Novel Trends in Synchrotron and FEL-Based Analysis Focus Topic

Room 202A - Session SA+MI-ThM

Ultra-fast Dynamics for Magnetic and Quantum Systems

Moderator: Claus Michael Schneider, Forschungszentrum Juelich GmbH, Germany

8:00am **SA+MI-ThM-1 New Opportunities at the APS: Using Intermediate Energy X-rays to Investigate Collective Behavior in Interacting Electron Systems**, ***Jessica McChesney***, *F Rodolakis,* Argonne National Laboratory

In an effort to address one of the grand challenges for condensed matter physics in the 21st century, namely to gain an understanding of the physics of materials which exhibit collective electronic phenomena, the Advanced Photon Source has developed the intermediate-energy x-ray (IEX) beamline. Now fully operational, this beamline enables the investigation of collective behavior in interacting electron systems using two distinct but complementary techniques: angle-resolved photoemission spectroscopy and resonant soft x-ray scattering. In this talk, I will discuss some of the unique capabilities of the beamline and present several examples of collective behavior in interacting electron systems including electron-phonon coupling, spin and charge density waves and orbital ordering in high-temperature superconductors, transition metal oxides, topological insulators and heavy fermion materials.

8:20am **SA+MI-ThM-2 Observation of Surface Recombination in Ultra-fast Carrier Dynamics of $La_{0.7}Sr_{0.3}MnO_3$ Thin Films**, ***Saeed Yousefi Sarraf***, *G Cabrera, R Trappen, N Mottaghi, S Kumari, C Huang, A Bristow, M Holcomb,* West Virginia University

Perovskite oxides (ABO_3) are a promising class of transition metal oxides that have attracted significant attention in material science due to diverse range of properties. Many studies on structural and magnetic properties have been done on perovskite oxides to base the multifunctional devices made by and proposed for these materials. Yet another very important property of perovskite oxides is that many of their band gaps are in the visible range. These gaps make these oxides a suitable choice for photovoltaic applications. However, despite the very critical role this property plays in light harvesting devices, there has been a limited understanding about the carrier dynamics of these materials, which inform us about the efficiencies of photovoltaic devices, especially in lower thicknesses. Since by decreasing the film thickness, the surface to bulk ratio increases and surface electrons dominate the bulk electrons, surface recombination might occur as an extra channel of energy relaxation, which decreases the device efficiency. Perovskite oxide $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) thin films were fabricated with different thicknesses by pulsed laser deposition on (100) $SrTiO_3$ single crystal substrates. Our films' quality were checked by in situ RHEED patterns and oscillations, X-ray diffraction and reflectivity, magnetometry and atomic force microscopy. Ultra-fast carrier dynamics were studied by a degenerate reflectivity pump probe setup at 800nm for different film thicknesses and different pump powers. For films with a thickness above ~20nm three different recombination were observed, attributed to electron phonon relaxation, spin lattice phonon assists relaxation and thermal diffusion relaxation. However, for films thinner than ~20nm an extra relaxation mechanism was observed, which we attributed to surface recombination. This optics work was funded by the American Chemical Society (PRF #56642-ND10); sample growth and optimization were supported by NSF (DMR-1608656).

8:40am **SA+MI-ThM-3 Non-equilibrium Control of Charge & Spin Motion in Quantum Materials**, ***Hermann Dürr***, Uppsala University, Sweden**INVITED**

A key driver of modern information technology is the quest for "smaller and faster" information processing and storage. The ultimate speed limit is the speed of light. Therefore, the idea to probe, change and control properties of materials with the help of light has long intrigued researchers in materials science. Of particular interest are magnetic materials which in nanostructured form are used for data storage, memory and processing. In this talk I will show several examples the unique potential of using femtosecond soft x-ray pulses from x-ray free electron lasers such as the LCLS to probe in real time ultrafast spin dynamics in nanoscale systems and during all-optical magnetic switching. Understanding and ultimately engineering the evolving electron, spin and lattice motion on the time- and lengthscales associated with the relevant interactions promises new ways for storing and processing of information.

9:20am **SA+MI-ThM-5 XUV-transient Grating: Probing Fundamental Excitations at the Nanoscale**, ***Laura Foglia***, *F Capotondi, R Mincigrucci, D Naumenko, E Pedersoli, A Simoncig, G Kurdi, M Manfredda, L Raimondi,* Elettra-Sincrotrone Trieste, Italy*; N Mahne,* IOM-CNR, Italy*; M Zangrando, C Masciovecchio, F Bencivenga,* Elettra-Sincrotrone Trieste, Italy **INVITED**

Nonlinear optical spectroscopies take advantage of multiple light-matter interactions via the Nth-order susceptibilities, to disentangle and selectively access the many interacting degrees of freedom that characterize complex systems. Indeed, the control on photon parameters (frequency, arrival time, polarization, etc.) for each field independently gives rise to a manifold of experimental techniques that allow to monitor, on ultrafast timescales, structural changes, spin and electron dynamics, collective phenomena as well as to selectively probe correlations among different excitations. Among the nonlinear processes, third order, or four-wave-mixing (FWM), interactions occur in all materials independently of their symmetry, and are thus the most widely used in applications. While nowadays these techniques are well established at optical wavelengths, their birth required the invention of the laser. Similarly, their extension to sub-optical wavelengths (XUV and X-ray), envisioned theoretically more than a decade ago, had to wait until the recent development of free electron lasers (FELs). XUV-FWM will allow exploiting core-hole resonances to address correlations among low-energy excitations and core states as well as monitoring charge and energy transfer processes. Additionally, it will extend the accessible wavevector range to the mesoscopic regime (0.1-1 nm−1), which is fundamental to investigate, e.g., lattice dynamics in nanostructures and disordered systems as well as transport phenomena at the nanoscale.

Here I report on the demonstration of XUV four-wave-mixing response in a transient grating (TG) approach, exploiting the unique properties of the seeded FEL source FERMI and of two dedicated setups: TIMER and mini-TIMER. All-XUV TG data allowed us to determine the phonon and thermal dynamics of several semiconductors in an uncharted length-scale range, extending down to ≈20 nm. The results are compared with those of XUV pump/optical probe TG, evidencing different couplings to the electronic subsystem.

Additionally, I present the first evidence of FWM processes stimulated by FEL pulses at different wavelengths, obtained exploiting the multi-color capabilities of FERMI and will discuss the foreseen implementation of second order wave-mixing techniques for the chemical and interface specific probing of electronic processes.

11:00am **SA+MI-ThM-10 Study of Photo-induced Dynamics in Quantum Materials using Femtosecond Time-resolved X-ray Scattering**, ***Wei-Sheng Lee***, SLAC National Accelerator Laboratory **INVITED**

It remains a great challenge to characterize and understand photo-induced dynamics in quantum materials when it is driven out-of-equilibrium by ultrafast photon pulses. Time-resolved x-ray scattering, enabled by x-ray free electron laser, can track the time-evolution of the magnetic, charge, and lattice degrees of freedom with femtosecond time resolution, providing new insights into the photo-induced dynamics. In this presentation, I will first highlight photo-induced dynamics of spin and charge orders in striped nickelate. The strong coupling between spin and charge orders still survives, despite that both orders have been strongly suppressed by photo-excitations [1,2]. In addition, by resonantly pumping a bond-stretching phonon using mid-IR pulses, we observed lattice-driven dynamics that is different from hot-electron-driven dynamics induced by optical pumping across the band gap [3]. Then, I will discuss phenomena associated with coherent lattice oscillations by highlighting the measurement of atomic displacement of a coherent A1g mode in an iron-based superconductor $BaFe_2As_2$, which can be correlated with accompanied electronic variations [4]. In particular, a quantitative "lock-in" comparison between electronic band structure obtained by time-resolved ARPES and the measured atomic displacements allows us to directly obtain orbital-specific electron-phonon coupling strength without any prior assumption of the electronic band structures [5]. Finally, I will showcase a proof-of-principle time-resolved RIXS experiment on CDW ordered 1T-$TiSe_2$ as an outlook for the future time-resolved x-ray scattering experiment in the next generation x-ray free electron laser, such as the LCLS-II.

1. W. S. Lee et al.. Phase fluctuations and the absence of topological defects in photo-excited charge ordered nickelate. Nature Communications 3, 838 (2012).

2. Y. D. Chuang, W. S. Lee et al., Real-Time Manifestation of Strongly Coupled Spin and Charge Order Parameters in Stripe-Ordered

La1.75Sr0.25NiO4 Nickelate Crystals Using Time-Resolved Resonant X-Ray Diffraction. Phys. Rev. Lett. 110, 127404 (2013).

3. W. S. Lee et al., Non-equilibrium Lattice-driven Dynamics of Stripes in Nickelates using Time-Resolved X-ray Scattering. Phys. Rev. B 95, 121105(R) (2017).

4. S. Gerber, et al., Direct characterization of photo-induced lattice dynamics in BaFe2As2. Nature Communication 6, 7377 (2015).

5. S. Gerber, S.-L. Yang et al., Femtosecond electron-phonon lock-in via photoemission and x-ray free-electron laser. Science 357, 71 (2017).

11:40am **SA+MI-ThM-12 HAXPES Lab- A Home Lab System for HAXPES Measurements***, S Eriksson,* Scienta Omicron*; **Anna Regoutz**,* Imperial College London, UK

During the past decade, increased attention has been shown to hard X-rays in the photoelectron spectroscopy field. This is mainly due to the increased information depth enabled by the higher photon energies. Such bulk sensitive measurements could previously only be performed at dedicated synchrotron radiation facilities. The beam lines providing this type of radiation are heavily booked, so access to the experimental setups is thus limited. Higher excitation energies also enables bulk sensitive measurements of deep core levels not accessible with standard XPS.

Here we present a newproduct featuring a monochromized X-ray source giving out Ga Ka radiation at 9.25keV and a wide acceptance angle hemispherical electron analyzer, both combined on a simple to use vacuum system. The base system can easily be customized by adding separate modules such as a MBE- or preparation chamber or a glove box. With this system, a new set of possible experiments opens up in the home laboratory: investigations of buried interfaces, in operando devices, real world samples, etc.

The X-ray source consists of a MetalJet X-ray tube and the electrons which are accelerated into this jet generate an intense Ga Ka radiation. These X-rays are monochromized using a newly developed monochromator. The small spot size of 20 µm provided by the liquid jet source is maintained throughout the passing of the monochromator and only slightly broadened to about 50µm. In order to allow for easy adjustment of the X-ray focal point relative to the electron analyzer, the entire assembly of monochromator and source can be moved down to a precision of a few micrometers. The hemispherical electron analyzer is configured for high kinetic energies allowing for detection of the full energy range the source provides and a large acceptance angle of +/- 30 degrees. The overall system resolution is shown to be <0.5 eV.

We present data taken from polycrystalline gold and silicon wafers with a surface layer of silicon dioxide with a controlled thickness as well as transistor stacks and energy related materials.

Author Index

Bold page numbers indicate presenter