Resonant photoemission in *f*-electron systems: Pu and Gd

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(Received 13 March 2003; revised manuscript received 30 June 2003; published 14 October 2003)

Resonant photoemission in the Pu $5f$ and Pu $6p$ states is compared to that in the Gd $4f$ and Gd $5p$ states. Spectral simulations, based upon an atomic model with angular momentum coupling, are compared to the Gd and Pu results. Additional spectroscopic measurements of Pu, including core level photoemission and x-ray absorption, are also presented.

DOI: 10.1103/PhysRevB.68.155109 PACS number(s): 71.20.Gj, 71.27.+a, 79.60.−i

I. INTRODUCTION

While chemically toxic and highly radioactive, Pu may be the most scientifically interesting element in the Periodic Table. It's properties include the following,[1] six different phases, close to each other in energy and sensitive to variations of temperature, pressure and chemistry; the face-centered-cubic phase (delta) is the *least* dense; Pu expands when it solidifies from the melt; and it is clearly the nexus of the actinide binary phase diagram. In a sense, it is the boundary between the light (delocalized $5f$ electrons) and heavy (localized or correlated $5f$ electrons) actinide elements, but this is an oversimplification. The localized atomic $5f$ states are naturally correlated, but important regimes of correlated electron states are conceivable as extended states on the delocalized side of the possible Mott transition. The proximity to this crossover may be the driving force behind these all exotic properties. Pu remains of immense technological importance and the advancement to a firm, scientific understanding of Pu and its compounds, mixtures, alloys and solutions is a crucial issue.[1,2]

As a result, computationally modeling Pu is a challenge. There have been substantial efforts made over the years.[3–5] Some of the more recent work includes pursuit of the structural parameters of alpha Pu from first principal calculations,[6] the electronic and phonon properties of the six phases,[7] the alloy electronic structure,[8] and the effects of localization in the $5f$ states.[9] Of particular interest is the dynamical mean field theory approach[10,11] and the models that predict the possibility of an internal magnetic ordering in the electronic structure.[12,13]

Furthermore, the development of computationally based predictive capabilities hinges upon experimental benchmarking of the variously proposed models. Perhaps the most direct method for testing models of electronic structure is photoelectron spectroscopy. In the recent past, there have been a number of photoelectron spectroscopy studies of Pu, using laboratory sources such as He I (21.22 eV) and He II (40.8 eV). For example, Gouder *et al*. have performed an investigation of $5f$ localization in the PuSe and PuSb.[14,15] Gouder *et al*.[16–18] and Havela *et al*.[19] have also used Pu thin film growth as a means to test for localization in the Pu $5f$ levels. At Los Alamos, a tunable laser plasma source[9,20–22] has been developed by Arko and Joyce, in order to supplement the studies performed with gas discharge lamps.[9] Joyce *et al*.[22] used the laser plasma source to perform a preliminary study of the $5d$-$5f$ resonant photoemission in Pu, but their work was limited by the poor signal to noise ratio of the plasma generated spectra. Earlier x-ray photoelectron spectroscopy included studies of alpha-Pu and delta-Pu by Cox *et al*.[23,24] plutonium metal by Baptist *et al*.,[25] and plutonium oxidation by Couteix *et al*.,[26] Veal *et al*.,[27] and Larson.[28]

This synchrotron-radiation-based study is another important step in such an experimental benchmarking. A summary of our resonant photoemission (RESPES) results for Pu is shown in Fig. 1. By comparing the Pu $5f$ and Pu $6p$ results to a RESPES investigation of the $4f$ system Gd, greater insight will be gained concerning the behavior of $5f$ electrons and Pu. Finally, the results of an atomic model calculation, for both Gd and Pu, will be presented.

II. EXPERIMENTAL AND COMPUTATIONAL DETAILS

The experiments were carried out at the SpectroMicroscopy Facility (Beamline 7.0) (Ref. 29) at the Advanced Light Source in Berkeley, CA, USA. In the photoelectron (x-ray absorption) measurements, the photoelectron (sample current) intensity was normalized to the photon flux by a Au

