PROBING ACTINIDE ELECTRONIC STRUCTURE USING FLUORESCENCE AND MULTI-PHOTON IONIZATION SPECTROSCOPY

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High-level theoretical models of the electronic structures and properties of actinide compounds are being developed by several research groups. This is a challenging problem due to the need for explicit treatment of relativistic effects, and the circumstance that many of these molecules exist in states where the f and/or d orbitals are partially filled. Theoretical models must be tested and evaluated through comparisons with experimental results. Gas phase data are most suitable for this purpose, but there have been very few gas phase studies of actinide compounds. We are addressing this problem by carrying out spectroscopic studies of simple uranium and thorium compounds (oxides and halides). Multiple resonance and jet cooling techniques are being used to unravel the complex electronic spectra of these compounds. Zero kinetic energy - pulsed field ionization measurements are being used to examine the cations. Recent results for the oxides will be discussed. Systematic errors in the accepted values for the ionization energies have been discovered, and the patterns of electronic states for these molecules provide information concerning the occupation of the $5f$ orbitals and their participation in bond formation.