An LAPW Study of d - Plutonium and the (001) Surface

A. K RAY, X. WU, The University of Texas at Arlington — The electronic structure properties of bulk fcc δ-Plutonium and the quantum size effects in the surface energies and the work functions of the (001) ultra thin films (UTF) up to 7 layers have been investigated with periodic density functional theory calculations within the full-potential linearized augmented-plane wave approach. Several levels of theory, namely NSP-NSO, NSP-SO, SP-NSO, and SP-SO, have been examined and our calculated equilibrium atomic volume of 178.3 a.u. and bulk modulus of 24.9 GPa at the fully relativistic level of theory are in good agreement with experimental results. The energy difference brought by spin-orbit coupling, about 7-8 eV, is dominant, but the energy difference brought by spin-polarization, from a few tenths to 2 eV, has a stronger dependence on the atomic volume. Density of states show that 5f electrons are more itinerant when the volume of δ-plutonium is compressed and they are more localized when the volume is expanded. The surface energy converges rapidly and the semi-infinite surface energy is predicted to be 0.692 eV. Quantum size effects for the work function is not pronounced for (001) surface. Work supported by the Department of Energy (Grant No. DE-FG02-03ER15409) and the Welch Foundation (Grant No. Y-1525). 1P. Blaha, K. Schwarz, G. K. H. Madsen, D. Kvasnicka and J. Luitz, WIEN2k (Technische Universitat Wien, Austria, 2001)