

Electronic Structure of Actinide Oxides

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An integrated approach to solving long-standing problems in the actinide oxide Mott insulators has yielded new insights into the electronic structure of these interesting materials. Our approach uses a new techniques in sample growth and electronic structure calculations combined with angle-resolved photoelectron spectroscopy (ARPES) measurements. We use single crystal, epitaxial films grown by the polymer-assisted deposition technique to access actinide oxide phases not readily available through standard bulk growth techniques.[1] We also take advantage of recent developments in Mott insulator calculations which use the hybrid functional approach to achieve very good agreement between calculations and experiments. [2, 3]

We have measured the electronic structure of thin film, single crystal actinide oxides including UO₂, U₃O₈ and ThO₂ by means of ARPES. The uranium ARPES data show two regions of interest in the valence band. The deeper binding energy region extends from 4 eV to 8 eV and is associated with the O 2p level. Strong dispersion in the normal emission data indicates the extensive role of the 2p levels in UO₂ bonding as well as details of the bulk electronic structure. The peak closer to the Fermi level (1.7 eV binding energy) is of predominant 5f character and shows less dispersion but does have a dispersive component not previously reported. The thin film results compare favorably with ARPES results reported for bulk single crystal UO₂ results.[4]

The dispersion in the 5f peaks indicates hybridization between the 5f electrons and the surrounding conduction states as the actinide-actinide separation distance in the actinide oxides exceeds any overlap between neighboring 5f radial wavefunctions. A comparison between these recent photoemission results with the new hybrid functional computational effort shows generally good agreement.[2] With both resonance ARPES and a broad survey of photon energy dependent data, we can make assessments of the degree of covalency in the actinide oxide electronic structure. With the detailed electronic structure information on UO₂ we can make reasonable speculations on bonding and hybridization trends across the light actinide oxide series.

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References:

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