

Actinides and Rare Earths

Room 123 - Session AC+MI-ThM

Superconductivity, Electron Correlation, Magnetism and Complex Behavior

Moderators: Tomasz Durakiewicz, Idaho National Laboratory, Ladislav Havela, Charles University, Czech Republic, David Shuh, Lawrence Berkeley National Laboratory, James G. Tobin, University of Wisconsin-Oshkosh

8:00am AC+MI-ThM-1 Electronic Structures of f-Electron Superconductors, Shin-ichi Fujimori, Japan Atomic Energy Agency, Japan **INVITED**

The unconventional superconductors in *f*-based materials attract much attention in recent years. In the present talk the electronic structures of *f*-electron superconductors UTe₂ and CeIr₃ will be presented.

Recently, it was discovered that UTe₂ has relatively high transition temperature ($T_c=2.1$ K), and it is classified into one of such class of materials [1,2]. The electronic structure of UTe₂ was studied by resonant photoelectron spectroscopy (RPES) and angle-resolved photoelectron spectroscopy (ARPES) with soft X-ray synchrotron radiation [3]. The partial U 5*f* density of states of UTe₂ were imaged by the U 4*d*-5*f* RPES and it was found that the U 5*f* state has an itinerant character, but there exists an incoherent peak due to the strong electron correlation effects. The band structure of UTe₂ was obtained by ARPES and its overall band structure was mostly explained by band structure calculations, except for the incoherent band at $E_F \sim 0.5$ eV. These results suggest that the U 5*f* states of UTe₂ have itinerant but strongly-correlated nature.

The electronic structure of the *f*-based superconductor CeIr₃ ($T_{sc}=3.1$ K) was also studied by photoelectron spectroscopy [4]. The energy distribution of the Ce 4*f* states were revealed by the Ce 3*d*-4*f* RPES. The contribution of the Ce 4*f* states to the density of states (DOS) at the Fermi energy was estimated to be nearly half of that of the Ir 5*d* states, implying that the Ce 4*f* states have a considerable contribution to the DOS at the Fermi energy. These results suggest that CeIr₃ is an *s*-wave-type superconductor with a considerable contribution from the Ce 4*f* states.

Work performed in collaboration with Ikuto Kawasaki, Yukiharu Takeda, Hiroshi Yamagami, Norimasa Sasabe, Yoshiki J Sato, Ai Nakamura, Yusei Shimizu, Arvind Maurya, Yoshiya Homma, Dexin Li, Fuminori Honda, and Dai Aoki.

References

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- [4] S. Fujimori *et al.*, Electron. Struct. **5**, 045009 (2023).

8:30am AC+MI-ThM-3 RIXS Study of the 5f Configuration in UTe₂: Low-Energy Excitations, Te₂ Chains and U Dimers, D. Christovam, M. Sundermann, A. Marino, Max Planck Institute for Chemical Physics of Solids, Germany; H. Gretarsson, PETRA III, Deutsches Elektronen-Synchrotron DESY, Germany; B. Keimer, Max Planck Institute for Solid State Research, Germany; A. Gloskovskii, PETRA III, Deutsches Elektronen-Synchrotron DESY, Germany; M. Haverkort, Institute for Theoretical Physics, Heidelberg University, Germany; I. Elfimov, University of British Columbia, Canada; E. Bauer, P. Rosa, Los Alamos National Laboratory; A. Severing, Institute of Physics II, University of Cologne, Germany; Liu Hao Tjeng, Max Planck Institute for Chemical Physics of Solids, Germany **INVITED**

UTe₂ is a recently discovered odd-parity superconductor, emerging as a promising candidate for topological superconductivity [1-3]. A key challenge in understanding its anisotropic properties and their dependence on tuning parameters is finding the appropriate *Ansatz* for describing such a complex electronic system, situated at the border between localization and itinerancy. It is essential to determine the extent to which local physics persists and to identify the governing electronic *fⁿ* configuration. Although the latter has been addressed using various methods, a fierce debate remains about whether the main configuration is *f²* or *f³*.

Discrepancies in determining the dominant *fⁿ* configuration are common in uranium-based intermetallics. We argue that this apparent contradiction arises from the challenge of accurately analyzing the spectra, given the complexity associated with the underlying many-body problem and uncertainties in the input model parameters.

In our study, we opt for a newly developed spectroscopic method for U materials, namely resonant inelastic x-ray scattering (RIXS) in the tender x-ray range [4]. Applying this method to UTe₂, we can unambiguously detect the presence of atomic-like U 5*f*-5*f* low-energy excitations. This establishes the correlated nature of UTe₂ despite the presence of strong covalence and band formation. Moreover, we can also fingerprint the U 5*f²* as the main configuration without the need to rely on accurate calculations, thus settling the debate about the 5*f* valence state [5].

The short Te₂-Te₂ distances, however, seem incompatible with a formal U⁴⁺ valence. We resolve this puzzle by utilizing band structure calculations and photon energy-dependent photoemission. We found that the charge transfer from the 5*p_y*_{||b} band of the Te₂ chain is directed not to the U 5*f* but to the bonding state of the U 6*d_{3z²-r²}* orbitals of the U dimer. Notably, these two band states are non-bonding to each other or to other non-5*f* states in the material. Yet, both bands hybridize with the U 5*f*, emphasizing that the description of the physical properties of UTe₂ must include the Te₂ 5*p_y*_{||b} and U 6*d_{3z²-r²}* bands plus the 5*f²* *Ansatz* [5] These are all very peculiar findings, making UTe₂ a very special material indeed.

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- [2] D. Aoki *et al.*, J. Phys. Soc. Jpn. **88**(4), 043702 (2019)
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- [4] A. Marino *et al.*, Phys. Rev. B **108**, 045142 (2023)
- [5] D. S. Christovam *et al.*, arXiv 2402.03852 [https://arxiv.org/abs/2402.03852] (2024)

9:00am AC+MI-ThM-5 Incommensurate Antiferromagnetism in UTe₂ Under Pressure, William Knafo, LNCMI, CNRS, France **INVITED**

The discovery of multiple superconducting phases in UTe₂ boosted research on correlated-electron physics [1,2,3,4]. The proximity to a ferromagnetic quantum phase transition was initially proposed as a driving force to triplet-pairing superconductivity [1], and this heavy-fermion paramagnet was rapidly identified as a reference compound to study the interplay between magnetism and unconventional superconductivity with multiple degrees of freedom.

Here, we present neutron diffraction experiments showing that long-range incommensurate antiferromagnetic order is established in UTe₂ under pressure [5]. The propagation vector $\mathbf{k}_m = (0.07, 0.33, 1)$ of the observed antiferromagnetic phase is close to a wavevector where antiferromagnetic fluctuations have previously been observed at ambient pressure [6,7]. Our work shows that superconductivity in UTe₂ develops in the vicinity of a long-range antiferromagnetic phase, which differs from the initial proposition of a nearby ferromagnetic phase. The nearly-antiferromagnetic nature of UTe₂ at ambient pressure and its relationship with superconductivity will be discussed.

This work was done in collaboration with T. Thebault, P. Manuel, D. Khalyavin, F. Orlandi, E. Ressouche, K. Beauvois, G. Lapertot, K. Kaneko, D. Aoki, D. Braithwaite, G. Knebel, and S. Raymond.

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- [2] Knebel *et al.*, J. Phys. Soc. Jpn. **88**, 063707 (2019).
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- [7] Knafo *et al.*, Phys. Rev. B **104**, L100409 (2021).

9:30am AC+MI-ThM-7 New Spectroscopic Insights into Correlation Effects and Covalency of U 5f Electrons in Uranium Intermetallic Compounds, A. Marino, M. Sundermann, D. Christovam, Max-Planck Institute for Chemical Physics of Solids, Physics of Correlated Matter, Dresden, Germany; H. Gretarsson, DESY/PETRA-III, Hamburg, Germany; B. Keimer, Max-Planck Institute for Solid State Research, Stuttgart, Germany; A. Gloskovskii, DESY/PETRA-III, Hamburg, Germany; J. Kunes, Department of Condensed Matter Physics, Masaryk University, Brno, Czechia; A. Hariki, Department of Physics and Electronics, Osaka Metropolitan University, Osaka, Japan; L. Tjeng, Max-Planck Institute for Chemical Physics of Solids, Physics of Correlated Matter, Dresden, Germany; Andrea Severing, Institute of Physics II, University of Cologne, Cologne, Germany **INVITED**

Uranium intermetallic compounds exhibit a wide range of fascinating physical phenomena that arise from the intricate interplay between atomic-

like correlations and band formation of the U 5f electrons. Describing their electronic structure is a challenge, particularly as atomic-like states have remained experimentally inaccessible. Ongoing disputes revolve around the existence of atomic-like states and, if present, around determining which configuration—U 5f^β or U 5f^α—provides the Ansatz or starting point for describing the low-energy properties. Furthermore, these correlated compounds exhibit strong intermediate valency, fuelling intense debates about the actual filling of the 5f shell and the degree of covalency.

This presentation introduces two novel and complementary methods: (1) valence band resonant inelastic x-ray scattering (RIXS) using tender x-rays and (2) photon energy-dependent photoelectron spectroscopy (PES & HAXPES) in combination with DFT+DMFT calculations.

(1) Valence Band RIXS: Using high-resolution RIXS at the U M-edges (3.5–3.7 keV), we observe low-energy excitations originating from atomic-like multiplet interactions [1]. The excellent signal-to-background ratio and the multiplet structure serve as a unique fingerprint for the U 5fⁿ configuration involved, offering clear evidence of the presence of local correlation physics. This method unambiguously determines the dominating configuration [1].

(2) Combination of DFT+DMFT calculations with soft and hard x-ray photoemission (PES & HAXPES): The energy dependencies of the photoionization cross-sections allow for the disentanglement of correlated 5f from band-like non-5f spectral contributions, enabling reliable tuning of parameters such as Hund's-J, Hubbard-U_{ff}, and, most importantly, the double counting correction (m_{dc}) to reproduce valence band spectra. We applied this method to UGa₂ and UB₂, two model materials representing the extreme ends of the localization-delocalization range. Despite their vastly different properties, we found the mean 5f shell filling to be almost the same. However, crucially, the distribution of uranium 5f configurations contributing to the ground state differs significantly: narrow in 'localized' UGa₂ and almost statistically broad in 'itinerant' UB₂. This method also reproduces the satellites in the 4f core-level spectra and explains the presence/absence of 5f-5f-multiplet excitations in the RIXS spectra [2].

- [1] A. Marino *et al.* Phys. Rev. B, **108** (2023) 045142
[2] A. Marino, A. Hariki *et al.*, to be published

11:00am **AC+MI-ThM-13 Unusual orders in the heavy-fermion superconductor CeRh₂As₂, Gertrud Zwicknagl**, TU Braunschweig, Germany
The heavy-fermion compound CeRh₂As₂ exhibits a complex phase diagram with rather unusual states at low temperatures. A prominent example is multi-phase superconductivity [1] which seems to develop inside a normal state characterized by itinerant multi-polar order. The present talk focusses on the instabilities of the strongly renormalized Fermi liquid state in the heavy-fermion compound CeRh₂As₂. The central focus is the role played by the non-symmorphic lattice structure and the consequences of the Crystalline Electric Field (CEF) which removes the orbital degeneracy of the Ce 4f states. The narrow quasiparticle bands which arise from the Ce-4f degrees of freedom via the Kondo effect are calculated by means of the Renormalized Band (RB) method. We conjecture that the quasi-quartet CEF ground state in combination with pronounced nesting features of the Fermi surface may give rise to ordered states involving multipolar degrees of freedom [2].

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1. S. Khim *et al.*, Science 373, 1012 (2021)
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11:15am **AC+MI-ThM-14 The Relation between Crystal Chemistry and Superconductivity in Actinide-Based Superconductors, Eteri Svanidze**, Max Planck Institute for Chemical Physics of Solids, Germany

Crystallographic features play an important role in the physical and chemical properties of a given solid-state material. In particular, actinide-based systems exhibit a wide range of properties – from unconventional superconductivity to peculiar magnetic orders. In this talk, I will highlight some of the old and new actinide-based superconductors, in which a delicate interplay between chemistry and physics is observed. A comprehensive characterization of properties of UBe₁₃ has revealed a deep interrelation between the physical and chemical features. Notably, single crystals of this material tend to include many defects which have a dramatic effect on superconducting state [1]. Motivated by this issue, an alternative method of studying intrinsic properties is investigated [2–4]. By creating a micro-scale device, it is possible to measure intrinsic superconductivity of UBe₁₃, which has so far remained out of reach [4]. The properties of UBe₁₃ are compared to those of other actinide-based superconductors – UTe₂ [5] and Th₄Be₃₃Pt₁₆ [6] – in which a strong coupling of lattice and

superconducting properties is observed. By studying these systems, it is possible to expand the understanding of crystal chemistry of solid-state materials, while simultaneously providing an insight into which crystallographic parameters impact the physical properties of a given solid-state material.

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[2] E. Svanidze *et al.*, "Revealing intrinsic properties of UBe₁₃", *in preparation* (2024)
[3] A. Amon *et al.*, "Interplay of atomic interactions in the intermetallic superconductor Be₅Pt", *Angew. Chem. Int. Ed.* **58**, 2 (2019).
[4] I. Antonyshyn *et al.*, "Micro-scale device - an alternative route for studying the intrinsic properties of solid-state materials: case of semiconducting TaGeIr", *Angew. Chem. Int. Ed.* **59**, 2 (2020)
[5] E. Svanidze *et al.*, "Intrinsic crystal structure of UTe₂", *in preparation* (2024)
[6] P. Kozelj *et al.*, "A noncentrosymmetric cage superconductor Th₄Be₃₃Pt₁₆", *Sci. Rep.* **11**, 22352 (2021)

11:30am **AC+MI-ThM-15 Thermoelectric Properties of Strongly Correlated Compounds NpPd₃ and PuPd₃, Krzysztof Gofryk**, Idaho National Laboratory; J. Griveau, 3DG Joint Research Centre, Directorate G-Nuclear Safety and Security, Germany; K. McEwen, University College London, UK; W. Nellis, Harvard University; J. Smith, Los Alamos National Laboratory
Actinides are characterized by the coexistence of localized and itinerant (delocalized) 5f-states near the Fermi energy. This dual nature of the 5f electrons leads to many exotic phenomena that are observed in these strongly correlated materials, spanning magnetic ordering, heavy-fermion ground state, unconventional superconductivity, and/or a "non-Fermi liquid" state. The unusual transport properties of the strongly correlated electron systems are related to the formation, near the Fermi level, of a narrow band with a large density of states. Therefore, the Seebeck coefficient in these materials, being proportional to the density of states at the Fermi level, often reaches large values and shows the characteristic temperature dependence [Phys. Rev. B 94, 195117 (2016)]. To explore the influence of electronic correlations on the transport properties in the AnPd₃ system, here we present low-temperature electrical resistivity and Seebeck coefficient measurements of NpPd₃ and PuPd₃ intermetallics. We show that the electrical resistivity shows characteristic behavior of systems with Kondo interactions. The magnitude and overall temperature dependence of the thermoelectric power of NpPd₃ and PuPd₃ are characteristic of 4f- and 5f-electron strongly correlated materials. For cubic NpPd₃ a sharp transition and change of sign in S(T) at the Néel temperature indicate a first-order nature of the magnetic transition, probably accompanied by a large change in the Fermi surface topology. We have also estimated the power factor (S²/ρ) of these materials.

11:45am **AC+MI-ThM-16 Magnetism and Electronic Transport of U₃Te₄: Contrasting Bulk and Thin Film Studies, S. Alex**, Institute of Physics of the Czech Academy of Sciences, Czechia; O. Koloskova, A. Cabala, Faculty of Mathematics and Physics, Charles University, Czechia; M. Mišek, Institute of Physics of the Czech Academy of Sciences, Czechia; L. Horák, Faculty of Mathematics and Physics, Charles University, Czechia; T. Gouder, F. Huber, European Commission, Joint Research Centre (JRC), Germany; **Evgenia Tereshina-Chitrova**, Institute of Physics of the Czech Academy of Sciences, Czechia

Since half a century ago, substantial research efforts were dedicated to exploring the magnetic properties of uranium pnictides and chalcogenides belonging to the Th₃P₄ type, including the examples of U₃P₄ and U₃As₄. However, U₃Te₄, a member of this group, was overlooked due to the scarcity of high-quality samples. The literature presents conflicting reports regarding the Curie temperature (T_c) of U₃Te₄, with reported values ranging from 105 K [1] to 38 K [2]. U₃Te₄ crystallizes in a body-centered cubic (bcc) cell (space group I4̄3d) with a lattice parameter of a = 9.412 Å. While the presence of non-collinear ferromagnetism, attributed to the unusual three-axial crystal-field anisotropy in the 3:4 compounds, has been proposed for U₃Te₄, the previously available data did not provide a definitive conclusion on this matter. In this study, we not only present first single-crystalline data of U₃Te₄ but also conduct a comparative analysis of its magnetic properties with U₃Te₄ thin films. To produce the single crystals, we utilized the molten salt flux (MSF) method. Our experimental approach involved exploring various U:Te ratios to yield a single-phase U₃Te₄. The resulting sample comprised crystallites of varying sizes, including needle-shaped crystals

measuring approximately 1 mm in length. For thin films production, the dc sputtering technique was employed, with details available in Ref. [3], where it was demonstrated that various U-Te compositions can be stabilized depending on the temperature of the Te target. We investigated the magnetization and compared the ordering temperature in both bulk and thin film U_3Te_4 to understand how the material's dimensionality affects its magnetic properties. Additionally, we studied the magnetic field effects on the electron transport properties of the crystals and films, exploring the influence of chemical and magnetic disorder on material behavior. We acknowledge the support of Czech Science Foundation under the grant no. 18-02344S. The thin films samples were prepared in the framework of the EARL project of the European Commission Joint Research Centre, ITU Karlsruhe. Physical properties measurements were performed in the Materials Growth and Measurement Laboratory (<http://mgml.eu/>) supported within the program of Czech Research Infrastructures (project no. LM2018096). [1] V. I. Chechernikov et al., *Sov. Phys. JETP* **25**, 4 (1967). [2] W. Suski et al., *Phys. Stat. Sol. (a)* **14**, K157-K160 (1972). [3] E.A. Tereshina-Chitrova et al. In *IEEE Int. Magn. Conf. - Short Papers*, pages 1–2, 2023.

12:00pm AC+MI-ThM-17 Theory of Valence-to-Core RIXS Measured at the Uranium M_5 Edge: Comparison of UO_2 and UF_4 . O. Stejskal, Jindřich Kolorenc, Institute of Physics (FZU), Czech Academy of Sciences, Czechia

Motivated by a recent experimental study [1], we model the valence-to-core resonant inelastic x-ray scattering (RIXS) measured at the uranium M_5 edge in insulating compounds UO_2 and UF_4 . We employ the Kramers–Heisenberg formula in conjunction with the Anderson impurity model extracted from the corresponding LDA+DMFT electronic-structure calculations [2], in which the double-counting correction is adjusted to best reproduce the experimental valence-band XPS [3,4]. In our simulations, we find two sets of excited states. One group is formed by excitations of the $5f^2$ shell that appear at energy losses $\lesssim 4$ eV. These excitations are not well resolved in the experimental data [1] as they are largely obscured by the elastic peak. The other group of excited states is formed by the charge-transfer excitations corresponding to a transfer of an electron from the oxygen/fluor 2p states to the uranium 5f shell. We identify these excitations with the spectral feature experimentally observed at an energy loss of roughly 8–10 eV, in agreement with other closely related investigations [5,6]. Our model estimates the intensity, with which the charge-transfer excitations appear in the RIXS spectra, to be larger in UO_2 than in UF_4 , just like it is observed in the experiment [1]. We analyze in some detail how this intensity depends on the strength of the metal-ligand hybridization and on other parameters of the model, such as the magnitude of the core-valence interaction acting in the intermediate state of the RIXS process.

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