LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

LLNL-JRNL-401237

Soft X-ray Studies of Pu Electronic Structure: Past Lessons and Future Directions

J. G. Tobin, S. W. Yu

February 11, 2008

Symposium Proceedings of the Materials Research Society

Disclaimer

Spring 2008 Materials Research Society Meeting
Actinides IV, Symposium NN
San Francisco, CA, USA
March 24-28, 2008

Soft X-ray Studies of Pu Electronic Structure: Past Lessons and Future Directions

J.G. Tobin and S.W. Yu

Lawrence Livermore National Laboratory, LLNS-LLC, Livermore, CA, USA

Photoelectron Spectroscopy (PES) [1] and X-ray Absorption Spectroscopy (XAS, Figure 1) [2-4] have contributed greatly to our improved understanding of Pu electronic structure. From these and related measurements, the following has been determined.

1. The Pu 5f spin-orbit splitting is large.
2. The number of Pu5f electrons is near 5.
3. The Pu 5f spin-orbit splitting effect dominates 5f itineracy.

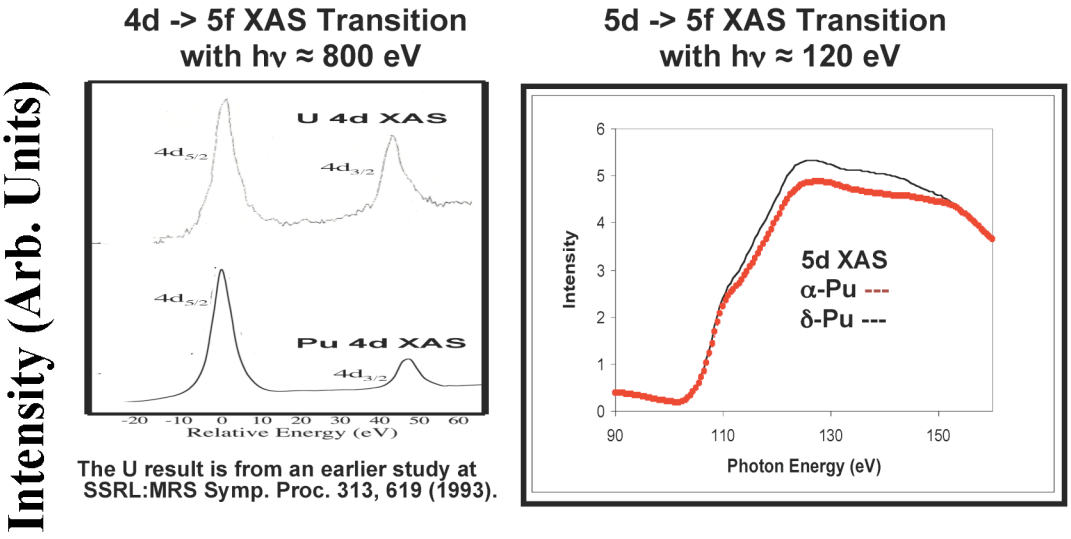

Figure 1

Significant questions remain concerning the nature of Pu electronic structure. Perhaps the missing piece of the puzzle is the direct experimental determination of the unoccupied electronic structure using high energy inverse photoelectron spectroscopy or Bremstrahlung Isochromat Spectroscopy (BIS). [5] Past BIS studies of Th and U indicate the feasibility and utility of Pu studies. [6]

To this end, a new BIS capability has been developed in our laboratory, as shown schematically in Figure 2 below. [7]

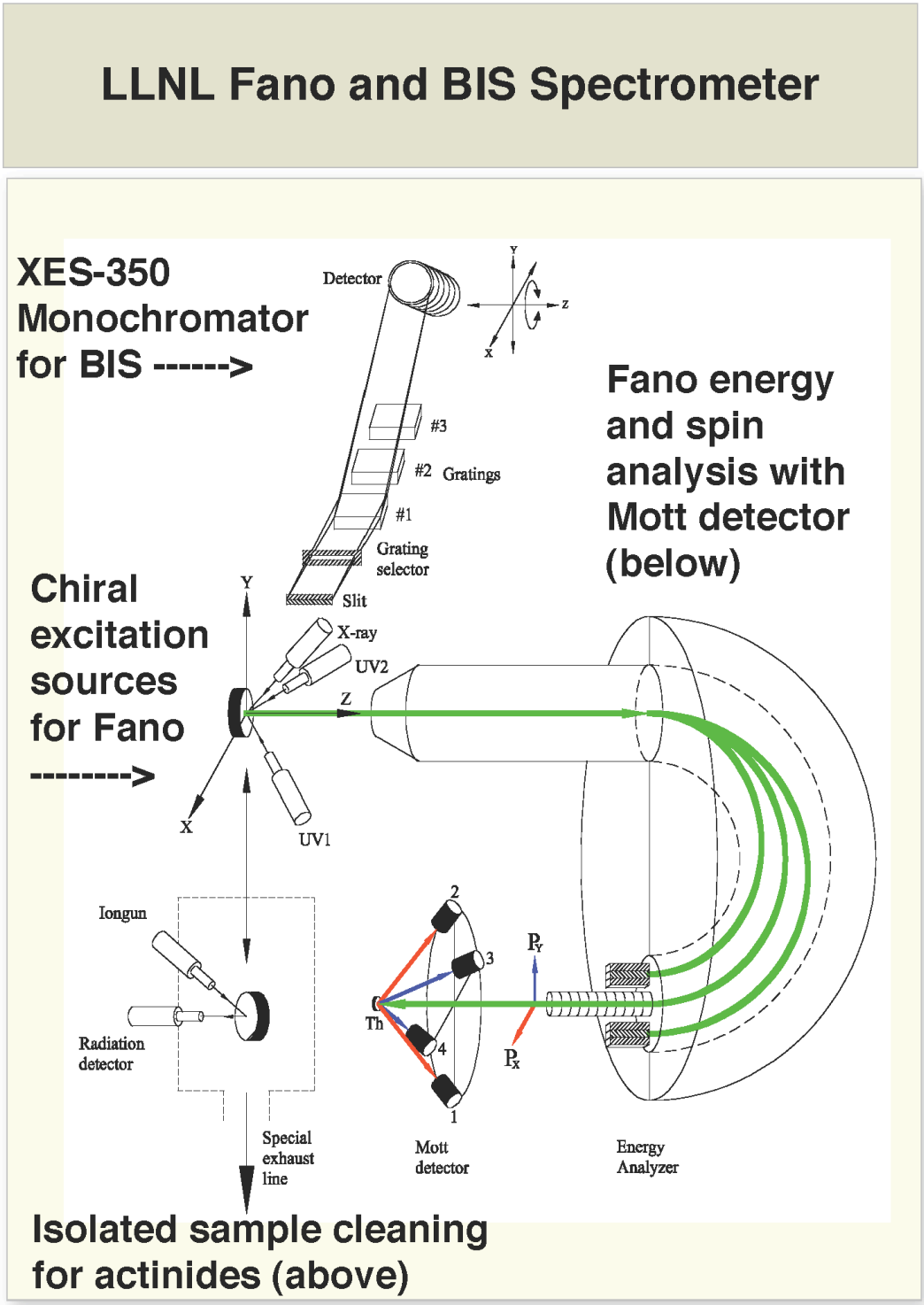

Figure 2

Electron stimulated emission of photons has been carried out using the XES-350 monochromator and detector system. Some of our preliminary results are shown below, using an electron excitation beam energy of 3000 eV.

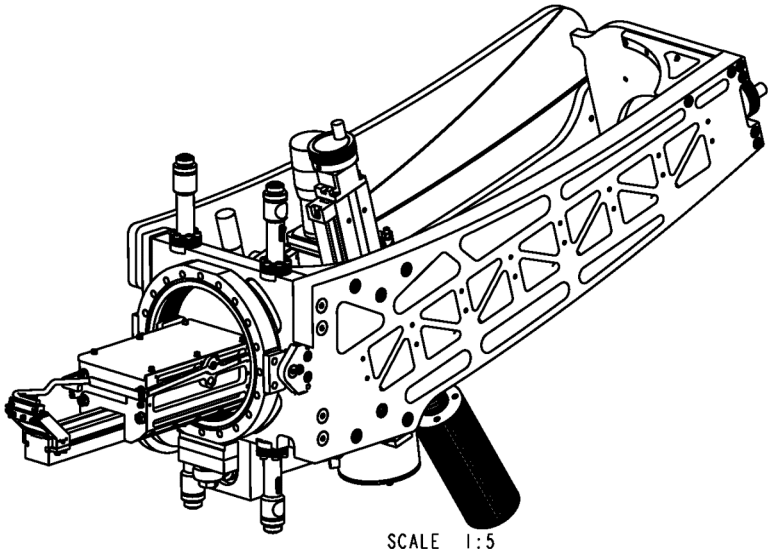

Figure 3
XES-350 A Schematic of the monochromator and detector is shown here. Figure provided by Scienta.

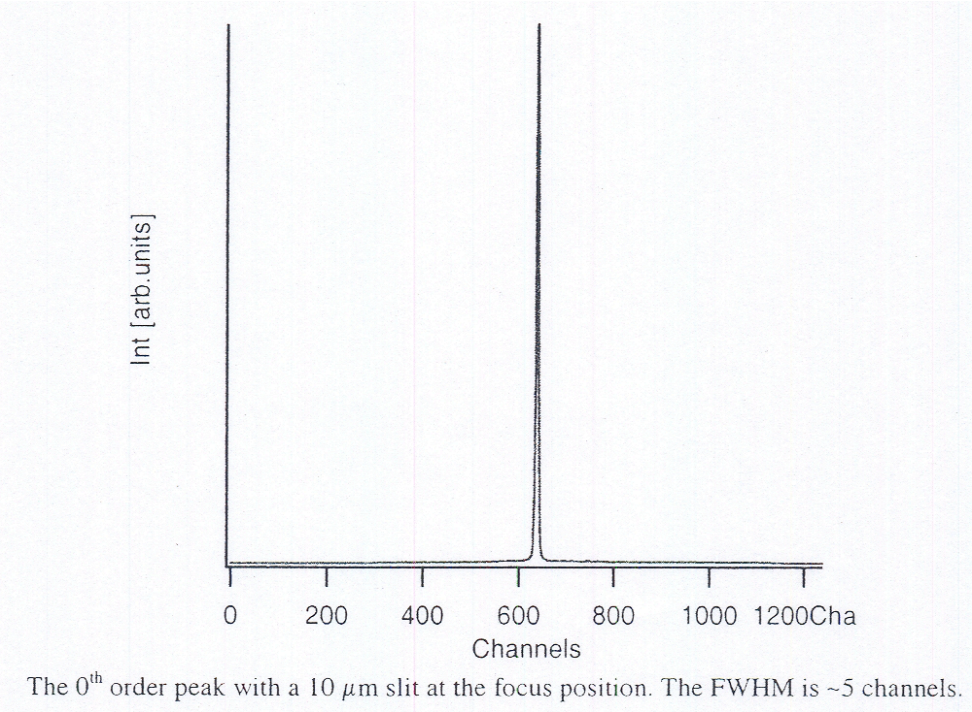

Figure 4

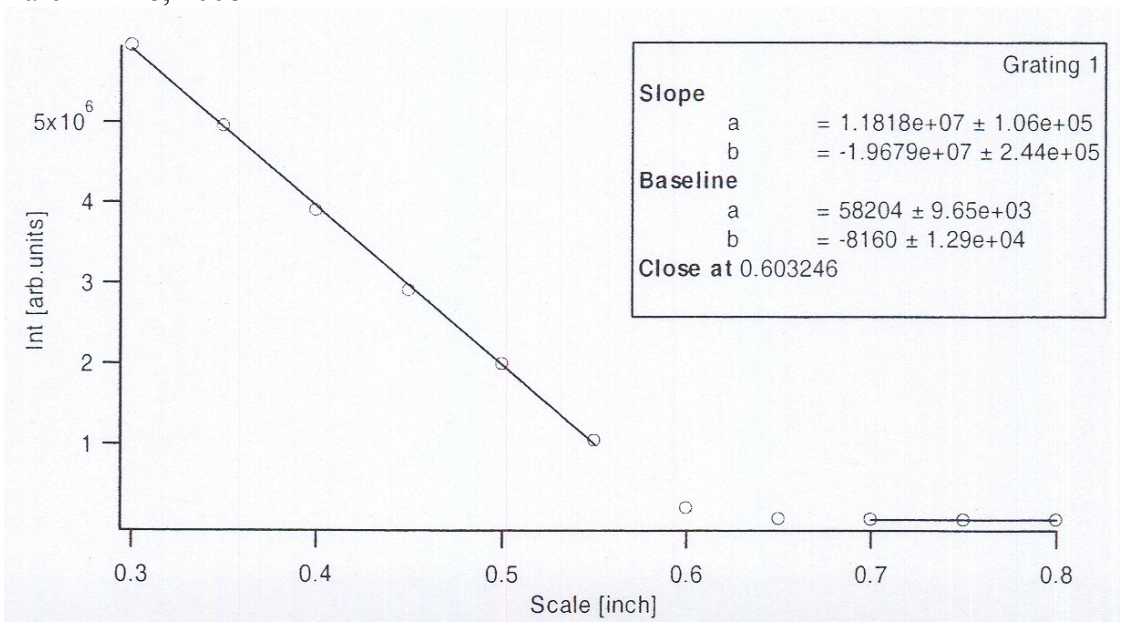

Figure 5
The intensity of the transmitted radiation as a function of the slit inch scale for Grating 1.

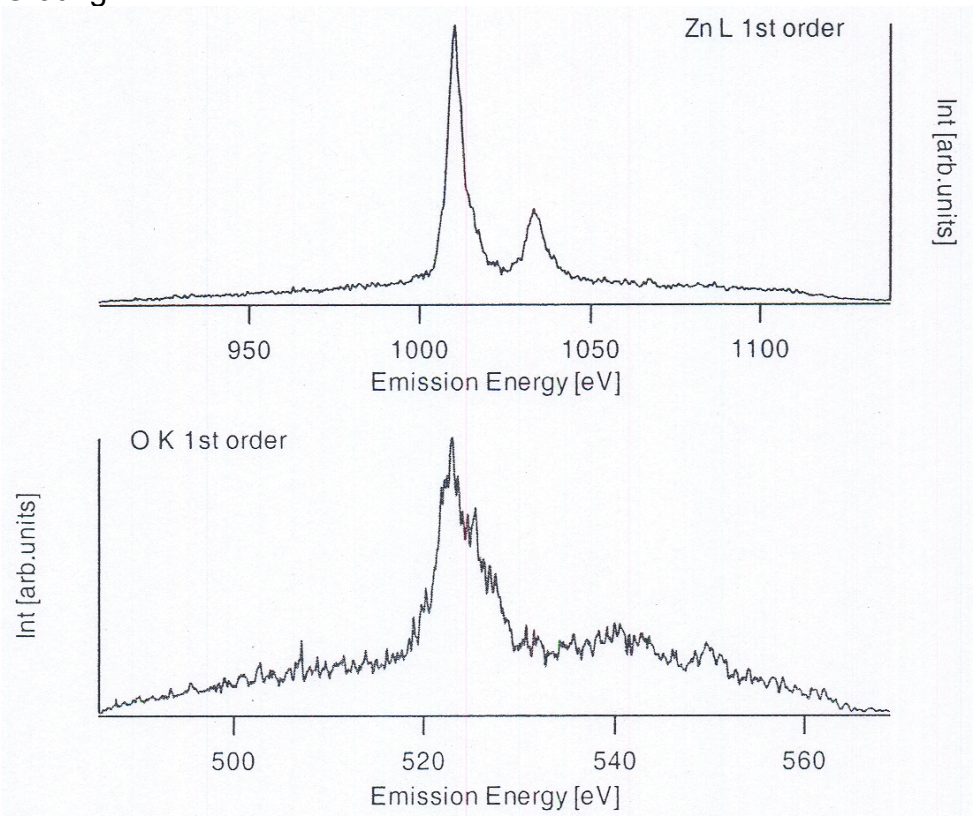

Figure 6
Zn L (2p) and O K (1s) emission in first order, measured at the calculated positions on Grating 1.

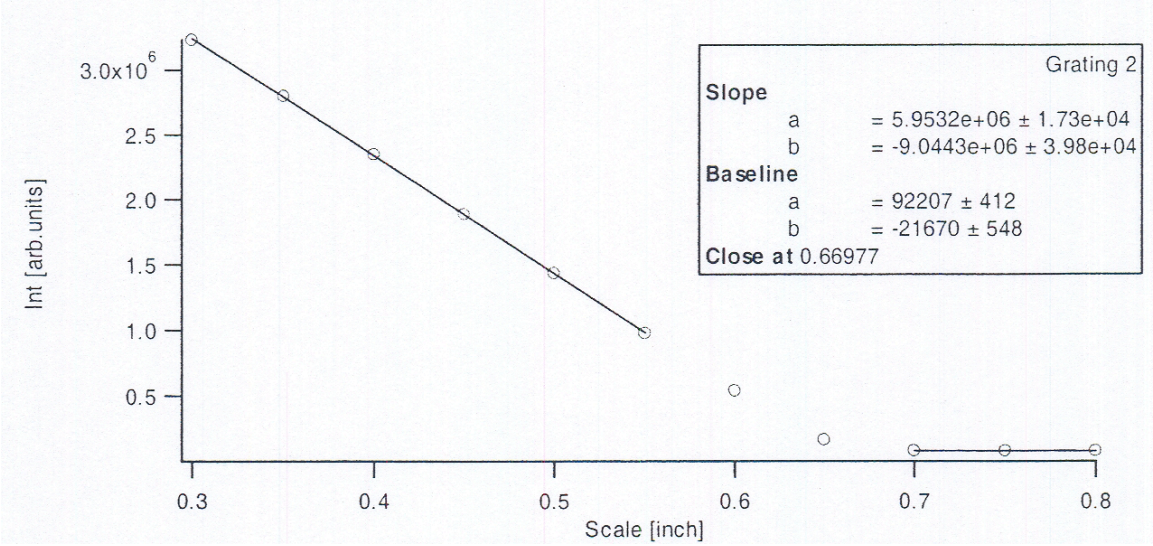

Figure 7
The intensity of the transmitted radiation as a function of the slit inch scale for Grating 2.

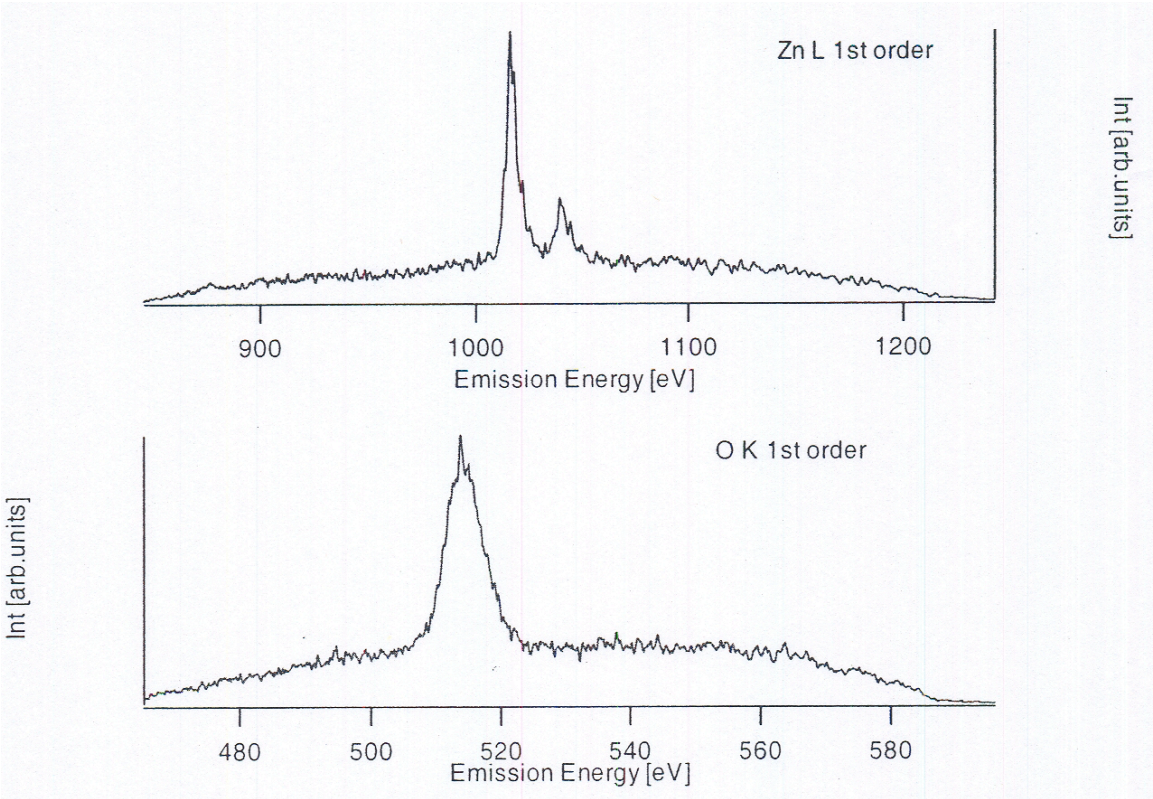

Figure 8
Zn L (2p) and O K (1s) emission in first order, measured at the calculated positions on Grating 2.

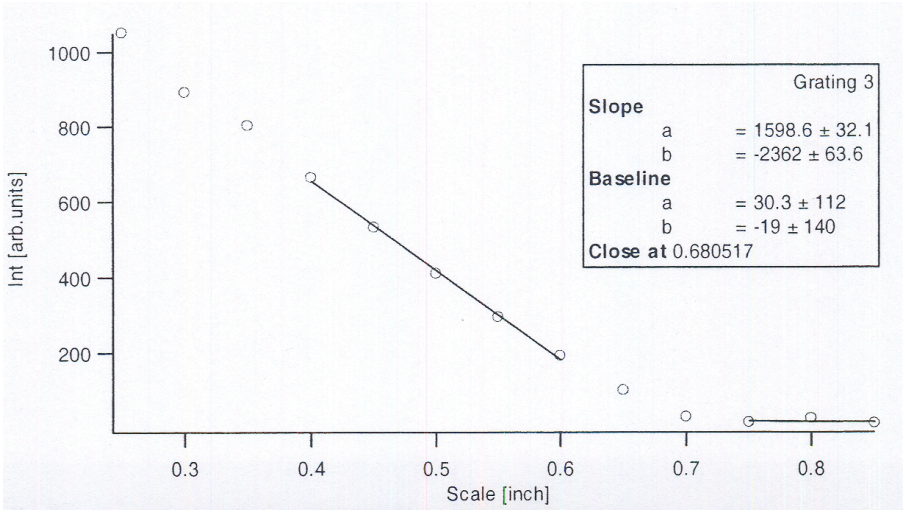

Figure 9
The intensity of the transmitted radiation as a function of the slit inch scale for Grating 3.

Acknowledgements:
Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract DE-AC52-07NA27344. Work that was performed by LLNL personnel was supported in part by the Office of Basic Energy Science at the U.S Department of Energy and Campaign 2 of WCI at LLNL. The X-ray emission data was collected at LLNL in collaboration with Gammadata Scienta personnel and M.T. Butterfield. Figures 4 - 9 and the corresponding captions were taken from “SAT XES 350,” a technical report from VG Scienta in 2006, provided under contract to LLNL and prepared by Marcus Agaker and Henrik Ohman.

References
1. J.G. Tobin et al, Phys. Rev. B **68**, 155109 (2003).
2. K.T. Moore et al , Phys. Rev. Lett. **90**, 196404 (2003).
3. G. van der Laan et al, Phys. Rev. Lett. **93**, 097401 (2004).
4. J.G. Tobin et al, Phys. Rev. B **72**, 085109 (2005).
5. 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,” Royal Society of Chemistry, ed. R. Alvarez, N.D. Bryan and I. May, page 773 (2006).
6. Y. Baer and J.K. Lang, Phys. Rev B **21**, 2060 (1980).
7. J.G. Tobin, S.W. Yu, T. Komesu, B.W. Chung, S.A. Morton and G.D. Waddill. MRS Symp. Proc. **986**, 63 (2007), Document # 0986-OO01-08.