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Self-Interaction Corrected Energies and the Solid-Solution-Like Partial 5f Localization in Expanded-Volume fcc Plutonium

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With increasing temperature, cerium and plutonium have phase transitions into expanded-volume fcc phases. For plutonium there is an overall expansion in atomic volume of 24% from the monoclinic ground state. This fcc phase has many unusual properties, including negative thermal expansion. On the basis of physics contained in a correlated-electron model, we have previously proposed that this phase transition is the result of an entropy-of-disorder-driven transition into an fcc phase which has a time varying homogenously disordered distribution of equal numbers of long-time-scale (stable) configuration and short-time-scale (fluctuating) interconfigurational sites. By a combination of LDA and (self-interaction-corrected) SIC-LDA LMTO electronic structure calculations, we have now successfully predicted a delocalized $5f$, i.e. bonding, monoclinic ground state and a partially disordered fcc excited state, and fcc density only 2.6% greater than experiment. Furthermore, on considering the entropy-generating process provided by our homogenously disordered model, the SIC-LDA calculations correctly predict a qualitatively excellent upper bound of the experimental transition temperature. The SIC-LDA fcc results indicate an unusual electronic structure having the lowest total energy with 4 out of a total of 5, $5f$ electrons being localized, and a nearby lying higher energy state with 3 localized $5f$ electrons. This provides an explanation for the negative thermal expansion.