

Actinides and Rare Earths Focus Topic

Room A215 - Session AC+LS+MI-MoM

Magnetism, Complexity, Superconductivity, and Electron Correlations in the Actinides and Rare Earths

Moderators: Krzysztof Gofryk, Idaho National Laboratory, Ladislav Havela, Charles University, Prague, Czech Republic

8:20am AC+LS+MI-MoM-1 Possible Structural Quantum Phase Transition in UCr₂Si₂ Accessed Through Cr → Ru Chemical Substitution, *Ryan Baumbach*, Florida State University **INVITED**

Materials with intertwined magnetic, electronic, and structural degrees of freedom often can be tuned (e.g., using pressure or chemical substitution) to induce novel behavior, including unconventional superconductivity. Examples include the cuprates, iron based superconductors, and lanthanide/actinide-based compounds, and despite their diversity of structure, chemistry and interaction mechanisms, their individual phase diagrams often conform to a semi-universal format that features a quantum phase transition. As a result, there have been prolonged efforts to develop new families of materials based on this paradigm. Even so, there still are few examples of f-electron intermetallics that combine both magnetic and structural quantum phase transitions. In this talk, we will present results from recent efforts to tune the ordered states of UCr₂Si₂, which is a Kondo lattice metal with antiferromagnetic ordering near $T_N \approx 24$ K and a structural phase transition near $T_S \approx 200$ K. In particular, we will focus on the influence of Cr to Ru chemical substitution, where we find that both T_N and T_S are rapidly suppressed towards separate quantum phase transitions. The impact of the quantum phase transitions on the structural, magnetic, and electronic properties will be examined in detail.

9:00am AC+LS+MI-MoM-3 Dynamic Spin Transport in Antiferromagnetic Insulators: Angular Dependent Spin Pumping in Y₃Fe₅O₁₂/NiO/Pt Trilayers, *Fengyuan Yang*, The Ohio State University **INVITED**

In recent years, pure spin transport driven by ferromagnetic resonance (FMR) spin pumping or a thermal gradient has attracted intense interest and become one of the most active frontiers in condensed matter and materials physics. Extensive research efforts have demonstrated pure spin currents in a broad range of materials, which enrich our understanding of dynamically-driven spin transport and open new paradigms for energy-efficient, spin-based technologies. Antiferromagnetic (AF) insulators possess various desired attributes, such as low loss and high speed up to THz frequencies, for future spintronic applications.

To probe the dynamic spin transport phenomena and the underlying mechanisms in AF insulators, we use high-quality Y₃Fe₅O₁₂ (YIG) epitaxial thin films excited by FMR as a source to inject spins into AF insulator NiO layers and detect the transmitted spin current using inverse spin Hall effect (ISHE) signals in YIG/NiO/Pt trilayers [1, 2]. We observed robust spin currents from YIG to Pt across AF insulators, which initially enhances the ISHE signals and can transmit spin currents up to 100 nm thickness, demonstrating highly efficient spin transport through an AF insulator carried by magnetic excitations. Recently, we studied the angular dependence of spin pumping in a series of YIG/NiO/Pt trilayers as the orientation of the applied magnetic field is rotated out of plane [3]. A simple sinusoidal angular dependence of V_{ISHE} has been viewed as a signature of spin pumping. Surprisingly, we observe an extensive plateau in the V_{ISHE} vs. ϑ_H plots with a pronounced peak feature at an out-of-plane angle of 45° to 60° when the measurement temperature is close to the Néel temperature (T_N) of NiO. This phenomenon can be understood as arising from the competition between the exchange coupling at the YIG/NiO interface, the easy-plane and in-plane easy-axis anisotropies of NiO, and the effect of the applied magnetic field. While insulating antiferromagnetic films can efficiently transmit spin currents and show promise for integration in spintronic devices, the underlying physics of spin ordering and dynamics is richer than currently understood.

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9:40am AC+LS+MI-MoM-5 Pressure Studies of Strongly Correlated Phases in Rare Earth Compounds, *Rena Zieve*, University of California, Davis **INVITED**

Various strongly correlated materials have complex low-temperature phase diagrams, exhibiting magnetism and superconductivity as well as spin glass, non-Fermi liquid, and other behaviors. Since each material has its own quirks, determining the underlying universal influences has been challenging. Pressure is a key tool in these efforts, since pressure can tune the interactions within a material without changing its chemical composition or impurities. I will discuss how hydrostatic or uniaxial pressure can probe strongly correlated materials by changing valence, breaking crystal symmetry, or altering the dimensionality of the electron system. I will draw examples from various rare earth compounds. Finally, I will mention recent pressure application techniques that expand the range of possible characterization measurements.

10:40am AC+LS+MI-MoM-8 Fermi Surface Reconstructions and Transport Properties in Heavy-fermion Materials, *Gertrud Zwicknagl*, Institut f. Mathemat. Physics, TU Braunschweig, Germany

The search for new types of exotic topological orders has recently rekindled the interest in Fermi surface reconstructions. Of particular interest are Electronic Topological (Lifshitz) transitions where the number of FS sheets changes abruptly under the influence of external parameters like chemical doping, pressure, or magnetic field. Lifshitz transitions (LTs) are generally associated with the presence of critical points in the electronic band structure, i. e., maxima, minima, or saddle points whose presence follows directly from lattice periodicity. As their separation from the Fermi energy is of the order of the bandwidth the critical points usually do not affect the low temperature behavior. In heavy-fermion materials, however, magnetic fields can drive LTs which are reflected in pronounced anomalies in thermodynamic and transport properties. Here we demonstrate that the magnetic field-dependent anomalies in the Seebeck coefficient provide detailed information not only on the critical points, i. e., their character and position relative to the Fermi energy but also on the quasi-particle dispersion in the vicinity of the critical points, i. e., the effective mass tensor. For lanthanide-based HFS, the theoretical analysis is based on Renormalized Band (RB) structure calculations assuming that the heavy quasiparticles result from a Kondo effect. For U-based HFS, on the other hand, we adopt the "dual nature" model which allows for a microscopic description of the heavy bands. The calculated Lifshitz transitions reproduce the observed positions of anomalies in the Seebeck coefficients surprisingly well.

11:00am AC+LS+MI-MoM-9 Direct Measurement the 5 f_{5/2} and 5 f_{7/2} Unoccupied Density of States of UO₂, *James G. Tobin*, University of Wisconsin-Oshkosh; *S Nowak*, SLAC National Accelerator Laboratory; *C Booth*, Lawrence Berkeley National Laboratory; *E Bauer*, Los Alamos National Laboratory; *S Yu*, Lawrence Livermore National Laboratory; *R Alonso-Mori*, *T Kroll*, *D Nordlund*, *T Weng*, *D Sokaras*, SLAC National Accelerator Laboratory

In a world of ever increasing population and diminishing resources, the need for abundant and inexpensive energy remains critical. [1] Despite the problems associated with radioactive contamination/disposal and nuclear proliferation, electricity generated by nuclear power remains immensely important, [2] providing for 20% of the electrical grid of the USA and 50% or more for several European nations. [3-6] Uranium Dioxide (UO₂) is by far the widely used nuclear fuel for the generation of electricity. [7] Thus, a fundamental understanding of the electronic structure of UO₂ is crucial, if only to provide the best theoretical models for its disposal and storage. [7, 8]

Using High Energy Resolution Fluorescence Detection (HERFD) in a Resonant Inelastic Scattering (RIXS) experiment and electric dipole selection rules, the U 5f_{5/2} and U 5f_{7/2} Unoccupied Densities of States (UDOS) were determined. Significant changes were observed in going from UF₄ (localized, 2 5f electrons) to UCd₁₁ (localized, 3 5f electrons), consistent with the predictions of the Intermediate Coupling Model. The results for UO₂ were experimentally confirmed by direct comparison with the Bremsstrahlung Isochromat Spectroscopy for Uranium Dioxide.

References

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11:20am **AC+LS+MI-MoM-10 Optimizing the Magnetic Performance of Tetragonal $\text{ReFe}_{12-x}\text{M}_x$ Phases by First Principles Computational Simulations, Heike Christine Herper, O Vekilova, P Thunström, O Eriksson, Uppsala University, Sweden**

The increase of environmentally friendly energy production is coupled to an increasing demand for new magnetic materials. Especially, new Rare earth (Re) lean permanent magnets are highly sought after as possible replacement for high-performance magnets based on Nd-Fe-B and Dy to limit costs and supply risk. In this context the tetragonal 1:12 phase (TmMn_{12}) which contains 35% less Re than commercial Nd-Fe-B magnets are rediscovered. To stabilize this phase with light Re and Fe instead of Mn a nonmagnetic phase stabilizing element is needed but this degrades the magnetic performance.

To identify new 1:12 phases being suitable for permanent magnet applications materials design based on computational simulations has become an important tool. Here we focus on $\text{ReFe}_{12-x}\text{M}_x$ with Re = Y, Ce, Nd, Sm and M = Ti and V. We use state of the art density functional theory methods (VASP; full potential LMTO (RSPt)). The phase stability and the magnetic properties were calculated depending on the M concentration. Aiming to reduce the Re amount we monitor the performance depending on the Nd/Y ratio.

The key quantities are the magnetocrystalline anisotropy (MAE) and the magnetization. To capture the correct magnetic behavior, it is crucial to describe the localization of the 4f electron properly for each Re. While for Sm-based systems the spin-polarized core approximation is sufficient to describe the localized 4f electrons, it fails for Nd, e.g. the low temperature MAE of $\text{NdFe}_{11}\text{Ti}$ would be uniaxial instead of conic. Using a DFT+U approach with $U = 5$ eV, $J = 1.1$ eV for $\text{NdFe}_{11}\text{Ti}$ reproduces the experimentally observed behavior. Ce is special since the uniaxial MAE of $\text{CeFe}_{11}\text{Ti}$ is obtained independent from the treatment of the 4f electron. However, an analysis of the hybridization function analogue to [1] shows that a spin polarized core approximation is more appropriate for Ce-based 1:12 phases. For a deeper insight additional studies are carried out to examine the crystal field splitting.

With SmFe_{11}V system a new phase was found leading to an increase of the magnetization by 17% compared to the commonly used concentrations of V. In view of the MAE a replacement of Nd by Y turned out to be preferable over a reduction of Ti. MAE values of 1.3 MJ/m^3 ($(\text{NdY})\text{Fe}_{11}\text{Ti}$) and 1.7 MJ/m^3 (SmFe_{11}V) are predicted [2]. The latter could already be verified in recent experiments [2].

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11:40am **AC+LS+MI-MoM-11 Optical Excitation Effect on Magnetodielectric and Photodielectric Properties of Rare Earth doped ZnO:Na Nanoparticles, W Jadwisieniczak, Mohammed Bsate, F Inbanathan, Ohio University; R Martinez, University of Puerto Rico; H Huhtinen, University of Turku, Finland; R Palai, University of Puerto Rico**

There have been ongoing efforts in developing rare earth (RE) ions doped semiconductors responsive to optical and magnetic stimuli for developing multifunctional devices. ZnO is considered as a promising semiconductor with wide range of applications in optoelectronics and spintronics due to its optical properties (i.e. direct band gap, high electron mobility, high thermal stability, strong absorption of UV), and at the same time being an attractive host for RE doping resulting in enhanced spin polarization in ZnO:RE matrix [1]. In this project, undoped and RE ions (Er and Yb) doped and co-doped ZnO:Na nanoparticles were studied with an aim to understand recently reported magnetodielectric and photodielectric effects stimulated by magnetic field and UV excitation stimuli [2]. After synthesizing well-defined Er and Yb-co-doped ZnO:Na by sol-gel route, evaluation of structural, optical, magnetic and electrical characteristics of

synthesized nanoparticles was performed. Structure, morphology, and composition of the samples were analyzed by XRD and SEM showing high quality hexagonal crystal structure. The absence of secondary phases was confirmed by Raman spectroscopy and XPS analysis of all elements in ZnO:NaErYb compound. Optical properties were investigated by optical reflectance spectroscopy, photoluminescence (PL), PL excitation, and PL kinetics with photons excitation energy corresponding to above and below bandgap energy. PL spectra were investigated under magnetic field of up to 2 Tesla in 7 K – 400 K temperature range. It was observed that PL spectra exhibit broad green-yellow defects emission band without NBE excitonic peak. PLE spectra revealed that both Er^{3+} and Yb^{3+} ions are optically active and involved in complex energy transfer between ZnO:Na host and 4f-shell levels of RE^{3+} ions dopants. PL spectra show features related to intra-shell 4f-4f transitions of the Er^{3+} ion (at 522 nm) and Yb^{3+} ion (at 980 nm). It was observed that luminescence decay of the ZnO defect band is unexpectedly long ($>$ tens of 10^{-3} second), strongly affected by RE^{3+} ions doping and depends on the magnetic field strength when excited at 3.814 eV. In the presentation we will attempt to correlate reported magnetodielectric and photodielectric properties of the Er,Yb-co-doped ZnO:Na [2] with observed optical characteristics.

References

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Actinides and Rare Earths Focus Topic

Room A215 - Session AC-MoA

Early Career Scientists

Moderators: Art Nelson, Lawrence Livermore National Laboratory, David Shuh, Lawrence Berkeley National Laboratory, Evgeniya Tereshina-Chitrova, Charles University, Prague, Czech Republic

1:40pm **AC-MoA-1 Advanced Characterization of Nuclear Fuels, Lingfeng He, T Yao**, Idaho National Laboratory; *V Chauhan*, The Ohio State University; *A Sen*, Purdue University; *Z Hua, M Bachhav*, Idaho National Laboratory; *M Khafizov*, The Ohio State University; *J Wharry*, Purdue University; *M Mann*, Air Force Research Laboratory; *T Wiss*, European Commission, Joint Research Centre (JRC); *J Gan, D Hurley*, Idaho National Laboratory

INVITED

Oxide nuclear fuels have been widely used in light water reactors. The thermal conductivity of nuclear fuels is closely related to energy conversion efficiency as well as reactor safety. Understanding the mechanisms that cause the degradation in thermal conductivity in a high radiation environment is important for the design and development of new high-burnup fuels. For oxide nuclear fuels, phonon scattering by point defects, extended defects such as dislocation loops and bubbles, and grain boundaries plays a significant role in limiting the thermal transport properties. In this work, detailed microstructural characterization of pristine and ion irradiated ThO₂ and UO₂ has been performed by using electron backscatter diffraction (EBSD), atomic-resolution scanning transmission electron microscope (S/TEM), atom probe tomography (APT) and time-domain Brillouin scattering (TDBS) techniques. The thermal conductivity before and after irradiation has been determined using laser-based modulated thermoreflectance (MTR) technique. This work is partially supported by the Center for Thermal Energy Transport under Irradiation, an Energy Frontier Research Center funded by the U.S. Department of Energy Office of Sciences.

2:20pm **AC-MoA-3 The Influence of Relative Humidity on the Oxidation of δ -Pu, Scott Donald, J Stanford, A Nelson, B McLean**, Lawrence Livermore National Laboratory

INVITED

The evolution of delta stabilized plutonium aged under a controlled environment composed of laboratory air and a range of relative humidities up to 95% was studied using Auger electron spectroscopy (AES). Linear-least squares analysis was performed on AES spectra acquired during Ar⁺ sputter depth profiles to gain insight on the thickness and any variation in the chemical speciation of the oxide. Sputter rates were calibrated from depth profiles obtained from an oxide of a known thickness from FIB/SEM measurements. At all relative humidities, the initial oxide layer was found to grow logarithmically, indicative of a diffusion-controlled process. The rate of oxide growth was also found to be independent of oxygen partial pressure (for pO₂ > 31.5 Torr) for all conditions studied.

The work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

3:00pm **AC-MoA-5 Magnetization and Transport Properties of Delta Phase Uranium, Xiixin Ding, N Poudel, T Yao, J Harp, K Gofryk**, Idaho National Laboratory

At room temperature, uranium metal is in its alpha form, the most common structural form of the element. It consists of corrugated sheets of atoms in an asymmetrical orthorhombic structure. However, the room temperature stabilized delta phase can be formed by alloying uranium with zirconium, which is known to have hexagonal crystal structure. It is important to know the physical properties of U-Zr alloys due to their technological importance. In this talk, we will present the first-time results of magnetization, transport and thermodynamic measurements of delta phase uranium at low temperatures and under high magnetic fields. The results obtained help us to understand the 5f ground state properties in different phases of U. We will discuss implications of these results.

3:20pm **AC-MoA-6 Using Fused Filament Fabrication to Develop Customized Materials which Attenuate Ionizing Radiation, Zachary Braunstein, E Murphy, J Dumont, S Talley, K Lee, A Labouriau**, Los Alamos National Laboratory

Ionizing radiation is of serious consideration in the nuclear industry because protecting workers and instrumentation is of utmost concern when operating equipment that emits potentially hazardous radiation.

Currently, commercial products are readily used as protective barriers, but there are circumstances when these are less than ideal at providing optimal shielding against neutrons and gamma rays^{[1],[2]}. As innovations to nuclear energy technologies continue to progress, developing new materials for radiation shielding grows in importance and need.

In the present work, we used an additive manufacturing (AM) technique known as Fused Filament Fabrication (FFF) to create novel 3D printed materials for radiation shielding. FFF is a layered AM process whereby thermoplastic filaments are heated up to their melting point and extruded into cross-sections of the end product^{[3],[4]}. Because FFF has the capability to create prototypes and end-use parts with fine resolution details and excellent strength-to-weight ratios, the technology is used throughout aerospace, automotive, and medical industries.

Difficulties in creating filaments for FFF arise from fabricating a homogeneous wire that has uniform thickness and a smooth surface. If a filament does not have these initial properties, then either the FFF process will not work or the end product will not be as desired. Creating a homogeneous wire proves more difficult when different base and filler materials are used in the fabrication process, however, this can be solved if the different materials are combined in a liquid solution. Creating a wire of uniform thickness relies heavily on the extrusion process, whereby the temperature and extrusion speed are controlled.

In this study, we have prepared homogeneous filaments with varying processing conditions such as the contribution of additives and the control of extrusion temperature and speed. Thus, we used FFF to create novel filaments to print sheets of customized materials for attenuating ionizing radiation. Irradiating the printed samples was performed at the Los Alamos Neutron Science Center and the Gamma Irradiation Facility by bombarding the customized materials with neutrons and gamma rays, respectively.

References

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3. Guo, N. and M.C. Leu, *Frontiers of Mechanical Engineering*, 2013. **8**(3): p. 215-243.
4. Srivatsan, T.S. *et al*, *Additive Manufacturing: Innovations, Advances, and Applications*. CRC Press. 2016. p. 1-48.

4:00pm **AC-MoA-8 Thermodynamic and Thermal Transport Properties of Thorium Dioxide single crystals, Narayan Poudel, X Ding**, Idaho National Laboratory; *J Mann*, Air Force Research Laboratory; *K Gofryk*, Idaho National Laboratory

Thorium dioxide (ThO₂) crystallizes into CaF₂-type (fluorite) cubic structure, similar to other members of AnO₂ (An = Th-Am) family. Thorium dioxide forms stoichiometrically and is a wide-gap transparent insulator (E_g~5-6 eV). This material is used as nuclear fuel in certain types of nuclear reactors (CANDU) that might have more advantages than conventional UO₂-based nuclear reactors. It is because of its higher thermal conductivity, higher corrosion resistance, and higher melting point. Despite its importance in nuclear technology, the thermodynamic and thermal transport properties of ThO₂ single crystals have not been studied extensively, especially at low temperatures where many different scattering mechanisms such as boundary, defects, and/or phonon-phonon dominate the heat transport. In this talk, we will present our recent measurements of the heat capacity and thermal conductivity of ThO₂ single crystals, obtained from room temperature down to 2 K. Large and good quality single crystals of ThO₂ have been synthesized by hydrothermal method for this study. We will also compare the result obtained on ThO₂ to UO₂, especially in the context of impact of 5f-electrons on thermodynamic and transport behavior in these materials.

4:20pm **AC-MoA-9 Magnetic Nanoparticles for Biomedical Applications, Iliana Medina-Ramirez, A Diaz de Leon Olmos**, Universidad Autonoma de Aguascalientes, Mexico; *J Zapien*, City University of Hong Kong

Magnetic nanostructured materials (MNMs) are attractive candidates for biomedical applications because of the highly desirable advantages of magnetic-guided targeting. Furthermore, MNMs can be produced by simple fabrication processes that enable flexibility to modulate their properties and desired bio-activity.

The simultaneous optimization of the bio-activity of interest while simultaneously preventing, or at least minimizing, deleterious side effects is of the utmost importance for critical applications such as the development of magnetic hyperthermia cancer treatment. However, this

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requires more complex multi-criteria optimization studies. We use Fe_3O_4 and CoFe_2O_4 MNMs prepared by microwave solvothermal method and a citrate surface modification, to modulate their toxicity and stability, as model systems to develop protocols for optimization of therapeutic MNMs. The interaction of surface-modified MNMs with HepG2 cells is evaluated by colorimetric, optical microscopy and Atomic Force Microscopy (AFM) studies for dose. We show that AFM presents important advantages over conventional techniques including information on the potential internalization of MNMs by the cells as function of surface modification. The optimized MNMs in this study present morphological, colloidal, biocompatibility and magnetic properties that make them promising candidates for the development of efficient therapeutic agents for hyperthermia cancer treatment applications.

Actinides and Rare Earths Focus Topic

Room A215 - Session AC+AS+LS-TuM

Chemistry and Physics of the Actinides and Rare Earths

Moderators: Melissa Denecke, University of Manchester, UK, James G. Tobin, University of Wisconsin-Oshkosh

8:00am AC+AS+LS-TuM-1 Study of the Early Actinide Oxides and Fluorides – Systematics of the Electronic Structure, Thomas Gouder, R Eloirdi, R Caciuffo, European Commission - Joint Research Centre, Germany INVITED Oxidation properties of uranium have a great influence on the stability of nuclear waste. U^{VI} has

a 1000 times higher solubility in water than U^{IV}, and so the dissolution of UO₂ based spent fuel nuclear waste strongly depends on the oxidation state of the surface. Oxygen incorporation into the surface of UO₂ first results in the formation of U(V), found in UO_{2+x} and mixed valent oxides, and then formation of U^{VI}. But despite early reports on the existence of a pure U(V) phase (U₂O₅) there are no spectroscopic data on pure U(V). This may be due to its position between the highly stable UO₂ and UO₃. Also, apparent mixed surface phases containing U^{IV}, U^V and U^{VI} may result from the strong redox gradient between the oxidizing environment and the reduced bulk, present in most corrosion experiments. Replacing bulk samples by thin films allows avoiding this problem, by confining the reaction to a thin region where final conditions are reached everywhere simultaneously, keeping the system homogeneous. This opened the path for a systematic study of the evolution of the electronic structure of uranium upon oxidation and follow the outer shell configuration from 5f² (U^{IV}), to 5f¹ (U^V) to 5f⁰ (U^{VI}).

We will present electron spectroscopy (XPS, UPS and BIS) and Electron Energy Loss (ELS) study of U₂O₅ compare results to the neighbouring oxides (UO₂ and UO₃). U₂O₅ has been produced by exposing UO₂ successively to atomic oxygen, leading to UO₃, and to atomic hydrogen. Films have been deposited on polycrystalline gold foils (inert substrate). During gas exposure the samples were heated to 400°C to ensure fast diffusion and equilibrium conditions.

Determination of the U oxidation states was based on the characteristic U-4f core level satellites, separated from the main lines by 6, 8 and 10 eV for U^{IV}, U^V and U^{VI}, respectively. We managed producing films which showed exclusively a single 8 eV satellite, indicating the presence of pure U^V. The formation of U^V was confirmed by the intensity evolution of the U5f valence emissions. Also the linewidth of the XPS 5f line decreases from UO₂, with the 5f¹ final state multiplet, to U₂O₅, with a 5f⁰ final state singlet. The U-5d line also displays a multiplet structure due to interaction with the open 5f shell (U^{IV} and U^V). Evolution of the O2p/O1s intensity ratio indicates increasing covalence of the U-O bond in higher oxides.

U oxide spectra will be compared to spectra of ThO₂. Multiplet splitting and its dependence on the open shell was followed by comparing 5f² (UO₂) and 5f⁰ (ThO₂) systems. Finally XPS/BIS of the valence region will be presented for various oxides (UO₂, U₂O₅, UO₃ and ThO₂) and fluorides (UF₄ and ThF₄).

8:40am AC+AS+LS-TuM-3 Broadening of the XPS Spectra of U Oxides, Paul S. Bagus, University of North Texas; C Nelin, Consultant INVITED

It is common to attempt to improve the energy resolution of XPS in order to obtain more information about the electronic structure. However, it may not be possible to improve the resolution because unresolved final states are present. When the energy separations of these states are less than or comparable to the lifetime of the core-hole, it will not be possible to resolve the states and there will only be a broadening, often quite significant, for the observed peak. The unresolved features may arise from closely spaced multiplets for the angular momentum coupling of the open shell electrons and from excitations to higher lying vibrational levels for the final ionic states; the later is especially important when bond distances for the core-ionized states are very different from those for the initial state; see, for example, Ref. [1]. Thus, for example, the U(5f_{7/2}) peak of UO₂ has a FWHM of 1.4 eV although the instrumental resolution was 0.3 eV. [2] Similar large FWHM have been observed for U in different oxidation states. [3-4] In order to be able to relate the widths of these broadened features to the chemical and physical interactions in the system, it is necessary to understand the separate contributions of the multiplet splittings and the vibrational excitations. It has been shown that, for U(IV) 4f XPS in UO₂, the contributions of the multiplet splitting and the vibrational excitations are comparable, each contributing ~0.5 eV to the FWHM. [2] In the present

work, the contributions of these mechanisms are examined for U(IV), U(V), and U(VI) oxidation states. In addition, the broadening is examined for the XPS of different core levels where the relative importance of multiplet and vibrational broadening is different from that for the U(4f) XPS. Furthermore, the importance of temperature for the vibrational broadening is also examined. The theoretical predictions for these different parameters can be validated [4] paving the way to extract chemical information from the measured FWHM. [5] The theoretical framework for these predictions is based on wavefunctions for embedded cluster models of the oxides which allows for a balanced treatment of spin-orbit, angular momentum coupling, and covalent bonding.

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9:20am AC+AS+LS-TuM-5 Multiscale Characterization of Lanthanide and Actinide Nanoparticles Embedded in Porous Materials, Stefan Minasian, S Alayoglu, S Aloni, Lawrence Berkeley National Laboratory; J Arnold, University of California at Berkeley; E Batista, Los Alamos National Laboratory; A Braun, C Booth, A Herve, Lawrence Berkeley National Laboratory; Y Liu, University of California at Berkeley; L Moreau, Lawrence Berkeley National Laboratory; T Lohrey, J Long, M Straub, S Robin, D Russo, University of California at Berkeley; D Shuh, Lawrence Berkeley National Laboratory; J Su, P Yang, X Zhang, Los Alamos National Laboratory INVITED Decades of synthetic work has shown that desirable physical properties can be obtained by tailoring the size and composition of main-group and transition metal nanoparticles. The origin of these phenomena can be traced to a variety of factors that are intrinsic to nanoparticles, including but not limited to (1) quantization of the electronic states and (2) increasing contributions from nanoparticle surfaces. Despite this progress, very few methods provide the level of synthetic control needed to prepare lanthanide and actinide nanoparticles with well-defined sizes and compositions. Furthermore, many conventional methods of characterization are unable to determine the relative roles of volume and surface chemistry on physical behavior involving the 4f- and 5f-electrons.

We established syntheses for lanthanide and actinide nanoparticles by adapting a conceptual approach employed previously by Fischer and Suh to form d-block transition metal and main-group element nanoparticles in metal organic frameworks (MOFs). Implantation of lanthanide and actinide precursor molecules in covalent organic frameworks (COFs) was achieved by sublimation, followed by decomposition to form nanoparticles via exposure to reactive gases and/or elevated temperatures. A multi-pronged imaging and spectroscopic strategy was developed to fully characterize the complex multi-component materials. The characterization techniques are complementary in that they provide atomic to micron-scale probes, element and bulk-level selectivity, and measurements that are individual and statistically significant (Figure 1). Transmission electron microscopy (TEM) showed that the COF-5 templates limited UO₂ nanoparticle growth to between 1-3 nm, which is similar to the COF-5 pore size (2.7 nm). In addition, small-angle X-ray scattering (SAXS) was used to determine the nanoparticle form factor (for size and polydispersity) in a globally-averaged manner. Scanning transmission X-ray microscopy (STXM) was used to probe micron-scale morphology and to evaluate composition with X-ray absorption spectroscopy at the B, C, and O K-edges. Finally, X-ray absorption fine structure (XAFS) spectroscopy was used to determine composition and structure because it does not require long-range crystallographic order, and because it is capable of probing the nanoparticles selectively despite the substantial background from the organic host. This presentation will also discuss our efforts to show how 4f and 5f-electron behavior changes with variations in size and composition at the nanometer level.

11:00am AC+AS+LS-TuM-10 Multiple Forms of Uranium Hydrides and their Electronic Properties, Ladislav Havela, V Buturlim, E Chitrova, O Koloskova, P Minarik, M Cieslar, M Dopita, L Horak, M Divis, I Turek, Charles University, Prague, Czech Republic; D Legut, VSB-Technical University of Ostrava, Czech Republic; T Gouder, European Commission - Joint Research Centre, Germany

Although the published U-H binary phase diagram contains only the UH₃ phase, a recent progress in sputter deposition synthesis led to the

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discovery of UH_2 , which is with its CaF_2 structure type analogous to e.g. PuH_2 or YH_2 [1]. In addition, a high pressure synthesis yielded several U hyper-hydrides (as UH_3), which are predicted to be a high-temperature superconductors [2]. As both UH_3 (appearing in two structure modifications, α - and β - UH_3) and UH_2 allow alloying with selected transition metals, we have a whole spectrum of U-H hydrides and certain general features can be deduced, which have a serious implications for our understanding of actinides in general.

The striking fact that both UH_3 phases have, despite very different crystal structure, practically identical size of U moments and magnetic ordering temperature $T_C \approx 165$ K, which do not change even in the nanostructured state (grain size several nm) is underlined by the fact that both parameters only weakly depend on substantial alloying with Mo and other d -metals. UH_2 with substantially larger U-U spacing is a ferromagnet, as well, its $T_C \approx 125$ K. Structure of all those forms contains identical building blocks, H atoms inside U tetrahedra of invariable size, which are only set up into different patterns. The U-H bonding seems to be therefore an ingredient, which is more important than the conventional U-U spacing. The nature of the bonds is revealed by ab-initio calculations. One general feature is the U- $6d$ hybridization with the H-1s states, which leads to an electron transfer towards H. However, unlike rare-earth trihydrides, UH_3 remains metallic, UH_2 naturally, too. The $5f$ states largely released from the $5f-6d$ hybridization can therefore develop a strong ferromagnetism despite low U-U spacings. The transfer could be traced even in the real space using the Bader analysis.

The ab-initio techniques, which are successful in capturing the equilibrium volumes and U magnetic moments, are so far less successful in description of photoelectron spectra close to the Fermi level, which indicates importance of electron-electron correlations.

This work was supported by the Czech Science Foundation under the grant No. 18-02344S.

[1] L. Havela et al., Inorg. Chem. 57 (2018) 14727.

[2] I.A. Kruglov et al., Sci. Adv. 4 (2018) eaat9776.

11:20am **AC+AS+LS-TuM-11 Hafnium L-Edge X-ray Absorption Near Edge Structure Spectra Reveals Crystal Field Splitting**, *David Shuh*, *D Calder*, Lawrence Berkeley National Laboratory; *L Davis*, Pacific Northwest National Laboratory; *M Mara*, University of California at Berkeley; *C Booth*, Lawrence Berkeley National Laboratory; *J Darab*, *J Icenhower*, *D Strachan*, Pacific Northwest National Laboratory

One method for the safe disposal of actinides currently under consideration by the U.S. Department of Energy is immobilization of radionuclides in a glass or ceramic wastefrom that will subsequently be stored in a geological repository. To develop successful, stable wasteforms, it is necessary to acquire an understanding of the fundamental chemistry of the actinides (U, Pu, etc.) and neutron absorbers (B, Gd, Hf) that will be incorporated into the glass or ceramic matrix. The X-ray absorption near edge structure (XANES) at the hafnium L_3 and L_1 absorption edges has been investigated to elucidate the local structure of Hf in wastefrom glasses that are currently being considered as candidates for the immobilization of radionuclides. During our investigations of the structure of Hf, a neutron absorber and potential Pu(IV) surrogate, we noted an unusual feature at the white line maximum at the L_3 absorption edge: The examination of a variety of Hf model compounds demonstrates that the second derivative of the Hf L_3 XANES displays structure characteristic of crystal field splitting of the empty d -manifold. The peralkaline glasses studied exhibit L_3 XANES spectra typical of Hf in an octahedral crystal field. The L_1 XANES data corroborate these results, as the L_1 spectra of the glasses also resemble that of an octahedral Hf siloxane model compound, distinct from spectra of other standards measured.

11:40am **AC+AS+LS-TuM-12 Electrical Resistivity in Uranium-based Thin Films**, *Evgeniya Tereshina-Chitrova*, *L Havela*, *M Paukov*, *M Dopita*, *L Horak*, *M Cieslar*, Charles University, Prague, Czech Republic; *Z Soban*, Institute of Physics, Academy of Sciences of the Czech Republic, Czech Republic; *T Gouder*, *F Huber*, *A Seibert*, Joint Research Center, European Commission, Germany

Metallic $5f$ materials have a very strong coupling of magnetic moments and electrons mediating electrical conduction. It is caused by strong spin-orbit interaction together with involvement of the $5f$ states in metallic bonding. In this work we investigate the electrical resistivity $\rho(T)$ of various uranium-based thin films using the Van der Pauw method. Thin film samples of U-Mo and their hydrides were prepared by reactive sputter deposition [1]. The basic in-situ diagnostics was performed using a photoelectron

spectroscopy (XPS). The films were further characterized using x-ray diffraction (XRD) methods and Transmission Electron Microscopy (TEM). The $\rho(T)$ of the films studied correspond to known bulk systems of equivalent composition but provide much more variability of concentrations and geometries.

We studied superconductivity in the U-Mo films by measuring low-temperature resistivity down to 0.4 K. The 100 nm-thick $\text{U}_{0.79}\text{Mo}_{0.21}$ film shows a transition to the superconducting state at 0.55 K. The transition is wide and extends to 0.4 K. The upper critical field for the thin U-Mo film is ~ 1 T, i.e. much lower than for the bulk U-Mo obtained by splat cooling [2].

The hydrides of the U-Mo films had to be prepared using a cooled substrate (≈ 177 K). The study of the $(\text{UH}_3)_{0.74}\text{Mo}_{0.26}$ of estimated thickness 210 nm showed that it is a ferromagnet with the Curie temperature of ≈ 165 K, i.e. equivalent to bulk UH_3 , which underlines the general insensitivity of U hydrides to structure modifications. Overall temperature dependence is very weak and the absolute value of 2 m Ω cm, estimated from the nominal thickness, is very high for a metallic system.

The net negative resistivity slope in the U-Mo films and hydrides can be attributed to the randomness on atomic scale, yielding very strong scattering of electrons and weak localization. In the hydrides it is supported also by random anisotropy, randomizing local directions of U moments.

The work is supported by the Czech Science Foundation under the project #18-02344S. Part of the work was supported by "Nano-materials Centre for Advanced Applications," Project No.CZ.02.1.01/0.0/0.0/15_003/0000485, financed by ERDF.

[1] Gouder, R. Eloirdi, F. Wastin, E. Colineau, J. Rebizant, D. Kolberg, F. Huber, Phys.Rev. B **70**, 235108 (2004).

[2] N.-T. H. Kim-Ngan, S. Sowa, M. Krupka, M. Paukov, I. Tkach and L. Havela, Adv. Nat. Sci: Nanosci. Nanotechnol. **6**, 015007 (2015).

Actinides and Rare Earths Focus Topic

Room A215 - Session AC+AS+LS-TuA

Forensics, Science and Processing for Nuclear Energy

Moderators: Paul S. Bagus, University of North Texas, Tomasz Durakiewicz, National Science Foundation, David Geeson, AWE

2:20pm **AC+AS+LS-TuA-1 Helium Implantation Studies in Metals and Ceramics for Nuclear Energy Applications, Microstructure and Properties**, *Peter Hosemann, M Baloach, S Stevenson, A Scott*, University of California, Berkeley; *Y Yang*, Lawrence Berkeley Lab, University of California, Berkeley

INVITED

Helium damage in materials is a concern for fission and fusion materials. The development of Helium bubbles in a solid can lead to fundamental property changes ranging from embrittlement to surface blistering. Especially shallow ion beam implantation is of interest to the fusion community since the surface degradation of materials is one of the main materials degradation mechanism. This work features a rapid Helium implantation and screening method based on nanometer precise helium implantation using the helium ion beam microscope (HIM). This tool allows multiple doses in the same grain with subsequent rapid materials examination. We evaluate single and polycrystalline SiC, W, Cu, and Ti in this work and find interesting channelling phenomena which have been fit to modelling results from the literature. Further we evaluate mechanical and microstructural property changes using TEM, nanoindentation, AFM and micropillar compression testing on these alloys.

3:00pm **AC+AS+LS-TuA-3 Origin of Element Selectivity during Solvent Extraction of Rare Earths: Studies of Model Interfaces using Synchrotron Radiation**, *M Miller, Y Liang, H Li, M Chu, S Yoo*, Northwestern University; *W Bu*, University of Chicago; *M Olvera de la Cruz, Pulak Dutta*, Northwestern University

INVITED

Solvent extraction, commonly used to remove lanthanides from aqueous solutions, is known to preferentially extract heavier lanthanides. To understand the underlying nanoscale mechanisms, we studied a model system: extractant monolayers floating on an aqueous solution containing lanthanide ions. X-ray fluorescence near Total Reflection (XFNTR) was used to detect interfacial cations, using synchrotron radiation at the ChemMatCARS facility, Advanced Photon Source.

We found that the interfacial density of adsorbed cations increases very sharply as the solution concentration is increased, reminiscent of a first-order phase transition. The threshold concentration is an order of magnitude lower for erbium ($Z=68$) compared to neodymium ($Z=68$). When the solution contains both ions equally, only erbium is found at the extractant interface, and the bulk erbium concentration must be decreased by one order of magnitude relative to neodymium before both species appear equally at the interface.

These results indicate that element selectivity during liquid-liquid extraction occurs at the extractant-laden interface. Electrostatic effects arising from the dielectric mismatch, ionic correlations, and the sizes of the cations can be used to theoretically model the observed behaviors.

4:20pm **AC+AS+LS-TuA-7 Analysis of Aged of Uranium Particles via X-ray Xpctromicroscopy**, *Andrew Duffin, J Ward*, Pacific Northwest National Laboratory

INVITED

Uranyl fluoride (UO_2F_2) particles are a common by-product when uranium hexafluoride (UF_6), a volatile form of uranium used in industry, is released into air. Uranyl fluoride particles continue to react in the environment, eventually moving towards uranium oxide. Understanding the chemical changes uranyl fluoride takes is important as the chemical state affects how this contaminant is transported in the environment. We employed scanning transmission X-ray microscopy (STXM) to map the chemical changes that occur in single particles of uranyl fluoride, aged under controlled conditions. We have shown that ligand K-edge X-ray absorption spectroscopy can fingerprint uranium chemical species at the nm spatial scale and we have used this specificity to gain insight into the transformation of uranyl fluoride to uranium oxide.

5:00pm **AC+AS+LS-TuA-9 Heat Transfer and Phase Stability of Early Actinides and Actinide Compounds**, *Dominik Legut, L Kývála, U Wdowik, G Jaglo, P Piekarczyk*, Technical University of Ostrava, Ostrava, Czechia; *L Havela*, Charles University, Prague, Czechia

INVITED

Actinides and especially their carbides as prospective nuclear fuel materials for the generation IV reactors were investigated using the density

functional theory. We demonstrate that their electronic, magnetic, elastic, and thermal properties can be at present well described if the spin-orbit interaction and partial delocalization 5f electrons is properly included in the computational approaches. One can well reproduce not only basic electronic structure but also elastic constants, phonon dispersions, and their density of states, provided by XPS, UPS, BIS, and inelastic neutron scattering data [1-4]. The localization of the 5f electrons could be captured using a moderate value of the on-site Coulomb interaction parameter. The case studies include a realistic description of the ground-state properties of elemental metals as Th, U and their monocarbides ThC and UC. The realistic description of the electronic structure and lattice dynamics (phonons) explains much higher thermal expansion and heat capacity in pure actinides (as Th) comparing with respective actinide monocarbides. The modelling also gives an insight up to which temperature the heat transport depends on lattice vibrations and where the electron transport starts to dominate. We can identify the vibration modes influencing the heat transport most. Carbides are compared with actinide oxides, where the major role is played by optical phonon branches [3]. The phase stability and elastic properties have been explored in a cascade of uranium hydrides [5], for which the requirement to reproduce the ferromagnetic moments gives an important feedback for the choice of on-site Coulomb U and Hund's exchange J.

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5. L. Havela, M. Paukov, M. Dopita, L. Horak, D. Drozdenko, M. Divis, I. Turek, D. Legut, L. Kývála, T. Gouder, A. Seibert, F. Huber, Inorg. Chem. 57, 14727 (2018).

5:40pm **AC+AS+LS-TuA-11 Reactivity of Potential TRISO Fuel Barrier Layers (SiC and ZrN) with H₂O Probed with Ambient Pressure Photoelectron Spectroscopy**, *Jeff Terry, M Warren*, Illinois Institute of Technology; *R Addou, G Herman*, Oregon State University

While the use of TRISO fuels has been long postulated within High Temperature Gas Reactors, another potential use for the TRISO fuels is as an accident tolerant fuel in Light Water Reactors (LWRs). Before TRISO fuels can be used in LWRs, the corrosion properties of the different layers of TRISO fuels must be well understood. Photoelectron Spectroscopy (PES) has long been utilized to study the oxidation behavior of materials due to its sensitivity to both element and chemical state. The problem with PES has been that it has historically been a technique that required Ultrahigh Vacuum conditions for measurements. This made it difficult to study corrosion in situ. New instruments have expanded the capabilities of PES. It is now possible to measure photoemission spectra at ambient pressure. We have measured the in situ corrosion of a SiC layer grown as a TRISO simulant at a pressure of 1 mbar of H₂O at temperatures up to 500 C using an ambient pressure photoemission system. We see no oxidation of the SiC layer by water at temperatures up to 350 C. Above 350 C, the SiC begins to oxidize with the formation of SiO₂. In contrast, ZrN reacts at a pressure of 1 mbar of H₂O at room temperature. As the temperature increases, the ZrN layer is completely converted to ZrO₂. In the TRISO fuel, the barrier layer is surrounded by pyrolytic carbon. We model the protective ability of the outer carbon layer by making multilayers SiC/C and ZrN/C and measuring these under the same conditions. We find that a 2 nm carbon layer prevents the underlying barrier layers from reacting with water.

Frontiers of New Light Sources Applied to Materials, Interfaces, and Processing Focus Topic Room A210 - Session LS+AC+HC+SS-ThA

Emerging Methods with New Coherent Light Sources

Moderator: Germán Rafael Castro, Spanich CRG BM25-SpLine Beamline at the ESRF

4:00pm **LS+AC+HC+SS-ThA-6 Resolving X-ray Based Spectroscopies in the Sub-nanometer Regime: Enabling Atomic Scale Insights into CO Adsorption on Thin Film Surfaces, *Heath Kersell, B Eren, C Wu***, Lawrence Berkeley National Laboratory; *I Waluyo, A Hunt*, Brookhaven National Laboratory; *G Somorjai, M Salmeron*, Lawrence Berkeley National Laboratory

X-ray based spectroscopies routinely yield detailed elemental, chemical, electronic, and magnetic information on a wide array of physically and chemically diverse samples. However, the spatial resolution of these techniques is limited, frequently by the size of the X-ray spot. Conversely, certain structural probes readily resolve sample topography with nanoscale- or even atomic-resolution. The union of X-ray based spectroscopies with nanoscale structural probes enables the acquisition of spectroscopic information at unprecedented length scales. We will demonstrate the combination of X-ray based spectroscopies (e.g. X-ray photoelectron spectroscopy {XPS}) with scanning tunneling microscopy (STM), and its application to CO adsorption and oxidation on model catalyst surfaces.

CO adsorption on various crystal surfaces plays a critical role in numerous chemical processes, including for example CO oxidation, the water gas shift reaction, and methanol oxidation. CO oxidation is widely used as a prototype reaction for studies of fundamental catalytic phenomena and is crucial in exhaust gas processing for automobiles and stationary CO sources. Recent studies demonstrate strikingly high activity for CO oxidation by Pt nanoparticles supported on cobalt oxide (CoO_x) as compared to either of the constituent materials. In the further development of these catalysts, a deeper understanding of the active sites and their deactivation is crucial. Using a combination of *operando* high pressure STM (HP-STM) and ambient pressure XPS (AP-XPS), we investigate the nature of catalytically active sites for CO oxidation on CoO-Pt catalysts at CO and O₂ pressures up to 130 mTorr. Our experiments showed very different behavior for the lattice oxygen (O_{lat}...) in CoO between fully oxidized and sub-stoichiometric cobalt oxides. At RT, fully oxidized Co films adsorbed CO in the form of stable surface carbonate species, poisoning the reaction until reaching higher temperatures where they decomposed. On sub-stoichiometric CoO_x the CO oxidation reaction proceeded at RT, reducing the oxide to the metallic state. We discuss these results in the context of structural transformations observed *in-situ*- via HP-STM, and demonstrate the behavior of surface sites under relevant gas mixtures.

As an outlook, we will discuss various *in-situ* multi-modal approaches which enhance the spatial resolution of X-ray based spectroscopies toward the nano- or even single atom scales. Such a union of spectroscopic and structural probes will provide a more accurate and complete picture of operating devices in the near future.

4:20pm **LS+AC+HC+SS-ThA-7 Imaging with XPS: Advanced Characterization for Advanced Materials and Devices, *Tatyana Bendikova, H Kaslasi, E Sanders, E Joselevich, D Cahen***, Weizmann Institute of Science, Israel

X-ray Photoelectron Spectroscopy (XPS), as a surface sensitive technique with the sensitivity down to single atomic layer, provides unique information about elemental composition and chemical and electronic states of elements in the material. For some research goals, however, this knowledge is not sufficient as it does not provide the entire information required for a comprehensive characterization of the investigated system. In addition to the basic functions of standard XPS, our instrument is equipped with advanced capabilities such as XPS imaging, which is particularly valuable in the analysis of patterned or inhomogeneous specimens. Following image acquisition, specific areas can thus be chosen and small spot XP spectra acquired at sites of particular interest. This information is useful in the characterization of patterned surfaces or inhomogeneous samples with surface features between several to hundreds of micrometers.

We present here two examples where XPS imaging is successfully used providing crucial information for understanding the investigated systems.

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In the first example bunches of GaN nanowires (50-100 nm each) randomly spread on Si substrate were monitored with XPS imaging. Then, focusing on the GaN bunch itself, small area XP spectra were obtained. This allowed to get precise top surface composition of the bunches significantly consuming the analysis time.

In the second example variations in chemical composition though dimensions of the Cs_xMa_{1-x}PbBr₃ (MA = CH₃NH₂)

crystal were studied using XPS imaging. Significant changes in the N/Cs ratio, depending on the distance from the crystal edge/center, were observed on the top surface. Variations in the N/Cs and Pb/(N+Cs) ratios were also observed along the crystal bulk.

4:40pm **LS+AC+HC+SS-ThA-8 Time-Resolved Photoemission with Free-Electron Lasers, *Kai Rossnagel***, CAU Kiel / DESY, Germany **INVITED**

Photoelectron spectroscopy is an essential analytical tool for learning about the properties and workings of quantum materials and functional interfaces, in which electrons are the main actors. In practice, photoelectron spectroscopy is a toolbox comprising three major techniques, where the momentum selectivity and atomic-site specificity of valence and core electron emissions are exploited, respectively: Angle-resolved photoelectron spectroscopy (ARPES) is the most powerful imaging technique for the energy-momentum space of the active electrons near the Fermi level, while x-ray photoelectron spectroscopy (XPS) is a universal tool for chemical analysis and x-ray photoelectron diffraction (XPD) an established surface structural probe. A dream is to combine all three techniques into a single experiment, make it complete by adding spin and femtosecond time resolution, and thus be able to shoot femto-stroboscopic movies of intertwined electronic, magnetic, chemical, and geometric structure dynamics and gain previously unachievable, direct "in operando" insight into dynamic structure-function relationships of materials and interfaces. Here, we aim to realize this dream by combining the soft x-ray SASE3 free-electron-laser (FEL) beam at the European XFEL with the most advanced photoelectron detection scheme currently available: the time-of-flight momentum microscope with efficient 3D energy-momentum detection and 2D spin filtering. The status of the project and of FEL-based photoelectron spectroscopy in general will be presented.

5:20pm **LS+AC+HC+SS-ThA-10 Ultrafast Magnetization Dynamics on the Nanoscale, *Bastian Pfau***, Max Born Institute, Germany **INVITED**

Nanometer-scale spin configurations are attractive as information entities for spintronic applications to realize nonvolatile and energy-efficient data storage and processing. In recent years, this research field was stimulated by the discovery that the spin can be effectively manipulated using ultra-short light pulses exciting suitably designed magnetic materials. Scattering and imaging methods based on sources delivering ultra-short x-ray pulses are particularly successful in revealing the magnetization dynamics on the relevant time and length scales. I will present research results on optically induced demagnetization and formation of nanoscale magnetic domains and skyrmions in Co-based multilayer systems. We investigate these processes using small-angle scattering signals or direct imaging via holography with femtosecond x-ray pulses delivered by free-electron laser sources. These methods additionally allow to address the influence of lateral nanoscale inhomogeneity and to work with laterally localized or structured excitation.

Frontiers of New Light Sources Applied to Materials, Interfaces, and Processing Focus Topic Room A210 - Session LS+AC+NS-ThA

Photon Science for Imaging Materials from the Meso- to the Nanoscale

Moderator: Maya Kiskinova, Elettra-Sincrotrone Trieste, Italy

2:20pm **LS+AC+NS-ThA-1 Triplet Dynamics in Photovoltaic Materials Measured with Time Resolved X-Ray Spectroscopies, *R Costantini***, University of Trieste; *R Faber*, Technical University of Denmark; *A Cossaro, A Verdini, L Floreano*, CNR - Istituto Officine Materiali; *C Haettig*, Ruhr-University Bochum, Germany; *A Morgante*, University of Trieste, Italy; *S Coriani*, Technical University of Denmark; *Martina Dell'Angela*, CNR - Istituto Officine Materiali, Italy **INVITED**

Singlet exciton fission (SF) is a multiexciton generation process in organic molecules, where an optically excited singlet exciton is converted into two triplet excitons. The exploitation of this mechanism has been shown

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capable of boosting the efficiency of solar energy conversion, and it has been proposed as a mean for exceeding the Shockley-Queisser limit of efficiency of solar cells. In the last decade, several studies have investigated different chromophores to identify the ones suitable to produce high yield SF and long living triplets. Such studies spanned from the fundamental to the applicative approach, also dealing with the optimization of the interfaces with the other materials in the device in order to achieve an overall increased efficiency of the charge transport. In particular, the study of the dynamics of the triplet states, when formed and transported across all the interfaces, is crucial for modelling the charge transport properties in a working device. Here we present a new experimental approach to measure the triplet dynamics at the picosecond timescale, that uses the advantage of chemical sensitivity with respect to conventional optical techniques, thus offering the possibility of tracking the dynamics of the triplet states across different materials. We exploit the chemical selectivity of X-ray absorption spectroscopy (XAS) in an optical pump/X-ray probe experiment at a pump-probe setup that we developed at the Elettra synchrotron and with the support of novel implemented calculation methodologies. We studied triplet dynamics in pentacene thin films (the prototypical singlet fission material) with lifetime of about 300ps.

3:00pm **LS+AC+NS-ThA-3 Synchrotron X-Ray Tomography to Understand Structure and Physical Transformations in Solid State Batteries, Kelsy Hatzell, M Dixit, Vanderbilt University** **INVITED**

The increasing demand for portable electronics, stationary storage, and electric vehicles is driving innovation in high-energy density batteries. Solid electrolytes that are strong enough to impede lithium dendrite growth may enable energy dense lithium metal anodes. Currently, the power densities of all-solid state batteries is limited because of ineffective ion transport and chemical and physical decomposition at solid|solid interfaces. The nature of ionic transport at intrinsic and extrinsic interfaces is important for mitigating chemical and structural instabilities. Extrinsic interface instabilities are responsible for high interfacial resistances. In order to displace liquid electrolytes, new materials and engineering strategies need to be developed to negate these degradation pathways. New insight into the governing physics that occurs at these interfaces are critical for developing engineering strategies for the next generation of energy dense batteries [1,2]. However, buried solid|solid interfaces are notoriously difficult to observe with traditional bench-top and lab-scale experiments. In this talk I discuss opportunities for tracking phenomena and mechanisms in all solid state batteries *in-situ* using advanced synchrotron techniques. Synchrotron techniques that combine reciprocal and real space techniques are best equipped to track relevant phenomena with adequate spatial and temporal resolutions.

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— G —
Gan, J: AC-MoA-1, 3
Gofryk, K: AC-MoA-5, 3; AC-MoA-8, 3
Gouder, T: AC+AS+LS-TuM-1, 5; AC+AS+LS-TuM-10, 5; AC+AS+LS-TuM-12, 6

— H —

Haettig, C: LS+AC+NS-ThA-1, 8
Harp, J: AC-MoA-5, 3
Hatzell, K: LS+AC+NS-ThA-3, 9
Havela, L: AC+AS+LS-TuA-9, 7; AC+AS+LS-TuM-10, 5; AC+AS+LS-TuM-12, 6
He, L: AC-MoA-1, 3
Herman, G: AC+AS+LS-TuA-11, 7
Herper, H: AC+LS+MI-MoM-10, 2
Herve, A: AC+AS+LS-TuM-5, 5
Horak, L: AC+AS+LS-TuM-10, 5; AC+AS+LS-TuM-12, 6
Hosemann, P: AC+AS+LS-TuA-1, 7
Hua, Z: AC-MoA-1, 3
Huber, F: AC+AS+LS-TuM-12, 6
Huhtinen, H: AC+LS+MI-MoM-11, 2
Hunt, A: LS+AC+HC+SS-ThA-6, 8
Hurley, D: AC-MoA-1, 3
— I —
Icenhower, J: AC+AS+LS-TuM-11, 6
Inbanathan, F: AC+LS+MI-MoM-11, 2
— J —
Jadwisieniczak, W: AC+LS+MI-MoM-11, 2
Jaglo, G: AC+AS+LS-TuA-9, 7
Joselevich, E: LS+AC+HC+SS-ThA-7, 8
— K —
Kaslasi, H: LS+AC+HC+SS-ThA-7, 8
Kersell, H: LS+AC+HC+SS-ThA-6, 8
Khafizov, M: AC-MoA-1, 3
Koloskova, O: AC+AS+LS-TuM-10, 5
Kroll, T: AC+LS+MI-MoM-9, 1
Kývala, L: AC+AS+LS-TuA-9, 7
— L —
Labouriau, A: AC-MoA-6, 3
Lee, K: AC-MoA-6, 3
Legut, D: AC+AS+LS-TuA-9, 7; AC+AS+LS-TuM-10, 5
Li, H: AC+AS+LS-TuA-3, 7
Liang, Y: AC+AS+LS-TuA-3, 7
Liu, Y: AC+AS+LS-TuM-5, 5
Lohrey, T: AC+AS+LS-TuM-5, 5
Long, J: AC+AS+LS-TuM-5, 5
— M —
Mann, J: AC-MoA-8, 3
Mann, M: AC-MoA-1, 3
Mara, M: AC+AS+LS-TuM-11, 6
Martínez, R: AC+LS+MI-MoM-11, 2
McLean, B: AC-MoA-3, 3
Medina-Ramirez, I: AC-MoA-9, 3
Miller, M: AC+AS+LS-TuA-3, 7
Minarik, P: AC+AS+LS-TuM-10, 5
Minasian, S: AC+AS+LS-TuM-5, 5
Moreau, L: AC+AS+LS-TuM-5, 5
Morgante, A: LS+AC+NS-ThA-1, 8
Murphy, E: AC-MoA-6, 3
— N —
Nelin, C: AC+AS+LS-TuM-3, 5
Nelson, A: AC-MoA-3, 3
Nordlund, D: AC+LS+MI-MoM-9, 1
Nowak, S: AC+LS+MI-MoM-9, 1

— O —

Olvera de la Cruz, M: AC+AS+LS-TuA-3, 7
— P —
Palai, R: AC+LS+MI-MoM-11, 2
Paukov, M: AC+AS+LS-TuM-12, 6
Pfau, B: LS+AC+HC+SS-ThA-10, 8
Piekarz, P: AC+AS+LS-TuA-9, 7
Poudel, N: AC-MoA-5, 3; AC-MoA-8, 3
— R —
Robin, S: AC+AS+LS-TuM-5, 5
Rossnagel, K: LS+AC+HC+SS-ThA-8, 8
Russo, D: AC+AS+LS-TuM-5, 5
— S —
Salmeron, M: LS+AC+HC+SS-ThA-6, 8
Sanders, E: LS+AC+HC+SS-ThA-7, 8
Scott, A: AC+AS+LS-TuA-1, 7
Seibert, A: AC+AS+LS-TuM-12, 6
Sen, A: AC-MoA-1, 3
Shuh, D: AC+AS+LS-TuM-11, 6; AC+AS+LS-TuM-5, 5
Soban, Z: AC+AS+LS-TuM-12, 6
Sokaras, D: AC+LS+MI-MoM-9, 1
Somorjai, G: LS+AC+HC+SS-ThA-6, 8
Stanford, J: AC-MoA-3, 3
Stevenson, S: AC+AS+LS-TuA-1, 7
Strachan, D: AC+AS+LS-TuM-11, 6
Straub, M: AC+AS+LS-TuM-5, 5
Su, J: AC+AS+LS-TuM-5, 5
— T —
Talley, S: AC-MoA-6, 3
Tereshina-Chitrova, E: AC+AS+LS-TuM-12, 6
Terry, J: AC+AS+LS-TuA-11, 7
Thunström, P: AC+LS+MI-MoM-10, 2
Tobin, J: AC+LS+MI-MoM-9, 1
Turek, I: AC+AS+LS-TuM-10, 5
— V —
Vekilova, O: AC+LS+MI-MoM-10, 2
Verdini, A: LS+AC+NS-ThA-1, 8
— W —
Waluyo, I: LS+AC+HC+SS-ThA-6, 8
Ward, J: AC+AS+LS-TuA-7, 7
Warren, M: AC+AS+LS-TuA-11, 7
Wdowik, U: AC+AS+LS-TuA-9, 7
Weng, T: AC+LS+MI-MoM-9, 1
Wharry, J: AC-MoA-1, 3
Wiss, T: AC-MoA-1, 3
Wu, C: LS+AC+HC+SS-ThA-6, 8
— Y —
Yang, F: AC+LS+MI-MoM-3, 1
Yang, P: AC+AS+LS-TuM-5, 5
Yang, Y: AC+AS+LS-TuA-1, 7
Yao, T: AC-MoA-1, 3; AC-MoA-5, 3
Yoo, S: AC+AS+LS-TuA-3, 7
Yu, S: AC+LS+MI-MoM-9, 1
— Z —
Zapien, J: AC-MoA-9, 3
Zhang, X: AC+AS+LS-TuM-5, 5
Zieve, R: AC+LS+MI-MoM-5, 1
Zwicknagl, G: AC+LS+MI-MoM-8, 1