Actinides and Rare Earths Focus Topic
Room 22 - Session AC+MI+SA+SU-MoM

Magnetism, Complexity, and Superconductivity in the Actinides and Rare Earths

Moderator: Tomasz Durakiewicz, Los Alamos National Laboratory

8:20am AC+MI+SA+SU-MoM-1 Magnetic and Transport Characteristics in the Uranium Intermetallic Compounds with the $HoCoGa_5$-type Structure, *Yoshinori Haga,* Japan Atomic Energy Agency, Japan INVITED
Physical properties of actinides compounds are mainly dominated by the 5f electrons behavior. The spatial extent of the 5f wave function and resulting hybridization with the neighboring atoms lead to itinerant characters. On the other hand, well isolated 5f electrons carry magnetic moment. Competition between those two characteristics often bring about peculiar electronic properties. Among a series of actinides compounds, so-called 115 compounds crystallizing in the tetragonal $HoCoGa_5$-type structure have extensively been studied because of unusual correlated electronic states, including heavy effective mass of conduction electrons, magnetic orderings and heavy fermion superconductivity. Those compounds are also good cases to study electronic transport properties in detail because of availability of extremely high quality samples. Among them, we discuss electronic properties of $URhX_5$ (X = Ga and In). While $URhIn_5$ orders antiferromagnetically at fairly high tempearture 98 K, isostructural and formally isoelectronic analogue $URhGa_5$ is a nonmagnetic semimetal[1,2,3]. Detailed transport measurements as well as the de Haas-van Alphen experiment revealed Fermi surface characteristics. We discuss origin of magnetism of uranium 5f electrons and its relationship between superconductivity realized in Pu-analogues.

[1] Y. Matsumoto et al., Phys. Rev. B 88, 045120 (2013).

[2] S. Ikeda et al., Physica B 359-361, 1039 (2005).

[3] Y. Matsumoto et al, JPS Conf. Proc. 3, 011097 (2014).

9:00am AC+MI+SA+SU-MoM-3 Magnetic Structures of Layered U_nRhIn_{3n+2} Materials, *Attila Bartha, M Klicpera,* Charles University, Prague, Czech Republic*; P Čermák,* Forschungszentrum Juelich GmbH, Germany*; B Ouladdiaf,* Institut Laue Langevin, France*; P Javorský, J Custers,* Charles University, Prague, Czech Republic
Materials of reduced dimensionality appear in many contemporary fields of research and technology, because they encompass a wide variety of interesting electronic phenomena. For instance carbon can be prepared in 3D (diamond), quasi-2D (graphite), 2D (graphene) or 1D (carbon nanotubes). All of these structures have distinct electronics. Diamond is an insulator. Graphene is semimetal. However, when the dimensionality is increased by putting several graphene layers together (eventually making graphite), the resulting band structure moves to that of a more trivial metal. Another example is high temperature superconductors being quasi-2D materials as well.

The role of dimensionality in f-electron systems has been mainly discussed in the context of quantum phase transitions and related phenomena. The series $Ce_nT_mIn_{3n+2m}$(n=1, 2; m=0, 1, 2; T=transition metal) of layered compounds, which can be viewed as m TIn_2–layers alternating with n–layers of $CeIn_3$ along the c-axis, has been extensively investigated. $CeIn_3$ is cubic (3D) and orders antiferromagnetically (AFM) at T_N = 10.2 K [1] with a propagation vector $\mathbf{k}$ = (0.5, 0.5, 0.5) [2]. Under hydrostatic pressure superconductivity appears with highest T_c = 0.3 K at p = 2.5 GPa. In $CeRhIn_5$, the anisotropic crystal structure leads to an incommensurate magnetic structure described with $\mathbf{k}$ = (0.5, 0.5, 0.297). The AFM order is reduced (T_N = 3.8 K) while superconductivity is supported, T_c increases to 1.9 K at p = 1.77 GPa [3-4].

We report on the magnetic structures of $URhIn_5$ and U_2RhIn_8, two new members of this intriguing $R_nT_mX_{3n+2m}$(R= Lanthanide, Actinide, X = In, Ga) family of compounds. Neutron diffraction measurements were performed on structurally well-defined single crystals. Both, $URhIn_5$ and U_2RhIn_8, adopt the tetragonal Ho_nCoGa_{3n+2}-type structure (P4/mmm) typical for this group of compounds. $URhIn_5$ orders antiferromagnetically below T_N = 98 K. The propagation vector equals $\mathbf{k}$ = (0.5, 0.5, 0.5) and we obtained a value of 1.65 μ_B/U^{3+} for the size of the ordered magnetic moment. The antiferromagnetic transition temperature of U_2RhIn_8 is higher yielding 117 K. We found that the magnetic structure can be described by propagation vector $\mathbf{k}$ = (0.5, 0.5, 0). The respective ordered magnetic moment amounts 1.7 μ_B/U^{3+}. In both materials the ordered magnetic moments are aligned along the tetragonal c-axis. Comparison with isostructural compounds and general conclusions for the series will be presented.

[1] C. Pfleiderer, Rev. Mod. Phys. 81 (2009) 1551

[2] A Benoit *et al.*, Solid State Commun. 34 (1980) 39

[3] H. Shishido *et al.* J. Phys. Soc. Jpn. Vol. 71 Suppl. (2002) 276

[4] H. Hegger *et al.,* Phys. Rev. Lett. 84 (2000) 4986

9:20am AC+MI+SA+SU-MoM-4 U_3Si_2 – Physical Properties and Resistance to Hydrogen, *Silvie Maskova,* Charles University, Prague, Czech Republic*; K Miliyanchuk,* Ivan Franko National University of Lviv, Lviv, Ukraine*; S Middleburgh,* Westinghouse Electric Sweden AB, Vasteras, Sweden*; L Havela,* Charles University, Prague, Czech Republic
U_3Si_2 (tetragonal structure with the space group of P4/mbm and two different U-positions, U1 and U2, in the unit cell) is considered to be promising material (due to its high density of uranium) as an accident-tolerant nuclear fuel [1] with rather high melting point (1938 K) indicating a high thermodynamic stability. From this point of view, it is very important to study its resistance to oxygen or hydrogen, as it can significantly influence the integrity of the material. U_3Si_2 was reported to oxidize at elevated temperatures [2].

We studied the H absorption and concomitant changes of basic electronic properties. We found that U_3Si_2 reversibly absorbs hydrogen (H can be released again by heating up to approx. 700 K) to the level of approximately 1.8 H/f.u., i.e. yielding $U_3Si_2H_{1.8}$. The H absorption proceeds at very low H pressures (kPa range) already, resulting in 10 % volume expansion. The temperature-induced desorption experiment showed that the hydrogen atoms are located in one specific position only. The crystal structure arrangement suggests that two different positions should be considered - U_3Si tetrahedra (similar to U_3T tetrahedra in U_2T_2X compounds [3] crystallizing in an ordered ternary derivative of the U_3Si_2 structure) and a split position in the U_6 octahedra consisting of 4 U1 and 2 U2 atoms (H atom shifts from the central position into the tetrahedra formed by 2 U1 and 2 U2 atoms. As H generally does not occupy two adjacent tetrahedra, we can assume only two from the 4 tetrahedra occupied). The later was confirmed as more plausible option by ab-initio calculations using Vienna Ab-initio Simulation Package (VASP) with the PBE-GGA exchange correlation.

Magnetic studies of U_3Si_2 confirmed that it is a Pauli paramagnet, as reported previously [4]. The volume expanded hydride reveals a Curie-Weiss behavior and a weak and inhomogeneous ferromagnetism arising gradually below T = 100 K. The low-temperature specific heat of $U_3Si_2H_{1.8}$ shows an upturn and a dramatic enhancement of the Sommerfeld coefficient of electronic specific heat γ, which reaches 440 mJ/mol f.u. K^2 (γ = 88 mJ/mol f.u. K^2 for U_3Si_2).

This work was supported by The Czech Science Foundation under the Grant No. 15-01100S.

[1] K.D. Johnson, A.M. Raftery, D.A. Lopes, J. Wallenius, J. Nucl. Mater. 477 (2016) 18-23.

[2] E. Sooby Wood, J.T. White, A.T. Nelson, J. Nucl. Mater. 484 (2017) 245-257.

[3] K. Miliyanchuk, L. Havela, A.V. Kolomiets, A.V. Andreev, Physica B 359–361 (2005) 1042-1044.

[4] T. Miyadai, H. Mori, T. Oguchi, Y. Tazuke, H. Amitsuka, T. Kuwai and Y. Miyako, J. Magn. Magn. Mater. 104-107 (1992) 47-48.

9:40am AC+MI+SA+SU-MoM-5 Understanding Surface Chemistry of f-element Oxides using First-principle Methods, *Ping Yang,* Los Alamos National Laboratory INVITED
Interfacial chemistry for f-element materials is critical for nuclear waste management and to address the environmental concerns associated with actinides. Cerium is often used as a surrogate for more radioactive actinide elements for thorough characterization using advanced spectroscopic techniques. Herein, we will present recent progress in characterizing interfacial interactions of ligands with CeO_2 and ThO_2 in both surface and nanoparticle forms. We will demonstrate how the synergy between theory and experiment has accelerated the progress in this field. The theoretical results are validated by spectroscopic characterizations. A better molecular-level understanding of the interfacial chemistry including coordination, energetics and reaction mechanisms will pave the way to a better strategy for waste management and environment remediation.

10:40am AC+MI+SA+SU-MoM-8 Inelastic X-ray Scattering Study of the Crystal Dynamics of Neptunium and Uranium Dioxide, *Roberto Caciuffo*, European Commission, Joint Research Centre, Karlsruhe, Germany; *P Maldonado*, Uppsala University, Sweden; *L Paolasini*, European Synchrotron Radiation Facility, France; *P Oppeneer*, Uppsala University, Sweden; *T Forrest*, European Synchrotron Radiation Facility, France; *A Prodi*, Consiglio Nazionale delle Ricerche, Italy; *N Magnani*, European Commission, Joint Research Centre, Karlsruhe, Germany; *A Bosak*, European Synchrotron Radiation Facility, France; *G Lander*, European Commission, Joint Research Centre, Karlsruhe, Germany **INVITED**

The energy-wavevector dispersion relations for normal modes of vibration propagating along high-symmetry lines in NpO_2 and UO_2 have been determined by measuring the coherent one-phonon scattering of X-rays from single-crystal specimens with mass varying from a few µg to ~1 mg. The inelastic X-ray scattering (IXS) experiments were carried out using the ID28 beamline at ESRF with an incident energy E=17.794 keV. The results are compared against *ab initio* phonon dispersion simulations computed within the first-principles density functional theory in the generalized gradient approximation plus Hubbard U correlation (GGA+U) approach, taking into account third-order anharmonicity effects in the quasiharmonic approximation.

In the case of NpO_2, the sample of dimension of $0.4x0.3x0.3mm^3$ was oriented with the specular direction along the (100) crystal axis and the (011) axis in the scattering plane. We have investigated different Brillouin zones in order to optimize the inelastic structure factor for the different optic branches. Optic phonons arise mainly from oxygen vibration modes and are very weak. Thermal expansion, heat capacity, thermal conductivity, phonon linewidth, and thermal phonon softening are calculated and compared with experimental data available for both NpO_2 and UO_2 [1]. We show that optical phonons contribute significantly to the heat transport due mainly to their large velocities and short lifetimes. Compared with UO_2, the main differences in the phonon density of states of NpO_2 are a softening of the optical modes and an increase of the peak centred around 55 meV, whereas the acoustic modes in NpO_2 are shifted to higher frequencies. The calculated value at 0 K of the bulk modulus in NpO_2 is in agreement with the experimental value and slightly smaller than the one determined by high-pressure X-ray diffraction for UO_2. NpO_2 has a smaller thermal conductivity than UO_2, at least in the temperature range 600 to 1000 K for which experimental values are available.

The lattice dynamics of UO_2 have been extensively investigated by neutron scattering both at low and high temperature. In this study we focussed our attention to the low temperature region, searching for vibronic contributions to the vibrational-magnetic-quadrupolar mixed modes that appear in UO_2 because of magnetoelastic and multipolar superexchange interactions. The situation is cleaner than with neutrons, as the latter see both magnons and phonons – the IXS spectra just the phonons. This observation opens a new window on these "hidden excitations", which drive much of the physics of UO_2.

[1] P. Maldonado *et al.,* Phys Rev B **93**, 144301 (2016)

11:20am AC+MI+SA+SU-MoM-10 Emergent Phenomena in 4f Heavy-Fermion Systems: from Bulk to Thin-Films, *Priscila Rosa*, Los Alamos National Laboratory **INVITED**

The interplay between magnetism and unconventional superconductivity in bulk materials has been one of the central topics in condensed matter physics for the past 40 years. In particular, the family of compounds $CeMIn_5$ (M = Co, Rh, Ir) provides an ideal platform for such investigation due to the small energy scales involved, and the high sample quality. In this talk, I will first discuss novel insights provided by substitution studies in bulk $CeRhIn_5$ under pressure. I will then present recent efforts to fabricate hybrid superlattices consisting of alternating superconducting $CeCoIn_5$ and antiferromagnetic $CeRhIn_5$.

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