52èmes Journées des Actinides

April 17-21 2023 Dresden, Germany

Ac Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr

Scope

The Journées des Actinides is an international conference series providing an informal forum for the discussion of current research on the physical and chemical properties of actinide elements, their alloys and compounds. Following previous events held in Santa Margherita (2022), Institut des Sciences Chimiques de Rennes (2021, online), Erice (Italy, 2019), Praia de Porto Novo (Portugal, 2018), Karpacz (Poland, 2017), Alpe d'Huez (France, 2016), we at the Max Planck Institute for Chemical Physics of Solids (Dresden, Germany) are happy to welcome you for the 14th School on the Physics and Chemistry of Actinides (SPCA) and the 52èmes Journées des Actinides (JdA).

This event series provides an informal and interdisciplinary forum for the discussion of current advances on the physical and chemical properties of lanthanide- and actinide-based alloys and compounds. The conference has a broad scope, spanning topics from fundamental to applied research:

- strongly correlated systems, superconductivity, quantum criticality
- inorganic and organometallic chemistry
- materials and nanomaterials
- theory and band structure
- actinide production and handling
- safety of the nuclear fuel cycle
- nuclear safeguards and security
- radiation protection
- environment contamination, remediation, and decommissioning

Local organizing committee

Eteri Svanidze (MPI CPfS, conference chair)
Amy Wright (MPI CPfS)

International advisory committee

- Éric Colineau (JRC Karlsruhe, Germany)
- Mauro Giovannini (University of Genova, Italy)
- Itzhak Halevy (NRCN, Be'er Sheva, Israel)
- Ladislav Havela (Charles University, Prague, Czech Republic)
- Dariusz Kaczorowski (INTIBS PAN, Wrocław, Poland)
- Mathieu Pasturel (ISCR, Rennes, France)
- Antonio Pereira Gonçalves (IST Sacavém, Portugal)
- Kristina Kvashnina (ESRF, France)

Conference sponsors

The organization of the 13th SPCA and 50èmes JdA was possible thanks to the support of:

European C-MetAC

ThermoFisher SCIENTIFIC

MBRAUN

DR.QUENDT

Quantum Design EUROPE

Awards

The committee will award four prizes:

1. Life-time research award – recognizes a researcher who have been contributing to the actinide community for a few decades. We would like to express our appreciation by awarding them with this award.
2. Young researcher award – recognizes a young researcher who made an excellent achievement in their early career, with emphasis on the actinide science.
3. Best oral presentation – aimed at recognizing a young researcher who has given an excellent presentation. The committee will consider the abstract, the presentation and the impact on our community.
4. Best poster presentation – aimed at recognizing a young researcher who has given the best poster presentation. We encourage the researchers to give a two-minute flash poster presentation. The committee will consider the abstract, the presentation and the impact on our community.

Previous awardees

	Life-time award	**Young researcher award**	**Best oral presentation**	**Best poster presentation**
2022 Santa Margherita, Italy	Roberto Caciuffo, Gertrud Zwicknagl	-	Rebecca Nicholls	-
2021 Brittany, France	Henri Noel	-	Evgenia Tereshina-Chitrova	-
2019 Erice, Italy	Dariusz Kaczorowski	Jean-Christophe Griveau	Daniel Chaney	Chris Sala
2018 Porto Novo, Portugal	Peter Rogl, Jim Tobin	Boris Dorado	Evgeny Gerber	Oleksandra Koloskova
2017 Karpacz, Poland	Robert Troc, Wojciech Suski	Anya Keatley	-	-
2016 Alpe d'Huez, France	Ladislav Havela	Silvie Maskova	Ross Springell	-
2015 Pruhonice, Czech Republic	-	-	-	-
2014 Ein Gedi, Israel	Gerry Lander	-	-	-
2013 Sestri Levante, Italy	-	Amir Hen	-	-
2012 Bristol, United Kingdom	Geoffrey Allen	-	-	-

Schedule

The 14th School on Physics and Chemistry of Actinides will take place from April 17th to April 19th. The aim of the school is to provide a unique opportunity for students and young researchers to gain knowledge from 16 expert master-classes in various areas of different actinide science aspects. We hope that an informal atmosphere will encourage many fruitful discussions.

We are grateful to the lecturers for agreeing to present their respective areas:

- Gertrud Zwicknagl (TU Braunschweig, Germany)
- Krzystof Gofryk (Idaho National Laboratory, USA)
- James Tobin (University of Winsconsin-Oshkosh, USA)
- Lucia Amidani (ESRF and HZDR, France and Germany)
- Andrea Severing (University of Cologne, Germany)
- Roberto Caciuffo (INFN, Italy)
- Mathieu Pasturel (University of Rennes, France)
- Ladislav Havela (Charles University, Czech Republic)
- Itzhak Halevy (NRCN, Be’er Sheva, Israel)
- Mauro Giovannini (University of Genoa, Italy)
- Antonio Pereira Gonçalves (University of Lisbon, Portugal)
- Yuri Grin (MPI CPfS, Germany)
- Gerry Lander (Grenoble, France)
- Dominik Legut (TU Ostrava, Czech Republic)
- Eric Colineau (JRC-Karlsruhe, Germany)
- Rory Kennefy (Idaho National Laboratory, USA)

The 52èmes Journées des Actinides will feature nearly 50 oral presentations and nearly 30 poster presentations, featuring predominantly young researchers in the field of actinide science.

We hope that you will enjoy the 14th School on Physics and Chemistry of Actinides and the 52èmes Journées des Actinides!

Poster presentations

	Poster presenter	Poster title
1	Alexander Andreev	Magnetism and magnetoacoustics of U3Ru4Al12 single crystal in high magnetic fields
2	Ine Arts	Corrosion modeling of spent fuel – adsorption of O, O_2, H_2O and H_2O_2 to the UO_2 surface
3	Dmitry Bocharov	Modelling of the crystalline scintillating materials from the first principles
4	Manuel Brando	Electronuclear Quantum Criticality
5	Eric Colineau	Laboratories for Actinides
6	Ross Colman	Evolution of local structure across the heavy rare earth zirconate oxides, $A_2Zr_2O_7$
7	Xiaolong Feng	Real Topologically Charged Nodal Lines in 2D
8	Mauro Giovannini	Magnetic phase separation in $EuPdSn_2$
9	Volodymyr Levytskyi	Crystal, electronic structure and physical properties of the $U_3Rh_4Ge_{13}$ and $Th_2Rh_3Ge_5$ metallic systems
10	Itzhak Halevy	Nuclear forensics – Fission track Analysis – Simulation for training and penetrating fluorescent colors.
11	Shu-Xian Hu	Theoretical Study on the Bonding Feature of Actinides
12	Rory Kennedy	The Glenn T. Seaborg Institute at Idaho National Laboratory
13	Dirk Lamoen	Charge order, frustration relief, and spin-orbit coupling in U_3O_8
14	Florence Legg	Epitaxial light actinide oxide thin films
15	Andreas Leithe-Jasper	Interplay of physical and chemical properties in complex non-centrosymmetric compounds
16	Orest Pavlosiuk	Temperature-dependent Fermi surface probed by Shubnikov-de Haas oscillations in topological semimetal candidates DyBi and HoBi

17	Meike Pfeiffer	High Pressure Study of the Unconventional Superconductor $CeRh_2As_2$
18	Jun Lin	Separation and self-conditioning of Plutonium through functionalized mesoporous silicas
19	Magdalena Majewicz	Novel ternary phases in the U-Pd-Ge ternary phase diagram
20	Simon Orlat	Unravelling the electronic structure of uranium: the case of MUO_3 (M = K, Na and Rb)
21	Matthieu Pasturel	Lattice dynamics and X-ray scattering studies of low temperature crystal- lattice modulation in CeSb
22	Malte Sachs	DFT-Prediction of the Crystal Structure and Magnetism of UIr
23	Philip Schröder	Magnetic structure of UNi_4B
24	Michael Shandalov	Radiation Damage in Electroless Deposited Silver Thin Films
25	Alpesh Rasikbhai Sheth	Modelling Oxygen and Rare-Earth Impurities in Nickelates
26	Thomas Stephant	Influence of the crystal structure and nature of the ligands on the valence of uranium in binary chalcogenides: a HERFD-XANES and RIXS study
27	William Thomas	Uranium hydride: Ferromagnetism and changes under pressure
28	Hao Tjeng	Why different spectroscopies give seemingly different answers on U compounds
29	David Wheeler	Cerium-Scandium: an oxidation-resistant Ce alloy
30	Andries van Hattem	Chemistry of clad breach in a Lead-cooled Fast Reactor

Each poster presenter will be given 1 minute to advertise their poster. Please prepare one PowerPoint slide that summarizes your poster (just a few key points, not the whole poster) and be ready to show it on Wednesday.

Below, you will find abstracts in the order of appearance for:

- Oral presentations (pages 8 – 42)
- Poster presentations (pages 44 - 73)

U f^2 multiplet states in low energy excitations of UTe$_2$

D.S. Christovam,[1] M. Sundermann,[1,2] A. Marino,[1] D. Takegami,[1] Johannes Falke,[1] Paulius Dolmantas,[1] Hlynur Gretarsson,[2,3] Bernhard Keimer,[3]
Priscila F. S. Rosa,[4] Andrea Severing,[1,5] and Liu Hao Tjeng[1]

1 Max Planck Institute for Chemical Physics of Solids, Dresden, Germany
2 PETRA III, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany
3 Max Planck Institute for Solid State Research, Stuttgart, Germany
4 Los Alamos National Laboratory, Los Alamos, United States of America
5 Institute of Physics II, University of Cologne, Cologne, Germany
e-mail: denise.christovam@cpfs.mpg.de

Here we have investigated UTe$_2$, proposed to be a topological spin-triplet superconductor, with high-resolution resonant inelastic x-ray scattering (RIXS) at the U $M_{4,5}$ edges. Using this new experimental method, we can clearly observe the presence of U 5*f* atomic multiplet states in the low energy excitation spectrum of UTe$_2$. This establishes the correlated nature of the material and questions the applicability of band theory to describe quantitatively its electronic structure. We find that the observed multiplet structure belongs to the U^{4+} 5f^2 configuration. This is also surprising since half of the Te in this material have very short Te-Te distances, suggesting that half of the Te has a valence close to 1- rather than 2-, with the consequence that the U is expected to be 3+ rather than 4+. We speculate that the U 6*d* shell absorbs the extra charge, and that the local U charge configuration could be written as 5$f^2$6d^1.

Impact of Fe substitution on the electronic structure of URu_2Si_2

Andrea Marino,[1] Denise S. Christovam,[1] Chun-Fu Chang,[1] Johannes Falke,[1] Chang-Yang Kuo,[2]
Chi-Nan Wu,[2] Martin Sundermann,[1,3] Andrea Amorese,[1,3] Hlynur Gretarsson,[4,5] Camilla Moir,[6]
M. Brian Maple,[6] Peter Thalmeier,[1] Liu Hao Tjeng[1] and Andrea Severing[1,3]

1 Max Planck Institute for Chemical Physics of Solids, Dresden, Germany
2 National Synchrotron Radiation Research Center, Hsinchu, Taiwan
3 Institute of Physics II, University of Cologne, Cologne, Germany
4 Max Planck Institute for Solid State Research, Stuttgart, Germany
5 PETRA III, Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany
6 Department of Physics, University of California, San Diego, USA
e-mail: marino@cpfs.mpg.de

The application of pressure as well as the substitution of Ru with Fe in the hidden order (HO) compound URu_2Si_2 lead to the formation of the large moment antiferromagnetic phase (LMAFM), with the respective phase diagrams bearing similarities for low Fe content (<0.3). For higher Fe content (>1) $URu_{2-x}Fe_xSi_2$ adopts the Pauli paramagnetic ground state of UFe_2Si_2. However, the question remains open what causes the suppression of HO in favour of the LMAFM phase. We investigate the $URu_{2-x}Fe_xSi_2$ series with 4*f* core-level photoelectron spectroscopy. The 4*f* satellite features are enhanced at low Fe content (x<0.4) and suppressed as x increases, following the trend shown by the ordered moment. We discuss this in terms of the variation of the filling of the 5*f* shell. Implications for the exchange interaction in a Doniach phase diagram picture are then also considered.

Consequences of multipolar interactions in f-electron oxides.

S. Khmelevskyi[1], L. P. Pourovskyi[2]

1 Research Center for Computational Materials Science and Engineering, Technical University of Vienna, Karlsplatz 13, A-1040, Vienna, Austria.
2 [2]CPHT, CNRS, École polytechnique, Institut Polytechnique de Paris, 91120 Palaiseau, France
e-mail: sk@cms.tuwien.ac.at

The nature of order in low-temperature phases of some materials is not directly seen by experiment. Such “hidden orders” (HOs) may inspire decades of research to identify the mechanism underlying those exotic states of matter. In insulators, HO phases originate in degenerate many-electron states on localized f - shells that may harbor high-rank multipole moments. We show how the ground-state order and magnetic excitations of a prototypical HO system NpO$_2$, can be fully described by a low-energy Hamiltonian derived by a many-body ab initio force theorem method. A primary non collinear order of time odd rank 5 (triakontadipolar) moments has been predicted [1,2]. We show also that exotic non-chiral magnetic order in PrO2 is a results of a strong high order multipolar interactions within whole |JM> ground state multiplet. The problem of magnetic order in PrO$_2$ cannot be reduced to the interactions within a ground state Kramer’s doublet.

As has been shown by Kotljar and Haule [3] in 2009 the canonical and perhaps most investigated metallic “hidden” order materials URu$_2$Si$_2$ can also develop a “hidden” multipolar order (hexadecapolar) due to localized 5f^2 configuration at low temperature. Whereas at higher temperatures a hybridization of the localized 5f-levels leads to the Kondo behavior (“Kondo arrest scenario”) and at very low temperature a hidden order phase coexists with superconductivity. We reveal on basis of correlated ab-intio calculations a close analogy between normal state behavior of URu$_2$Si$_2$ and newly discovered heavy fermion UTe$_2$ superconductor.

References

[1] S. Khmelevskyi, L. P. Pourovskyi., PNAS, **118**, e2025317118)2020)
[2] L. V. Pourovskyi, S. Khmelevskyi Phys. Rev. B **99**, 094439 (2019)
[3] K. Haule and G. Kotliar, Nature Phys. **5**, 796 (2009)

Accessing the U–O phase diagram using topotactic oxidation

E. Lawrence Bright,[1] C. Giacobbe[1], L. M. Harding[2], R. Springell[2], and G. H. Lander[2]

1 European Synchrotron Radiation Facility, Grenoble, France
2 University of Bristol, Bristol, United Kingdom
e-mail: e.lawrencebright@esrf.fr

The oxidation of uranium dioxide, UO_2, is of major interest to the nuclear industry. As UO_2 is oxidised to U_4O_9 and U_3O_8, either during burn-up or long-term storage, the physical and chemical properties of the fuel are altered, reducing their economic and safety performance. Allen and Holmes theorized a transformation of UO_2 into U_3O_8 where shear mechanisms allow (111) UO_2 planes to change their (ABC) stacking to (A) stacking transforming to the U_3O_8 (100) plane [1]. However, recent lab-based work has shown that the UO_2 (001) surfaces oxidise to form U_3O_8 (130) in a topotactic transformation [2].

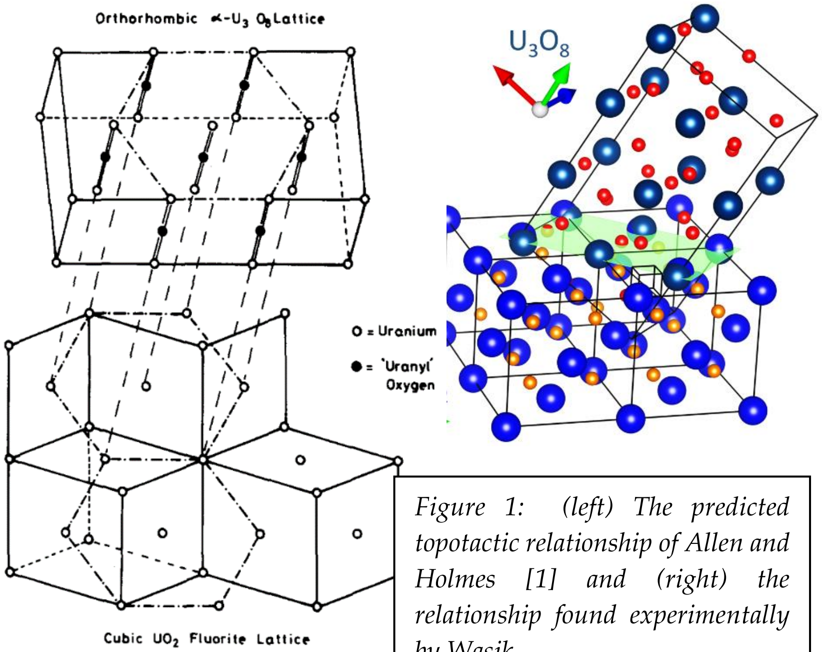


Figure 1: (left) The predicted topotactic relationship of Allen and Holmes [1] and (right) the relationship found experimentally by Wasik.

We present work from an experiment at the ID11 beamline at ESRF, where x-ray diffraction data was obtained during in-situ diffraction of a (001) UO_2 thin film as it topitactically oxidised to (130) U_3O_8. With this data, we show new insight into the mechanisms of this topotactic transformation, revealing that the intermediate oxide phases (UO_{2+x}, β-U_4O_9, U_3O_7) form during this process. This provides a novel route for obtaining single crystal (multi-domain) samples of these intermediate oxides, allowing single crystal x-ray diffraction analysis of their crystallographic structures.

References

[1] G. C. Allen, N. R. Holmes, *J. Nucl. Mater.* **231-237**, 223 (1995).
[2] J. M. Wasik (2009) ''Oxidation of Uranium Dioxide'' (Doctoral thesis, University of Bristol, Bristol, UK)

Charge order, frustration relief, and spin-orbit coupling in U_3O_8

Dirk Lamoen,[1] Rolando Saniz,[2] Gianguido Baldinozzi, Ine Arts, Gregory Leinders, and Marc Verwerft

1 EMAT, Physics Department, University of Antwerp, Groenenborgerlaan 171, B-2020 Antwerpen, Belgium
2 CMT, Physics Department, University of Antwerp, Groenenborgerlaan 171, B-2020 Antwerpen, Belgium
3 Université Paris-Saclay, CentraleSupélec, CNRS, SPMS, 91190 Gif-sur-Yvette, France
4 Belgian Nuclear Research Centre (SCK CEN), Institute for Nuclear Materials Science, Boeretang 200, B-2400 Mol, Belgium
e-mail: Dirk.Lamoen@uantwerpen.be

Research efforts on the description of the low temperature magnetic order and electronic properties of U_3O_8 have been inconclusive so far. Reinterpreting neutron scattering results, we use group representation theory to show that the ground state presents collinear out-of-plane magnetic moments, with antiferromagnetic coupling both in-layer and between layers. Charge order relieves the initial geometric frustration, generating a slightly distorted honeycomb sublattice with Néel type order. The precise knowledge of the characteristics of this magnetic ground state is then used to explain the fine features of the band gap. In this system, spin-orbit coupling (SOC) is of critical importance, as it strongly affects the electronic structure, narrowing the gap by ~38%, compared to calculations neglecting SOC. The predicted electronic structure actually explains the salient features of recent optical absorption measurements, further demonstrating the excellent agreement between the calculated ground state properties and experiment.

Hunting the Lifshitz point in UPd_2Si_2 induced by magnetic field and pressure

M. Szlawska, M. Ohashi,* M. Majewicz, K. Wochowski, and D. Kaczorowski

Institute of Low Temperature and Structure Research, Polish Academy of Sciences,
50-422 Wrocław, Poland
**on leave from Institute of Science and Engineering, Kanazawa University, Kakuma-machi,*
Kanazawa 920-1192, Japan

UPd_2Si_2 crystallizes with the tetragonal unit cell of $ThCr_2Si_2$ type (space group *I*4/*mmm*). The compound orders magnetically at low temperatures with magnetic moments oriented parallel to the tetragonal axis. It exhibits large magnetic anisotropy in both ordered and paramagnetic (P) states. The magnetic phase diagram constructed along the easy magnetic direction is complex and consists of four phases - P and three types of ordered states [1]. In zero field, with decreasing temperature, UPd_2Si_2 undergoes two successive transitions, at T_N from P to incommensurately modulated (IC) phase and at T_1 to simple antiferromagnetic (AF) state. Application of magnetic field results in the formation of ferrimagnetic (Fi) phase [1]. Remarkably, in a field of about 20 T, the P, IC and Fi phases meet each other at a point that was suggested to be a Lifshitz point (LP) [2], a multicritical point that differs from any other [3]. Furthermore, even in zero magnetic field, LP can be induced in UPd_2Si_2 by applying hydrostatic pressure of 3.4 GPa [4].

Aiming to study the effect of pressure on the magnetic phase diagram of UPd_2Si_2, and in particular to look for the occurrence of LP, we performed electrical resistivity measurements in magnetic fields up to 14 T under pressures up to 2.5 GPa. Our experiments revealed that pressure stabilizes the AF phase. We confirmed that in zero field, T_1 increases and tends to T_N, which remains nearly constant. We found that the boundary line between the AF and Fi phases shifts towards higher fields with increasing pressure. In turn, the boundary line between the IC and Fi phases hardly changes. Our findings thus suggest that pressure-induced LP may occur in UPd_2Si_2 below 2.5 GPa only in magnetic fields stronger than 14 T.

References

[1] T. Honma, H. Amitsuka, S. Yasunami, K. Tenya, T. Sakakibara, H. Mitamura, T. Goto, G. Kido, S. Kawarazaki, Y. Miyako, K. Sugiyama, M. Date, *J. Phys. Soc. Japan* **67**, 1017 (1998);
[2] Plackowski, D. Kaczorowski, J. Sznajd, *Phys. Rev. B* **83**, 174443 (2011);
[3] R. M. Hornreich, M. Luban, S. Shtrikman, *Phys. Rev. Lett.* **35**, 1678 (1975);
[4] H. Hidaka, A. Tanaka, S. Takahashi, T. Yanagisawa, H. Amitsuka, *J.Phys.:Conf. Series* **273**, 012032 (2011);

This work was supported by the National Science Centre of Poland, grant no. 2018/31/D/ST3/03295.

Magnetism and superconductivity in Hg-based binary compounds

Mitja Krnel, Yuri Prots, Yuri Grin, Eteri Svanidze

Max Planck Institute CPfS, Nothnitzer str. 40, 01187 Dresden, Germany
e-mail: mitja.krnel@cpfs.mpg.de

Crystallographically complex compounds containing 4*f* and 5*f* electrons often have peculiar chemical and physical properties. I will present the discovery and characterization of two binary Hg-based compounds that have complex crystal structure and exhibit superconductivity or antiferromagnetic order.

The $\text{LaHg}_{6.4}$ [1] and $\text{UHg}_{6.4}$ [2] crystallize in a new structure type that can be represented by La/U centered polyhedra with coordination numbers 13 and 14. An examination of crystallographic and physical properties of $\text{LaHg}_{6.4}$ reveals that this material enters the superconducting state below $T_c = 2.4$ K (Fig. 1a), while showing crystallographic disorder in one dimension. The same disorder is found in $\text{UHg}_{6.4}$, which orders antiferromagnetically below $T_{N1} = 35.5$ K and displays another transition at $T_{N2} = 47.3$ K, which is also likely antiferromagnetic (Fig. 1b). Given the exceptional air and moisture sensitivity of both compounds, special synthesis and characterization techniques were necessary to avoid decomposition of the samples.

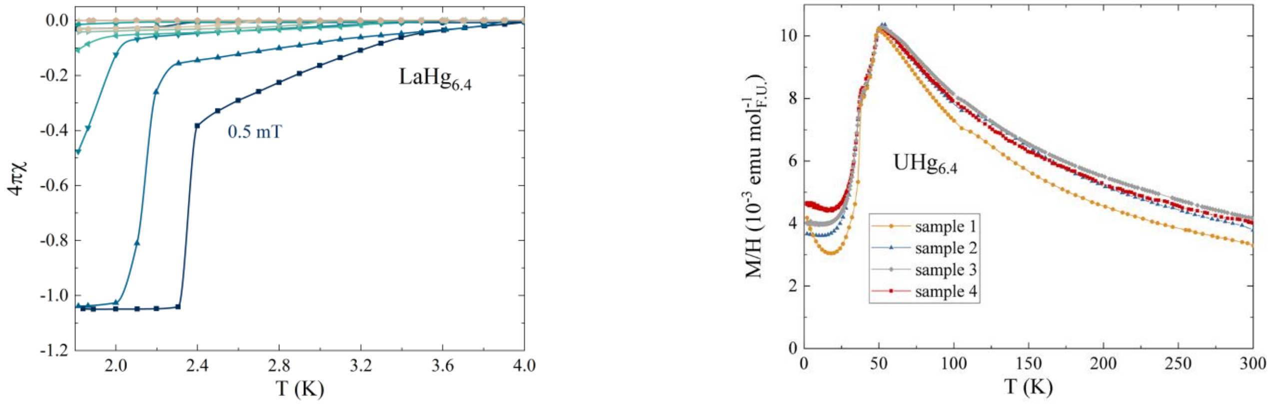


Figure 1: (a) Magnetic susceptibility as a function of temp. for $\text{LaHg}_{6.4}$ *for various fields, showing Meissner effect below T = 2.4 K and (b) Magnetic susceptibility as a function of temp. for* $\text{UHg}_{6.4}$ *in* $\mu_0H = 0.1$ *T for four samples.*

References

[1] Yu. Prots et al., *Inorg. Chem.* **61**, 15444-15451 (2022).
[2] Yu. Prots et al., *Phys. Rev. B* **106**, L060412 (2022).

Searching for new uranium-based arsenides

N. Zaremba, M. Krnel, Yu. Prots, A. Leithe-Jasper, Yu. Grin, and E. Svanidze

Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzer Strasse 40, Dresden 01187, Germany, svanidze@cpfs.mpg.de
e-mail: Nazar.Zaremba@cpfs.mpg.de

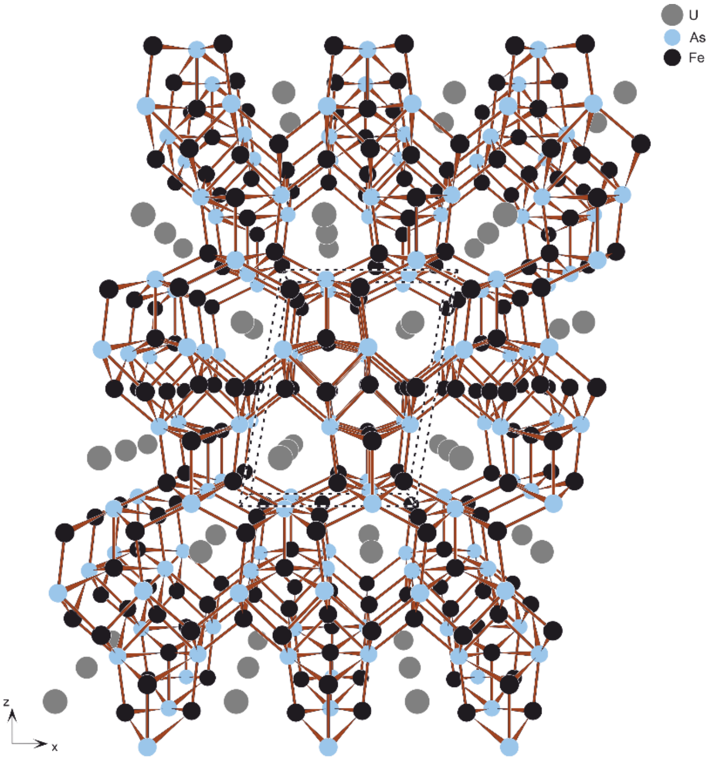


Figure 1: Crystal structure of UFe_5As_3 compound, represented by a Fe-As framework.

As new classes of superconducting materials emerge, puzzles of high-temperature superconductivity continue to be one of the pressing issues in condensed matter physics and solid-state chemistry. In particular, iron pnictide superconductors still pose many open questions. Surprisingly, very few actinide-based analogues of iron pnictide high temperature superconductors have been reported so far, perhaps as a result of synthesis complications imposed by toxicity, reactivity, and high vapor pressure of constituent elements. In this work, we revisit the U-Fe-As ternary, in which only one compounds has been reported to exist so far – $UFeAs_2$ ($P4/nmm$ space group) [1]. By implementing flux synthesis, we were able to grow large single crystals of UFe_5As_3 compound, which adopts UCr_5P_3 structure type [2] ($P2_1$/m, $mP18$) with lattice parameters a = 7.0501(17) Å, b = 3.8582(9) Å and c = 9.6342(13) Å, β = 100.2(8)° [3]. The refined composition agrees well with the elemental ratio established by the EDX analysis – $U_{9.9(2)}Fe_{58.7(2)}As_{31.4(2)}$. The magnetic behavior of UFe_5As_3 was studied by magnetic susceptibility measurements in the temperature range 1.8 - 600 K and an antiferromagnetic ordering below T_N = 56 K was established. The Sommerfeld coefficient γ = 138 mJ/mol K^2 indicates enhanced effective electron mass. More in-depth investigations of this and other U-Fe-As ternary compounds are currently underway.

References:

[1] D. Kaczorowski, J. Alloys. Compd. 186, 333-338 (1992).
[2] W. Jeitschko, R. Brink, P.G. Pollmeier, Z. Naturforsch. (1993) 48b, p. 52-57.
[3] N.Zaremba, M. Krnel, Yu. Prots, A. Leithe-Jasper, Yu. Grin, E. Svanidze, in preparation (2023)

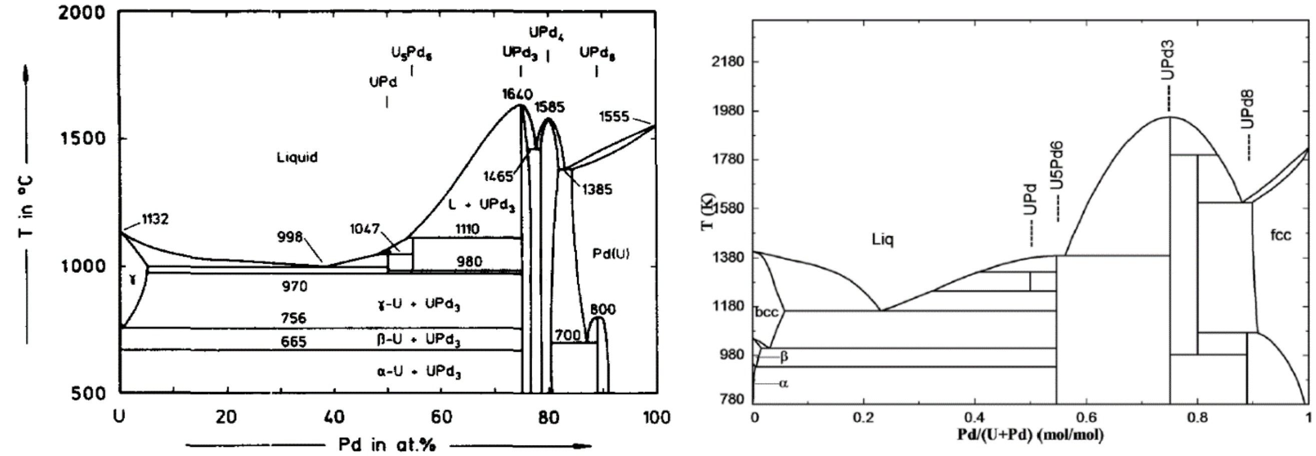
New assessment of U-Pd phase diagram

Théo Croze,[1,2] A. Quaini[1], S. Gosse[1], O. Tougait[2]

1 ISAS, S2CM, CEA – Université Paris-Saclay, Gif-sur-Yvette, France
2 Univ. Lille, CNRS, Centrale Lille, Univ. Artois, UMR 8181 - UCCS - F-59000, Lille, France
e-mail: theo.croze@cea.fr

U-Pd binary phases have retained considerable attention for both technological and fundamental studies, reagarding that, Pd constitutes one of the main fission products of the U-based nuclear fuels, and the specific electronic structure of UPd_3 described as a localized model. Despite an abundant literature, a comprehensive constitution of the U-Pd system is still missing. As an example of the absence of an accepted description of the phase equilibria, an experimental [1] (left) phase diagram and an optimized one using the Calphad method [2] (right) are depicted on Fig. 1. The comparison outlines conflicting results about the invariant reactions, phase formation and stability as well as solubility domains. In addition, it should be mentioned that the existence of UPd, U_5Pd_6 and UPd_8 intermediate compounds hasn't been fully proved with their crystalline structure remaining undetermined up to now.

Fig. 1: Sketches of U-Pd phase diagram, based on experimental data [1] (left) and calculated (right) [2]



The aim of the present study is to assess the U-Pd system using a coupled approach combining an experimental investigation of phase relations and the optimization of a new thermodynamic model. For that, the Pd rich region was investigated by metallography, X-ray microanalysis and X-ray diffraction. The presentation will show a new assessment of the U-Pd binary system by the Calphad method.

References

[1] H.Kleykamp, S. G.Kang, J. Nucl. Mater., **280-286**, 230 3 (1996).
[2] L. C.Wang, University of Ontario Institute of Technology, Oshawa, Ontario, Canada (2018).

Hide and Seek: where and what is the magnetic order?

Lars Nordström

Uppsala University, 751 21 Uppsala, Sweden
e-mail: lars.nordstrom@physics.uu.se

We will concentrate on magnetic materials with Uranium atoms, which means that the spin orbit coupling is significant. This strong coupling leads to that the spin and orbital degrees of freedom of the valence electrons are strongly mixed and intertwined which require a careful treatment where these degrees are handled on equal footing. Such a method [1] is conceptually introduced and its implications is discussed. In this context we will put some effort to discuss and seek the possibility that the formation of complex magnetic order may be not observable, which is often referred to as hidden order. [2] Lastly, we will also briefly discuss the fact that systems that are candidates for hidden order often displays an instability to unconventional superconductivity at low temperature. Here we will discuss the example of the newly discovered superconductor which is attracting large interests and showing several striking properties, UTe_2. [3]

References

[1] Bultmark et al, Phys Rev B **80**, 035121 (2009) and Cricchio et al, Euro Phys Lett **94**, 57009 (2011)
[2] Cricchio et al, Phys Rev Lett **103**, 107202 (2009)
[3] Ran et al, Science **365**, 684 (2020)

Neutron Scattering Techniques Applied to Actinide Materials

G. H. Lander[1], and Roberto Caciuffo,[2]

1 Institut Laue Langevin, 38042 Grenoble, France
2 Istituto Nazionale di Fisica Nucleare, Via Dodecaneso 33, IT-16146 Genova, Italy
e-mail: lander@ill.fr

Neutron scattering has been applied to nuclear materials for the last 60 years. Overwhelming advantages over other scattering techniques include the ability to detect light atoms in the presence of heavy ones, measure disordered and ordered magnetism, and measure both phonon and magnon dispersion curves if large enough single crystals are available. In this talk the basic cross sections will be presented and some examples discussed.

Starting in the 1960s work on UO_2 represents a fascinating historically study of the importance of neutron scattering, from the early work of Willis (at Harwell, UK) on the structural aspects of both the pure compound and UO_{2+x}, to inelastic scattering (at Chalk River, Canada) of the phonons and magnons (and their interaction). Inelastic scattering in 1980s at the ISIS, UK, spallation source established the crystal-field parameters. To bookend the work on UO_2 we consider some of the work performed on single crystals in the early 2000s at the Institut Laue Langevin using advanced techniques with polarization analysis.

Other notable experiments have been the discovery (1980 at ORNL US, and ILL, France) of the charge-density wave in uranium metal, and the search for magnetism in plutonium, which despite many theoretical predictions, has never been found. Instead, inelastic scattering at the ORNL spallation source in 2015 was able to detect the Kondo gap in this complex material, providing definitive support for the theoretical predictions of the dynamical mean-field theory.

A serious disadvantage of neutron scattering is the amount of material required for experiments. Increasing safety demands have unfortunately almost halted further neutron experiments on transuranium materials.

Fundamentals of the U halides

C. L. Silva[1,2], L. Amidani[1,2], F. Otte[1,2], E. F. Bazarkina[1,2], S. Weiss[2], F. Kraus[3], K. O. Kvashnina[1,2]

1 The Rossendorf Beamline at ESRF – The European Synchrotron, CS40220, 38043 Grenoble Cedex, France
2 Helmholtz-Zentrum Dresden-Rossendorf (HZDR), Institute of Resource Ecology, 01314 Dresden, Germany
3 Fachbereich Chemie Philipps-Universität Marburg, Hans-Meerwein-Str. 4, 35032 Marburg, Germany
e-mail: clara-lisa.e-silva@esrf.fr

Assessing the nature of bonding as ionic versus covalent is a subject of strong interest in f-element complexes [1]. While spectroscopic techniques reveal the characteristics of f-element bonding, the effort to understand 5f electron systems has been harmed by the lack of high-quality experimental data on actinide compounds to be studied [2-4]. We report here the high energy resolution fluorescence detected (HERFD) recorded for the family of the U(III) and U(IV) halides, named UX$_3$ and UX$_4$ (X=F, Cl, Br, I). The electronic structure of both 5f^3 and 5f^2 halide families was investigated by HERFD at the U M$_4$ edge which reveals differences in the spectral shape in both families. To interpret the changes and gain insight into the nature of the 5f orbitals, we applied crystal-field multiplet theory to calculate the 3d-4f RIXS maps, and extract the HERFD spectra as cuts of the calculated RIXS maps. We investigated the effect of reducing different contributions to the electron-electron interactions involved in the RIXS process to rationalize the spectral changed observed. We found that that the spectral shape is primarily due to electron-electron interactions, with little influence from crystal field interactions. Small variations of the Slater-Condon integrals result in very good agreement with the changes observed for UX$_4$, while stronger variation of the interaction between the final core-hole and the 5f electrons is needed to partially reproduce changes of the UX$_3$ set. Our analysis indicate the sensitivity of HERFD at the M4 edges to weak changes of the intra-atomic interactions in strongly ionic systems.

References

[1] Tanti, J. et al., *Inorganics* **6**, 88 (2018).
[2] Kvashnina, K. O., and S. M. Butorin. *Chemical Communications* **58**, 327 (2022).
[3] Butorin, S. M. *Journal of Chemical Physics* **155**, 164103 (2021).
[4] Amidani, L. et al., *Inorg. Chem.* **60**, 16286 (2021).

Theoretical Study on the Bonding Feature of Actinides

Shu-Xian Hu

University of Science and Technology Beijing, 100083 Beijing, China
e-mail: hushuxian@csrc.ac.cn

Actinide metal oxo clusters are of vital importance in actinide chemistry, as well as in environmental and materials sciences. They are ubiquitous in both aqueous and nonaqueous phases and play key roles in nuclear materials (e.g., nuclear fuel) and nuclear waste management. Despite their importance, our structural understanding of the actinide metal oxo clusters, particularly the transuranic ones, is very limited because of experimental challenges such as high radioactivity. Herein we report a systematic theoretical study on the structures and stabilities of seven actinide metal oxo−hydroxo clusters $[An^{IV}_6O_4(OH)_4L_{12}]$ (1-An; An = Th−Cm; L = O_2CH-) along with their group 4 (Ti, Zr, Hf, Rf) and lanthanide (Ce) counterparts $[M^{IV}_6O_4(OH)_4L_{12}]$ (1-M). The work shows the Td-symmetric structures of all of the 1-An/M clusters and suggests the positions of the −OH functional groups, which are experimentally challenging to determine. Furthermore, by removing six electrons from 1-An, we found that oxidation could happen on the AnIV metal ions, producing $[An^{V}_6O_4(OH)_4L_{12}]^{6+}$ (2-An; An = Pa, U, Np), or on the O^{2-} and OH^{-} ligands, producing $[An^{IV}_6(O^{\bullet-})_4(OH^{\bullet})_2(OH)_2L_{12}]^{6+}$ (3-An; An = Pu, Am, Cm). On the basis of 2-An, we constructed a series of tetravalent and pentavalent actinide metal oxo clusters $[An^{IV}_6O_{14}]^{4-}$ (4-An) and $[An^{V}_6O_{14}]^{2+}$ (5-An), which proves the feasibility of the highly important pentavalent actinyl clusters, demonstrates the f orbital's structure-directing role in the formation of linear $[O{=}An^{V}{=}O]^{+}$ actinyl ions, and expands the concept of actinyl−actinyl interaction into pentavalent transuranic actinyl clusters.

References

[1] S.-X. Hu et al., *Inorganic Chemistry* **59**, 18068 (2020).
[2] P. Zhang and S.-X. Hu et al., *European Journal of Inorganic Chemistry* **38**, 3926 (2021).
[1] S.-X. Hu et al., *Inorganic Chemistry* **61**, 15589 (2022).

Role of electronic interactions on valence fluctuations and structural properties of actinides

Bernard Amadon

1 CEA, DAM, DIF, F-91297 Arpajon, France
2 Université Paris-Saclay, CEA, Laboratoire Matière en Conditions Extrêmes, 91680 Bruyères-le-Châtel, France
e-mail: bernard.amadon@cea.fr

The description of pure actinides is complex for electronic structure calculations because of the interplay of electronic interaction, spin orbit coupling and complex crystal structure. Being able to describe the variety of the phases of plutonium, their spectral properties and their non magnetic ground state is thus a challenge and represents a striking example of this complexity. We discuss the theoretical description of the experimental transition from low volume early actinides (uranium, neptunium, α-plutonium) to high-volume late actinides (δ-plutonium, americium, and curium) and the volume of phases of plutonium. We will discuss methods using an explicit 5f electronic interactions, such as the combination of Density Functional Theory (DFT) with Dynamical Mean Field Theory (DMFT) and DFT+U.

We will comment the role of Coulomb interaction U. We will in particular discuss the link between valence fluctuations and structural properties.

References

[1] Bernard Amadon Phys. Rev. B **94**, 115148 (2016), Phys. Rev. B **97**, 039903 (2018)
[2] B. Amadon and B. Dorado, J. Phys.: Condens. Matter **30**, 405603 (2018).

Endohedral Actinidofullerenes: U-U bonds

Yannick Roselló, Antonio Rodríguez-Fortea, Josep M. Poblet

Departament de Química Física i Inorgànica, Universitat Rovira i Virgili, 43007, Tarragona, Spain
e-mail: yannick.rosello@urv.cat

Endohedral metallofullerenes (EMFs) are hybrid compounds that encapsulate metal atoms or clusters in their inner void space. EMFs are stabilized by a charge transfer from the internal cluster to the carbon cage. Several mono-actinidofullerenes as $U@C_{80}$, $Th@C_{80}$, $Th@C_{82}$, $U@C_{76}$, $U@C_{82}$ have been characterized.[1] Two actinides (U_2 and Th_2) and actinide clusters (U_2C, U_2N…) have been also found inside different carbon cages, showing characteristic metal-metal and metal-cage interactions.[2-4]

To evaluate the feasibility of covalent U-U bonds, we have investigated the formation of small diuranium EMFs by laser ablation using mass spectrometric detection. Computational studies show that, thanks to the formation of strong $U(5f^3)$-$U(5f^3)$ triple bonds, two U^{3+} ions can be incarcerated inside the fullerene. The formation of U-U bonds competes with U-cage interactions that tend to separate the U ions. Although 5f-5f interactions are responsible for the covalent interactions at distances close to 2.5 Å, overlap between 7s6d orbitals is still detected above 4 Å in larger cages as $U_2@C_{80}$. In general, metal ions within fullerenes should be regarded as templates in cage formation, not as statistically confined units that have little chance of being observed.[5]

References

[1] Cai, W.; Abella, L.; Zhuang, J.; Zhang, X.; Feng, L.; Wang, Y.; Morales-Martínez, R.; Esper, R.; Boero, M.; Metta-Magaña, A.; Rodriguez-Fortea, A.; Poblet, J. M.; Echegoyen, L.; Chen, N. *J. Am. Chem. Soc.* **140**, 18039-18050 (2018).
[2] Zhang, X.; Wang, Y.; Morales-Martínez, R.; Zhong, J.; de Graaf, C.; Rodríguez-Fortea, A.; Poblet, J. M.; Echegoyen, L.; Feng, L.; Chen, N. *J. Am. Chem. Soc.* **140**, 3907−3915 (2018).
[3] Li, X.; Roselló, Y.; Yao, Y.R.; Zhuang, J.; Zhang, X.; Rodríguez-Fortea, A.; de Graaf, C.; Echegoyen, L.; Chen, N. *Chem. Sci.* **12**, 282−292 (2021).
[4] Yao, Y. R.; Roselló, Y.; Ma, L.; Puente Santiago, A. R.; Metta-Magaña, A.; Chen, N.; Rodríguez-Fortea, A.; Poblet, J. M.; Echegoyen L. *J. Am. Chem. Soc.* **143**, 15309−15318 (2021).
[5] Moreno-Vicente, A.; Roselló, Y.; Chen, N.; Echegoyen, L.; Dunk, P. W.; Rodríguez-Fortea, A.; de Graaf, C.; Poblet, J. M. *J. Am. Chem. Soc.* ACCEPTED ON 14/02/2023

Actinide-MOF Chemistry and Applications

Moritz Schmidt,[1] Kai Lv[2], Juliane März,[1] Sebastian Fichter,[3] Michael Patzschke,[1] and Peter Kaden[1]

1 Helmholtz-Zentrum Dresden - Rossendorf, Institute of Resource Ecology, Bautzner Landstr. 400, 01328 Dresden, Germany
2 China Academy of Engineering Physics, Institute of Nuclear Physics and Chemistry, 621900 Mianyang, Sichuan, China
3 Helmholtz-Zentrum Dresden - Rossendorf, Institute of Ion Beam Physics, Bautzner Landstr. 400, 01328 Dresden, Germany
e-mail: moritz.schmidt@hzdr.de

Actinide metal-organic frameworks (An-MOFs) have garnered considerable attention in recent years.[1] Yet, this class of compounds remains understudied compared with their transition metal or lanthanide homologues and this, despite a wide range of interesting properties, which may be turned into promising applications, from radiation detection to tailor-made nuclear waste forms. Moreover, this novel group of metal-organic compounds allows for straightforward comparison with molecular complexes, which have been the target of fundamental studies in actinide science for some time.

We will discuss a series of isoreticular MOFs based on mononuclear An(IV) (Th, U, Np, Pu) primary building units in which each actinide is coordinated by six molecules of 9,10-anthracenedicarboxylic acid (ADC).[2] This arrangement leads to an unusually large coordination number of 12 in icosahedral symmetry. Quantum chemical calculations indicate that this large coordination number is only feasible in the high-symmetry environment provided by the An-MOFs. Moreover, these MOFs not only demonstrate autoluminescence but also wide-bandgap (2.84 eV) semiconducting properties. In addition, we will present recent findings, illustrating how selective crystallization from mixed-metal solutions may present a viable pathway for the production of actinide waste forms from specialized waste streams. As an example we will present the formation of Th-MOFs containing isonicotinic acid linkers in the presence of a wide range of metals representative of fission products.

References

[1] K. Lv et al., *Coordination Chemistry Reviews* **446**, 214011 (2021) and references therein.
[2] K. Lv et al., *Journal of the American Chemical Society* **144**, 2879 (2022).
[3] K. Lv et al., *ACS Materials Letters* **5**, 536 (2023).

Itinerant-localized dichotomy in magnetic anisotropic properties of U-based ferromagnets

Dominik Legut[1], Alexander Shick[2,3,1], Itzhak Halevy[4], Maxim Tchaplianka[2]

[1]IT4Innovations, TU-Ostrava,17. listopadu 2172/15, CZ-70800 Ostrava, Czech Republic ,
[2]FZU-Institute of Physics CAS, Na Slovance 2, 18221 Prague, Czech Republic
[3]Weizmann Institute of Science, 76100 Rehovoth, Israel
[4]Ben Gurion University of the Negev, 84105 Beer-sheva, Israel
e-mail: dominik.legut@vsb.cz

Exploration of the electronic and magnetic character of quantum matter is one of the most fundamental fields of modern condensed matter physics. Traditionally, two competing itinerant and localized characteristics distinguish magnetic and heavy Fermi-liquid behavior. Only a handful of quantum materials that show this itinerant-localized dichotomy are the uranium-based compounds UGe_2, UTe_2, URhGe, and UCoGe, where ferromagnetism coexists with unconventional superconductivity.

In this work, we examine the itinerant-localized dichotomy in selected U-based ferromagnets making use of the correlated band theory implemented in a local-spin-density plus Coulomb-U approach (LSDA+U). At first, we demonstrate that the LSDA+U with exact atomic limit LSDA+U(HIA) which combines the relativistic density functional theory with the Anderson impurity model provides a good quantitative description for spectroscopic and magnetic properties, including the magnetic anisotropy energy (MAE), of UGa_2. These results are compared with previous experimental and theoretical works and additional features are pointed out.

Next, the method is applied to UFe_{12} and $UFe_{10}Si_2$ ferromagnets. We estimate the thermodynamic stability for these materials in terms of the enthalpy of formation, and illustrate that the Si atom substitution into UFe_{12} stabilizes the $ThMn_{12}$-type crystal structure. Furthermore, the LSDA+U(HIA) calculations yield the uniaxial MAE of $UFe_{10}Si_2$ in agreement with available experimental data, and suggest $UFe_{10}Si_2$ ferromagnet as a candidate for magnetically hard material.

In the resent years there is an extensive research targeting new permanent magnets with reduced amount of the rare-earth elements. A good permanent magnet ought to have reasonably high Curie temperature, a large magnetization for a high energy product, and a substantial uniaxial MAE to resist demagnetization. Our results suggest that replacing a part of U atoms by some rare-earth like Sm in $UFe_{10}Si_2$ will further enhance the magnetization and MAE. This can facilitate a development of the new rare-earth-lean permanent magnets.

We acknowledges partial support provided by EU OPVV project SOLID21-CZ.02.1.01/0.0/0.0/16_019/0000760, and the Czech Science Foundation (GACR) Grant No. 22-22322S. DL acknowledges the project e-INFRA CZ (ID:90140) by the Ministry of Education, Youth and Sports of the Czech Republic. ABS acknowledges financial support from the Israeli Ministry of Aliyah and Integration Grant 140636.

Corrosion modeling of spent fuel – adsorption of O, O_2, H_2O and H_2O_2 to the UO_2 surface

Ine Arts,[1] Rolando B. Saniz[2], Selma Mayda[1], Gianguido Baldinozzi[3], Gregory Leinders[4], Marc Verwerft[4], Dirk Lamoen[1]

1 EMAT, Physics Department, University of Antwerp, B-2020 Antwerpen, Belgium
2 CMT, Physics Department, University of Antwerp, B-2020 Antwerpen, Belgium
3 Université Paris-Saclay, CentraleSupélec, CNRS, SPMS, F-91190 Gif-sur-Yvette, France
4 Belgian Nuclear Research Centre (SCK CEN), Institute for Nuclear Materials Science, B-2400 Mol, Belgium
e-mail: ine.arts@uantwerpen.be

Uraniumdioxide (UO_2) is the main component of nuclear fuels. This compound is very susceptible to oxidation, which results in formation of the mixed-valence oxide U_3O_8.[1] From an application point of view, this interest relates to the safe and sustainable management of nuclear fuel. Our goal is an understanding of this oxidation process at the atomic level, using ab initio electronic structure calculations.

The focus in this work lies on the early oxidation of stoichiometric UO_2, where excess oxygen can be treated as interstitial point defects. In order to study this oxidation we investigate the adsorption of the O atom and oxygen containing molecules (O_2, H_2O, H_2O_2) on the three most stable UO_2 surfaces ((111), (110) and (100)). The calculations are performed within DFT, using PBE+U including spin-orbit coupling and taking into account the non-collinear 3k (transverse) magnetic ground state.

Dissociative adsorption is found for H_2O and H_2O_2 on the (111) and (100) UO_2 surface respectively, forming hydroxyl groups on the surface. Adsorption on the UO_2 surface lowers the barrier for dissociation of O_2. Defect formation energy is calculated for interstitial oxygen, up to several layers into the material starting from the surface. Diffusion energy barriers are presented. Additionally adsorption energies are studied as a function of temperature and partial pressure.

References

[1] E. De Bona, K. Pope, O. Walter, M. Cologna, C. Hennig, A. C. Scheinost, and D. Prieur., *Inorg. Chem.* **61**, 1843 (2020).

Structural Incorporation of Dopants into Uranium Oxides – The Effect of Temperature, Size, and Oxidation State

S. K. Potts,[1] P. Kegler,[1] K. Popa,[2] W. Bonani,[2] J.-Y. Colle,[2] O. Walter,[2] G. Modolo,[1]
M. Klinkenberg,[1] I. Niemeyer,[1] D. Bosbach,[1] S. Neumeier[1]

1 Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research – Nuclear Waste Management (IEK-6), 52428 Jülich, Germany
2 European Commission, Joint Research Centre (JRC), Karlsruhe, Germany
e-mail: s.potts@fz-juelich.de

To support the International Atomic Energy Agency's (IAEA's) sustainable quality control system in particle analyses in nuclear safeguards [1], a process has been implemented in the laboratories at the Forschungszentrum Jülich (FZJ) to produce well-defined uranium oxide microparticulate reference materials for mass spectrometric verification measurements [2], [3]. The enhancement of mass spectrometric analytical methods for the detection of even small amounts of dopants (e.g., lanthanides and actinides), requires the development of novel mixed uranium oxide microparticulate reference materials. Due to the yield limitations in the µg range of the aerosol-based process implemented at FZJ, the structural characterization of these materials is challenging. Therefore, in the present work doped materials at the bulk-scale were used as "internal reference materials" to investigate the incorporation mechanism of dopants, such as lanthanides [4], Th, or Pu, into the uranium-oxide structure. Various doped ammonium diuranate materials were co-precipitated and investigated by TG-DSC in order to identify the temperature ranges in which the phase transformation from UO_3 to U_3O_8 phase occurs. According to these results, the materials were calcined and systematically characterized with XRD as well as Raman and IR to investigate the long-range as well as the short-range order phenomena of the materials' structure. This presentation will provide an insight regarding the incorporation of dopants, such as lanthanides, Th or Pu, into the uranium oxide structures.

References

[1] K.G.W. Inn et al., *J. Radioanal. Nucl. Chem* **296**, 5 (2012).
[2] R. Middendorp et al., *Anal. Chem.* **89**, 4721 (2017).
[3] S. Neumeier et al., *MRS Adv.* **3**, 1005 (2018).
[4] S. K. Potts et al., *MRS Adv.* **7**, 128 (2022).

Evolution of local structure across the heavy rare earth zirconate oxides, $A_2Zr_2O_7$

Ross H. Colman,[1] Milan Klicpera[1], Kristina Vlášková[1]

1 Charles University, Ke Karlovu 5, 121 16, Prague 2, Czech Republic
e-mail: ross.colman@mag.mff.cuni.cz

Oxide ion conductors require migration pathways for ions to pass through the lattice, typically via hopping through vacancy sites. The ease of this migration is dictated by both the bonding strength, and the structure (relative vacancy locations), with these characteristics guiding the ion conductivity and subsequently the minimum useful working temperature. The rare-earth oxides, with general formula $A_2B_2O_7$ (where A^{3+} is a rare-earth ion, and B^{4+} is a transition or p-block metal), have shown reasonable ion conductivity properties. These oxides show distinct structure types dependent on the A/B ion size ratio: $r_A/r_B > 1.46$ result in an ordered pyrochlore structure, and $r_A/r_B < 1.46$ results in a disordered defect-fluorite structure [1]. Probes of the ion local structure (spectroscopic methods), however, don't show an abrupt change across this distinct *structural-border* [2]. Understanding the subtleties of the structural changes through this family is important for explaining the observed ion-conductivities [3].

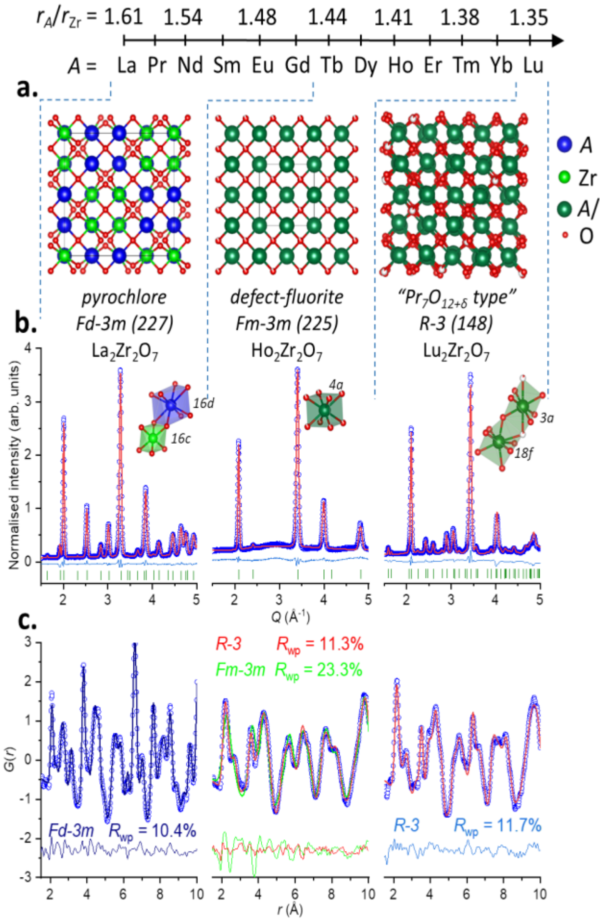


*Fig 1. The rare-earth zirconate series, $A_2Zr_2O_7$, showing: **a.** the long-range average structures; **b.** refined powder neutron diffraction data (with inset cation environments); **c.** nPDF data with fits.*

In this poster I will show the results of our recent efforts to use a combination of neutron and X-ray diffraction, with small box pair-distribution-function analysis (xPDF and nPDF) to better understand the local structure evolution across the heavy rare-earth zirconates, $A_2Zr_2O_7$ (Fig 1).

References

[1] J. Shamblin, M. Feygenson, J. Neuefeind, et al., Nat. Mater. **15**, 507 (2016).
[2] P. E. R. Blanchard, R. Clements, B. J. Kennedy, et al., Inorg. Chem. **23**, 41 (2012).
[3] M. Shafique, et al., *J. Alloys Compd*. **671**, 226 (2016).

The oxidation of single-phase uranium silicide thin films

L. M. Harding,[1] E. Lawrence Bright[2], J. Laverock[3], D.T. Goddard[4], R. Springell[1]

1 School of Physics, H.H Wills Physics Laboratory, University of Bristol, Bristol, UK
2 European Synchrotron Radiation Facility, 71 Avenue des Martyrs, 38040 Grenoble, France
3 School of Chemistry, University of Bristol, Cantocks Close, Bristol, UK
4 National Nuclear Laboratory, Springfields, Preston, Lancs., PR4 0XJ, UK
e-mail: lottie.harding@bristol.ac.uk

Advanced Technology Fuels (ATFs) are a key concept in the drive to improve overall performance and safety in the nuclear industry. Uranium silicide (U_3Si_2) has been highlighted as a potential ATF, having improved thermal properties and increased uranium densities when compared with UO_2. However, there are significant discrepancies within the literature regarding the corrosive properties of U_3Si_2. Some studies indicate that uranium preferentially oxidises, resulting in a U-Si fuel type with a higher Si content [1]. Whereas other studies suggest the formation of a uranium silicate, $USiO_4$ [2]. Understanding the surface oxidation is vital for assessing the structural integrity of the fuel, and is beneficial for dissolution studies. Furthermore, conducting analysis on this material is challenging, as the process required to produce U_3Si_2 in the bulk often results in the formation of multi-phased silicides [3]. In this regard, thin films offer significant potential, providing idealised surfaces on which single parameter studies can be conducted. Through varying the growth parameters, stoichiometry, grain size, and crystallographic orientation can also be controlled.

Here we present an investigation into the ambient oxidation of uranium silicide single crystals. Using DC magnetron sputtering, the U-Si phase diagram has been delicately mapped as a function of uranium content [4]. The oxidation of U_3Si, U_3Si_2, U_3Si_5, USi_2, and USi_3 surfaces has been probed using x-ray diffraction and x-ray photoelectron spectroscopy; allowing for the oxidation thickness to be measured, and for an investigation into the valency of each oxidised phase to be conducted. These studies have been vital in understanding the oxidation products that form on U-Si phases as a function of uranium content.

References

[1] E. Jossou, et al., Physical Chemistry Chemical Physics 207, 4708-4720, (2018)
[2] T Yan et al., Journal of Nuclear Materials 520, 1-5, (2019)
[3] Ki Hwan Kim et al., Journal of nuclear science and technology, (1997)
[4] Harding et al., Thin Solid Films, 768, 139690, (2023)

Lattice dynamics and X-ray scattering studies of low temperature crystal- lattice modulation in CeSb

L. Paolasini[1], E. Mariani[1], F. de Bergevin[1], P. Maldonado[2], T. Forrest[3] and G. Lapertot[4]

1 ESRF, 38043 Grenoble France,
2 Department of Physics and Astronomy, Uppsala University, Sweden
3 Diamond Light Source Ltd, Harwell Science and Innovation Campus, UK
4 CEA-IMAPEC, 38000 Grenoble, France
e-mail: paolasini@esrf.fr

The Ce monopnictide compounds with the NaCl-type structure like CeSb exhibit unusual transport and magnetic properties, and have been extensively studied with neutron and synchrotron scattering techniques [1-4]. They have extremely low carrier densities, and the single electron of the 4f orbital of Ce^{3+} ion determines the unusual magnetic properties that have been ascribed to the strong magnetic polaron effect, produced by the combination of localization of the low-density carriers and mixing effects between Ce ions and the p state of the neighbouring pnictogens (p-f mixing) [5].

We have determined the phonon dispersion curves in all the main crystallographic directions by high-resolution inelastic X-ray scattering at ID28 beamline at ESRF. Ab-initio calculations, using the Vienna Ab-initio Simulation Package (VASP) and including an additional Hubbard term (DFT+ U) to obtain a good description of the electronic structure of this highly correlated system.

At low temperature in correspondence of the dominant magnetic modulation called AFP3 phase [1] with the magnetic propagation vector q=4/7 (T=14-15K), elastic X-ray scattering shows charge satellite peaks of type (G+◎$_{n}$,0,0) with ◎$_{n}$= n x 2/7 (n=±1, ±2,+3 and G=2,4,6) up to 3rd order. Surprising, a detailed analysis over different Brillouin zones revealed a systematic absence of satellite peak ◎$_{-3}$. We propose a displacement model which take into account ferromagnetic, antiferromagnetic and anti-ferro-para-magnetic magnetostriction coupling between different ◎$_{7}$ and ◎$_{8}$ Ce planes, and that reproduce the experimental data with great accuracy. The resulting model can be used to rationalize different conflicting results reported for this material in literature.

References

[1] J. Rossat-Mignod et al., J.M.M.M. 52, (1985) 111-121
[2] D. McMorrow *et al.*, J. Phys. Cond. Matter 9, (1997) 1133
[3] K. Iwasa *et al.*, Phys. Rev. Lett. **88** (2002) 207201
[4] A. Stunault *et al.*, Physica B 345 (2004) 74-77
[5] T. Kasuya, T. Suzuki: J. Phys. Soc. Jpn. **61**, 2628 (1992)

Even and Odd Parity Superconductivity in $CeRh_2As_2$

J. F. Landaeta[1], P. Khanenko[1], D. Agterberg[2], P. Brydon[3], D. Cavanagh[3], T. Lühmann[1], M. Brando[1], C. Geibel[1], S. Khim[1], and E. Hassinger[3]

1 Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany
2 Department of Physics, University of Wisconsin-Milwaukee, Milwaukee, Wisconsin 53201, USA
3 Department of Physics and MacDiarmid Institute for Advanced Materials and Nanotechnology, University of Otago, P.O. Box 56, Dunedin 9054, New Zealand
4 Technical University Dresden, Institute for Solid State and Materials Physics, 01062 Dresden, Germany
e-mail: javier.landaeta@cpfs.mpg.de

Multiple new forms of unconventional superconductivity have been discovered over the past four decades. In most cases, the strong electronic correlations and the interplay between superconductivity and other phases (magnetic ones, among others) lead to the understanding that the simple frame of electron-phonon superconductivity breaks down. In that case, complex phase diagrams with multiple superconducting phases become possible. Nevertheless, many materials only show a single superconducting phase. CeRh2As2 is one of the few materials with multiple superconducting phases [1] with a critical temperature Tc = 0.26 K. When H||c, this material shows a field-induced transition from a low field superconducting phase SC1 to a high field phase SC2 at H* = 4 T, showing an upper critical field about Hc2 = 14 T. On the other, when H||ab, only the SC1 phase is observed with Hc2 = 2 T. The large anisotropy of Hc2 and the field-induced transition were explained within the frame of the local inversion-symmetry breaking and the natural appearance of the Rashba interaction. The latter led to identifying SC1 as an even-parity and SC2 as odd-parity superconducting phase. In this talk, I will give a general overview of the superconducting properties of CeRh2As2, including a comprehensive study of the field-angular dependence of Hc2, which corroborates that the SC2 phase is an odd-parity superconducting phase [2]. Additionally, I will discuss the effect of local inversion-symmetry breaking and Rashba spin-orbit coupling by comparing CeRh2As2 with the isostructural superconductor LaRh2As2 [3].

References

[1] S. Khim & J. F. Landaeta et al., *Science*, **373**, 1012–1016 (2021)
[2] J. Landaeta et al., *Phys. Rev* .X, **12**, 031001 (2022).
[3] J. F. Landaeta et al. *Phys. Rev*. B. **106**, 014506 (2022)

Crystal growth of locally-noncentrosymmetric intermetallic compounds

Seunghyun Khim

Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany
e-mail: seunghyun.khim@cpfs.mpg.de

$CeRh_2As_2$ has been attracting attention to the unusual two-phase superconductivity [1]. Interestingly, the field-induced high-field phase is revealed to be a pseudo-spin triplet state. And staggering spin-orbit interaction given by the lack of local inversion symmetry in the Ce site is suggested to play a crucial role as the origin. This structural feature is inherent to the centrosymmetric $CaBe_2Ge_2$-type structure. As this structure promises interesting low-temperature phenomena associated with spin-orbit interactions, further exploration of intermetallic compounds in this type of the structure would be highly intriguing. In this work, we report on the crystal growth of some intermetallic compounds in the $CaBe_2Ge_2$-type structure and present their low-temperature characterizations [2].

References

[1] S. Khim, J. Landaeta *et al*, Science **373**, 1012 (2021).
[2] J. Landaeta, S. Khim *et al*., Phys. Rev. B **106**, 014506 (2022).

Real phonons needed to unravel magnetic entropy in UCu_2P_2

Ladislav Havela,[1] Dominik Legut,[2] Fuminori Honda,[3] Dariusz Kaczorowski[4]

1 Charles University, Ke Karlovu 3, Prague, Czech Republic
2 VŠB-Technical University, 17.listopadu 2172/15 Ostrava, Czech Republic
3 Tohoku University, Oarai, Ibaraki 311-1313, Japan
4 Institute of Low Temperature and Structure Research, 50-422 Wrocław, Poland
e-mail: havela@mag.mff.cuni.cz

The highest T_C in a purely 5*f* ferromagnet was found in UCu_2P_2. Its T_C = 216 K [1] can be still dramatically enhanced under pressure. It belongs to the layered Zintl phases with the trigonal structure of the $CaAl_2Si_2$ type (*P*-3*m*), formed by alternating cationic (Ca) and anionic (Al-Si) layers. Our computations show that the polar character of the bonding is a vital ingredient in reaching such high T_C. U moments are aligned along the *c*-axis. The Ising character of magnetism was, however, difficult to reconcile with heat capacity measurements. An optimum Debye model (grey dots) used to describe the phonon contribution was leaving lot of magnetic entropy at low temperatures. Therefore we performed ab-initio calculation of phonons using harmonic (magenta) and quasi-harmonic (red) approximations, which indicate that the lattice is strongly non-Debye like and there is no magnetic entropy up to ≈ 80 K. Its temperature dependence can be described by an exponential function assuming a magnon gap of $\Delta \approx 500$ K. The total magnetic entropy (inset) is however still high, pointing to local-moment character of 5*f* magnetism.

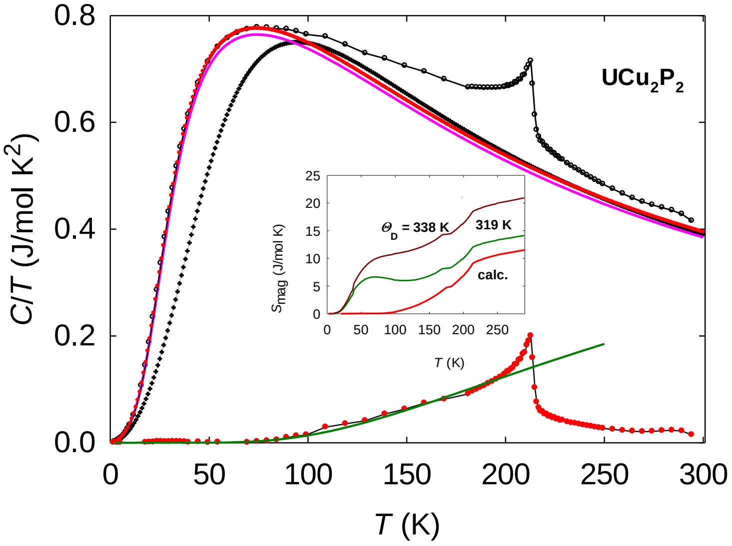


Fig.1: C/T(T) representation of experimental data, Debye background (Θ_D = 338 K) (grey), two types of calculated phonon background (magenta line – C_V, red line – C_P) and the difference of experimental and calculated phonon $C_p(T)$ data (at the bottom). The inset displays variations of magnetic entropy assuming the Debye model with two different Θ_D values and the calculated phonon heat capacity. In all cases the small electronic specific heat $C_e = \gamma^* T$, γ = 0.5 mJ/mol K^2 was subtracted for entropy determination.

This work was supported by the Grant Agency of the Czech Republic under the grant No. 21-09766S.

References

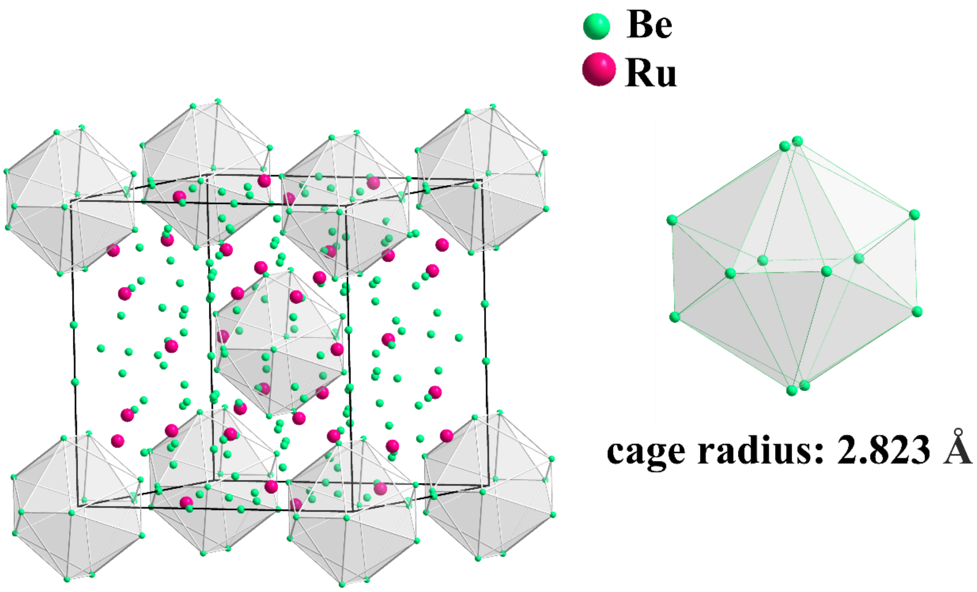
[1] D. Kaczorowski and R. Troc, *J.Phys.: Condens. Matter* **2**, 4185 (1990).

Crystal structure of $Be_{17}Ru_3A_{0.25}$ (A = Ce, Pr, Th, Gd, U, Eu, Y)

Laura Agnarelli, Yurii Prots, Marcus Schmidt, Mitja Krnel, Eteri Svanidze, Ulrich Burkhardt,
Yuri Grin, Andreas Leithe-Jasper

Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzer Str. 40, 01187 Dresden, Germany
e-mail: laura.agnarellicpfs.mpg.de

The crystal structure of the binary $Be_{17}Ru_3$ compound, was for the first time investigated by Donald E. Sands et al. in 1962. [1] In the crystal structure ruthenium atoms are found at the center of a distorted truncated tetrahedron formed by beryllium atoms. According with the literature report, the crystal structure presents a cavity in (0,0,0) position with radius that is approximately 2.82 Å. Our refinement of the crystal structure from single crystals, show that the cavity is not completely empty, but occupied by a very little amount of Ru (~2 at. %). Some preliminary results show that the cavity is large enough so that it can be occupied by a third element like lanthanides or actinides and that, depending on the atomic size of the element inserted in the cavity, the cubic lattice parameter *a*, tends to increase. The magnetic properties of the ternary compounds with Ce, U and Y were measured, indicating paramagnetic for the first two and diamagnetic for the last one behaviour.



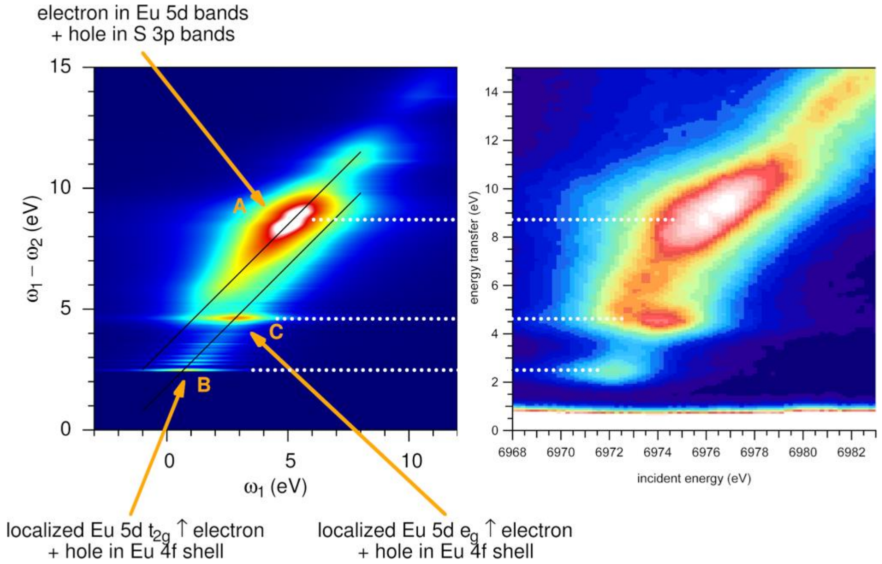
References

[1] D.E Sands et al. *Acta Crystallographica* **15**, 1191 (1962)

Localized excitons in EuS seen by valence-to-core RIXS at Eu L_3 edge

Jindřich Kolorenč,[1] Lucia Amidani[2]

1 Institute of Physics (FZU), Czech Academy of Sciences, Na Slovance 2, 182 00 Praha, Czech Republic
2 Rossendorf Beamline at the European Synchrotron, CS40220, 38043 Grenoble Cedex 9, France
e-mail: kolorenc@fzu.cz



Left: RIXS intensity found as a combination of two independent calculations using the band-theory formula [3]. One calculation assumes $4f^7$ final states at the absorbing Eu atom (feature A), the other assumes $4f^6$ final states (features B and C). Right: Measured RIXS spectrum.

We investigate the electronic structure of europium sulfide (EuS) with the aim to understand the valence-to-core resonant inelastic x-ray scattering (RIXS) spectra measured at the Eu L_3 edge. We show that the main signal (feature A in the figure) comes from the direct RIXS: an Eu 2p core electron is excited to an empty Eu 5d band above the Fermi level, and then another electron from an Eu 5d state hybridized with the S 3p bands (located below the Fermi level and hence occupied) fills back the core hole. Besides this straightforward channel, the measured RIXS spectra display two satellite features (B and C in the figure). After considering several candidates (such as excitations of the 4f shell excited by indirect RIXS processes, that is, by the interaction with the core hole in the intermediate state), we arrive at the conclusion that the satellites are excitons formed by a 4f hole and an 5d electron localized at the same Eu atom. Such excitons were suggested in the context of optical absorption a long time ago [1], but the concept is not widely accepted. It was argued that existence of these excitons is incompatible with photoconductivity measurements [2]. Our observations indicate that these arguments may need revisiting.

References

[1] T. Kasuya and A. Yanase, *Rev. Mod. Phys.* **40**, 684–696 (1968), DOI: 10.1103/RevModPhys.40.684
[2] P. Wachter and B. Bucher, *Physica B* **408**, 51–57 (2013), DOI: 10.1016/j.physb.2012.09.018
[3] J. Jiménez-Mier et al., *Phys. Rev. B* **59**, 2649–2658 (1999), DOI: 10.1103/PhysRevB.59.2649

Nuclear forensics – Fission track Analysis – Simulation for training and penetrating fluorescent colors.

Babayew Rami,[1] Yaacov Yehuda-Zada,[1] Lorincik Jan,[2] Orion Itzhak,[3] Katarivas-Levy Galit,[4]
Itzhak Halevy [1,3,*]

1 Physics Department, Nuclear Research Centre Negev, Beer-Sheva, Israel
2 Nuclear Fuel Cycle department, Research Centre Rez, Czech Republic
3 Unit of Nuclear Engineering, Faculty of Engineering Sciences, Ben-Gurion Uni. of the Negev, Israel
4 Department of Biomedical Engineering, Faculty of Engineering Sciences, Ben-Gurion Uni. of the Negev, Israel
E-mail: halevy.itzhak.dr@gmail.com

To answer nuclear forensics questions, we are developing new innovative techniques and approaches to make this analysis more reliable and accurate. Currently, only trained researchers can analyze microscope images. Since this analysis is dependent on the researcher's own abilities and skills, it is obvious that different researchers will produce results that are slightly distinct. A new worker's certification period is quite long, and it must cover numerous examples from previously measured data as well as some that we can only predict. A good simulation software can aid with training and provide a tool for grading new researchers.[1] The fission tracks were simulated by Monte-Carlo software, GEANT4, which uses all the physics behind the nuclear fission tracks, such as thermal neutrons flux, fission cross-section, radiation time, particle size, enrichment, etc. In this study, our Trainer2.0 software calculates the tracks on our Lexan detector and its projection, according to the physical parameters like neutron flax, size of the particle, the isotope, and radiation time. The result is a “star” centered on the simulated particle. Our full software is written with MatLab code.

We can simulate an extreme condition and learn new aspect in the fission track technique. From the simulation we can learn about the proper amount of material to use as a sample in the FTA technique. The simulation can predict and compared to the mini-bulk and the micro-bulk analysis. New idea of using penetrating fluorescent colors give as the ability to scan our detector in 3D instead of 2D. In this case we used the Dapi marker as a first shoot, this marker is well known for biomedical research. This new idea to investigate the FT Star more than just by his projection.

Identifying the length of the tracks and their distribution allow us to determine the element source isotope be the shape of “fission products distribution” and the density of the impurities in the source.

References

[1] Halevy I., Admon U., Chinea-Cano E., Weiss A.M., Dzigal N., E. Boblil, Dagan M., Orion I. and Radus R. Investigations, Progress in Nuclear Science and Technology, 2018, v. 5; p. 175-178 3.3.

5f Covalency from X-Ray Resonant Raman Spectroscopy

J. G. Tobin,[1,*] S. Nowak,[2] S.-W. Yu[3], R. Alonso-Mori,[2]
T. Kroll,[2] D. Nordlund,[2] T.-C. Weng,[2] and D. Sokaras[2]

1 University of Wisconsin-Oshkosh, Oshkosh, WI, USA 54901, USA
2 SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA
3 Lawrence Livermore National Laboratory, Livermore, CA, 94550, USA
e-mail: tobinj@uwosh.edu

X-Ray Resonant Raman Spectroscopy (XRRS), a variant of Resonant Inelastic X-ray Scattering (RIXS), has been used to investigate the two prototype systems, UF_4 and UO_2. [1] Both are $U5f^2$ and each is an example of 5f localized, ionic behavior and 5f localized, covalent behavior, respectively. (Figure 1) From the M_5 XRRS measurements, the 5f band gap in each can be directly determined and, moreover, a clear and powerful sensitivity to 5f covalency emerges.

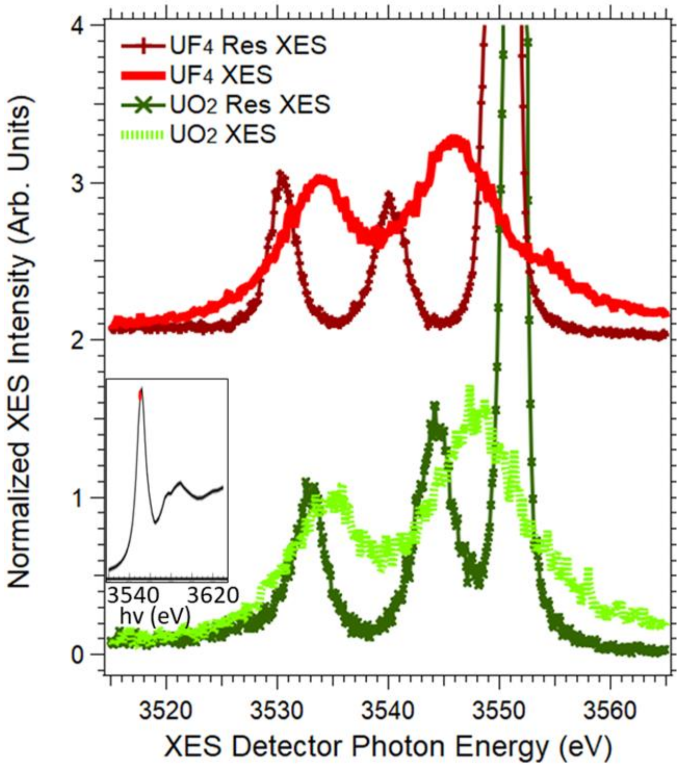


Figure 1 Shown here are the XRRS (Res XES) and off resonance XES spectra for UF_4 and UO_2. Each spectrum is normalized to unity peak height at the lowest energy peak. The UF_4 spectra are shifted upwards by 2 units, to aid in comparison. Inset: This is UF_4 M_5 XAS, with the red dot showing the excitation energy for Res XES (XRRS), at the maximum of the 5f x-ray absorption. hv is the photon energy.

XES = X-ray Emission Spectroscopy. XAS = X-Ray Absorption Spectroscopy.

References

[1] J. G. Tobin, S. Nowak, S.-W. Yu, R. Alonso-Mori, T. Kroll, D. Nordlund, T.-C. Weng, D. Sokaras, "5f Covalency from X-Ray Resonant Raman Spectroscopy," J. Phys. Cond. Matter (2022), https://iopscience.iop.org/article/10.1088/1361-648X/ac9bbd.

Unravelling the electronic structure of uranium: the case of MUO_3 (M = K, Na and Rb)

S. Orlat[1], R. Bes[2], F. Tuomisto[2], P. Martin[1], P. Moisy[1], G. Leinders[3], K. Kvashnina[4,5]
1 CEA, DES, ISEC, DMRC, University of Montpellier, Marcoule, France
2 Department of Physics, University of Helsinki, P.O. Box 64, FI-00014 Helsinki, Finland
3 Belgian Nuclear Research Centre (SCK CEN), Institute for Nuclear Materials Science, Boeretang 200, B-2400 Mol, Belgium.
4 The Rossendorf Beamline at ESRF, The European Synchrotron, CS40220, 38043 Grenoble Cedex 9, France
5 Institute of Resource Ecology, Helmholtz Zentrum Dresden-Rossendorf (HZDR), PO Box 510119, 01314 Dresden, Germany
e-mail: simon.orlat2@cea.fr

Uranium is a key chemical element of the nuclear industry and has a complex chemistry due to the large number of oxidation states allowed by its $[Rn]7s^2 6d^1 5f^3$ ground state electronic configuration. Even if this element has been the focus of substantial research, its electronic structure remains difficult to study theoretically due to the same magnitude of the crystal field splitting, spin orbit coupling and electron-electron interaction. The core of this contribution, is to present recent effort made to overcome this difficulty by studying U(V) compounds such as alkali metal uranates KUO_3, $NaUO_3$ and $RbUO_3$. More precisely, the analysis of high energy resolution fluorescence detected data obtained at the U-L_3 edge measuring the $L\beta_5$ emission line for enhanced spectral resolution [1,2] will be discussed in the light of new theoretical calculation results obtained using the Finite Difference Method for Near-Edge Structure (FDMNES) code [3] as illustrated in [4]. As shown by recent results obtained on KUO_3 [4] and on $NaUO_3$ [5], such combined experimental and theoretical approach allows to better understand the impact on the uranium valence electronic structure induced by the change in the U-O interatomic distances and the associated local structural distortions.

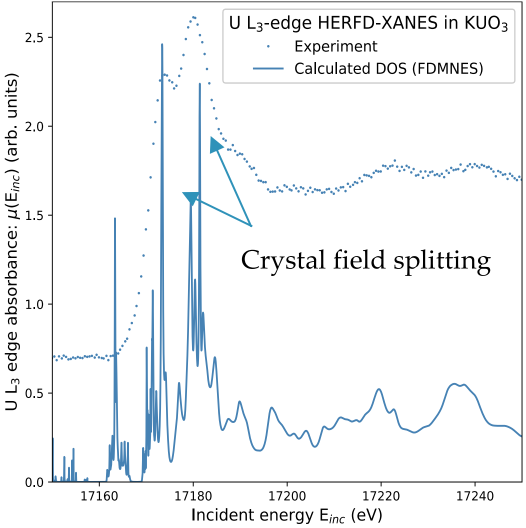


Figure 2: uranium L_3-edge HERFD-XANES in KUO_3 and corresponding simulated spectrum using FDMNES [3]

References

[1] K. O. Kvashnina et al., J. Electron Spectrosc. Relat. Phenom. 194, 27 (2014).
[2] K. O. Kvashnina et al., Anal. Chem. 87, 8772 (2015).
[3] O. Bunău et al., J. Phys. Condens. Matter. 21, 4260 (2009).
[4] R. Bes et al., J. Synch. Rad. 29, 21 (2022) 21.
[5] S. M. Butorin et al., Chem. Eur. J. 22, 9693 (2016).

Large-Scale Quantum Mechanical Simulations of Actinide Oxides

Nabeel Anwar[1], Robert M. Harker[2], Mark T. Storr[2], Marco Molinari[3], Chris-Kriton Skylaris[1]
1 School of Chemistry, University of Southampton, Southampton, SO17 1BJ, UK
2 AWE, Aldermaston, Reading, RG7 4PR, UK
3 Department of Chemical Sciences, University of Huddersfield, Huddersfield, HD1 3DH, UK
e-mail: na5g20@soton.ac.uk

Actinide oxides (AnO_2) play an important role within the nuclear fuel cycle, due to their thermal and chemical stability. Irradiation-induced defects and the formation of fission products changes the composition of these materials over time. Neutrons and charged particles produced by the fission process can collide with and displace thousands of atoms in the AnO_2 lattice. The formation of defects influences the physical properties of actinide oxide materials, degrading their thermal and mechanical performance as a nuclear fuel [1].

Experimental studies of AnO_2 materials are difficult due to their high toxicity and radioactivity. Neutron or X-ray diffraction requires relatively large volumes of material and electron microscope experiments enhance the thermal motion the of defects [2, 3]. Computational studies of such materials include the application of interatomic potentials to study defect behaviour, but the quality of the predictive capability can be dependent upon the data used in the fitting process. In contrast density functional theory (DFT) does not rely upon such a fitting process and as such provides a 1st principles approach to simulate the properties of AnO_2 materials. However, conventional DFT methods are limited to calculations to just tens of atoms by unfavourable (cubic) scaling of computational effort with the number of atoms.

The development of linear-scaling DFT methods within the ONETEP package [4], allows simulations approaching thousands of atoms in size. We present large-scale DFT simulations of AnO_2 materials, investigating defects at different concentrations, for which formation energies are calculated. Our investigation may then be extended, to explore transport mechanisms of intrinsic and extrinsic defects in the material.

References

[1] H. Matzke, *Radiation Effects in Solids*, Springer, Dordrecht, 2007, ch.14, pp. 401-420.
[2] Y. Satoh, H. Matsui, and T. Hamaoka, *Phys. Rev. B*, 2008, **77**, 094135.
[3] Y. Matsukawa and S. J. Zinkle, *Science*, 2007, **318**, 959-962.
[4] J. C. A. Prentice, J. Aarons, J. C. Womack, A. E. A. Allen, L. Andrinopoulos, L. Anton, R. A. Bell, A. Bhandari, G. A. Bramley, R. J. Charlton, R. J. Clements, D. J. Cole, G. Constantinescu, F. Corsetti, S. M.-M. Dubois, K. K. B. Duff, J. M. Escartín, A. Greco, Q. Hill, L. P. Lee, E. Linscott, D. D. O'Regan, M. J. S. Phipps, L. E. Ratcliff, Á. Ruiz Serrano, E. W. Tait, G. Teobaldi, V. Vitale, N. Yeung, T. J. Zuehlsdorff, J. Dziedzic, P. D. Haynes, N. D. M. Hine, A. A. Mostofi, M. C. Payne, C.-K. Skylaris, *J. Chem. Phys.*, 2020, **152**, 174111.

Stability studies of the AmSel extraction system

P. Vacas-Arquero[1], I. Sanchez-García[1], L.J. Bonales[1], H. Galán[1]

1 Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT)
e-mail: Pablo.Vacas@ciemat.es

One of the most sustainable solutions toward the proper management of nuclear waste is the so-called Advanced Nuclear Fuel Cycle. This requires the separation of the most relevant minor actinides (MA: Np, Am, and Cm) from fission products like lanthanides (Ln) in order to create new fuel or transmutation blanks for advanced nuclear reactors [1]. Within the MA recovery, an important interest of the recovery of only Am is increasing due to the complication in the fabrication of new fuel containing Cm [2]. The separation of Am from Cm continues being one of the current challenges due to their similar chemical behaviour, reason why new strategies are being designed and developed. This is the case of the AmSel (Americium Selective Extraction) process, based on liquid-liquid extraction methodologies and designed to work after the recovering of uranium and plutonium by PUREX process [3].

The AmSel process implies a first step (co-extraction step) where the Ln(III) and An(III) from PUREX raffinate are co-extracted by using an organic phase containing a TODGA based solvent. Then, a selective stripping of Am(III) using a water-soluble BTBP separates Am(III) from Cm(III) and the Ln(III) thanks to the inverse selectivity of BTBP ligands and TODGA towards Am and Cm (stripping step) [3]. A limiting point for the development of these processes is their degradation resistant to the harsh operation conditions (the high [HNO_3] where nuclear fuel is dissolved and the high radioactive field), since its degradation may produces undesirables effects such as loss of selectivity, third phase formation, etc.

In this work, it is presented the stability studies performed for the reference stripping step of the AmSel process, simulating the most relevant process conditions. For that, the extraction behavior and the composition after gamma radiation has been evaluated by gamma-ray and alpha spectrometry (using ^{152}Eu, ^{241}Am and ^{244}Cm) and by HPLC-MS and Quantitative Raman Spectroscopy, respectively.

References

[1] Baron, P. et al., *Progress in Nuclear Energy* **117**, 103091 (2019).
[2] Geist, A. et al., *Separation science and technology* **56.11**, 1866-1881 (2021).
[3] Wagner, C. et al., *Solvent Extraction and Ion Exchange* **34.2**, 103-113 (2016).

The U-Cr-O ternary system: preliminary experimental results of the thermodynamic investigation.

Jinjiang Cui[1,2,3], Pierre Benigni[2], Roland Ducher[1], Marc Barrachin[1], Olivier Tougait,[3]

1 Institut de Radioprotection et Sûreté Nucléaire, Service des Accidents Majeurs, 13115, Saint Paul Lez, Durance, France
2 Aix Marseille Univ, Université de Toulon, CNRS, IM2NP, Marseille, France
3 Univ. Lille, CNRS, Centrale Lille, Univ. Artois, UMR 8181 - UCCS - Unité de Catalyse et Chimie du Solide, F-59000, Lille, France
e-mail: olivier.tougait@univ-lille.fr

Metal doped UO_2 fuels are regarded with high interest to improve both the efficiently, in terms of burn-up and safety of nuclear system in light water reactors. These additives promote the crystalline growth of the grains yielding in the improvement of the fission gas accommodation and reduction of the fuel swelling. Among the various metals which have been considered, chromium oxide as additive is regarded of prim interest for its benefit on the microstructural and mechanical properties of sintered pellets. Despite numerous studies dedicated to Cr-U-O materials and transformations regarding preparation [1, 2], phase equilibria and stability [2, 3], physical [4, 5], mechanical [6] and chemical [7, 8] properties, the investigations of the thermodynamic features remain rather limited and this even though they constitute relevant data for in-use conditions (control of the O partial pressure) and for the so-called accidental scenarios to anticipate the consequences of loss of the coolant accident (LOCA).

The present project aims to optimize with the help of the Calphad method the ternary U-Cr-O phase diagram in the whole concentration range which is not available in the open literature to the best of knowledge to the authors. To do so, a well-established procedure based on a chain of steps is applied among which the 2 first legs, (i) a critical assessment of the thermodynamic data and (*ii*) our preliminary experimental results regarding the phase relations, Cr solubility in uranium oxides and some thermal properties, will constitute the heart of the presentation.

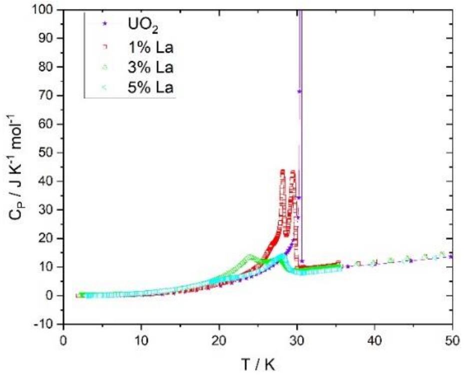
References

[1] P. Kegler, *et. al.*, Materials, 2021, 14, 6160.
[2] X. Guo, *et. al.*, Dalton Trans., 2016, 45, 4622.
[3] A. Leenaers, *et. al.*, J. Nucl. Mater.,2003 317, 62.
[4] M.W.D. Cooper *et. al.*, Acta Mater., 2018, 150 , 403.
[5] L. Bourgeois, *et. al.*, J. Nucl. Mater., 2001, 295, 73.
[6] R.W. Rice, J. Mater. Sci. 1997, 32, 3071.
[7] C. Riglet-Martial, *et. al.*, J. Nucl. Mater., 2014, 447, 63.
[8] H. Smith, et al., Nature Chemistry, 2022, 5, 163.

Low-temperature heat capacity of the lanthanide-doped UO_2

Octavian S. Vălu, Jean-Christophe Griveau, Jean- François Vigier, Karin Popa,Eric Colineau, Rudy J.M. Konings

European Commission, JRC Karlsruhe, Hermann-von-Helmholtz-Straße 1, 76344 Eggenstein-Leopoldshafen, Germany
e-mail: octavian.valu@ec.europa.eu



Substitution of atoms in the uranium dioxide fluorite crystal lattice can affect their thermodynamic properties. Replacement of atoms with different characteristics can lead to lattice modification and disorder, affecting the phonon states and their distribution with direct impact on the thermal behavior of the material.

The antiferromagnetic transition of UO_2 occurs at 30.44 K [1] as a λ-type anomaly in the low-temperature heat capacity. Previous measurements have shown [2] that the tetravalent ions substitution on the uranium sublattice has a strong impact on the transition peak even at low concentrations. When adding trivalent ions (Am^{3+}) in the crystal structure [3], a more complicated lattice is obtained with a higher degree of disorder due to the M^{3+}/U^{5+} charge compensation, all leading to an evident change in shape and position of the anomaly. On one hand, the matrix dilution and on the other one the radiation damage, both have a strong impact even at low concentrations and small-accumulated radiation doses.

In the current work, the low-temperature heat capacity measurements of UO_2 doped with La and Nd were performed by calorimetry [4]. The obtained results show that the substitution of trivalent ions lead to changes that are more complex. Aside the reduced peak intensity and shift, the λ-shaped antiferromagnetic transition of UO_2 is also altered by a split of the peak (figure 1). Also the effect of substitution of a divalent ion is investigated.

References

[1] J. J. Hunzicker and E. F. Westrum Jr., *J. Chem. Thermodyn.* **3**, 61 (1971).
[2] O. S. Vălu, E. De Bona, K. Popa, J.-C. Griveau, E. Colineau and R. J. M. Konings, *Sci. Rep.* 9, 15082 (2019).
[3] O. S. Vălu, O. Beneš, E. Colineau, J.-C. Griveau and R. J. M. Konings, *J. Nucl. Mater.* **126–134**, 507 (2018).
[4] J.-C. Griveau, J.-F. Vigier, K. Popa, S. O. Vălu, E. Colineau and R. J. M. Konings, *J. Appl. Phys.* **132**, 185103 (2022).

Magnetic, thermodynamic and transport properties of $PuAlO_3$

Jean-Christophe Griveau, Karin Popa, Jean-Francois Vigier, Eric Colineau, Dragos Staicu, Ondrej Benes, Olaf Walter, Rudy J.M. Konings

European Commission, Joint Research Centre, P.O. Box 2340, D-76125 Karlsruhe, Germany

Here we report properties at low and high temperature properties of polycrystalline $PuAlO_3$. This perovskite compound has been recently reported with an orthorhombic configuration (space group Imma) [1], similarly to $CeAlO_3$ and $PrAlO_3$ lanthanide analogues. It is also an impurity phase reported in Pu-Al systems and more specifically in spin-fluctuator $PuAl_2$ [2] possibility at the origin of magnetic transitions above T_{sf}.

Several well structurally characterized polycrystalline samples of $PuAlO_3$ have been examined by DC magnetization (MPMS-3 Quantum Design) showing a modified Curie Weiss law from 300 K down to 50 K. An antiferromagnetic transition T_N is clearly visible around 15 K

Heat capacity using PPMS-9T Quantum Design has been determined down to 6 K and up to room temperature. Self-heating of ^{239}Pu isotope has prevented accessing very low temperature features of the compound. The antiferromagnetic transition is confirmed also at 15 K and slightly decreases in temperature with magnetic field applied up to 9 T.

Interestingly, an anomaly of unknown origin is visible around 140 -150 K and confirmed simultaneously by heat capacity and magnetization measurements on several samples without any change of the crystallographic (4-circles single crystal Diffractomer Bruker).

Finally, high temperature heat capacity (DHTC 96 type), thermal diffusivity (Home Made Laser Flash) and thermal conductivity derived from these results up to 1700 K are presented and compared with calculated data available in the literature for rare earth and actinide aluminates.

References

[1] Vigier JF, Popa K, Martel L, Manara D, Dieste Blanco O, Freis D, Konings RJM. Plutonium and Americium Aluminate Perovskites. Inorg Chem. 2019 Jul 15; 58(14):9118-9126.

[2] A. J. Arko, F. Y. Fradin, and M. B. Brodsky, Magnetic, Transport, and Nuclear-Magnetic-Resonance Properties of U1−xPuxAl2, Phys. Rev. B 8, 4104.

Poster #1

Magnetism and magnetoacoustics of $U_3Ru_4Al_{12}$ single crystal in high magnetic fields

A.V. Andreev,[1] D.I. Gorbunov[2], T. Nomura[2], S. Zherlitsyn[2]

[1]*Institute of Physics, Academy of Sciences, Na Slovance 2, 182 21 Prague, Czech Republic,*
[2]*Hochfeld-Magnetolabor Dresden (HLD-EMFL), Helmholtz-Zentrum Dresden-Rossendorf,*
01328 Dresden, Germany
e-mail: a.andreev@seznam.cz

Competing interactions in geometrically frustrated compounds attract a lot of attention due to a rich variety of phases and their interesting physics. Applied magnetic field can act as a tuning parameter between different magnetic states. Frustrated compounds often exhibit field-induced anomalies and offer a rich playground for studying competing interactions. $U_3Ru_4Al_{12}$ crystallizes in a hexagonal crystal structure of $Gd_3Ru_4Al_{12}$ type (space group $P6_3/mmc$). The U atoms form a distorted kagome lattice in the basal plane. $U_3Ru_4Al_{12}$ orders into a non-collinear antiferromagnetic structure at T_N = 9.5 K with the U moments confined to the basal plane.

We report on magnetization and ultrasound measurements of $U_3Ru_4Al_{12}$ single crystal in static and pulsed magnetic fields up to 58 T. At 2 K, the magnetization, M, shows a significant anisotropy between the basal plane and the [001] axis. For field applied along [100] and [120], M displays small field-induced anomalies below 20 T. The $\mathrm{d}M/\mathrm{d}H$ derivative shows maxima between 10 and 20 T. At higher fields, the magnetization approaches a saturation of approx. 3.6 μ_B per formula unit, or 1.2 μ_B per U atom. No anomalies are observed for field applied along the [001] axis.

The relative-sound velocity changes, $\Delta v/v$, for a transverse acoustic mode (propagation $\boldsymbol{k} \parallel [100]$, polarization $\boldsymbol{u} \parallel [001]$) also show anomalies in applied field. When field is applied along the [100] direction, $\Delta v/v$ displays a total softening of 0.4%. A maximum and a small dip can be resolved at 4 and 12 T, respectively. For field applied along the [120] direction, the sound velocity displays hardening. One observes a minimum at 4 T and a change of slope at 20 T. All anomalies in ultrasound correlate with $M(H)$.

Our data show a significant magnetoelastic coupling in $U_3Ru_4Al_{12}$. We followed the temperature evolution of the field-induced anomalies in magnetization and ultrasound. A magnetic phase diagram of $U_3Ru_4Al_{12}$ will be presented. A large number of phases likely indicates that a geometric frustration plays an important role in the physics of $U_3Ru_4Al_{12}$.

Poster #2

Corrosion modeling of spent fuel – adsorption of O, O_2, H_2O and H_2O_2 to the UO_2 surface

Ine Arts,[1] Rolando B. Saniz[2], Selma Mayda[1], Gianguido Baldinozzi[3], Gregory Leinders[4], Marc Verwerft[4], Dirk Lamoen[1]

1 EMAT, Physics Department, University of Antwerp, B-2020 Antwerpen, Belgium
2 CMT, Physics Department, University of Antwerp, B-2020 Antwerpen, Belgium
3 Université Paris-Saclay, CentraleSupélec, CNRS, SPMS, F-91190 Gif-sur-Yvette, France
4 Belgian Nuclear Research Centre (SCK CEN), Institute for Nuclear Materials Science, B-2400 Mol, Belgium
e-mail: ine.arts@uantwerpen.be

Uraniumdioxide (UO_2) is the main component of nuclear fuels. This compound is very susceptible to oxidation, which results in formation of the mixed-valence oxide U_3O_8.[1] From an application point of view, this interest relates to the safe and sustainable management of nuclear fuel. Our goal is an understanding of this oxidation process at the atomic level, using ab initio electronic structure calculations.

The focus in this work lies on the early oxidation of stoichiometric UO_2, where excess oxygen can be treated as interstitial point defects. In order to study this oxidation we investigate the adsorption of the O atom and oxygen containing molecules (O_2, H_2O, H_2O_2) on the three most stable UO_2 surfaces ((111), (110) and (100)). The calculations are performed within DFT, using PBE+U including spin-orbit coupling and taking into account the non-collinear 3k (transverse) magnetic ground state.

Dissociative adsorption is found for H_2O and H_2O_2 on the (111) and (100) UO_2 surface respectively, forming hydroxyl groups on the surface. Adsorption on the UO_2 surface lowers the barrier for dissociation of O_2. Defect formation energy is calculated for interstitial oxygen, up to several layers into the material starting from the surface. Diffusion energy barriers are presented. Additionally, adsorption energies are studied as a function of temperature and partial pressure.

References

[1] E. De Bona, K. Pope, O. Walter, M. Cologna, C. Hennig, A. C. Scheinost, and D. Prieur., *Inorg. Chem.* **61**, 1843 (2020).

Poster #3

Modelling of the crystalline scintillating materials from the first principles

Anna Ivanova, Anatoli I. Popov, Andrew Chesnokov,
Aleksander Platonenko, and Dmitry Bocharov

Institute of Solid State Physics, University of Latvia, Kengaraga 8, LV-1063, Latvia
e-mail: dmitrijs.bocarovs@cfi.lu.lv

$Y_2Si_2O_5$ (YSO) is one of the scintillator materials that is successfully used in Compact Muon Solenoid (CMS) detectors in the Large Hadron Collider. YSO can be modified by substituting Y^{3+} metal ion(s) of elementary formula with Lu^{3+} ions ($Lu_xY_{1-x}SiO_5$, Lu_2SiO_5), also adding Ce^{3+} impurity to a crystalline lattice in various concentrations. Oxy-orthosilicate materials exhibit high stability against radiation and favourable luminescence properties. [1]

In this work, density functional theory (DFT) methods with LCAO approximation were applied as implemented in CRYSTAL17 code. [2]

Computational models of YSO as an ideal and Ce-doped crystals were made to describe and predict their structural and electronic properties. Corresponding crystalline structures were described in detail on atomic and electronic levels; infra-red (IR) and Raman spectra were calculated also.

Obtained data was compared to the data available in the literature. As a conclusion, the developed computational model well reproduces structural and electronic properties of YSO and is in a good agreement with the literature. Further, this model can be used to research YSO-like structures with various defects incorporated.

This research is supported by Latvian Research Program “High-energy physics and accelerator technologies” (Agreement No: VPP-IZM-CERN-2020/1-0002).

References

[1] T. Yanagida, Inorganic Scintillating Materials and Scintillation Detectors, Proceedings of the Japan Academy. Ser. B: Physical and Biological Sciences **94**, 75 (2018).
[2] R. Dovesi, A. Erba, R. Orlando, C. M. Zicovich-Wilson, B. Civalleri, L. Maschio, M. Rerat, S. Casassa, J. Baima, S. Salustro, B. Kirtman. WIREs Comput Mol Sci. **8**, e1360 (2018).

Poster #4

Electronuclear Quantum Criticality

J. Banda[1], D. Hafner[1], J. Landaeta[1], E. Hassinger[1], K. Mitsumoto[2], M. Giovannini[3], J. Sereni[4], C. Geibel[1] and M. Brando[1]

1 Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany
2 Liberal Arts and Sciences, Toyama Prefectural University, Imizu, Toyama 939-0398, Japan
3 Department of Chemistry and Industrial Chemistry (DCCI), University of Genova, 16100 Genova, Italy
4 Department of Physics, CAB-CNEA, CONICET, 8400 San Carlos de Bariloche, Argentina
e-mail: brando@cpfs.mpg.de

We present here a rare example of electronuclear quantum criticality in a metal. The compound $YbCu_{4.6}Au_{0.4}$ is located at an unconventional quantum critical point (QCP). In this material the relevant Kondo and RKKY exchange interactions are very weak, of the order of 1K. Furthermore, there is a strong competition between antiferromagnetic and ferromagnetic correlations, possibly due to geometrical frustration within the fcc Yb sublattice. This causes strong spin fluctuations which prevent the system to order magnetically. Because of the very low Kondo temperature the Yb^{3+} 4*f*-electrons couple weakly with the conduction electrons allowing the coupling to the nuclear moments of the ^{171}Yb and ^{173}Yb isotopes to become important. Thus, the quantum critical fluctuations observed at the QCP derive not from purely electronic states but from entangled electronuclear states. This is evidenced in the anomalous temperature and field dependence of the specific heat at low temperatures.

Poster #5

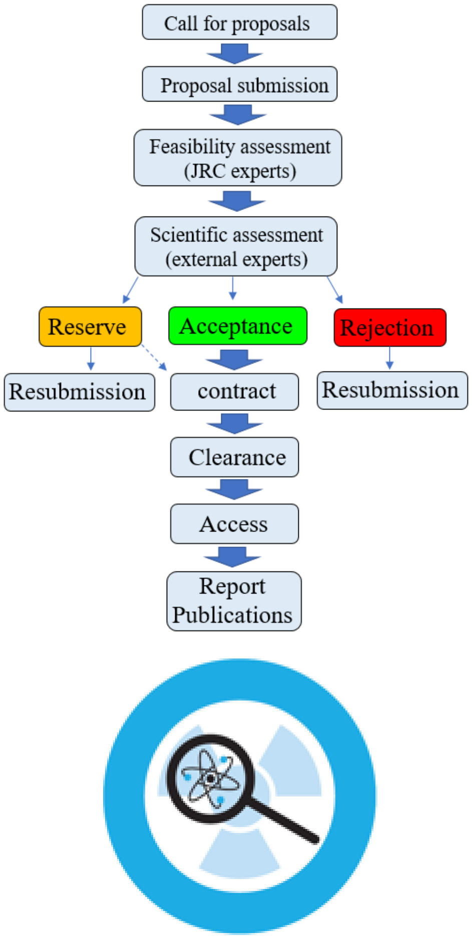
Laboratories for Actinides

Eric Colineau, Konstantinos Boboridis, Laura Aldave de las Heras, Rachel Eloirdi

European Commission, Joint Research Centre (JRC), Karlsruhe, Germany, e-mail: JRC-ACTUSLAB-KARLSRUHE@ec.europa.eu

Large research infrastructures are expensive to build and operate and are often beyond the resources of a single institution or country. In particular, nuclear research infrastructures need an expensive licence and access to strategic materials.

JRC operates licensed laboratories in Karlsruhe, where actinide materials including transuranics can be handled within a safe and secure infrastructure and their chemical and physical properties explored. The "Actinide User Laboratory" (ActUsLab) provides free access for external scientists from eligible countries to three laboratories : PAMEC (Properties of Actinide Materials under Extreme Conditions), FMR (Fuels and Materials Research) and HC-KA (Hot Cells).



Call for proposals

Proposal submission

Feasibility assessment (JRC experts)

Scientific assessment (external experts)

Reserve

Acceptance

Rejection

Resubmission

contract

Resubmission

Clearance

Access

Report Publications

Figure 1: Procedure for Open Access to ActUsLab

Acknowledgements

We are grateful to Ms. Krisztina Varga for administrative support to the programme and to the colleagues of PAMEC, FMR and HC-KA facilities for scientific and technical support to Users. We also thank the support Units of JRC-Karlsruhe for making the access possible.

References

[1] Actinide User Laboratory (ActUsLab): joint-research-centre.ec.europa.eu/knowledge-tools-laboratories/open-access-jrc-research- infrastructures/actinide-user-laboratory-actuslab_en

[2] JRC Open Access web portal: https://joint-research-centre.ec.europa.eu/knowledge-tools-laboratories/open-access-jrc-research-infrastructures_en.

Poster #6

Evolution of local structure across the heavy rare earth zirconate oxides, $A_2Zr_2O_7$

Ross H. Colman,[1] Milan Klicpera[1], Kristina Vlášková[1]

1 Charles University, Ke Karlovu 5, 121 16, Prague 2, Czech Republic
e-mail: ross.colman@mag.mff.cuni.cz

Oxide ion conductors require migration pathways for ions to pass through the lattice, typically via hopping through vacancy sites. The ease of this migration is dictated by both the bonding strength, and the structure (relative vacancy locations), with these characteristics guiding the ion conductivity and subsequently the minimum useful working temperature. The rare-earth oxides, with general formula $A_2B_2O_7$ (where A^{3+} is a rare-earth ion, and B^{4+} is a transition or p-block metal), have shown reasonable ion conductivity properties. These oxides show distinct structure types dependent on the A/B ion size ratio: $r_A/r_B > 1.46$ result in an ordered pyrochlore structure, and $r_A/r_B < 1.46$ results in a disordered defect-fluorite structure [1]. Probes of the ion local structure (spectroscopic methods), however, don't show an abrupt change across this distinct *structural-border* [2]. Understanding the subtleties of the structural changes through this family is important for explaining the observed ion-conductivities [3].

In this poster I will show the results of our recent efforts to use a combination of neutron and X-ray diffraction, with small box pair-distribution-function analysis (xPDF and nPDF) to better understand the local structure evolution across the heavy rare-earth zirconates, $A_2Zr_2O_7$ (Fig 1).

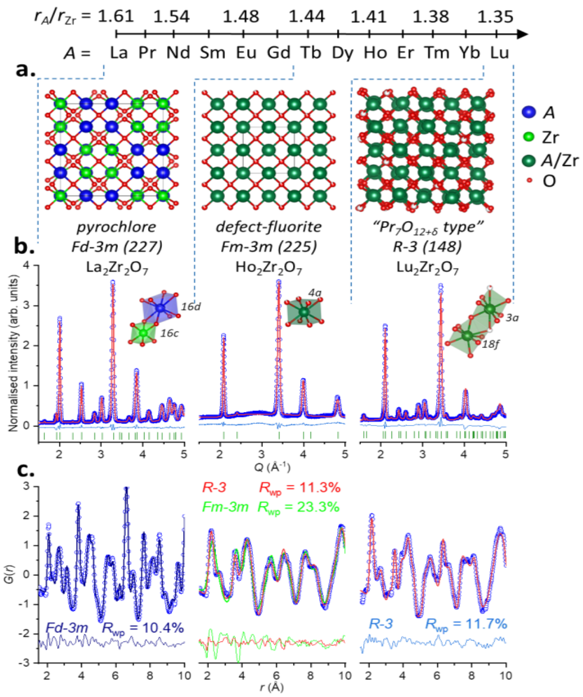


Fig 1. The rare-earth zirconate series, $A_2Zr_2O_7$, showing: ***a.*** *the long-range average structures;* ***b.*** *refined powder neutron diffraction data (with inset cation environments);* ***c.*** *nPDF data with fits.*

References

[1] J. Shamblin, M. Feygenson, J. Neuefeind, et al., Nat. Mater. **15**, 507 (2016).
[2] P. E. R. Blanchard, R. Clements, B. J. Kennedy, et al., Inorg. Chem. **23**, 41 (2012).
[3] M. Shafique, et al., *J. Alloys Compd*. **671**, 226 (2016).

Poster #7

Real Topologically Charged Nodal Lines in 2D

Xiaolong Feng,[1,2] Weikang Wu[3], Zhi-Ming Yu[4], Yu Xin Zhao[5], Shengyuan A. Yang[2]

1 Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 40, 01187 Dresden, Germany
2 Singapore University of Technology and Design, 8 Somapah Rd., 487372 Singapore
3 Shandong University, 250061 Jinan, China
4 School of Physics, Beijing Institute of Technology, 100081 Beijing, China
5 Department of Physics, Nanjing University, 210093 Nanjing, China
e-mail: xiaolong.feng@cpfs.mpg.de

Different from Weyl points, conventional nodal lines are not stable against perturbations. Here, we demonstrate the realization of $\mathbb{Z}$-charged nodal lines in 2D, which is robust against weak perturbations. Such nodal lines can only be canceled out by merging nodal lines of opposite topological charge, which is featured by a local π_1 charge. The local π_1 charge can be interpreted as the topological charge of the linking Weyl nodes. Besides, a nontrivial topological charge of the global path traversing BZ can be obtained to capture the topological edge states, connecting the projections of linking Weyl nodes. Moreover, multiple charged nodal lines can be obtained by merging nodal lines of the same chirality as Weyl points. An interesting concentric structure is constructed to understand the topologically charged nodal line.

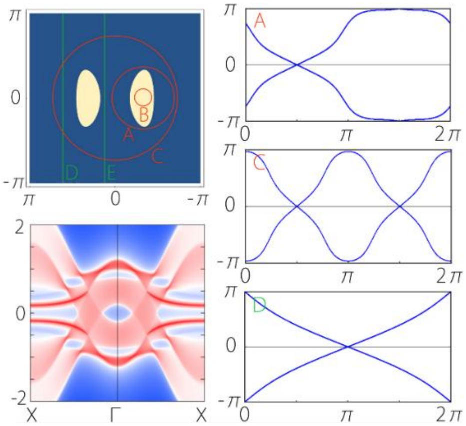


Fig. 1 Topological Nodal Lines featured by double topological charges, π_0 charge and π_1 charge. The local π_1 charge can protect single nodal line from perturbations. They can be canceled out only by merging with nodal lines of opposite π_1 charge.

References

[1] Fang et al., *Phys. Rev. B* 92, 081201 (R) (2015).
[2] Tomas et al., *Phys. Rev. B* **96**, 155105 (2017).
[3] Ahn et al., *Phys. Rev. Lett.* **121**, 106403 (2018).
[4] Wang et al., *Phys. Rev. Lett.* **125**, 126403 (2020).

Poster #8

Magnetic phase separation in $EuPdSn_2$

M. Giovannini,[1] A. Martinelli[2], D.H. Ryan[3], J.G. Sereni[4], C. Ritter[5], A. Leineweber[6], I. Čurlik[7], R. Freccero[1]

1 Department of Chemistry, University of Genova, Via Dodecaneso 31, 16146 Genova, Italy
2 SPIN-CNR, Corso F.M. Perrone 24, 16152 Genova, Italy
3 Physics Department and the Centre for the Physics of Materials, McGill Universit, Quebec, Canada
4 Department of Physics, CAB-CNEA, CONICET, 8400 San Carlos de Bariloche, Argentina
5 Institut Laue -Langevin, 71 Avenue des Martyrs, 38042 Grenoble Cedex 9 – France
6 Inst. of Mater.Science, TU Bergakademie Freiberg, Gustav-Zeuner-Str. 5, D-09599 Freiberg, Germany
7 Faculty of Humanities and Natural Sciences, University of Prešov, 17. novembra 1, Prešov, Slovakia
e-mail: mauro.giovannini@unige.it

In the course of our systematic investigation on Eu-Pd-Sn, several novel compounds have been discovered, namely Eu_2Pd_2Sn, $EuPd_2Sn_4$ and $EuPdSn_2$[1-3]. A common feature shown by these (and other analogous) compounds is the complex magnetic behavior and magnetic anisotropy despite these systems have L = 0 and no crystal electric field effects. Neutron diffraction is by far the best method to determine magnetic structures. However, in case of europium these measurements are hampered by the rather large adsorption cross section for thermal neutrons. In our study to overcome this problem a large-area-flat-plane geometry was used to collect neutron diffraction patterns. Here we combine ^{151}Eu Mössbauer spectroscopy and neutron powder diffraction to show that antiferromagnetic and ferromagnetic domains are in competition and coexist in the ground state of $EuPdSn_2$ as supported also by theoretical calculations. This study is complemented by a chemical bonding analysis in positional space which revealed unusual features such as Eu-Pd polar covalent interactions building up graphite-like distorted honeycomb layers and an almost neutral Sn as revealed by QTAIM effective charges.

References

[1] M. Giovannini et al, *Inorg. Chem.* **60**, 8085 (2021).
[2] I. Čurlik et al. Acta Physica Polonica A, **131**, 1003 (2017).

Poster #9

Crystal, electronic structure and physical properties of the $U_3Rh_4Ge_{13}$ and $Th_2Rh_3Ge_5$ metallic systems

Volodymyr Levytskyi,[1] Eteri Svanidze[2], Andreas Leithe-Jasper[2], Roman Gumeniuk[1]

1 TU Bergakademie Freiberg, Leipziger Str. 23, 09596 Freiberg, Germany
2 Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzer Str. 40, 01187 Dresden, Germany
e-mail: volodymyr.levytskyi@physik.tu-freiberg.de

Crystal structure of the rhombohedral (space group $R3c$) Remeika phase $U_3Rh_4Ge_{13}$ adopting the HT-$Y_3Pt_4Ge_{13}$ structure type has been refined by powder x-ray diffraction (PXRD) method (unit cell parameters a = 5.6004(6) Å, c = 15.5158(1) Å, reliability factors R_B = 0.022, R_p = 0.021). The shortest interatomic distances observed are mainly close to the sums of atomic radii [i.e. d(U-Ge) = 2.98 Å, d(Rh-Ge) = 2.57 Å, d(Ge-Ge) = 2.46 Å] and d(U-U) = 4.468 Å exceeds significantly Hill limit. Despite an absence of the isostructural $Th_3Rh_4Ge_{13}$ phase, a new compound $Th_2Rh_3Ge_5$ has been found to form in the Th-Rh-Ge system. The full-profile PXRD refinements of $Th_2Rh_3Ge_5$ confirmed its isomorphism with $U_2Co_3Si_5$ structure type (sp. gr. $Ibam$, a = 10.0572(1) Å, b = 11.9142(1) Å, c = 6.0997(1) Å, R_B = 0.027, R_p = 0.037). The structural relationships of $U_3Rh_4Ge_{13}$ with the primitive cubic $Yb_3Rh_4Sn_{13}$, and of $Th_2Rh_3Ge_5$ with the body-centred tetragonal $ThCr_2Si_2$ structure types are discussed.

The ac- and dc-magnetic susceptibility measurements on $U_3Rh_4Ge_{13}$ sample revealed an antiferromagnetic ordering at T_N = 22 K followed by a metamagnetic transition below 5 K, which agrees well with earlier report [1]. Long-range magnetic ordering is reflected by λ-anomally in the specific heat capacity temperature dependence. Both experimental and theoretical calculations hint toward localized nature of 5f-states in the studied germanide.

As follows from the electrical resistivity and thermopower characteristics, $U_3Rh_4Ge_{13}$ reveals metallic behaviors. Besides, it displays low thermal conductivity (i.e. 0.4–0.6 W m−1 K−1) below ~250 K as well as undergoes a structural phase transition in the temperature range of 292–315 K.

Both $\rho(T)$ measurements and DFT calculations indicated $Th_2Rh_3Ge_5$ to be a simple metal.

References

[1] B. Lloret et al., *J. Phys. Colloq.* **49**, C8-487 (1988).

Poster #10

Nuclear forensics – Fission track Analysis – Simulation for training and penetrating fluorescent colors.

Babayew Rami,[1] Yaacov Yehuda-Zada,[1] Lorincik Jan,[2] Orion Itzhak,[3] Katarivas-Levy Galit,[4] Halevy Itzhak[1,3,*]

1 Physics Department, Nuclear Research Centre Negev, Beer-Sheva, Israel

2 Nuclear Fuel Cycle department, Research Centre Rez, Czech Republic

3 Unit of Nuclear Engineering, Faculty of Engineering Sciences, Ben-Gurion Uni. of the Negev, Israel

4 Department of Biomedical Engineering, Faculty of Engineering Sciences, Ben-Gurion Uni. of the Negev, Israel

E-mail: halevy.itzhak.dr@gmail.com

To answer nuclear forensics questions, we are developing new innovative techniques and approaches to make this analysis more reliable and accurate. Currently, only trained researchers can analyze microscope images. Since this analysis is dependent on the researcher's own abilities and skills, it is obvious that different researchers will produce results that are slightly distinct. A new worker's certification period is quite long, and it must cover numerous examples from previously measured data as well as some that we can only predict. A good simulation software can aid with training and provide a tool for grading new researchers.[1] The fission tracks were simulated by Monte-Carlo software, GEANT4, which uses all the physics behind the nuclear fission tracks, such as thermal neutrons flux, fission cross-section, radiation time, particle size, enrichment, etc. In this study, our Trainer2.0 software calculates the tracks on our Lexan detector and its projection, according to the physical parameters like neutron flax, size of the particle, the isotope, and radiation time. The result is a "star" centered on the simulated particle. Our full software is written with MatLab code.

We can simulate an extreme condition and learn new aspect in the fission track technique. From the simulation we can learn about the proper amount of material to use as a sample in the FTA technique. The simulation can predict and compared to the mini-bulk and the micro-bulk analysis. New idea of using penetrating fluorescent colors give as the ability to scan our detector in 3D instead of 2D. In this case we used the Dapi marker as a first shoot, this marker is well known for biomedical research. This new idea to investigate the FT Star more than just by his projection.

Identifying the length of the tracks and their distribution allow us to determine the element source isotope be the shape of "fission products distribution" and the density of the impurities in the source.

References

[1] Halevy I., Admon U., Chinea-Cano E., Weiss A.M., Dzigal N., E. Boblil, Dagan M., Orion I. and Radus R. Investigations, Progress in Nuclear Science and Technology, 2018, v. 5; p. 175-178 3.3.

Poster #11

Theoretical Study on the Bonding Feature of Actinides

Shu-Xian Hu

University of Science and Technology Beijing, 100083 Beijing, China
e-mail: hushuxian@csrc.ac.cn

Actinide metal oxo clusters are of vital importance in actinide chemistry, as well as in environmental and materials sciences. They are ubiquitous in both aqueous and nonaqueous phases and play key roles in nuclear materials (e.g., nuclear fuel) and nuclear waste management. Despite their importance, our structural understanding of the actinide metal oxo clusters, particularly the transuranic ones, is very limited because of experimental challenges such as high radioactivity. Herein we report a systematic theoretical study on the structures and stabilities of seven actinide metal oxo−hydroxo clusters $[\mathrm{An^{IV}_6O_4(OH)_4L_{12}}]$ (1-An; An = Th−Cm; L = $\mathrm{O_2CH}$−) along with their group 4 (Ti, Zr, Hf, Rf) and lanthanide (Ce) counterparts $[\mathrm{M^{IV}_6O_4(OH)_4L_{12}}]$ (1-M). The work shows the Td-symmetric structures of all of the 1-An/M clusters and suggests the positions of the −OH functional groups, which are experimentally challenging to determine. Furthermore, by removing six electrons from 1-An, we found that oxidation could happen on the AnIV metal ions, producing $[\mathrm{An^{V}_6O_4(OH)_4L_{12}}]^{6+}$ (2-An; An = Pa, U, Np), or on the $\mathrm{O^{2-}}$ and $\mathrm{OH^{-}}$ ligands, producing $[\mathrm{An^{IV}_6(O^{\bullet-})_4(OH^{\bullet})_2(OH)_2L_{12}}]^{6+}$ (3-An; An = Pu, Am, Cm). On the basis of 2-An, we constructed a series of tetravalent and pentavalent actinide metal oxo clusters $[\mathrm{An^{IV}_6O_{14}}]^{4-}$ (4-An) and $[\mathrm{An^{V}_6O_{14}}]^{2+}$ (5-An), which proves the feasibility of the highly important pentavalent actinyl clusters, demonstrates the f orbital's structure-directing role in the formation of linear $[\mathrm{O{=}An^{V}{=}O}]^{+}$ actinyl ions, and expands the concept of actinyl−actinyl interaction into pentavalent transuranic actinyl clusters.

Poster #12

The Glenn T. Seaborg Institute at Idaho National Laboratory

J. Rory Kennedy

Glenn T. Seaborg Institute, Idaho National Laboratory, Fremont Avenue, Idaho Falls, Idaho, 83415, USA
e-mail: rory.kennedy@inl.gov

Established in 2017, the Glenn T. Seaborg Institute at Idaho National Laboratory (INL-GTSI) focuses on advancing fundamental research in the actinide sciences by providing unique opportunities to early career scientists and engineers to gain experience studying the actinide elements and their associated systems. The INL-GTSI is built from three focus areas that are based on the expertise and supporting infrastructure at INL and include solid state chemistry and physics, solution phase chemistry and physics, and forensic and isotope science. The INL-GTSI offers our young researchers the unique chance to work in a U. S. national laboratory as an INL Distinguished Postdoctoral Associate, a highly competitive, highly selective program intended to promote the top echelon of recent Ph.D. graduates. The high expectations for our Seaborg Institute postdoctoral associates are rewarded with a top level salary and strong benefits, direct access to advanced instrumentation in radiological facilities, both discretionary and programmatic research funds, and a very active mentoring program.

Topical areas that are of interest to the INL-GTSI include, but not limited to, fundamental actinide properties, structure/property (electronic, magnetic, thermal) relations, actinide quantum criticality, f-electron interactions, electron correlations, computational studies, nuclear fuels, new phases, defect effects, interface interactions, isotope production and separation, forensic analytical chemistry, gamma spectroscopy, surrogate nuclear debris, structure and dynamic properties of actinides in non-aqueous solvents, separations chemistry and kinetics for advanced nuclear fuel cycles, radiation effects on the chemical behavior of actinides in solution, innovative industrial and medical applications of actinides, and innovative and advanced ligand design for complexation of the actinides.

Capabilities supporting the INL-GTSI for both radioactive and non-radioactive samples include electron microscopy (SEM, TEM with EDS, WDS, EBSD), focused ion beam (FIB), electron probe microanalyzer (EPMA), physical properties measurement system (PPMS), X-ray diffraction, atom probe tomography, 3D X-ray microscopy, plasma FIB secondary ion mass spectrometry (PFIB-SIMS), 2 peta-flop scale supercomputer systems, an artificial intelligence system, molecular beam epitaxy (MBE), multiscale mechanical properties testing, thermal properties including high spatial resolution thermal conductivity, laser resonant ultrasound spectroscopy, electrochemistry, etc.

Poster #13

Charge order, frustration relief, and spin-orbit coupling in U_3O_8

Dirk Lamoen,[1] Rolando Saniz,[2] Gianguido Baldinozzi, Ine Arts, Gregory Leinders, and Marc Verwerft

1 EMAT, Physics Department, University of Antwerp, Groenenborgerlaan 171, B-2020 Antwerpen, Belgium
2 CMT, Physics Department, University of Antwerp, Groenenborgerlaan 171, B-2020 Antwerpen, Belgium
3 Université Paris-Saclay, CentraleSupélec, CNRS, SPMS, 91190 Gif-sur-Yvette, France
4 Belgian Nuclear Research Centre (SCK CEN), Institute for Nuclear Materials Science, Boeretang 200, B-2400 Mol, Belgium
e-mail: Dirk.Lamoen@uantwerpen.be

Research efforts on the description of the low temperature magnetic order and electronic properties of U_3O_8 have been inconclusive so far. Reinterpreting neutron scattering results, we use group representation theory to show that the ground state presents collinear out-of-plane magnetic moments, with antiferromagnetic coupling both in-layer and between layers. Charge order relieves the initial geometric frustration, generating a slightly distorted honeycomb sublattice with Néel type order. The precise knowledge of the characteristics of this magnetic ground state is then used to explain the fine features of the band gap. In this system, spin-orbit coupling (SOC) is of critical importance, as it strongly affects the electronic structure, narrowing the gap by ~38%, compared to calculations neglecting SOC. The predicted electronic structure actually explains the salient features of recent optical absorption measurements, further demonstrating the excellent agreement between the calculated ground state properties and experiment.

Poster #14

Epitaxial light actinide oxide thin films

Florence Legg,[1] Ross Springell [1], Helen Steele [2]

1 University of Bristol, Bristol, BS8 1TL, England
2 Sellafield Ltd., Seascale, Cumbria, CA20 1PG, England
e-mail: florence.legg@bristol.ac.uk

Epitaxial thin films of thoria (ThO_2) and $(U,Th)O_2$ mixed oxides (MOX), across the U-Th range, have been synthesised by DC magnetron sputtering. These films can be implemented in the research of thorium-based nuclear fuels, mixed actinide oxide nuclear fuel and the long term storage of nuclear waste forms. The samples were characterized using x-ray diffraction (XRD) and spectroscopic ellipsometry (SE). Three epitaxial ThO_2 samples of different orientations have been synthesised and confirmed by XRD analysis. The samples are [1 1 1], [1 1 0] and [1 0 0] oriented, with lattice parameters determined to be 5.71 ± 0.01 Å, 5.606 ± 0.005 Å and 5.62 ± 0.01 Å respectively. The epitaxial $(U,Th)O_2$ samples are [1 0 0] oriented and four samples have been fabricated, across the U-Th range. All samples synthesised were found to have a unique specular direction. Values for the band gap and optical constants of the epitaxial ThO_2 and MOX samples have also been determined by SE. The mixed oxide sample series has also been exposed to a hydrogen peroxide solution, to investigate the effect of ThO_2 on the dissolution of UO_2.

Poster #15

Interplay of physical and chemical properties in complex non-centrosymmetric compounds

A. Amon, Yu. Prots, E. Svanidze, Yu. Grin, A. Leithe-Jasper

Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzer Strasse 40, Dresden 01187, Germany
e-mail: Andreas.Leithe-Jasper@cpfs.mpg.de

Crystallographically complex compounds often possess peculiar physical properties, the evolution of which can be tracked by changing one of the constituent elements at a time. In this poster, we will share recent discovery and synthesis of isotypic $R/A_4Be_{33}Pt_{16}$ (R = Y, La - Nd, Sm – Lu; A = U or Th) compounds, which crystallize with the noncentrosymmetric cubic space group I43d. The lattice parameters vary from a = 13.6682(4) Å for R = La to a = 13.4366(3) Å for R = Lu analogues. $R_4Be_{33}Pt_{16}$ phases exhibit a wide range of ground states. R = Y, La, and Lu analogues display superconductivity. Their calculated electronic structures show nonzero density of states at the Fermi level, with the value of the Sommerfeld coefficient consistent with those obtained experimentally. The rest of the $R_4Be_{33}Pt_{16}$ compounds exhibit magnetic ground states with ordering temperatures ranging from T_{mag} = 0.4 K (R = Yb) to T_{mag} = 40 K (R = Pr). The A = Th system exhibits superconductivity below T_c = 0.9 K, which is strongly affected by minute changes of the lattice parameter a. The A = U analogue does not show any transitions down to T = 0.35 K. Its large value of the Sommerfeld coefficient γ_n = 200 mJ $mol_U^{-1}K^{-2}$ indicates effective electron mass enhancement. The diversity of physical properties of $R/A_4Be_{33}Pt_{16}$ compounds can likely be attributed to the nature of the individual rare-earth elements, structural non-centrosymmetricity, the large number of atoms per unit cell (212), as well as the complex multi-centre interactions within the Be-Pt framework.

References

[1] A. Amon, E. Svanidze, Yu. Prots, M. Nicklas, U. Burkhardt, A. Ormeci, A. Leithe-Jasper, Yu. Grin, *Dalton Trans.* 2020, 49, 9362

[2] E. Svanidze, A. Amon, M. Nicklas, Yu. Prots, M. Juckel, H. Rosner, U. Burkhard, M. Avdeev, Yu. Grin, A. Leithe-Jasper *Phys. Rev. Mater.* 2021, 5, 074801

[3] P. Kozelj, M. Juckel, A. Amon, Yu. Prots, A. Ormeci, U. Burkhard, M. Brando, A. Leithe-Jasper, Yu. Grin, E. Svanidze, *Sci. Rep.* 2021, 11, 22352

Poster #16

Temperature-dependent Fermi surface probed by Shubnikov-de Haas oscillations in topological semimetal candidates DyBi and HoBi

Orest Pavlosiuk, Paulina Nowakowska, Piotr Wiśniewski, Dariusz Kaczorowski

Institute of Low Temperature and Structure Research, Polish Academy of Sciences,
ul. Okólna 2, 50-422 Wrocław, Poland
e-mail: o.pavlosiuk@intibs.pl

Materials exhibiting extreme magnetoresistance combined with a possible non-trivial topology of the electronic structure have captured extensive research interest. Among such materials are rare-earth monopnictides. Recently, a new magnetic splitting effect has been discovered in several rare-earth monobismuthides [1, 2]. This splitting appears only below the Néel temperature and can lead to a change in the shape of the Fermi pockets.

In this study, we thoroughly investigated the magnetotransport properties of high-quality single crystals of DyBi and HoBi grown from Bi-flux. Both studied compounds exhibit extremely large magnetoresistance (MR~10^4% at T = 2 K and in B = 14 T) and a magnetic field-induced electrical resistivity plateau at low temperatures, both features are often observed in semimetals with nearly perfect charge carrier compensation. In the temperature range 50-300K, the standard Kohler's rule is obeyed for both compounds, while at lower temperatures the extended Kohler's rule [3] must be invoked. The quantum oscillations of electrical resistivity, i.e. Shubnikov-de Haas (SdH) effect, were observed in DyBi and HoBi. They are composed of several different frequencies indicating the coexistence of electron- and hole-type Fermi pockets. Interestingly, a quite rare temperature-dependence of SdH oscillation frequencies was discovered in both materials. This observation, along with the successful application of the extended Kohler's rule, points to a strong temperature dependence of carrier concertation and mobility, which is consistent with a possible effect of novel magnetic band splitting [1,2] on the shape of the Fermi surface.

References

[1] B. Schrunk et al., Nature **603**, 610 (2022).
[2] Y. Kushnirenko et al., *Physical Review B* **106**, 115112 (2022).
[3] J. Xu et al., *Physical Review X* **11**, 041029 (2021).
*This work was supported by the National Science Centre (Poland) under research grant 2021/40/Q/ST5/00066.

Poster #17

High Pressure Study of the Unconventional Superconductor $CeRh_2As_2$

Meike Pfeiffer,[1] Konstantin Semeniuk[2], Seunghyun Khim[2] and Elena Hassinger[1]

1 Technische Universität Dresden, IFMP, 01062 Dresden, Germany
2 Max Planck Institute of Chemical Physics of Solids, Nöthnitzer Straße 40, 01187 Dresden, Germany
e-mail: meike.pfeiffer@tu-dresden.de

The heavy-fermion superconductor $CeRh_2As_2$ hosts two distinct superconducting states which are currently understood as even- and odd-parity states. [1,2] The compound crystallizes in a lattice that is globally centrosymmetric but lacks local inversion symmetry at the Ce sites. This likely results in a strong spin-orbit coupling, leading to a very high ratio of upper critical field (> 15 T) to critical temperature (T_c = 0.26 K). The normal state of the compound hosts an additional phase below 0.4 K, believed to be a quadrupole-density wave (QDW) order. [3] We present a high-pressure electrical resistivity study of the superconducting and normal state properties of $CeRh_2As_2$. The QDW order is highly sensitive to lattice compression and gets fully suppressed at P_c approx. 0.6 GPa. The superconducting transition temperature T_c decreases at a significantly lower rate, and both superconducting phases persist up to 2.7 GPa. Thus, we find that the QDW is not responsible for the phase switching from even- to odd-parity state.

References

[1] S. Khim and J. Landaeta et al., *Science* **373**, 1012-1016 (2021)

Poster #18

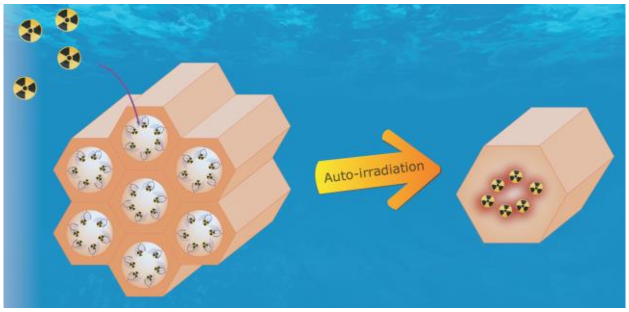
Separation and self-conditioning of plutonium through functionalized mesoporous silicas

Jun Lin,[1] Olaf Walter,[2] Xavier Deschanels,[1] Jérémy Causse,[1] Sandrine Dourdain[1]

1 ICSM, CEA, CNRS, ENSCM, Univ Montpellier, Marcoule, France
2 European Commission, Joint Research Centre, 76344 Karlsruhe, Germany
e-mail: jun.lin@cea.fr

The decontamination and dismantling of nuclear facilities generate significant quantities of effluents contaminated with radionuclides (RN). The present study aims to develop a treatment strategy for RNs based on the use of functionalized mesoporous silicas. Furthermore, this research work aims to demonstrate the dual functionality of this material, which allows for the separation of RN, in the case of this study plutonium, and also promotes its self-conditioning by closing the mesoporosity due to irradiation damage.

Two conventional mesoporous silicas (SBA-15 and MCM-41) have shown a significant decrease in their porosity under external irradiation [1-3]. Efficient capture of actinides present in aqueous solutions by functionalized silicas (PropPhos-SBA-15 and AcPhos-SBA-15) has also been demonstrated through experiments conducted at JRC-Karlsruhe. Current experiments involving ^{239}Pu and ^{238}Pu are ongoing, and preliminary findings derived from synchrotron-based SAXS corroborate that the mesoporous network begins to contract under self-irradiation of ^{238}Pu.



References

[1] Lin, J., et al., *Microporous Mesoporous Mater.* **111454**, 328 (2021).
[2] Lin, J., Radiation-induced Alterations in Mesoporous Silicas: The Effect of Electronic Processes Involving Ions and Electrons. *École doctorale ED-459, Université Montpellier*, (2022).
[3] Lou, Y., Behavior of mesoporous silica under ionic irradiation. *Université Montpellier*, (2016).

Poster #19

Novel ternary phases in the U-Pd-Ge ternary phase diagram

Magdalena Majewicz,[1] Mathieu Pasturel[2], Maria Szlawska[1], Dariusz Kaczorowski[1]

1 Institute of Low Temperature and Structure Research, Polish Academy of Sciences, ul. Okólna 2, 50-422 Wrocław, Poland

2 Univ Rennes, CNRS, Institut des Sciences Chimiques de Rennes - UMR 6226, 35042 Rennes, France

e-mail: m.majewicz@intibs.pl

Intermetallic compounds that contain uranium are known to show unique properties due to interactions between 5*f* and conduction band electrons. They exhibit a variety of interesting phenomena such as coexistence of superconductivity and long-range ferromagnetism in germanides like UGe_2 [1] or URhGe [2], or a complex magnetic phase diagram with the possibility of a Lifshitz point in UPd_2Si_2 [3]. UPd_2Ge_2 [4-6] was suspected to have a rich magnetic phase diagram but has never been studied in a single-crystalline form. Preliminary syntheses using the Czochralski pulling technique and the flux method resulted in the formation of multiphase samples, suggesting that UPd_2Ge_2 does not melt congruently. The difficulties in preparing pure UPd_2Ge_2 single crystals suggested that the study of the U-Pd-Ge phase diagram could lead to the discovery of a wealth of new ternary phases in addition to the already known UPdGe [7] and UPd_2Ge_2.

Several syntheses of polycrystalline samples have been performed with different compositions in the U-Pd-Ge phase diagram. The ingots were then annealed at temperatures between 527 and 827°C for 7 to 28 days. The phases present in the samples were identified using powder X-ray diffraction and EDS microanalysis. New phases such as $U_3Pd_4Ge_4$, $U_{13}Pd_{47}Ge_{25}$ and $U_{34}Pd_{2,5}Ge_{33}$ have been identified. Their crystal structure and preliminary results of magnetization and heat capacity measurements will be presented. Moreover, the coexistence of 1-1-1, 3-4-4 and 1-2-2 phases in a few samples suggests that only the first phase melts congruently, while the others are formed during peritectic transformations. Analysis of the relationship between these phases leads to the conclusion that the preparation of single crystals of pure 3-4-4 and 1-2-2 phases requires significant modifications in the crystal growth procedure.

This work was partially supported by the National Science Centre (Poland), grant no. 2018/31/D/ST3/03295 and by Campus France SSHN scholarship for research stay.

References

[1] S. Saxena et al., *Nature* **406**, 587 (2000).
[2] D. Aoki et al., *Nature* **413**, 613 (2001).
[3] T. Plackowski et al., *Phys. Rev. B* **83**, 174443 (2011).
[4] H. Ptasiewicz-Bąk et al., *J. Phys. F: Metal Phys.* **11**, 1225 (1981).
[5] H. M. Duh et al., *Journal of Magnetism and Magnetic Materials* **145**, 337 (1995).
[6] G. André et al., *Solid State Communications*, Vol. **97**, No. 11, 923 (1996).
[7] S. El-Kahatib et al., *Journal of Applied Physics* **93**, 8352 (2003).

Poster #20

Unravelling the electronic structure of uranium: the case of MUO_3 (M = K, Na and Rb)

S. Orlat[1], R. Bes[2], F. Tuomisto[2], P. Martin[1], P. Moisy[1], G. Leinders[3], K. Kvashnina[4,5]
1 CEA, DES, ISEC, DMRC, University of Montpellier, Marcoule, France
2 Department of Physics, University of Helsinki, P.O. Box 64, FI-00014 Helsinki, Finland
3 Belgian Nuclear Research Centre (SCK CEN), Institute for Nuclear Materials Science, Boeretang 200, B-2400 Mol, Belgium.
4 The Rossendorf Beamline at ESRF, The European Synchrotron, CS40220, 38043 Grenoble Cedex 9, France
5 Institute of Resource Ecology, Helmholtz Zentrum Dresden-Rossendorf (HZDR), PO Box 510119, 01314 Dresden, Germany
e-mail: simon.orlat2@cea.fr

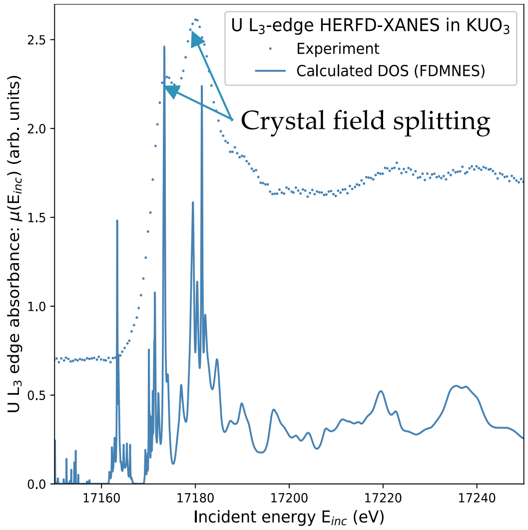


Figure 3: uranium *L_3*-edge HERFD-XANES in KUO_3 and corresponding simulated spectrum using *FDMNES* [3]

Uranium is a key chemical element of the nuclear industry and has a complex chemistry due to the large number of oxidation states allowed by its $[Rn]7s^2 6d^1 5f^3$ ground state electronic configuration. Even if this element has been the focus of substantial research, its electronic structure remains difficult to study theoretically due to the same magnitude of the crystal field splitting, spin orbit coupling and electron-electron interaction. The core of this contribution, is to present recent effort made to overcome this difficulty by studying U(V) compounds such as alkali metal uranates KUO_3, $NaUO_3$ and $RbUO_3$. More precisely, the analysis of high energy resolution fluorescence detected data obtained at the U-L_3 edge measuring the $L\beta_5$ emission line for enhanced spectral resolution [1,2] will be discussed in the light of new theoretical calculation results obtained using the Finite Difference Method for Near-Edge Structure (FDMNES) code [3] as illustrated in [4]. As shown by recent results obtained on KUO_3 [4] and on $NaUO_3$ [5], such combined experimental and theoretical approach allows to better understand the impact on the uranium valence electronic structure induced by the change in the U-O interatomic distances and the associated local structural distortions.

References

[1] K. O. Kvashnina et al., J. Electron Spectrosc. Relat. Phenom. 194, 27 (2014).
[2] K. O. Kvashnina et al., Anal. Chem. 87, 8772 (2015).
[3] O. Bunău et al., J. Phys. Condens. Matter. 21, 4260 (2009).
[4] R. Bes et al., J. Synch. Rad. 29, 21 (2022) 21.
[5] S. M. Butorin et al., Chem. Eur. J. 22, 9693 (2016).

Poster #21

$U_3Os_4Al_{12}$: a new magnetically ordered member of the breathing Kagome lattice $Gd_3Ru_4Al_{12}$ structure-type

Mathieu Pasturel, Vincent Dorcet, Thierry Guizouarn, Stéphanie Lamy, Carmelo Prestipino

Univ Rennes, CNRS, Institut des Sciences Chimiques de Rennes – UMR6226, 35042 Rennes, France
e-mail: mathieu.pasturel@univ-rennes1.fr

Magnetic competitions in the distorted or breathing Kagome lattice do not only induce magnetic frustrations, but they are predicted to lead to other outstanding physical properties such as topological insulation and skyrmion phases [1]. Such a network is formed by *4f* or *5f* elements in the $Gd_3Ru_4Al_{12}$ structure-type. Accordingly, a spin-glass behaviour has been reported for the uranium bearing $U_3Fe_4Al_{12}$ [2] and $U_3Co_4Al_{12}$ [3]. Opposite, an antiferromagnetic order below $T_N = 8.4$ K has been found for $U_3Ru_4Al_{12}$ [4]. Single crystal investigations using neutron diffraction [5] or high magnetic field experiments highlight the formation of an unusual magnetic structure with magnetic moments lying inside the Kagome-lattice, that undergoes several reorientations inside this plane with magnetic field, giving birth to a rich magnetic phase diagram [6].

Following our investigation of this series, the osmium-bearing $U_3Os_4Al_{12}$ phase has been isolated. It crystallizes as expected in the hexagonal $Gd_3Ru_4Al_{12}$-type with U-atoms located at the apexes of the triangles forming the breathing Kagome lattice. Magnetic and specific heat measurements on a polycrystalline sample indicate the occurrence of a magnetic ordering below 5.5 K, most probably of antiferromagnetic nature. The M(B) curve measured at 2 K up to 12 T shows several changes in its slope attributed to reorientations of the magnetic moments in the magnetically ordered state, evidencing a complex magnetic phase diagram for this aluminide.

References

[1] M. Ogunbunmi et al., *Crit. Rev. Solid State Mater. Sci.* (2022) and references therein.
[2] A. P. Gonçalves et al., *Intermetallics*, **17**, 25-31 (2009)
[3] O. Tougait et al., *J. Solid State Chem*., **177**, 2053-2057 (2004)
[4] M. Pasturel et al., *J. Phys. Cond. Matter*, **21**, 125401 (2009)
[5] R. Troć et al., *Phys. Rev. B*, **85**, 064412 (2012)
[6] D. Gorbunov et al., Phys. Rev. B, **99**, 054413 (2019)

Poster #22

DFT-Prediction of the Crystal Structure and Magnetism of UIr

Dr. Malte Sachs, Prof. Dr. Florian Kraus

Philipps-Universität Marburg, Hans-Meerwein-Str. 4, 35032 Marburg
e-mail: malte.sachs@chemie.uni-marburg.de

The correlated metal UIr is discussed as a rare example of a non-centrosymmetric, ferromagnetic superconductor[1] crystalizing in the acentric PdBi structure type ($P2_1$, $mP16$)[2].

We tested the quality of density functional theory (DFT) for predicting the crystal structure and magnetic properties of UIr. Scalar-relativistic calculations with the evolutionary algorithm USPEX[3] find not the observed but the NaTl structure type ($Fd\bar{3}m$, $cF16$) as the energy minimum demonstrating the failure of DFT to reproduce the global energy landscape of UIr.

Helpful DFT predictions are still possible around the local minimum of the experimentally observed structure. Structural optimizations of UIr motivated us to redetermine and correct its crystal structure from the acentric space group $P2_1$ to the centrosymmetric space group $P2_1/c$.[4] We explain the discrepancy with the previous model by the occurrence of twinning. Accordingly, UIr *is not* an acentric superconductor, as has been widely discussed. We further predict the magnetic easy-axis of UIr to be along $[10\bar{1}]$ in agreement with experimental observations.[5,6] However, our calculations hint towards additional antiferromagnetic components along [010] not observed in the experiments yet.

We hope that our results will have an impact on all those theoretical models that investigated potentially novel superconducting coupling mechanisms of UIr based on the non-centrosymmetric and purely ferromagnetic model.

References

[1] Akazawa *et al.*, *J. Phys. Soc. Jpn.* **2004,** *130*, 3149-3134.
[2] Siegrist *et al.*, *J. Less-Common Met.* **1986**, *125*, 167-174.
[3] Oganov et al., *Journal of Chemical Physics* **2006**, *124*, 244704.
[4] Sachs *et al., Inorg. Chem.* **2021**, *60,* 16686-16699.
[5] Sakarya *et al., J. Phys. Soc. Jpn.* **2010,** *79*, 014702.
[6] Galatanu *et al., J. Phys. Soc. Jpn.* **2004,** *73*, 766-767.

Poster #23

Magnetic structure of UNi_4B

Philip Schröder[1], J. Willwater[1], S. Süllow[1], M. Reehuis[2], R. Feyerherm[2], H. Amitsuka[3], B. Ouladdiaf[4], E. Suard[4], R. Sibille[5], M. Klicpera[6], M. Vališka[6], J. Pospíšil[6] and V. Sechovský[6]

[1] IPKM,TU Braunschweig, Germany
[2] HZB, Berlin, Germany
[3] Hokkaido University, Sapporo, Japan
[4] ILL, Grenoble, France
[5] PSI, Villigen, Switzerland
[6] Charles University, Prague, Czech Republic
e-*mail: philip.schroeder@tu-braunschweig.de*

UNi_4B is a frustrated metallic *f*-electron magnet that has been proposed to show a highly unusual form of partial antiferromagnetic ordering at $T_N = 19.5$ K. A long-range magnetic order consisting of a vortex-like structure in the pseudo-hexagonal plane [1,2] is formed by only two thirds of U-ions carrying magnetic moments, whereas the remaining one third is assumed to stay in the paramagnetic state. More recently, the magnetic structure of UNi_4B was discussed in the context of toroidal order, which was argued to lead to novel types of magnetoelectric effects like the current dependent magnetization observed in UNi_4B [3].

Here, we summarize the results of a comprehensive study of the magnetic structure of UNi4B [2]. We have determined the unique type of magnetic order, i.e., a vortex-type spin structure with a magnetic moment of $\mu_{ord} = 0.99(1)\ \mu_B$/(U Atom). The refinement of the magnetic structure suggests that the magnetic moments are slightly canted within the close-to-hexagonal plane. Recent measurements of the elastic constants in magnetic fields verified a complex magnetic phase diagram [4]. In order to establish the evolution of the magnetic structure in the high field phases of UNi_4B, we performed neutron diffraction experiments in high magnetic fields. We will present our first results, which show a significant field dependent change of intensity and hysteresis for fields B||b and ||c axis and discuss this observation in terms of field induced modifications of the partial ordering in UNi_4B.

References

[1] S. Mentink et al., Phys. Rev. Lett. 73, 1031 (1994).
[2] J. Willwater et al., Phys. Rev. B 103, 184426 (2021).
[3] H. Saito et al., J. Phys. Soc. Jpn. 87, 033702 (2018).
[4] T. Yanagisawa et al., Phys. Rev. Lett. 126, 157201 (2021).

Poster #24

Radiation Damage in Electroless Deposited Silver Thin Films

Michael Shandalov,[1] Shachar Moskovich [2], Yuval Golan [2], Eyal Yahel [3]

1 Materials Department, NRCN, Beer Sheva POB 9001, Beer Sheva, Israel
2 Department of Materials Engineering, BGU, Be'er Sheva 8410501, Beer Sheva, Israel
3 Physics Department, NRCN, Beer Sheva POB 9001, Beer Sheva, Israel
e-mail: michash1234@gmail.com

Radiation damage is studied mostly through irradiation with external radiation sources experiments and research of self-irradiated bulk solids [1]. Self-irradiation damage in alpha emitting atoms occurring due to decay into an alpha particle and a recoiling daughter has been studied mostly in bulk actinides, which in most cases involves work in gloveboxes, high levels of radiation and large amounts of radioactive waste. To reduce radioactive waste and allow for work in an unrestricted chemical lab, the radiating element was introduced in minute concentration in a thin film, below the allowed radiation threshold. Previous work on self-irradiating materials focused on thorium alloying of semiconductors such as lead sulfide [2-4]. The goal of the present work was to extend this methodology to metals, and silver, an abundant and widely used metal, which was chosen as a model system for self-irradiation studies. Subsequently, ^{232}Th was incorporated within metallic silver thin films using electroless deposition from acidic bath [4]. By varying deposition parameters, it was possible to control film morphology while introducing controlled amounts of ^{232}Th within the films. After optimization of the ^{232}Th system, ^{232}Th was substituted with ^{228}Th to induce self-radiation damage within the films. Preliminary results on characterization of Ag thin films doped with ^{228}Th presented in this work.

References

[1] V. Dubinko, "Radiation damage and recovery of crystals: Frenkel vs. Schottky defect production", *Nova Science Publishers Inc*., New York, 2011.
[2] M. Biton *et al.*, *Thin Solid Films*, **556**, 223 (2014).
[3] T. Templeman *et al.*, *Sci. Rep.*, **7**, 1 (2017).
[4] M. Perez *et al.*, *RSC Materials Chemistry Frontiers*, **5**, 2860 (2021).

Poster #25

Modelling Oxygen and Rare-earth impurities in Nickelates

Alpesh Sheth[1], Claudine Lacroix [1], Sébastien Burdin[2]

1 Univ. Bordeaux, CNRS, LOMA, UMR 5798, F-33400 Talence, France
2 Institut Néel, CNRS and Université Grenoble-Alpes, Boite Postale 166, 38042 Grenoble,France
e-mail: alpesh-rasikbhai.sheth@u-bordeaux.fr

Recent discovery of Superconductivity in hole doped infinite layered nickelate $R_{1-x}R'_xNiO_2$ opens up the need to revisit the electronic structure and physical properties of rare-earth nickelates.(e.g. $Nd_{1-x}Sr_xNiO_2$). Experimentally this consists of two critical steps 1) Reducing perovskite rare-earth Nickelates ($R\text{-}NiO_3$) to obtain layered rare-earth nickelates ($R\text{-}NiO_2$). 2) Substitutional doping of layered rare-earth nickelates in relevant stoichiometric ratio. This leads to emergence of new effects from disorder and electronic correlations from the rare-earth.

These systems also exhibit a plethora of conventional and unconventional phenomena. Some of them are absence of superconductivity in the bulk nickelate and limited thickness of the infinite-layer phase in thin film, absence of long range anti-ferromagnetism and signature of weakly insulating behavior, absence of Meissner effect and suspect of the origin of superconductivity from the interface, absence of superconductivity in the *La*-nickelate and the role of **4f** orbital in the recipe of superconductivity. Numerous theoretical hypotheses involving the Kondo effect, anti-Kondo effect, multi-orbital effect, Hund's coupling effect, and disorder effect are being put forth in light of the observed facts, but the question of *"What could the crucial ingredients manipulating the superconductivity in film and bulk nickelates be?"* has not yet been answered.

Emphasizing the origin of these emergence phenomena, Friedel oscillations (FO) are simulated in the dilute impurity regime with two kinds of impurities with an extra apical Oxygen(O) impurity and/or Strontium (Sr) atom substituting Neodymium (Nd). The aim is to model disorder and the impact of electronic correlations from rare-earth materials using the Tight Binding (TB) model. The model parameters are first chosen phenomenologically and further intending to use the empirical parameter to ensure the bench marking of the previous available results.

Poster #26

Influence of the crystal structure and nature of the ligands on the valence of uranium in binary chalcogenides: a HERFD-XANES and RIXS study.

T. Stephant[1,2], M. O.J.Y. Hunault[2], P. L. Solari[2], M. Pasturel[1], C. Prestipino[1]

1 Univ. Rennes, CNRS, ISCR-UMR6226, F-35000, Rennes, France
2 SOLEIL Synchrotron, L'Orme des merisiers, Départementale 128, 91190 Saint-Aubin, France
e-mail: thomas.stephant@univ-rennes.fr

Thanks to its *5f* orbitals, uranium benefits from several valence states, from U^{3+} to U^{6+}, in inorganic compounds and possesses a wide crystal-chemistry. The radial expansion of these orbitals leads to energetically close crystal field (dominating in the case of *3d* elements) and spin-orbit (dominating in the case of *4f* rare earths) interactions, and subsequent rich and exotic physical properties [1] (*e.g.* coexistence of superconductivity and ferromagnetism). Associated with a chalcogen element (Q = S, Se, Te), uranium forms inorganic compounds characterized by various crystallographic structures leading to unique uranium polyhedral structures [2] with *e.g.* the presence of $(S_2)^{2-}$ dimers.

However, due to the limited number of known uranium chalcogenides, practically no experimental information on the nature of the U-Q bond is available in the literature. As a consequence, understanding the localization of 5*f* electrons of actinides in solid-state, a great challenge for theoretical physicists, remain limited to the study of oxides [3] and intermetallic materials.

To increase the field of investigation, some binary uranium chalcogenides (S, Se, Te) have been characterized and studied by HERFD-XANES and RIXS spectroscopies at the U M_4 edge. These preliminary measurements enabled to determine the oxidation states of uranium for these binary compounds. The results will be presented and discussed towards the influence of crystal structure and ligands on this oxidation state. For example, HERFD-XANES suggests different U valence states for isostructural compounds UQ_3 ($P2_1/m$).

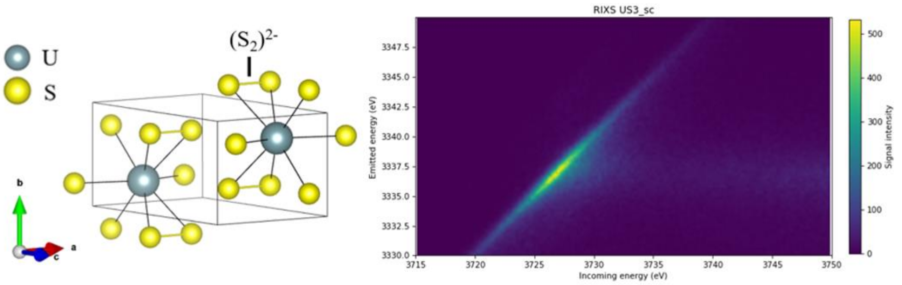


Figure 1: Crystal structure and U 3d4f RIXS map of US_3 ($P2_1/m$).

In addition, some ternary compounds from U-M-S systems (M = 3*d* metal) have been synthetized for further investigation of their physical properties.

References

[1] Aoki, D. et al., *J. Phys. Soc. Jpn.* **88**, 022001 (2019).
[2] Mesbah, A. et al., *Dalton Trans.* **45**, 16067–16080 (2016).
[3] Kvashnina, K. O. et al., *Chem. Commun.* **58**, 327–342 (2022).

Poster #27

Uranium hydride: Ferromagnetism and changes under pressure

William Thomas, Ross Springell, Sven Friedemann
University of Bristol, Bristol, BS8 1TL, UK, email: william128.thomas@bristol.ac.uk

Uranium and hydrogen readily react to form uranium hydride (UH_3). Despite being discovered as the first 5f ferromagnet in 1952, there are still some UH_3 properties that are not well understood such as the easy axis of magnetization [1]. There is a remarkable paucity of information in literature around how ferromagnetism arises in the material and how the magnetic properties change under pressure. This poster will explore the two UH_3 phases at the atomic level and discuss similarities in crystal structure that may explain the ferromagnetism present. The work will also present previous studies into the effects of pressure on the magnetic properties of β-phase UH_3, with an insight into future magnetism investigations that will be conducted at the University of Bristol.

As well as exhibiting strong ferromagnetism in the common α and β UH_3-phases, uranium hydride has found growing attention in the solid state physics community surrounding the proposed superconductivity of uranium "super-hydrides" [2]. These "super-hydrides" are synthesized by applying large pressure to a uranium hydride and hydrogen environment, to achieve these pressures at the University of Bristol a diamond anvil cell (DAC) is used. The specific "super-hydrides" predicted for superconductivity will be presented in this poster alongside evidence of the successful synthesis of the UH_7 and UH_8 phases [3].

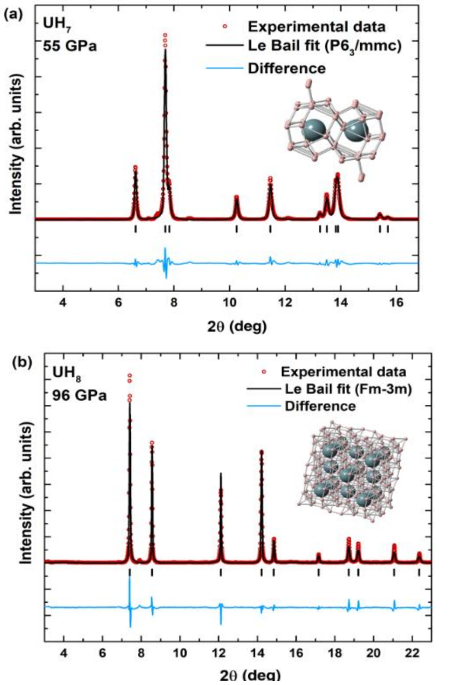


Fig 1: XRD pattern of successfully synthesized UH_7 and UH_8 super-hydrides (Accompanied with simulated crystal unit cells) [3]

The predicted and measured synthesis conditions are compared and discussed with comment on their similarities. Unfortunately, despite being successfully synthesized and characterized, the UH_7 and UH_8 samples were never tested for superconductivity. The future work in this area will follow on from crystal structure determination of the UH7 and UH8 phases by conducting transport and magnetic measurements to investigate the superconductivity claims.

References

[1] L. Havela et al., "UH3-based ferromagnets: New look at an old material," Journal of Magnetism and Magnetic Materials, vol. 400, pp. 130–136, Feb. 2016.

[2] A. Kruglov et al., "Uranium polyhydrides at moderate pressures: Prediction, synthesis, and expected superconductivity," Science Advances, vol. 4, no. 10, Oct. 2018.

[3] B. Guigue, A. Marizy, and P. Loubeyre, "Synthesis of UH7 and UH8 superhydrides: Additive-volume alloys of uranium and atomic metal hydrogen down to 35 GPa," Physical Review B, vol. 102, no. 1, Jul. 2020

Poster #28

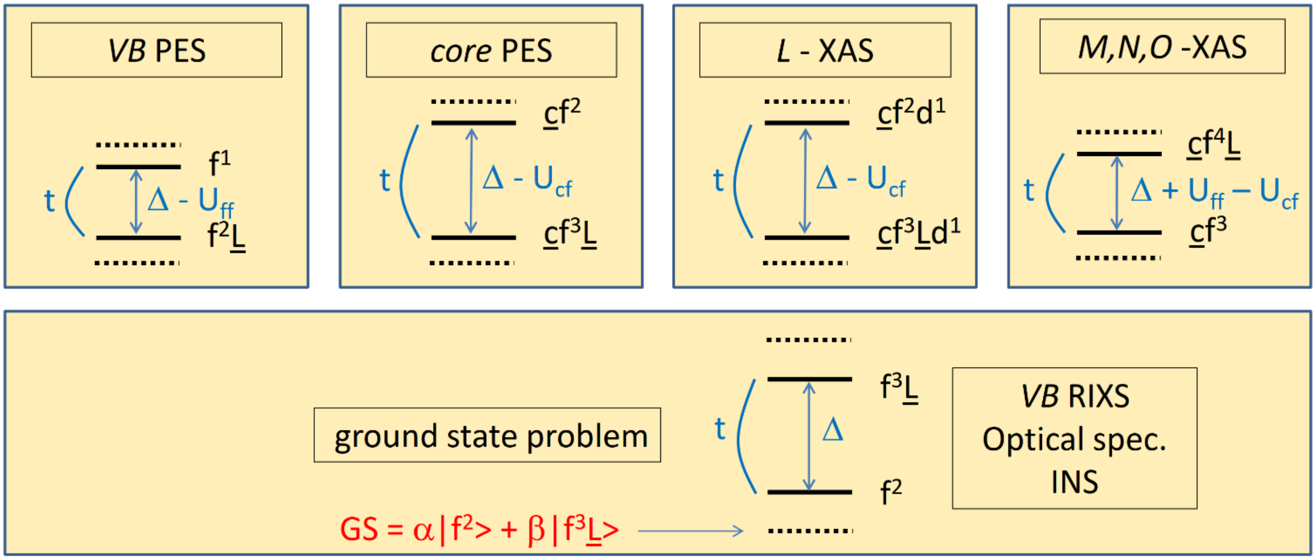
Why different spectroscopies give seemingly different answers on U compounds

Hao Tjeng

Max Planck Institute of Chemical Physics, Nöthnitzerstr. 40, 01187 Dresden, Germany
e-mail: hao.tjeng@cpfs.mpg.de

A central issue in the research field of uranium-based materials is the charge state of the U 5f since it provides a good (and probably even a necessary) starting point to model the chemical and physical properties of these materials. Numerous spectroscopic studies have been carried out, utilizing a wide range of methods such as photoelectron spectroscopy (PES, ARPES, RPES), x-ray absorption spectroscopy (XAS, XMCD, HERFD), x-ray Raman spectroscopy, and RIXS. Yet, there is a considerable debate and confusion in the community since different methods give seemingly different answers about the U 5f valency.

Here we argue that the different methods should give the same answer. The key ingredient is that the analysis of the spectra must include explicitly electron correlation effects at the U sites. An interpretation in terms of one-electron density of states and "convolutions" thereof with various core level or added states is not sufficient and leads to erroneous results. Instead, one must at least use a configuration-interaction based approach in order to arrive at a consistent and unified picture, and include the "quantum interference" effects that occur when analyzing the intensities of the various peaks in the spectra.



Poster #29

Cerium-Scandium: an oxidation-resistant Ce alloy

D.W. Wheeler

AWE, Aldermaston, Reading, Berkshire, RG7 4PR, United Kingdom

Cerium (Ce) is often considered to be a non-radioactive surrogate for delta-phase plutonium (δ-Pu) alloys [1] on account of the many similarities, including relatively low melting temperature; similar crystal structure (face centred cubic) at ambient temperature and pressure; and a propensity to undergo phase transitions when subjected to pressure or cooling to sub-ambient temperatures. However, the rapid oxidation of Ce, especially in the presence of water, can present difficulties in handling Ce and some of its alloys such as cerium-lanthanum (Ce-La) [2,3]. One way by which the oxidation resistance may be improved is by alloying Ce with Scandium (Sc) which has been suggested as an alloying element that could reduce the oxidation of Ce in air [4]. However, very few studies of this alloy system have been found in the published literature. Given the dearth of information available, a Ce-Sc alloy has been prepared and its structure and properties investigated.

A Ce-13 wt.% Sc alloy was arc melted and subsequently given an homogenising heat treatment, initially at 575 °C for 24 hours, and followed by a second heat treatment of 650 °C for another 24 hours. The oxidation behaviour of the alloy was studied by exposing a free-standing specimen to the open laboratory atmosphere (RH ~30%) at ambient temperature (~21 °C) for up to 40 days, during which it was analysed by X-ray diffraction (XRD) at regular intervals. This initial work has indicated that alloying Ce with Sc has increased its oxidation resistance. Time-resolved XRD patterns have shown that although the CeO_2 reflections do increase in intensity over this timescale, the rate at which they increase is markedly reduced compared with unalloyed Ce or Ce-5%La. The data suggest that the presence of Sc does improve the oxidation resistance of Ce, which may benefit experimental studies of the latter, particularly when specimen oxidation may complicate data analysis.

References

1. C.J. Boehlert, J.D. Farr, R.K. Schulze, R.A. Pereyra, J.A. Archuleta, *Phil. Mag.*, **83** (2003), 1735-1744.
2. D.W. Wheeler, *Corros. Sci.*, **111** (2016), 52-60.
3. D.W. Wheeler, *Solid State Ionics*, **313** (2017), 22-31.
4. K.A. Gschneidner, B.J. Beaudry, ASM Metals Handbook (9th ed.), Vol. 2: Non-Ferrous Alloys and Pure Metals (1982), 722-723.

Poster #30

Chemistry of clad breach in a Lead-cooled Fast Reactor

Andries van Hattem,[1] Karin Popa, [2] Jean-Christophe Griveau,[2] Eric Colineau, [2] Anna L. Smith, [1] Rudy J.M. Konings[1,2]

1 Technische Universiteit Delft, Mekelweg 16, 2629 JB, Delft, The Netherlands
2 European Commission, Joint Research Centre (JRC), Karlsruhe, Germany
e-mail: a.vanhattem@tudelft.nl

Lead-cooled Fast Reactors are one of the Generation IV nuclear reactor designs, that are to be cooled with either liquid Pb or a liquid eutectic mixture of Pb and Bi. In Europe, mixed oxide fuel $(U,Pu)O_2$ is currently the reference for such type of reactors. During irradiation, several classes of fission products form, making the fuel a multi-component system: metallic precipitates, volatile fission products, oxide products and fission products that dissolve in the fuel matrix [1].

In case of clad breach, the fuel and fission products can come into contact with the coolant, giving rise to a potential very complex chemistry. The interaction chemistry can be distinguished in among others coolant-volatile fission products phase interaction (Pb,Bi)-(Cs,Mo,Te,I,O), coolant-fuel interaction (Pb,Bi)-(U,Pu,O) and the interaction between coolant and radionuclides with a potential long term radiological impact (Pb,Bi)-(Ba,Sr,U,Pu,O).

As part of our systematic studies into several of these (sub)-systems, the compounds Ba_2PbUO_6 [2] and Ba_2PbPuO_6 [3] are being synthesised and characterised. We will here report the results of the compound synthesis, structure analysis by XRD and low temperature heat capacity measurements. The measurements are both of interest for the thermochemistry of the fuel-coolant interaction (yielding the standard entropy values at 298 K) and the fundamental physical understanding of the behavior of the f-shell electrons: Ba_2PbUO_6 contains U^{6+} with electronic structure $[Rn]5f^0$, while Ba_2PbPuO_6 has Pu^{6+} with an electronic structure of $[Rn]5f^2$.

Acknowledgements

The experimental data used in this research were generated through access to the ActUsLab under the Framework of access to the Joint Research Centre Physical Research Infrastructures of the European Commission (PARTICULAR, FMR/PAMEC Access Agreement N°36345/02).

References

[1] Pelletier, Michel, and Yannick Guérin. "Fuel Performance of Fast Spectrum Oxide Fuel." (2020): 72-105. In: Konings, Rudy, and Roger Stoller. *Comprehensive nuclear materials*. Elsevier, 2020.

[2] Awasthi, S. K. et al. *Journal of Inorganic and Nuclear Chemistry* 29.5 (1967): **1225-1228**.

[3] Awasthi, S. K. et al. *Journal of Inorganic and Nuclear Chemistry* 30.3 (1968): **819-821**

List of participants

	First name	Surname	Affiiliation
1.	Laura	Agnarelli	MPI CPfS
2.	Beatriz	Aguiar M. V. dos Santos	Centro de Ciências Tecnológicas e Nucleares
3.	Bernard	Amadon	CEA, Université Paris Saclay
4.	Lucia	Amidani	Rossendorf Beamline at the European Synchrotron (France)
5.	Alexander	Andreev	Czech Academy of Sciences
6.	Nabeel	Anwar	University of Southampton
7.	Ine	Arts	University of Antwerp
8.	Arushi	Arushi	MPI CPfS
9.	Rami	Batrice	Los Alamos National Laboratory
10.	Dmitry	Bocharov	University of Latvia
11.	Manuel	Brando	MPI CPfS
12.	Sébastian	Burdin	Bordeaux University
13.	Roberto G. M.	Caciuffo	Istituto Nazionale di Fisica Nucleare
14.	Jacopo	Carbone	Ecole Polytechnique Fédérale de Lausanne (EPFL),
15.	Irene	Cardaio	Helmholtz-Zentrum Dresden-Rossendorf (HZDR)
16.	Jorge	Cardenas	MPI CPfS
17.	Denise	Christovam	MPI CPfS
18.	Eric	Colineau	European Commission, Joint Research Centre (JRC)
19.	Ross	Colman	Charles University, Prague
20.	Timothy	Coons	Los Alamos National Laboratory
21.	Théo	Croze	ISAS, S2CM, CEA – Université Paris-Saclay
22.	Shanmukh V V U K	Devanaboina	Charles University, Prague
23.	Xiaolong	Feng	MPI CPfS
24.	Mauro	Giovannini	University of Genova
25.	Krzyzstof	Gofryk	Idaho National Laboratory
26.	Yuri	Grin	MPI CPfS
27.	Jean-Christophe	Griveau	European Commission, Joint Research Centre

28.	Itzhak	Halevy	Ben-Gurion Uni. of the Negev
29.	Lottie	Harding	University of Bristol
30.	Andries	Hattem, van	Technische Universiteit Delft
31.	Ladislav	Havela	Charles University
32.	Shu-Xian	Hu	University of Science and Technology Beijing
33.	Dariusz	Kaczorowski	Institute of Low Temperature and Structure Research, Polish Academy of Sciences
34.	Rory	Kennedy	Glenn T. Seaborg Institute, Idaho National Laboratory
35.	Seunghyun	Khim	MPI CPfS
36.	Sergii	Khmelevskyi	Technical University of Vienna
37.	Jindrich	Kolorenc	Czech Academy of Sciences
38.	Mitja	Krnel	MPI CPfS
39.	Dirk	Lamoen	University of Antwerp
40.	Javier	Landaeta	MPI CPfS
41.	Gerard	Lander	Institut Laue Langevin
42.	Eleanor	Lawrence Bright	European Synchrotron Radiation Facility (ESRF)
43.	Florence	Legg	University of Bristol
44.	Dominik	Legut	VSB - Technical University of Ostrava
45.	Andreas	Leithe-Jasper	MPI CPfS
46.	Jun	Lin	CNRS - ICSM, CEA, CNRS, ENSCM, Univ Montpellier,
47.	Magdalena	Majewicz	Institute of Low Temperature and Structure Research, Polish Academy of Sciences
48.	Andrea	Marino	MPI CPfS
49.	Juliane	März	Helmholtz-Zentrum Dresden-Rossendorf
50.	Vincent Paul	Munar	Institute Mines Telecom Atlantique / Universidad Politécnica de Madrid
51.	Lars	Nordström	Uppsala University
52.	Pavlosiuk	Orest	Institute of Low Temperature and Structure Research, PAS
53.	Simon	Orlat	CEA Marcoule
54.	Luigi	Paolasini	ESRF
55.	Mathieu	Pasturel	Institut des Sciences Chimiques de Rennes
56.	António	Pereira Gonçalves	C2TN, DECN, Instituto Superior Técnico, Universidade de Lisboa

57.	Meike	Pfeiffer	Technical University Dresden
58.	Shannon Kimberley	Potts	Forschungszentrum Jülich GmbH
59.	Sushma	Sankar	MPI CPfS
60.	Martin	Sundermann	MPI CPfS & DESY
61.	Katharina	Reinhold	Ecole Polytechnique Fédérale de Lausanne (EPFL),
62.	Yannick	Roselló	Universitat Rovira i Virgili
63.	Malte	Sachs	Philipps-Universität Marburg
64.	Moritz	Schmidt	Helmholtz-Zentrum Dresden - Rossendorf
65.	Philip	Schröder	Universität Braunschweig
66.	Andrea	Severing	University of Cologne
67.	Michael	Shandalov	NRCN
68.	Alpesh Rasikbhai	Sheth	Université de Bordeaux CNRS, LOMA
69.	Theo	Siegrist	Florida State University, FAMU-FSU College of Engineering
70.	Clara	Silva	Rossendorf Beamline at the European Synchrotron
71.	Thomas	Stephant	Université de Rennes
72.	Eteri	Svanidze	MPI CPfS
73.	Maria	Szlawska	Institute of Low Temperature and Structure Research, Polish Academy of Sciences
74.	William	Thomas	Interface Analysis Centre, University of Bristol
75.	Hao	Tjeng	MPI CPfS
76.	James	Tobin	University of Wisconsin Oshkosh
77.	Olivier	Tougait	Université de Lille
78.	Pablo	Vacas-Arquero	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT)
79.	Sorin-Octavian	Vălu	European Commission, JRC Karlsruhe,
80.	Levytskyi	Volodymyr	TU Bergakademie Freiberg
81.	William	Watson	Imperial College London
82.	David	Wheeler	AWE (Atomic Weapons Establishment)
83.	Steffen	Wirth	MPI CPfS
84.	Nazar	Zaremba	MPI CPfS
85.	Gertrud	Zwicknagl	TU Braunschweig