## Novel Trends in Synchrotron and FEL-Based Analysis Focus Topic

## Room 103C - Session SA+AS-MoA

## Frontiers of Photoemission with Synchrotron and XFEL Radiation/Advances in High-resolution Imaging Techniques

**Moderators:** Maya Kiskinova, Elettra-Sincrotrone Trieste, Italy, Olivier Renault, CEA-University Grenoble Alps, France

1:40pm **SA+AS-MoA1 Photoemission with Soft and Hard X-Rays: Past, Present, and Future***, **Charles Fadley,*** University of California, Davis **INVITED**

In this talk, I will begin by briefly reviewing some of the key early developments in soft x-ray photoelectron spectroscopy (XPS), angle-resolved XPS (ARXPS), x-ray photoelectron diffraction (XPD), and soft x-ray angle-resolved photoemission (ARPES). I will then consider combining these well-established methods with more recent techniques involving the tailoring of the x-ray wavefield through standing-wave (SW) excitation or total-reflection (TRXPS) to provide enhanced depth resolution and the use of hard x-ray excitation in the multi-keV regime (HXPS, HAXPES) to study bulk materials and buried layers and interfaces [1-5]. Applications to semiconductor- [1], oxide- [2.4,5], and magnetic- [2] heterostructures, as well as liquid/solid interfaces [3] will be considered. Future possibilities combining these approaches with variable polarization, as well as spin-, space-, and time- resolution will also be discussed.

**Acknowledgements:**

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**References:**

[1] "Bulk Electronic Structure of the Dilute Near-Ferromagnetic Semiconductor $Ga_{1-x}Mn_xAs$ via Hard X-Ray Angle-Resolved Photoemission" A. X. Gray, J. Minar, S. Ueda, P. R. Stone, Y. Yamashita, J. Fujii, J. Braun, L. Plucinski, C. M. Schneider, G. Panaccione, H. Ebert, O. D. Dubon, K. Kobayashi, and C. S. Fadley, Nature Materials 11, 957 (2012).

[2] "Interface properties of magnetic tunnel junction $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3$ superlattices studied by standing-wave excited photoemission spectroscopy", A. X. Gray et al., Phys. Rev. B 82, 205116 (2010);"Momentum-resolved electronic structure at a buried interface from soft X-ray standing-wave angle-resolved photoemission",A.X. Gray et al., Europhysics Letters 104, 17004 (2013).

[3] "Chemical-state resolved concentration profiles with sub-nm accuracy at solid/gas and solid/liquid interfaces using standing-wave ambient-pressure photoemission (SWAPPS)", S. Nemsak et al., Nature Communications 5, 5441 (2014).

[4] "Depth profiling charge accumulation from a ferroelectric into a doped Mott insulator",M. Marinova, J. E. Rault, et al., Nano Letters 15, 2533−2541 (2015).

[5] "Energetic, spatial and momentum character of a two-dimensional electron gas at the buried interface between $GdTiO_3$ and $SrTiO_3$", S. Nemšák et al., Phys. Rev. B, to appear, http://arxiv.org/abs/1508.01832 .

2:20pm **SA+AS-MoA3 Honorary Session for Prof. Charles Fadley***, O.J. Renault,* CEA-University Grenoble Alps, France*; **Julien Rault,*** Synchrotron SOLEIL, France

This contribution is intended to honor Prof. Charles Fadely for his unvaluable work in the field of photoemission over the past 50 years and will celebrate his 75th birthday. It will immediately follow his invited talk and will take the form of 3 short talks given by some of his former students.

2:40pm **SA+AS-MoA4 Ultrafast Magnetization Relaxation Dynamics in $La_{0.66}Sr_{0.33}MnO_3$ Films***, **Tommaso Pincelli,*** Università di Milano, Italy*; A.Yu. Petrov, G. Panaccione,* Laboratorio TASC, IOM-CNR, Italy*; M. Oura,* RIKEN SPring-8, Japan*; T.L. Lee,* Diamond Light Source Ltd., UK*; G. Rossi,* Università di Milano, Italy

Hole-doped rare-earth manganites, like $La_{0.66}Sr_{0.33}MnO_3$ (LSMO), display exotic phenomena such as concurrent colossal magnetoresistance and half-metallicity which originate from the interplay of charge, spin, and orbital degrees of freedom [1]. The peculiar transport properties of LSMO thin films combined with the ferromagnetic order that persists up to about 350 K [2] render such system a most technologically attractive material for spin injection: the spin polarization at the Fermi level reaches about 100% for $T<T_{Curie}$ [3].

The ultrafast manipulation of spin states in LSMO can be tested by state-of-the-art time-resolved pump-probe techniques. Previous studies by optical pump-probe spectroscopy have given evidence of photoinduced effects in ferromagnetic manganites [4].

Photo-Electron Spectroscopy (PES) allows a direct measurement of the electronic structure; time-resolved PES is able to disentangle the delicate out-of-equilibrium interplay between electronic, spin and lattice degrees of freedom [5], an essential feature in the case of highly correlated materials. HArd X-ray PhotoElectron Spectroscopy (HAXPES) extends the probing depth of PES to the bulk of the solid (tens of nm), and therefore does not suffer of the modification induced by the surface.

We present here a pump-probe HAXPES study of the relaxation dynamics of LSMO thin films. We study the structure of the Mn 2p core level and, in particular, the bulk-only screening channel proportional to the metallic and ferromagnetic state in LSMO. We observe a large and 'slow' reduced lineshape change up to 200 picoseconds after the IR pumping. By comparison with all-optical techniques (Time-Resolved Magneto-Optical Kerr effect, TR-MOKE) we are able to attribute the observed quenching to a collapse of magnetic order. The sudden demagnetization reduces the mobility of electrons in the solid, inducing a localization similar to a metal-insulator transition.

Since LSMO is half-metallic, the direct exchange of energy between the optically excited electrons and the magnetic order is inhibited by the absence of final states for spin-flip scattering [3]. So we can follow the relaxation dynamics as the energy is first dissipated in the lattice and then in a reduction of the magnetic order.

**References**

[1] Y. Tokura et al. J. Magn. & Magn. Mater. 200, 1 (1999).

[2] K. Horiba et al. Phys. Rev. B 71, 155420 (2005).

[3] G.M. Müller et al. Nat. Mat. 8, 56 (2009); J.-H. Park, et al. Nature, 392, 794 (1998).

[4] A.I. Lobad et al. Appl. Phys. Lett. 77, 4025 (2000); K. Matsuda et al. Phys. Rev. B 58, 4203 (1998).

[5] B. Frietsch et. al. Nature Communications, 6, 8262 (2015).

3:00pm **SA+AS-MoA5 Inelastic Background Analysis of Haxpes Spectra for Device Technology: A Non-Destructive Tool for Accessing Deeply Buried Interfaces***, **Charlotte Zborowski,*** *O.J. Renault, E. Martinez, A. Torres,* CEA, LETI, MINATEC Campus, France*; Y. Yamashita,* NIMS, Japan*; G. Grenet,* Inl, Ecl, France*; S. Tougaard,* SDU, Denmark

Recently, the advent of Hard X-ray Photoelectron Spectroscopy (HAXPES) has enabled to study deeply buried interfaces [1]. It was shown that by combining HAXPES with inelastic background analysis [2], structures at a depth >50 nm can be studied. Here, we present a study on technologically relevant High Electron Mobility power Transistors Ta/Al.

The study was performed on stacks of two metal layers of aluminum and tantalum with different thicknesses deposited on an $Al_{0.25}Ga_{0.75}N$/AlN/GaN heterostructure [Fig. 1a]. We have used the technique to non-destructively study the activation annealing. HAXPES was performed at the Spring-8 synchrotron (Japan) using 8 keV photons.

The figure shows spectra measured around Al, Ga and Ta peaks for an as deposited sample [Fig. 1b]. The calculation of inelastic background was performed using two input parameters; the IMFP, calculated using the TPP-2M formula [3] and as the spectra present marked plasmons, after the elastic peaks, we used an average of individual inelastic cross-sections, σ, which can be determined from reflection electron energy-loss spectra. The calculation of this cross-sections' average has been made according to a mixtures rule, involving the different crossed layers, which consists in a relevant way to analyze deeply buried layers. The figure shows how the modelling of the inelastic background is used to determine the in-depth distributions, which are found in good agreement with the TEM results. We have also successfully used this technique to study the effect of annealing on the diffusion of the elements at the interfaces.

**Bibliography**

[1] P. Risterucci et al., *Applied Physics Letters* 104, (2014).

[2] S. Tougaard, *Journal of Electron Spectroscopy and Related Phenomena*, 178–179 (2010).

[3] H. Shinotsuka et al., *Surface and Interface Analysis* 47, 871-888 (2015).

*Part of this work was performed at the Nanocharacterization Platform of CEA-MINATEC (PFNC).*

*NIMS and Spring-8 is acknowledged for providing beamtime and the staff of the BL15-XU beamline for their assistance during the experiment. Cyril Guedj is acknowledged for providing TEM analysis and expertise.*

3:20pm **SA+AS-MoA6 Soft X-ray ARPES Investigation of the Spin-polarized n-$BaTiO_3$/$SrRuO_3$ Buried interface**, ***Julien Rault**, P. Le Fèvre, F. Bertran, J. Rebellato,* Synchrotron SOLEIL, France*; T. Maroutian, P. Lecoeur,* Université Paris-Sud - CNRS, France

The electric field control of functional properties such as spin injection is a crucial goal in oxide-based electronics. Non-volatile switching between different electron and spin transport in a tunnel junction channel can be achieved through charge accumulation or depletion at the interfaces [1, 2]. It has been recently suggested [3] that polarization-dependent spin-injection is expected at the interface between ferromagnetic $SrRuO_3$ (SRO) and semiconducting, ferroelectric n-$BaTiO_3$ (n-BTO), paving the way for adjustable spin-injection in full-oxide devices. This fascinating effect is due to the matching of the spin-dependent Fermi surface of SRO with n-doped BTO tube-like Fermi-surface [2, 3].

To investigate this phenomenon experimentally, we use angle-resolved photoemission spectroscopy (ARPES) to access the band structure of a buried n-BTO/SRO interface. A well-known limitation of ARPES comes from its very low probing depth (< 2 nm) due to the very low electron inelastic mean free path in the usual photon range of ARPES (1-100 eV). To overcome this limitation, we use soft x-ray photons and are able to access the interface Fermi surface below a 2-nm, upward polarized BTO thin film deposited on SRO.

Using 600 eV linearly-polarized photons, we measure the band dispersion of the interface SRO through the BTO band gap along with BTO bands for higher binding energies (see Fig. 1). The SRO-related bands were not visible at lower probing depth (photon energy ca. 250 eV) showing they actually come from the interface. Clear light-polarization dependence on the energy-momentum cuts along ΓX high-symmetry direction is shown in Figure 1. This is used to assign some parts of the Brillouin zone to specific orbitals with different spin-polarization. The in-plane Fermi surface of the interface SRO for $k_z$ = Z was also acquired and showed some clear feature fitting well the calculated band structure from Liu *et al.*, see Figure 2.

These set of results is indicative of how soft x-ray ARPES is a technique of choice to probe the band structure of functional oxide interfaces. Combined with spin-resolved photoemission, which is available at our laboratory, it will help to better understand the spin polarization predicted in ferromagnetic/ferroelectric heterostructures.

[1] Marinova, M. *et al., Nano Letters* **15**, 2533–2541 (2015)

[2] Liu, X., Burton, J. D., Zhuravlev, M. Y. & Tsymbal, E. Y., *Physical Review Letters* **114**, 46601 (2015)

[3] Liu, X., Wang, Y., Burton, J. D. & Tsymbal, E. Y., *Physical Review B* **88**, 165139 (2013)

4:00pm **SA+AS-MoA8 Progress and Perspectives in Photoemission using XFEL Radiation**, ***Serguei Molodtsov**,* European XFEL GmbH, Germany **INVITED**

Photoemission is today one of the most powerful techniques for investigating low-energy properties of matter from the aerosol and adsorbate nanoparticles and molecules to the surface and bulk of solid state matter with many dedicated and specialized beamlines at synchrotron radiation facilities. The upcoming ultra-brilliant FEL sources, giving access to ultrashort timescales in the fs range, in combination with the outstanding peak brilliance achieved, set the stage for novel science. Experiments on X-ray FEL sources are being pioneered and planned at low repetition rate facilities (FLASH, LCLS, SACLA, FERMI). The European XFEL that will come in early user operation already in 2017 will be characterized by laser action from 260 eV to 25 keV photon energy and above. The facility will also yield an extremely high mean brilliance with a repetition rate suitable for different modes of photoemission detection. Together with the planned for 2021 source LCLS II this will make the European XFEL unique FEL facility for photoemission studies worldwide.

In this presentation an overview of time-resolved photoemission experiments on solids that were done at XFELs so far will be given and perspectives related to high repetition rate XFEL facilities, particularly European XFEL, will be provided.

4:40pm **SA+AS-MoA10 Revealing the Origins of Non-Joulian Magnetism with High-Resolution Photoemission Microscopy**, ***Alexander Gray**, R.U. Chandrasena,* Department of Physics, Temple University*; H.D. Chopra,* Department of Mechanical Engineering, Temple University **INVITED**

All magnets elongate and contract anisotropically when placed in a magnetic field, an effect referred to as Joule magnetostriction. The hallmark of Joule magnetostriction is volume conservation, which is a broader definition applicable to self-accommodation of ferromagnetic, ferroelectric or ferroelastic domains in all functional materials. Recently, a new class of single-crystalline magnets exhibiting a 'giant' non-volume-conserving or non-Joulian magnetostriction was discovered [1]. In this talk I will discuss the results of our recent investigations of non-Joulian $Fe_3Ga$ alloys using high-resolution polarization-dependent photoelectron microscopy. Our results suggest that non-Joulian magnetism arises from an unusual nearly-equipartition of the crystal into nm-scale lamellar domains and domain walls within highly periodic magnetic micro-cells. We suggest that this high-energy configuration is stabilized by the strain gradients arising from CDW that offsets electronic energy by a greater amount. High-resolution x-ray magnetic circular dichroism measurements at the Fe and Ga *L* absorption edges further provide evidence of weak iron-induced magnetism on gallium atoms via negative exchange. The results are in excellent agreement with the state-of-the-art theoretical electronic-structure calculations. Our findings open up new ways for the design of alloy systems having functional magnetic properties similar to $Fe_3Ga$ where non-Joulian magnetostriction was first reported.

[1] H. D. Chopra and M. Wuttig, Non-Joulian magnetostriction, *Nature* **521**, 340 (2015).

# Author Index

## Bold page numbers indicate presenter