Ab initio study of the electronic structure of plutonium oxides

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Problem under investigation

variation of *Pu* magnetic properties in dependence on its valency

Objects under investigation

$Pu^{+2}O$, $Pu_2^{+3}O_3$, $Pu^{+4}O_2$

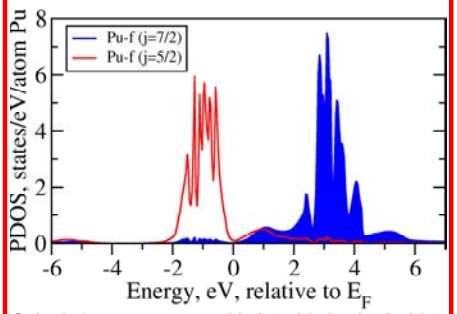
Method for investigation

LDA+U+SO – ***L***ocal ***D***ensity ***A***pproximation with explicit account for Coulomb (***U***) and ***S***pin-***O***rbital interactions

With reference to

A.O. Shorikov, A.V. Lukoyanov, M.A. Korotin, and V.I. Anisimov. *Magnetic state and electronic structure of the δ and α phases of metallic* ***Pu*** *and its compounds.* // PRB 72 (2005) N 024458.

PuO

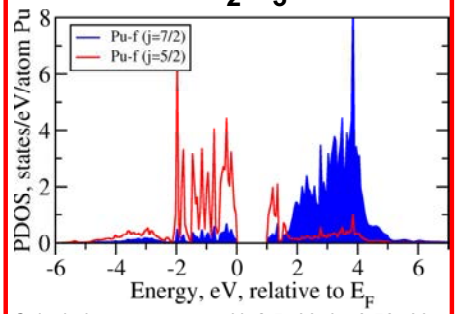


Calculation parameters: U=2.5 eV, J_H=0.52 eV

Calculated moments: S=0.60, L=0.84, J=0.24

Calculated occupancy: n_f=5.34 e, config.: f^6

Pu_2O_3

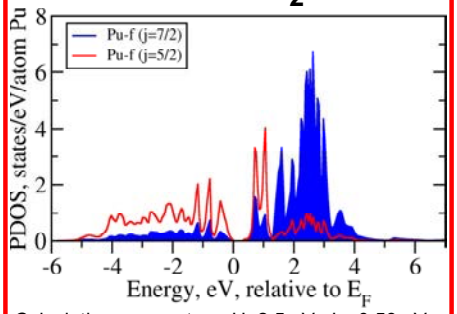


Calculation parameters: U=2.5 eV, J_H=0.53 eV

Calculated moments: S=1.60, L=3.76, J=2.19

Calculated occupancy: n_f=5.09 e, config.: f^5

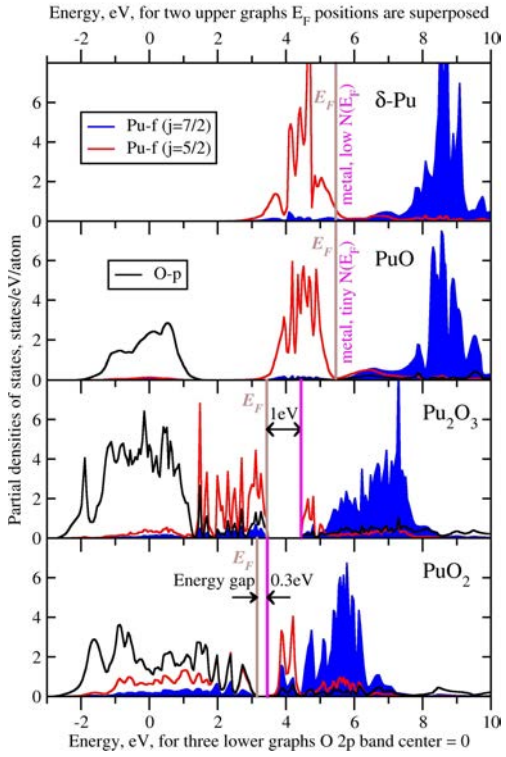
PuO_2



Calculation parameters: U=2.5 eV, J_H=0.53 eV

Calculated moments: S=1.61, L=4.21, J=2.60

Calculated occupancy: n_f=4.84 e, config.: f^4



PuO

The calculated small **Pu** moments make plutonium monoxide similar to nonmagnetic **δ-Pu** (with n_f=5.81). Close proximity of **PuO** to the metallic state is reflected in its density of states (DOS), that differs from **δ-Pu** DOS mainly by the hybridized *j=5/2* and *j=7/2* states whereas the centers of gravity of subbands are located approximately at the same energies.

Pu_2O_3 and PuO_2

To provide f^5 and f^4 configurations, the Fermi level in these oxides separates partially filled *j=5/2* subband. With the decrease of oxidation number, the hybridization of the *j=5/2* and *j=7/2* subbands increases and leads to the opening of a gap in both Pu_2O_3 and PuO_2 (from 1 eV to 0.3 eV) that agrees with insulating ground state of PuO_2 obtained in the previous studies*.

*L.Petit, A.Svane, and W.M.Temmerman, Science **301** (2003) 498.

Basing on oxygen *p*-states DOS near the Fermi level, we conclude that **PuO** demonstrates Mott character of its spectrum whereas Pu_2O_3 has charge-transfer gap. Moreover, PuO_2 is in negative charge-transfer regime.

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