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Photoemission Studies at the Advanced Light Source Shed Light on Plutonium Phase Characteristics

The physical characteristics of any given material are largely derived from the behavior of its valence electrons. Valence electrons are the lowest-energy electrons in a material and are responsible for the formation of chemical bonds. Typically, in a metal, electrons are either localized around a particular atom or are delocalized (i.e., shared by all the atoms in the crystal) throughout the entire metal. The actinide series is interesting because as the atomic number increases across the series, the electrons in the actinide metals transit from delocalized 5f electrons (Ac–Pu) to localized 5f electrons (Pu–No). Plutonium (element 94) is located right at this transition. This placement in the series leads to plutonium metal being one of the most complex materials known. Metallic plutonium displays six allotropic phases (α , β , γ , δ , δ' , and ϵ) at standard pressure. A 20% volume expansion occurs during the change from the α phase to the δ phase. These physical properties have been attributed to the 5f valence electrons changing from delocalized states to localized states as the crystal structure changes from the α to the δ phase.

Soft x-ray techniques (photon energy in the range of 10–1000 eV) such as photoelectron; x-ray emission; and near-edge x-ray absorption spectroscopies have been used to determine the electronic structure of many (in fact most) materials. However, these techniques have not been fully utilized on the actinides. The safety issues involved in handling the actinides make it necessary to minimize the amount of radioactive materials used in the measurements. To our knowledge, the only synchrotron radiation source in the world where soft x-ray measurements have been performed on plutonium is the Spectromicroscopy Facility at Beam Line 7.0.1 at the Advanced Light Source (ALS). The ALS is the premier synchrotron radiation facility in the world for the study of the electronic structure of materials using soft x-ray photons. The Spectromicroscopy Facility was first used for resonant photoemission measurements on plutonium oxide in 1993. Our first measurements on plutonium were conducted in the fall of 1998.

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