Electronic structure theory of Pu-based alloys and compounds: Pu-Am, Pu-Ce-alloys and $PuCoGa_5$

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In the present work, we study the electron correlation effects in the electronic structure and spectra of Pu-Am, Pu-Ce alloys and $PuCoGa_5$. We focus on comparison between the theory and available experimental results for valence-band photoelectron spectra (PES), as well as XAS and EELS. We make use of the "local density matrix" approximation (LDMA) to DMFT [1], that combines the Hubbard-I approximation with the full-potential linearized augmented plane wave (FP-LAPW) method , including self-consistency over the charge density. This implementation is all-electron, includes spin-orbit interaction, and makes no shape approximations for the charge density.

We consider the δ-Pu alloys, and study the electronic structure of ordered Pu_3Ce and Pu_3Am in the same *fcc* supercell. The Coulomb parameter U_{Ce} = 6.1 eV, exchange J_{Ce} = 0.7 eV, U_{Am} = 4.5 eV, exchange J_{Am} = 0.67 eV, and U_{Pu} = 4.5 eV, J_{Pu} = 0.64 eV were used. The experimental lattice parameter of Pu_3Am (473.3 pm) was used in the calculations.

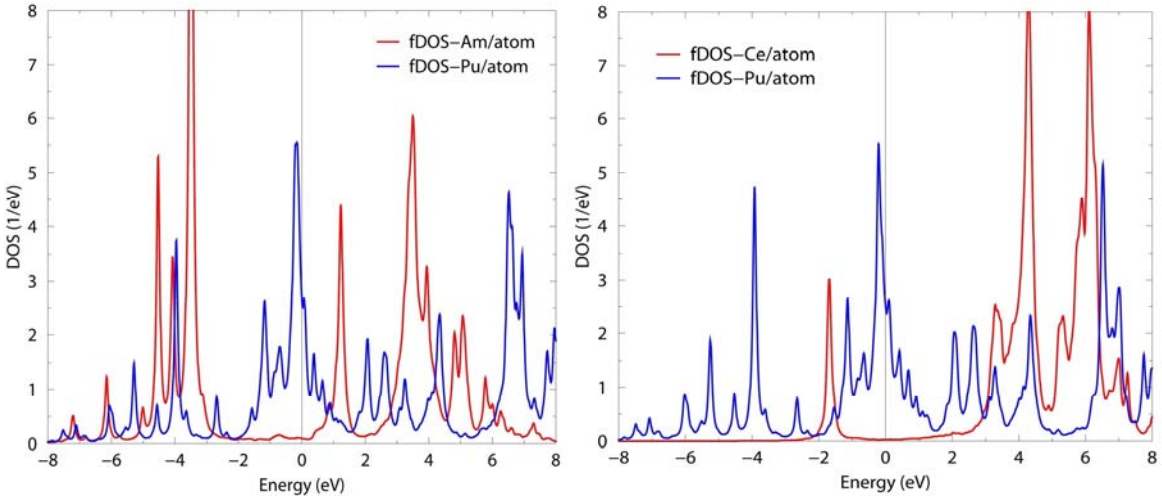

Fig.1. Element-specific *f*-projected densities of states (fDOS) calculated for Pu-Am and Pu-Ce

We plot in Fig.1 the total and *f*-projected spectral densities for Pu and Am atoms in Pu_3Am, and Pu and Ce in Pu_3Ce alloys. The Pu-atom fDOS is very similar to the fDOS for δ-Pu , in agreement with valence-band PE spectra [2]. We conclude that alloying with Ce and Am does not induce any significant changes in the Pu electronic structure. The Am-atom fDOS is very similar to the fDOS for pure Am. Calculated PE spectrum of PuAm-alloy corresponds to the weighted average of spectra of pure Pu and Am, in agreement with experiment . The Ce-atom fDOS resembles the positions of lower and upper Hubbard bands in the experimental photoemission and inverse photoemission data for α-Ce [3], but fails to reproduce the quasi-

particle peak at the Fermi level. This is due to the use of HIA which does not accurately include hybridization between *f* and non-*f* states.
The Sommerfeld γ-coefficient was calculated. The results are included in Tab.I. There is only a very small contribution to γ coming from Ce (as well as Am) atoms, and the γ–values are mostly due to the Pu-*f* states. They are a bit smaller than the γ–value of 30.1 mJ/mol K^2 for δ-Pu obtained from the LDA+HIA calculations of Ref. [1]. This slight decrease is evidently caused by an increase of volume with alloying. Note that LDA+HIA yields somewhat higher γ-value than 20.4 mJ/mol K^2 obtained from LDA+DMFT(QMC) [4]. The theoretical γ–values explain a large part of the experimental γ–value per mol Pu of approx. 40 mJ/mol K^2.

	Pu-n_{5f}	γ_{Pu} (mJ/mol K^2)	Ce/Am-n_f	$\gamma_{Ce/Am}$ (mJ/mol K^2)
Pu_3Ce	5.24	24.0	0.76	0.21
Pu_3Am	5.24	26.0	6.00	0.41

Table I.: The *f*-shell occupations and *f*-shell contributions into the Sommerfeld coefficient γ in Pu_3Ce and Pu_3Am alloys resulting from the charge density self-consistent LDA+HIA.

Finally we discuss the electronic structure of $PuCoGa_5$. There is fairly good agreement between calculated PE spectra shown in Fig. 2 and experimental results of [5] for the bulk $PuCoGa_5$. Furthermore, we study the electronic structure of thin $PuCoGa_5$ films in order to determine the effect of Ga excess at the surface.
This work was supported by the Grant Agency of Czech Republic (Project P204/10/0330) and German-Czech collaboration program (436TSE113/53/0-1, GACR 202/07/J047).

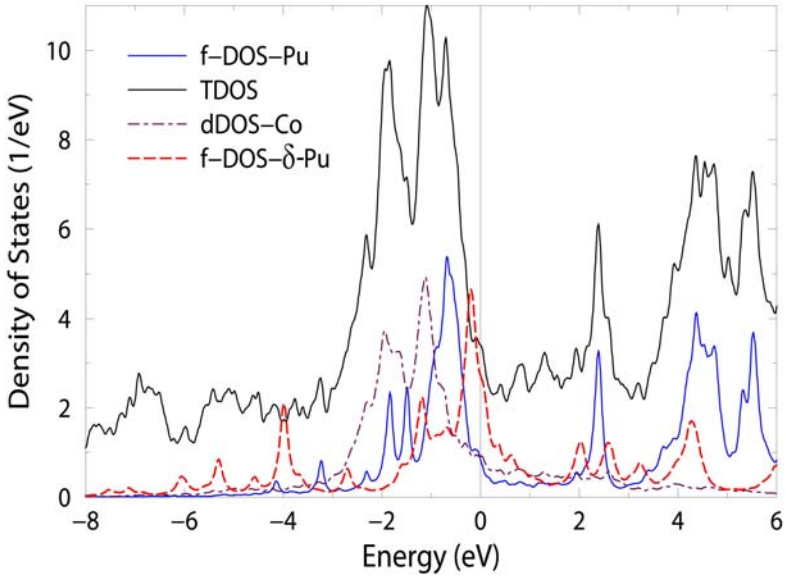

Fig. 2 Total and *f*-projected densities of states (DOS) for bulk $PuCoGa_5$ in comparison with δ-Pu.

References

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