Wednesday Afternoon, November 2, 2011

Actinides and Rare Earths Focus Topic
Room: 209 - Session AC+MI-WeA

Magnetic and Electron Correlation Effects in Actinides and Rare Earths

Moderator: J.G. Tobin, Lawrence Livermore National Laboratory

2:00pm **AC+MI-WeA1 Electronic Structure Theory of Complex Ordered Actinide Materials**, ***P.M. Oppeneer***, Uppsala University, Sweden **INVITED**

Actinide materials display many complex and correlated behaviors that originate from the special properties of the open f-shell atom embedded in a specific material's environment. First-principles investigations provide a route to assess these anomalous phenomena in a materials specific way, providing direct, fundamental insight.

Here we consider recently obtained *ab initio* modeling results for actinide materials that are in the focus of current interest: actinide oxides, such as NpO_2, PuO_2, and higher-oxides, U_3O_8, and Np_2O_5, the hidden order (HO) material URu_2Si_2, and correlated plutonium compounds.

NpO_2 is one of the very few materials in which complex multipolar order has been identified. Using the density-functional theory (DFT)-based LDA+*U* method we provide a first-principles theory of multipolar order and superexchange in NpO_2. DFT+*U* calculations offer a precise microscopic description of the 3***q***-antiferro ordered phase. We find that the usually neglected higher-order multipoles (electric hexadecapoles and magnetic triakontadipoles) are at least equally significant as the electric quadrupoles and magnetic octupoles [1].

We further investigate light actinide oxides in higher oxidation states, such as U_3O_8, PuO_{2+x}, and Np_2O_5, for which non-collinear magnetic ordering is predicted. The possible further oxidation of PuO_2 to PuO_{2+x} is studied using DFT+*U* calculations in combination with x-ray absorption measurements [2].

The Pu monochalcogenides are intriguing materials, in which a correlated temperature gap develops, reminiscent of the behavior seen in Kondo insulators. Using dynamical mean field theory (DMFT) in comparison to LDA+*U* calculations, we show that dynamical self-energy fluctuations are important for the formation of an unusual gap. Static approximations to the self-energy as the LDA+*U* fail to provide a gap.

For URu_2Si_2 we report extensive electronic structure investigations [3], using full-potential LSDA, LSDA+*U*, and DMFT approaches to assess the origin of the hidden order. Our investigation show that the itinerant f-electron picture provides an excellent description of the materials properties of this fascinating compound. The Fermi surface which is crucial for the HO transition and the occurrence of unconventional superconductivity is accurately given. Our study points to the formation of long-lived spin fluctuations that are the driving quasiparticles for the HO.

1. M.-T. Suzuki, N. Magnani, and P.M. Oppeneer, Phys. Rev. B **82**, 241103(R) (2010).

2. A. Modin, Y. Yun, M.-T. Suzuki et al., Phys. Rev. B **83**, **075113** (2011).

3. P.M. Oppeneer, J. Rusz, S. Elgazzar, M.-T. Suzuki, T. Durakiewicz, and J.A. Mydosh, Phys. Rev. B **82**, 205103 (2010).

2:40pm **AC+MI-WeA3 Anomalous Quasiparticle Dynamics in the Hidden Order state of URu_2Si_2**, ***T. Durakiewicz***, *G.L. Dakovski, Y. Li, S.M. Gilbertson, G. Rodriguez, A.V. Balatsky, J.X. Zhu, K. Gofryk, E.D. Bauer, P.H. Tobash, A. Taylor, J.L. Sarrao,* Los Alamos National Lab, *P.M. Oppeneer,* Uppsala Univ., Sweden, *P.S. Riseborough,* Temple Univ., *J.A. Mydosh*, Leiden Univ., the Netherlands **INVITED**

An exotic phase of unknown nature emerges from a heavy fermion state in URu_2Si_2 at $T_0 = 17.5$ K. The nature of this hidden order (HO) state is being vigorously debated, while the massive removal of entropy due to the HO transition evades explanation. Here we use time-and angle-resolved photoemission spectroscopy (tr-ARPES) to elucidate the itinerant nature of HO. We show how the Fermi surface is renormalized by shifting states away from the Fermi level at specific hot spots. By measuring the ultrafast dynamics we identify the location and lifetime of the quasiparticle states forming at the hotspots. We find that the quasiparticle lifetime increases from 42 fs to few hundred fs across the HO transition, and the hidden order parameter is related to the anisotropic gapping of the Fermi surface.

4:00pm **AC+MI-WeA7 Advanced X-ray Spectroscopies on 4f and 5f Systems**, ***J. Bradley***, *M. Lipp,* Lawrence Livermore National Laboratory, *A. Sorini*, SLAC National Accelerator Laboratory

Photon-in photon-out x-ray spectroscopies allow for a bulk-sensitive, high-pressure compatible look at rare earth and actinide electronic structure. The techniques couple to well-defined and meaningful quantum mechanical observables, including orbital occupation number and magnetic moment. These observables are key differentiators between theoretical treatments of strongly correlated systems, and they also provide meaningful and correct intuitive understanding. Here we will present an selection of measurements, both at ambient and high pressure, that exemplify the kind of insight these techniques can provide. In particular, we will address the question of rare earth volume collapse, where considerable controversy has existed between competing (Mott vs. Kondo) theoretical treatments.

4:20pm **AC+MI-WeA8 Hard X-Ray Photoelectron Spectroscopy and Electronic Structure of Single Crystal UPd_3, UGe_2, and USb_2**, ***M.F. Beaux***, *T. Durakiewicz, J.J. Joyce, E.D. Bauer, J.L. Sarrao,* Los Alamos National Laboratory, *L. Moreschini, M. Grioni,* Ecole Polytechnique Federale, Switzerland, *F. Offi,* Universita Roma Tre, Italy, *M.T. Butterfield,* KLA-Tencor, *G. Monaco,* European Synchrotron Radiation Facility, France, *G. Panaccione,* Laboratorio Nazionale TASC-INFM-CNR, Italy, *E. Guziewicz*, Polish Academy of Sciences

Hard X-ray Photoelectron Spectroscopy (HAXPES) with 7.6 keV photons has been performed on single crystals of UPd_3, UGe_2, and USb_2 at the European Synchrotron Radiation Facility (ESRF). The greatly reduced surface sensitivity of HAXPES enabled observation of the bulk core levels in spite of surface oxidation. An 800 meV splitting within the Sb 3d core level was observed. The splitting of the Sb core levels is attributed to manifestations of two distinct Sb binding sites within the USb_2 single crystal as supported by consideration of interatomic distances and enthalpy-of-formation. Photoelectron mean-free-path vs oxide layer thickness considerations were used to model the effectiveness of HAXPES for probing bulk features of in-air cleaved samples.

4:40pm **AC+MI-WeA9 Actinide Dioxides under Pressure**, ***L. Petit***, Daresbury Laboratory, UK

The self-interaction corrected local spin density approximation is used to investigate the oxidation of actinide dioxides under pressure. The methodology enables us to determine the ground state valency configuration of the actinide 5f electrons and to study the localization/delocalization transition that occurs under pressure. We argue that this delocalization facilitates the oxidation of the actinide dioxides and present results for the estimated transition pressures.

5:00pm **AC+MI-WeA10 Hybridization and Electronic Structure in Pu Compounds**, ***J.J. Joyce***, *T. Durakiewicz, K.S. Graham, M.F. Beaux, E.D. Bauer, J.N. Mitchell, T.M. McCleskey, E. Bauer, Q.X. Jia, R.L. Martin, J.X. Zhu, J.M. Wills,* Los Alamos National Laboratory, *L. Roy,* Savannah River National Laboratory, *G.E. Scuseria*, Rice University

The electronic structure of Pu materials is directly tied to the details of the 5*f* electron bonding and hybridization. In compounds where direct 5*f*-5*f* bonding is negligible due to crystal structure and wavefunction overlap, hybridization is the key component for 5*f* electron influence on electronic properties. We examine two strongly correlated materials, PuCoGa5 and PuO2 that span the range of interesting materials from Mott insulator to heavy fermion superconductor. The synergy between synthesis, spectroscopy and modeling has provided a unique opportunity to explore details of the energy and crystal momentum dependence of Pu compound electronic structure through angle-resolved photoemission on single crystal samples and advanced modeling based on theories beyond density functional theory.

The strength of the 5*f* electron hybridization may be quantified through dispersion in 5*f* electron peaks from the angle-resolved photoemission. In the case of PuO2, we see over two eV of dispersion in the hybridized (O 2p - Pu 5*f*) valence band. For PuCoGa5, the quasiparticle peak at the Fermi energy shows 50 meV or more of dispersion in reciprocal space over a range covering slightly less than half the zone center to zone boundary. We are unable to follow the peak dispersion beyond this point as it crosses above the Fermi energy. These energy dispersions place significant constraints on models, which might be used to describe the electronic structure of these strongly correlated materials. For PuCoGa5, models, which place the 5*f* electrons in a localized configuration without significant hybridization, would not agree with the experimental results. In the case of PuO2, the dispersion measured in photoemission agrees well with the

hybrid functional calculations for PuO2 and further support the increase in hybridization moving from ionic UO2 to covalent PuO2.

5:20pm **AC+MI-WeA11 Structure and Magnetic Properties of Actinide-Based Thin Films**, ***L. Havela***, Charles University, Czech Republic, *N.-T. Kim-Ngan*, Pedagogical University Cracow, Poland, *A. Adamska*, Charles University, Czech Republic, *A.G. Balogh*, TU Darmstadt, Germany, *T. Gouder*, European Commission, JRC Institute for Transuranium Elements, Germany **INVITED**

Actinide-based sputter deposited films were so far used in the context of surface-science studies (such as [1]) and for exploration of electronic structure by photoelectron spectroscopy (e.g. [2,3]). In addition, sputter deposition was used in attempts to synthesize amorphous uranium alloys for ex-situ studies of magnetic properties. Such early (late 1980's) attempts in U.S. [4] or Japan [5] were undertaken in simple setups and lack proper diagnostics of the deposited material. Considering strong electropositivity of U, oxidation has to be suspected for films prepared in HV conditions. More recently, U metal in multilayers with possibility of epitaxial growth were sputter deposited with the aim to induce uranium magnetic moments [6]. We have used sputter deposition to investigate structure and magnetic properties of various U-based compounds as a function of deposition conditions (deposition rate, substrate type and temperature). Employing diagnostics by XPS, Glancing Angle XRD, and RBS, it was established that UN films have a long-term stability, which allows comfortably to make ex-situ studies over months. The reason can be seen in pronounced compressive residual strains, imposed during the deposition, which prevent progressing the surface oxidation into the bulk of several hundred nm thick films. Departing more from a fully crystalline state, the antiferromagnetism of UN is masked by a weak ferromagnetism, as usual for nanograined AF structures, and finally both moments and their order disappears [7]. Similar suppression of magnetism was found for ferromagnetic US [8].

Recently we undertook sputter-deposition experiments on Fe-rich U-Fe alloys derived from the Laves phase UFe_2, which combines the 3*d* and 5*f* magnetism in a compound with a relatively high Curie temperature (T_C = 162 K). An Fe-excess is expected to increase the T_C value markedly. Nanocrystalline material obtained up to the stoichiometry $UFe_{2.3}$ by splat cooling, with the excessive Fe atoms entering the U sublattice has T_C enhanced up to 230-240 K. More Fe leads to the segregation of α-Fe. We succeeded to synthesize amorphous films by U and Fe co-sputtering, with stoichiometry up to UFe_3. T_C is enhanced up to 450 K in this case.

[1] T. Gouder et al., Surface Science 601 (2007) L77.

[2] T. Gouder et al., Phys.Rev.Letters 84 (2000) 3378.

[3] L. Havela et al., Phys.Rev.B 65 (2002) 235118.

[4] P.P. Freitas et al., J.Appl.Phys. 63 (1988) 3746.

[5] S. Itoh et al., Physica B 281-282 (2000) 230.

[6] R. Springell et al., Phys. Rev. B 77 (2008) 064423.

[7] D. Rafaja et al., J.Alloys Comp. 386 (2005) 87.

[8] L. Havela et al., J. Alloys Comp. 408-412 (2006) 1320.

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