Actinides and Rare Earths Focus Topic
Room 103C - Session AC+MI-WeM

Magnetism, Complexity, and Superconductivity in the Actinides and Rare Earths (8:00-11:00 am)/Actinide and Rare Earth Theory (11:00 am-12:20 pm)

Moderators: Tomasz Durakiewicz, Los Alamos National Laboratory, Ladislav Havela, Charles University, Prague, Czech Republic, Alexander Lichtenstein, University of Hamburg

8:00am **AC+MI-WeM1 Local Magnetic Properties of Uranium Compounds Probed with XMCD**, ***Fabrice Wilhelm***, *A. Rogalev,* ESRF, France **INVITED**

In these last years actinides and actinide compounds, mainly Uranium based compounds, have been the subject of increasing interest due to their very different magnetic behaviors, such as Pauli paramagnetism, localized and itinerant magnetism, and heavy fermions. The element specific X-ray Magnetic Circular Dichroism (XMCD) spectroscopy technique has been proven over two decades to be the best suitable technique to probe the orbital and spin magnetism and to isolate those different contributions separately using the magneto-optical sum-rule under given assumption regarding the magnetic dipole contribution. XMCD experiments have been reported for a great number of uranium compounds and have permitted to understand which is the ground state. It has also revealed differences between localized and itinerant systems that are not yet fully understood. This talk reviews recent advances in use of polarized x-rays to study local magnetic properties and electronic structure of uranium based compounds.

8:40am **AC+MI-WeM3 Neutron and X-ray Scattering as a Probe of Complex Order in Actinides**, ***Helen Walker,*** STFC, UK **INVITED**

Neutron and X-ray scattering are powerful probes of complex ordering in the actinides, giving access to the spatial and temporal fluctuations of structural and electronic degrees of freedom. To demonstrate the vast array of different types of information obtainable using different scattering techniques, I will focus on the question of the elusive hidden order parameter in URu_2Si_2.

Innumerable different theories have been proposed for the nature of the hidden order [1], with a preponderance directed towards multipolar order. With this in mind I will discuss how X-ray Resonant Elastic Scattering has shown that no quadrupolar ordering is present [2], and future opportunities for probing higher order multipoles. I will also discuss the valence state, which is of vital significance for any multipolar model, and how this might be investigated by probing the f-states at the M-edges using Resonant X-ray Emission Spectroscopy.

More recently, there is a growing consensus pointing towards some electronic nematic state, resulting in an orthorhombic distortion [3,4,5]. Inelastic neutron and X-ray scattering experiments, which have searched for a change in the band structure associated with such a distortion, will be discussed.

[1] J.A. Mydosh and P.M. Oppeneer, *Rev. Mod. Phys.***83**, 1301 (2011)

[2] H.C. Walker, *et al.*, *Phys. Rev. B***83**, 193102 (2011)

[3] R. Okazaki, *et al.*, *Science***331**, 439 (2011).

[4] S. Kambe, *et al.*, *Phys. Rev. Lett.***110**, 246406 (2013).

[5] S. Tonegawa et al., *Nat. Comms.* **5**, 4188 (2014)

9:20am **AC+MI-WeM5 Hydrogen Contributing to 5*f*-localization in UTX Compounds**, ***Silvie Maskova***, Charles University in Prague, Czech Republic; *K. Miliyanchuk,* Ivan Franko National University of Lviv, Ukraine; *S. Danis, B. Vondrackova,* Charles University in Prague, Czech Republic; *O. Stelmakhovych,* Ivan Franko National University of Lviv, Ukraine; *L. Havela,* Charles University in Prague, Czech Republic

U-based ternary compounds, which were studied in large isostructural series, allow large variations of the 5*f* localization due to variable 5*f*-5*f* overlap and the 5*f* hybridization with the *d*- and *p*-states. An additional tuning parameter is the H absorption, working primarily as a negative pressure. At 5*f*-band systems it leads to enhancement of magnetic properties in most of cases [1]. However, this tool is restricted to cases in which substantial H absorption can be achieved. In the system of UTX compounds (T = later transition metal, X = p-metal) with the ZrNiAl structure type, a high absorption was registered only for UNiAl, leading to $UNiAlH_{2.3}$, in which the Neel temperature is enhanced from 19 K to 95 K. Recently we discovered a new UTX compound, UNiZn [2], which is itself non-magnetic, as the 5*f*-*p* hybridization is stronger in the case of smaller Zn than for larger Al. This material forms an equivalent hydride, $UNiZnH_{2.3}$, which is antiferromagnetic (below 50 K). This opened a possibility to study systematically the onset of antiferromagnetism in the system UNiZn-UNiAl and variations of properties of related hydrides. We found that the magnetic order in the precursors sets in between 10 and 30 at.% Al. The Sommerfeld coefficient of electronic specific heat γ increases from 94 mJ/mol.K^2 in UNiZn and reaches 360 mJ/mol.K^2 in the critical area and then it decreases for a higher Al concentration to 164 mJ/mol.K^2 in UNiAl, which is known as a mid-weight heavy fermion. [3]. The hydrides exhibit lower γ-values. γ = 100 mJ/mol.K^2 in the UNiZn hydride decreases monotonously to 67 mJ/mol.K^2 in the hydride of UNiAl. The situation implies that reducing the 5*f*-*p* hybridization even more, we may proceed with strengthening of magnetism in the sense of further increasing T_N and decreasing γ. The natural counterpart of UNiAl is UNiGa with the same structure type but larger X-ligand. However, no attempt to synthesize a hydride of UNiGa was successful in the past. In the present work we undertook a study of the hydrides of quasi-ternary system UNiAl-UNiGa [4], and found that the H absorption can be reached even for high Ga concentrations. Physical properties of Ga-substituted hydrides are currently explored.

[1] S. Maskova, L. Havela, E. Santava, and K. Miliyanchuk, J. of Physics: Conf. Ser.200 (2010) 032040.

[2] S. Maskova, S. Danis, K. Miliyanchuk, O. Stelmakhovych, B. Vondrackova, A.V. Kolomiets, L. Havela, J. Alloys Comp. 646 (2015) 885-892.

[3] L. Havela, K. Miliyanchuk, A. Kolomiyets: Int.J.Mat.Res. 100 (2009) 1182-1186.

[4] H. Maletta, V. Sechovsky, P.A. Veenhuizen, F.R. de Boer, L. Havela, and G. Hilscher, Z.Phys. B – Cond. Matter 72 (1988) 455-460.

9:40am **AC+MI-WeM6 Radiation damage: Experimental Investigation of Aluminum Containing Helium Bubbles at Static High Pressure in a Diamond Anvil Cell**, ***Itzhak Halevy***, Physics Department; *B. Glam,* NRC Soreq, Israel; *S. Maskova,* Charles University, Prague, Czech Republic; *D. Moreno,* NRC Soreq, Israel; *S. Eliezer,* NRC Soreq Shalom Eliezer2, Israel

High pressure investigation of the crystallographic structure and bulk modulus of aluminum containing helium bubbles is presented. The targets were obtained by melting pure aluminum with 0.15% wt.^{10}B powder. The solid targets were neutron irradiated in the Soreq nuclear reactor to get homogeneous helium atoms inside the aluminum boron 10 matrix according to the reaction $^{10}B+n\rightarrow{}^7Li+{}^4He$. The irradiated aluminum was heat treated for accumulation of helium atoms into nanometric bubbles by diffusion . The helium bubbles formation in Al–^{10}B metal was observed by TEM.

The crystallographic structure and the bulk modulus of aluminum loaded up to pressure of 39 GPa in a diamond anvil cell (DAC) were studied in different samples: (1) Al-^{10}B, (2) Irradiated Al-^{10}B with helium bubbles (3) The irradiated Al-^{10}B foil with helium bubbles that was used for TEM observation.

The data was analyzed by the X'Pert Plus (Philips) Rietveld analysis software packages. The high-pressure energy dispersive X-ray diffraction studies were taken at the X17-C beam-line of the National Synchrotron Light Source (NSLS) at BNL [1]. The energy dispersive data was collected with a high purity germanium detector at a fixed Bragg angle (2Θ~=12°). The high-pressure X-ray powder diffraction measurements were taken at discrete pressure steps in the range of 0– 35 GPa. The data wascollected by the EDS technique, using the white beam of the superconducting wiggler magnet at the X17-C beam-line. Typical data collection time was about 10 min. Inthe lower range of the high-pressure range, angle dispersive measurements were carried out in transmission configuration using the image plate technique. The data was analyzed using acommercial Rietveld analysis software packages [2].

The Pressure as function of V/V_0 was fitted by Vinet equation to yield the B-$_0$ and B_0' of the EOS. B_0 and B_0' are the bulk modulus and its pressure derivative at the equilibrium volume V_0 at zero pressure. For our pure Al B_0=68.6GPa while B_0' was kept at 3.6 .

Our early work with SEM and TEM showed He bubals as the sample was radiated.

Quantum Design SQUID equipment was used for magnetic studies of bulk Al-15wt.% B and pure Al. Magnetic measurements were performed in the temperature range 2-300 K and fields up to 7 T.

Results of the susceptibility measurements, *c*(*T*), are summarized . The susceptibility is field independent in the whole temperature range studied. All “samples” have similar weak temperature dependence, with *c*-values somewhat below $1*10^{-10} m^3$/mol.

References

[1] Hu, J.: High pressure X-ray beam line X17C. www.bnl.gov/x17c. Updated 07/01/2007

[2] Rietveld, H.M.: J. Appl. Crystallogr. 2, 65 (1969)

11:00am **AC+MI-WeM10 AVS Gaede Langmuir Award Talk: Multiplets and More for Core-Level Spectra***, Paul Bagus*[*]*,* University of North Texas **INVITED**

Multiplets are an essential aspect of electronic spectroscopies and they must be taken into account to correctly describe these spectra. In particular this is true for the core-level spectra of open-shell systems including both X-Ray Photoelectron Spectroscopy, XPS, and Near-edge X-Ray Adsorption Fine Structure, NEXAFS. Unfortunately, the significance of multiplets for the interpretation and analysis of XPS and NEXAFS is often neglected, quite possibly because of the complex mathematical formalism that is required obtain formal expressions for the multiplets and their energies. In this talk, the focus will be on using the familiar rules for the quantum mechanical addition of angular momentum and for multiplet energies, as given, for example, by Hund’s rules, to provide a qualitative guide to the importance of these splittings. The combination of these qualitative guides with rigorous electronic structure calculations of energies and intensities allows us to understand the origins of the XPS and NEXAFS features in terms of the chemical and physical interactions in a system. This combination also allows us also to understand how ligand field mixings modify atomic descriptions of these core-level spectra. For XPS, we show that the value of multiplet splittings to identify oxidation states depends on the choice of core-level ionized. [1] For NEXAFS, we show that decomposing a spin-orbit split level, or multiplet, into its composition in terms of Russell-Saunders, RS, multiplets gives insight into the origin of the often complex features of the spectra.[2] This is because the selection rules are stricter for the RS multiplets. The role of “shake” excitations from occupied bonding orbitals into un-occupied anti-bonding orbitals will also be considered. Several examples for the spectra of heavy metal oxide systems will be presented to illustrate the value of the methods described.

We acknowledge support for this work by the Geosciences Research Program, Office of Basic Energy Sciences, U.S. DOE.

1. P. S. Bagus*, et al.*, Surf. Sci. **643**, 142 (2016).

2. P. S. Bagus, M. J. Sassi, and K. M. Rosso, J. Electron Spectrosc. Relat. Phenom. **200**, 174 (2015).

11:40am **AC+MI-WeM12 Complex Magnetism of Gd Intermetallics: Ab-initio Theory and Experiment.***, Leon Petit,* Daresbury Laboratory, UK*; D. Paudyal, Y. Mudryk, K.A. Gschneidner, V.K. Pecharsky,* Ames Laboratory, Iowa State University*; M. Lueders, Z. Szotek,* Daresbury Laboratory, UK*; J.B. Staunton,* Warwick University, United Kingdom of Great Britain and Northern Ireland

Using an ab-initio electronic structure theory which includes disordered local moments and strong f-electron correlations, we have investigated the magnetic ordering and critical temperatures of Gd-intermetallics.[1] The theory correctly finds GdZn and GdCd to be simple ferromagnets and predicts a remarkably large increase of Curie temperature with pressure (+1.5 K $kbar^{-1}$) for GdCd, confirmed by our experimental measurements . In our calculations for GdMg, a transition from ferromagnetic to AF1 is observed with increasing pressure, whilst a canted magnetic state is seen to emerge from either the ferromagnetic or anti-ferromagnetic state with lowering the temperature. Replacing 35% of the Mg atoms with Zn removes this transition, in excellent agreement with long-standing experimental data. We conclude that despite being filled and situated at low binding energies, the non-lanthanide metal d-states strongly influence the electronic structure at the Fermi level as well as the magnetic ordering.

[1] L. Petit, D. Paudyal, Y. Mudryk, K. A. Gschneidner, Jr., V. K. Pecharsky, M. Lueders, Z. Szotek, R. Banerjee, and J. B. Staunton, Phys. Rev. Lett **115**, 207201 (2015)

12:00pm **AC+MI-WeM13 Thermodynamics of the Doped $Sm(Co_{1-x}Fe_x)_5$ Alloys: *Ab Initio* Study***, Alexander I. Landa, A. Söderlind, E.A. Turchi,* Lawrence Livermore National Laboratory

$SmCo_5$ (in the hexagonal $CaCu_5$–type structure) magnets exhibit enormous uniaxial magnetocrystalline anisotropy (K_1 ~ 17.2 MJ/m_3) substantially higher than for the $Nd_2Fe_{14}B$ (Neomax) magnets (K_1 ~ 4.9 MJ/m3), and $SmCo_5$ magnets have almost twice higher Curie temperature (T_c ~ 1020 K) than $Nd_2Fe_{14}B$ magnets (T_c ~ 588 K). However the world market of permanent magnets is currently dominated by Neomax magnets (~ 62 %), which possess the highest energy performance with a record energy product of 470 kJ/m_3 that is twice as high as the energy product of $SmCo_5$ magnets, of 231 kJ/m_3. Although $SmCo_5$ magnets are more suitable for high temperature applications than Neomax, due to their relatively low energy performance $SmCo_5$ magnets occupy only ~ 3% of the world market. From a cost point of view, it would be beneficial to substitute Co atoms with Fe because Fe in the Earth’s crust is ~ 2000 times more abundant than Co and consequently much cheaper. In addition, Fe is a ferromagnetic metal with the largest magnetization at room temperature. However $SmFe_5$ is unstable, and does not appear in the equilibrium Sm-Fe phase diagram. Our presentation show results of *ab initio* Density Functional Theory based on the Exact Muffin-tin Orbital (EMTO) method for the heat of formation of $Sm(Co_{1-x}Fe_x)_5$ alloys doped with different metals (Me). The Coherent Potential Approximation (CPA) implemented in the *ab initio* EMTO method allows us to gradually substitute the Co atoms by Fe atoms on the Cu-types sites of the $CaCu_5$–type structure. Previous neutron diffraction studies of $Th(Co_{1-x}Fex)_5$ alloys show that the larger Fe atoms prefer to occupy the 3g-type sites whereas the smaller Co atoms prefer to occupy the 2c-type sites. EMTO-CPA calculations reveal very small region ($x \leq 0.05$) of stability of $Sm(Co_{1-x}Fe_x)_5$ alloys. The Full-Potential Linear Muffin-tin Orbital (FPLMTO) calculations for $SmCo_5$ and $SmFe_5$ end points of $SmCo_5$-$SmFe_5$ phase diagram give similar results to those given by the EMTO method. We calculate the heat of formation of the pseudo-binary $SmFe_3(Me_{1-x}Co_x)_2$ alloys where Fe atoms occupy all the 3g-type sites and the occupation of the 2c-type sites gradually changes from pure Me ($SmFe_3Me_2$ compound) to pure Co ($SmFe_3Co_2$ compound) within the CPA. Our calculations show that $SmFe_3(Me_{1-x}Co_x)_2$ alloys could remain stable until approximately half of Me atoms are substituted by Co atoms. This work performed under the auspices of the US DOE by LLNL under Contract DE-AC52-07NA27344. This research is supported by the Critical Materials Institute, an Energy Innovation Hub funded by the US Department of Energy, Office of Energy Efficiency and Renewable Energy, Advanced Manufacturing Office.

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[*] **Gaede Langmuir Award Winner**

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