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Experimental Bench-marking of Pu Electronic Structure

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Title: Experimental Bench-marking of Pu Electronic Structure

Two deliverables

1. Complete commissioning of the combined Fano (Spin) and BIS Spectrometer in Room 1226, B235.
2. Perform a detailed study of Ce using Fano and BIS measurements.

Plan Description:

Our plan is to do Ce (as a Pu surrogate) this year and be ready to do Pu next year. The Fano (Spin-resolved Photoelectron Spectroscopy) measurements are essential to testing electron correlation in the occupied 5f states. BIS (Bremstrahlung Isochromat Spectroscopy or high energy Inverse Photoelectron Spectroscopy) experiments are crucial to a quantitative determination of the 5f unoccupied density of states (5f-UDOS). The 5f UDOS is the key to differentiation between a myriad of models of 5f electronic structure. During this time, we will work to converge to a solution for the Pu safety issues, with the plan to implement these in the next FY. Acceleration of this schedule and implementation of the safety plan in this FY will require a very significant increase in funding. Ultimately, results from the Pu experiments will be fed into calculations performed by P. Soderlind, A. Landa, and others.

Q3 Progress Report

Q3 Status: Yellow – significant issues regarding completion

%Complete – 75%

Budget: > 25% over, based upon \$300K/yr budget

Costs for October 2006 – June 2007 = \$290K

Based upon \$300K/yr budget – 29% over spent

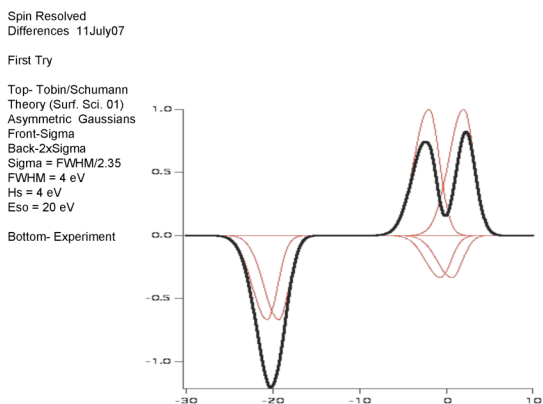
Q3 Status - significant achievements, reports, documents, presentations, etc.

- i. Fano spectra: observation of nuclear mag effects in “nonmag” mat’ls
- ii. XES/BIS construction and implementation
- iii. Publication – Pt Fano Manuscript accepted by Surface Science Letters
- iv. Manuscript – new approach to Pu PES with Per Soderlind
- v. Russian Workshop: talk and two posters

i. Fano spectra: observation of nuclear mag effects in “nonmag” mat’ls

Ultimately we will use Fano measurements of the valence states of Ce and Pu to probe electron correlation in these systems. The Fano Effect is the observation of spin polarized photoelectron emission from NONMAGNETIC materials, under chirally selective excitation, such as circularly polarized photons. Observation of strong Fano dichroic effects have been made in the past for core levels in non-magnetic systems, with both circular and linear polarization. Because of the vectorially chiral selection rules for these processes, unpolarized radiation can produce the same effect, albeit with about twice the background (and thus about 1/2 of the percentage dichroism) relative to properly linearized polarization.

During our calibration studies of the core levels of the non-magnetic materials Cu, Au and Pt, we came upon a very intriguing and novel result: the possible observation of NUCLEAR MAGNETIC splitting in the core levels of Cu and Au, and absence in Pt. This makes sense: Cu and Au have large magnetic nuclear spins and Pt has a much smaller value, based upon a statistical averaging of the available nuclear spin ground states. Although these experiments are completed and present lab work is focused upon the BIS and Fano of Ce, further “theoretical” analysis is in progress.



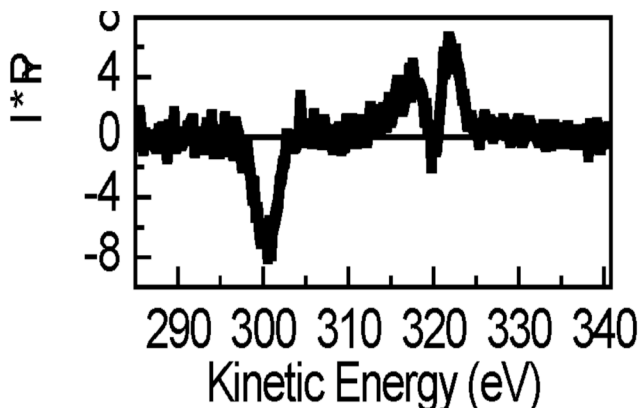
Core Level Fano measurements of Cu, using the Cu2p states.

Spin Resolved Differences

Top-Theory

Bottom-Experiment

This data was collected in Room 1226 in B235.



ii. XES/BIS implementation and testing

We will be using Bremsstrahlung Isochromat Spectroscopy to probe the unoccupied electronic structure of Ce and Pu. The photon detection will be performed with a commercially available soft x-ray spectrometer, the XES350, that we have already purchased from Gammadata Scienta. The mechanical supports for the XES-350 have been constructed and the XES-350 has been implemented into the vacuum system in Room 1226, B235. Its present position is shown below.

Experimental Configuration for Fano PES and BIS

Top

Schematic of the new XES-350 monochromator-detector system for BIS.

Near middle

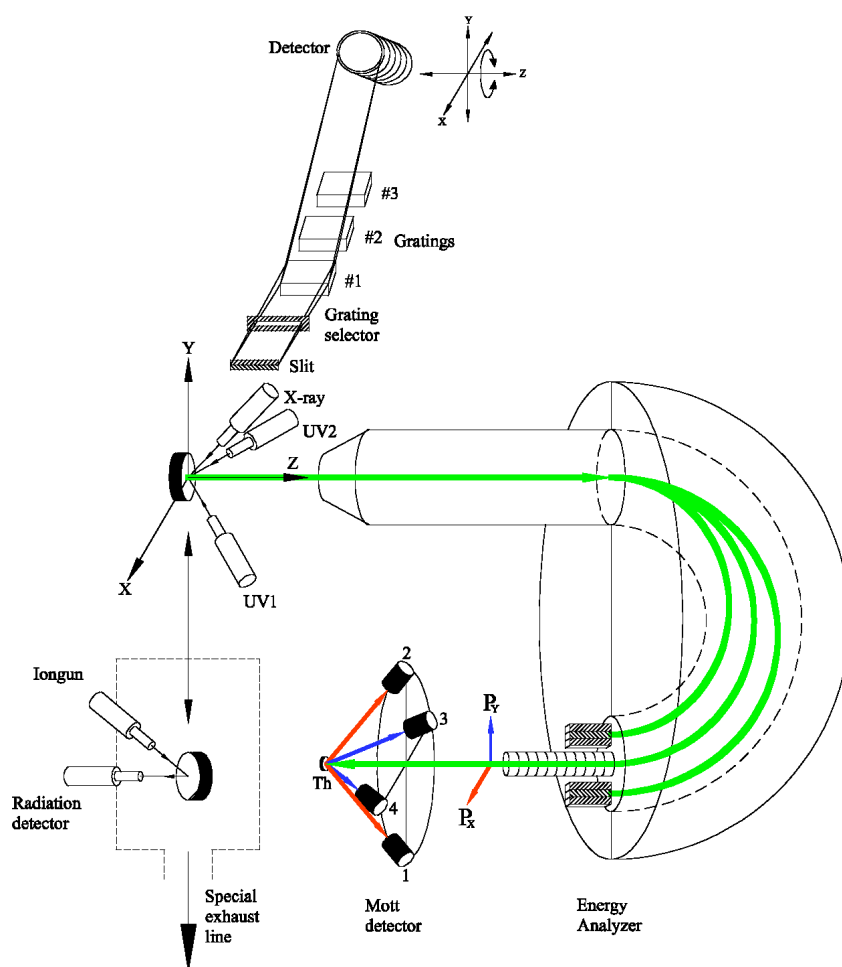
Analysis position including two VUV sources for chirality reversal and an x-ray source.

Far middle and bottom

Hemispherical electron energy analyzer with multichannel detection and true spin analysis using a Mott detector for Fano PES.

Near bottom

Isolated sample cleaning position for actinides.



iii. Publication – Pt Fano Manuscript accepted by Surf. Science Lett.

“Observation of an underlying relativistic effect in the valence bands of Pt”

S.-W. Yu and J. G. Tobin, LLNL, Livermore, California 94550, USA

We have measured the photoelectron spin polarization emitted by unpolarized UV radiation from the valence bands of the well ordered Pt(001)-(5x1) surface and the disordered surface destroyed by Ar ions bombardment. Almost identical spin polarizations have been observed in both cases. This observation suggests that the electron spin polarization in photoemission caused by unpolarized light is determined by a short-range order of atoms. This finding has an obvious implication that the electron spin polarization in photoemission caused by unpolarized light can be used to study the bulk electronic structure of the nonmagnetic materials.

iv. Manuscript – new approach to Pu PES with Per Soderlind

JGT and Per Soderlind are working on a new approach to testing the electronic structure of Pu, combining Per's LDA magnetic calculations with magnetic cancellation with JGT's spectroscopic model, as published earlier in the Journal of Alloys and Compounds.

v. Russian Workshop: talk and two posters

Dates of Travel: 6/21/07 – 6/30/07

DOE Trip Number(s): 706284

Funding Source*
(DOE/HQ Sponsor): DNT

Destinations (Installation, City, and Country): Frankfurt, Germany, Nizhniy Novgorod, Russia and Sarov, Russia

Statement of Trip Purpose: To attend 7th Russian-US Pu Workshop at VNIIEF in Sarov, Russia

List of Persons Contacted: Please see attendee list, especially Boris Nadykto (VNIIEF), Lidia Timofeeva (VNIINM), Victor Pushkov (VNIIEF) and Andrey Kutepov (VNIITF).

List of Facilities Visited: VNIIEF, Sarov, Russia: Hall of Scientists, ISKRA Laser Fusion Facility, Bomb Museum

Abstract (i.e., major highlights, benefits of the travel, results of meetings): JGT presented one talk and two posters. They were entitled: (Oral Presentation) “Observation of Dynamical Spin Shielding in Ce: Why It Matters for Pu Electronic Structure,” (Poster Presentation) “On the Electronic Configuration in Pu,” and (Poster Presentation) Spectroscopic Signature of Aging in δ -Pu(Ga) for the Roundtable on Aging and Phase Stability.”
JGT met with Andrey Kutepov (VNIITF) and planned a resumption of the interlab contract.