2.1.2 Report on IWOSMA-3

International Workshop on Orbital and Spin Magnetism of Actinides (IWOSMA-3)

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Timing
Number of days : 2
Start : 2006-06-01
end : 2006-06-02

Location of the activity
CECAM, 46 allée d'Italie, 69007 Lyon, France

Support

CECAM
Psi-k

Description

This International Workshop on Orbital and Spin Magnetism of Actinides (IWOSMA) is the third in a series. The first workshop took place in Daresbury in 1999 and the second in Berkeley, CA, USA in 2002. These workshops are informal gatherings of theoreticians and experimentalists addressing the latest issues in the electronic and magnetic properties of actinides.

Scientific Objectives

The magnetism of transition metal systems and lanthanide systems is now fairly well understood, where d and f electrons can be described in a delocalized and localized model, respectively. On the other hand, actinide systems do not fit in such a description. The localization of the 5f is in between that of the 3d and 4f and the strong spin-orbit interaction necessitates a relativistic approach. Furthermore, electron

correlation effects play a major role in these compounds. Recently, it has become possible to determine element-specific magnetic moments using neutron diffraction and x-ray scattering and absorption. The latter technique makes it even possible to separate the orbital and spin contribution to the total magnetic moment. The results are very interesting but difficult to reproduce with present state-of-art calculations. Not only a very large orbital polarization but also a large magnetic dipole term has been measured in cubic compounds, such as US. This allows for severe testing of the extra terms included in band theory to account for orbital polarization. It is also clear that deeper insight in magnetism can be obtained by studying the unusual behaviour of the actinides. The recent development and application of such techniques as DMFT could contribute to the understanding of magnetism in actinides. Despite the fact that actinides for health reasons will find less application in technological market products, the understanding of their magnetic and electronic properties will no doubt provide key elements for a general description of electron correlation and relativistic effects.

Presentation List

How to calculate the critical temperature of superconductors

E.K.U. Gross

Freie Universitat Berlin, Germany

Abstract

The traditional model of Bardeen, Cooper and Schrieffer (BCS) properly describes the universal features that all conventional superconductors have in common, but it is not able to make accurate predictions of materials specific properties such as the critical temperature. To tackle this problem, a novel density-functional approach is developed [1] which treats both the electron-phonon interaction and the electron-electron repulsion on the same footing. The formalism can be viewed as the superconducting generalization of the multi-component density-functional theory [2] for electrons and nuclei. Approximations of the universal xc functionals are derived on the basis of many-body perturbation theory [1,3]. In this way, a true ab-initio description is achieved which does not contain any empirical parameters. Numerical results for the critical temperature, the gap, the jump of the specific heat, and the exponent of the isotope effect will be presented for simple metals [4], for MgB_2 [5], and for Li, Al and K under pressure. In particular, for MgB_2, the two gaps and the specific heat as function of temperature are in very good agreement with experimental data. For Li and Al under pressure the calculations explain why these two metals behave very differently, leading to a strong enhancement of superconductivity for Li and to a clear suppression for Al with increasing pressure [6]. Finally, the peculiar properties of Pb are found to arise from two-gap superconductivity, somewhat similar to MgB_2.

References

[1] M. Lüders et al, Phys. Rev. B **72**, 024545 (2005).

[2] T. Kreibich, E.K.U. Gross, PRL **86**, 2984 (2001).

[3] S. Kurth, M. Marques, M. Lüders, E.K.U. Gross, PRL **83**, 2628 (1999).

[4] M. Marques et al, Phys. Rev. B **72**, 024546 (2005).

[5] A. Floris et al, PRL **94**, 037004 (2005).

[6] G. Profeta et al, PRL **96**, 047003 (2006).

Orbital Ordering in actinide oxides: new perspectives on old problems

Gerry Lander
JRC-ITU, France

Abstract

The behaviour of the actinide oxides has been of major interest since the 1940s. UO_2 is known to order with antiferromagnetic order since neutron experiments of the 1960s. However, the puzzle of the lack of ordered magnetism in NpO_2 (also dating from the same period) was recently solved by the discovery [1,2] that the ordering phenomenon in NpO_2 is not connected with dipole moments (as in UO_2), but rather with the long-range ordering (at 25 K) of the anisotropic 5f charge distribution surrounding the Np^{4+} ion. Such ordering involves the 5f quadrupole charge distributions, and is commonly known as "orbital ordering". It cannot be observed with neutrons (or muons) but can be measured by resonant x-ray scattering. Following the work on pure NpO_2, we have re-examined single crystals of $(U_{0.75}Np_{0.25})O_2$ and shown that both the U and Np ions in this material exhibit dipole and quadrupole ordering [3]. All theoretical models of UO_2 emphasize the importance of the interplay between the Jahn-Teller and quadrupolar interactions, but direct evidence for such quadrupolar ordering was difficult to find in UO_2 as the quadrupolar interactions are weaker than the dominating dipole ones. We have now provided that evidence [4] and also shown definitively the triple-q nature of the magnetic ordering in UO_2. [5] This talk will review these experiments, some of the theories currently being advanced, and what remains to be done in understanding these complex actinide oxides at low temperature.

References

[1] J. A. Paixao et al., Phys. Rev. Lett. **89**, 187202 (2002)
[2] R. Caciuffo et al., J. Phys. CM **15**, S2287 (2003), and references therein
[3] S. B. Wilkins et al., Phys. Rev. B **70**, 214402 (2004)
[4] S. B Wilkins et al., Phys. Rev. B **73**, 060406 (R) (2006)
[5] E. Blackburn et al., Phys. Rev. B **72**, 184411 (2005)

Determining the Electronic Structure of Pu using Unorthodox Spectroscopies

James Tobin
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Abstract

The standard method to determine the band structure of a condensed phase material is to (1) obtain a single crystal with a well defined surface and i (2) map the bands with angle resolved photoelectron spectroscopy (occupied or valence bands) i and inverse photoelectron spectroscopy (unoccupied or conduction bands). i Unfortunately, in the case of Pu, the single crystals of Pu are either nonexistent, i very small and/or having poorly defined surfaces. i Furthermore, effects such as electron correlation and a large spin-orbit splitting in the 5f states i have further complicated the situation. i Thus, we have embarked upon the utilization of unorthodox electron spectroscopies, i to circumvent the problems caused by the absence of large single crystals of Pu with well-defined surfaces. i The talk will include a discussion of resonant photoelectron spectroscopy [1], i x-ray absorption spectroscopy [1,2,3,4], i electron energy loss spectroscopy [2,3,4], i Fano Effect measurements [5], i and bremstrahlung isochromat

spectroscopy [6], including the utilization of micro-focused beams to probe single-crystallite regions of polycrystalline Pu samples. [2,3,6] This work was performed under the auspices of the U.S. DOE by Univ. of California, Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

References

1. 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, "Resonant Photoemission in f-electron Systems: Pu and Gd", Phys. Rev. B **68**, 155109 (October 2003).

2. K.T. Moore, M.A. Wall, A.J. Schwartz, B.W. Chung, D.K. Shuh, R.K. Schulze, and J.G. Tobin, "The Failure of Russell-Saunders Coupling in the 5f States of Plutonium", Phys. Rev. Lett. **90**, 196404 (May 2003).

3. G. van der Laan, K.T. Moore, J.G. Tobin, B.W. Chung, M.A. Wall, and A.J. Schwartz, ,"Applicability of the spin-orbit sum rule for the actinide 5f states," Phys. Rev. Lett. **93**, 097401 (Aug 2004).

4. J.G. Tobin, K.T. Moore, B.W. Chung, M.A. Wall, A.J. Schwartz, G. van der Laan, and A.L. Kutepov, "Competition Between Delocalization and Spin-Orbit Splitting in the Actinide 5f States," Phys. Rev. B **72**, 085109 (2005).

5. S.W. Yu, T. Komesu, B.W. Chung, G.D. Waddill, S.A. Morton, and J.G. Tobin, "f-electron correlations in nonmagnetic Ce studied by means of spin-resolved resonant photoemission," Phys. Rev. B **73**, 075116 (2006); J.G. Tobin, S.A. Morton, B.W. Chung, S.W. Yu and G.D. Waddill, "Spin-Resolved Electronic Structure Studies of Non-Magnetic Systems: Possible Observation of the Fano Effect in Polycrystal Ce," Physica B, 378-380, xxxxx (May 2006).

6. J.G. Tobin, M.T. Butterfield, N.E. Teslich Jr., R.A. Bliss, M.A. Wall, A.K. McMahan, B.W. Chung and A.J. Schwartz, "Using Nano-focussed Bremstrahlung Isochromat Spectroscopy (nBIS) to Determine the Unoccupied Electronic Structure of Pu," in "Recent Advances in Actinide Science," published by the Royal Society of Chemistry, as part of the Actinides 2005 Meeting in Manchester, UK, July 05; accepted 2005

Electronic properties of Pu-Am

Ladia Havela

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Abstract

The specific position of Pu just at the localization threshold makes it very sensitive to external variables. Surprizingly, various allotropic phases have nearly identical weak magnetic susceptibility, i despite a large volume expansion exceeding 20% for the fcc delta-Pu comparing to monoclinic alpha-Pu. i This makes the scenario, in which the volume expansion is the way to formation of magnetic moments, rather questionable.

To resolve the issue whether an additional lattice expansion can lead to a 5f localization beyond the delta-Pu level and/or to onset of magnetism we focused on the Pu-Am system. The solid solution of Am in Pu stabilizes the fcc phase down to the lowest temperatures, expanding the volume. Available experimetal data indicate that the magnetic properties remain weakly paramagnetic up to at least 30% Am. Similarly, the gamma coefficient of the specific heat per Pu atom does not increase, and very little variations of the character of the 5f states are indicated by photoelectron spectroscopy.

The experimental findings can be understood on the basis of LSDA+U calculations of the Pu-Am system,

which reveal very little impact of varying the Am concentration and expanding the lattice on the Pu-5f states and their non-magnetic state.

Some aspects of the calculation of spin and orbital magnetization densities

Mike Brooks
Uppsala, Sweden

Abstract

The relationships between current, magnetic moment and magnetization densities are discussed. The approximations made in obtaining these densities from Ab initio energy band calculations are outlined. Particular emphasis is placed upon the interpretation of neutron diffraction experiments on intermetallics containing cerium or actinides with small magnetic moments, where the bonding electron density and spin density from the broad 5d or 6d bands becomes important. Since the bonding spin densities are absent in atomic calculations the entire interpretation of the experiments may be changed. Examples are $CeFe_2$ and UFe_2 where consistency between neutron diffraction and XMCD experiments is obtained.

Strong Correlations Across the Actinide Series:

Gabi Kotliar
Rutgers, United States

Abstract

The actinides realizations of many interesting many body physics phenomena such as the Mott transition, and provide the motivation for the developments of first principles methods to correlated materials.

In this talk we will discuss some of the lessons learned from applying realistic Dynamical Mean Field Theory to Plutonium and Americium and contrast the Mott transition in these two systems.

Calculation of Spectroscopic Properties of Actinide compounds

Hubert Ebert
Univ. München, Germany

Abstract

Electron spectroscopies are the most direct experimental tool to probe the electronic structure of a material. However, to allow for a one-to-one comparison with theory the calculation of corresponding spectra has to deal in an adequate way with all aspects of the electronic structure and also to account properly for the experimental situation. For actinide compounds this requires in particular an adequate treatment of relativistic as well as correlation effects. In our contribution we present a corresponding theoretical description of magnetic Compton scattering that is used to probe the spin magnetic properties. In a second study dynamical mean field theory (DMFT) is used to achieve at an improved description of the optical and magnetooptical properties of actinide compounds.

Spectral Density Functional Calculations in d and f electron systems

Sergej Savrasov

University of California, United States

Abstract

We will discuss spectral density functional method used to calculate total energies and one electron spectral functions of strongly correlated systems. The method employs dynamical mean field theory and exact diagonalization technique as a self-consistent algorithm to find redistribution of spectral weight originally deduced from density functional LDA band structures. As a result, a much wider class of systems showing phenomena such as Mott transition, atomic multiplets, Kondo effect, ordered and disordered atomic magnetism, etc can be explored. Aplications to Mott insulating oxides, actinides and heavy fermion superconductors will be dicussed and contrasted to experimental data and predictions of LDA band structures.

High Pressure Structural Phase Transformations in Curium, Americium and Americium Curium alloys

Rajeev Ahuja

University of Uppsala, Sweden

Abstract

Density-functional electronic structure calculations have been used to investigate the high pressure behavior of curium (Cm), americium (Am) and americium-curium alloys (AmCm). The phase transitions from fcc to the low symmetry structures are shown to originate from a drastic change in the nature of the electronic structure induced by the elevated pressure. For the low density phases, an orbital polarization correction to the local spin density(LSD) theory was applied. Gradient terms of the electron density were included in the calculation of the exchange/correlation energy and potential, according to the generalized gradient approximation (GGA). Theory compares rather well with our recent experimental data (Science 309, 110 (2005)). The new phase (Cm-III) phase is linked to magnetism.

Ab initio study of the electronic properties and Fermi surface

Sebastien Lebegue

LCM3B (UMR UHP -CNRS 7036), France

Abstract

The electronic structure of the uranium dipnictides UX_2 (X=As, Sb, and Bi) is investigated by means of it ab initio calculations based on density functional theory. The calculated Fermi surfaces are presented and compared to available experimental models obtained from de Haas-van Alphen experiments. In agreement with experiments they are found to have a significant two dimensional character. Also, the change of the electronic properties through the series is discussed.

References

S. Lebègue, P. M. Oppeneer, and O. Eriksson
Phys. Rev. B 73, 045119 (2006)

Localization of 5f Electrons and Phase Transitions in Americium

Michel Penicaud

CEA, France

Abstract

Density-functional electronic calculations have been used to investigate the high-pressure behavior of americium. The phase transitions calculated agree with the recent sequence obtained experimentally under pressure; double hexagonal close packed → face centered cubic → face centered orthorhombic → primitive orthorhombic. In the first three phases the 5f electrons are found localized, only in the fourth phase (Am IV) the 5f electrons are found delocalized. The localization of the 5f electrons is modeled by an anti-ferromagnetic configuration which has a lower energy than the ferromagnetic ones. In this study the complex crystal structures have been fully relaxed.

The microscopic 5f magnetisation of the superconductor PuCoGa5

Arno Hiess

ILL, France

Abstract

$PuCoGa_5$ is a superconductor with a critical temperature of T = 18 K and an upper critical field estimated to exceed Bc2 $\rangle$ 70 T; both parameters are records in this class of intermetallic strongly correlated electron systems [1]. The superconductivity is thought to be unconventional and carried by strongly correlated electrons involving the 5f electrons of plutonium [2]. No magnetic order has been observed in $PuCoGa_5$ down to T = 1 K, the lowest temperature measured. Those measurements suggest the bulk magnetic susceptibility to follow a modified Curie-Weiss behaviour in the normal state with an effective moment μ = 0.5 μ_B, and the induced magnetic moment at B = 10 T and T = 20 K had been proposed to correspond to about μ = 0.01 μ_B. To investigate the microscopic magnetic properties further and possibly to shed light on the relevance of the magnetism for the superconductivity, we here report on experiments using polarised neutron diffraction. This experimental technique is sensitive to periodically arranged magnetic moments only and allows determination of the Pu 5f form factor. The method is it insensitive to any magnetic contribution from random impurities or sample mount. The polarised neutron experiment has been performed on the hot neutron diffractomater D3 at the ILL i after determination of crystallographic parameters under similar experimental conditions by non-polarised neutron diffraction on D9. We used a large single crystal (2 * 2 * 4 mm^3 grown from 242Pu. We discovered that the induced magnetic moment is even smaller than previously reported. New bulk susceptibility measurements performed on the same single crystal are consistent with the neutron investigations. This result challenges the currently discussed models for the superconductivity in this weakly magnetic compound.

References

[1] J. L. Sarrao et al., Nature 420, 297 (2002)
[2] N. J. Curro et al., Nature 434, 622 (2005)

Lattice dynamics of light actinides

Johann Bouchet
CEA-DIF, France

Calculated de Haas-van Alphen frequencies of NpCoGa5

Ingo Opahle
IFW Dresden, Germany

Abstract

The electronic structure and magnetic properties of $NpCoGa_5$ are investigated in the framework of relativistic density functional theory in the local spin density approximation (LSDA) with and without orbital polarization (OP) corrections. A detailed analysis of the Fermi surface is presented. Comparison of the calculated angular dependence of the de Haas-van Alphen frequencies with recent experimental data shows that LSDA reproduces the main features of the Fermi surface topology, while the spin and orbital moments of $NpCoGa_5$ are less well described. The inclusion of OP corrections leads to a very good agreement between calculated and measured de Haas-van Alphen frequencies, but does not yield a significant improvement of the calculated magnetic properties. We predict that $NpCoGa_5$ shows an intrinsic GMR effect at moderate magnetic field.

References

I. Opahle, S. Elgazzar, V. D. P. Servedio, Manuel Richter, and
P. M. Oppeneer, Europhys. Lett. 74 (2006) 124.

Excess Magnetic Susceptibility Arising From Self Damage in Pu and Pu Alloys

Scott McCall
LLNL, United States

Abstract

The 5f-electrons of both α-Pu and δ-Pu occupy a narrow f-band as indicated by both the magnitude of the electronic specific heat and the large Pauli magnetic susceptibility ($\sim$ 500 μemu/mol Pu), with no indication of local magnetic moments in the "pure" metal. Pu decays by emission of a 5 MeV alpha particle and corresponding 86 keV U recoil, generating a damage cascade of $\sim$2500 vacancies and interstitials. Most of these immediately recombine, but a few hundred defects remain, and at low temperatures (T$\langle$ 35K), they become frozen within the lattice. This damage is observable as an increase in the magnetic susceptibility with time (damage) that returns to the undamaged initial (t=0) value after annealing to 350K. Thus the excess magnetic susceptibility (EMS) arises from the defects, and not the decay products which are unaffected by thermal annealing. The EMS is well described by a two component function: χ_ν(1-exp(-t/τ))+χ_D't, where both χ_ν and χ_D' are functions of temperature, while the characteristic time, τ, is temperature independent with $\tau_\alpha \rangle \tau_\delta$. These characteristic times are inversely proportional to a volume of magnetic influence arising from the damage cascades that is significantly

larger than the region of physical damage predicted by simulations. The temperature dependence of χ_ν obeys a Curie-Weiss law which suggests there are evolving localized magnetic moments and hinting at characteristics of the 5f electrons hidden in the undamaged material. This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

Probing the magnetism of acinides with μSR techniques

Alain Yaouanc

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Abstract

As the nuclear magnetic resonance (NMR) techniques, the positive muon spin rotation and relaxation (μSR) techniques provide information on the static and dynamical properties of the magnetic field at the probe site. Taking examples from the published literature, we shall illustrate the possibilities offered by these techniques.

One of their key characteristics is their ability to detect magnetic moments which may not be dipole-like. NpO_2 offers a typical example. One may observe magnetic reflections in diffraction techniques while no magnetic moments are detected by μSR because of their relatively fast fluctuation rate. UPt_3 is an example. In contrast to NMR, measurements can be carried out in truely zero field. This allows to detect quasi-static small itinerant magnetic moments as in UGe_2. Finally, we shall discuss the investigation of the superconductivity of materials, in particular the symmetry and the temperature dependence of the London penetration depth.

Magnetic Susceptibility of $PuCoGa_5$ from ITU-SQUID measurements

Franck Wastin

Karlsruhe, Germany

Abstract

Magnetism and unconventional superconductivity are interdependent in strongly correlated electron systems. $PuCoGa_5$ displays superconducting behavior, with a critical temperature of Tc = 18.5 K and an upper critical field Bc2(0) estimated to exceed 70 T [1], considered to be unconventional and carried by strongly correlated 5f electrons [2]. No magnetic order has been observed down to T = 1 K but the temperature dependence of the magnetic susceptibility of $PuCoGa_5$ is indicative of local-moment behavior close to that expected for Pu^{3+}. Initial bulk measurements on a single crystal were reported to follow a modified Curie-Weiss behavior in the normal state with an effective moment μ_{eff} = 0.68 μ_B, and an induced magnetic moment at B = 1 T and T = 20 K corresponding to about 5.55 mμ_B. However, further unpublished experiments on various single and polycrystalline samples led to a large discrepancy in the induced moment measured. In this work, we present the efforts conducted at ITU to distinguish the intrinsic magnetic susceptibility of $PuCoGa_5$.

References

[1] J. L. Sarrao et al., Nature 420, 297 (2002)
[2] N. J. Curro et al., Nature 434, 622 (2005)

X-ray Magnetic Circular Dichroism study of U/Fe multilayers

Fabrice Wilhelm
ESRF, France

Abstract

X-ray magnetic circular dichroism (XMCD) measurements have been performed at the U M4,5-edges and Fe K-edge on well-defined uranium/iron multilayers with different compositions. i The multilayers have layer thicknesses in the range 9 to 40 A for uranium and 9 to 34 A for iron. i Temperature and element-specific magnetic hysteresis curves show that the easy magnetization axis lies in-plane. i At both 10 K and room temperature, the U layers have been found to be magnetically polarized in all of the multilayers studied. An induced U magnetic moment (maximum value $\sim 0.1\ \mu_B$, coupled parallel to the Fe magnetic layers through the exchange coupling, has been observed and is mainly located at the interfaces. The value of the orbital to spin ratio for the induced U moments is ~ 2.1, suggesting moderate hybridization between the U 5f and Fe 3d states.

Theory of Am and Pu systems.

Axel Svane
Aarhus, Denmark

Abstract

Two aspects of the theory of electronic structure of actinides are addressed. Equilibrium lattice constants and effective valencies are calculated on the basis of the self-interaction corrected local spin density total energy functional. We consider the actinide elements Pu, Am, Cm and Bk, as well as alloys of Pu and Am. Secondly, the spectral function of well localized actinide systems is evaluated on the basis of the multiplet Hubbard-I approach, where an additional interaction is introduced to represent fluctuations of the f-shell configuration. The approach is applied to Am and PuSe, and gives good agreement with photoemission data.

Program

Day 1: June 01 2006

Session : 1 Experimental Aspects

09:30 to 10:15 : Presentation

Orbital Ordering in actinide oxides: new perspectives on old problems
Gerry Lander

10:15 to 11:00 : Presentation
Determining the Electronic Structure of Pu using Unorthodox Spectroscopies
James Tobin

11:00 to 11:30 : Coffee Break

11:30 to 11:55 : Presentation

11:55 to 12:20 : Presentation
Magnetic Susceptibility of PuCoGa5 from ITU-SQUID measurements
Franck Wastin

12:20 to 14:00 : Lunch Break

Session : 2 Theoretical Aspects
14:00 to 14:40 : Presentation
Some aspects of the calculation of spin and orbital magnetization densities
Mike Brooks

14:40 to 15:20 : Presentation
Theory of Am and Pu systems.
Axel Svane

15:20 to 16:00 : Presentation
Spectral Density Functional Calculations in d and f electron systems
Sergej Savrasov

16:00 to 16:30 : Coffee Break

16:30 to 17:00 : Presentation
Calculation of Spectroscopic Properties of Actinide compounds
Hubert Ebert

Day 2: June 02 2006

Session : 3
09:00 to 09:40 : Presentation

Strong Correlations Across the Actinide Series:
Gabi Kotliar

09:40 to 10:05 : Presentation
Excess Magnetic Susceptibility Arising From Self Damage in Pu and Pu Alloys
Scott McCall

10:05 to 10:30 : Presentation
High Pressure Structural Phase Transformations in Curium, Americium and Americium Curium alloys
Rajeev Ahuja

10:30 to 11:00 : Coffee Break

11:00 to 11:15 : Presentation
Ab initio study of the electronic properties and Fermi surface
Sebastien Lebegue

11:15 to 11:40 : Presentation
Electronic properties of Pu-Am
Ladia Havela

11:40 to 12:05 : Presentation
Localization of 5f Electrons and Phase Transitions in Americium
Michel Penicaud

12:05 to 12:30 : Presentation
X-ray dichroism studies of the crystalline and magnetic properties of UFe2
Andrei Rogalev

12:30 to 14:25 : Lunch Break

Session : 4
14:25 to 14:50 : Presentation
The microscopic 5f magnetisation of the superconductor PuCoGa5
Arno Hiess

14:50 to 15:15 : Presentation
How to calculate the critical temperature of superconductors
E.K.U. Gross

15:15 to 15:30 : Presentation
Lattice dynamics of light actinides

Johann Bouchet

15:30 to 16:00 : Coffee Break

16:00 to 16:15 : Presentation
X-ray Magnetic Circular Dichroism study of U/Fe multilayers
Fabrice Wilhelm

16:15 to 16:40 : Presentation
Calculated de Haas-van Alphen frequencies of NpCoGa5
Ingo Opahle

Organizer's report

Conclusions

1. Great meeting.

2. Exciting science.

3. Expanding field.

4. Are we starting to understand the electronic structure of Pu and other actinides ? The valence electronic structures of the actinide metals and alloys in general and plutonium (Pu) in particular remain mired in controversy. Interestingly, the various phases of Pu metal provide a microcosm of the metallic actinides as a whole. Thus, unravelling the nuances of the interplay of electronic and geometric structures in Pu will illuminate the properties of all transuranic metals. In a sense, the behavior of the Pu 5f electrons is completely counter-intuitive. The dense phase, alpha, has some semblance of delocalization in the 5f valence bands and can be treated theoretically within single electron models such as the Local Density Approximation. The alpha phase is monoclinic, which is a low symmetry ordering. The less dense delta-phase is fcc and exhibits evidence of localized and/or correlated electronic behavior. The fcc is a high symmetry phase which is normally associated with superior wavefunction overlap in d state metals. But herein is the key: the linear combinations of the 5f's do not produce the nicely lobed wavefunctions with symmetry about the x, y, z and diagonal axes, as occurs for d states. Instead, the linear combinations of f -states have oddly lobed and badly directed wavefunctions which match very poorly with the high symmetry of the fcc structure. In the rare earths and lanthanides, this odd lobing is of no consequence: generally, the 4f valence states are inside the outer 5d, 6p and 6s electrons and can be treated as isolated, atomic-like orbitals that do not participate in chemical bonding. In the actinides, the 5f's are less well shielded. Shielding is undoubtedly a key issue and relates to the volume changes. In fact, the 20% volume increase of Pu between the alpha and delta phases is a reflection of the discontinuous 30% volume jump between Pu and Am, as one moves along the row of actinide elements. Furthermore, the fcc delta- Pu can not be treated theoretically without ad hoc corrections, e.g., the addition of Coulombic repulsion terms to LDA formalisms. Finally, we have not even begun to deal with the issue of dilute alloy formation and its impact upon the electronic structure of metallic actinides. The

upshot of this is that the valence electronic structure of Pu in particular and the actinides in general is only poorly understood.

5. There are still a lot of challenges ahead

Recommendations

1. Let's meet again: IWOSMA-4 is planned to be held in Prague and to be organized by Ladia Havela.
2. Do more experiments on actinides.
3. Work on clean samples.
4. Use a multi-technique approach and explore core-hole spectroscopy.
5. Do more calculations for these systems taking their correlated nature into account.

Participant List

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