Holcomb Group
Capabilities

Synchrotron Radiation & Ultrafast Optics

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"The interface is the device."

- Herbert Kroemer, beginning of his Nobel Lecture in 2000
Complex Oxides

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

Perovskites

• Superconductors (YBa$_2$Cu$_3$O$_7$-$\delta$)
• Ferroelectrics (BaTiO$_3$)
• Colossal Magnetoresistance (((La,Sr)MnO$_3$)
• Multiferroics (BiFeO$_3$)
• High $\varepsilon_r$ Insulators (SrTiO$_3$)
• Low $\varepsilon_r$ Insulators (LaAlO$_3$)
• Conductors (Sr$_2$RuO$_4$)
• Thermoelectrics (doped SrTiO$_3$)
• Ferromagnets (SrRuO$_3$)

All perovskite-related structures $a, b \sim 3.8-4.0$ Å
Ferroelectric Perovskites (e.g. PbZrTiO₃)
- Non-cubic, spontaneous polarization
- “d⁰-ness”
- 6s electrons on A-site

Magnetic Perovskites (e.g. La₀.7 Sr₀.3MnO₃)
- 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

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.

Identify the Major Contributor(s) to and Influencers on Interface Coupling. 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)

- 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

Van der Laan, et al., PRB 34, 6529 (1986)
Gopalan, et al., ARMS 37 (2007)
Optical Techniques

- MSHG
- Nonlinear MOKE
- Carrier Dynamics
- Angular SHG

- Linear MOKE
- Reflectivity
- Band Offsets

Compensating Optics

Ti:Sapphire
50 fs pulse
700-1080 nm

Can reach 350-540 nm
with frequency doubling
(0.57 – 1.77 eV total range)

In addition to x-ray absorption spectroscopy at national labs
Unlike the Kerr effect, SHG is interface sensitive.

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

- Excited electrons move to surface
- Separation of electrons and holes creates EM field.
Angular SHG – YMnO$_3$

$T = 310$ K

$\sim \chi_{zzz}$

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

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

Incident angle $\sim 35^\circ$ with sample normal; Reflection geometry

$\varphi = 0$ correspond to incident s-polarized and reflected p-polarized light
Nonlinear polarization at the frequency $2\omega$ in the presence of quasi-dc interfacial field

$$P^{NL}_{\pm}(2\omega,t) = \left[ \chi^{(2)} + \chi^{(3)}_e \epsilon(t) \pm \chi^{(3)}_m M(t) \right] [E(\omega)]^2$$

Total Intensity

$$I^{(2\omega)}_\pm \propto |P^{NL}_{\pm}|^2$$

Induced SHG intensity

$$\Delta I^{(2\omega)}_\pm = I^{(2\omega)}_\pm - I^{(2\omega)}_0$$

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 Studies to Reveal the Coupling Dynamics. 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.

**Time Dynamics**

- **GaAs** 100 nm
- **GaSb** 500 nm
- **InAs** 20 nm

**Thermalization/cooling of electrons and holes**

\[ \tau_{R1} \approx 2 \text{ ps} \]

**Transport of Spins and Carriers across Heterostructure**

\[ \tau_{R2} \approx 15 \text{ ps} \]

**Relaxation of the Interfacial Magnetic Fields**

\[ \tau_D \approx 100 \text{ ps} \]
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.

$$\Delta R/R \propto A e^{-t/\tau} \cos(2\pi t/T_p + \phi),$$

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

Determine Band Alignment for Promising Magnetolectric Systems. 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

(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

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)

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

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

$T_{Mn} = 2 \text{ nm}$

LSMO (FM)

$T_{Mn} = 4.3 \text{ nm}$

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.
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.