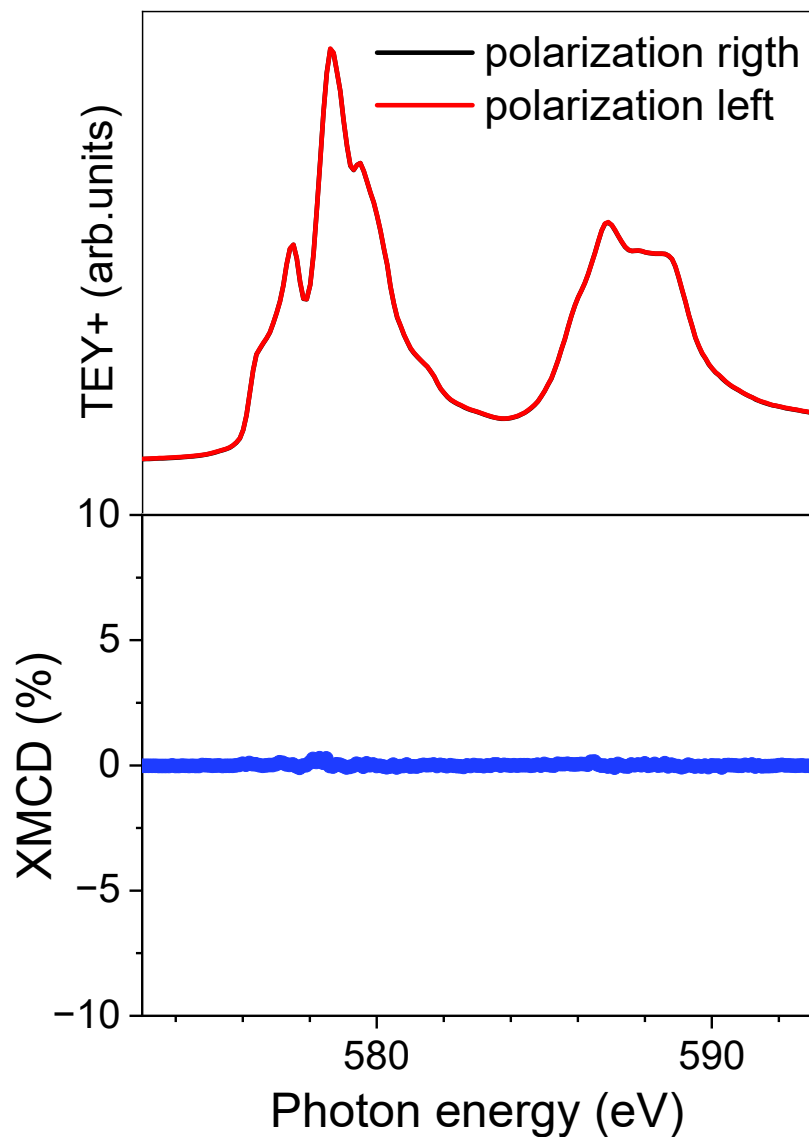
Experiments performed at
the APE beamline of Elettra
Sincrotrone Trieste





Tracking the XMCD intensity (difference between the two XAS for opposite circular polarized light) at selected energies one can follow the magnetization behaviour separately for each ionic component

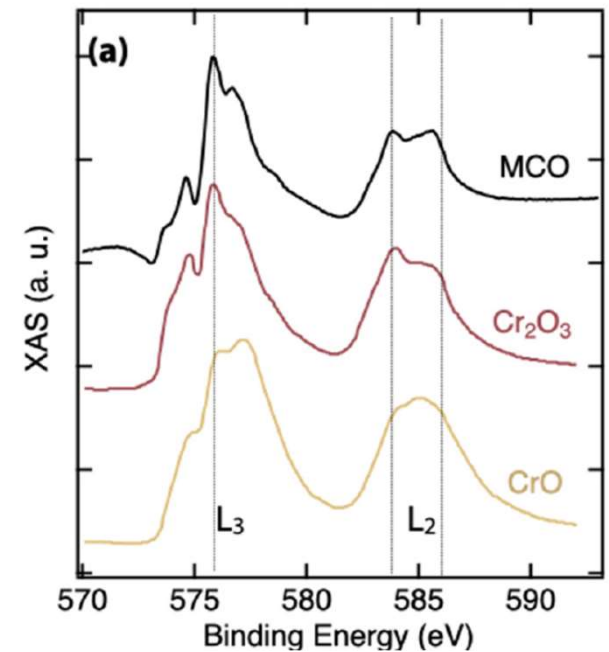
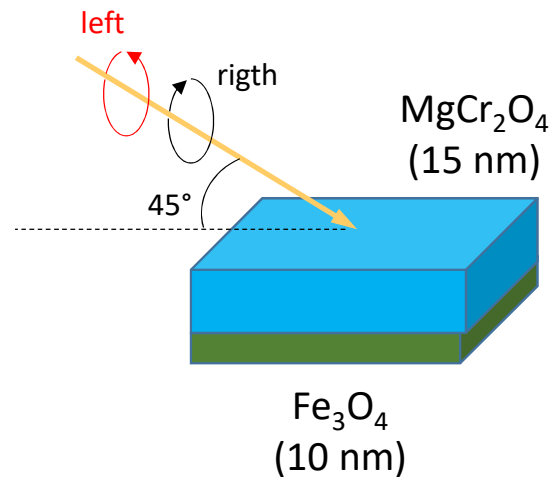




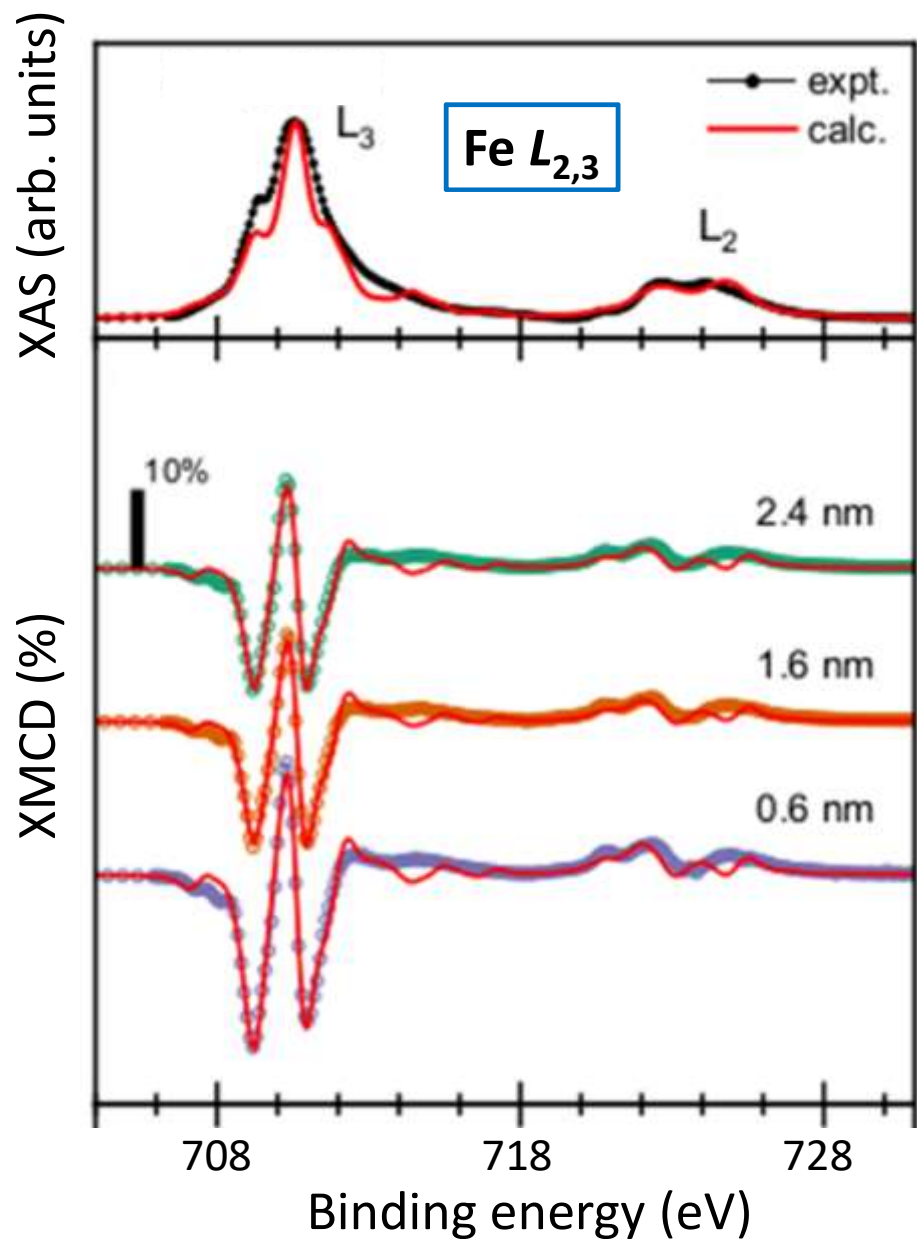
XAS AND XMCD IN CR

Thick MgCr_2O_4 film

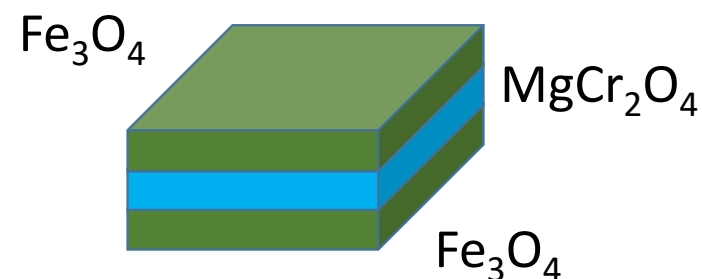
- XAS of Cr is comparable to what obtained in literature
- No XMCD in chromium, as expected



Wen *et al.*, PRB **102**, 165426 (2020)

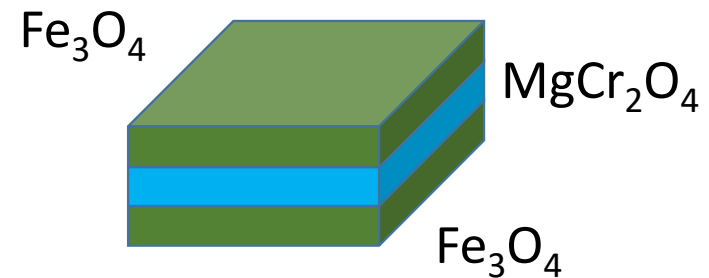
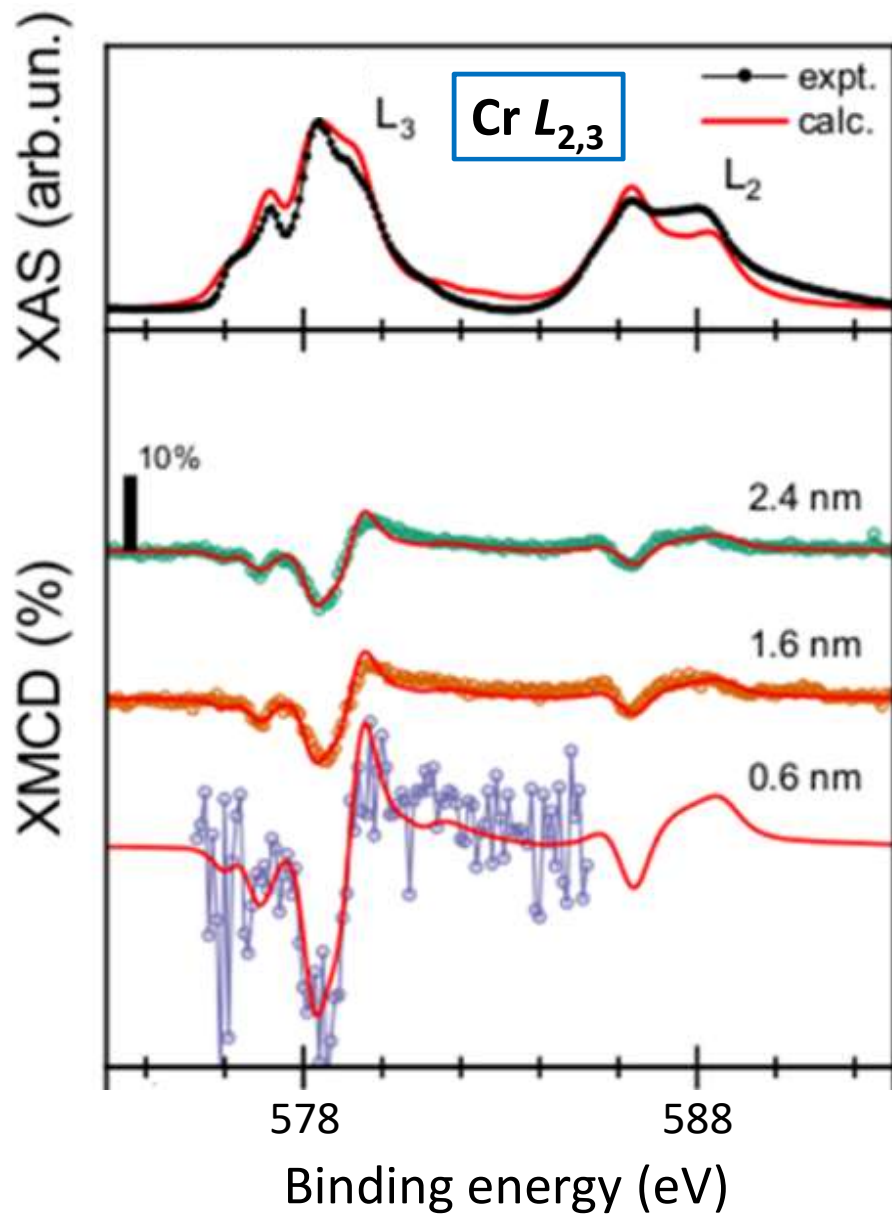


What happens in trilayers?

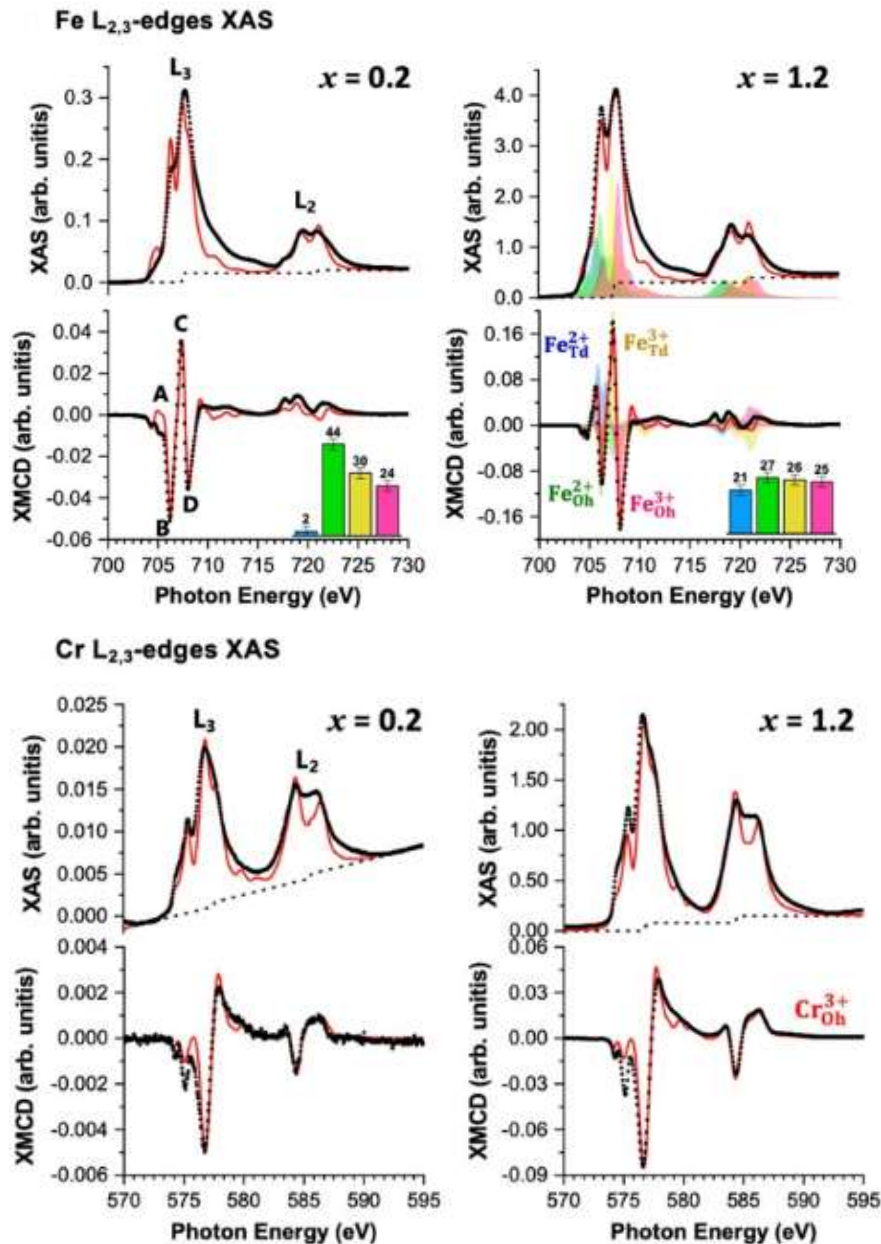


- XAS and XMCD spectra are from Fe_3O_4
- Contribution of the three iron ionic species
- Larger XMCD for thinner spacer

In red are calculations of multiplet configuration

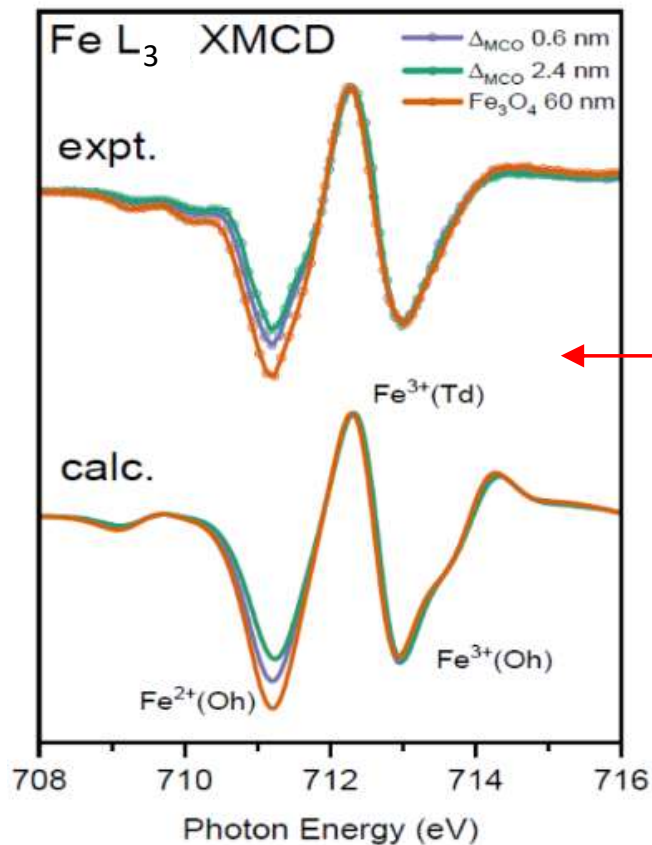


- XAS compatible with MgCr_2O_4 (only Cr^{3+}) present
- Sizable Cr XMCD! Not compatible with the magnetic behaviour of (paramagnetic) MgCr_2O_4
- Noisier and larger XMCD for thinner MgCr_2O_4



Comparison with Fe_{3-x}Cr_xO₄ thin films

- The results obtained at Fe₃O₄/MgCr₂O₄ interface are compatible with the formation of a Fe/Cr oxide mixed phase
- Chromium content in such an interface alloy has to be low, because Fe XMCD in our experiments is still compatible with Fe₃O₄
- The strength of XMCD in chromium is higher at low chromium content



Interface intermixing with mainly Fe^{2+} substitution by Cr^{3+}

Fe magnetic moment with XMCD sum rules

Sample	$\mu (\mu_B)$
60 nm Fe_3O_4	1.98
7 nm Fe_3O_4	1.41
0.6 nm MgCr_2O_4	2.01
1.6 nm MgCr_2O_4	1.58
2.4 nm MgCr_2O_4	1.51

- At low MgCr_2O_4 thickness, magnetite films are not separated – small amount of chromium in the mixed interface alloy result in high XMCD strength. Magnetic moment as high as thick magnetite film
- At larger MgCr_2O_4 thickness, magnetite films are separated. Magnetic moment similar to single 7 nm film.

Sample	$\text{Fe}^{2+}(\text{Oh}) : \text{Fe}^{3+}(\text{Td}) : \text{Fe}^{3+}(\text{Oh})$
60 nm Fe_3O_4	1 : 1 : 0.94
0.6 nm MgCr_2O_4	0.92 : 1 : 1
2.4 nm MgCr_2O_4	0.85 : 1 : 1

RESULTS

XAS/XMCD ALLOWS **ELEMENT SENSITIVE** INVESTIGATION
OF MAGNETIC PROPERTIES AT **COMPLEX INTERFACES**

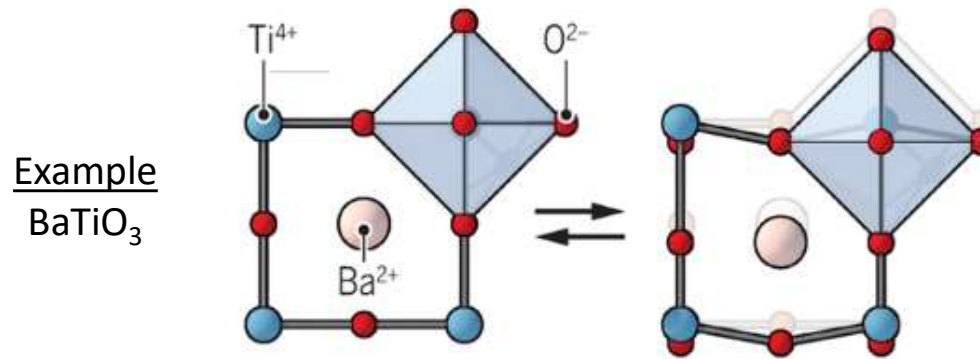
LIMITED CATION INTERDIFFUSION OCCURS AT THE $\text{Fe}_3\text{O}_4/\text{MgCr}_2\text{O}_4$ INTERFACE,
RESULTING IN THE FORMATION OF A **MIXED Cr/Fe SPINEL OXIDE PHASE**
LOCALIZED AT THE INTERPHASE BOUNDARIES.

FOR SPACER THICKNESSES ≥ 1.6 NM, THE TWO FERROMAGNETIC LAYERS
ARE DECOUPLED AND BEHAVE AS **INDEPENDENT FERROMAGNETIC LAYERS**

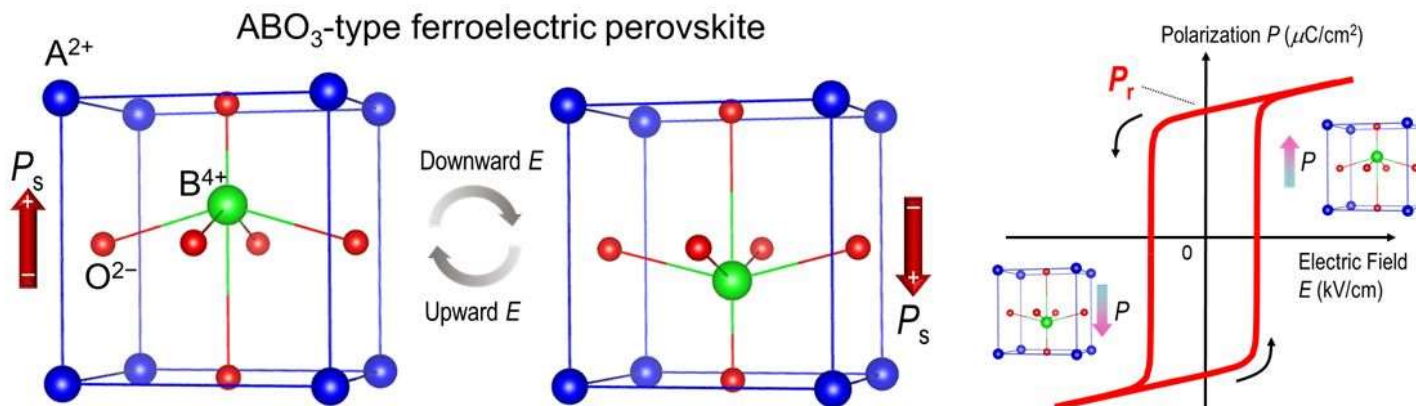
INSIGHTS FOR THE **DESIGN OF COMPLEX ALL-OXIDE SYSTEMS**

INVESTIGATING FERROELECTRIC MATERIALS EXPLOITING LINEAR POLARIZED RADIATION

In oxides ferroelectricity arises from atomic displacement of positive metallic ions and negative oxygen ions in opposite direction. This happens below a transition temperature (Curie temperature) concomitant with a reduction of the symmetry of the crystal lattice (e.g. from cubic to tetragonal).



Li and Ji, Science **361**, 132 (2018)



Polarization in ferroelectric materials can be switched by applying an electrical external field, which changes the ionic position.

⇒ **Hysteresis loop**

Can one characterize ferroelectric materials by detailing their electronic properties?

⇒ Yes! Exploiting differences in absorption of linearly polarized light

A case study:

Piezoelectric films of $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ (PZT) 30 nm thick were deposited on two different substrates, SrRuO_3 (SRO) and $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO), where the ferroelectric polarization point away from the substrate (SRO) or towards the substrate (LSMO)

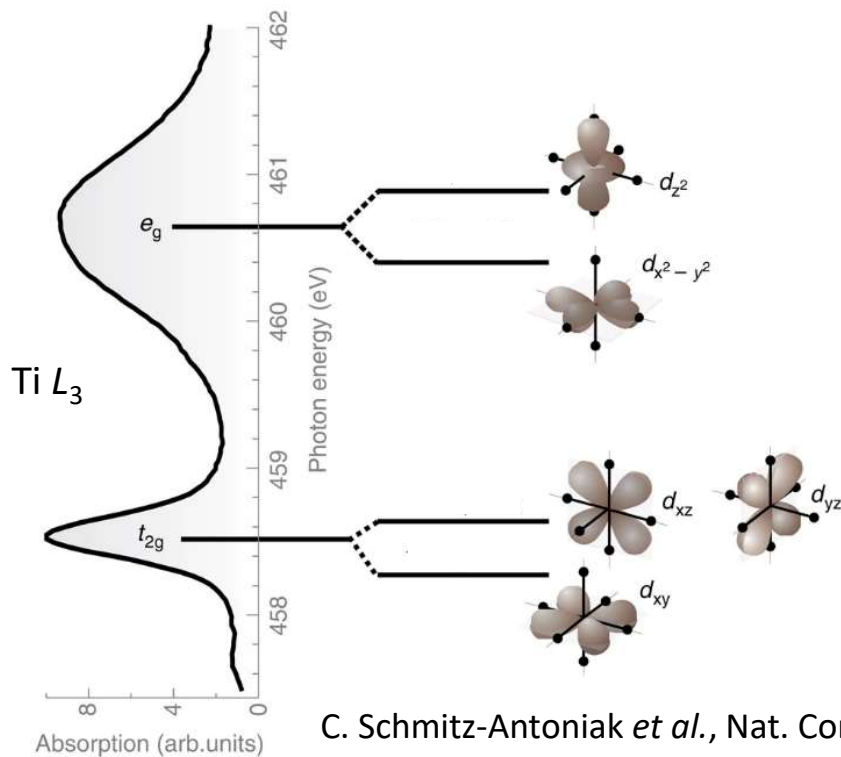


To uncover the electronic properties of PZT X-ray absorption spectroscopy (XAS) around Ti 2*p* core level was used, recorded at Advanced Light Source (USA) and Swiss Light Source (Switzerland), with linearly polarized X-ray excitation

E. Arenholz, *et al.*, Phys. Rev. B **82**, 140103(R) (2010)

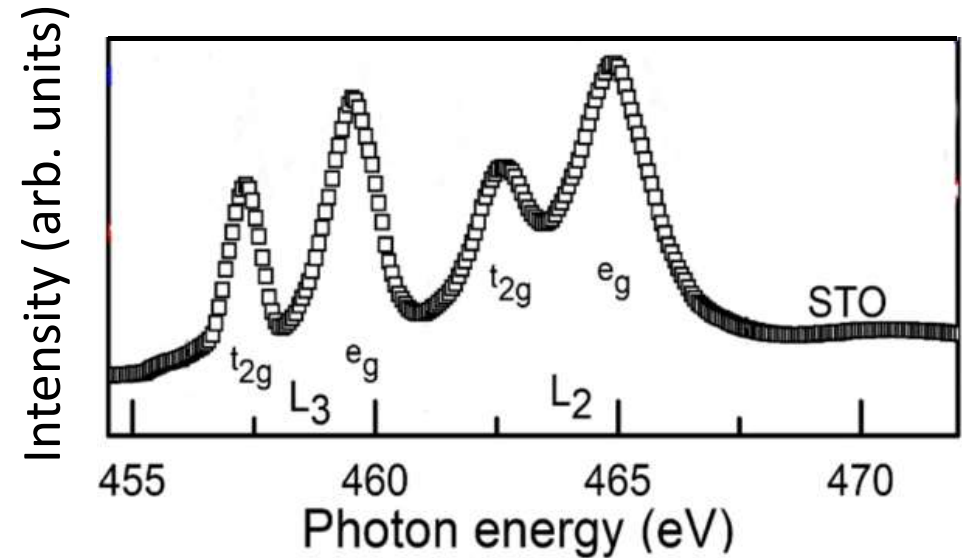
Ti valence in PZT is Ti^{4+} as in SrTiO_3 and share a similar XAS 2p spectrum as cubic strontium titanate.

The 2p spin-orbit interaction splits the spectrum into $2p_{3/2}$ (L_3) and $2p_{1/2}$ (L_2) structures with energy separation of 5.5 eV each spin-orbit component is split by the crystal-field interaction (the electrostatic potential, V , due to the neighboring lattice sites acting on the 3d orbitals) in e_g and t_{2g} components (none of these states is pure due to mixing by 2p-3d electrostatic interactions)

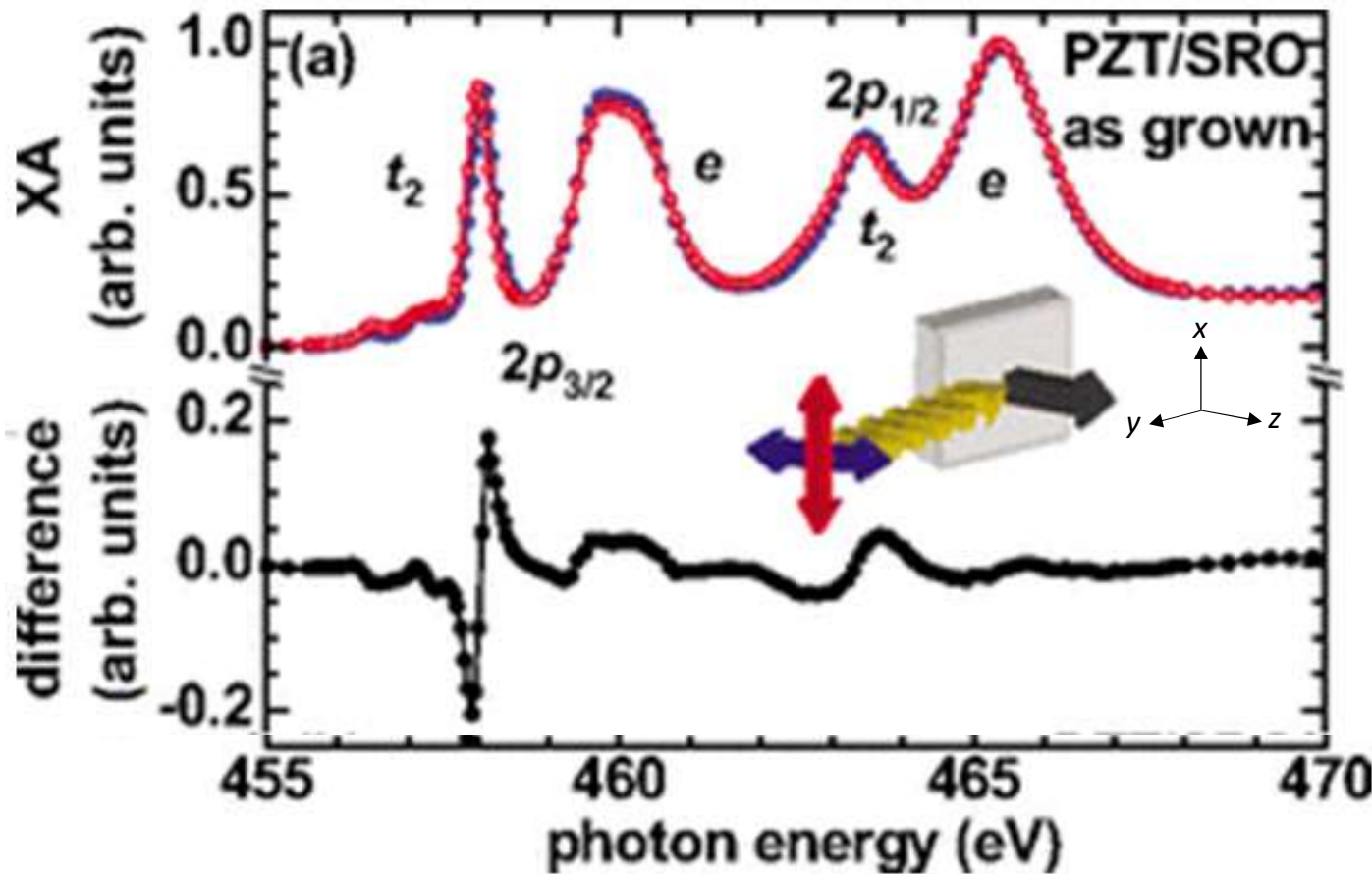


C. Schmitz-Antoniak *et al.*, Nat. Comm. **4**, 2051 (2013)

A. Bhogra *et al.*, Sci. Rep. **9**, 14486 (2019)



The presence of tetragonal symmetry further split both e_g and t_{2g} states: the t_{2g} state in d_{xy} and d_{xz} - d_{yz} components, and the e_g state into $d_{x^2-y^2}$ and d_{z^2} components. Their energy position depend on the amount of contraction and elongation of the distortion, and their energy splitting is often too small to be directly visible in the absorption spectrum, requiring to be analysed by measuring the dichroism between two different directions of linear polarization.

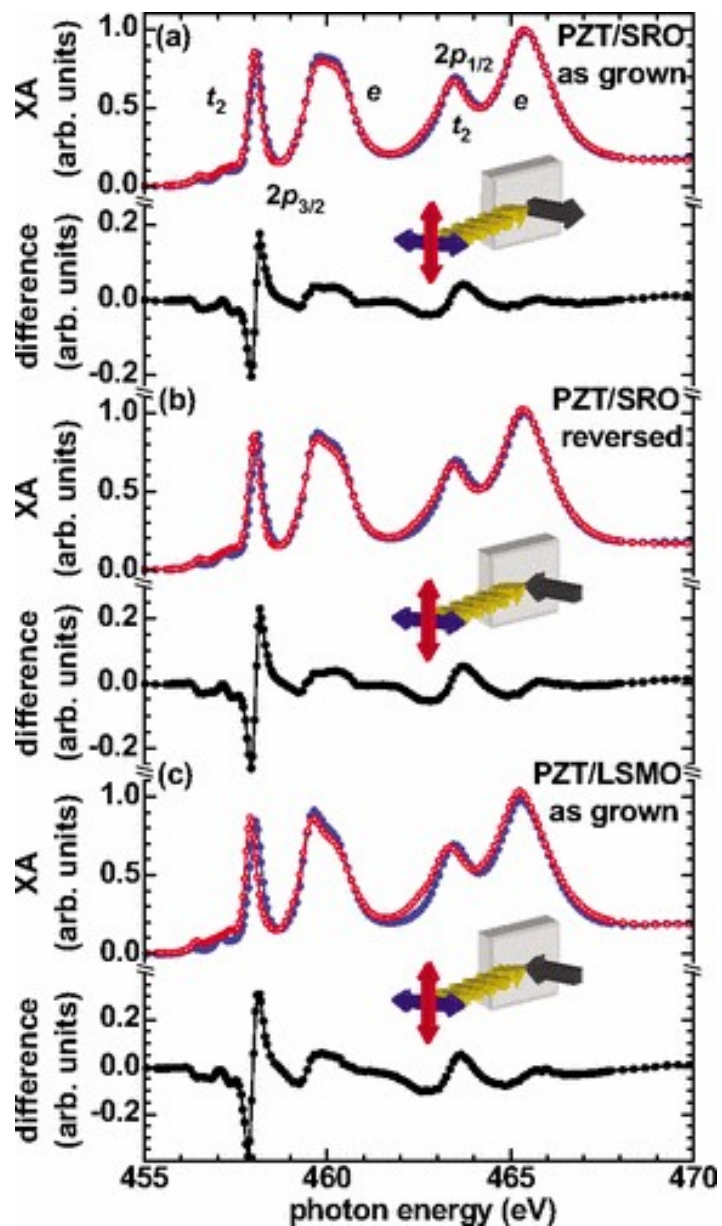


Two adsorption spectra:

- **Red**: X-ray linear polarization in the film plane
- **Blue**: X-ray linear polarization nearly perpendicular to the film (incidence angle 84° from surface normal)

According to the *symmetry* of each component the spectral feature is enhance or suppressed.

The difference spectra (linear dichroism) highlight such symmetry, with differences value of about $\pm 20\%$ mostly visible in the energy range of the $2p_{3/2} t_2$ component



Spectra recorded in different conditions

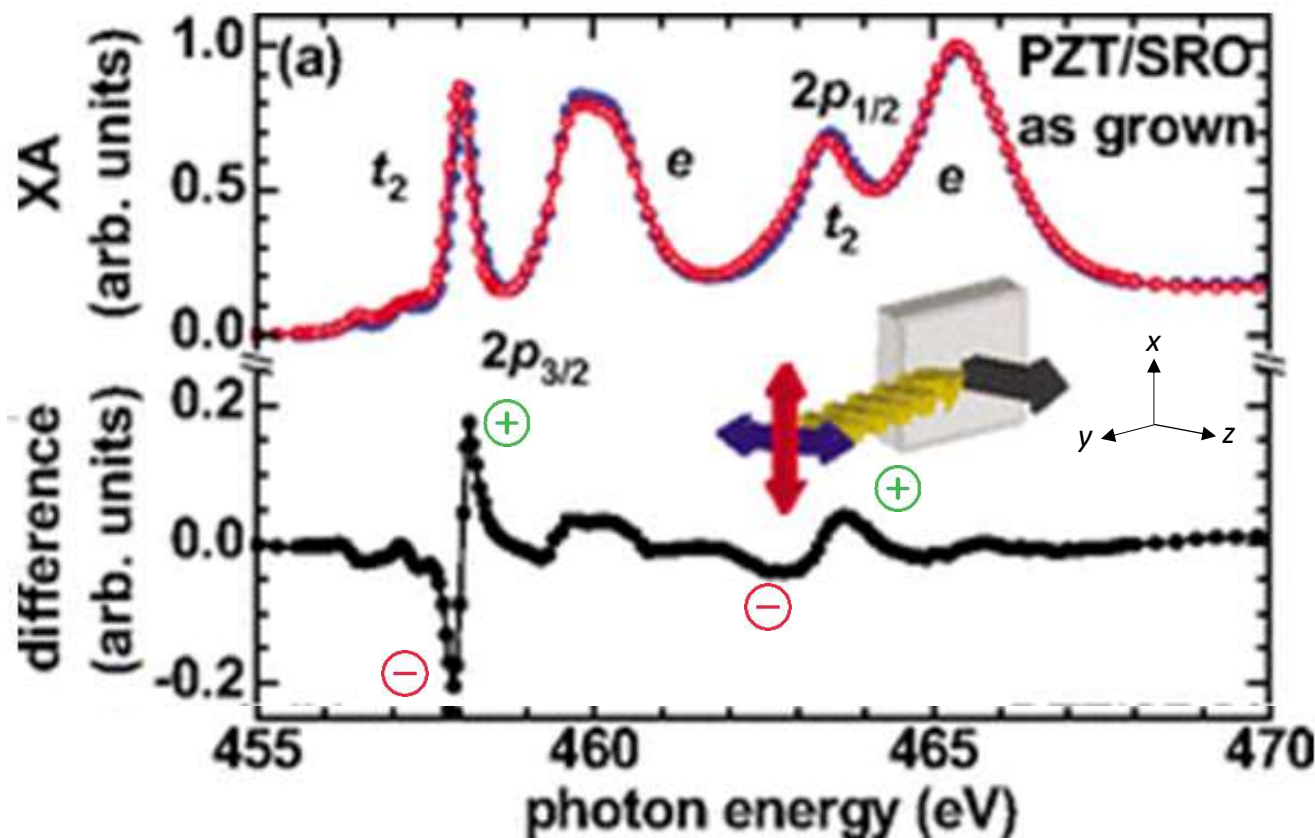
PZT/SRO as grown - Polarization pointing away from the substrate

PZT/SRO reversed - Polarization pointing towards the substrate

PZT/LSMO as grown - Polarization pointing towards the substrate

Spectra and their differences show very similar features

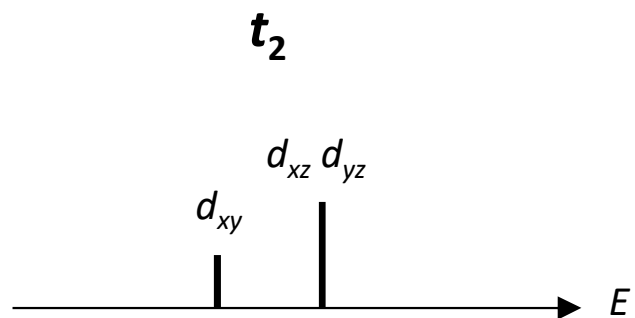
Let's consider the difference spectrum from the XAS intensity with the X-ray polarization vector parallel to the ferroelectric polarization (I_z) and perpendicular to it (I_{xy}) in PZT/SRO.



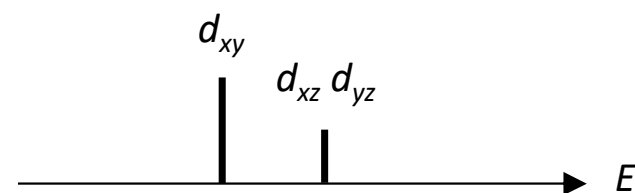
In general, if we take the difference spectrum as $I_z - I_{xy}$ states with mainly (xz,yz) and z^2 character shows up as positive signals, while those with mainly xy and $x^2 - y^2$ character shows up as negative signal.

In the difference spectrum main structures are $-/+$ peaks in correspondence of the t_2 components. Thus by acquiring spectra with different linear polarization and calculating the difference we can distinguish the different “sub” components. Note that such difference is highlighted when the plane normal to the z axis is different from the planes parallel to the z axis.

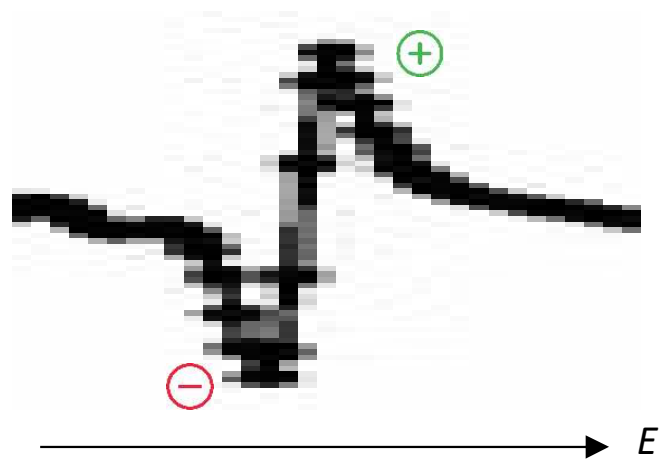
Spectrum with vertical
polarization I_z

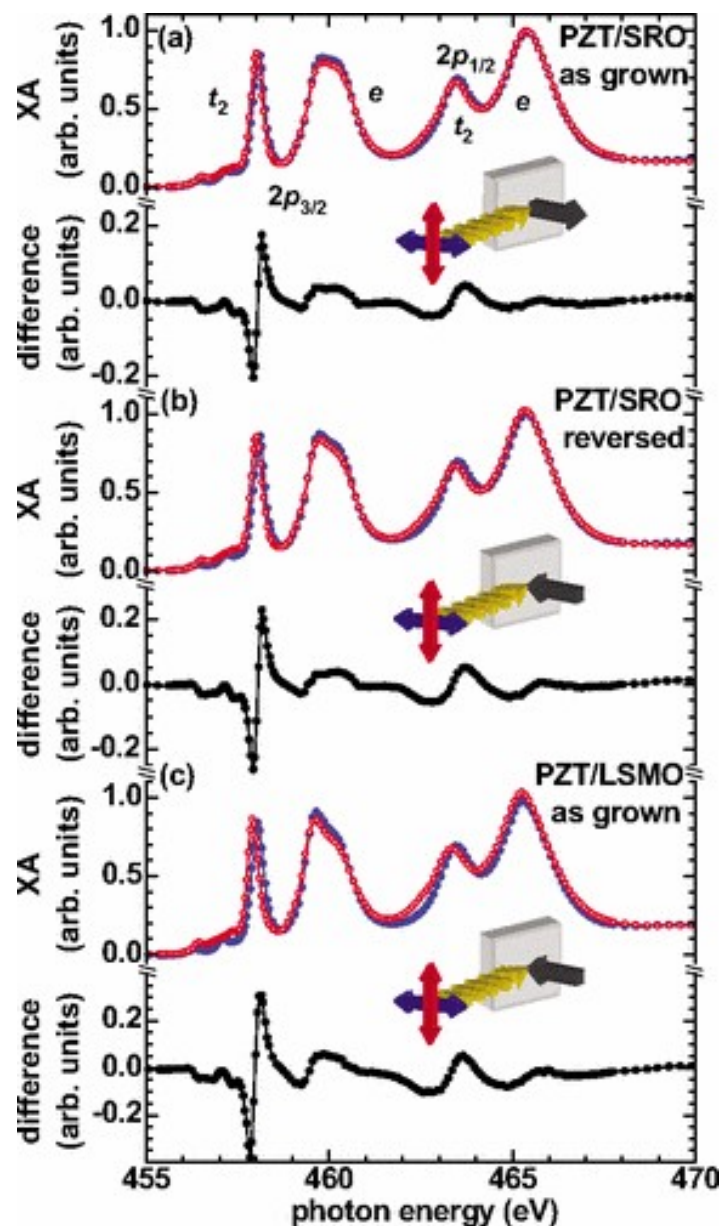


Spectrum with horizontal
polarization I_{xy}



Difference spectrum
 $I_z - I_{xy}$

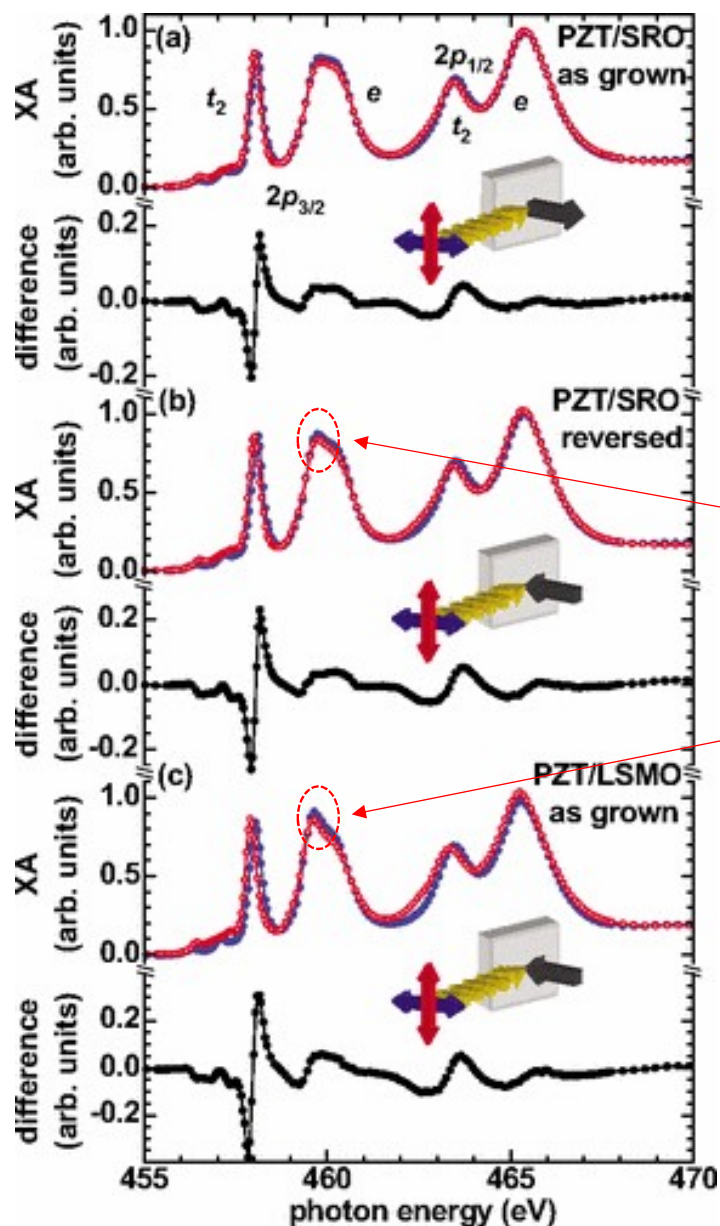




The observed difference in the XAS spectra mainly originate from the splitting in the t_{2g} component, by the fact that the Ti bond directions point along the tetragonal z axis, out of the xy plane.

Even by reversing the polarization direction, which changes somehow the profiles of the XAS spectra, does not change the asymmetry on the charge distributions, since the difference spectrum is practically unchanged.

The observed difference spectrum does not depend from structural variation: by comparing the spectra from PZT/SRO and PZT/LSMO we observe the same differences in both the XAS spectra and in the difference spectra.



PZT/SRO as grown
Polarization pointing
away from the substrate

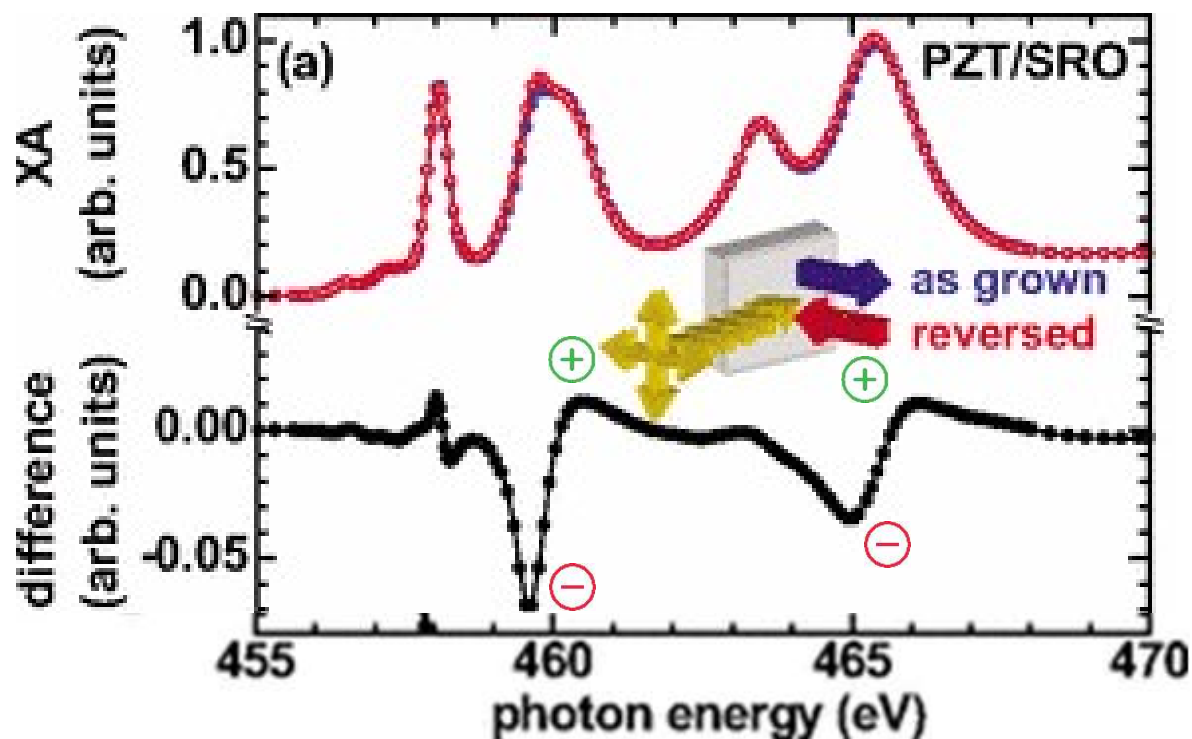
PZT/SRO reversed
Polarization pointing
towards the substrate

PZT/LSMO as grown
Polarization pointing
towards the substrate

Note that spectra and their differences show indeed very similar features, but a slight difference is visible in the structure at the low binding energy side of the $2p_{3/2}$ e feature, which is somehow larger in the PZT/SRO reversed and even larger in the PZT/LSMO, where the polarization was originally pointing towards the substrate.

**What is going on when the
polarization is reversed?**

Let's now consider the difference spectrum from the XAS intensity obtained averaging the two X-ray polarization directions by varying the ferroelectric polarization.

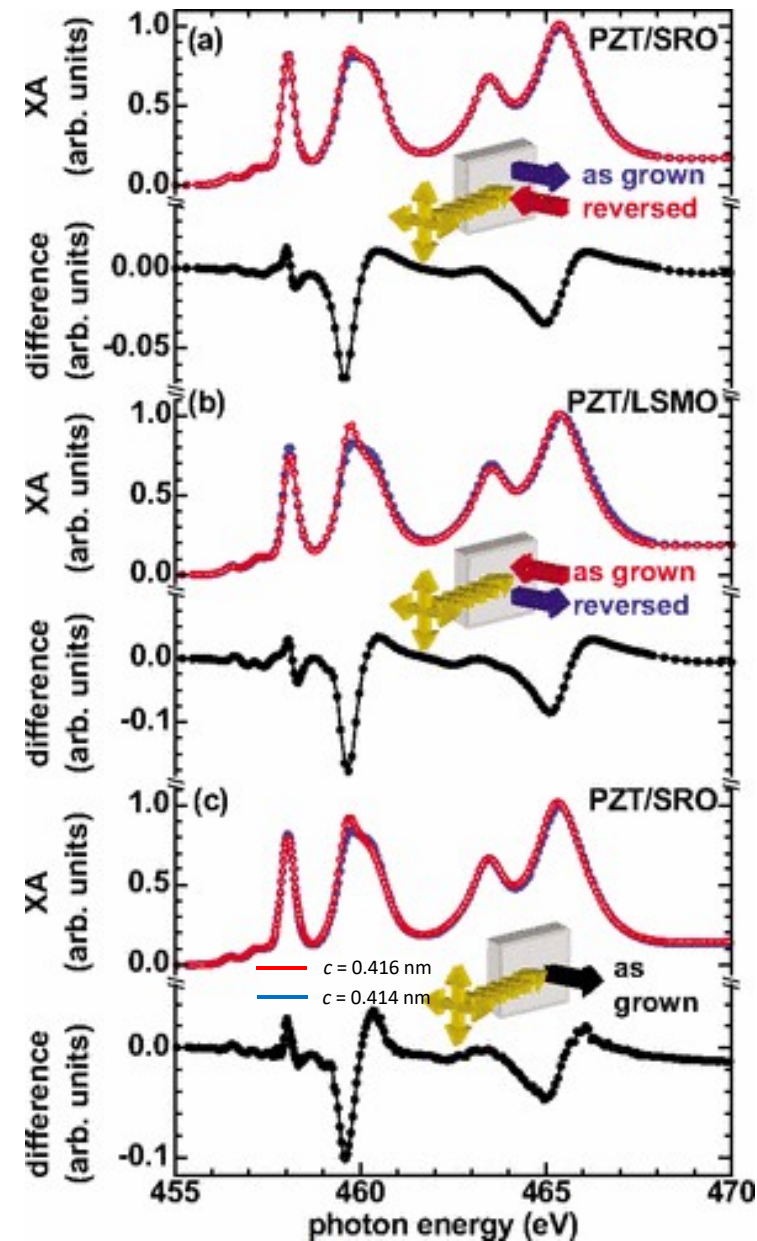


Polarization averaged XAS spectra obtained in the two following situations:

- **Red**: polarization pointing towards the substrate electrode
- **Blue**: polarization pointing away from the substrate electrode
- **Black**: difference spectrum

In this case the main differences appear as $-/+$ peaks in correspondence of the e_g components. This e -split difference spectrum is obtained by introducing an energy splitting between the $d_{x^2-y^2}$ and the d_{z^2} .

This means the z axis is distinct from the x and y axes, i.e., the electrostatic energy is different along z from the x and y axes, corresponding to contraction or elongation of the z axis.



To explain these differences one has to assume that upon reversing electric polarization also the structure of the PZT has slightly changed.

The $d_{x^2-y^2}$ has lower energy than the d_{z^2} level for the polarization pointing toward the bottom electrode than away from it for both systems, PZT/SRO and PZT/LSMO. This is equivalent to an increase in the electrostatic potential along the z axis, such as caused by contraction along the z direction, when the polarization points away from the bottom electrode compared to when the polarization points toward the bottom electrode.

To confirm the presence of the structural changes, one can compare the polarization averaged XAS spectra of PZT/SRO systems with varying PZT layer thickness resulting in lattice constants $c = 0.414$ nm and 0.416 nm (ferroelectric polarization points away from the bottom electrode): the sample with $c = 0.416$ (the sample with elongated c axis) shows the anticipated changes in the intensity of the $2p_{3/2} e_g$ feature, confirming the finding of the structural changes associated with the reversal of the ferroelectric polarization in PZT.

RESULTS

XAS/XLD ALLOWS **ELEMENT SENSITIVE** INVESTIGATION
OF CHARGE ANISOTROPY IN FERROELECTRIC SYSTEMS

THE FERROELECTRIC ORDER IN PZT THIN FILMS IS REVEALED BY THE PRONOUNCED DIFFERENCES IN THE X-RAY ABSORPTION BETWEEN SPECTRA MEASURED WITH THE **X-RAY POLARIZATION PERPENDICULAR AND PARALLEL** TO THE FERROELECTRIC POLARIZATION, DUE TO THE REDUCTION IN SYMMETRY AROUND THE Ti^{4+} IONS.

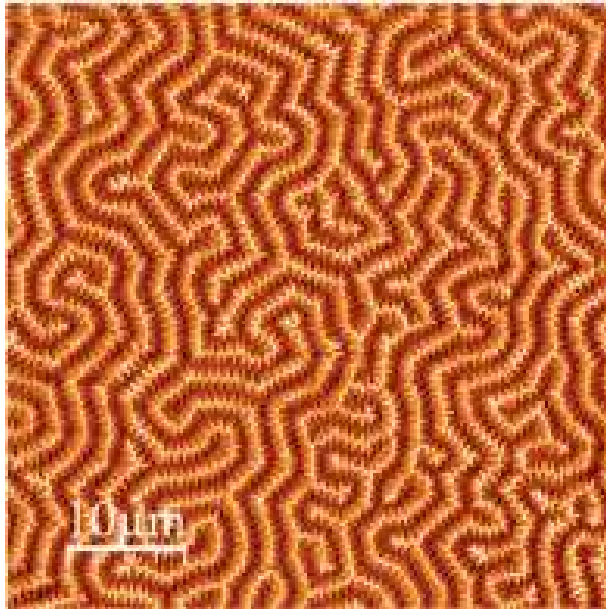
EXPERIMENTAL **EVIDENCE IS FOUND FOR CHANGES IN THE PZT STRUCTURE** INDUCED BY THE REVERSAL OF THE FERROELECTRIC POLARIZATION.

THIS **PRONOUNCED LINEAR POLARIZATION DEPENDENCE** IS A GENERAL PHENOMENON EXPECT TO BE FOUND IN ANY FERROELECTRIC AND MULTIFERROIC SYSTEM.

MICROSCOPY EXPLOITING POLARIZED RADIATION: IMAGING MAGNETIC AND FERROELECTRIC DOMAINS

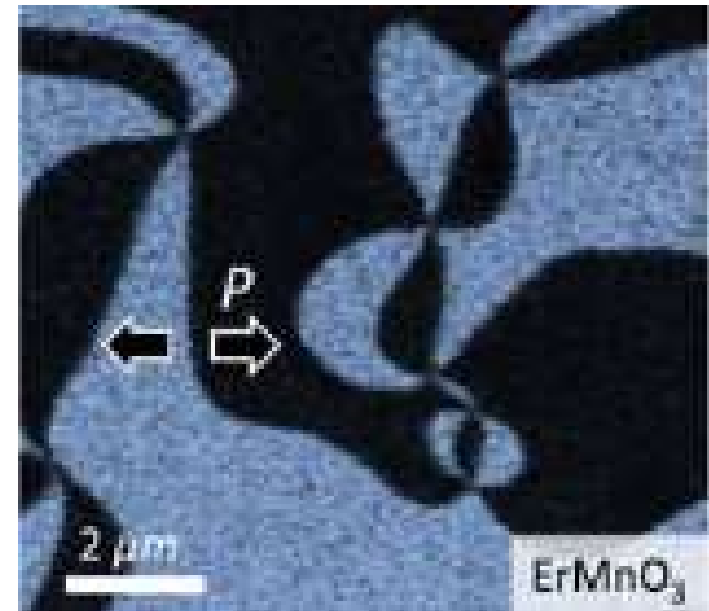
Magnetic/ferroelectric domains are area with uniform magnetization or electrical polarization and carry a wealth of information of the investigated materials

Magnetic domains in Fe_3Sn_2 observed at room temperature by a magnetic force microscopy



K. Heritage *et al.*, Adv. Funct. Mater. **30**, 1909163 (2020)

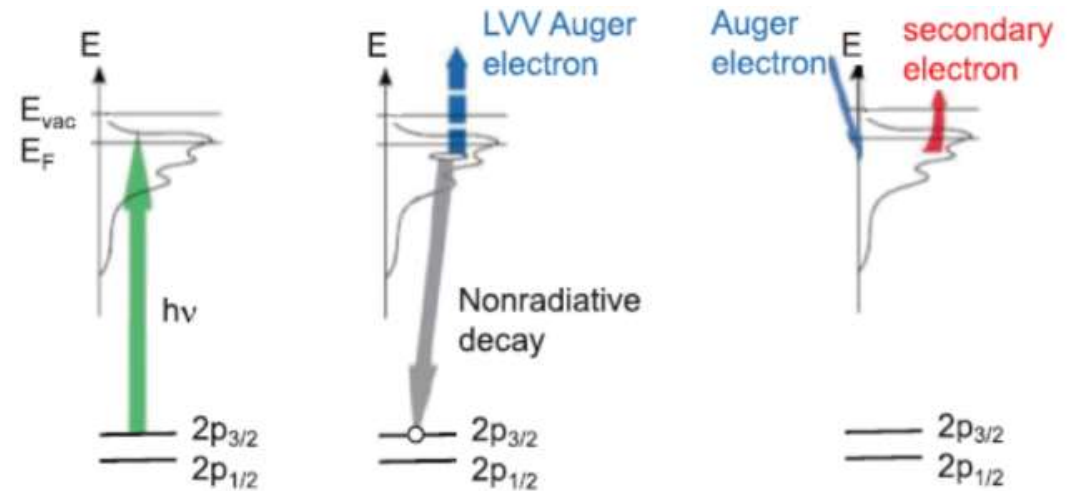
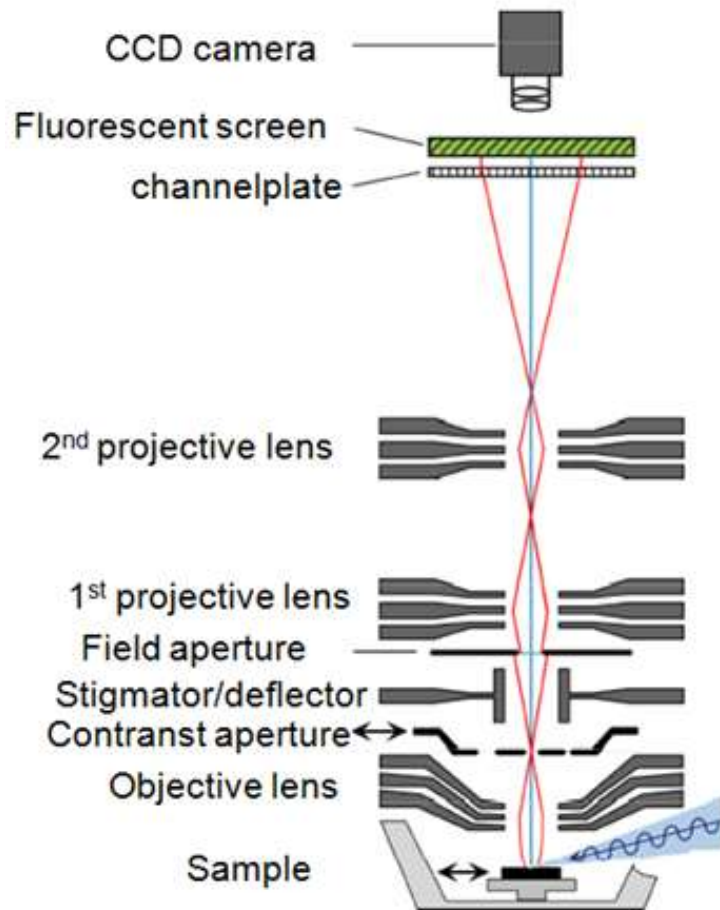
Ferroelectric domains in ErMnO_3 observed at room temperature by a piezoresponse force microscopy



E. Hassanpour *et al.*, New J. Phys. **18**, 043015 (2016)

With a photoemission electron microscope (PEEM), exploiting the polarization dependent X-ray absorption, one can image both ferroelectric and magnetic domain and investigate the interaction in these class of materials

PHOTOEMISSION ELECTRON MICROSCOPE (PEEM)



M. Ghidini, *et al.*, Adv. Electron. Mater. **8**, 2200162 (2022)

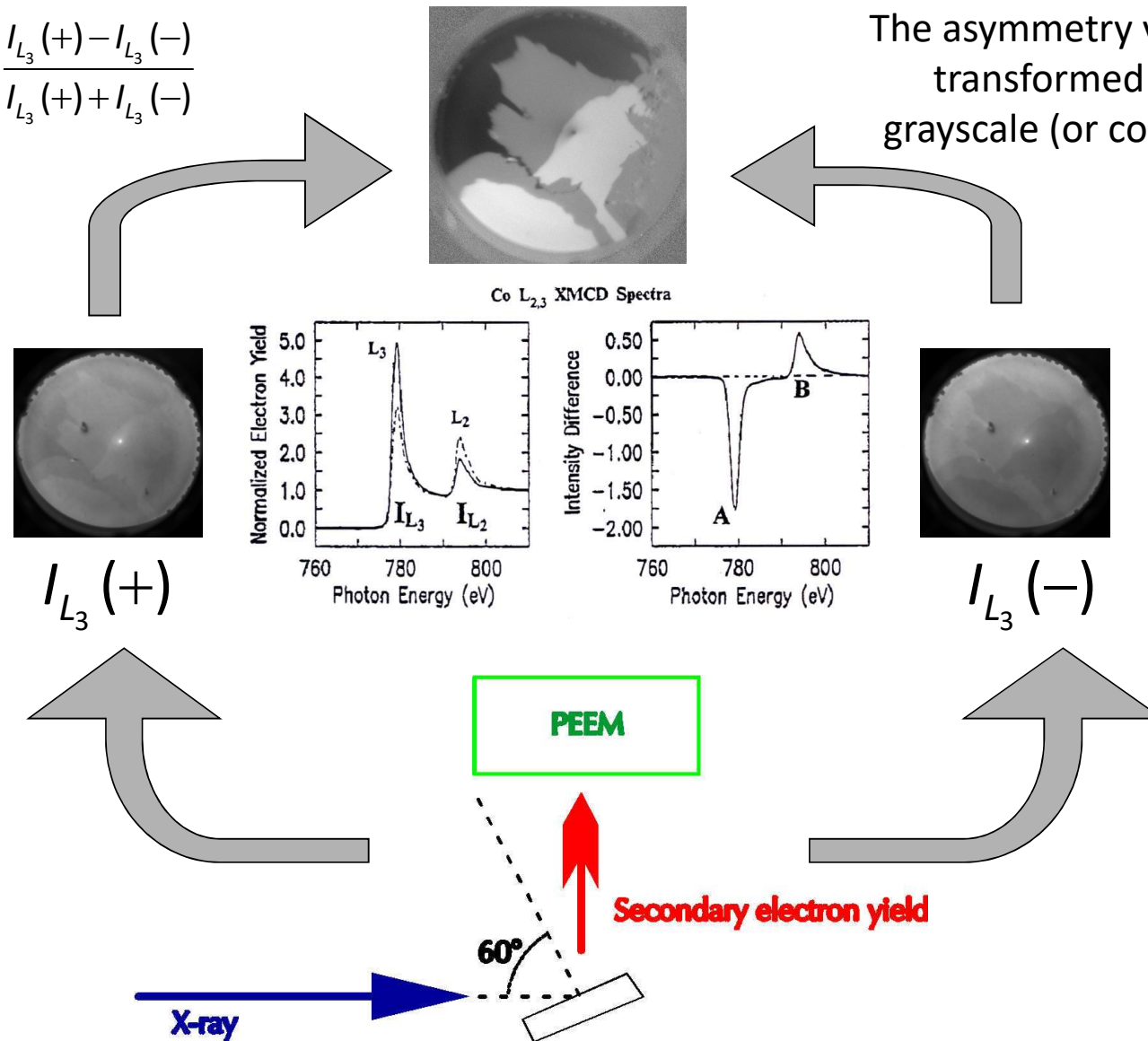
- ✓ Sensitivity to chemical elements
- ✓ Access to buried layers (within the secondary electron information depth)
- ✓ Good lateral resolution (typically a few hundred nm for ferro-electric/magnetic structures)

Magnetic domain imaging with XMCD-PEEM

Asymmetry is calculated as

$$Asy = \frac{I_{L_3}(+) - I_{L_3}(-)}{I_{L_3}(+) + I_{L_3}(-)}$$

The asymmetry values are transformed into a grayscale (or color scale)



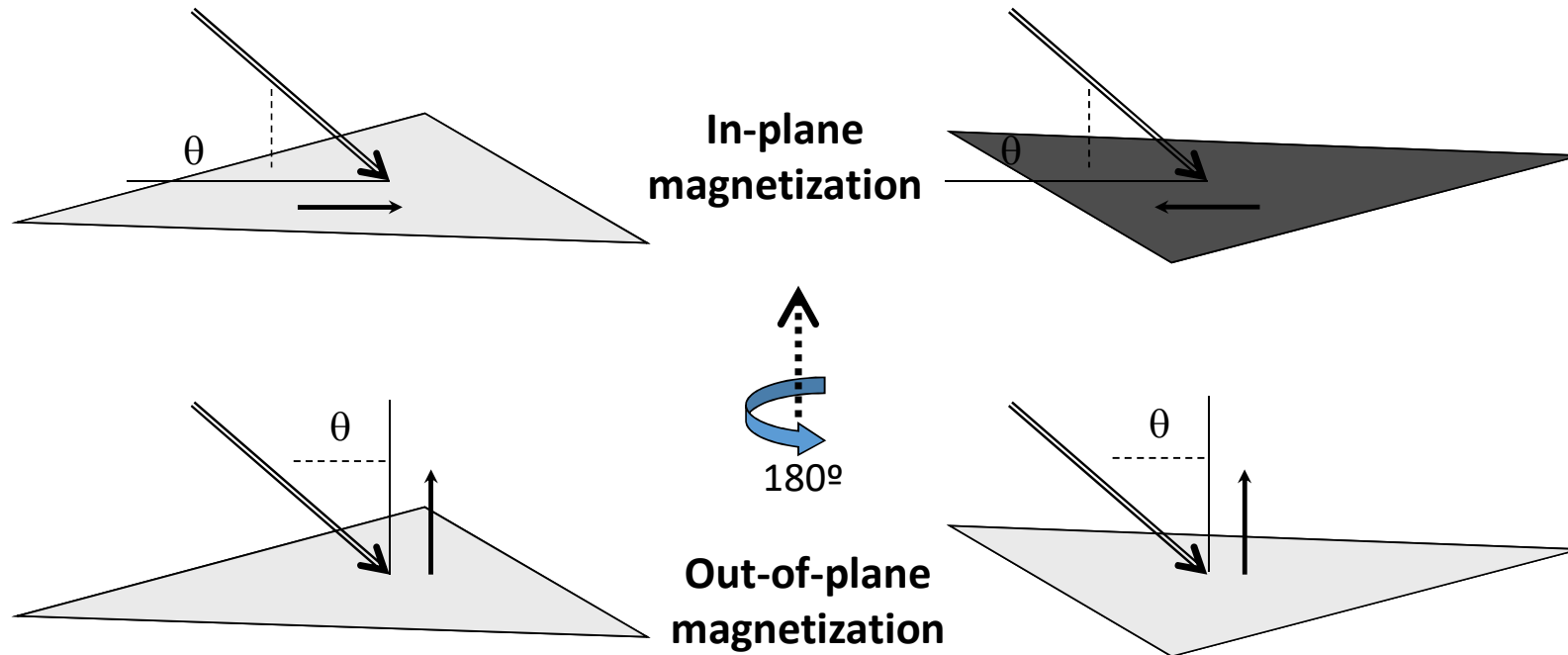
Magnetic contrast

$$A \sim \mathbf{q} \cdot \mathbf{M} \sim \cos\theta$$

$\mathbf{q}$ = X-ray propagation direction

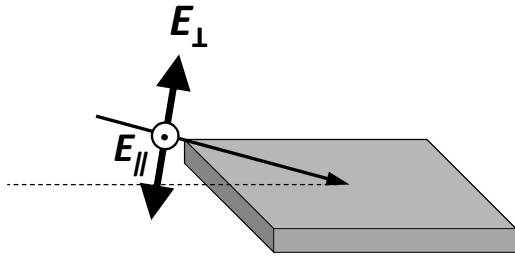
$\mathbf{M}$ = magnetization direction

θ = angle between X-ray propagation direction and magnetization direction



By fixing the X-ray incidence direction and rotating the sample by 180°, we find that **magnetic domains with in-plane magnetization exhibit opposite asymmetry values** (bright domains become dark and *vice versa*) whereas **magnetic domains with out-of-plane magnetization do not change their magnetic contrast**

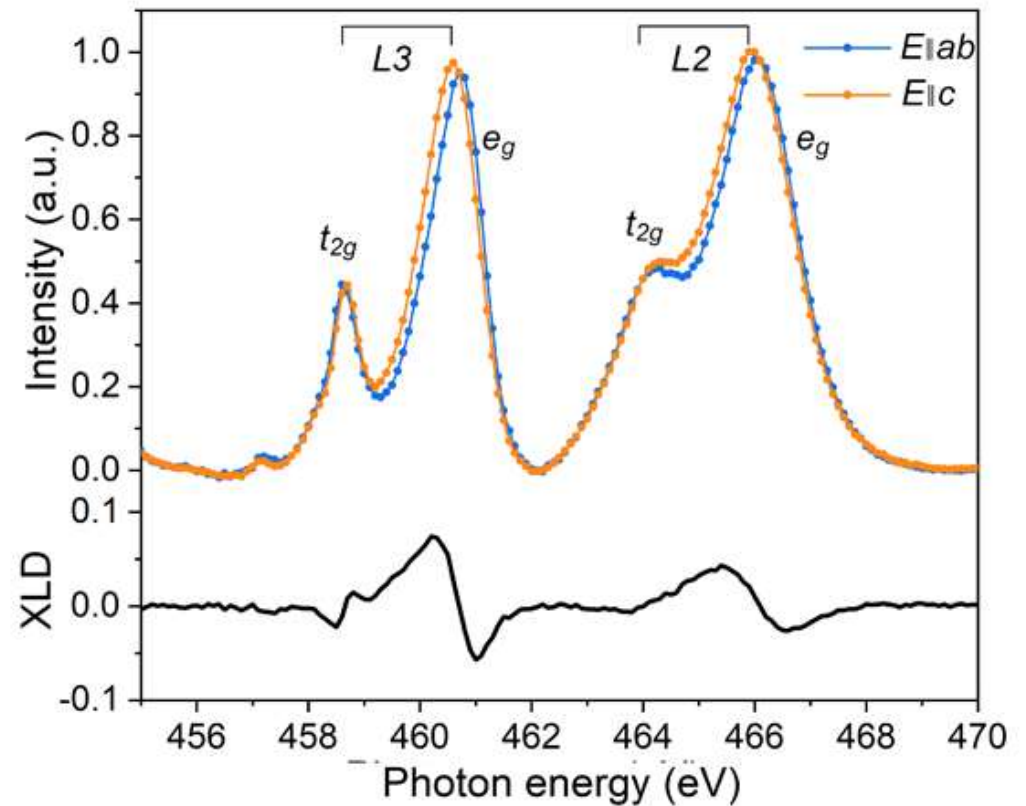
Ferroelectric domain imaging with XLD-PEEM



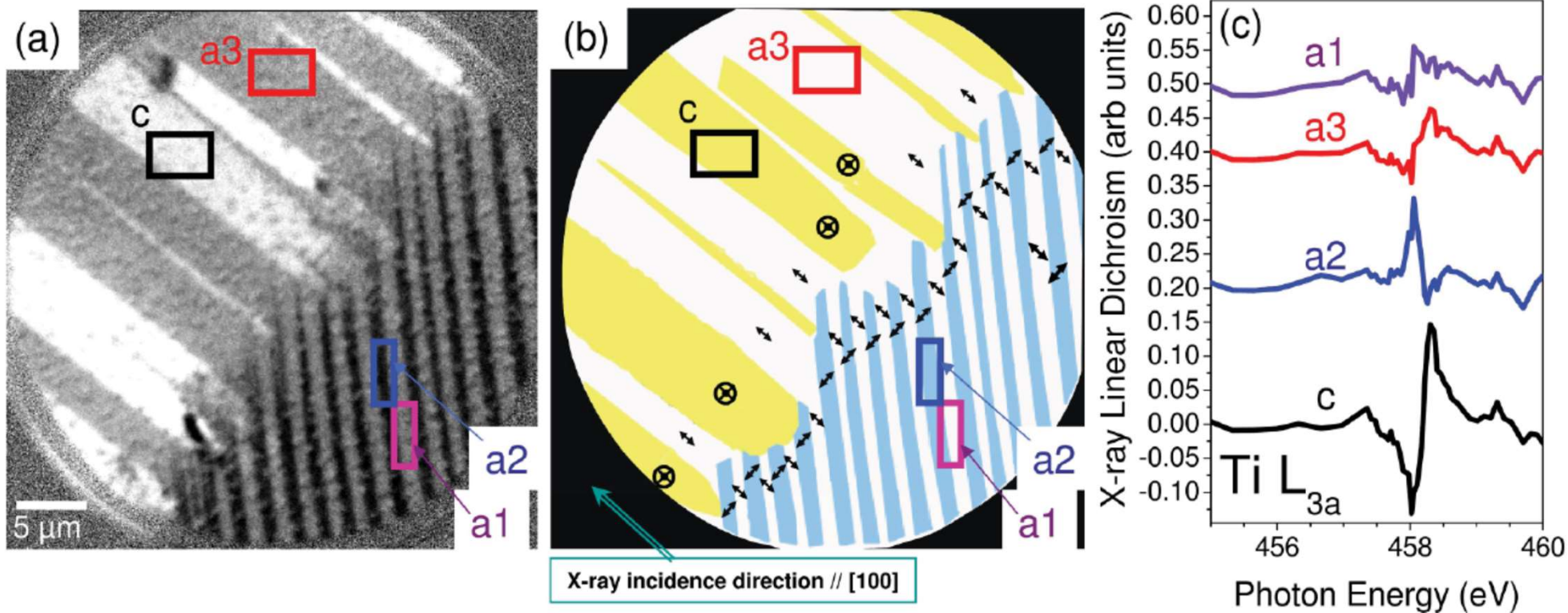
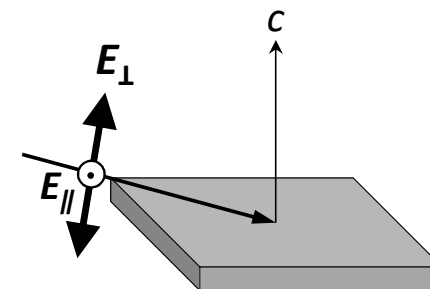
One of the way to calculate, at a given photon energy, the **asymmetry** is

$$Asy = \frac{I_{L_3}(ab) - I_{L_3}(c)}{I_{L_3}(ab) + I_{L_3}(c)}$$

XAS and XLD spectra of Ti in BaTiO₃



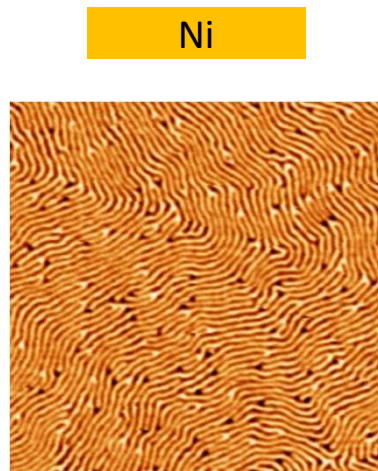
Example: Ferroelectric domains configuration in a BaTiO_3 single crystal showing perpendicular (c domains) and in-plane (a domains) polarized domains



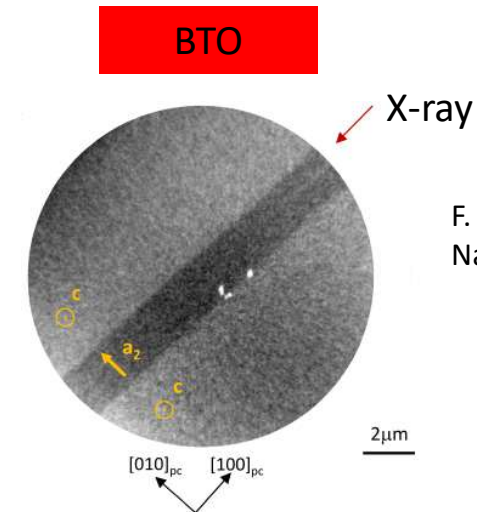
R. V. Chopdekar *et al.*, Phys. Rev. B **86**, 014408 (2012)

Electrical control of the magnetization

Interaction at the interface between a ferromagnetic Ni thin film and a ferroelectric BaTiO₃ (BTO) substrate



Due to the presence of stress in Ni ferromagnetic film out of plane (or canted) easy magnetization axis can develop, showing as stripes domain with alternating out-of-plane magnetization



F. Maccherozzi *et al.*,
Nat. Comm. **16**, 8445 (2025)

In a BTO crystal different kind of ferroelectric domains develop, mainly out-of-plane c domain and in-plane a domain

What happens at the interface of a nickel film on top of a BTO substrate? Can one control the magnetization of the nickel film solely via electrical interaction of the substrate?

100 nm



We will see that ferromagnetic and ferroelectric domains are correlated!

How can we change the magnetization configuration in nickel without applying a magnetic field?

APPLYING VOLTAGE



https://www.dreamstime.com/chudtsankov_info

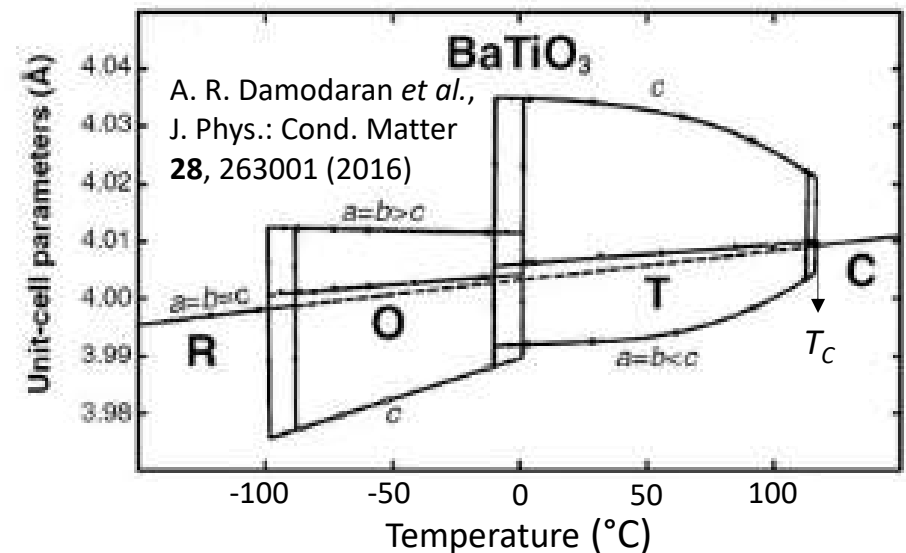
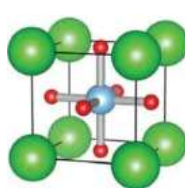
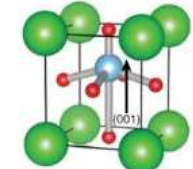
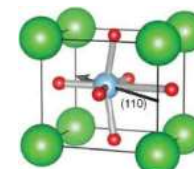
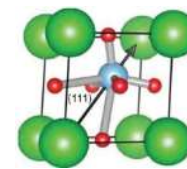
CHANGING TEMPERATURE

Rhombohedral

Orthorhombic

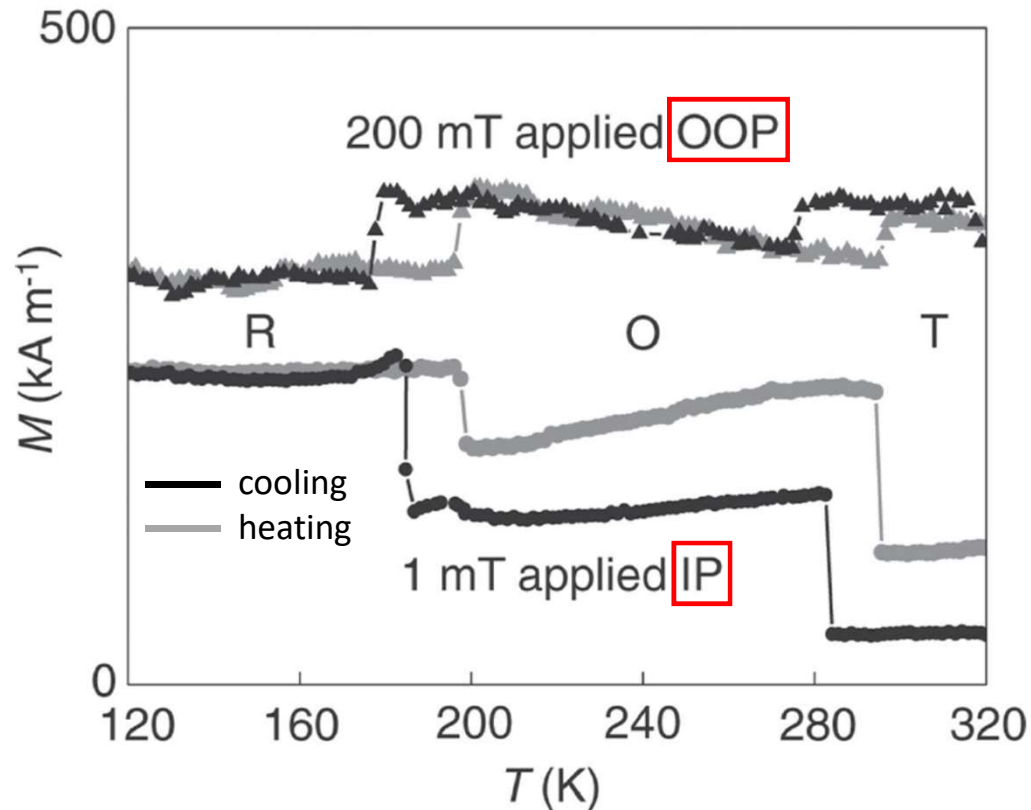
Tetragonal

$T > T_c$
Cubic



Exploiting XMCD and XLD in a PEEM microscope we can look at the changes in domain configuration on both nickel and BTO

OOP = out-of-plane magnetization
IP = in-plane magnetization



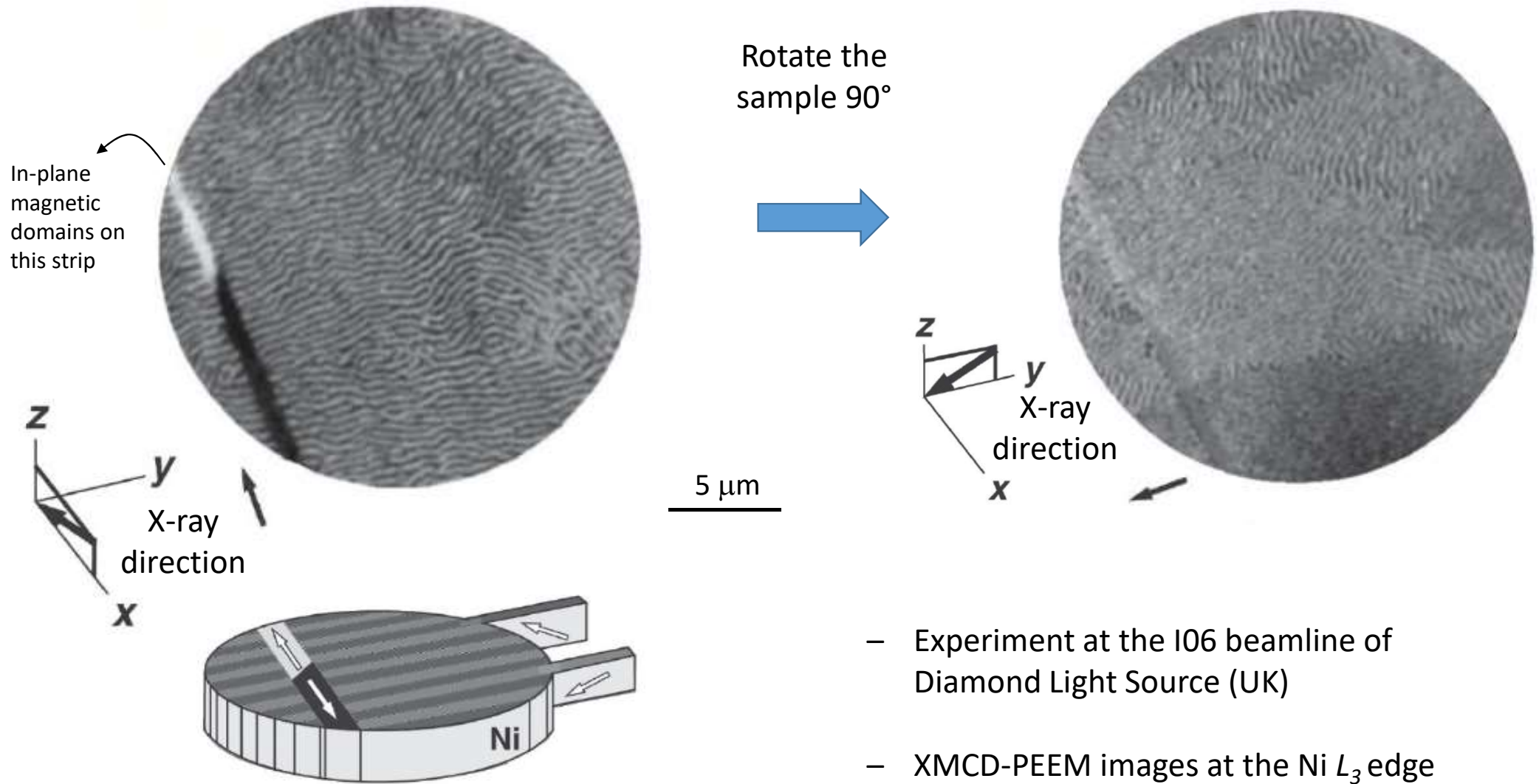
Thermally driven structural phase transitions in the substrate yield sharp hysteretic jumps of OOP and IP magnetization: On cooling, these jumps represent an increase of IP magnetization at the expense of OOP magnetization.

After returning to room temperature in non-saturating magnetic fields, IP and OOP magnetizations are slightly modified due to the stochastic nature of magnetic domain-wall depinning, but easy and hard-axis loops are recovered.

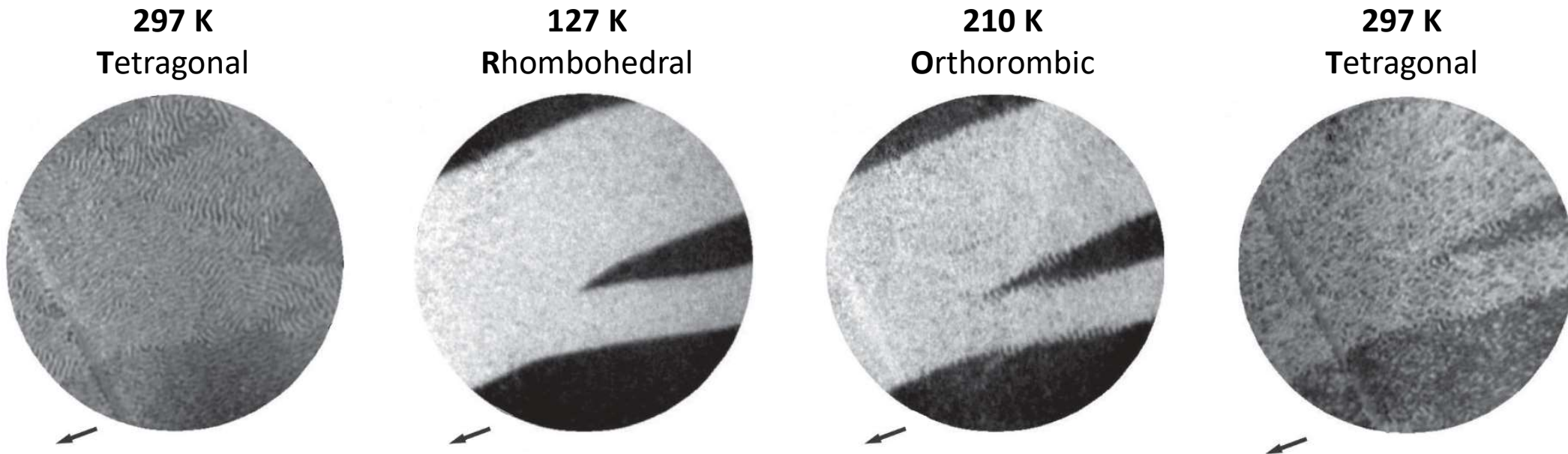
Therefore strains effects observed on cooling to R-phase BTO appear (in average) to be reversible.

What is going on?

Out-of-plane *stripes* domain in Ni: room temperature



Magnetic domains in Ni: temperature evolution

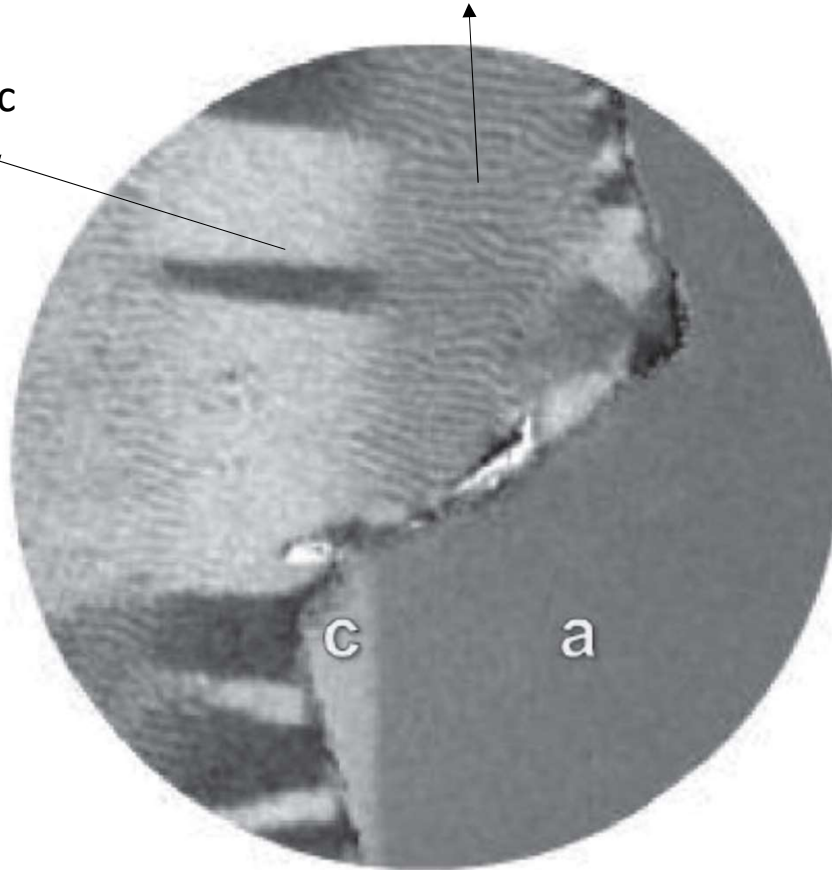


On cooling to the R-phase of the substrate, stripe domains are annihilated in favor of a uniform IP magnetization. This annihilation is due to suppression of the OOP anisotropy responsible for stripe domains, consistent with the magnetization data. The resulting IP domains are antiparallel, evidencing the development of a uniaxial IP anisotropy in the imaged field of view. Subsequent warming to the O-phase of the substrate nucleates stripes at IP domain walls. Returning to the room temperature T-phase recovers stripe domains that are less wavy and more straight than before.

Relation between ferroelectric and magnetic domains configuration

In-plane magnetic domain in Ni

Out-of-plane magnetic domain in Ni



Build up a picture combining two images: a PEEM image obtained with XMCD contrast, and a PEEM image of the exposed substrate obtained with XLD contrast

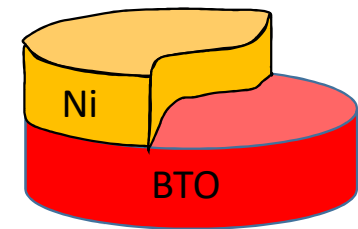


Out-of-plane c domain in BTO



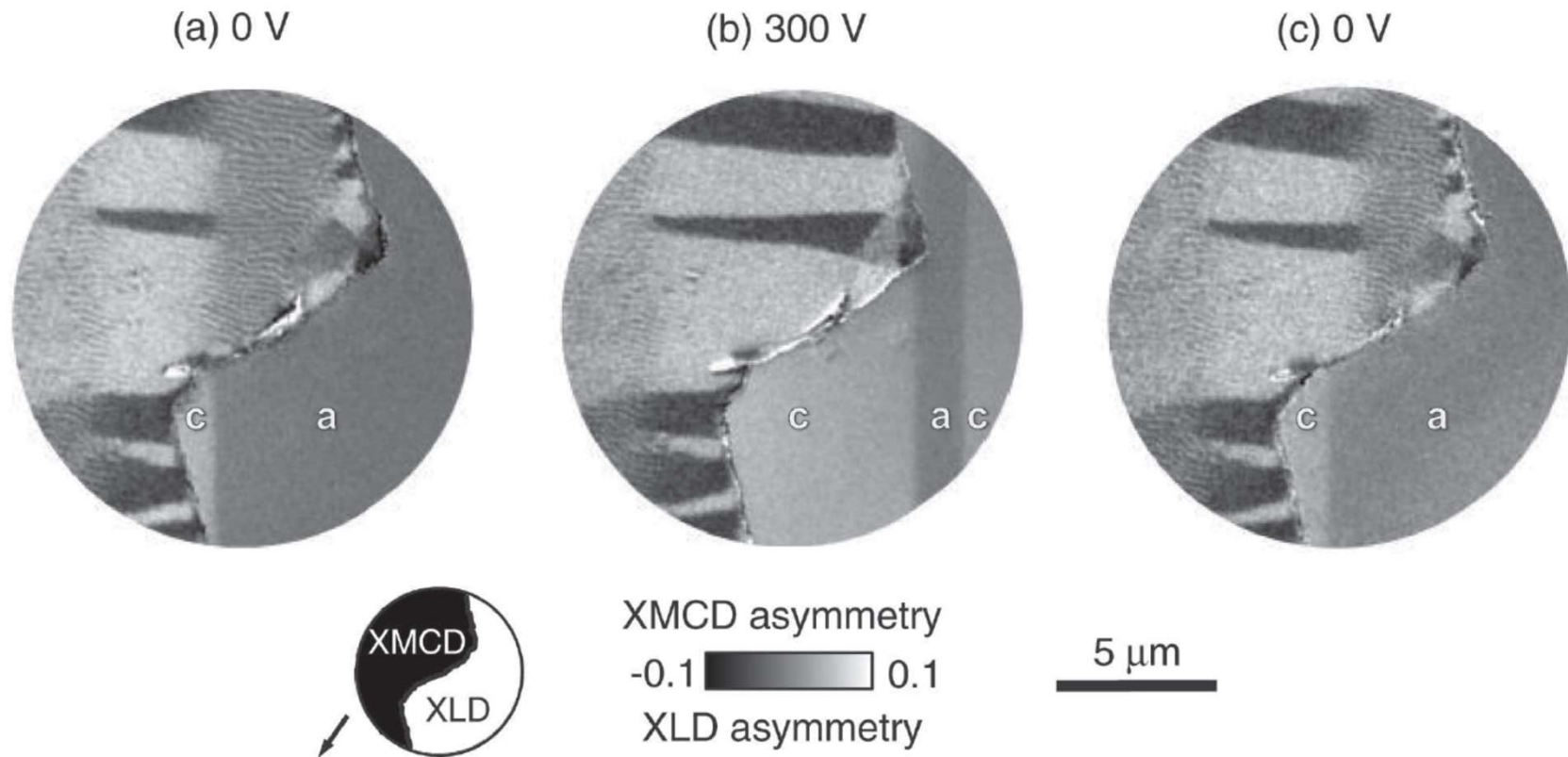
In-plane a domain in BTO

Look at an area where there is an edge, leaving the BTO substrate uncovered



- Out-of-plane magnetized stripe domains in nickel exist on the a in-plane polarized ferroelectric domains in BTO
- In-plane magnetized domains in nickel exist on the c out-of-plane polarized ferroelectric domains in BTO

Magnetic domains in Ni: evolution through electric field



The *c* ferroelectric domain is associated to IP ferromagnetic domains in Ni (IP domains near the zig-zag edge arise due to relief of growth stress by delamination). The application of an electric field impose a displacement of the domain wall, and an increase of the *c* domain at the expense of the *a* domain, concomitant with the annihilation of the stripes magnetic domains in Ni. The process is essentially reversible: the electrical annihilation of stripe domains in favor of IP domains is due to the conversion of underlying *a* to *c* domains.

M. Ghidini, *et al.*, Advanced Materials **27**, 1460 (2015)

RESULTS

IMAGING TECHNIQUES BASED ON POLARIZATION-DEPENDENT X-RAY ABSORPTION SPECTROSCOPY ARE WELL SUITED FOR THE INVESTIGATION OF FERROMAGNETIC AND FERROELECTRIC SYSTEMS

INTERFACIAL INTERACTIONS BETWEEN FERROMAGNETIC AND FERROELECTRIC MATERIALS CAN BE ELUCIDATED THROUGH THE INVESTIGATION OF THEIR RESPECTIVE FERROMAGNETIC AND FERROELECTRIC **DOMAIN STRUCTURES**

LOCAL **VARIATIONS IN THE MAGNETIZATION** OF A FERROMAGNETIC SYSTEM CAN ALSO BE INDUCED BY THE APPLICATION OF AN **EXTERNAL ELECTRIC FIELD**

SUMMARY

- ✓ Exploiting the strong electric polarization of X-rays produced by synchrotron radiation, detailed information can be obtained on the structural, electronic, and magnetic properties of condensed matter
- ✓ Circular and linear dichroism can be employed to detail the electronic and magnetic properties of ferromagnetic and ferroelectric heterostructures
- ✓ This opens up the possibility of performing a comprehensive characterization of nanostructures and heterostructures of interest for spintronics and electronic applications.