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Multiconfigurational nature of 5f orbitals in uranium and plutonium and their intermetallic compounds[1]

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The structural, electronic, and magnetic properties of U and Pu elements and intermetallics remain poorly understood despite decades of effort, and currently represent an important scientific frontier toward understanding matter. The last decade has seen great progress both due to the discovery of superconductivity in $PuCoGa_5$ and advances in theory that finally can explain fundamental ground state properties in elemental plutonium, such as the phonon dispersion curve, the non-magnetic ground state, and the volume difference between the α and δ phases. A new feature of the recent calculations is the presence not only of intermediate valence of the Pu 5f electrons, but of multiconfigurational ground states, where the different properties of the α and δ phases are primarily governed by the different relative weights of the $5f^4$, $5f^5$, and $5f^6$ electronic configurations. The usual method for measuring multiconfigurational states in the lanthanides is to measure the lanthanide L_{III}-edge x-ray absorption near-edge structure (XANES), a method that is severely limited for the actinides because the spectroscopic features are not well enough separated. Advances in resonant x-ray emission spectroscopy (RXES) have now allowed for spectra with sufficient resolution to resolve individual resonances associated with the various actinide valence states. Utilizing a new spectrometer at the Stanford Synchrotron Radiation Lightsource (SSRL), RXES data have been collected that show, for the first time, spectroscopic signatures of each of these configurations and their relative changes in various uranium and plutonium intermetallic compounds. In combination with conventional XANES spectra on related compounds, these data indicate such states may be ubiquitous in uranium and plutonium intermetallics, providing a new framework toward understanding properties ranging from heavy fermion behavior, superconductivity, and intermediate valence to mechanical and fundamental bonding behavior in these materials.

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