Actinides and Rare Earths
Room 320 - Session AC-ThM

Actinides and Rare Earths Early Career

Moderators: Dr. MIles Beaux, LANL, **Dr. Krzysztof Gofryk**, Idaho National Laboratory, **Prof. Itzhak Halevy**, Ben Gurion Uni. Be'er Sheva, **Prof. Dr. Gertrud Zwicknagl**, Technical University Brauschweig

11:00am **AC-ThM-13 Scanning Tunneling Spectroscopy Reveals Extreme Stoichiometric Sensitivity in Actinide Oxides***, **Benjamin Heiner**, Miles Beaux,* Los Alamos National Laboratory

With the recent availability of single crystals, we have characterized the surface of Np2O5 using scanning tunneling spectroscopy (STS) at cryogenic temperatures. In addition to the band gap, two electronic features were observed on the surface: finite electron density in the previously observed band gap and a semimetallic suppression of the density of states at the Fermi Energy. These were unexpected and unpredicted, but using a proven theoretical model, we attribute these behaviors to areas of the crystal surface with variable oxygen vacancies. The model predicts that as oxygen vacancies are added to the calculation, the neptunium 5f orbital density fills in the band gap of pure Np2O5 until the crystal eventually becomes semimetallic. This work contextualizes the characterization of plutonium substoichiometric sesquioxide by STS, which contradicts calculation of plutonium oxides to be semiconductors. That the band gap of Np2O5 collapses into semimetallic behavior leads us to hypothesize that stoichiometric plutonium oxides would also have band gaps, but the electronic structure of actinide oxides are sensitive to even slight deviations to atomic ratios.

LA-UR-26-23359

11:15am **AC-ThM-14 Structural and Magnetism of Lanthanide- and Actinide-based Pyrochlores***, **Binod Rai**, Gia Thinh Tran,* Savannah River National Lab

Pyrochlore oxides are of significant interest because of their structural flexibility, radiation tolerance, and complex magnetic behavior arising from geometric frustration. In lanthanide- and actinide-based pyrochlores, the corner-sharing tetrahedral network promotes competing magnetic interactions that can lead to unconventional magnetic ground states and emergent quantum phenomena. These materials are also promising candidates for nuclear waste immobilization due to their ability to accommodate radionuclides while maintaining structural stability.

In this work, we investigate the structural, magnetic, and thermodynamic properties of the solid-solution series $(Tb_{1-x}Er_x)_2Ti_2O_7$. The end-member compounds exhibit contrasting magnetic behaviors: $Tb2Ti_2O_7$ remains magnetically disordered at very low temperatures despite strong antiferromagnetic interactions, whereas $Er2Ti_2O_7$displays antiferromagnetic ordering. By systematically varying the Tb/Er ratio, we examine how competing anisotropies and exchange interactions influence the evolution of magnetic frustration and phase behavior. Structural characterization is correlated with magnetic susceptibility and heat-capacity measurements to establish composition-dependent trends. In addition, recent results on actinide-based pyrochlores will be presented, highlighting their structural stability and magnetic properties relevant to strongly correlated 5f-electron systems and nuclear materials applications.

11:30am **AC-ThM-15 Role of Surface Oxidation in the Interdiffusion Behavior of Uranium-Niobium Diffusion Couples***, **Michelle Greenough**, Joseph Boro, Adrian Gonzales, W. Preston Cole, Debra Rosas, Erik Oerter, Scott Donald, Tien Roehling, Tae Wook Heo,* Lawrence Livermore National Laboratory

Understanding uranium oxidation behavior under controlled environmental conditions is important for predicting the stability and long-term performance of uranium-based materials. In this study, oxidized pieces of uranium were then incorporated into uranium-niobium diffusion couples to serve as a system for evaluating the impact of oxide chemistry and thickness on uranium-niobium diffusion. Both the oxidized specimens and U-Nb diffusion couples were characterized using Auger electron spectroscopy (AES), X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), and electron probe microanalysis (EPMA). These complementary techniques allowed for correlation between oxide characteristics and the nature of the formed diffusion couple. Oxide chemistry and thickness were demonstrated to influence the resulting diffusion material after testing several different relative humidities.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and was supported by the LLNL-LDRD Program under Project No. 25-ERD-005.

11:45am **AC-ThM-16 Laboratory-Based X-Ray Absorption Spectroscopy of Actinide Materials***, David Shuh, Alexander Ditter,* Lawrence Berkeley National Laboratory (LBNL)*; **Haisley Windsor**,* Lawrence Berkeley Lab, University of California, Berkeley

Synchrotron radiation spectroscopy of actinide materials has led to marked improvement in the fundamental understanding of their *f*-electron chemistry and physics. Beamlines and endstations with unique spatial resolution, energy resolution, and intensity characteristics have made it possible to determine the chemical and physical properties of a range of actinide materials. The recent development of tunable laboratory-based x-ray instruments with sufficient performance, albeit not fully to the level of synchrotron radiation facility beamlines/endstations, are beginning to provide a reasonable alternative for x-ray measurements of actinide materials. The first results from the LBNL Sigray QuantumLeap x-ray absorption spectroscopy (XAS) spectrometer will be presented.

At the same time as interest and capabilities are growing in laboratory-based actinide XAS, there has been already been great activity in the tender x-ray region for high-resolution studies of actinide materials. As synchrotron radiation light source upgrades get underway at the Advanced Light Source (ALS-U) and at the National Synchrotron Light Source II (NSLS IIU), it is worthwhile to assess and present the possible impact of new, fully-optimized tender x-ray beamline/endstation capabilities on the field of actinide science conducted in energy region with respect to current capabilities.

12:00pm **AC-ThM-17 Scanning Tunneling Spectroscopy of Pu Compound Crystals***, **Miles Beaux**, Benjamin Heiner,* Los Alamos National Laboratory*; Andrew Yost,* Scienta Omicron*; William DeBenedetti, Filip Ronning, Eric Bauer, David Arellano, Derek Prada, Paul Tobash,* Los Alamos National Laboratory

Scanning tunneling spectroscopy (STS) has been performed on a variety of Pu compound crystals including PuB_4, $PuCoGa_5$, and $PuIn_3$, with the results revealing distinct characteristics of semiconductive, semimetallic, and metallic, respectively. Specifically, the STS measurement showed the PuB_4 to have a band gap of approximately 0.042 eV, $PuCoGa_5$ to have no band gap with an electron density of zero at (and only at) the Fermi energy (E_F), and $PuIn_3$ to have no band gap with electron density at E_F. STS measurements were performed at temperatures ranging from 25 Kelvin (above the supercritical transition temperature of $PuCoGa_5$) to room temperature. In addition to the STS results, structural scanning tunneling microscopy (STM) images were successfully obtained for the $PuIn_3$ and $PuCoGa_5$ surfaces. STM images of the $PuIn_3$ surface revealed step edges of approximate multiples of 0.8 nm consistent with a 111-surface orientation. STM images of the $PuCoGa_5$ revealed two distinct Moiré patterns consistent with different faces of the crystal structure.

Actinides and Rare Earths
Room Ballroom A - Session AC-ThP

Actinides and Rare Earths Poster Session

AC-ThP-1 The Application of Occam's Razor to Bremsstrahlung Isochromat Spectroscopy*, J G Tobin,* University of Wisconsin - Oshkosh

In philosophy and science, Occam's Razor is the problem-solving principle that recommends searching for the simplest possible explanation, consistent with the facts. [1] As put forth by Ptolemy (circa AD 90 -180): "We consider it a good principle to explain the phenomena by the simplest hypothesis possible." [2] More colloquially, "When you hear hoofbeats, think of horses not zebras," as suggested by Theodore Woodward. [3] In the case of Bremsstrahlung Isochromat Spectroscopy (BIS), [4-9] there have been many different interpretation methods based upon different theoretical models. Here, a simple picture will be presented, founded upon one underlying principle: the 5f electrons of the actinides are a slightly perturbed jj-coupled system. [10-15] This simple picture will include (1) the demonstration that many uranium systems are 5f3-localized and (2) a model for 5f mixing and delocalization. [16 -17] ACKNOWLEDGEMENTS: JGT wishes to thank the University of Wisconsin - Oshkosh for its on-going support. It is a gem set beside the Fox River and Lake Winnebago. References1.https://en.wikipedia.org/wiki/Occam%27s_razor2.Franklin, James (2001). The Science of Conjecture: Evidence and Probability before Pascal. The Johns Hopkins University Press. Chap 9. p. 2413.Sotos, John G. (2006) [1991]. Zebra Cards: An Aid to Obscure Diagnoses. Mt. Vernon, VA: Mt. Vernon Book Systems. ISBN 978-0-9818193-0-3.4.J.K. Lang and Y. Baer, Rev. Sci. Instrum. 50, 221 (1979).5.Y. Baer and J. Schoenes, Solid State Commun. 33, 885 (1980).6.Y. Baer and J.K. Lang, Phys. Rev. B 21, 2060 (1980).7.Y. Baer, Physica 102B, 104-110 (1980).8.E. Wuilloud et al., Phys. Rev. B 29, 5228 (1984).9.Y. Baer, "Electron Spectroscopy Studies," Chapter 4, "Handbook of the Physics and Chemistry of the Actinides," eds. A.J. Freeman and G.H. Lander, North Holland, Amsterdam (1984)10.J. G. Tobin et al., Phys. Rev. B 105, 125129 (2022).11.J.G. Tobin et al., Solid State Sciences 160, 107779 (2025).12.J. G. Tobin, "An Empirical Analysis of alpha-U Bremsstrahlung Isochromat Spectroscopy," MRS Advances 7, 783-788 (2022).13.J.G. Tobin et al., Phys. Rev. B 72, 085109 (2005).14.A. L. Kutepov, J. G. Tobin, S.-W. Yu, B. W. Chung and P. Roussel, J. of Phys.: Cond. Matter, 36, 045601 (2024).15.J. G. Tobin et al., MRS Bulletin 47, 1078–1083 (2022).16.J.G. Tobin, J. Vac. Sci. Technol. A 43, 063206 (2025).17.J.G. Tobin and A. Kutepov, J. Electron Spect. Rel. Phen. 284, 147577 (2026).

AC-ThP-2 Uranium 2D Ferromagnetism at Ambient Temperatures in Layered Zintl Phase UCu_2P_2*, Ladislav Havela,* *Alexandre Kolomiets, Volodymyr Buturlim,* Charles University, Czechia*; Fuminori Honda,* Kyushu University, Japan*; Fabrice Wilhelm,* ESRF Grenoble, France*; Jiri Prchal,* Charles University, Czechia*; Andrei Rogalev,* ESRF Grenoble, France

Light-actinide magnetism is based on 5*f* electronic states, which are typically involved in metallic bonding, in contrast with the localized 4*f* states in lanthanides. Hence they are rather an analogy to transition-metal 3*d* materials, characterized by itinerant character. There is, however, a special ingredient in actinides, the strong spin-orbit interaction. It gives rise to orbital magnetic moments, yielding the strong anchor to the crystal lattice, providing thus new functionalities, as enormous magnetocrystalline anisotropy or giant magnetoresistance effects.

However, weaker Coulomb *U* restricts the 5*f* magnetism to the low temperature range. Combining with 3*d* metals, efficiently enhancing ordering temperatures in compounds with rare earths, does not work with actinides, as the 3*d* and 5*f* band states coexist at similar energies and hybridize between each other, suppressing typically both 3*d* and 5*f* magnetism.

There exists an alternative route of further reducing the 5*f* delocalization in compounds with large U-U spacings and weak 5*f*-ligand hybridization. The trick is to use polar bonding to reduce also the 5*f*-6*d* hybridization by withdrawing the U-6*d* states from the energy range of 5*f*'s by the bonding to an anion in Zintl phases. The most successful case is UCu_2P_2, a layered compounds with trigonal crystal structure, formed by alternating slabs of U (cation) and Cu-P (polyanion). The interactions can be further tuned by hydrostatic pressure, increasing the Curie temperature from 216 K to 296 K at pressures reaching 15 GPa. To describe variations of Curie temperature and spin and orbital moments, we applied X-ray magnetic circular dichroism at U-$M_{4,5}$ edges (ID 12 at ESRF) using perforated diamond anvil cells, minimizing the absorption in diamond. The results are in a good agreement with ab-initio calculations by means of the GGA+*U* method, which reveal other surprising features of the compound.

This work was supported by the Czech Science foundation under grant No. 25-16339S.

AC-ThP-3 Recent Studies of Actinide Materials by Soft X-Ray Synchrotron Radiation Methods*, David Shuh,* Lawrence Berkeley National Laboratory*; Haisley Windsor, Cambell Conour,* Department of Chemistry, University of California Berkeley*; Emma Archer, Nic Cicchetti, Olivia Gunther,* Lawrence Berkeley National Laboratory*; Jennifer Wacker,* Lawrence Berkeley National Lab*; Sergei Butorin,* Uppsala University, Sweden*; Polly Arnold,* Department of Chemistry, University of California Berkeley*; Hendrik Ohldag, Matthew Marcus,* Lawrence Berkeley National Laboratory

The capability to conduct x-ray synchrotron radiation spectroscopy on actinide materials has led to marked improvement in the fundamental understanding of *f*-electron behavior and chemistry of *f*-electron materials. Specialized beamlines and endstations capable of utilizing the unique spatial resolution, energy resolution, and intensity provided by the advent of near and third generation synchrotron radiation x-ray beams from storage ring user facilities, coupled to new sample holding techniques, has provided unmatched opportunities to characterize the chemical and physical properties for a range of actinide materials. This has been particularly evident in the soft x-ray region of synchrotron radiation where the scanning transmission x-ray microscope (STXM) has been used to determine electronic structure from the ligand perspective via ligand K-edge spectroscopy and resonant inelastic x-ray scattering (RIXS) has been employed to garner information about actinide metal centers Both have emerged as uniquely powerful tools to elucidate the electronic structure of actinide materials.

The results of recent actinide soft x-ray STXM and RIXS studies will be presented. These will include recent studies of the actinyl tris(benzoate) complexes with a range of donating/withdrawing substituents studied using scanning transmission x-ray microscopy (STXM) and time dependent density functional theory (TDDFT). The results from RIXS studies of the Am oxides will be recapped and prospects for future studies highlighted. Lastly, an update on the status of the laboratory-based x-ray absorption system used for f-element studies at LBNL will be presented.

AC-ThP-5 5f Electronic Structure of van der Waals Uranium-Tellerium Alloys*, Jonathan Denlinger,* Lawrence Berkeley National Laboratory*; Christopher Broyles, Justin Shotton, Sheng Ran,* Washington University, St. Louis

Low dimensionality of magnetism and of correlated f-electron physics are current high interest research topics in condensed matter. Here, we compare and contrast the properties of two layered van der Waals uranium alloys with a focus on the diversity of their U 5f electronic structures measured by angle-resolved photoemission (ARPES), and on new f-spectroscopic features not previously reported.

UOTe crystallizes in (Te-U-O-U-Te) quintuplet layers and exhibits bilayer (uddu) antiferromagnetic ordering of local moments below 165K [1]. The U 5f states are found to be well localized at ~1 eV below E_F similar to UO_2, but with an additional weak satellite peak at -0.2 eV of unexplained origin that exhibits a Kondo-like temperature dependence. Moreover, the AFM ordering has a distinct effect on the relative energy alignment of the localized f-states between the zone center and the zone boundary. ARPES of UOTe also exhibits unusual superstructure replicas of the Fermi surface that result from an exotic incommensurate structural modulation of its vdW gap between Te layers.

The UTe_3 crystal structure contains two Te square-net planes that ubiquitously promote charge density wave (CDW) formation in the rare-earth tritellurides. Here strong U 5f character near E_F with strong temperature-dependent Kondo coherence is found in a shallow electron pocket whose high density of states is consistent with its ferromagnetic ordering below 20K [2]. The strong f-scattering dominates the otherwise delicate imperfect Fermi surface nesting of light mass Te p-bands, that results in the suppression of CDW ordering. Finally, an unusual momentum-dependent 'negative' spectral weight profile for zone-folded Te p-bands is observed and discussed as an interference phenomena.

[1] C. Broyles *et al.*, Adv. Mater. **37**, 2414966 (2025).
[2] J. Shotton *et al.*, arXiv: 2603.03509 (2026).

AC-ThP-6 Development of a Novel Aerogel-Assisted Ssntd for Automated Fissile Isotope Identification, *Rami Babayew, Yaacov Yehuda-Zada,* Nuclear Research Center Negev, Israel*; Galit Bar,* Soreq Nuclear Research Center, Israel*; Noam Elgad,* Nuclear Research Center Negev, Israel*; Danny Dayan,* Ben Gurion University Be'er Sheva, Israel*; Jan Lorincik,* Centre Řež, Czech Republic*; Itzhak Orion,* Ben Gurion University Be'er Sheva, Israel*; Shay Dadon,* Nuclear Research Center Negev, Israel*; Aryeh Weiss,* Bar Ilan University, Israel*; Galit Katarivas Levy,* ***Itzhak Halevy,*** Ben Gurion University Be'er Sheva, Israel

Accurate identification of fissile isotopes is a central challenge in nuclear forensics and safeguards. Fission Track Analysis (FTA) using solid-state nuclear track detectors (SSNTDs) provides high sensitivity but is limited by manual processing and restricted isotopic discrimination. This work presents an automated, physics-informed framework for fissile isotope characterization combining Monte Carlo simulations, synthetic data generation [1], and image-based analysis.

GEANT4 simulations were performed for ^{235}U, ^{233}U, and ^{229}Th to model fission fragment transport and cluster formation in LEXAN® SSNTDs with silica aerogel spacers. Synthetic track images were generated to study the effect of aerogel thickness on cluster morphology. Results show that increasing aerogel thickness enhances cluster separation and improves signal-to-noise ratio up to an optimal range, beyond which performance saturates due to pixel-resolution constraints and track truncation effects.

An automated image-processing pipeline (FTA-Finder) was developed to identify fission sites, separate overlapping clusters, and suppress background tracks. The fraction of well-resolved clusters increases with aerogel thickness, indicating improved reconstruction robustness.

Isotopic discrimination was evaluated using reconstructed track-length histograms and double-Gaussian fits for heavy and light fission fragments. A complementary analysis based on Real Flight Path (RFP) distributions showed clear isotope-dependent peak shifts. For 0.1 μm grains embedded in 50 μm aerogel (ρ = 0.033 g/cm^3), ^{235}U exhibited higher RFP peak positions than ^{233}U, while ^{229}Th showed the lowest values due to differences in fission-fragment energy and mass distributions.

Statistical analysis over 30 synthetic realizations showed significant separation between ^{235}U and ^{229}Th at ~3σ, while discrimination between ^{235}U and ^{233}U approached the resolution limit (~1σ). Peak deviations remained below 0.69%, confirming stable reconstruction performance.

All results are based on simulation and synthetic data. Future work will focus on experimental validation using neutron irradiation in reactor environments and extension to additional detector configurations. The proposed framework provides a scalable pathway toward quantitative isotopic fingerprinting for nuclear forensic and safeguards applications.

Reference:

[1] R. Babayew, Y. Yehuda–Zada, N. Elgad, J. Lorince, I. Orion, A. Weiss, G. Katarivas Levy, I. Halevy, *Simulation tools for improvement of the fission track analysis method for nuclear forensics* (2024), JRNC, Akadémiai Kiadó, Budapest, Hungary, DOI: 10.1007/s10967-023-09313-5.

Actinides and Rare Earths
Room 320 - Session AC-FrM

Actinides and Rare Earths Science

Moderators: Dr. Krzysztof Gofryk, Idaho National Laboratory, **Prof. Dr. Ladislav Havela**, Charles University, Czech Republic, **Dr. Paul Roussel**, AWE, **Dr. David Shuh**, Lawrence Berkeley National Laboratory

8:15am **AC-FrM-1 Environmental Radioactivity at the Department of Energy's Hanford Site: Sources and Sinks**, ***Carolyn Pearce***, Pacific Northwest National Laboratory; *Jay LaVerne,* University of Notre Dame; *Hilary Emerson, Amanda Lawter, James James Szecsody, Rob Mackley, Xin Zhang, Zheming Wang, Kevin Rosso, Gregory Schenter,* Pacific Northwest National Laboratory; *Linda Young,* Argonne National Laboratory; *Thomas Orlando,* Georgia Institute of Technology; *Aurora Clark,* University of Utah; *Xiaosong Li,* University of Washington; *Lynn Francesconi,* Hunter College, City University of New York **INVITED**

Plutonium production for the U.S. weapons program at the Department of Energy's Hanford Site created one of the world's largest environmental remediation challenges. Cleanup of 200 million liters of radioactive and chemically complex waste stored in 177 underground tanks will take decades and cost billions of dollars. Addressing the sources and sinks of environmental radioactivity requires: (i) characterization of radioactive tank waste chemistry and (ii) understanding contaminant transport and reactivity in the subsurface. The Ion Dynamics in Radioactive Environments and Materials (IDREAM) Energy Frontier Research Center provides mechanistic insights into precipitation and dissolution processes central to Hanford tank waste treatment. Hanford sludge contains water-insoluble aluminum oxides/ hydroxides that influence waste processing. Atomic force microscopy (AFM) showed that gibbsite dissolution in alkaline solutions proceeds through release of aluminate dimers rather than classical monomer detachment mechanisms. Gibbsite can also selectively adsorb or incorporate rare earth elements in tank waste. IDREAM developed an AFM integrated with an X-ray source to directly observe radiolytically driven interfacial reactions with unprecedented spatial resolution. These studies revealed how ionizing radiation, adsorbed water, and surface impurities alter mineral reactivity under highly alkaline conditions. Adsorbed carboxylates or chromium on boehmite nanoplates significantly slowed dissolution despite incomplete surface coverage. In parallel with tank waste retrieval, legacy disposal to cribs and trenches during plutonium separations operations produced vadose zone contamination and groundwater plumes that continue to impact the environment. Key groundwater contaminants include technetium-99, present as highly mobile pertechnetate (TcO_4^-), and uranium. Current remediation strategies include treatment of perched water zones and permeable reactive barriers. Technologies under evaluation include tin apatite particles, sulfur-modified iron particles, bismuth subnitrate particles, calcium polysulfide, and polyphosphate amendments. Proposed immobilization mechanisms include reduction, adsorption, incorporation into mineral phases, and in situ precipitation/coating reactions. Their effectiveness for sequestration of Tc-99 and uranium was evaluated through batch and column studies using Hanford sediments. Advances in understanding radioactive waste chemistry, radiolytic interfacial processes, and contaminant immobilization mechanisms are critical for reducing environmental risks associated with Hanford waste retrieval, treatment, and disposal.

8:45am **AC-FrM-3 Reaching the Monolayer Limit in a van der Waals Actinide Magnet**, ***Priscila Rosa***, Colorado State University **INVITED**

The discovery of local-moment magnetism in van der Waals (vdW) semiconductors down to the single-layer limit has led to a paradigm shift in the understanding of two-dimensional (2D) magnets and unleashed their potential for applications in microelectronic and optoelectronic devices. The incorporation of strong electronic and magnetic correlations in 2D vdW metals remains a sought-after platform not only to enable control of emergent quantum phases, such as superconductivity, but also to achieve more theoretically tractable microscopic models of complex materials. To date, however, there is limited success in the discovery of such metallic vdW platforms, and *f*-electron monolayers remain out of reach. In this talk, I will discuss experimental and theoretical studies of β-UTe_3, an actinide ferromagnet that can be exfoliated to the monolayer limit. A sizable electronic specific heat coefficient provides the hallmark of strong correlations. Remarkably, β-UTe_3 remains ferromagnetic in the half-unit-cell limit with an enhanced ordering temperature of 35 K, a factor of two larger than its bulk counterpart. Time permitting, I will also share recent spectroscopic insights into this system. Our work establishes β-UTe_3 as a novel materials platform for investigating and modeling correlated behavior in the monolayer limit and opens numerous avenues for quantum control with, e.g., strain engineering.

9:15am **AC-FrM-5 Theory of High-Resolution X-Ray Absorption and Resonant Inelastic X-Ray Scattering: Focus on Intermediate Valency**, ***Jindrich Kolorenc***, Institute of Physics (FZU), Czech Academy of Sciences, Prague, Czechia **INVITED**

I will discuss modeling of the high-energy-resolution x-ray absorption and the resonant inelastic x-ray scattering in actinide compounds by means of the charge-transfer multiplet theory, that is, using the Anderson impurity model constructed on the basis of first-principles electronic-structure calculations. As one of the applications, I will analyze uranium L_3 and M_4 x-ray absorption spectra taken on UPd_2Cd_{20}. These spectra indicate that the uranium 5f shell undergoes valence fluctuations in this compound [1].

The regime of valence fluctuations, known from intermetallics based on the anomalous rare-earth elements (such as Ce, Yb, Sm, Eu), has not yet been identified by microscopic techniques in any uranium compound. It was only suggested in a few of them on the basis of thermodynamic properties (such as the magnetic susceptibility) and on their similarity to the properties of the rare-earth valence fluctuators [2]. Such arguments, however, are hardly unequivocal. The x-ray absorption spectroscopy certainly provides a more direct probe of the 5f-shell behavior. Although UPd_2Cd_{20} has only one type of uranium site in its primitive cell, the absorption spectra obtained in the high-energy-resolution fluorescence-detection (HERFD) mode display two edges, each of which can be associated with a different valence state, $5f^2$ and $5f^3$. The shape of the L_3 spectrum is well approximated by two shifted copies of the uranium 6d density of states obtained in density-functional calculations, the shape of the M_4 spectrum is well approximated by a combination of UCl_3 ($5f^3$) and UCl_4 ($5f^2$) spectra reported previously [3]. This points to the $5f^2 \leftrightarrow 5f^3$ fluctuations on a timescale longer than the characteristic time of the spectroscopy technique. Unlike 4f systems, in which such fluctuations typically lead to a destruction of magnetism due to one of the involved $4f^n$ configurations being non-magnetic [4], the $5f^2$ and $5f^3$ states are both magnetic, allowing for a magnetic ordering in the fluctuating state—UPd_2Cd_{20}, in particular, is antiferromagnetic.

1. Y. Hirose *et al*., *Experimental observation of valence fluctuations by high-resolution x-ray absorption spectroscopy in UPd_2Cd_{20}*, Phys. Rev. B (2026), DOI: 10.1103/g9y8-7lq9
2. R. Troć, *The possibility of the mixed valence state in the uranium intermetallic compounds: $UCoGa_5$, U_2Ru_2Sn and U_2RuGa_8. The scaling properties*, J. Alloys Compd. **442**, 34 (2007).
3. C. L. Silva *at al.*, *On the origin of low-valent uranium oxidation state*, Nat. Commun. **15**, 6861 (2024).
4. D. I. Khomskiĭ, *The problem of intermediate valency*, Sov. Phys. Usp. **22**, 879 (1979).

9:45am **AC-FrM-7 Electronic Structure Studies of Tetravalent Actinides by Solid State NMR Spectroscopy**, ***Herman Cho***, *Khyati Anand,* Pacific Northwest National Laboratory; *Sejun Park,* Los Alamos National Laboratory; *Khusboo Rana, Eric Walter,* Pacific Northwest National Laboratory **INVITED**

The magnetic properties of tetravalent actinide centers differ markedly within the actinide row and with isoelectronic counterparts from the lanthanide row. Notable examples include the temperature independent paramagnetism of PuO_2,[1,2] exotic forms of multipolar order in UO_2 and NpO_2,[3,4] and the existence of multiple magnetic transitions in UF_4 and NpF_4.[5] These studies have provided definitive evidence that the assignment of the valence electrons of actinides to 5f orbitals oversimplifies the underlying electronic structure. With magnetic resonance spectroscopic techniques we have shown that local magnetic and electric fields at ligand sites and the metal itself can be measured in the solid state with better than part per thousand resolution. We apply this approach in systematic studies of electronic structure in an isomorphous series of An(IV) compounds including the tetrafluorides (Fig. 1)[6,7] and the dioxides (Fig. 2).[8] Plutonium, which is positioned in the 5f row between the light actinides -- characterized by itinerant valence electrons -- and higher *Z* elements that have valence electrons found to be in localized (atom-like) states, presents examples of both types of behavior.

REFERENCES

1. Raphael, G.; Lallement, R. *Solid State Commun*. **1968**, *6*, 383−385.
2. Gendron, F.; Autschbach, J. *J. Phys. Chem. Lett*. **2017**, *8*, 673−678.

3. Santini, P.; Carretta, S.; Amoretti, G.; Caciuffo, R.; Magnani, N.; Lander, G. H. *Rev. Mod. Phys*. **2009**, *81*, 807−863.
4. Rai, B. K.; Bretaña, A.; Morrison, G.; Greer, R.; Gofryk, K.; zur Loye, H.-C. *Rep. Prog. Phys*. **2024**, *87*, 066501.
5. Kern, S.; Hayward, J.; Roberts, S.; Richardson, J. W.; Rotella, F. J.; Soderholm, L.; Cort, B.; Tinkle, M.; West, M.; Hoisington, D.; Lander, G. H. *J. Chem. Phys*. **1994**, *101*, 9333–9337.
6. Capan, C.; Dempsey, R. J.; Sinkov, S.; McNamara, B.; Cho, H.; *Phys. Rev. B* **2016**, *93*, 224409.
7. Walter, E. D.; Capan, C.; Casella, A. J.; Carter, J. C.; McNamara, B. K.; Soderquist, C. Z.; Sinkov, S. I.; Clark, R. A.; Heller, F. D.; Sweet, L. E.; Corbey, J. F.; Cho, H. *J. Chem. Phys*. **2021**, *154*, 211101.
8. Rana, K.; Anand, K.; Park, S.; Sinkov, S. I.; Sweet, L. E; Cho, H. *Inorg. Chem*. **2025**, *64*, 21791-21795.

10:30am **AC-FrM-10 High-Resolution X-ray Spectroscopic Studies on Protactinium and Actinium***, **Jacob Branson**, Bianca Schacherl, Kiara Maurer, Tim Prüßmann, Kathy Dardenne,* Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal, Germany*; Rikard Malmbeck, Olaf Walter,* European Commission, Joint Research Center, Karlsruhe, Germany*; Tonya Vitova,* Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal, Germany **INVITED**

Despite occurring in nature, the chemistry of protactinium remains relatively unstudied due to its low abundance, radioactivity, and difficulty of isolation. However, existing studies on protactinium reveal chemical behavior and electronic structure that is unique even amongst the actinides, and this behavior has been attributed to the relatively large radial extent of the Pa 5f orbitals in tandem with their similar energy to the Pa 6d orbitals. In this work, the nature of the involvement of the Pa 5f and 6d orbitals is probed using high-resolution X-ray spectroscopy (HR-XANES) and resonant inelastic X-ray scattering (RIXS) experiments at the Pa $M_{3,4}$-edges, which have been demonstrated to be valuable probes of actinide electronic structure. For these studies, K_2PaF_7 and $Pa_2O_5{\cdot}xH_2O$ were synthesized in the solid state and characterized using Pa L_3-edge EXAFS. The Pa $M_{3,4}$-edge HR-XANES and RIXS experiments on these samples were conducted using two different scattering geometries and reveal dramatic differences in Pa 5f and 6d molecular orbital energetics and bond covalency between these coordination environments. We relate these differences to the competing influences of orbital energy and orbital overlap. These results form a basis of understanding for the electronic structure and chemistry of more complex Pa systems and demonstrate the high potential of the Pa 5f and 6d orbitals to participate in bonding interactions. The first experimental results on Ac M_3-edge HR-XANES of Ac-DOTA, relevant for understanding the bonding properties of Ac in radiopharmaceuticals, will also be presented and discussed. All experiments were conducted at the ACT end station of the CAT-ACT beamline at the KIT Light Source in Karlsruhe, Germany.

11:00am **AC-FrM-12 Manufacturing to Performance: Advanced Characterization of Nuclear Fuels***, **Robert Harrison**, Jonathan Morgan,* University of Manchester, UK*; Angus Wylie,* Massachusetts Institute of Technology*; Samira Bostanchi, Chris Green, David Pearmain,* Lucideon, UK*; Dave Goddard,* UKNNL, UK*; Mike Short,* Massachusetts Institute of Technology **INVITED**

Development of novel manufacturing techniques and advanced nuclear fuel materials offers significant economic and safety benefits for current and future reactor systems. However, linking processing to in-pile performance requires correlative advanced characterisation across multiple length scales. This work presents two case studies demonstrating this approach.

Firstly, field-assisted sintering (FAS) techniques, including spark plasma sintering (SPS) and flash sintering (FS), are being developed for UO_2 and mixed uranium–plutonium oxide (MOx) fuels. These methods significantly reduce sintering temperatures and processing times compared to conventional routes. However, understanding the impact of these rapid sintering processes on microstructure and chemical homogeneity—particularly plutonium distribution—is critical for fuel performance and reprocessability. A bespoke uranium-active FS system has been optimised (873-1173 K), achieving >95% theoretical density and ~5µm grain sizes, representing ~50% reductions in temperature and cycle time for UO_2. Extension to MOx surrogate systems (CeO_2-doped UO_2) revealed significant heterogeneity, including Ce-rich inclusions and degraded pellet quality. To resolve this, a correlative characterisation framework combining XRD, SEM-EDS, EPMA and Raman microscopy is employed to probe phase composition, chemical distribution and local defect chemistry. This multi-modal approach provides insight into the formation of heterogeneous regions and their relationship to processing conditions and pellet quality.

Secondly, uranium nitride (UN) is studied as an advanced and accident tolerant fuel. Ion irradiation (Ar, up to 100 DPA at 573 K) is used as a surrogate for neutron damage. Post-irradiation examination integrates XRD and TEM-EELS to characterise defect evolution, including dislocation loops, lattice swelling and gas bubble formation with Transient grating spectroscopy (TGS) to measure near-surface thermal diffusivity. This revealed a significant reduction of thermal conductivity at doses >10 DPA, directly correlated with nanoscale defect populations. In-situ annealing using TGS and TEM shows recovery of thermal transport above ~773 K, linked to point defect recombination. Overall, this work demonstrates how correlative, multi-technique characterisation links manufacturing, defect evolution and functional performance, supporting predictive understanding and accelerated development of advanced nuclear fuels.

11:30am **AC-FrM-14 Cooperative Phenomena in Locally Non-Centrosymmetric Heavy-Systems and Raman Effect in Cecosi***, **Gertrud Zwicknagl**,* TU Braunschweig, Germany

Heavy-fermion systems with magnetic ions in non-centrosymmetric sites have gained high attention during the past years. A prominent example is CeRh2As2 which exhibits a complex phase diagram with many unusual phases [1,2]. Theoretical discussions mainly focused on the role of the Rashba effect, i. e., on the consequences of local inversion symmetry breaking on the spins of the heavy quasiparticles [3].

In the present contribution, I will discuss consequences of local inversion symmetry breaking on the orbital degrees of freedom, i. e., the possible hybridization of orbitals with different parities. The inter-site interaction resulting from local Ce 4f-Ce 5d hybridization and d-d- hopping can lead to new cooperative phenomena like the lattice instability in CeCoSi [4].

I will discuss the consequences of these cooperative phenomena for the Raman spectra of CeCoSi [5].

[1] S. Khim et al, Science **373**, 1012 (2021)

[2] D. Hafner et al, Phys. Rev. **X 12**, 011023 (2022)

[3] see e. g. B. K. Nally and P. M. R. Brydon, New J. Phys. **26**, 093015 (2024) and references therein

[4] Takeshi Matsumura et al., Phys. Rev. **B 113**, 014415 (2026)

[5] Owen Moulding et al., arXiv: 2505.03249

11:45am **AC-FrM-15 Statistical Enhancement of Fissile Isotope Discrimination via Superposition of Fission-Track Clusters***, Rami Babayew, Yaacov Yehuda-Zada,* Nuclear Research Center Negev, Israel*; Galit Bar,* Soreq Nuclear Research Center, Israel*; Noam Elgad,* Nuclear Research Center Negev, Israel*; Danny Dayan,* Ben Gurion University Be'er Sheva, Israel*; Jan Lorincik,* Centre Řež, Czech Republic*; Itzhak Orion,* Ben Gurion University Be'er Sheva, Israel*; Shay Dadon,* Nuclear Research Center Negev, Israel*; Aryeh Weiss,* Bar Ilan University, Israel*; Galit Katarivas Levy, **Itzhak Halevy**,* Ben Gurion University Be'er Sheva, Israel

Accurate fissile isotope identification using Fission Track Analysis (FTA) is fundamentally limited by stochastic variations in individual fission-fragment trajectories and by the finite statistics obtainable from single-cluster measurements. While recent advances in automated image processing and synthetic-data-driven reconstruction have improved the robustness of FTA, isotopic discrimination between fissile materials with similar fission-fragment distributions remains challenging.

In this work, we propose a novel statistical enhancement framework based on superposition of reconstructed fission-track clusters originating from the same fissile grain. The method combines multiple realizations of track topologies into a unified statistical representation, effectively increasing counting statistics without increasing detector area or irradiation complexity.

Monte Carlo simulations using Geant4 were performed for ^{235}U, ^{233}U, and ^{229}Th embedded in aerogel-assisted LEXAN® solid-state nuclear track detectors (SSNTDs). Synthetic microscope images were generated and processed using an automated reconstruction pipeline to extract track morphology, cluster centroids, and Real Flight Path (RFP) distributions.

Statistical analysis demonstrated that the proposed superposition framework reduces stochastic uncertainty approximately according to: $\sigma_{eff} = 1/\sqrt{N}$

where N is the number of superposed cluster realizations. For example, superposition of N = 9 realizations improves discrimination between ^{235}U and ^{229}Th from ~3σ to ~9σ, and between ^{235}U and ^{233}U from ~1σ to ~3σ,

consistent with the expected √N scaling. More distinct isotope pairs exhibited even stronger enhancement, exceeding 5σ significance for modest superposition levels.

The proposed methodology transforms FTA from a primarily single-event morphological analysis into an ensemble-based statistical fingerprinting technique. This approach provides a scalable pathway toward improved isotopic sensitivity, automated nuclear forensic analysis, and enhanced safeguards verification, particularly for isotopes with partially overlapping fission-fragment signatures.

Future work will focus on experimental validation under neutron irradiation conditions and implementation of physics-informed alignment algorithms for real detector datasets.

Author Index

Bold page numbers indicate presenter