Electronic and Geometric Structure of Pu Metal: A High-Resolution Photoelectron Spectromicroscopy Study

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Introduction

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 make a transition 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 a 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 phase 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 Spectromicroscopy Facility is designed so that measurements can be made on small quantities of hazardous material. This facility has a photon flux of 10^{13} photon/sec at a photon energy of 100 eV with 0.01 eV resolution. The high photon flux allows one to focus the beam down to a size of 50 microns and still have enough light intensity at the sample for measurements to be conducted in a reasonable time frame 1–10 minutes per spectrum. Therefore, the sample size can be on the order of 100 microns in diameter. This greatly minimizes the amount of plutonium on site during the experiment.

Results

We performed core-level photoemission, valence band photoemission, and near-edge x-ray absorption spectroscopy on both polycrystalline α-plutonium and δ-plutonium microcrystals. Only the photoemission experiments will be described here. Photoelectron spectroscopy is predicated upon the photoelectric effect first described by Einstein in 1905. An incident photon is absorbed by an atom in the solid, and an electron is ejected. An electron energy analyzer is used to measure the direction and kinetic energy of the emitted electron. The kinetic energy of the photoelectron is directly related to its binding energy in the solid.

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