Holcomb Group Capabilities

Synchrotron Radiation & Ultrafast Optics

West Virginia University

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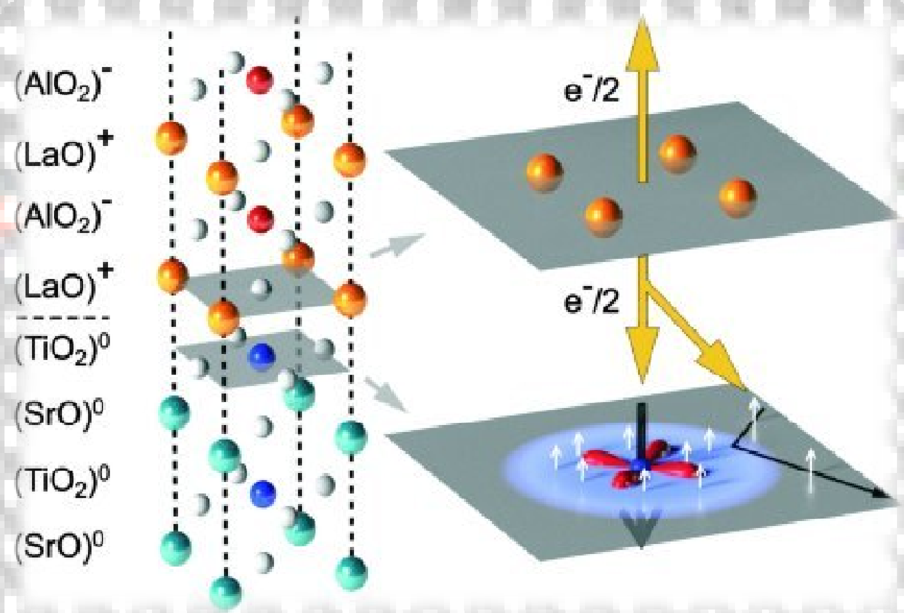
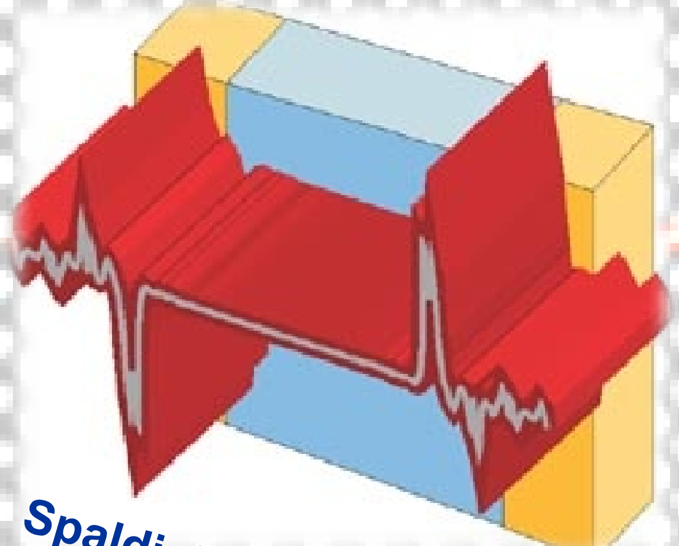
The Physicists' New Playground

"The interface is the device."

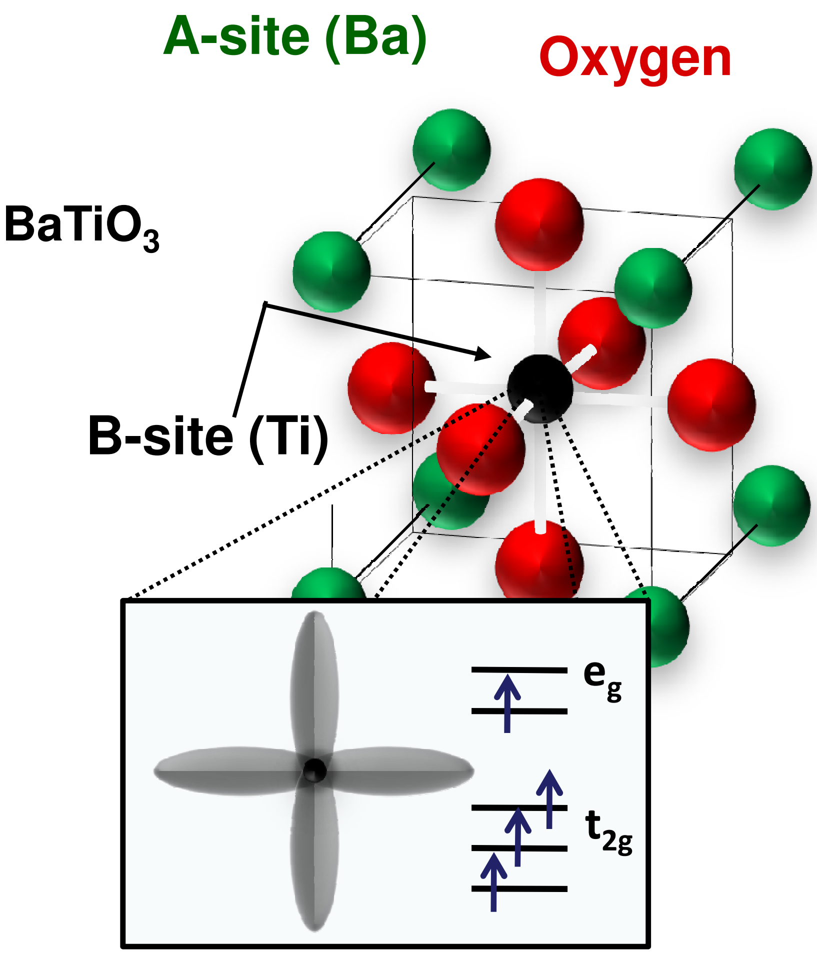
- Herbert Kroemer, beginning of his Nobel Lecture in 2000





Spaldin's spin capacitor

Complex Oxides



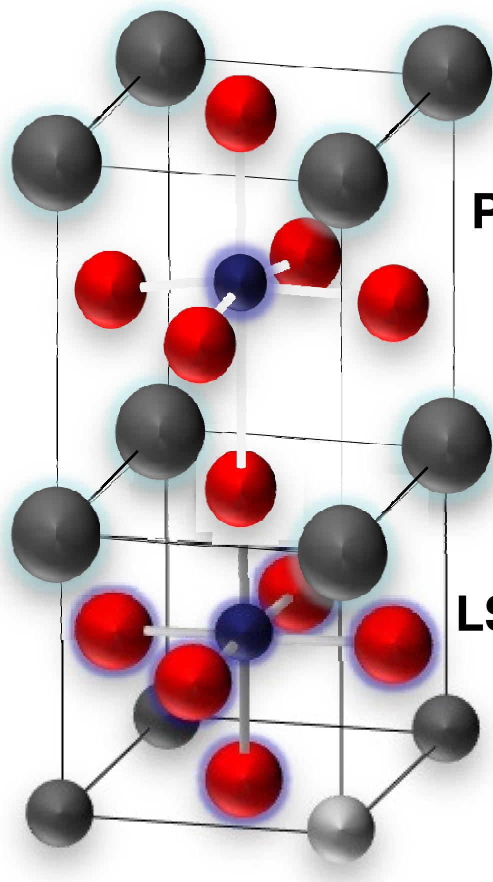
Perovskites

- Superconductors ($YBa_2Cu_3O_{7-\delta}$)
- Ferroelectrics ($BaTiO_3$)
- Colossal Magnetoresistance (($(La,Sr)MnO_3$)
- Multiferroics ($BiFeO_3$)
- High ε_r Insulators ($SrTiO_3$)
- Low ε_r Insulators ($LaAlO_3$)
- Conductors (Sr_2RuO_4)
- Thermoelectrics (doped $SrTiO_3$)
- Ferromagnets ($SrRuO_3$)

All perovskite-related structures *a,b* ~ 3.8-4.0 Å

A huge range of oxide crystals : pyrochlores, layered structures, spinels, rock salt, …

Magnetic & Ferroelectric Heterostructures



Hill (Spaldin), *J. Phys. Chem. B* 104, 6694 (2001)
Kimura, *et al.*, *Nature* 426, 55 (2003)
Hur,…, Cheong, *Nature* 429, 392 (2004)
Feibig, *J. Phys. D* 38, R123 (2005)
Eerenstein, *et al.*, *Nature* 442, 759 (2006)
Cheong & Mostovoy, *Nature Mater.* 6, 13 (2006)

Ferroelectric Perovskites (e.g. $PbZrTiO_3$)

- Non-cubic, spontaneous polarization
- "d^0-ness"
- *6s* electrons on A-site

Magnetic Perovskites (e.g. $La_{0.7}Sr_{0.3}MnO_3$)

- Spontaneous magnetization
- Partially filled d-orbitals
- Doping through A-site substitution

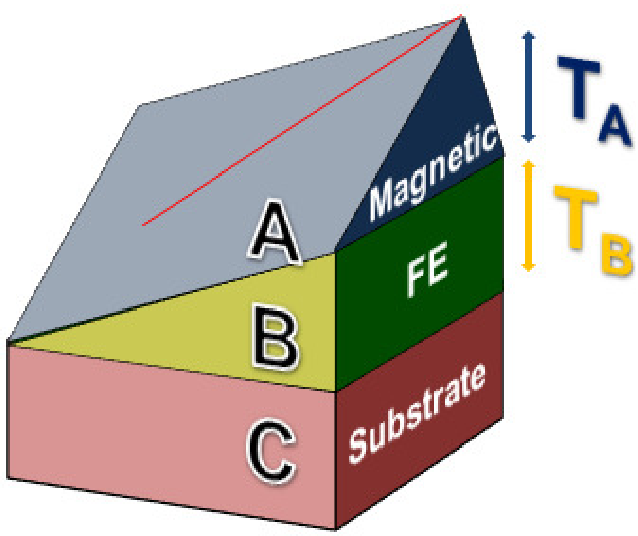
Multiferroic Perovskites

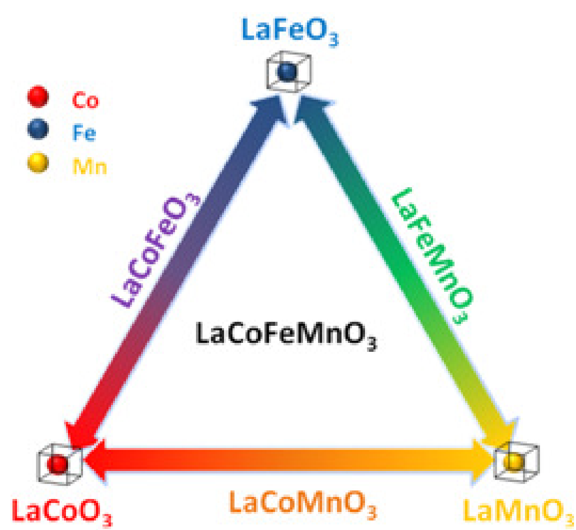
- Multiple order parameters
- *Magnetoelectricity* – coupling between electric and magnetic order parameters
- Many pathways to create and influence coupling (strain, thickness, roughness)
- Allow understanding of interfacial physics

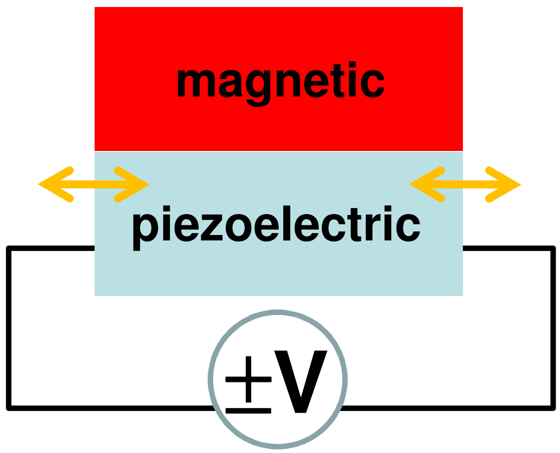
Sample Strategies

The following the heterostructures platforms has been chosen to allow the study of many parameters (including strain and thickness dependence) in the same sample. In this manner we may rule out variations that occur when samples are grown at different times, such as on various substrates. We also compare these results to more standard heterostructures.

<u>Identify the Major Contributor(s) to and Influencers on Interface Coupling</u>. The effects of thickness, strain and material choice will be investigated by studying wedged, compositionally spread, and piezoelectrically strained (Biegalski, 2010) materials.



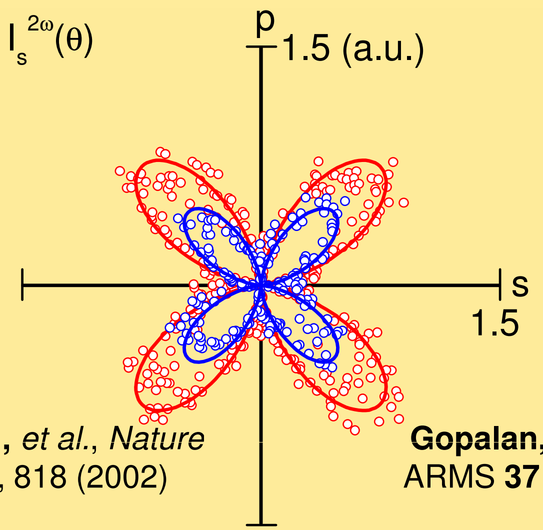




Surface/Interface Characterization

- Imaging domains and determining magnetic and ferroelectric directions in multifunctional materials
- Magnetic dead layers
- Interface imaging

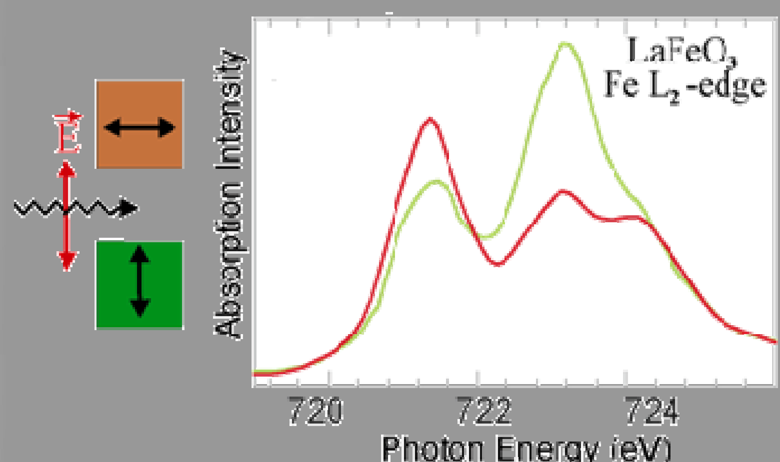
Ultrafast Optics (i.e. SHG)



Fiebig, *et al.*, *Nature* **419**, 818 (2002)

Gopalan, *et al.*, ARMS **37** (2007)

X-ray Dichroism



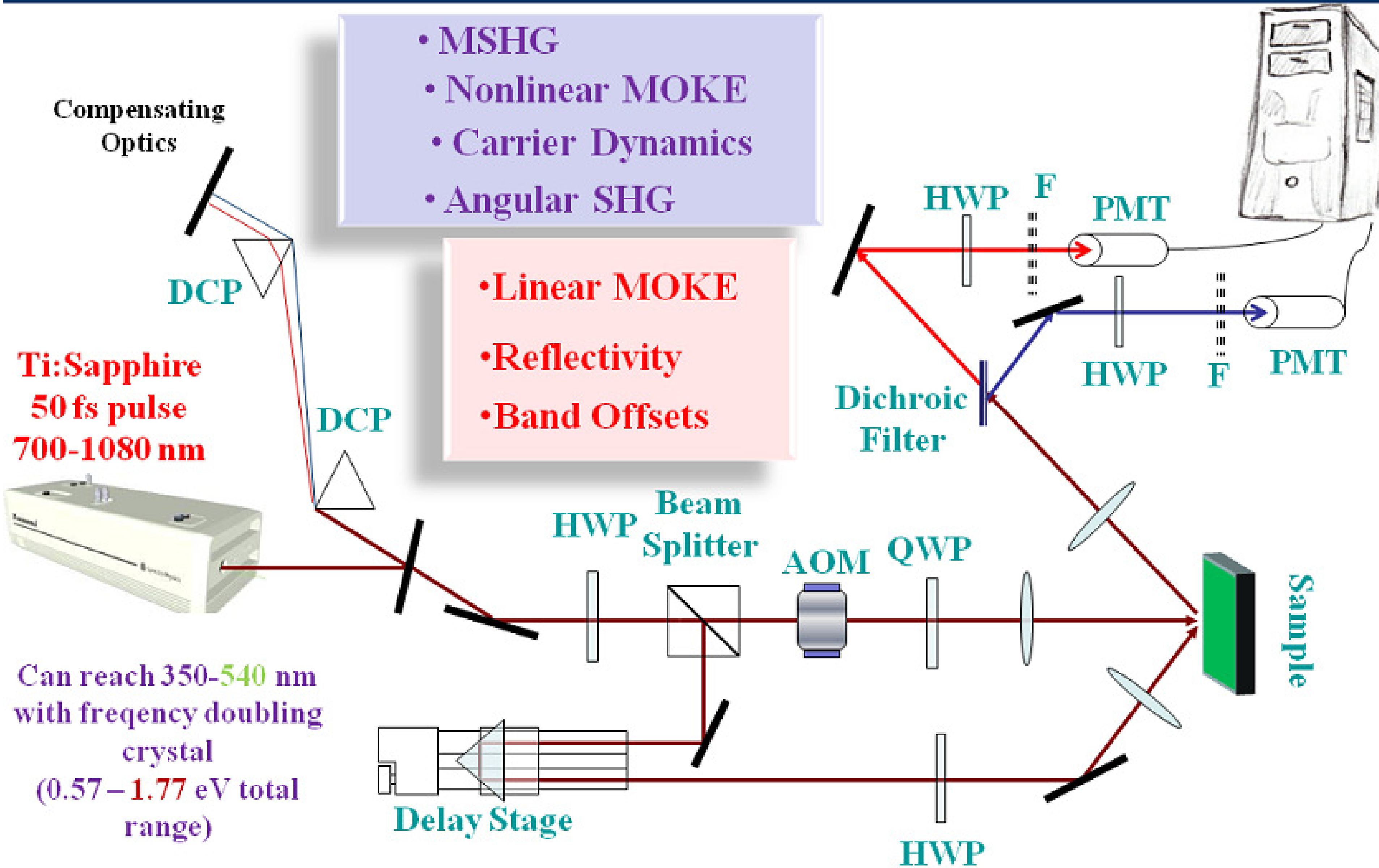
Van der Laan, *et al.*, PRB **34**, 6529 (1986)
Scholl, *et al.*, Science **287**, 1014 (2000)

- Carrier dynamics (i.e. rise times and lifetimes)
- Depth-dependent electric and magnetic fields at the surface, interface and bulk
- Symmetry determination
- Band offset measurements

Optical Techniques

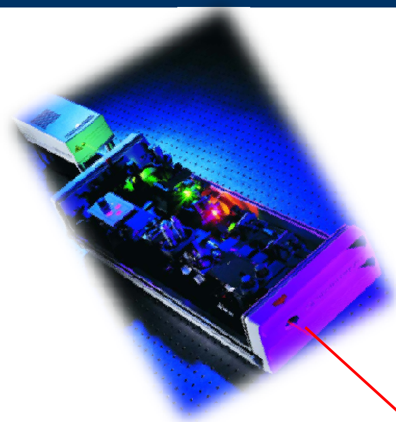
- MSHG
- Nonlinear MOKE
- Carrier Dynamics
- Angular SHG

- Linear MOKE
- Reflectivity
- Band Offsets



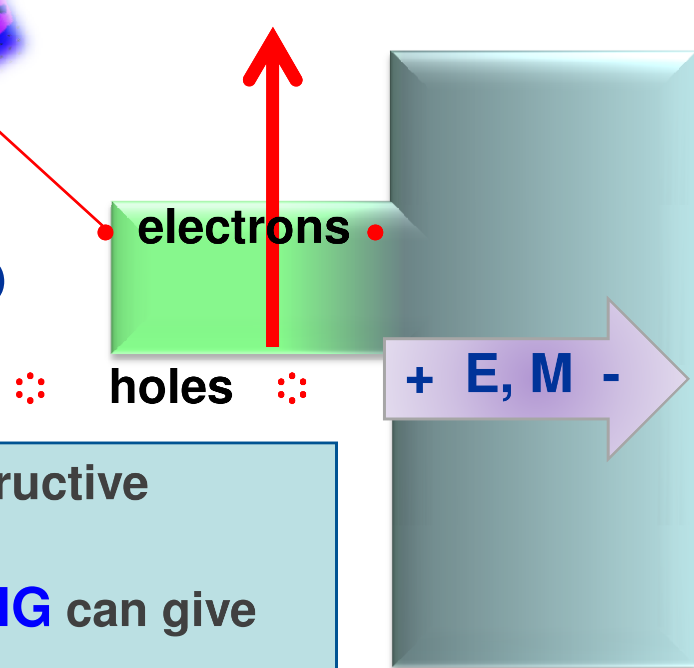
In addition to x-ray absorption spectroscopy at national labs

Carrier Dynamics



Unlike the Kerr effect, SHG is interface sensitive

Band Gap

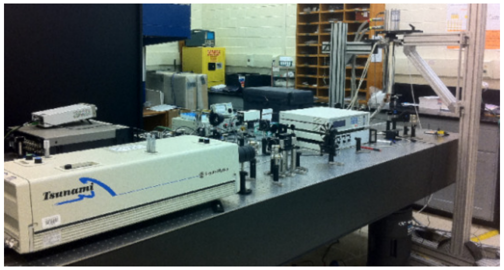


Surface

- Contactless, nondestructive technique.
- **Time-dependent SHG** can give information on:
  - Trap Densities and Lifetimes
  - multiphoton electron-hole injection and dynamics.

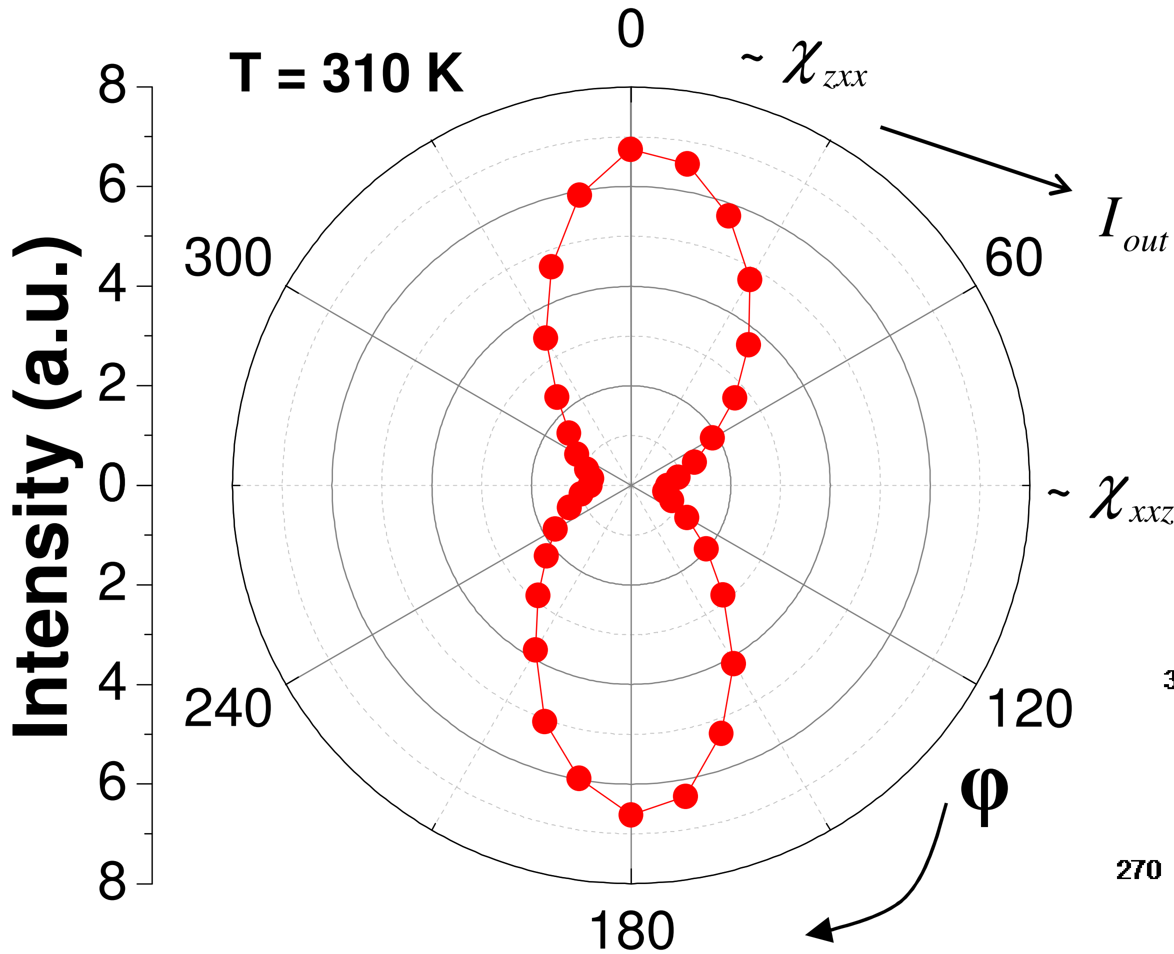
Insulator

•Excited electrons move to surface
•Separation of electrons and holes creates EM field.



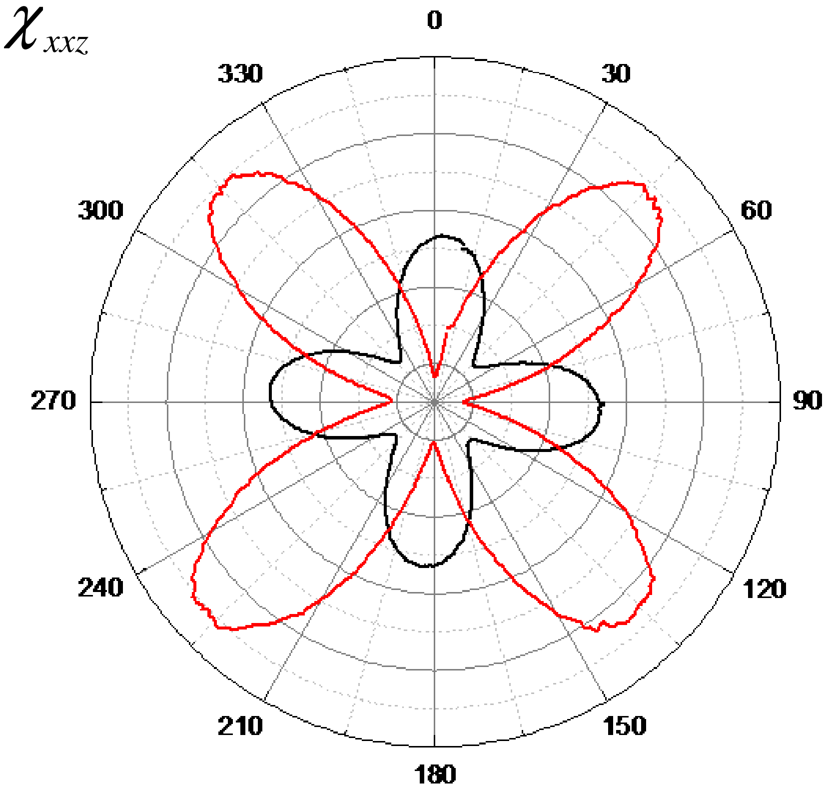
Band Diagram

Angular SHG – $YMnO_3$



$$I_{out}(2\omega) \propto \left|E_{out}(2\omega)\right|^2 \propto \cos^6 \varphi$$

One complete rotation of polarizer/analyzer @ fixed analyzer/polarizer angle



Incident angle ~ 35º with sample normal; Reflection geometry

φ=0 correspond to incident s-polarized and reflected p-polarized light

Interfacial Electric & Magnetic Fields

Nonlinear polarization at the frequency 2ω in the presence of quasi-dc interfacial field

$$P_{\pm}^{NL}(2\omega,t)=[\chi^{(2)}+\chi_e^{(3)}\epsilon(t)\pm\chi_m^{(3)}M(t)][E(\omega)]^2$$

Total Intensity

$$I_{\pm}^{(2\omega)}\propto\left|P_{\pm}^{NL}\right|^2$$

Induced SHG intensity

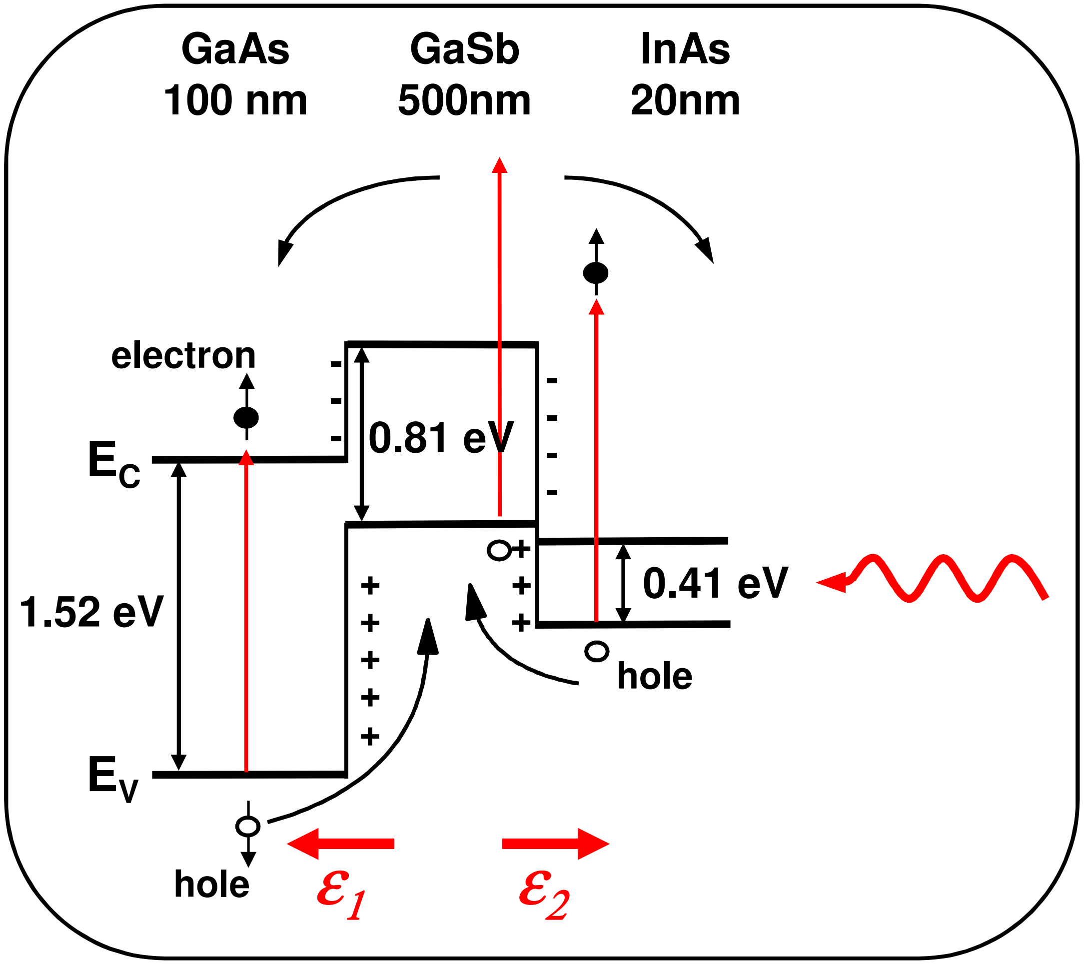
$$\Delta I_{\pm}^{(2\omega)}=I_{\pm}^{(2\omega)}-I_0^{(2\omega)}$$

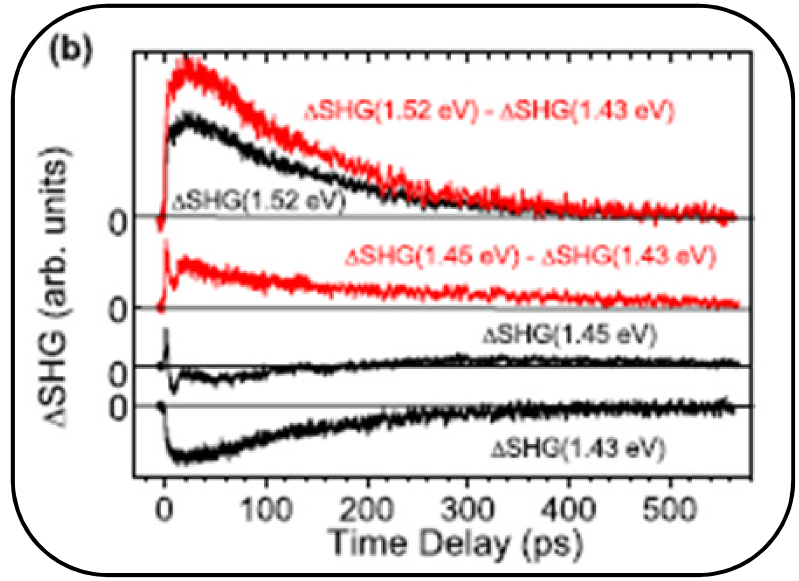
Extract electric- and magnetic-field-induced contributions

$$\begin{cases}\Delta I_{-}^{(2\omega)}+\Delta I_{+}^{(2\omega)}\propto\epsilon(t)\\ \Delta I_{-}^{(2\omega)}-\Delta I_{+}^{(2\omega)}\propto M(t)\end{cases}$$

Time Dynamics

<u>Time Studies to Reveal the Coupling Dynamics</u>. The excitation of an ensemble of spins by a circularly polarized laser light tuned just above the band gap gives rise to a net magnetization at the interface. By varying the time delay, we obtain valuable information about the dynamics of the carriers across interfaces, such as recombination and spin lifetimes. These times are expected to vary between different compositions and over different thicknesses.





Thermalization/cooling of electrons and holes $\tau_{R1} \sim 2\text{ps}$

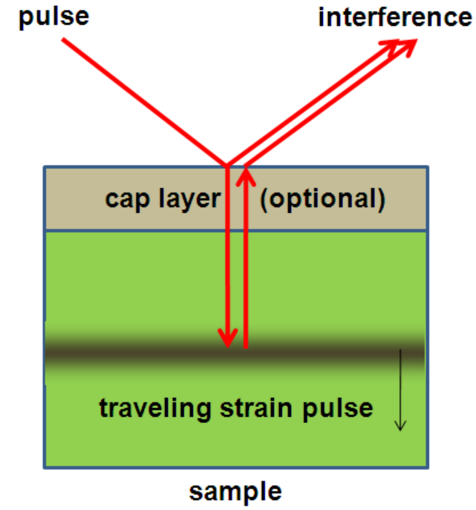
Transport of Spins and Carriers across Heterostructure $\tau_{R2} \sim 15\text{ps}$

Relaxation of the Interfacial Magnetic Fields $\tau_{D} \sim 100\text{ps}$

Dynamic Strain

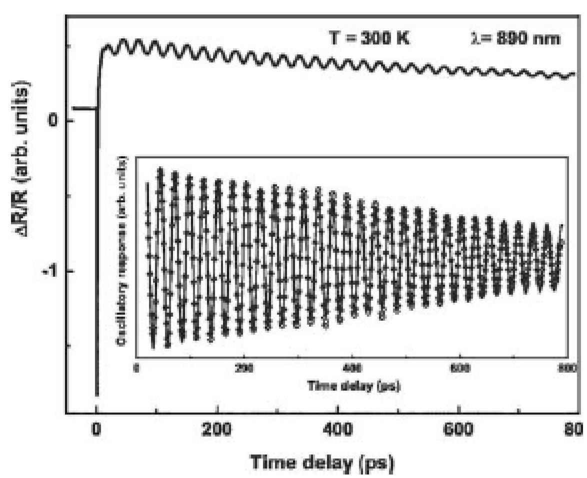
Dynamic Strain Studies to Illuminate Coupling Mechanisms. We should control strain on an ultrashort time scale in order to understand the time dynamics involved. Similar to sound pulses from beating a drum, strain pulses can be generated with intense femtosecond laser pulses.

Reflectivity oscillations originate from the interference of probe beams reflected from the top sample surface and the propagating strain pulse.



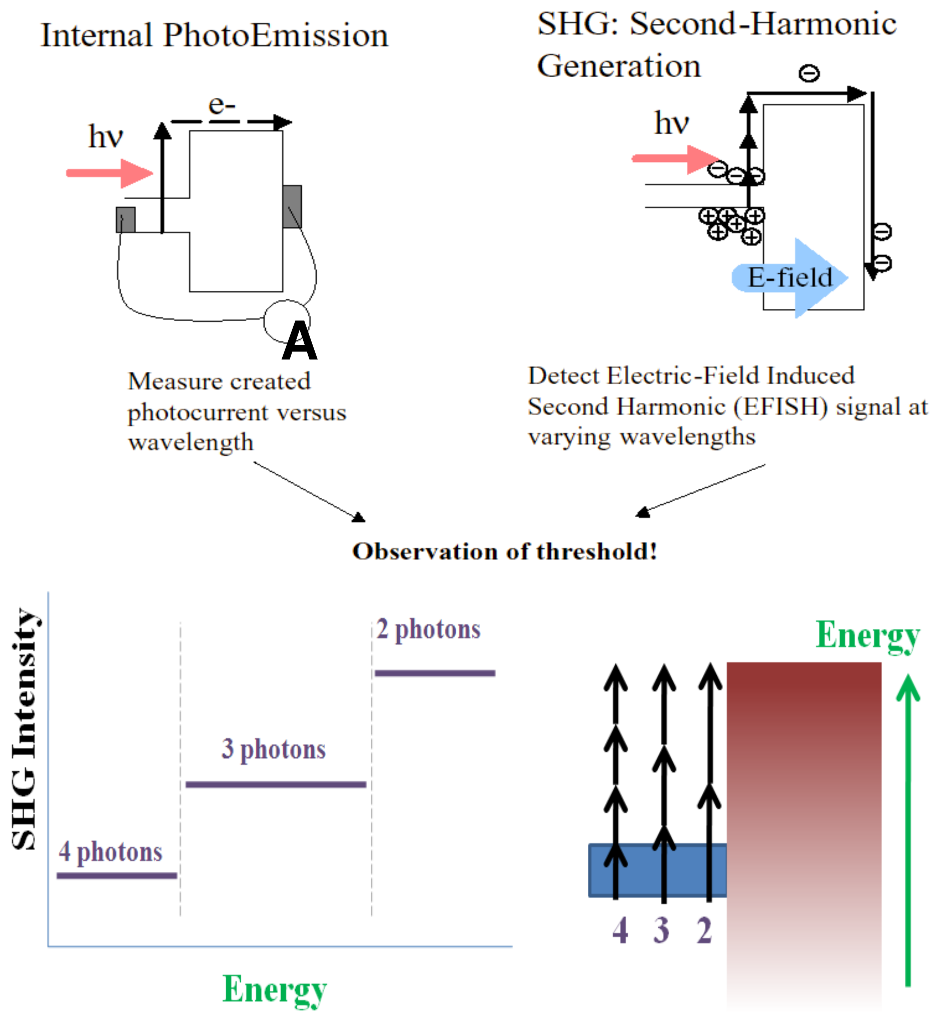
$$\Delta R/R \ \alpha \ Ae^{-t/\tau} \cos(2\pi t/T_p + \varphi),$$

where A is the amplitude, τ is the damping time and T_p is the oscillation period. τ is related to the absorption properties of the material by $\tau = 1/(2\alpha Vs) = \lambda(4\pi V_s\kappa)$, where α is the absorption coefficient and κ is the imaginary part of the complex refractive index N (=n+iκ). The amplitude A is connected to the change in the local complex refractive index through the z strain (η_{33}) by the relation $A \ \alpha \ |\delta N / \delta\eta_{33}|$.



Band Alignment

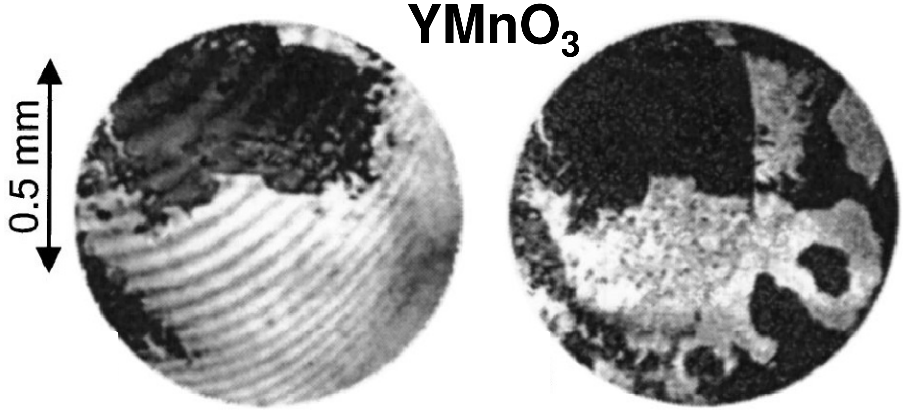
<u>Determine Band Alignment for Promising Magnetoelectric Systems</u>. SHG can also be manipulated to determine band offsets between materials (Jiang *et. al*., 2003).



This band alignment can have a strong effect on the interfacial fields and carrier dynamics. Band offsets can be determined by varying the incident light energy and observing a sharp jump in the SHG intensity. This sharp jump occurs when the number of incident photons required to excite an electron to the bottom of the conduction band of the adjoining material changes.

Future Goal: SHG Imaging

$YMnO_3$



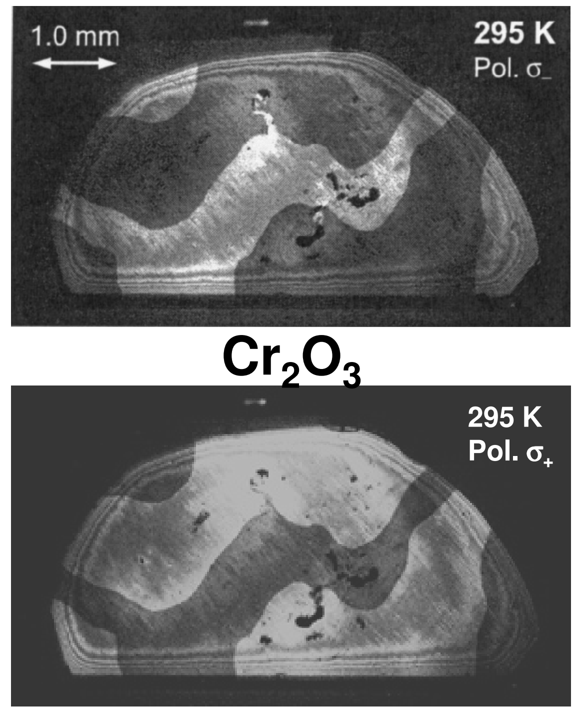
(a) ferroelectric domains and

(b) antiferromagnetic domains

Imaging of both ferroelectric and magnetic domains allows the investigation of multiferroics at various energies

Near field imaging can be purchased for low temperatures and large magnetic fields

Domain imaging using SHG light

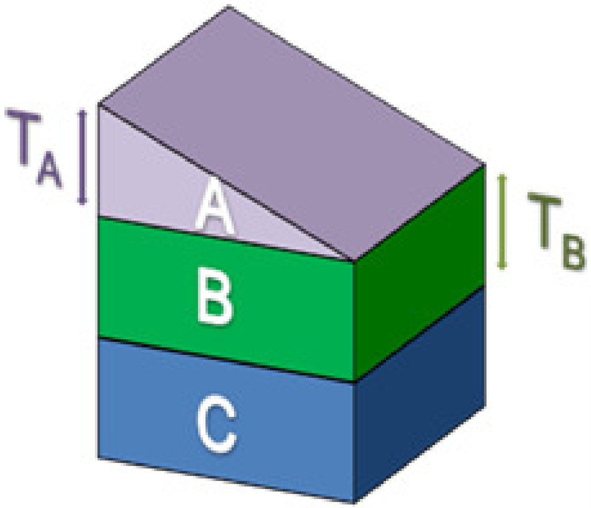


Cr_2O_3

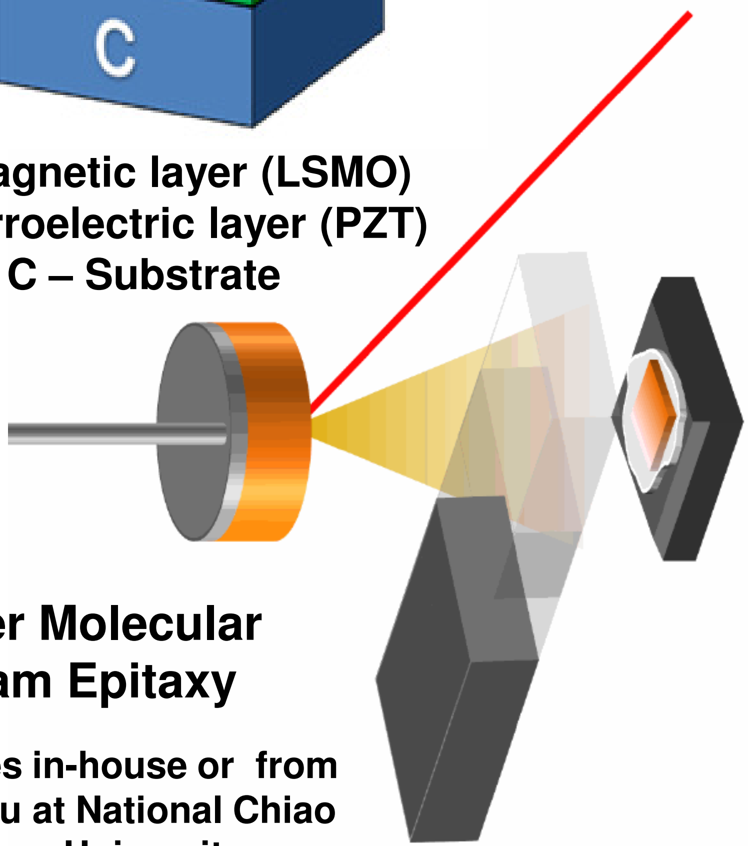
Antiferromagnetic domains

Manfred Fiebig *et al*, J. Opt. Soc. Am. B 22, 2005

Synchrotron Measurements

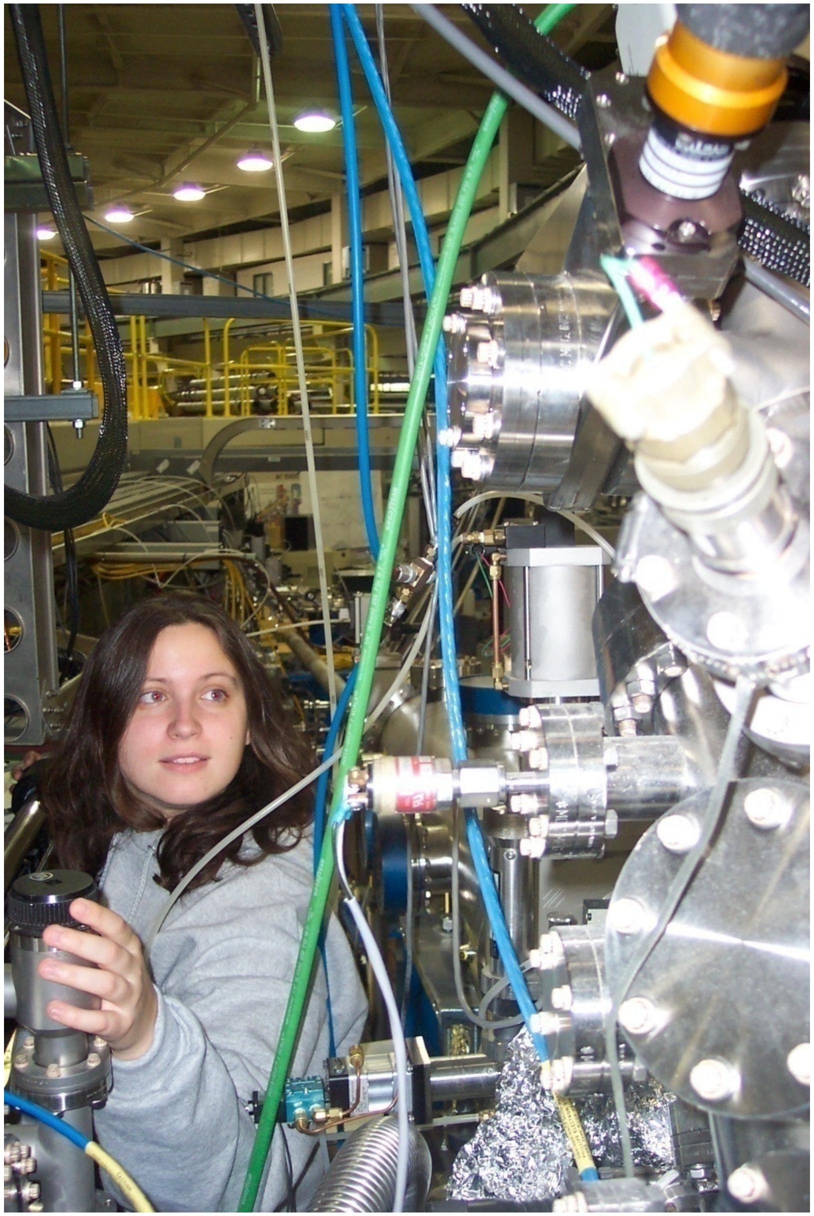


A – Magnetic layer (LSMO)
B – Ferroelectric layer (PZT)
C – Substrate



Laser Molecular Beam Epitaxy

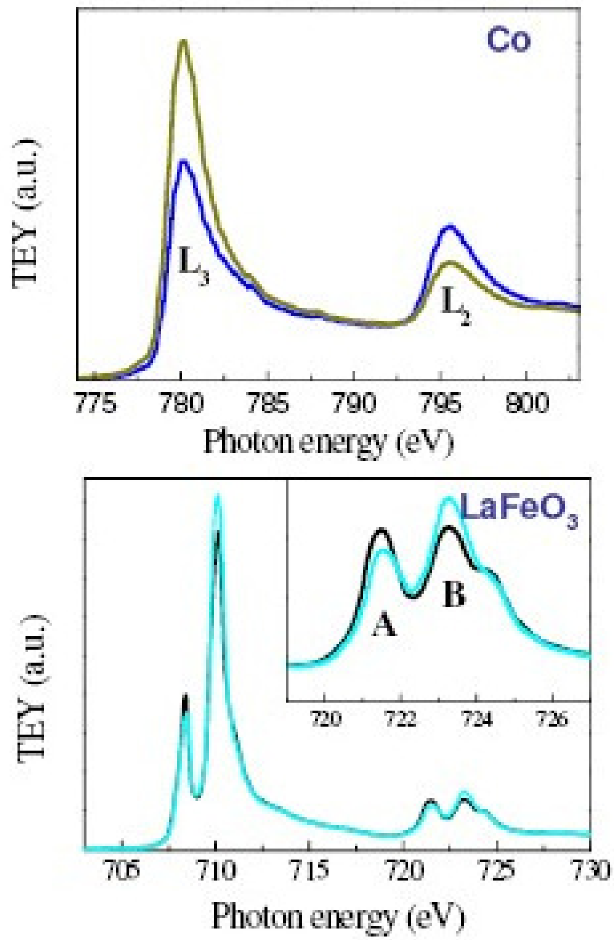
Samples in-house or from Y.H. Chu at National Chiao Tung University

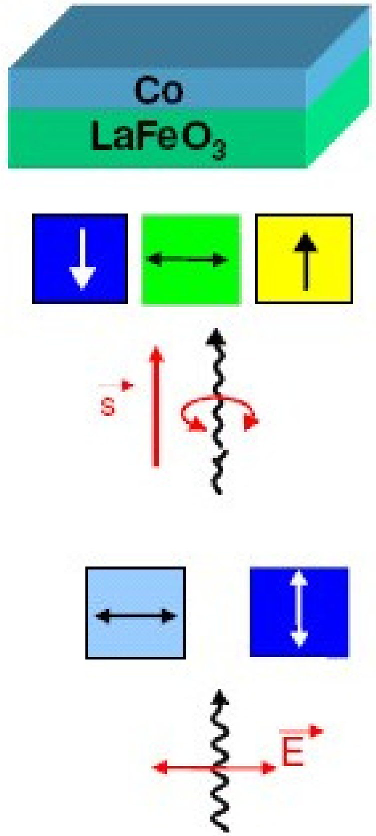


Photoemission Electron Microscopy (PEEM)

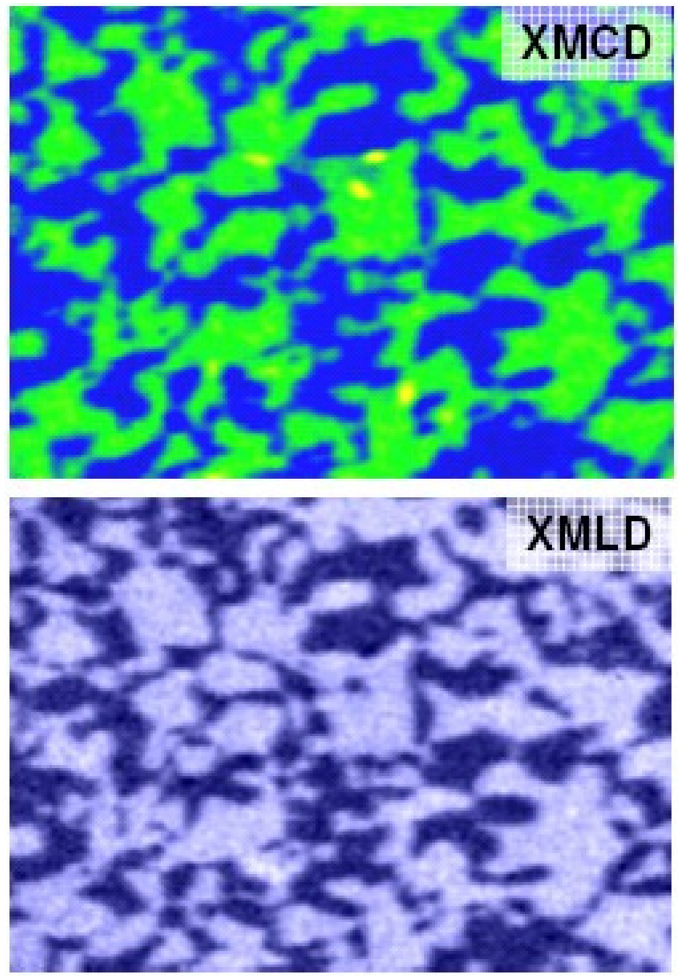
F. Nolting *et al.* *Nature* **405** (2000)

X-ray Absorption Spectroscopy



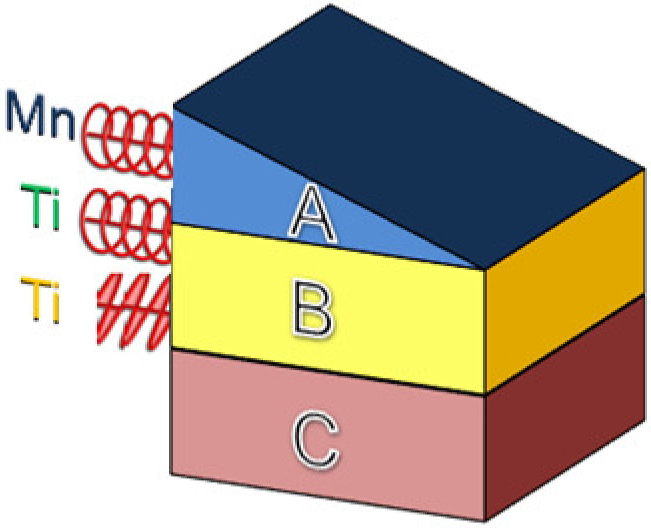


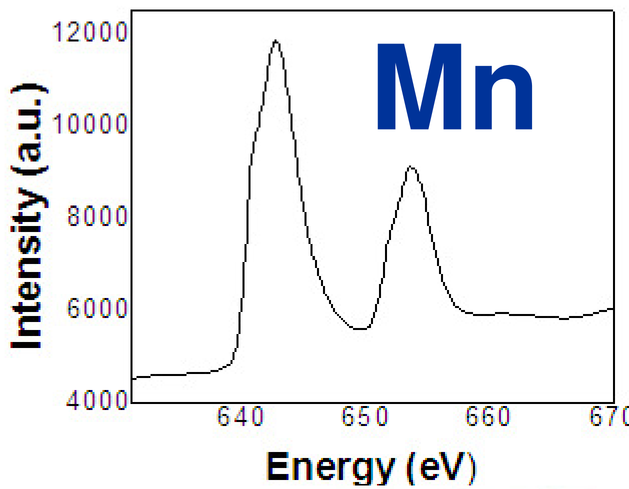
Microscopy

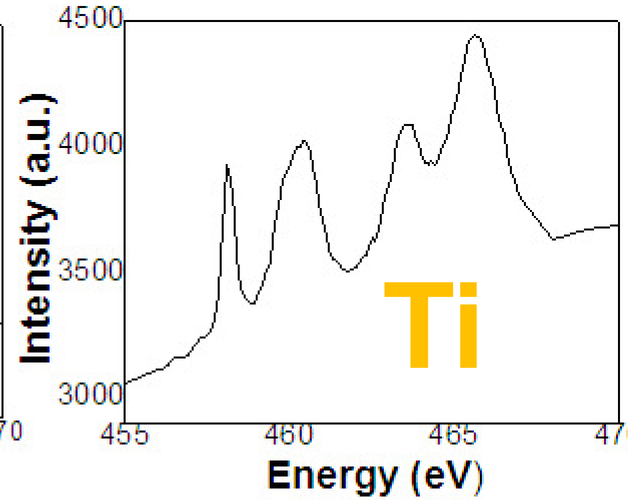


This technique is element specific!
Can we do also see coupling for a magnetic/ferroelectric system?

ME Interface Imaging

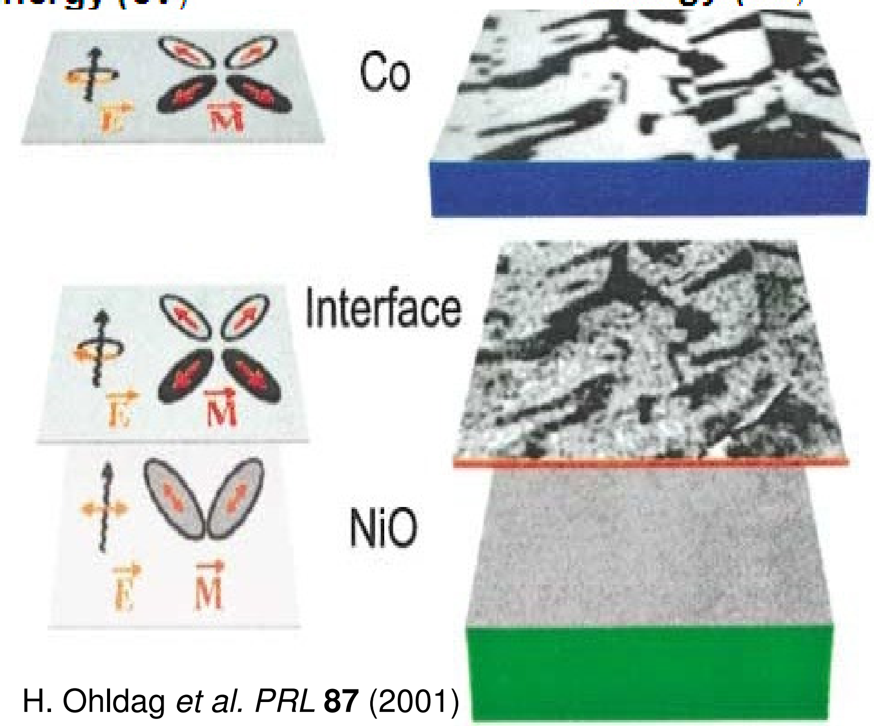






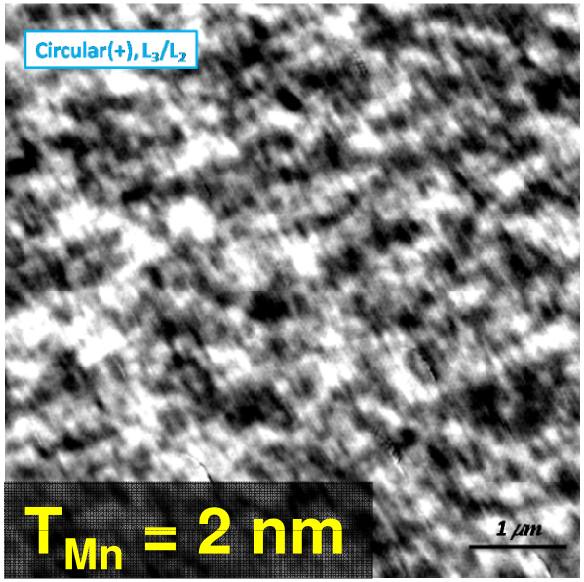
Similar to exchange bias measurements (shown right), we use circularly polarized light on the Ti edge to image any interface magnetism.

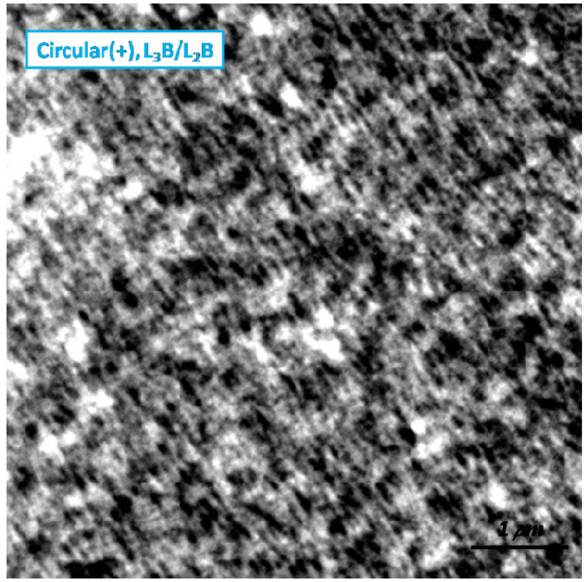
Expect interface image to mimic one—if not both—individual layers.

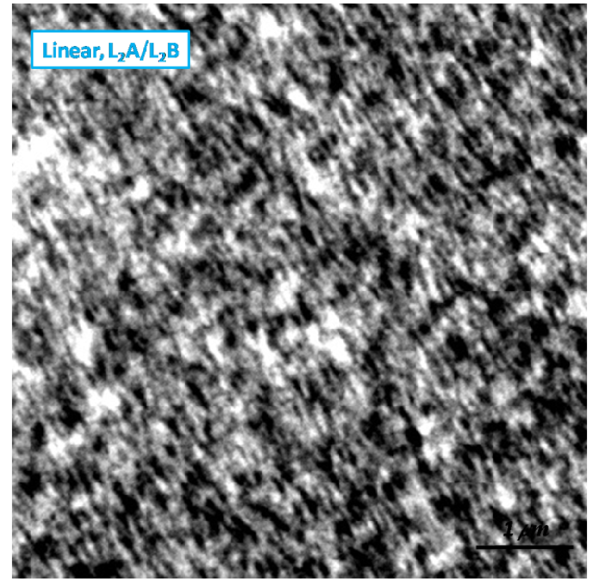


H. Ohldag *et al.* *PRL* **87** (2001)

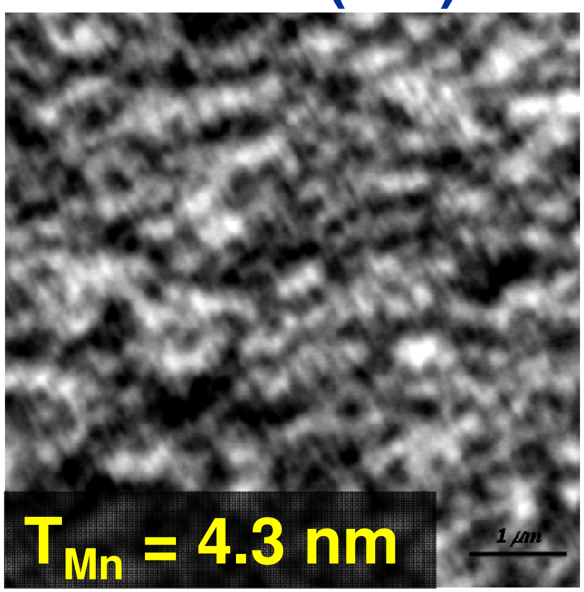
LSMO Thickness Dependence

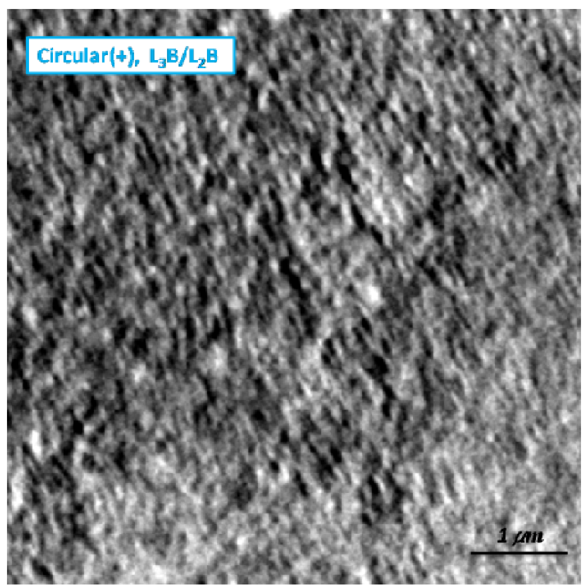


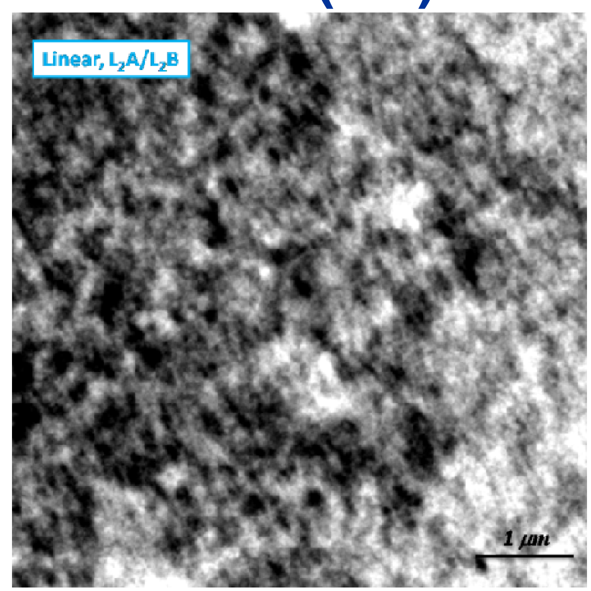




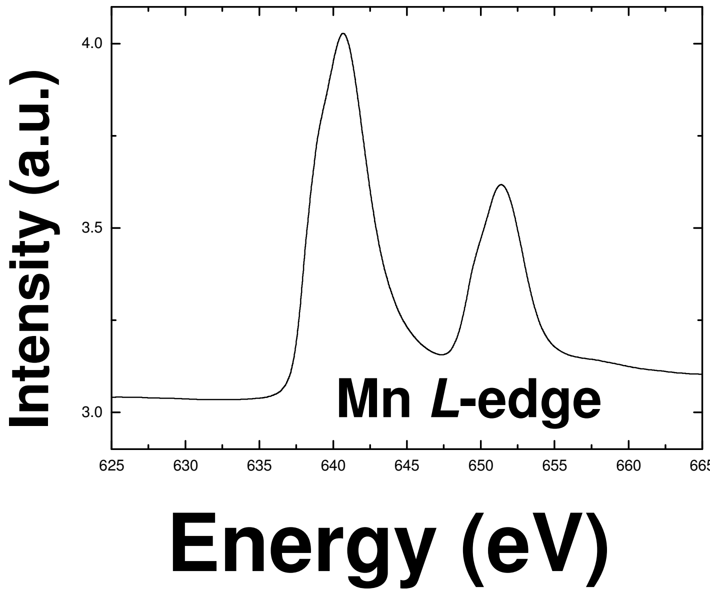
LSMO (FM) **Interface** **PZT (FE)** T=140K



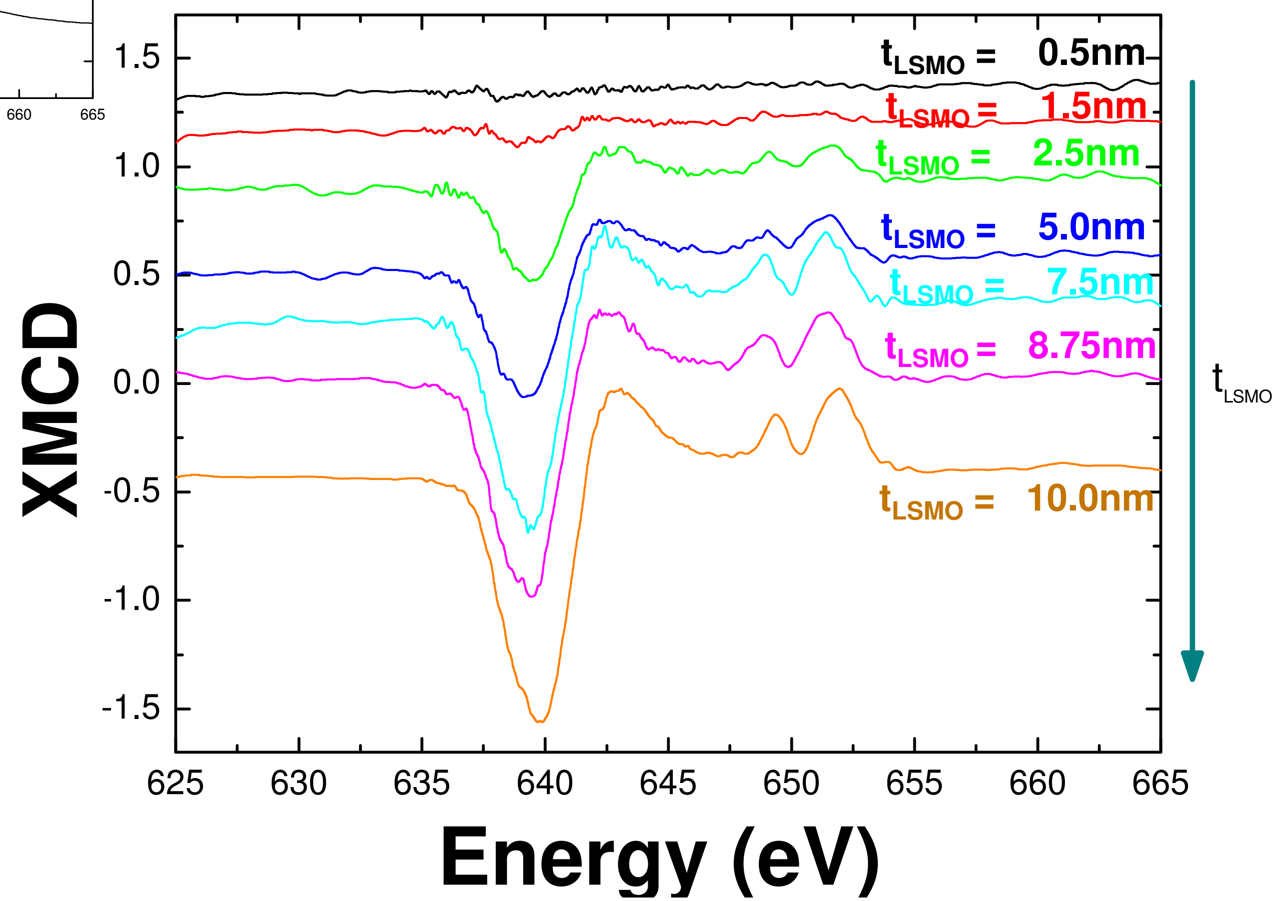




Magnetism of LSMO



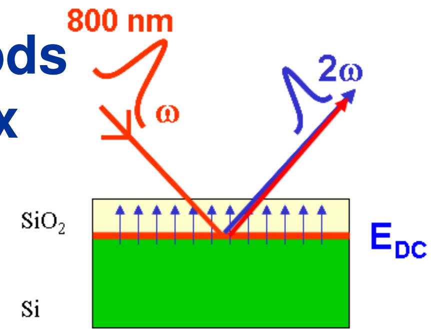
Magnetization gradually decreases as the LSMO thickness decreases.

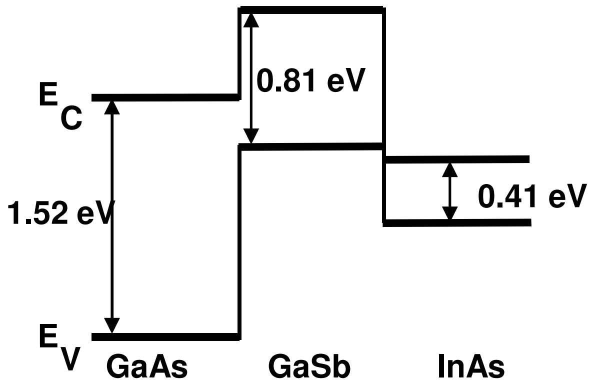


Consistent with LSMO magnetic dead layer around 1.2 nm.

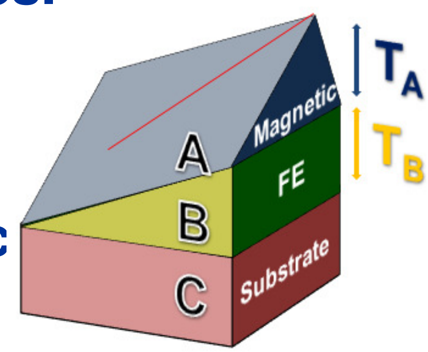
Conclusions

XAS and optics techniques provide many good methods for studying magnets, multiferroics and other complex materials.

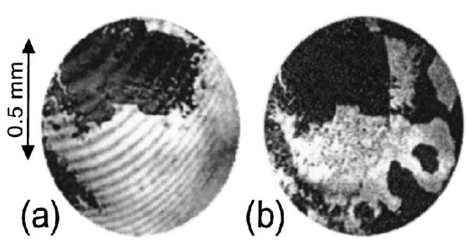




Fields, field dynamics, strains, and band offsets can be obtained at interfaces or through depth-dependent studies.



Wedged, compositionally spread or piezoelectrically strained samples of layered ferroelectric and/or magnetic materials allow characterization of interface coupling.



XAS and imaging studies of the heterostructure interfaces can show coupling between the layers and domains.