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The Spectroscopic Signature of Aging in (delta)-Pu(Ga)

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The electronic structure of Pu is briefly discussed, with emphasis upon Aging effects.

Introduction: Pu Electronic Structure

Photoelectron Spectroscopy [1,2] and X-ray Absorption Spectroscopy [2-4] have contributed greatly to our improved understanding of Pu electronic structure. (See Figure 1.) 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 5.
3. The Pu 5f spin-orbit splitting effect dominates 5f itineracy.

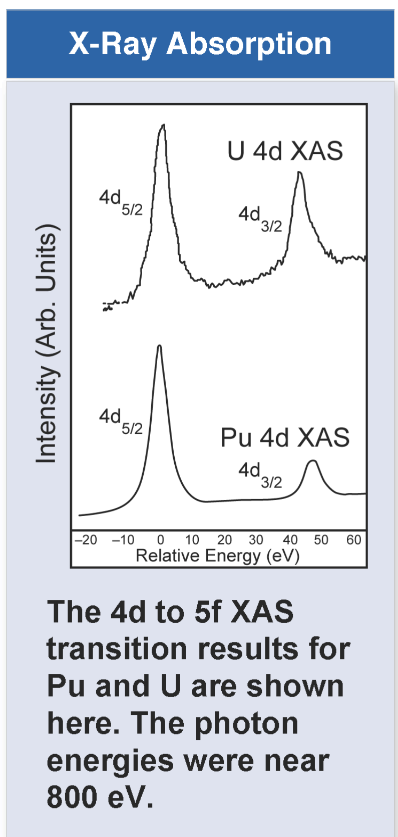

Figure 1. The X-ray absorption spectra (XAS) of alpha-U and alpha-Pu are above.

Past Method: Resonant Photoemission

Resonant Photoemission, a variant of Photoelectron Spectroscopy, has been demonstrated to have a sensitivity to aging in Pu samples. (See Figure 2.) The spectroscopic results have been correlated with resistivity measurements and shown to be the fingerprint of mesoscopic or nanoscale internal damage in the Pu physical structure. This means that a spectroscopic signature of internal damage due to aging in Pu has been established. [5]

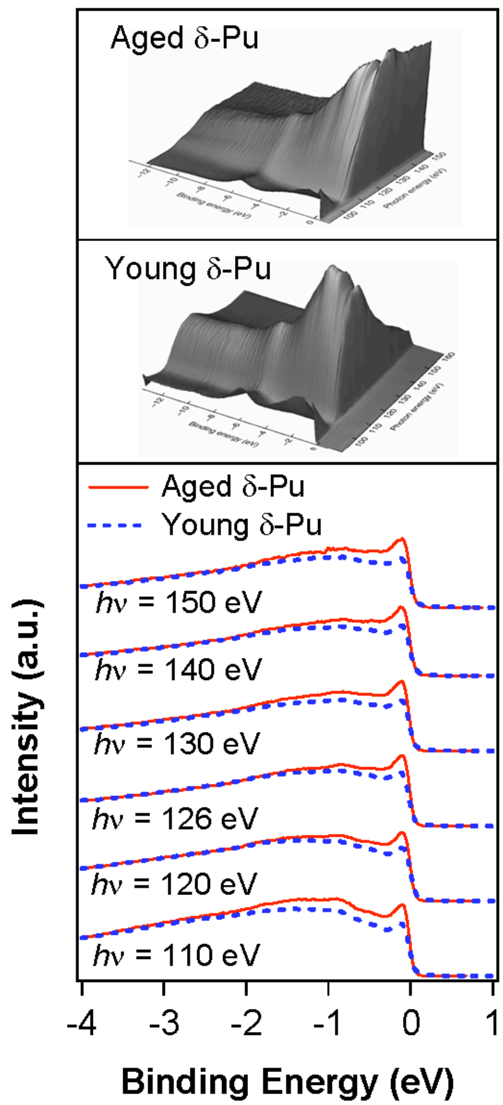

Figure 2. Resonant photoemission of young and aged delta-Pu.

Discussion: Present state

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). [6]

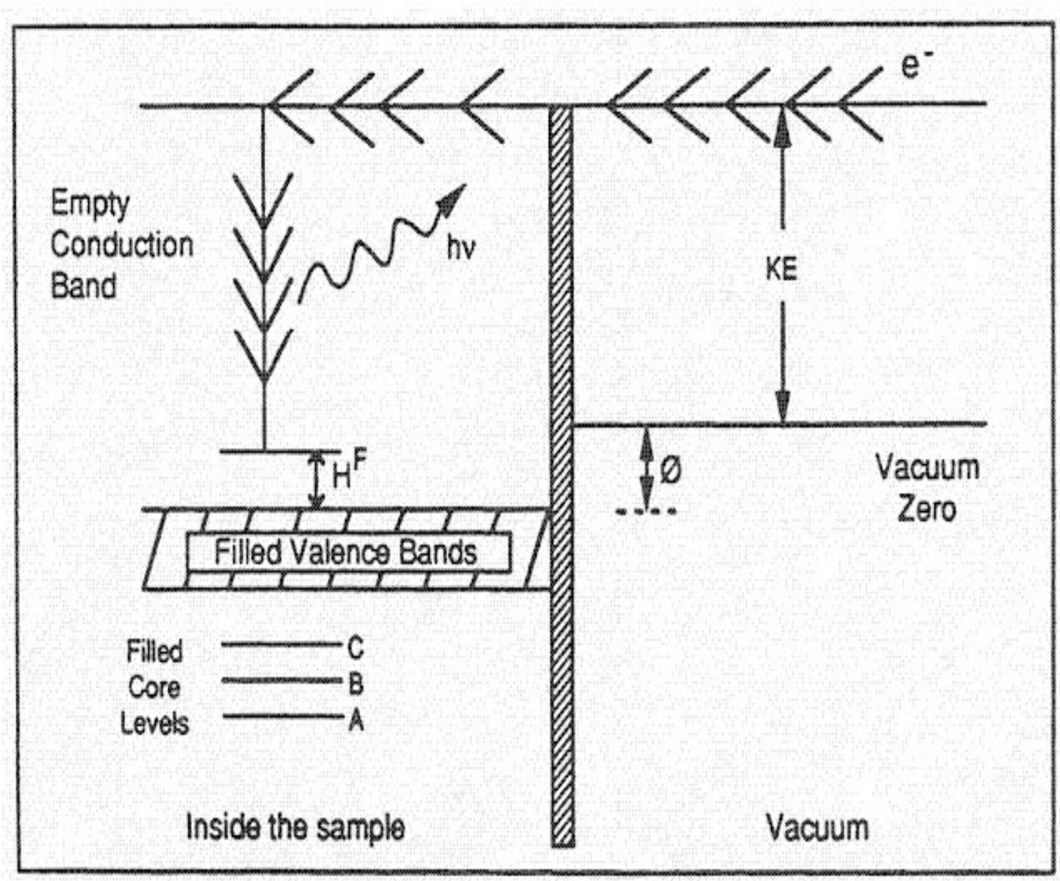

Figure 2.
Shown here is a schematic of the inverse photoelectron process. KE is kinetic energy of the incoming electron, θ is the work function, H^F is the energy of the state relative to the Fermi Level, and hv is the energy of the emitted electron. A defining characteristic of IPES/BIS is that hv ≈ KE.

Past BIS studies of Th and U indicate the feasibility and utility of Pu studies, [7] as confirmed by our simulations shown below. [6]

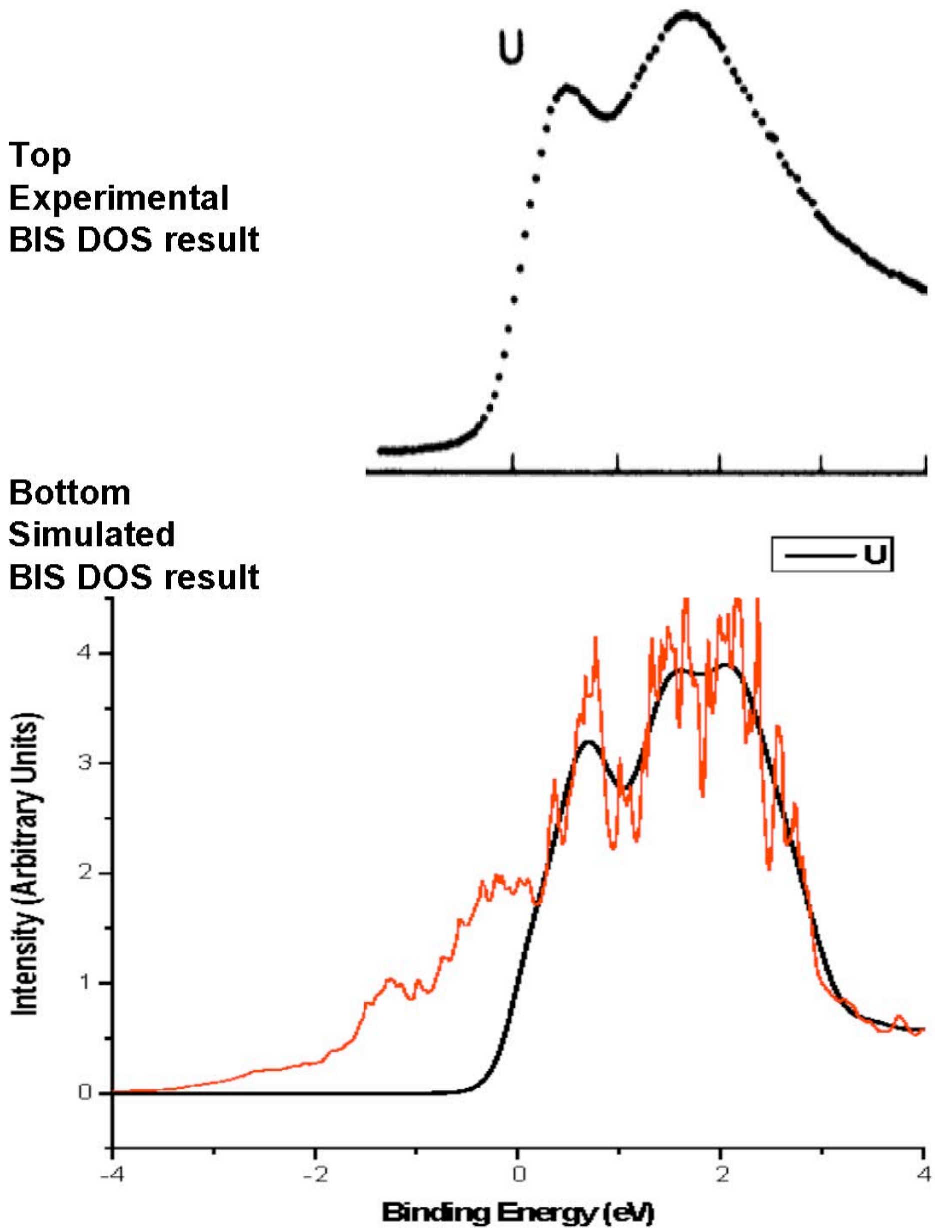

Figure 4. This is a comparison of an earlier BIS measurement by Baer and Lang [7] of Uranium with a simulated Density of States generated by starting with a calculation by Kutepov (in red), [4] which is the truncated at the Fermi Energy (only unoccupied states can contribute to BIS) and then smoothed to reflect broadening from the instrumental band-pass (in black).[8] Top: Experimental BIS result of Baer and Lang,. Bottom: Red: DOS calculations by A.L. Kutepov; Black: DOS calculation times inverse Fermi function, with some instrumental broadening. [8]

Conclusions: Future Directions

A new BIS capability has been developed in our laboratory. [6] Electron stimulated emission of photons has been carried out using the XES-350 monochromator and detector system.

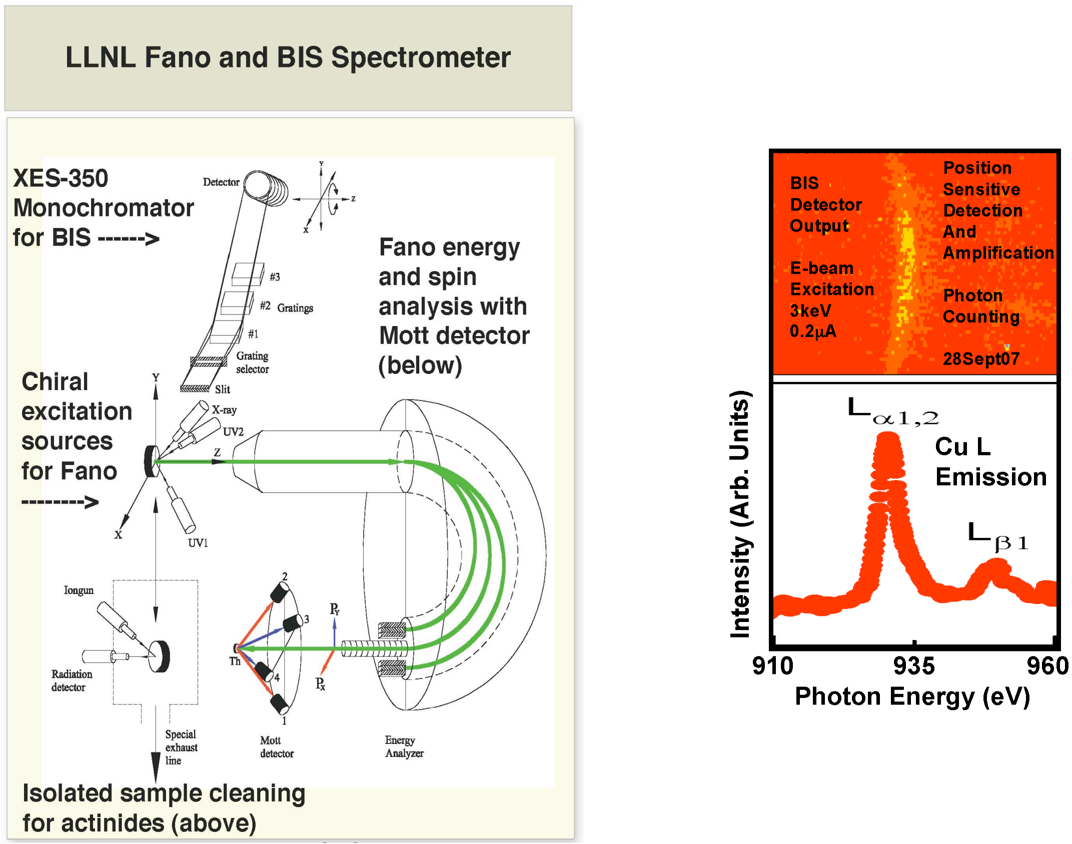

Figure 5.

Left: Sketch for BIS and spin resolved photoelectron spectroscopy (SRPES) experimental setup installed recently at Lawrence Livermore National Lab for the electronic structure study of actinides. For BIS, the detection of the photons is performed with the XES-350 monochromator and multi-channel detector. For SRPES, unpolarized light hits sample at an angle of 45 degrees with respect to the surface normal. The energies and the spins of the normally emitted photoelectrons are analyzed by hemispherical electron energy analyzer and Mott detector which has a thorium target operated at 25 keV with Sherman function of 0.16 $\pm$ 0.04, respectively. Two transversal spin components *PX* and *PY* can be measured in Mott detector simultaneously.

Right: X-ray Emission Spectroscopy (XES) of Cu using an electron excitation beam of E = 3000eV

Acknowledgements

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