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New Trends in Structural Electronic Characterization of Materials, Interfaces, and Surfaces Using Synchrotron and FEL Based Light Sources Focus Topic
Room On Demand - Session LS-Contributed On Demand

New Trends in Structural Electronic Characterization of Materials, Interfaces, and Surfaces Using Synchrotron and FEL Based Radiation Sources Contributed On Demand Session

LS-Contributed On Demand-1 Resolving Interfacial Electronic Structure in Thin Film Heterostructures using Resonant X-ray Reflectometry*, Ryan Need,* University of Florida

The interfaces between electronic materials are critically important for the operation of modern electronic devices and the pursuit of new knowledge in solid state physics. Today, the interest in electronic interfaces is fueled by emerging technologies like quantum computing, spintronics, and energy storage, along with fundamental questions about how tuning symmetry and band topology at interfaces can modify functional properties. Answering these questions requires the development of characterization probes that have high spatial and spectral resolution, a wide range of sample environments, and are compatible with in-situ or in-operando experiments.

In this talk, we argue that resonant X-ray reflectometry (RXR), which combines the element-specificity and local structure sensitivity of X-ray absorption spectroscopy with the Angstrom depth-resolution of reflectometry, is the probe best suited to this challenge. As an example, we show how RXR can be used to extract electronic structure depth profiles in complex oxide superlattices and visualize interfacial charge transfer with atomic layer resolution. Specifically, we present measurements collected from $SmTiO_3$(SmTO)/$SrTiO_3$ (STO) superlattices with STO quantum wells varying in thickness from five SrO planes to a single SrO plane. At the polar-nonpolar SmTO/STO interface, a known electrostatic discontinuity leads to approximately half an electron per areal unit cell transferred from the interfacial SmO layer into the neighboring STO quantum well. We observe this charge transfer as a suppression of the Ti t_{2g} absorption peaks that minimizes contrast with the neighboring SmTO layers at those energies and leads to a pronounced absence of superlattice peaks in the reflectivity data. Critically, our results demonstrate the sensitivity of RXR to electronic reconstruction in the monolayer limit and prove RXR to be a powerful means of characterizing interfacial electronic structure in quantum materials.

LS-Contributed On Demand-4 HAXPES Study of Surface/Interface Effects Induced by Heavy Alkali Post Deposition Treatment of (Ag,Cu)(In,Ga)se2 Thin Film Solar Cell Absorbers*, Natalia Martin,* Uppsala University, Sweden*; T. Törndahl,* Uppsala University , Sweden*; K. Simonov,* Department of Materials and Process Development Swerim AB, Sweden*; H. Rensmo, C. Platzer-Björkman,* Uppsala University, Sweden

Solar cells based on Cu(In,Ga)Se_2 (CIGS) thin film absorbers have shown high and stable efficiencies for both laboratory cells and industrial modules with recent record cell efficiencies of 23.4% [1]. In order to reach closer to the theoretical maximum efficiency of around 30%, further reductions of optical and electrical losses are needed. Some recent works show that silver (Ag) alloying in CIGS to form (Ag,Cu)(In,Ga)Se_2 (ACIGS) leads to higher device efficiencies as compared to similar CIGS devices without Ag [2]. More, a postdeposition treatment (PDT) based on the alkali metal fluorides (i.e., KF, RbF or CsF) [3] after absorber formation has been studied recently and is known to improve the efficiency in CIGS solar cells, mainly by an increase in the open circuit voltage, V_{OC}. The exact role of the silver and/or alkali elements is intensively discussed, but it is clear that interface and grain boundary effects are important in (A)CIGS solar cells. It is likely that a redistribution of the absorber elements near the surface region may occur during the alkali PDT and some works show interface formation in CIGS solar cells subjected to a metal fluoride post deposition treatment [4].

In this work we employed hard X-ray photoelectron spectroscopy (HAXPES) to study the interface between CdS buffers and PDT-ACIGS absorbers, which have been exposed to different metal fluoride PDT. Two different alkali PDT are compared, RbF and CsF, to investigate how the choice of the metal fluoride treatment affects the buffer/absorber interface. In particular, the chemical and electronic properties of CdS/ACIGS interfaces in terms of intermixing, energy band alignment and composition for a set of device relevant samples have been studied.

The results show that the electronic structure at the CdS/ACIGS interface does not change for the different PDTs. However, the ACIGS composition at the near surface region seems to change depending on the applied PDT process. Possible reasons for this will be discussed. The results give insights into how selective alkali PDT could change the ACIGS surface composition, which may influence the solar cell behavior.

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LS-Contributed On Demand-7 Surface Action Spectroscopy With Inert Gas Messenger Atoms*, Hans-Joachim Freund,* Fritz Haber Institute of the Max Planck Society, Germany

Action spectroscopy is well established in the field of cluster physics and chemistry, where sample densities are very low. The action spectroscopy approach is based on the idea, that messenger species are attached to the object under investigation, and that laser light of varying frequency in the infrared is used to detach the messengers, which are detected with a mass-spectrometer. By varying the infrared frequency, one is able to record the vibrational spectrum of the object under study without having to record a reference spectrum. It has not been applied to surfaces due to the need for high intensity frequency variable laser sources. On the other hand, desorption induced by electronic transition as well as photo-desorption are well established fields surface science. We have over the last few years developed surface action spectroscopy employing the Free Electron Laser installed at the Fritz-Haber Institute. In the presentation I report on the conceptual design and some first results on surface vibrational spectroscopy using the approach (1-3), as well as the pros and cons of the techniques in comparison to other vibrational spectroscopy techniques.

References:

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LS-Contributed On Demand-13 X-ray Magnetic Linear Dichroism Studies of Electrical Switching of Antiferromagnetic Order in α-Fe2O3 Epitaxial Films*, Egecan Cogulu,* *N. Statuto,* New York University*; Y. Cheng,* Department of Physics, Ohio State University*; S. Yu, F. Yang,* Ohio State University*; R. Chopdekar, H. Ohldag,* Advanced Light Source, Lawrence Berkeley National Laboratories*; A. Kent,* New York University

Recently manipulation of antiferromagnetic (AFM) order has been gaining the attention of the spintronics community. Magnetic switching has been reported in AFM thin films based on electronic transport methods, which provide only spatially averaged information on AFM states. Other techniques, such as x-ray magnetic linear dichroism (XMLD), can provide local information on AFM domains. In this study, we report direct observation of spin reorientation in response to current pulses in (0001) α-Fe_2O_3/Pt heterostructures. Our experiment combines the application of current pulses and spatially resolved photoemission electron microscopy (PEEM) with X-ray magnetic linear dichroism to detect antiferromagnetic contrast. Pulses were applied in two different configurations A and B (Fig. 1a), and XMLD images were interleaved with current pulse sequences to reveal changes in the AFM domains (Fig. 1c). Our analysis shows that electrical pulses do not only reorient the Néel vector within the easy plane as previous work indicates. Instead, we conclude that most of the change current induced change associated with the Néel vector is a rotation out of the film plane, and different pulse directions can bring the AFM order in and out of plane deterministically.

This work is supported in part by Air Force Office of Scientific Research under Grant FA9550- 19-1-0307.

LS-Contributed On Demand-16 Probing Interfacial Ferromagnetism in Oxide Superlattices Using Depth Resolved X-Ray Spectroscopic and Scattering Techniques, ***Jay Paudel,*** Temple University*; M. Terilli,* Rutgers University*; I. Vobornik, P. Orgiani, G. Panaccione,* CNR-IOM, TASC Laboratory, Italy*; C. Klewe, P. Shafer,* Advanced Light Source, LBNL*; V. Strocov,* Swiss Light Source, PSI, Switzerland*; J. Chakhalian,* Rutgers University*; A. Gray,* Temple University

The origins of emergent interfacial ferromagnetism in $CaMnO_3$/$CaRuO_3$ and $CaMnO_3$/$LaNiO_3$ heterostructures have been investigated for nearly two decades [1-4]. Several experimental and theoretical studies suggest that this phenomenon is stabilized by charge transfer across the interface and suggest different thicknesses of the resultant ferromagnetic layers. In this comprehensive study, we have carried out polarization-dependent x-ray absorption spectroscopy and x-ray resonant magnetic scattering to probe the depth-resolved magnetic profile at the interfaces in these material systems. Furthermore, we have utilized bulk-sensitive hard x-ray photoemission spectroscopy and soft x-ray standing-wave photoemission spectroscopy in both first- and second-order Bragg geometries to probe the depth-dependent chemical profile and interfacial charge reconstruction in $CaMnO_3$. Finally, high-resolution angle-resolved photoemission spectroscopy with in-situ pulsed-laser deposition synthesis were used to track the topology of the Fermi surface of $LaNiO_3$ in the superlattice across the thickness-dependent metal-insulator transition. The results were compared to the first-principles theoretical calculations.

This work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences, and Engineering Division under Award DE-SC0019297

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New Trends in Structural Electronic Characterization of Materials, Interfaces, and Surfaces Using Synchrotron and FEL Based Radiation Sources Invited On Demand Session

LS-Invited On Demand-1 Soft X-ray Resonant Inelastic Scattering (RIXS) to Study the Magnetic and Electronic Properties of Materials, ***Nicholas Brookes,*** ESRF, France **INVITED**

Soft x-ray resonant inelastic x-ray scattering (RIXS) spectroscopy has made many advances in the past few years mainly due to improved experimental facilities at third generation synchrotron sources [1].

In this talk the technique will be introduced and the current possibilities offered by soft x-ray RIXS will be demonstrated using results from the ESRF. In particular, some of the work on transition metal oxides like the nickelates [2,3], the cuprates [4] and iron [5] will be highlighted.

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LS-Invited On Demand-7 Bulk and Interface Hard-X-ray Bandmapping with Spin Resolution Combining Full-field Momentum Imaging with ToF-recording, ***Gerd Schönhense,*** Johannes Gutenberg University of Mainz, Germany **INVITED**

Photoelectron momentum microscopy (MM) is an alternative approach for ARPES, combining full-field k-space imaging with hemispherical or time-of-flight (ToF) energy analyzers. The heart of ToF-MM is a fast delay-line detector (DLD), recording position and time t of each counting event (at rates up to ~$5x10^6$ cps). 3D (k_x,k_y,t)-recording is advantageous in low-intensity experiments with soft or hard X-rays, spin mapping, or time-resolved ARPES at FEL sources. This contribution gives an overview with examples and an outlook on ongoing developments.

MM with X-ray excitation gives access to 4D bulk spectral functions $\rho(E_B,\mathbf{k})$, yielding bands, Fermi surfaces and -velocity distributions [1]. The band structure is captured in a tomographic-like mode via direct transitions to a final-state sphere, whose k-radius is tuned via the photon energy. PETRA III (Hamburg) provides 50ps photon pulses with 192ns period (40-bunch mode) at beamlines P04 (hv=300-1700eV; resolving power >10^4) and P22 (2300-7500eV; r. p. up to 10^5 with Si(333) monochromator). Information depths of 10-20nm allow studying buried structures like a 2D e-gas at an inner interface or bulk bands of a Heusler compound capped with 2nm MgO or 1nm Au.

Imaging spin filters are powerful tools complementing MM [2]; the effective figure-of-merit increases when exploiting ToF as third "coordinate" [3]. Using circ.-pol. soft X-rays, we uncovered a relation between the *Fano* spin component (along the photon helicity), the perpendicular spin component (oriented like spin in *Mott* scattering) and the circular dichroism [4]. Hard X-ray spin-MM allowed quantifying the spin gap in the half-metallic Heusler ferromagnet Co_2MnSi and the degree of (bulk) spin polarization close to E_F in magnetite, being controversially discussed for decades.

First experiments at FLASH (DESY, Hamburg) suggest a large potential of ToF-MM for fs pump-probe photoemission [5]. Event coordinates (E_B,**k**), arrival time and intensity of the corresponding FEL pulse, delay and parameters of the pump pulse are streamed in real-time and sorted into a multidimensional histogram memory. Current technical improvements concern a new objective lens without extractor field, correction and suppression of space-charge shifts, and a novel *dispersive-plus-ToF* hybrid MM. A drift section and fast DLD (<80ps time res.) behind a large single hemisphere will facilitate ToF at synchrotrons with 500MHz multibunch filling.

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LS-Invited On Demand-13 My Adventures with Synchrotrons: From Discovering New Types of Magnetism to Helping NASA, ***Mikel Holcomb,*** West Virginia University, USA **INVITED**

In many areas of science and the world, competition is seen as an opportunity to obtain improved performance or results. Utilizing many techniques (bulk magnetometry, neutron reflectometry and resonant x-ray magnetic scattering), we have discovered and explored the existence of competing magnetic phases in many single layer thin films that results in giant negative magnetization. We have focused on the system of complex oxide $La_{0.7}Sr_{0.3}MnO_3$. While transmission electron microscopy images show pristine epitaxial growth, the data supports that there are regions of different magnetic order. This results in interesting magnetic measurements, that share similarities with ferrimagnets with competing magnetic lattices. This competition results in spontaneous negative magnetization that aligns counter to a small applied magnetic field and inverted hysteresis loops near room temperature. This behavior has much in common with superparamagnetic nanoparticles. In this talk, the time, field and temperature dependence of these samples will be discussed to help understand this phenomenon. The switch from negative to positive magnetization effectively doubles the change in magnetization, important for some types of devices. We acknowledge funding support from NSF (DMR-1608656) for growth and optimization and DOE (DE-SC0016176) for the magnetic characterization of our films.

LS-Invited On Demand-19 Extending Time-Resolved X-Ray Diffraction using Coherence, ***Mark Sutton,*** McGill University, Canada **INVITED**

The use of x-ray diffraction for in-situ or in-operando measurements is now relatively common. It generally combines the power of x-ray structural measurements with x-ray's penetrating abilities to be able to perform other experimental techniques in combination. This allows one to observe how the atomic structure is or is not coupled to other material properties or underlies the processing of the material. Small angle x-ray scattering is often used, but when there is predominately crystalline order, wide angle x-ray scattering provides more detailed information. Often a trade off between better time-resolution and lower structural resolution is made to study the fastest processes. Over the last several years coherence has begun to play an important role in x-ray diffraction. Techniques like x-ray correlation spectroscopy (XPCS) and coherent diffraction imaging (CDI)

being primary examples and the trade-off between the two techniques is similar to the trade-off between higher time resolution and higher structural resolution. In this talk I will explore information that is available from speckle patterns that is beyond conventional XPCS but is not as complete as CDI.

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