

Tuesday Morning, October 19, 2010

Actinides and Rare Earths Topical Conference

Room: Isleta - Session AC+MI-TuM

Actinide and Rare Earth Magnetic Interfaces & Nanostructures

Moderator: T. Durakiewicz, Los Alamos National Laboratory

8:20am **AC+MI-TuM2 An Alternative Model for Electron Correlation in Pu**, S.W. Yu, J.G. Tobin, P. Soderlind, Lawrence Livermore National Laboratory

8:40am **AC+MI-TuM3 Correlation Effects in Gadolinium Compounds: Comparison of Theory and Experiment**, P.A. Dowben, University of Nebraska - Lincoln **INVITED**

The apparent Gd 4f binding energy in photoemission spectroscopy is strongly affected by both correlation energy contributions, and final state effects in photoemission. The latter are clearly strongly influenced by metallicity and screening [1]. To understand these various contributions to the observed electronic structure of gadolinium compounds, we compare a series of studies on gadolinium compounds and clusters with pure and alloyed gadolinium films [2]. These materials range from the metallofullerenes, in which a metal atom or cluster is encapsulated inside a fullerene, such as in the “buckyball” C60, to more conventional rare earth compounds like GdN, Gd2O3, Gd doped HfO2 and Gd-Ni alloys. The Gd3+ ion exhibits bonding and hybridization that is quite different for these various compounds. While the total spin multiplicity of Gd is maximum because of its half-filled 4f state, and the atomic orbitals are strongly hybridized with the C60 molecules in the example of Gd@C60 [3] or oxygen in the case of Gd doped HfO2 [4], while the highest possible symmetry site for gadolinium is generally not favored. These results are to some extent a confirmation of theoretical expectations but there are complications. There is experimental evidence for intra-atomic hybridization with both the unoccupied and occupied Gd 4f states, as well as band dispersion even for the occupied Gd 4f states in some systems [2] and spin dependent screening [5].

[1] J.E. Ortega, F.J. Himpsel, Dongqi Li and P.A. Dowben, *Solid State Commun.* **91** (1994) 807-811

[2] T. Komesu, H. K. Jeong, David Wooten, Ya. B. Losovyj, J. N. Crain, M. Bissen, F. J. Himpsel, J. Petrosky, Jinke Tang, Wendong Wang, I.N. Yakovkin, and P. A. Dowben, *Physica Status Solidi B* **246** (2009) 975-980

[3] R. F. Sabirianov, W. N. Mei, Jing Lu, Y. Gao, X. C. Zeng, R.D. Bolskar, P. Jeppson, Ning Wu, A.N. Caruso, and P. A. Dowben, *J. Phys. Cond. Matter* **19** (2007) 082201

[4] Ya. B. Losovyj, I. Ketsman, A. Sokolov, K. D. Belashchenko, P.A. Dowben, J. Tang, Z. Wang, *Appl. Phys. Lett.* **91** (2007) 132908

[5] Hae-Kyung Jeong, R. Skomski, D. Wisbey, P.A. Dowben, *Physics Letters A* **341** (2005) 508-515

This work has been undertaken with Y. Losovyj, T. Komesu, Hae-Kyung Jeong, I. Ketsman, N. Wu, A. Sokolov, J. Colon Santana, W. Choe, K. D. Belashchenko, R. F. Sabirianov, W. N. Mei, Jinke Tang, A.N. Caruso, D. Wooten, R. Skomski, F. Himpsel, E. Vescovo

9:20am **AC+MI-TuM5 Electronic Phase Diagram of Rare Earth Mono-Pnictides and Mono-Chalcogenides**, L. Petit, R. Tyer, Daresbury Laboratory, UK, A. Svane, Aarhus University, Denmark, Z. Szotek, W.M. Temmerman, Daresbury Laboratory, UK **INVITED**

The self-interaction corrected local spin density (SIC-LSD) approximation is used to predict the ground state valency configuration of the manifold of rare earth mono-pnictides and mono-chalcogenides. This is a first-principles methodology that adequately describes the dual character of f-electrons, itinerant vs. localized, and the goal is to combine it with high throughput computing, in order to achieve predictive capability in the search for and design of new materials with specific physical properties. For the rare earth mono-pnictides and mono-chalcogenides we predict an electronic phase diagram composed of metallic, semi-conducting and heavy fermion like regions, and exhibiting valency transitions brought about by a complex interplay between ligand chemistry and lanthanide contraction. Our findings, including the ground state lattice parameters, and the “intermediate valent” character of compounds such as SmO and TmSe, are in excellent agreement with the available experimental data.

10:40am **AC+MI-TuM9 Nanotechnology in the Actinides: Uranium Thin Films and Multilayers**, R.S. Springell, University College London, UK **INVITED**

From the modern scientific landscape of nanotechnology research, the manipulation of elements and compounds on a near-atomic scale has contributed to innovations, some of which affect our everyday life. The growth of thin films and multilayers has allowed systematic studies of reduced dimensionality and the electronic interplay between dissimilar materials. Important applications have emerged, notably the GMR effect. However, such studies of systems containing actinide elements are, to our knowledge, absent, but offer exciting possibilities because of potential hybridization involving the 5f electronic states.

The samples are prepared by a dedicated sputtering facility at Oxford University, UK2.

Our investigation into uranium multilayers, which involves both bulk as well as element specific techniques, compares the structural properties² and electronic interactions^{3,4,5} of uranium interleaved with layers of ferromagnetic Fe, Co, Ni, and Gd. Only in the case of U/Fe is a large magnetic moment induced on the U atoms, close to the interface⁵.

The study of epitaxial uranium films is initially concerned with the nature of the structural constraints incurred by the substrate and buffer⁶ and the potential for stabilizing structures otherwise absent in the bulk. a-U (which is the normal ambient orthorhombic phase) grows well on a Nb buffer on sapphire. In the bulk it exhibits a charge-density wave (CDW) at 43 K, so in epitaxial films an interesting question is the minimum thickness for the CDW to appear; we have already observed the CDW down to 10 nm. Using epitaxial hcp-Gd as a buffer, we have produced hcp-U for the first time⁷. The observed c/a = 1.90 is larger than for any other hexagonal element. We report also our first efforts to prepare epitaxial UO2 films.

11:20am **AC+MI-TuM11 Growth and Characterization of Dy/Y Superlattices**, J. Yu, H. Sato, P.R. LeClair, G.J. Mankey, University of Alabama, J.L. Robertson, R.S. Fishman, Oak Ridge National Laboratory

Inelastic neutron scattering is the world's most powerful tool to study the magnetic excitations of solids. We aim to study spin wave dispersion relations in multilayer structures and how they are affected by the indirect RKKY exchange interaction through nonmagnetic spacers. Dy was chosen as the magnetic film since it has a huge magnetic moment of 10.2 Bohr magnetons and the neutron scattering cross section is directly proportional to the atomic moment. For elastic scattering, a film with thickness of order 100 nm is sufficient, but inelastic neutron scattering has a considerably lower cross-section requiring much more material for measurement. To enable such a study, many samples with tightly-controlled thickness and crystallinity must be fabricated. The neutron experiments can then be performed by stacking 8-10 samples, each with 50-100 superlattice repeats. The number of repeats for each sample is limited by the accumulation of defect structures over the growth front. Using a home-built sputtering system, we have achieved the ability to make suitable samples. The system has computer-controlled magnetron sputtering guns and a sample transfer system which enables continuous sample rotation of a 2" diameter wafer at 900°C during sputtering in an ultra-pure environment. Maintaining a good vacuum is essential since both Y and Dy are highly reactive and form stable oxides with only a minute amount of oxygen in the sputtering environment. In addition, the dispersion relations of the spin wave modes are highly sensitive to thickness variations, so it is essential to create samples with uniform and repeatable layer thicknesses. We report the growth and characterization of epitaxial Dy/Y multilayer samples and our optimization of the growth temperature to produce samples with well-defined superlattice Bragg peaks in high-angle x-ray diffraction.

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11:40am **AC+MI-TuM12 Study of f Electron Correlations in Uranium- and Cerium-Oxides by BIS and XES**, S.W. Yu, B.W. Chung, Lawrence Livermore National Lab, D.G. Waddill, Missouri University of Science and Technology, J.G. Tobin, Lawrence Livermore National Lab

When a high energy electron beam impinges on a material, some electrons of the incident electron beam are decelerated into the unoccupied states of the material with a spontaneous emission of electromagnetic radiation

(bremsstrahlung). This bremsstrahlung process can be considered as the inverse of the photoemission process if the initial and final states are exchanged and the occupied state is replaced by the unoccupied one. BIS (Bremsstrahlung Isochromat Spectroscopy) is a very powerful tool to study the bulk unoccupied electronic structures of materials, minimizing the impact of surface effects.

Recently, we have developed a BIS capability, using an XES-350 system from VG SCIENTA, at the Lawrence Livermore National Laboratory, to investigate the electronic structure of actinides, in which 5f electrons play important roles to determine their physical and chemical properties. Similar to the resonant photoemission, the measured BIS signal can be resonantly enhanced when the electron energy reaches the threshold of the binding energy of a core electron, as a result of the interference between the BIS signal and the x-ray emission signal emitted by a radiative decay of the core hole. In case of cerium-oxide, the BIS signal is enhanced significantly at the $3d \rightarrow 4f$ resonance. However, in the case of uranium-oxide, the BIS signal is enhanced only slightly at the $4d \rightarrow 5f$ resonance.

The x-ray emission can be described as a spontaneous emission of photons in a transition that are allowed by the dipole selection rules between two electronic states. The implication of the dipole selection rules is that, for example, in the case of K-emission, where 1s holes are created, only states with p-character are allowed to decay and the partial p-type density of state is measured in a x-ray emission spectrum. Therefore, the x-ray emission spectra from a variety of core levels allow us to study the symmetries of the occupied electronic states.

We will present the resonant BIS spectra and x-ray emission spectra (U 4f, 4d, O1s, Ce 3d) of the uranium- and cerium-oxides. Based on these data, we will discuss the detailed electronic structures of f electrons in uranium- and cerium-oxides.

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