Site-dependent correlation strength and occupation number of 5f electrons in alpha phase plutonium metal

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August 26, 2022

Abstract

In order to quantify the site-dependent correlation strengths in terms of the quasi-particle weights and the occupation numbers of 5f electrons in alpha phase plutonium metal with eight crystallographically nonequivalent atomic sites Pun (n=1˜8), we perform a first principles calculation by using a many-body method merging density functional theory (DFT) with dynamical mean-field theory (DMFT) plus the relativistic and correlation effects. The quasi-particle weight, the electronic spectrum function, the hybridization function, as well as the occupation number of Pu 5f electrons all suggest that Pu1 and Pu8 atoms have the most itinerant and the most localized 5f electrons, respectively, while the other atoms Pun (n=2˜7) exhibit an intermediate correlation strength. The quasi-particle weights demonstrate that only the jj or intermediate coupling scheme is more appropriate for Pu 5f electrons in comparison with the Russel-Saunders (LS) scheme. The electronic spectrum function and the occupation analysis establish that Pu 5f electrons simultaneously have dual itinerant and localized regimes with average 5f occupation numbers of nf between 4.8994 and 4.9628, which are mainly composed of 5f4, 5f5 and 5f6 configurations, irrespective of different atomic sites. Finally, we also estimate the so-called quasi-particle band structure to directly compare with an experimental angle-resolved photoemission spectrum (ARPES).

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