Actinides and Rare Earths Focus Topic
Room: 6 - Session AC+MI+SS+TF-MoM

Electronic Structure and Spectroscopy of Actinides

Moderator: A.J. Nelson, Lawrence Livermore National Laboratory

9:00am **AC+MI+SS+TF-MoM3 Strong Correlations and the Electronic Structure of the Actinide Dioxides**, ***R.L. Martin***, Los Alamos National Laboratory **INVITED**

The series of actinide dioxides (AnO2, An=Pa, ... Cm) are difficult challenges for electronic structure theory. The early members of the series are Mott insulators, the band gap corresponding to *f-f* transitions, while the later members, beginning with PuO2, are O*2p* -> An*5f* charge transfer insulators. I will review recent experimental results (X-ray absorption, photoemission and optical band gaps) which now allow us to distinguish among several many-body approximations to their electronic structure, including the SIC, DFT+U, DMFT+U and hybrid DFT (HSE) approaches.

9:40am **AC+MI+SS+TF-MoM5 Synchrotron Radiation Studies of Actinide Compounds**, ***S.M. Butorin***, Uppsala University, Sweden **INVITED**

Core-to-core resonant inelastic x-ray scattering (RIXS) and valence-to-core RIXS techniques are two complimentary ways for probing the electronic structure in actinide systems. Specific cuts of the core-to-core RIXS maps around *Mβ* and *Lα*lines of actinides represent remarkably improved high-resolution x-ray absorption spectra of actinide *3d* and *2p* edges, respectively, as a result of limited lifetime broadening of core holes present in shallower levels in the final state of the spectroscopic process. That allows for more detailed studies of unoccupied states and better oxidation states assignments. In turn, the valence-to-core RIXS spectra are only limited by the instrumental resolution and provide information about actinide chemical bonding and interactions between valence electrons.

A comparison of experimental data with results of model calculations shows that the resonant spectra of actinide systems recoded at the actinide *M*(*3d*) and *O* (*5d*) thresholds which probe the *5f* states can be interpreted using the many-body theory, such as the Anderson impurity model, while the data obtained at the *L3* threshold and representing the *6d* states of actinides can be described within a single-particle approach, such as LDA+*U* (local density approximation with supplemented Coulomb interaction *U*) framework.

In course of discussion of the above statements, we present the RIXS data for a number of actinide systems with emphasis on the results contributing to understanding of the U-O and Pu-O phase diagrams, in particular data for UO2+x, U4O9, U3O8 and PuO2+x. The influence of the Coulomb interaction between *5f* electrons on the electronic structure of actinides is also discussed.

10:40am **AC+MI+SS+TF-MoM8 Quasiparticle Dynamics in Uranium Systems from Ultrafast Spectroscopies**, ***T. Durakiewicz***, Los Alamos National Laboratory

Every time we add a new dimension to an experimental method, we open a window to novel, unexpected and fascinating phenomena. Here we show the results of our focused effort of adding time-domain to the powerful experimental techniques of Angle Resolved Photoelectron Spectroscopy (ARPES) and reflectivity. The novel tools are applied to actinides and help us understand the details of the electronic structure of the correlated f-electron materials.

In the hidden order system URu_2Si_2 we investigate the massive renormalization of the Fermi surface at specific k values. The application of time-resolved ARPES allowed a direct measurement of the momentum-resolved quasiparticle lifetime which was shown to increase by an order of magnitude at the hidden order transition. Time-resolved ARPES together with the ultrafast reflectivity results provided evidence for forming a multiple gap structure, including the hybridization gap, pseudogap and HO gap [1, 2].

Another actinide system of interest is a Mott insulator UO_2, where we have investigated the complex dynamics of the Hubbard excitons. We have found that the dynamics can be divided into four distinct processes: instantaneous hop, picosecond lattice deformation, phonon emission and relaxation, and the slow relaxation related to the propagation of Hubbard excitons [3]. We have also obtained the first direct measurement of Hubbard gap in 5f system [4].

The novel femtosecond pump-probe methods provide unique information about the dynamics of 5f quasiparticles, and open novel possibilities in addressing the long-standing questions about the role of near-Fermi level band renormalization in establishing the physical properties of correlated materials.

References

[1] Physical Review B 84, 161101(Rapid Comm.) (2011)

[2] Physical Review B 84, 161103(Rapid Comm.) (2011)

[3] Physical Review Letters 106, 207402 (2011)

[4] manuscript in preparation

11:00am **AC+MI+SS+TF-MoM9 Comparison of Spectroscopic Data with Cluster Calculations of Plutonium, Plutonium Dioxide and Uranium Dioxide**, ***J.G. Tobin***, *S.W. Yu, B.W. Chung,* Lawrence Livermore National Laboratory, *M.V. Ryzhkov,* Russian Academy of Science-Ekaterinburg, *A. Mirmelstein*, Russian Federation Nuclear Center-Snezhinsk

Using spectroscopic data produced in the experimental investigations of bulk systems, including X-Ray Absorption Spectroscopy (XAS), Photoelectron Spectroscopy (PES) and Bremstrahlung Isochromat Spectroscopy (BIS) [1-5], the theoretical results within for $U0_2$ [6], PuO_2 [6] and Pu [7] clusters have been evaluated. The calculations of the electronic structure of the clusters have been performed within the framework of the Relativistic Discrete-Variational Method (RDV). [6,7] The comparisons between the LLNL experimental data and the Russian calculations are quite favorable. The cluster calculations may represent a new and useful avenue to address unresolved questions within the field of actinide electron structure, particularly that of Pu. Observation of the changes in the Pu electronic structure as a function of size suggests interesting implications for bulk Pu electronic structure.

Acknowledgements

Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract No. DE-AC52-07NA27344. JGT and SWY were supported by the DOE Office of Science, Office of Basic Energy Science, Division of Materials Science and Engineering. Work at the RAS and VNIITF was supported in part by Contract B590089 between LLNL and VNIITF. The Advanced Light Source (ALS) in Berkeley and the Stanford Synchrotron Radiation Laboratory are supported by the DOE Office of Science, Office of Basic Energy Science .

References

1. J.G. Tobin and S.-W. Yu, Phys. Rev. Lett, **107,** 167406 (2011).

2. S.-W. Yu,J. G. Tobin, J. C. Crowhurst, S. Sharma, J. K. Dewhurst, P. Olalde-Velasco, W. L. Yang, and W. J. Siekhaus, Phys. Rev. B **83**, 165102 (2011).

3. J.G. Tobin, B.W. Chung, R. K. Schulze, J. Terry, J. D. Farr, D. K. Shuh, K. Heinzelman, E. Rotenberg, G.D. Waddill, and G. Van der Laan, Phys. Rev. B **68**, 155109 (2003).

4. J.G. Tobin, P. S ö derlind, A. Landa, K.T. Moore, A.J. Schwartz, B.W. Chung, M.A. Wall, J.M. Wills, R.G. Haire, and A.L. Kutepov, J. Phys. Cond. Matter **20**, 125204 (2008) .

5. S.-W. Yu,J. G. Tobin, P. Olalde-Velasco, W. L. Yang,and W. J. Siekhaus, J. Vac. Sci. Tech. A. **30**, 011402 (2012).

6. M.V. Ryzhkov and A.Ya. Kupryazhkin, J. Nucl. Materials **384**, 226 (2009).

7. M.V. Ryzhkov, A. Mirmelstein, S.-W. Yu and J.G. Tobin, "Probing Actinide Electronic Structure through Pu Cluster Calculations," submitted to Phys. Rev. B, Feb 2012.

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