

Monday Morning, October 28, 2013

Actinides and Rare Earths Focus Topic

Room: 102 C - Session AC+MI+SA+TF-MoM

Actinides and Rare Earths: Experiment and Electron Correlation

Moderator: T. Durakiewicz, Los Alamos National Laboratory

8:20am AC+MI+SA+TF-MoM1 Our Understanding of the Condensed-Matter Physics of Actinides: What have we Learned in 50 Years?, G. Lander, ITU, Karlsruhe, Germany **INVITED**

By 1950 the Manhattan Project and the early nuclear industry had a large legacy of new materials that were poorly understood from a physics perspective. The physics of uranium and plutonium are good examples.

By the mid-1960s progress had been made in applying many physical techniques (many of which, such as sensitive transducers to measure elastic constants, had also been a development of WW II) on the actinide elements and many of their compounds, particularly the simple dioxides. Most theoretical treatments considered the elements and their metallic compounds within the framework of *d* transition-metals, as many properties seemed to follow these metals, rather than those of the *4f* lanthanide series.

By the mid-1970s the group at Argonne National Laboratory had shown, *inter alia*, that a large orbital moment existed in the actinides even if many properties followed itinerant-electron behavior, and the first band-structure calculations showed how difficult it was to resolve this dichotomy.

The discovery of so-called heavy-fermion superconductors, such as UBe₁₃, at Los Alamos National Laboratory in the early 1980s brought considerable prominence to the field and was a precursor, although not recognized at the time, to the discovery of high-T_c materials in 1986. The further discovery (in 2001) of superconductivity at 18 K in PuCoGa₅, also at Los Alamos, shows the key importance of the electronic ground state of the *5f* electrons and how this drastically affects the physical properties.

Theory has always been “behind” experiments in the actinides; however, the experimental results have proved a sensitive test to the most advanced electronic-structure calculations, such as dynamical mean-field theory (DMFT) within the local-density approximation, so that in some respects the actinides have become a “test bed” for the newest theoretical models.

60 years after some of the pioneering condensed-matter experiments on these materials, we have a far better picture of the actinides, the importance of the orbital moments, the relevance of intermediate coupling, and the criterion that determine whether the *5f* states behave as localized or itinerant. However, we do not have *predictive* theories – they are all *reactive*. This implies that we still need to maintain an experimental capability, as these materials will be with us a very long time, even if we abandon nuclear energy.

The challenge today is how to maintain and nurture that experimental capability in a climate where even depleted uranium is regarded with suspicion and its handling demands kilograms of paperwork? Without experiments will theory follow?

9:00am AC+MI+SA+TF-MoM3 5f Electron Localization, J.L. Smith, Los Alamos National Laboratory **INVITED**

The light actinide elements show a large number of crystal structures and low-melting points. At the element americium, this *5f*-electron series finally settles down and looks like the rare-earth series. This occurs because the *5f* electrons have ceased to form energy bands and have localized. The superconductivity of americium proved this. I will review how the tug of war between itinerant and localized behavior leads to all of the interesting properties and touch on such things as quantum critical points. This has impact on the understanding of the other long rows in the periodic table.

9:40am AC+MI+SA+TF-MoM5 Historical Aspects and Perspectives of X-Ray Spectroscopy in Lanthanide and Actinide Materials, G. Kaindl, Freie Universität Berlin, Germany **INVITED**

The talk addresses some aspects in the use of tunable x-rays in studies of electronic and magnetic properties of lanthanide and actinide materials by x-ray absorption fine-structure (XANES) and resonant elastic x-ray scattering (REXS), emphasizing related features of these *4f* and *5f* materials.

Based on early applications of L- and M-edge XANES to lanthanide valence studies, the method was applied to U and Th compounds, exploring XANES at various thresholds (L, M, N, O) [1], and then to the more

radioactive actinides Np, Pu, and Am as well as to high-pressure studies. Subsequently, the method has been improved both experimentally (e.g. high-resolution XANES [2]) and theoretically [3], and even compounds of Pa, Cm, and Cf in solid and aqueous environments have been investigated by now.

Resonant magnetic x-ray scattering was first applied to Ho metal, where the magnetic scattering cross-section is enhanced by a factor of 50, when the x-ray energy is tuned across the L_{III} absorption edge of Ho [4]. Subsequently, much larger enhancements up to 5 magnitudes were observed at the M_V edges of U in UAs [5] and of Eu in the antiferromagnetic (AFM) semiconductor EuTe [6]. In the latter, an epitaxial thin EuTe(111) film was studied, and virtually background-free magnetic Bragg-peaks with pronounced Laue oscillations were found. From these, the AFM order in the EuTe(111) film could be derived with atomic-layer resolution. The magnetization of the outermost layer was found to decrease significantly stronger with temperature than that of the bulk layer [6]. In a further pioneering experiment that employed a synchrotron slicing source for the production of tunable soft x-rays with pulse widths as short as 100 fs (probe pulses) and synchronized pump pulses of 400 nm wavelength, the dynamics of the AFM order could be studied by fs soft x-ray diffraction [7].

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11:00am AC+MI+SA+TF-MoM9 High Energy Resolution X-ray Spectroscopy of f-electron Systems, K.O. Kvashnina, European Synchrotron Radiation Facility (ESRF), France, J.G. Tobin, Lawrence Livermore National Laboratory

This contribution will provide an overview of the possible spectroscopic techniques and experiments that become available for f-electron systems using high energy resolution X-ray emission spectrometer^[1]. As an example we will show the studies of the electronic structure of cerium (Ce) and uranium (U) nanostructured materials by means of high energy resolution fluorescence detection (HERFD) and resonant inelastic X-ray scattering (RIXS) via transitions between core levels and between core and valence levels^[2-5]. The experimental spectral features will be characterized using a variety of theoretical codes including the LDA+*U* approximation within DFT^[6], atomic multiplet theory^[7] and full multiple scattering FEFF^[8].

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11:20am AC+MI+SA+TF-MoM10 Signatures of the γ - α Volume Collapse in Cerium, *M.J. Lipp*, Lawrence Livermore National Laboratory
INVITED

High-pressure x-ray emission measurements of the $L\gamma_1$ (L_2N_4) emission line were used to decide the longstanding debate over the nature of the famous iso-structural (γ - α) volume collapse at 0.75 GPa in elemental cerium that ends in a critical point at 1.5 GPa and 480 K. The satellite structure of this line offers direct access to the total angular momentum observable $\langle J^2 \rangle$ as shown by extended local atomic model calculations and experiences a 30% step-like decrease across the volume collapse. This validates the Kondo model in conjunction with previous measurements of the equation of state at high temperature that were also well fit by the Kondo volume collapse model plus a quasiharmonic representation of the phonons - but could also be reproduced within the Hubbard-Mott framework. The remaining satellite in the α -phase after the volume collapse does not change significantly over the pressure range studied. Direct comparison is made with previous predictions by dynamical mean field theory.

This work was performed under the auspices of the US Department of Energy by Lawrence Livermore National Laboratory under Contract No. DE-AC52-07NA27344 and funded by the Laboratory Directed Research and Development Program at LLNL under project tracking code 12-LW-014. Portions of this work were performed at HPCAT (Sector 16), Advanced Photon Source (APS), Argonne National Laboratory. HPCAT operations are supported by DOE-NNSA under Award No. DE-NA0001974 and DOE-BES under Award No. DE-FG02-99ER45775, with partial instrumentation funding by NSF. APS is supported by DOE-BES, under Contract No. DE-AC02-06CH11357.

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