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

The Utilization of Spin Polarized Photoelectron Spectroscopy as a Probe of Electron Correlation with an Ultimate Goal of Pu

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We are developing the technique of spin-polarized photoelectron spectroscopy as a probe of electron correlation with the ultimate goal of resolving the Pu electronic structure controversy. Over the last several years, we have demonstrated the utility of spin polarized photoelectron spectroscopy for determining the fine details of the electronic structure in complex systems such as those shown below.

The proposed half-metallic ferro-magnet Fe3O4 [Figure 1, Ref 2]

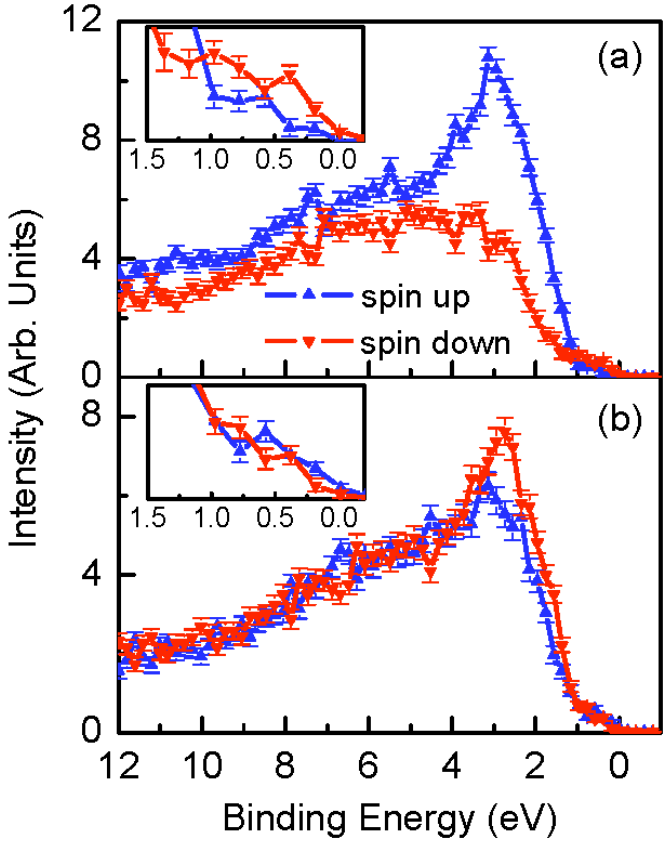

Figure 1
a) Spin resolved photoemission spectra from a) "as received" sample hν=160eV.
b) Spin resolved photoemission spectra from a sample after Ne+ sputtering, hν=160eV.
Note the large effect observed in the as received sample in (a) and the reduction of the effect in (b)

The 2p core levels of ultra-thin films such as Fe [Figure 2, Ref 3].

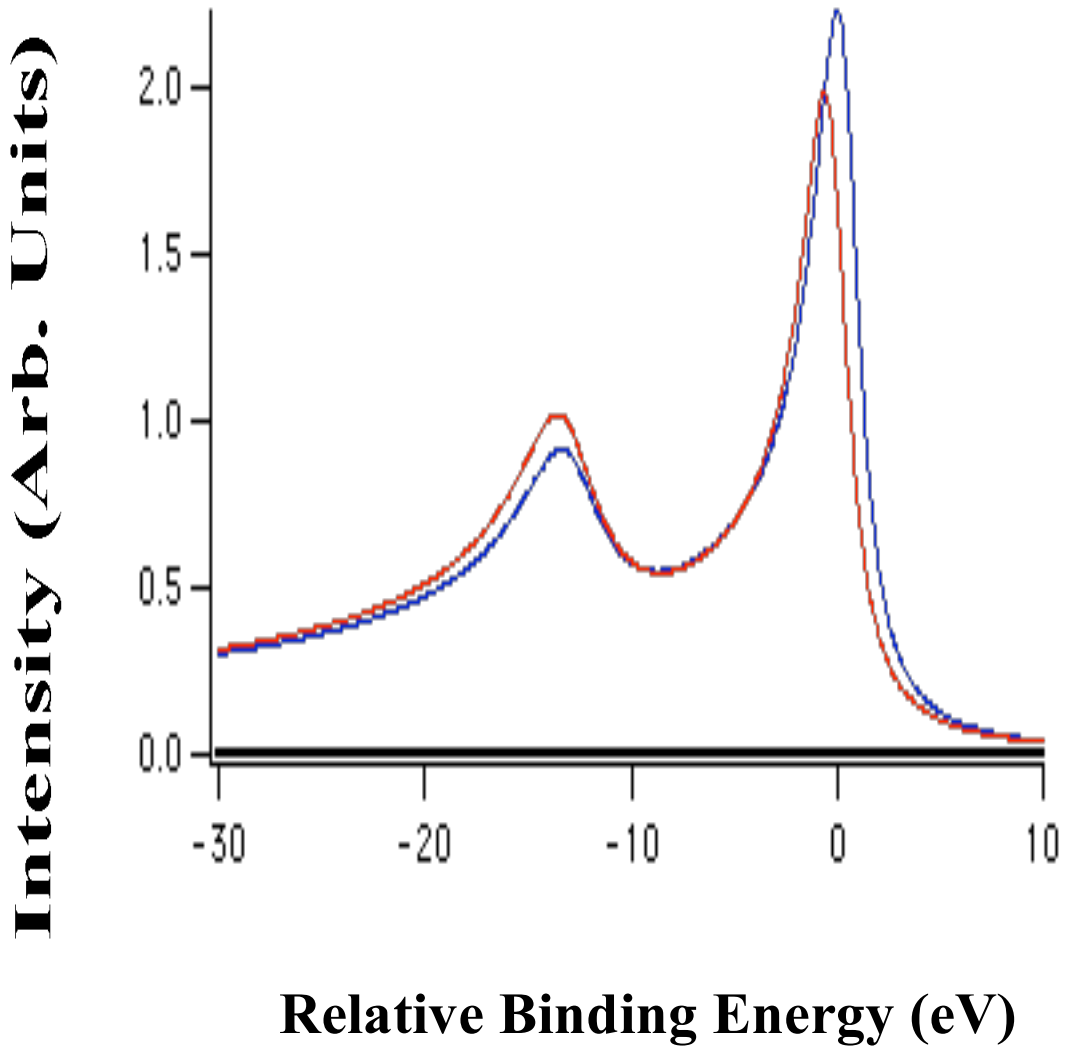

Figure 2
Simulated spin-resolved spectrum of the Fe 2p doublet, following Reference 3. Red is spin up and blue is spin down.

The valence states of Fe on compound semiconductors [Figure 3, Ref 4].

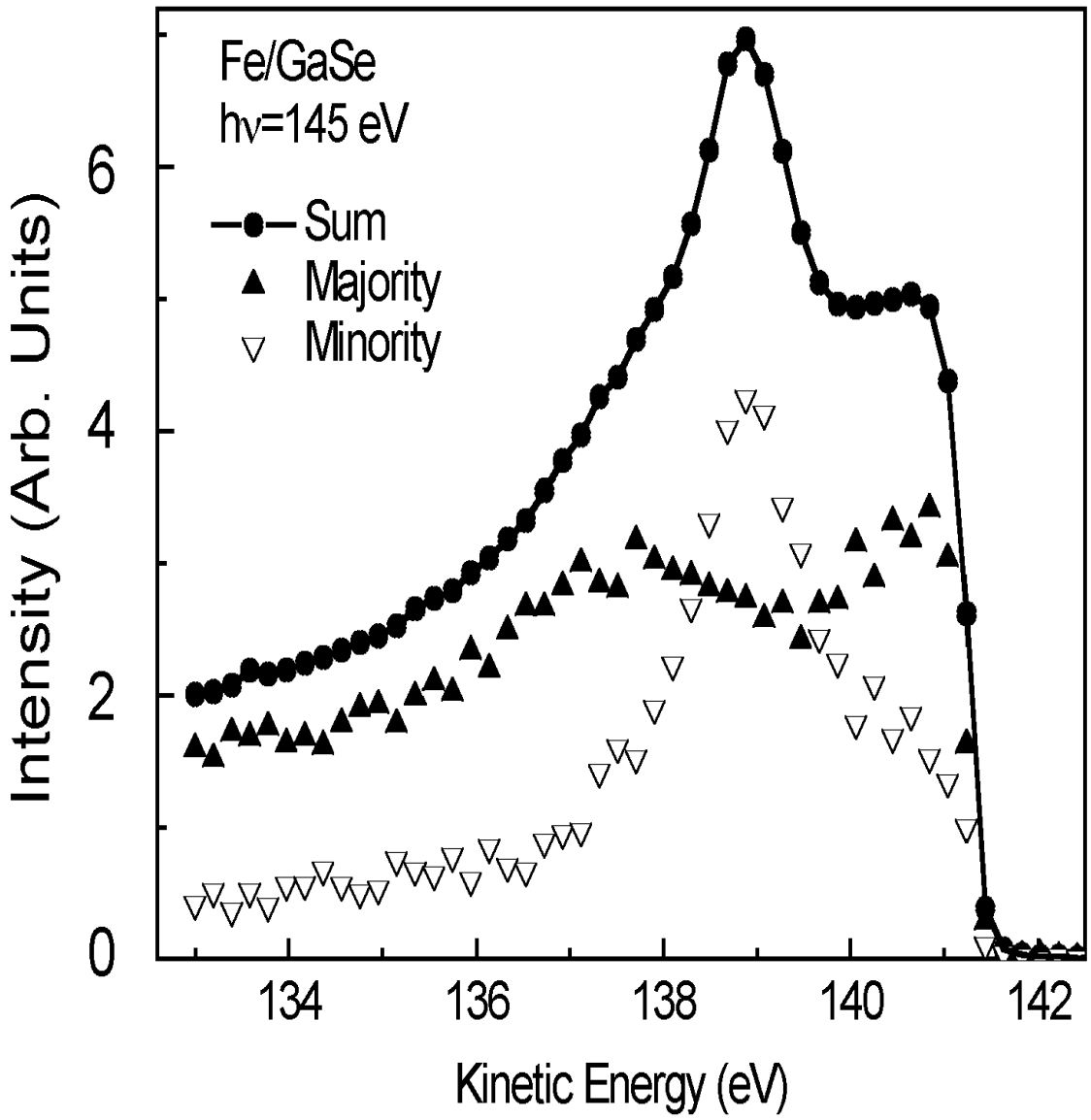

Figure 3
An example of some data collected on Beamline 7 at the ALS is shown here. The photon energy was 145 eV and the photon polarization is linear. The spin-resolved valence band photoemission results are from Fe overlayers, as part of a larger collaboration. [4] The Fermi edge is near 141 eV Kinetic Energy.

The non-magnetic system Ce [Figure 4, Ref 5-7].

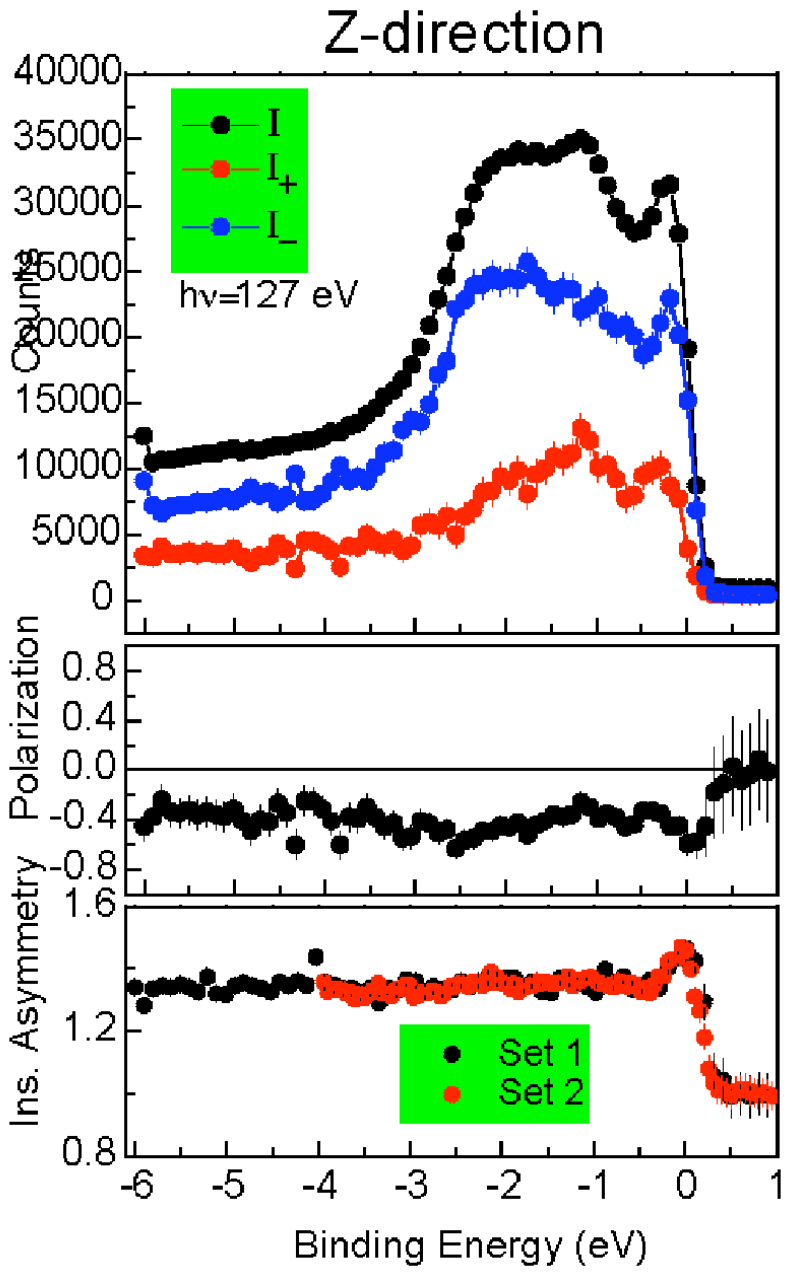

Figure 4
Spin resolved results for a polycrystalline film of Ce deposited upon W(110). The photon energy was 127 eV. Top panel: spin polarized spectra. Middle panel: polarization. Lowest panel: Instrumental asymmetry. Circularly polarized photons were used to generate the spin dependence.

The non-magnetic system Pt [Figure 5, Ref 8].

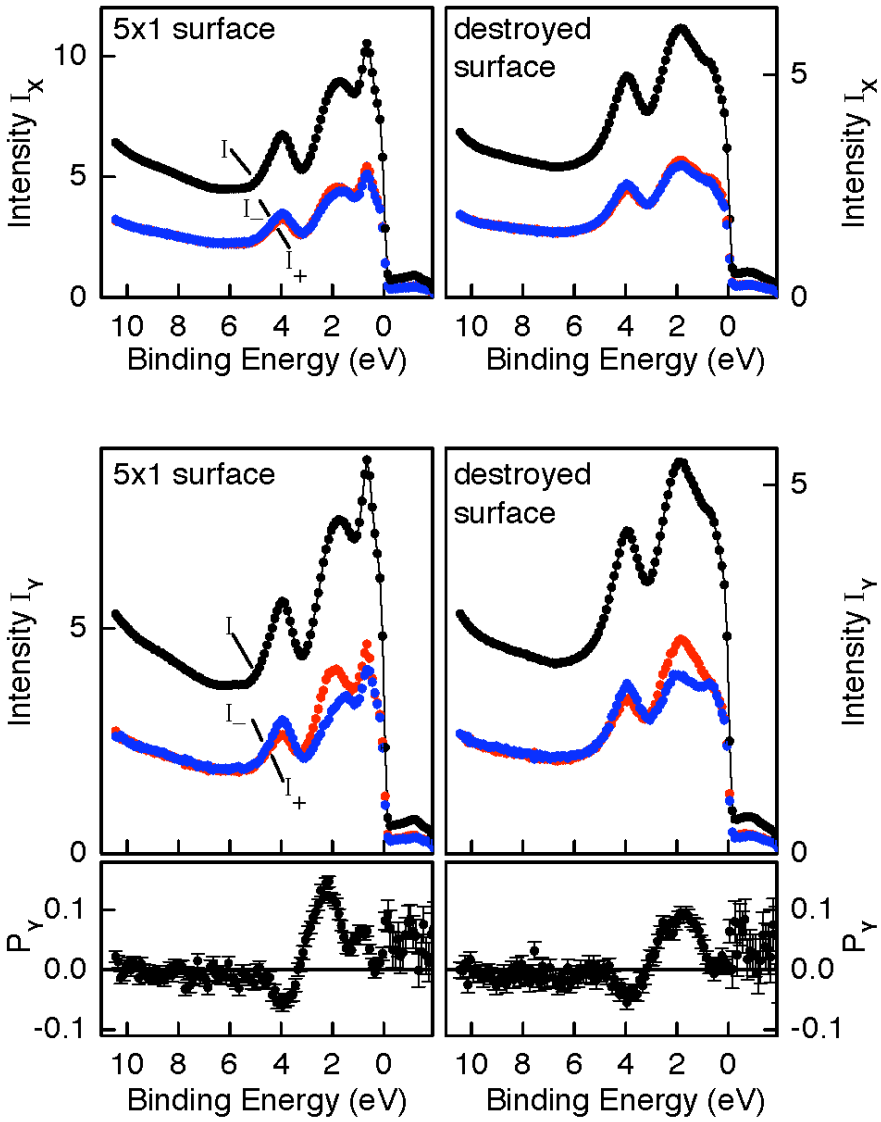

Figure 5
Spin-resolved photoemission spectra obtained with unpolarized He I(h*v* =21.2 eV) light for normal emission from the valence bands of Pt(001). Upper panel for P_X component: The spin integrated total intensity *I*(black) and spin separated partial intensities *I*+ in red and I_i in blue are shown for 5x1 surface and destroyed surface. Since the measured spin polarization P_X is zero, the partial intensities *I*+ and I_i are identical. Lower panel for P_Y component: The spin integrated total intensity *I*(black) and spin separated partial intensities *I*+ in red and I_i in blue are shown for 5x1 surface and destroyed surface. The measured spin polarizations P_Y for 5x1 surface and destroyed surface are shown. Note the loss of the 5x1 reconstructed surface state intensity at the binding energy of 0.66 eV with the destruction of the surface order for both *X* and *Y* components.

Various recent publications have addressed the ongoing question of the nature of the Pu electronic structure. [9,10] One possibility is the existence of a Kondo-like shielding of the 5f electrons. [1] We propose that spin resolved PES is the most promising approach to resolving this question. [7,11]

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 UMR personnel was supported in part by the Office of Basic Energy Science at the U.S Department of Energy. 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 Berkeley Lab contribution was done under DOE Contract DE-AC02-05CH11231.

- Present Address, Spring8 Synchrotron Radiation Facility, Japan

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