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Photoelectron Spectroscopy of Plutonium at the Advanced Light Source

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We are developing a program to perform Photoelectron Spectroscopy and X-Ray Absorption Spectroscopy upon highly radioactive samples, particularly Plutonium, at the Advanced Light Source in Berkeley, CA, USA. First results from alpha and delta Plutonium are reported as well as plans for a dedicated spectrometer for actinide studies.

KEYWORDS: ***Synchrotron radiation, photoelectron emission, x-ray absorption, Plutonium***

I. Introduction

Photoelectron Spectroscopy and X-Ray Absorption are being used to investigate the electronic structure of alpha and delta Pu. [It is generally believed that alpha is more free electron like and delta is possibly a correlated electronic system, although this has yet to be unequivocably proven and the details of which remain clouded.] Our preliminary results[1], where Resonant Photoemission was used to probe large grain polycrystalline delta and polycrystalline alpha, have lead us to modify our initial plans. For example, during the last year we have embarked upon a building project, developing a dedicated Pu Spectrometer at the Advanced Light Source. One result of our first studies is that we believe that minimization of sample oxidation is a key to successful experimentation and we are pursuing that vigorously. Additionally, it now appears that a new experiment, based upon "Double Polarization," may be the key to differentiating between the several models now being proposed to explain the electronic structures of alpha and delta Pu. Here the combination of a chiral xray environment and true spin detection will allow us to test whether spin-orbit, exchange, coulombic repulsion or other multielectronic effects drive the differences between alpha and delta Pu.

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II. Experimental

The first experiments were performed at the Spectromicroscopy Facility (Beamline 7.0) at the Advanced Light Source in Berkeley, CA[2]. The Pu samples were taken from a specially purified batch of Pu metal. The plutonium was zone refined and vacuum distilled while magnetically levitated[3] . The product of the purification process was α-Pu containing a total of 170 ppm impurities. A portion of the refined metal was alloyed with gallium to form the δ-phase (fcc symmetry). The sample surfaces were prepared by repeated room-temperature, sputter-annealing cycles to minimize the amount of oxygen and other impurities dissolved in the sample or at grain boundaries, in a specially designed chamber attached to the sample introduction and analysis systems on Beamline 7.0. The transfer, preparation, and analysis chambers ensured that the Pu metal samples did not experience pressures greater than 10^{-8} torr. This minimized any surface contaminants that could adversely effect the soft x-ray measurements.

III. Discussion

Using the tunabilty of synchrotron radiation, it is possible to perform many variants of photoelectron spectroscopy and x-ray absorption, including accessing the core levels of the sample constituents. One of the variants that was pursued was Resonant Photoemission.[4] Photoelectron spectroscopy is a "photon in, electron out" process. Often, it can be simplified down to a single electron phenomenon, where the energy of the photon is absorbed and transferred over entirely to a single electron, while all other "spectator" electrons essentially remain frozen. An advantage of this is its simplicity of interpretation. But in many