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Ab-Initio Study on Plutonium Compounds Pu3M (M=Al, Ga, In), PuNp and Elemental Neptunium

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Auspices Statement

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REPORT

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AB-INITIO STUDY ON PLUTONIUM COMPOUNDS Pu_3M (M=AL, GA, IN), PUNP AND ELEMENTAL NEPTUNIUM

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Abstract

Using spin-polarized relativistic density functional theory the electronic and magnetic structures for the plutonium compounds $Pu_3M(M = Al, Ga, In)$ and $PuNp$ have been investigated. For the first group of compounds the enhanced hybridization between Pu $5f$ and p-states of alloying element, as it has been found in spin-polarized calculations, is believed to be the main reason for the higher formation energies obtained in such kind of studies in comparison with the non-spin-polarized case. Also, comparative analysis of the actinides U, Np, Pu, Am, and Cm has been performed based on their electronic and magnetic structure. Some noticeable difference in the calculated magnetic structure was discovered between the actinide with local magnetic moments (Cm) and the actinides $(Pu$, $Am)$ in which magnetic moments were found only in the calculations.

I. INTRODUCTION

There is no need to repeat words about the importance of plutonium for the nuclear weapons. It is also wide known that this metal is very interesting itself due to its particular properties and yet not unsolved electronic structure.

As for the engineering applications the cubic delta phase of Pu is the most convenient due to its ductility. Though the phase is only stable from 593 to 736K the small additions of some elements (for example Ga, Al, Ce, Am) stabilize it at room temperature. So, it is very important to understand the mechanism of δ-Pu stabilization.

From the theoretical point of view even pure δ-Pu is a complicated object because both LDA (local density approximation to density functional theory) and GGA (generalized gradient approximation) failed to reproduce the equilibrium properties of it. Some success was achieved with LDA+U approach [1] but using the adjustable Hubbard U parameter defined for the pure Pu makes it difficult to use this approach for the study of plutonium alloys.

On the other hand, taking the spin polarization into account leads to the almost ideal description of the ground state properties of δ-Pu [2–12]. The problem is that the local magnetic moments, which appear in the calculations, have not been found in the Pu experimentally, [13]. An opinion exist that local magnetism is present in δ-Pu but is screened due to the Kondo interactions, [13]. Also, M.Fluss, [14], has proposed a defect-based spin mediated Kondo impurity mechanism of stabilization of δ-Pu. Landa and Söderlind, [11], explained the stability of δ-Pu with the model in which δ-Pu is a disordered magnet that undergoes transformation to an antiferromagnetic (AFM) structure with a mechanical destabilization and phase transition to a lower symmetry phase upon cooling.

Thus, the question of magnetism in Pu is still open. But independently on the physical meaning of magnetism in this metal, the introduction of magnetic ordering in calculations leads to major improvements comparing to the nonmagnetic LDA or GGA results. It seems as if the magnetic interactions were some additional degrees of freedom in the variational principle of density functional theory, and the variation of them would let us improve some results (such as equilibrium volume and bulk modulus) but lead to the wrong magnetic moments.

However not very successful the density functional theory might be for the direct description of plutonium electronic structure, this theory may be still useful for comparative analysis. Keeping that in mind and trying to shed more light upon the magnetism in pure Pu, we have planned a series of calculational studies on Pu compounds. Some of the compounds planned to be calculated are δ-Pu-stabilizers. So, we hope that this study will be useful for the understanding of the δ-Pu stabilization too. The question of magnetism in the plutonium compounds is also interesting because Robert et al., [8], have shown that only inclusion of spin-polarization allows us to bring up the calculated structural hierarchy of $Pu_{1-x}M_x(M = Al, Ga, In)$ in agreement with the trends observed in the experimental phase diagrams.

As a first step in this direction we have concentrated on the alloys $Pu_3M(M = Al, Ga, In)$ too. In the present work the question has been studied within fully relativistic spin-polarized density functional theory. Using Dirac-relativistic quantum numbers allows us to consider the interplay of spin-orbit interaction and magnetic splitting in more details than it was done in Ref. [8] where spin-orbit interaction was treated as perturbation.

We have also planned a series of calculations aimed to the study of neighbors (Np, Am) of Pu in the Periodic Table of elements and their compounds. The purpose of doing that is also our wish to have a look at Pu from different points of view. In this first report the electronic and magnetic structures of Np and the compound PuNp have been investigated.

This paper is organized as follows. In section II the computational method and its parameters are described briefly. The results obtained for the compounds $Pu_3M(M = Al, Ga, In)$ are discussed in section III. The calculations for neptunium and comparison of them with previous results for another actinides are presented in sections IV and V. In the section VI some results of studying on $PuNp$ are given. Lastly, in section VII the conclusions and future plans are offered.

II. THE METHOD OF CALCULATIONS

The density functional theory in the generalized gradient approximation, [15], has been used in this work. The calculations have been performed with the full potential, Dirac relativistic (j, κ) basis, spin-polarized linear-augmented-plane-wave method (RSPFLAPW+LO). The details of the method were presented earlier, [12].

All calculations for the compounds $Pu_3M(M = Al, Ga, In)$ have been carried out with cubic $AuCu_3$ structure, neglecting by a slight tetragonal distortion which exist in Pu_3Al.

Experimental lattice parameters (4.507 Å, 4.507 Å, and 4.702 Å correspondingly) have been applied. For α-Np, the geometry optimization has been performed for all the volumes studied. Also, the theoretical equilibrium volume has been exploited to study the electronic and magnetic structure for PuNp. For this compound fcc structure with 50% Pu atoms changed by Np atoms has been studied. The MT-spheres have been almost touching in all the cases.

Within the muffin-tin spheres the charge density and potential have been expanded in spherical harmonics with a cutoff L_{max} equal to 4. The angular momentum cutoff $L_{max} = 10$ has been used for the basis functions. The basis set has also included the semicore orbitals - $5d, 6s$ and $6p$ for Pu and Np. The plane wave expansion of the wave functions has been terminated in such a manner that the convergence of the total energy has been better than 1 mRy per atom in all the cases.

The integration over Brillouin zone has been carried out by the improved tetrahedron method, [16]. 27, 480, and 910 irreducible points have been used for the calculations on α-Np, bcc-Np, and fcc-Np correspondingly. 294 and 476 irreducible points have been used in the self-consistent $Pu_3M(M = Al, Ga, In)$ and $PuNp$ calculations.

III. THE RESULTS OF STUDYING ON Pu_3Al, Pu_3Ga, Pu_3In

A. Formation energies

As a first step, the energies of formation has been calculated for the compounds Pu_3M. Non-spin-polarized and spin-polarized calculations have been performed. As antiferromagnetic configurations are difficult to generate in complex alloy structure, the ferromagnetic ordering was used in the spin-polarized calculations. The formation energies have been found via the relation [8]

TABLE I: Formation energies for plutonium compounds (in ev/atom) from nonmagnetic (NM) and ferromagnetic (FM) calculations.

	Pu_3Al	Pu_3Ga	Pu_3In
NM	-0.005	-0.041	0.213
FM	-0.226	-0.296	-0.235

TABLE II: The atomic magnetic moments (in Bohr's magnetons) on the actinides atoms in the plutonium's compounds.

Compound	Atom	M_{spin}	M_{orb}	M_{total}
Pu_3Al	Pu	4.751	−1.989	2.762
Pu_3Ga	Pu	4.858	−1.985	2.873
Pu_3In	Pu	5.066	−2.171	2.895
$PuNp$	Pu	3.952	−1.798	2.154
	Np	2.235	−1.595	0.640

$$E^f_{Pu_{1-x}M_x} = E_{Pu_{1-x}M_x} - ((1-x)E_{Pu} + xE_M) \tag{1}$$

The results have been collected in the Table I. As it can be seen, only spin-polarized calculations lead us to the conclusion about stability of the compounds. This fact is in full agreement with the result obtained in [8]. The most noticeable effect of magnetism is seen in Pu_3In. It may be related to the more distant positions of Pu atoms in this compound as compared with Pu_3Al and Pu_3Ga.

B. Electronic and magnetic structure

The partial DOSs for the compounds Pu_3M are presented in the Figures 1, 2, and 3. Total densities of states are given in the Figure 4. Calculated magnetic moments have been placed in the Table II. The spin-resolved DOSs (differential with respect to energy magnetic moments) are presented in the Figures 5 and 6. Also, in these figures the so called magnetic-moment functions are presented. The magnetic-moment function is the contributions to the total moment ($M_{spin} + M_{orb}$) from one-particle states integrated up to the given energy. So, the magnetic-moment function can be considered as some generalization of the definition of atomic magnetic moment, the latter just being the value of the above function at the Fermi energy.

As it is seen from Figures 1 - 3, in the non-spin-polarized calculation the $5f_{5/2}$ and $5f_{7/2}$ states are well defined. On the other hand, in the spin-polarized case there is an intricate interference between spin and spin-orbit splittings. One result of this interference is that

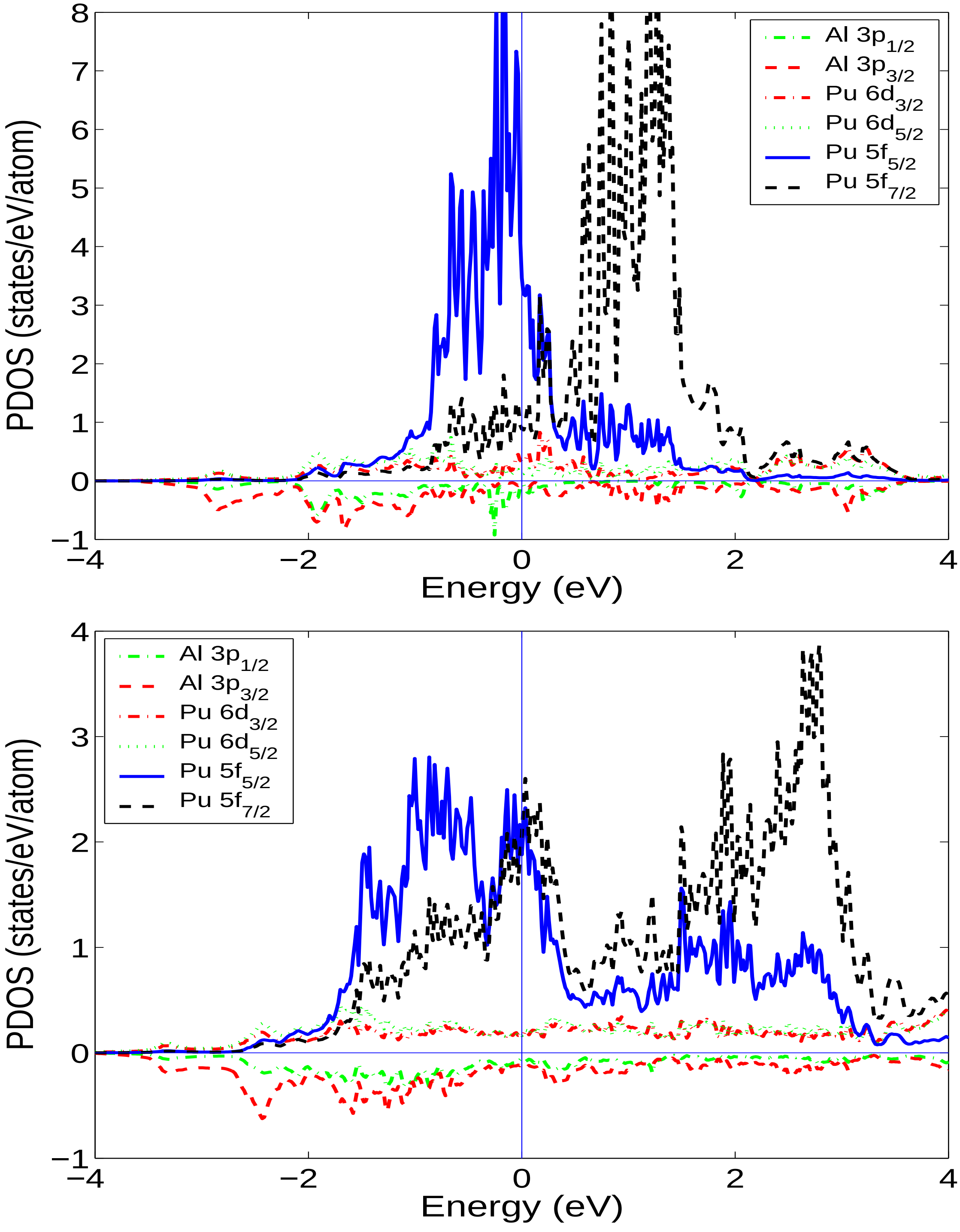

FIG. 1: Partial densities of states (PDOS) for nonmagnetic (upper panel) and ferromagnetic (lower panel) Pu_3Al. Fermi level is shifted to zero energy. The states of Al are given with negative sign for convenience.

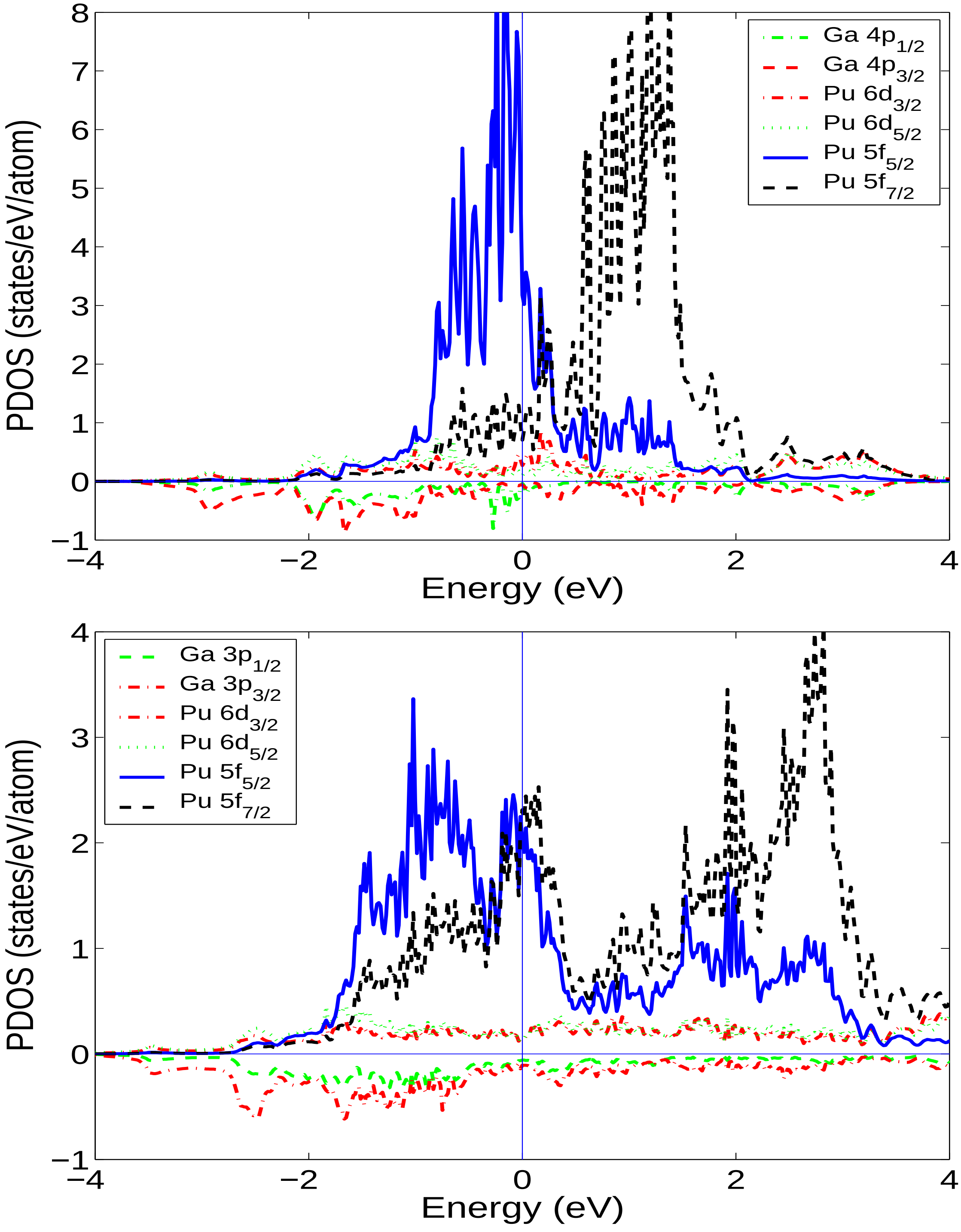

FIG. 2: Partial densities of states (PDOS) for nonmagnetic (upper panel) and ferromagnetic (lower panel) Pu_3Ga. Fermi level is shifted to zero energy. The states of Ga are given with negative sign for convenience.

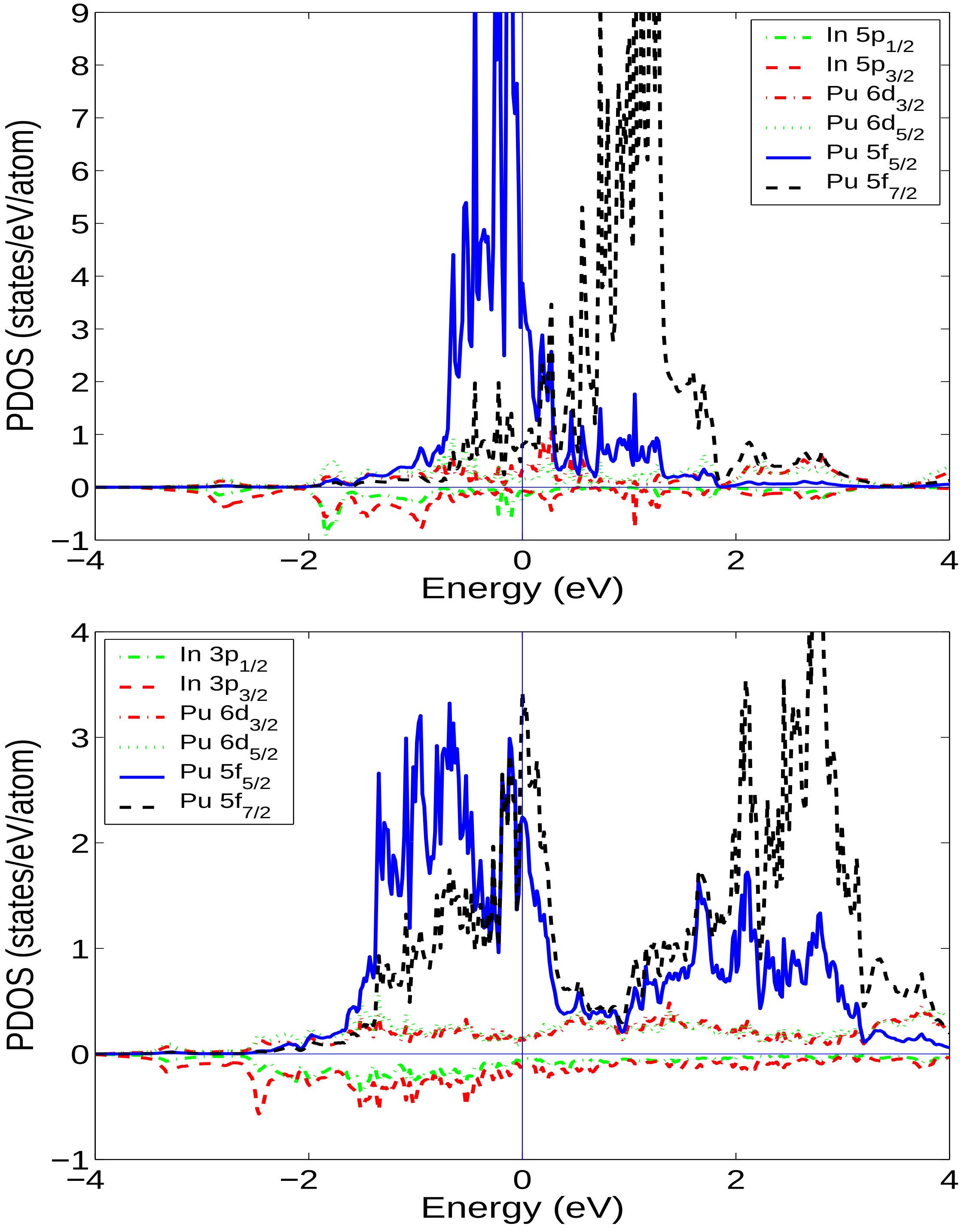

FIG. 3: Partial densities of states (PDOS) for nonmagnetic (upper panel) and ferromagnetic (lower panel) Pu_3In. Fermi level is shifted to zero energy. The states of In are given with negative sign for convenience.

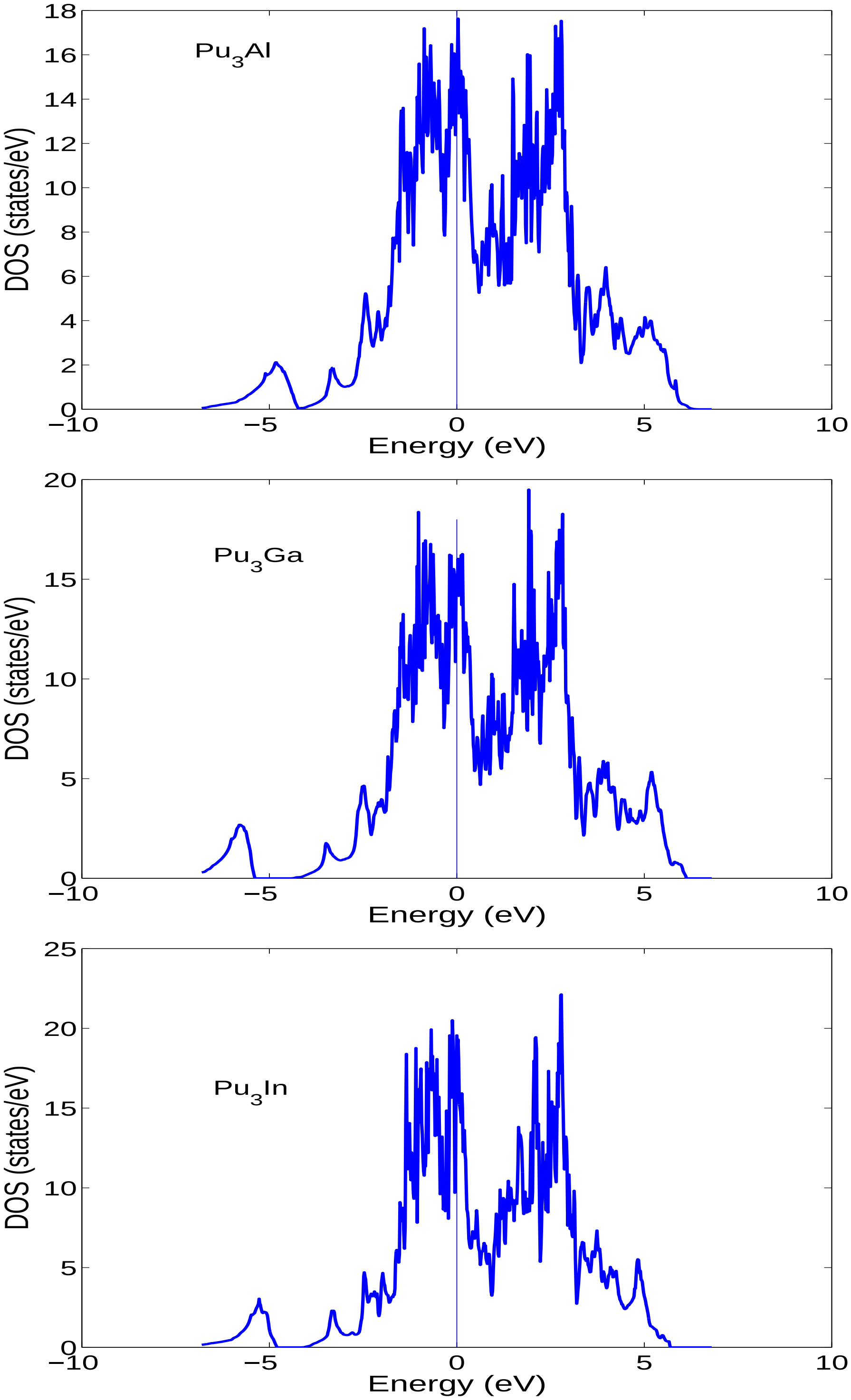

FIG. 4: Total densities of states (DOS) for compounds Pu_3M from ferromagnetic calculations. Fermi level is shifted to zero energy.

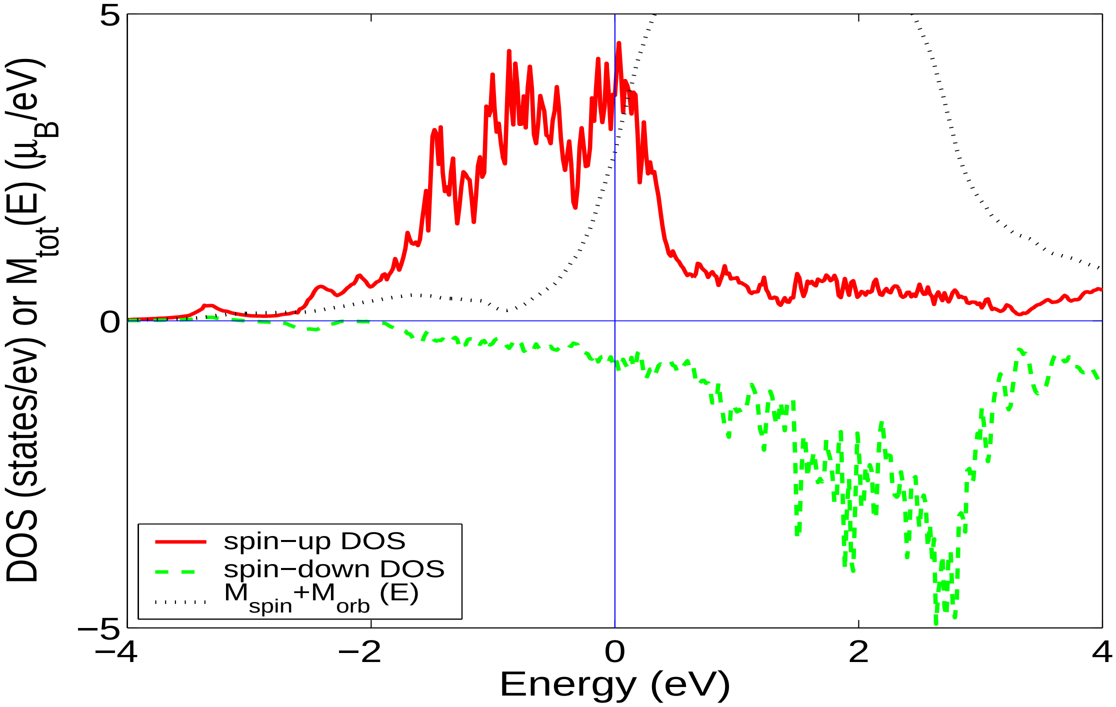

FIG. 5: Spin-resolved DOS and magnetic-moment function (see text for the explanation) for Pu atoms in Pu_3Al. Fermi level is shifted to zero energy. Spin-down DOS is shown with negative sign.

the peaks of DOS at the Fermi level are formed which consist mostly of spin-up states and in which both $5f_{5/2}$ and $5f_{7/2}$ states are well presented. The fact, that the peaks at E_f are formed mostly with spin-up states is very well seen from the figures 5 and 6. Another effect of above interference is that the bottom of $5f$ states is shifted to the lower energies as compared with the results from non-spin-polarized calculations and this fact leads to the stronger hybridization between Pu $5f$ and p-levels of alloying elements. It increases the binding energy obtained in the spin-polarized calculations in comparison with the non-spin-polarized case and can be considered as a main reason for the stabilization of δ-Pu by adding the above elements.

Turning back to the peculiarity of Pu_3In we can notice that $5f_{7/2}$-peak falls exactly on the Fermi level (Fig. 3) while in Pu_3Al and Pu_3Ga this peak is slightly shifted up from E_f and doesn't take part in the binding. We think this difference in the electronic structure as a good explanation for the prominent effect of inclusion of magnetism in the modelling of Pu_3In.

The magnetic-moment functions (Fig. 5 and 6) give a quite interesting food for thought.

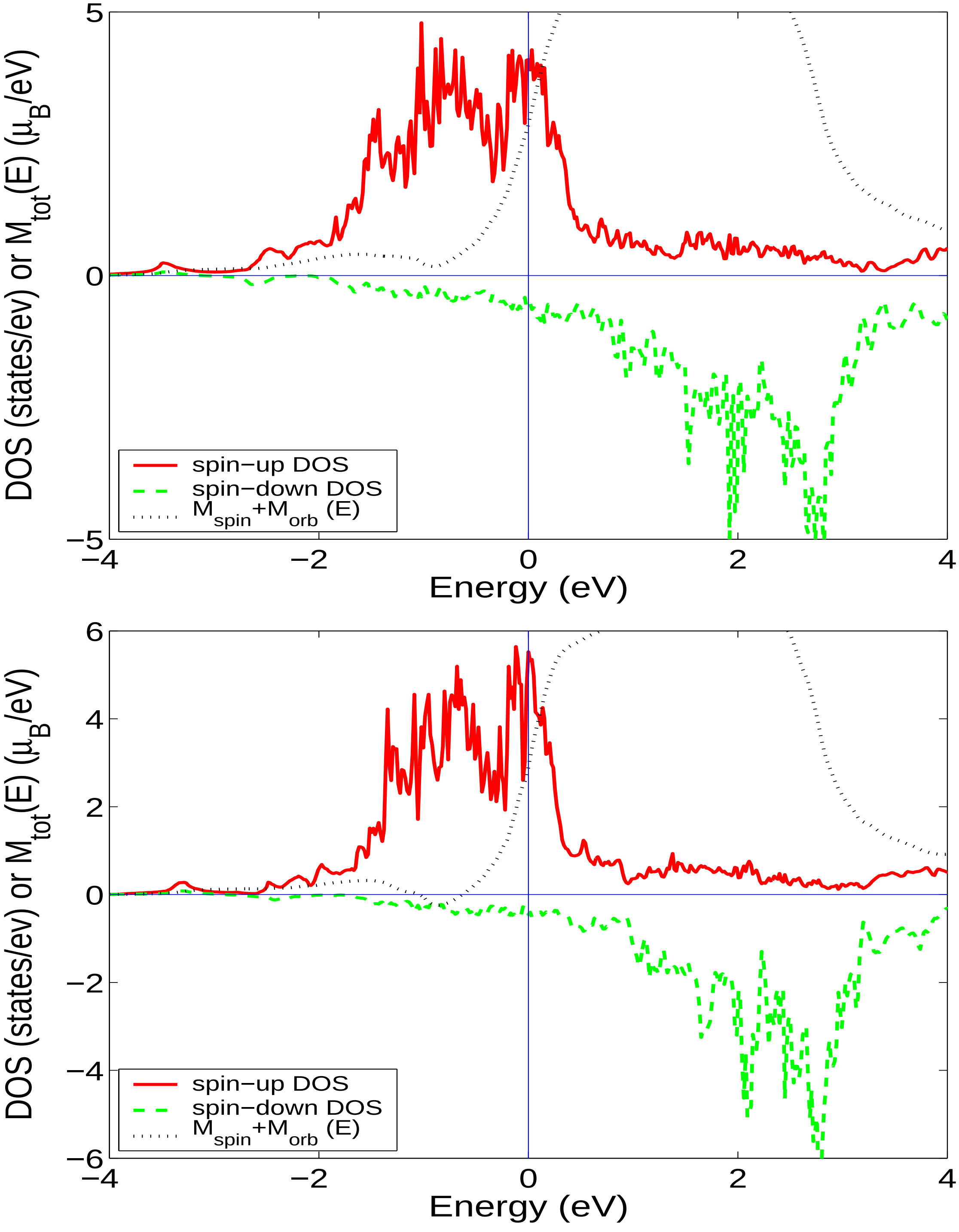

FIG. 6: Spin-resolved DOS and magnetic-moment function (see text for the explanation) for Pu atoms in Pu_3Ga (upper panel) and Pu_3In (lower panel). Fermi level is shifted to zero energy. Spin-down DOS is shown with negative sign.

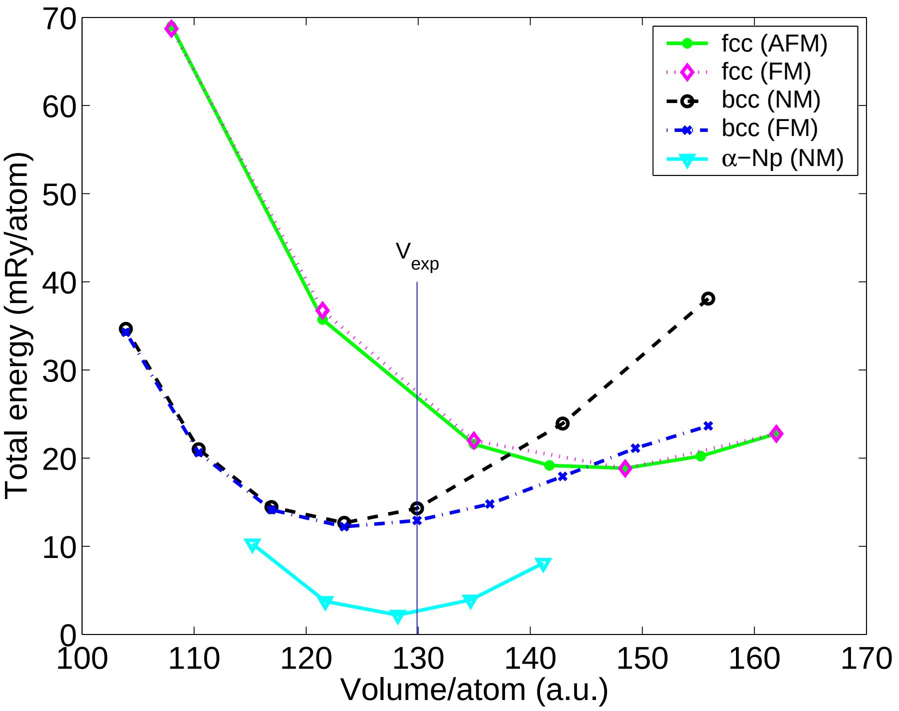

FIG. 7: Total energy versus volume for Np.

It's easily to see that if Fermi energy had been lower by about $1eV$, the total magnetic moments on Pu atoms would have been very close to zero due to cancellation between M_{spin} and M_{orb}. This observation is important in view of the fact that local magnetic moments have not been found experimentally in the Pu_3M compounds as they haven't been found in pure Pu. However, if we speculate that the wrong thing with the spin-polarized GGA is its excessive spin-splitting of the $5f$-states we'll possibly be able to explain the origin of the wrong local moments in calculations. Indeed, if we artificially bring down the spin-splitting, the gravity centers of spin-up and spin-down DOSs will be closer to each other and the peak of magnetic-moment function will go up relatively E_f resulting in zero magnetic moments on the plutonium atoms.

IV. EQUILIBRIUM GEOMETRY AND ELECTRONIC STRUCTURE OF NP

Three different crystal structures have been studied for neptunium in this work: α-Np (orthorombic), bcc, and fcc. Self-consistent spin-polarized calculations for α-Np have resulted in zero magnetic moments, so in the following the results for α-Np will be marked as

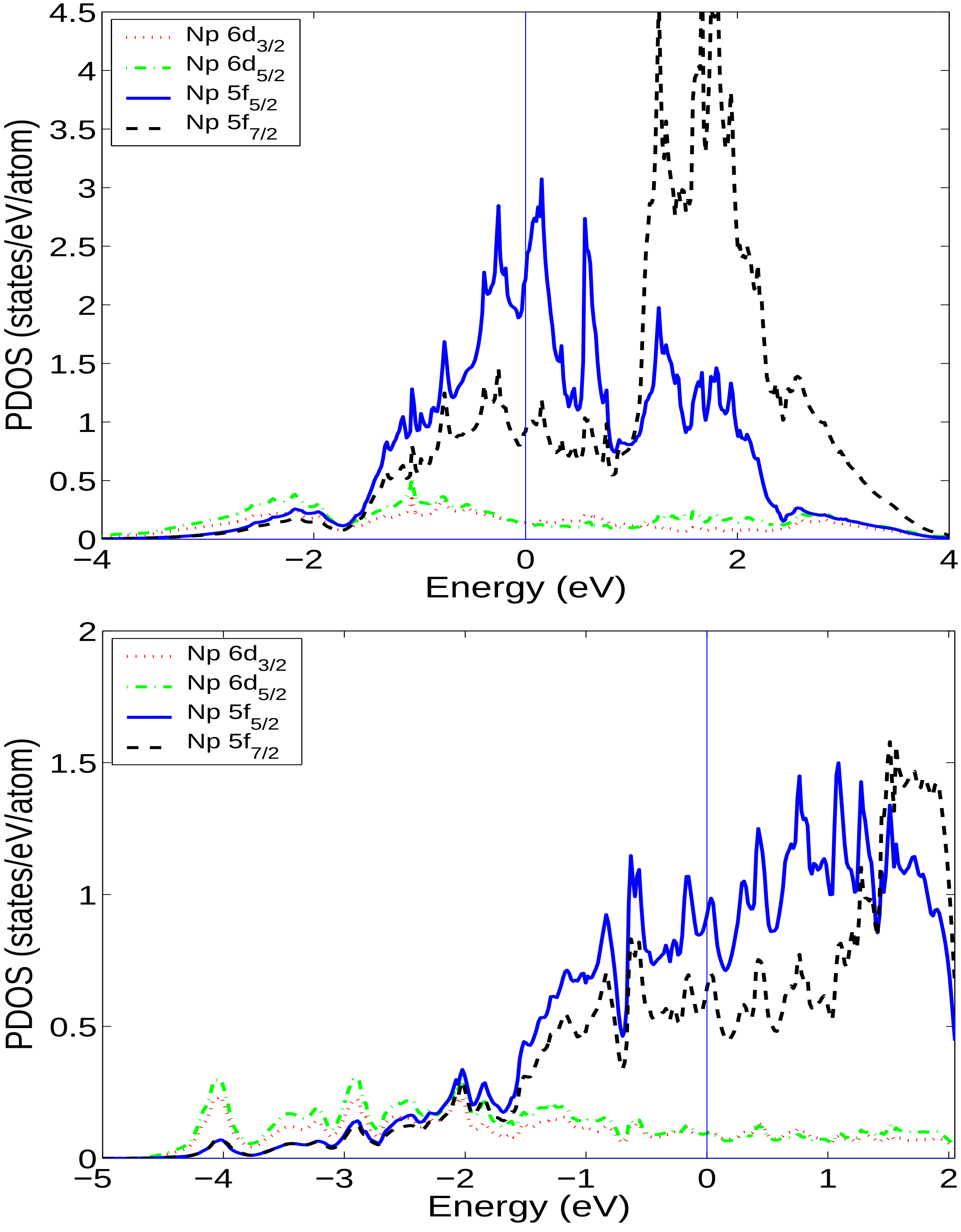

FIG. 8: Partial densities of states (PDOS, states/eV) for ferromagnetic *bcc*-Np (upper panel) and α-Np (lower panel). Fermi level is shifted to zero energy.

NM (nonmagnetic). On the other hand, for *bcc* and fcc crystal structures non-zero magnetic moments (though for *bcc* quite small at the equilibrium volume) on atoms have been found. For fcc structure the FM and AFM orderings have been studied, while for the *bcc* structure only FM ordering of magnetic moments has been investigated. Obtained atomic magnetic moments can be seen in the Table III.

Total energies as functions of volume for above crystal structures are presented in the Figure 7.

Partial electronic DOS for *bcc* (FM) Np and α-Np are presented in the Figure 8 and differential magnetic moments (spin-resolved DOS) of ferromagnetic *bcc* Np are given in the Figure 9.

As it can be seen the energy of α-Np is the lowest and the experimental equilibrium volume for this structure is well reproduced in the calculations. The energy difference between α-Np and *bcc*-Np is about 10 mRy, which is in accordance with the temperature 900K near which *bcc*-Np is stable.

It is easily to see from Fig. 7 that the role of magnetism in $bcc - Np$ which is practically absent at 0K volume will evidently grow up with the increasing of temperature as the volume will increase. For $fcc - Np$ it is not important whether its ordering is ferro or antiferromagnetic. This fact is completely opposite to the case of $fcc - Pu$, [12], in which there is a clear distinction between FM and AFM orderings. This difference between fcc Np and fcc Pu is interesting, but the cause for such a difference is not clear yet.

The fact that some magnetic structure in the high-temperature phase (*bcc*) of Np has been found in the calculations is important. This is on the analogy of Pu, in which the magnetic moments in the high-temperature phase (fcc) are much bigger than in the α-phase, [12]. It seems that magnetism plays the same role in neptunium as it does in plutonium though the effect of magnetism in Np is less than in Pu. Such a thought is supported by the experiments on the electric resistivity, [17,18], in which similar behavior for neptunium and plutonium has been found. The temperature dependance of the resistivity of these metals has been explained in [18] as a result of enhanced electron scattering from the spin fluctuations.

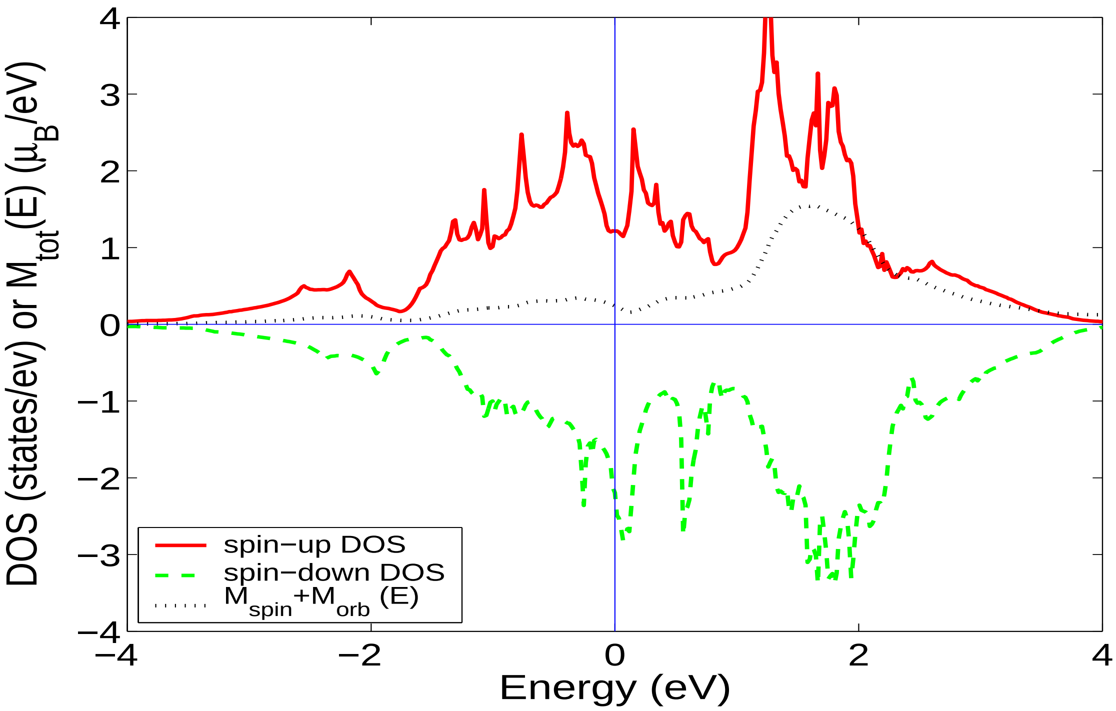

FIG. 9: Spin-resolved DOS and magnetic-moment function (see Section III B for the explanation) for *bcc*-Np. Fermi level is shifted to zero energy. Spin-down DOS is shown with negative sign.

V. COMPARATIVE ANALYSIS OF ELECTRONIC AND MAGNETIC STRUCTURE OF U, Np, Pu, Am, AND Cm

First of all, let us consider the equilibrium volumes of the actinides. In the figure 10 they are presented in comparison with the corresponding experimental volumes.

As it can be seen, the calculations reproduce experimental volumes for actinides quite well except for plutonium. This fault in the case of Pu is, obviously, due to some correlation effects not accounted for in the present DFT calculations. Obtained magnetic moments (Table III) may also be considered as an evidence of correlations missed for Pu, because magnetic moments have not been found in it at low temperatures, [13]. Similar conclusion about the importance of correlations in Pu has also been obtained by other researchers, [19–22], and is in accord with the work by Savrasov *et al.*, [23], which argued, using many-body dynamical mean-field approach (DMFT), that both α-Pu and δ-Pu phases are strongly correlated.

Table III contains calculated and experimental magnetic moments. In the above row of

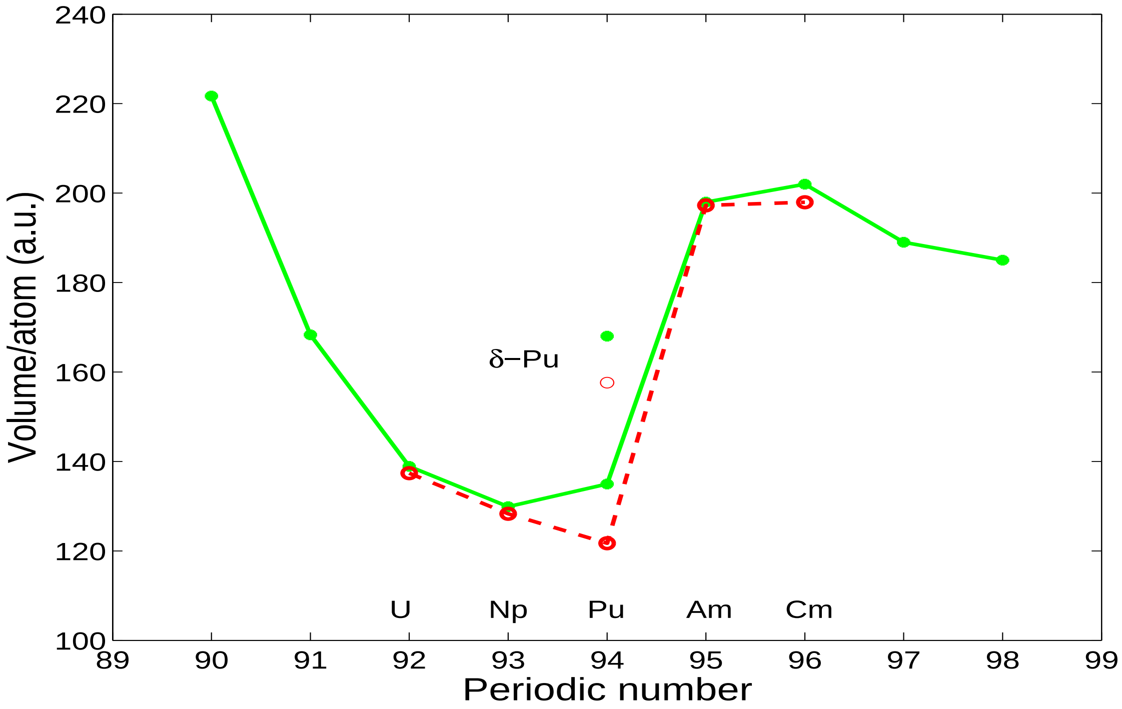

FIG. 10: The equilibrium volumes of the the actinides. Experimental data are shown with solid line and theoretical results - with dashed line.

actinides the static magnetic moments have been found experimentally only for curium and for this element the theory is seemed to reproduce them quite well.

It is interesting to note that in all actinides with magnetic ground state the ground state is antiferromagnetic. Also, in transition from α-Pu to Cm there is a tendency of increasing of the energy difference between NM and AFM phases (3.2, 40, 107, 146 mRy/atom for α-Pu, δ-Pu, Am, and Cm respectively).

The fact deserves to be mentioned that in americium a very good description of ground state properties was found only in the calculations in which the magnetism was taken into account. But the formation of the spin moments in Am, as obtained in our calculations, is in contrast to the often presented in the papers claim about nonmagnetic ground state for Am. Nonetheless, spin polarization cannot be ignored for Am because the NM calculation gives too bad ground state properties for it. Also, our result is in full accord with the result by Söderlind *et al.*, [24].

The partial DOSs of the actinides are presented in the Figures 11, 12, 13.

The spin-resolved DOSs from the spin-polarized calculations for α-Pu, δ-Pu, Am, and

Cm are presented in the Figure 14.

The calculated densities of states (DOS) show the progressive separation of the 5f manifold into two groups of states. In α-U and α-Np this separation is fully spin-orbit. In α-Pu both spin-orbit and exchange splittings are responsible for the separation. Beginning from δ-Pu the spin (exchange) origin of splitting prevails more and more.

One more interesting speculation might be drawn from the magnetic-moment function (Fig. 14). As it has already been said for the compounds Pu_3M (section III) the shifting of E_f down might result in zero magnetic moments. The same is true for pure actinides α-Pu, δ-Pu, and Am: if we bring down the internal magnetic field (exchange splitting) so that Fermi level was lower by about $0.5eV$ we will (possibly) obtain very small total magnetic

TABLE III: The atomic magnetic moments (in Bohr's magnetons) of actinides with AFM magnetic structure. All values are from the calculations at experimental equilibrium volumes.

Structure	Atom	M_{spin}	M_{orb}	M_{tot}	M_{exp}
α-U	1	0	0	0	0
α-Np	1	0	0	0	0
bcc-Np (FM)	1	0.74	−0.50	0.24	
fcc-Np (FM)	1	2.47	−1.67	0.80	
fcc-Np (AFM)	1	2.58	−1.65	0.83	
α-Pu	1	−0.46	0.18	−0.28	0
	2	2.22	−1.00	1.22	0
	3	−2.62	0.99	−1.63	0
	4	2.07	−0.94	1.13	0
	5	2.20	−1.09	1.11	0
	6	−2.80	1.24	−1.56	0
	7	−2.44	1.03	−1.41	0
	8	3.54	−1.65	1.89	0
δ-Pu	1	4.09	−1.75	2.34	0
Am	1	6.23	−0.82	5.41	0
Cm	1	6.69	0.16	6.85	8.1

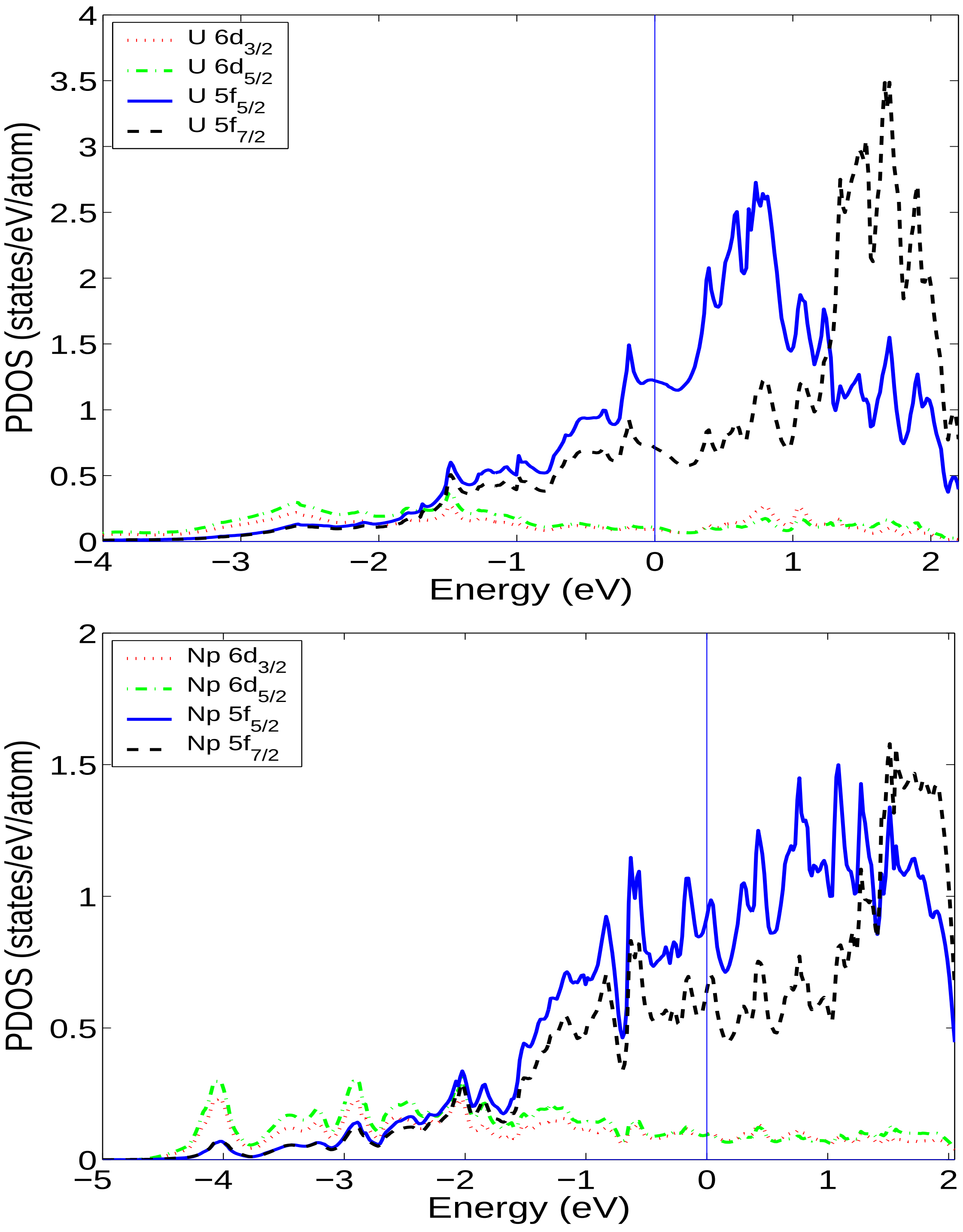

FIG. 11: Partial densities of states for α-U (upper panel) and α-Np (lower panel).

moments. The same is not true however in the case of Cm: there is a high plato in magnetic-moment function of this element which goes to zero only at energy $2.5eV$ below E_f. This observation is in full agreement with the experimental information: the absense of magnetic

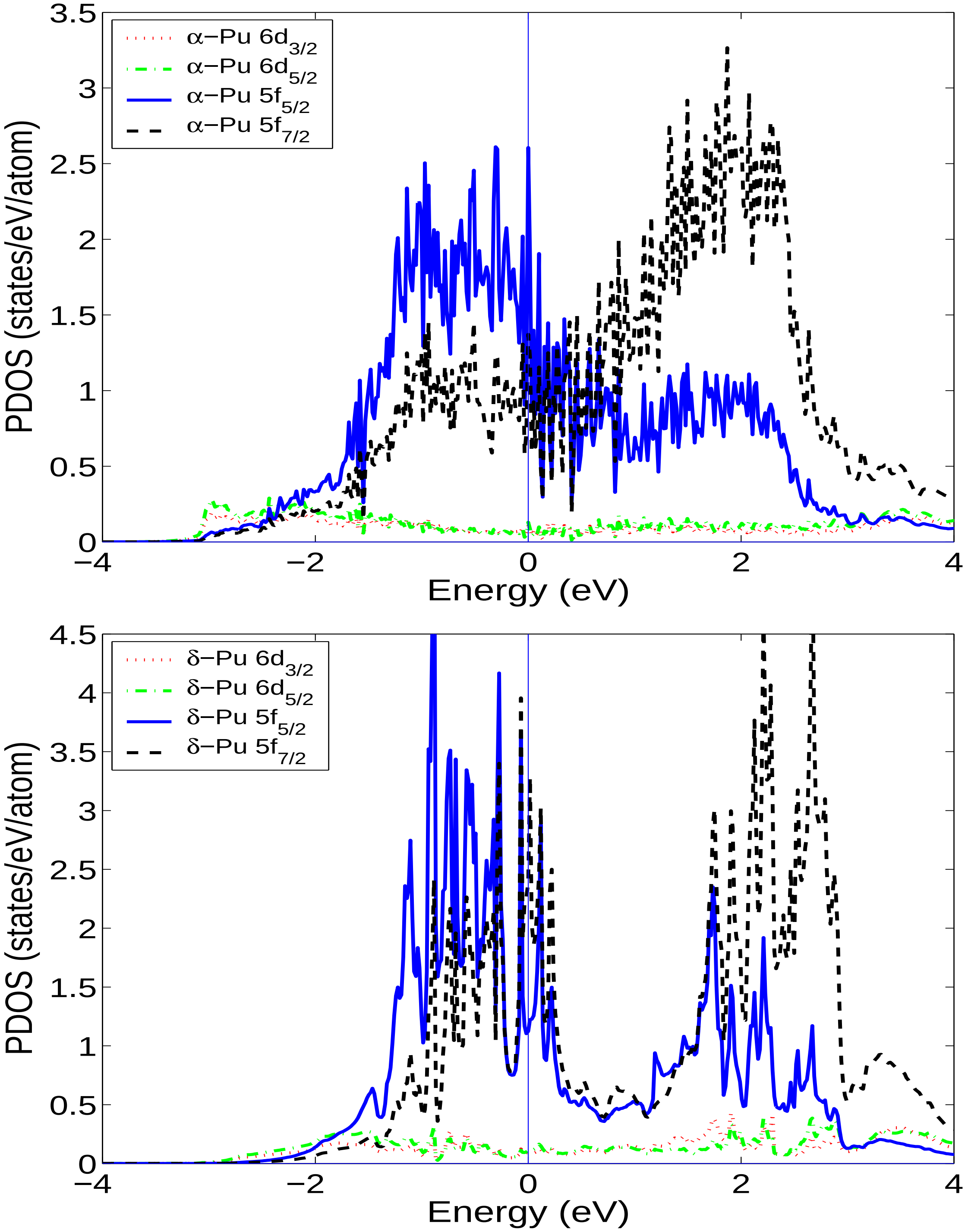

FIG. 12: Partial densities of states for α-Pu (upper panel, representative atom) and δ-Pu (lower panel).

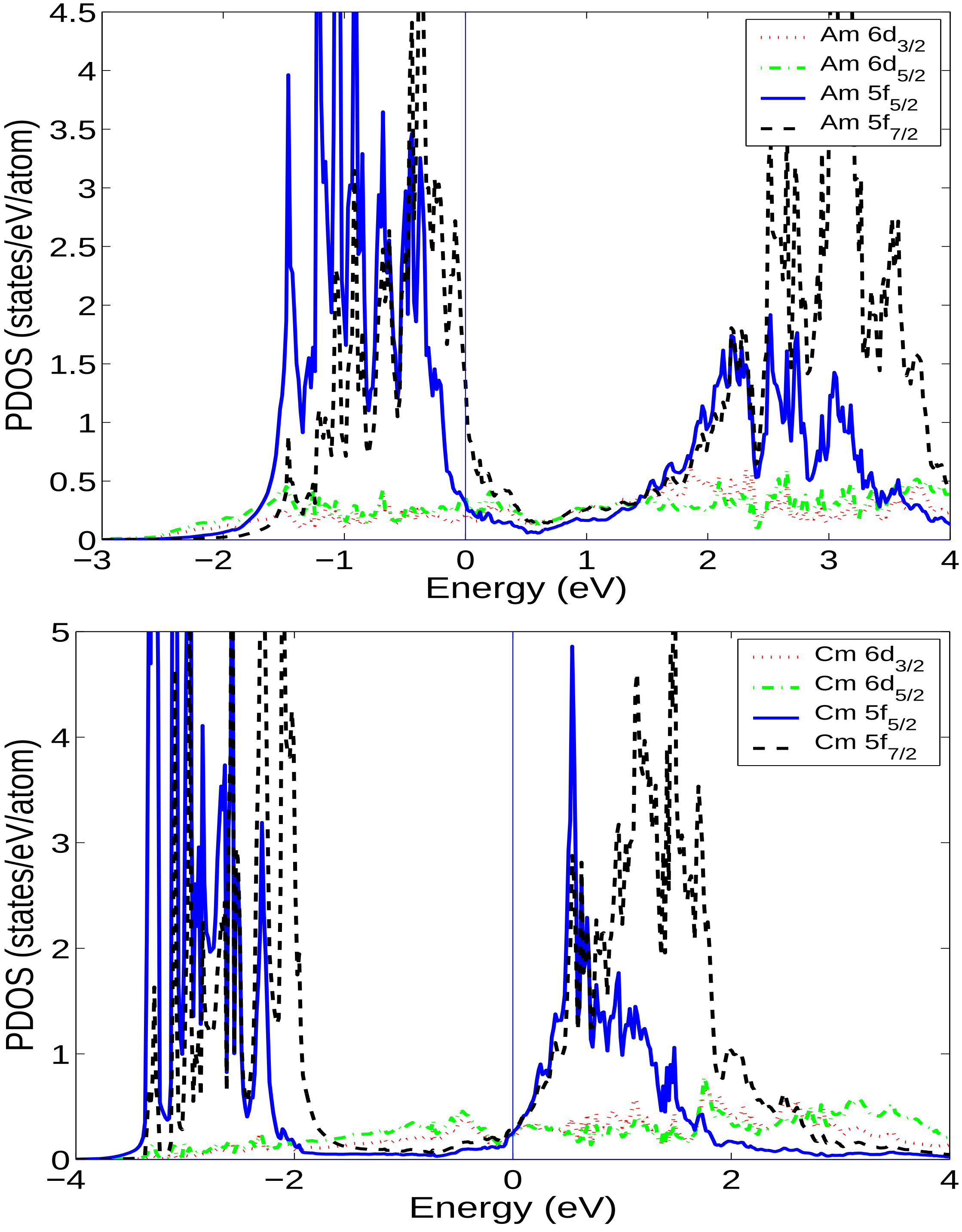

FIG. 13: Partial densities of states for antiferromagnetic dhcp Am (upper panel) and Cm (lower panel).

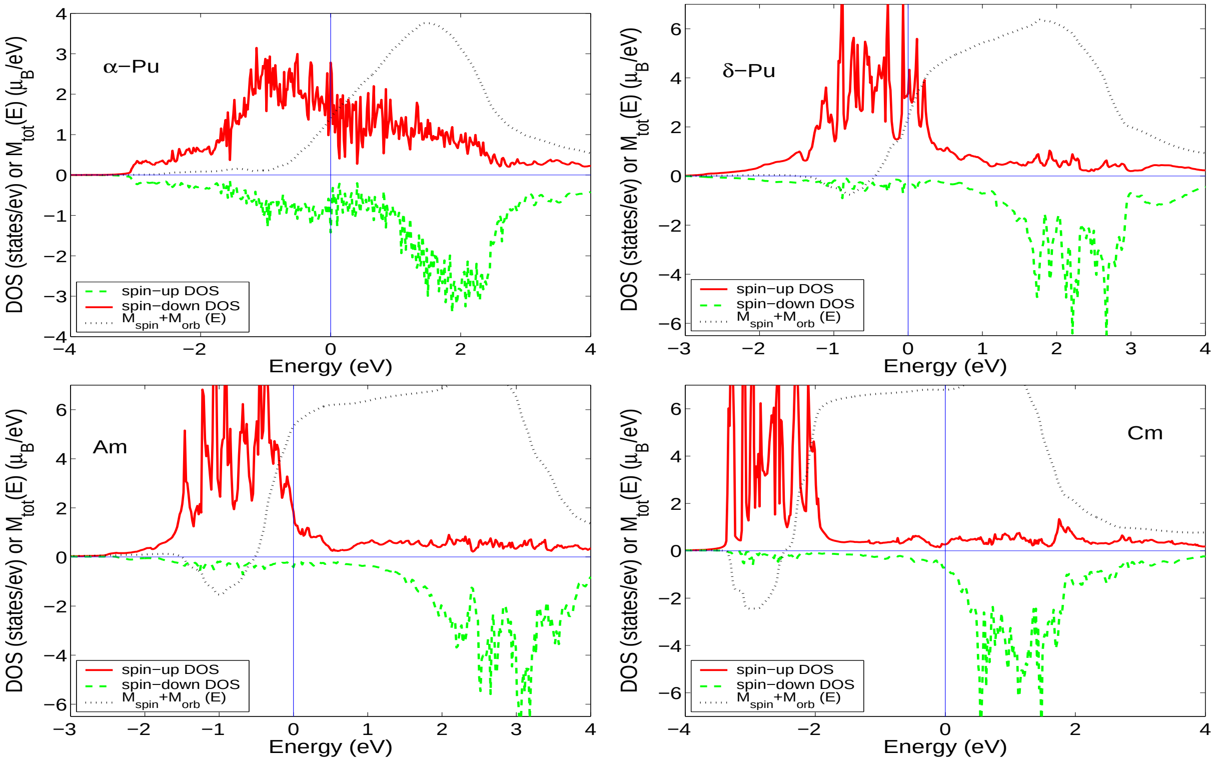

FIG. 14: Spin-resolved DOS and magnetic-moment function (see Section III B for the explanation) for α-Pu, δ-Pu, Am, and Cm. Fermi level is shifted to zero energy. Spin-down DOS is shown with negative sign.

moments in Pu and Am and the big moment found for Cm. So, this consideration can also be thought as some evidence of too strong spin-splitting in the present DFT-GGA approach.

VI. $PuNp$: ELECTRONIC AND MAGNETIC STRUCTURE

In the present work the calculational study for the compound PuNp has also been performed. This studying (together with the calculations on PuAm, which are going to be finished during next quarter) is aimed to give a comparative analysis of the influence of the changing some Pu atoms with Np (or Am) on the calculated electronic and magnetic properties.

In the Figure 15 the total energy of PuNp is given as a function of volume from both non-spin-polarized and spin-polarized calculations. The formation energy calculated following to the receipts given in chapter I appeared to be positive: $0.211 eV/atom$ in the nonmagnetic case, and $0.177 eV/atom$ in the spin-polarized case. So, these calculations result in the fact that pure Pu and Np are more stable than their compound $PuNp$. In this respect it is interesting to calculate energy of formation for the compound of Pu with its another neighbor - Am which is known to be δ-Pu - stabilizer. This task is expecting to be performed in the next quarter.

The electronic (partial DOS) and magnetic (spin-resolved DOS) structures are presented in the Figures 16 and 17 correspondingly. Total DOS is given in the Figure 18. Calculated magnetic moments for spin-polarized case are given in the Table II. The partial DOSs of Np atoms in non-spin-polarized calculation are almost the same as partial DOSs of Pu. But the difference increases in the spin-polarized case. Plutonium states are more affected by magnetic effects. As it is seen from partial DOS and spin-resolved DOS, both spin-orbit splitting and exchange splitting play their roles in the formation $PuNp$ electronic structure, though for Pu atoms the magnetism is more responsible while for Np atoms the spin-orbit interaction prevails. And again, the magnetic-moment function for Pu tends to zero at the energy about $1eV$ below Fermi level. The question arises: how does this function behave for the compounds of Pu in which Pu has real (i.e. experimentally found) local magnetic

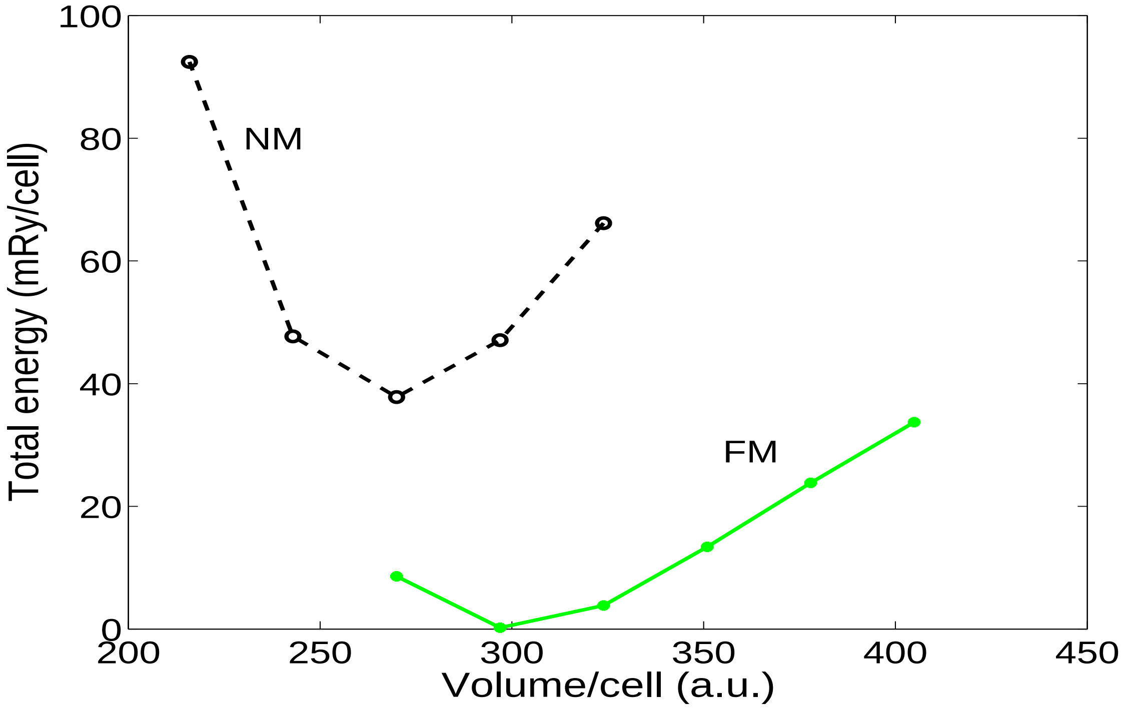

FIG. 15: Total energy versus volume for PuNp from nonmagnetic and ferromagnetic calculations. Unit cell contains one Pu atom and one Np atom.

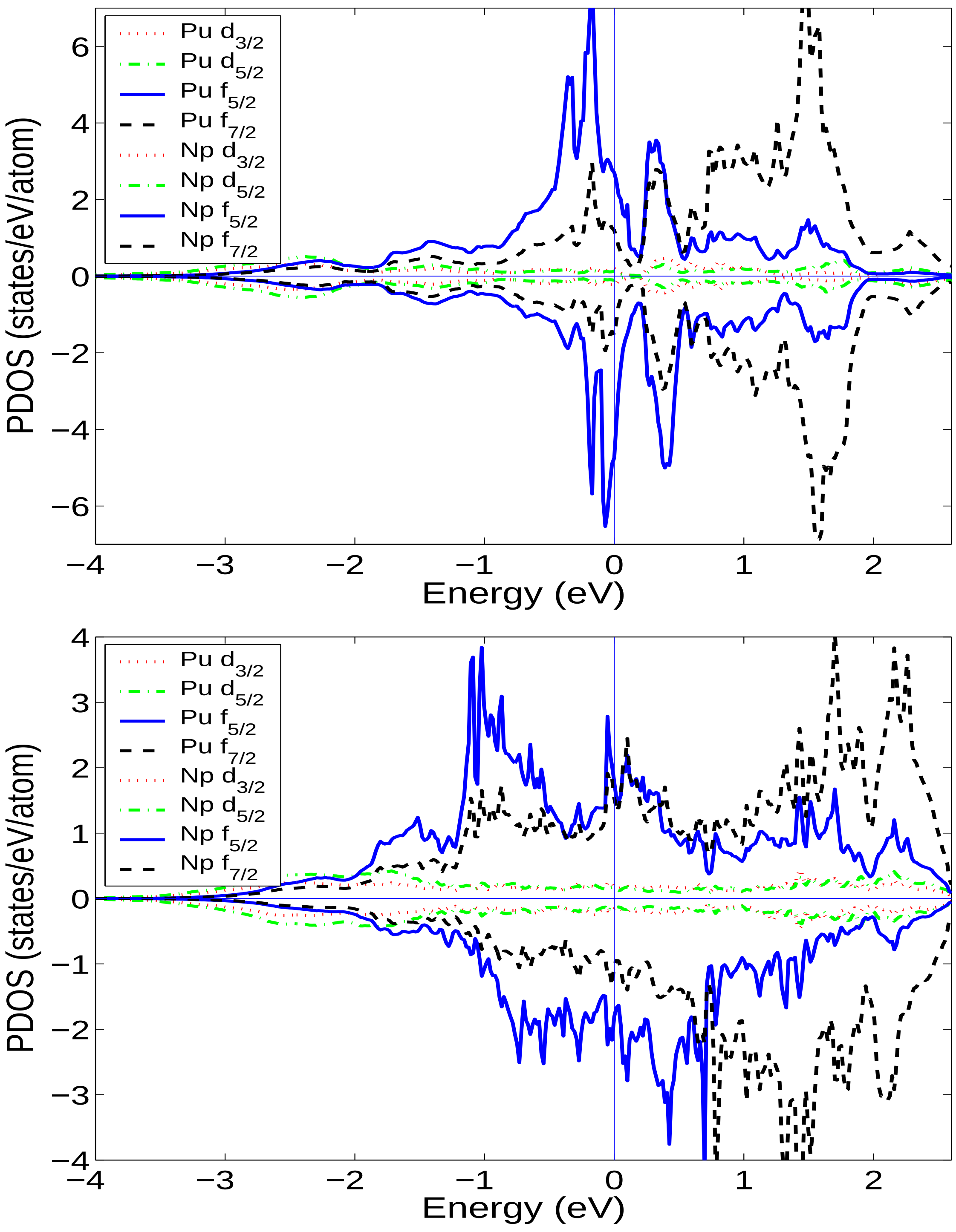

FIG. 16: Partial densities of states (PDOS, states/eV) for nonmagnetic (upper panel) and ferromagnetic (lower panel) $PuNp$. Fermi level is shifted to zero energy. The states of Np are given with negative sign for convenience.

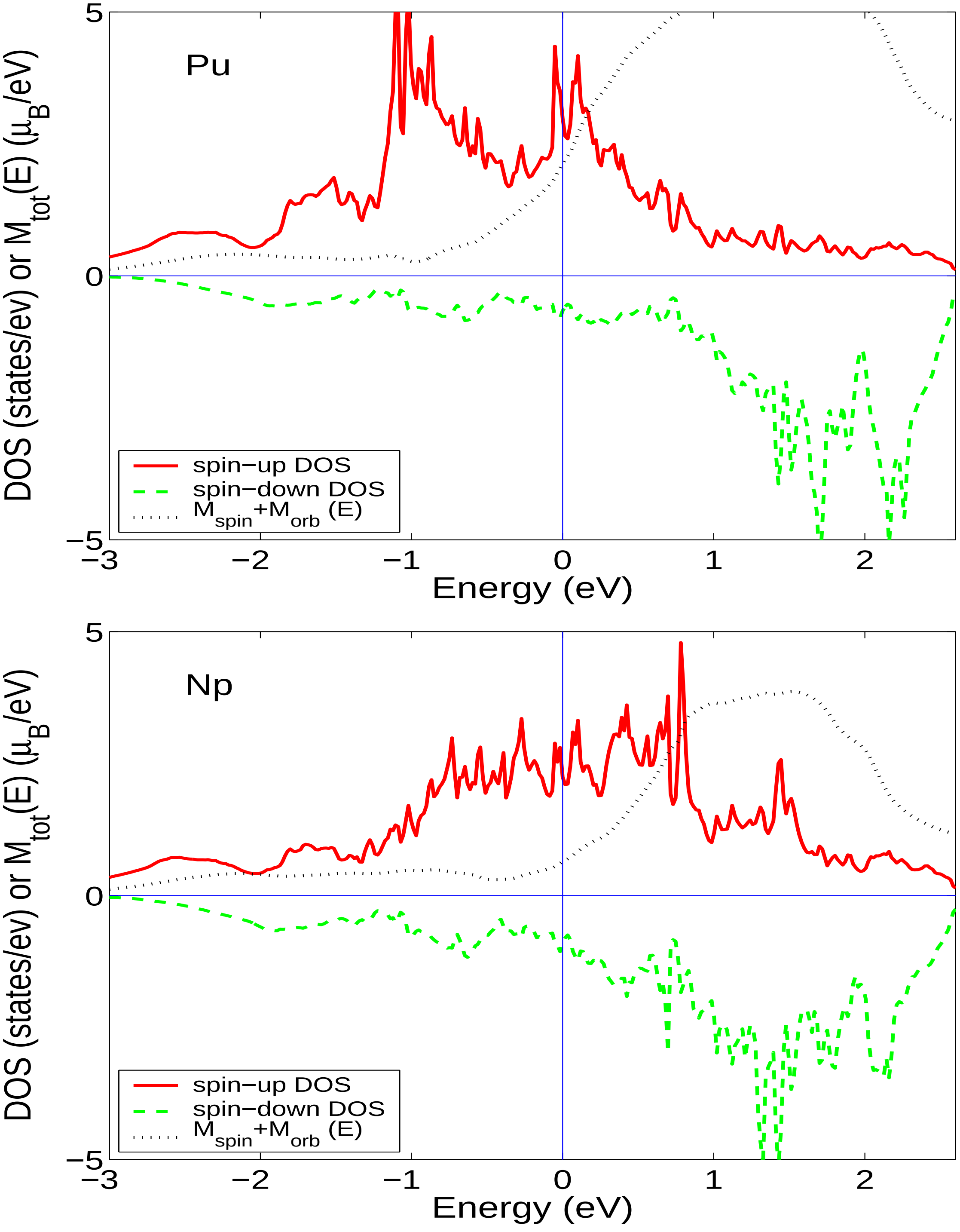

FIG. 17: Spin-resolved DOS and magnetic-moment function (see Section III B for the explanation) for $PuNp$. Fermi level is shifted to zero energy. Spin-down DOS is shown with negative sign.

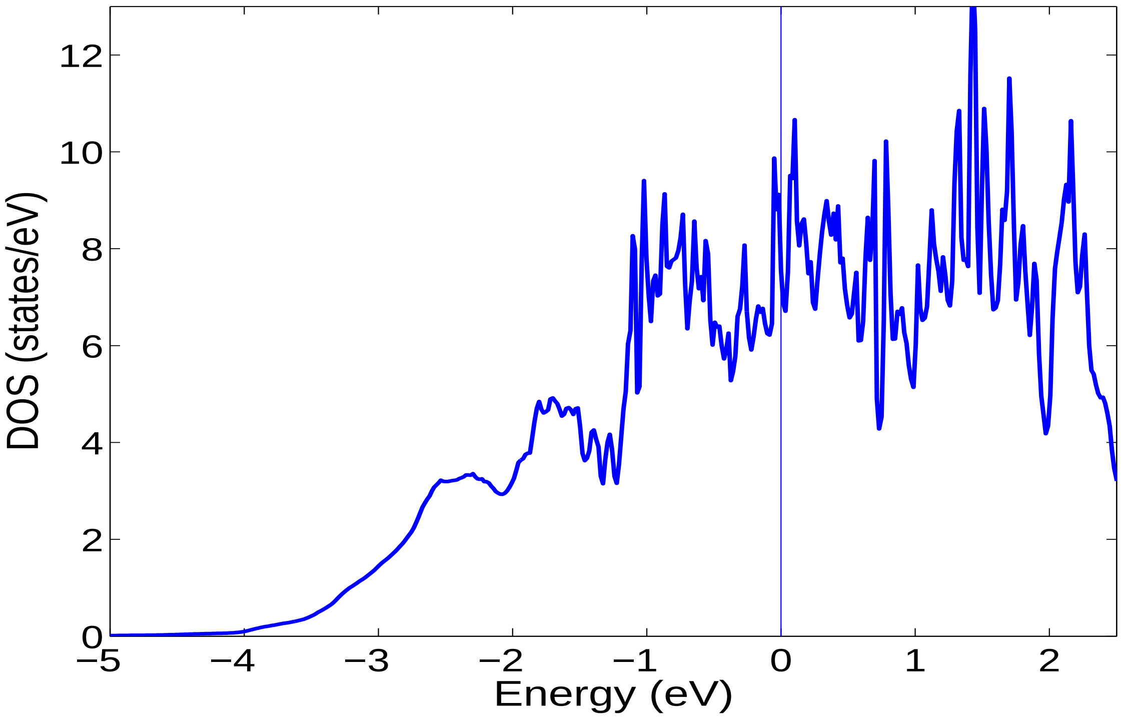

FIG. 18: Total densities of states (DOS, states/eV) for compound $PuNp$ from ferromagnetic calculations. Fermi level is shifted to zero energy.

moments?

VII. CONCLUSIONS AND PLANS FOR THE FUTURE

In summary, first-principles relativistic GGA-based calculations have been applied to describe the electronic and magnetic structure for compounds $Pu_3M (M = Al, Ga, In)$ and to find their formation energies. In accordance to the work [8] the key role of spin (magnetic) polarization in the binding has been found for these compounds. The future plans related with the above calculations consist in performing the same studying for the compounds Am_3M and Np_3M $(M = Al, Ga, In)$, i.e. for the compounds of neighbors of Pu in the Periodic Table.

Also, the comparative analysis of the actinides U, Np, Pu, Am, and Cm has been performed based on their electronic and magnetic structure. Some noticeable difference in the calculated magnetic structure was discovered between the actinide with local magnetic moments (Cm) and the actinides $(Pu,\ Am)$ in which magnetic moments were found only in calculations.

In general, these calculations suggest that the present version of spin-polarized DFT gives excessive spin-splitting when applied to the actinides. So, it is interesting to check this suggestion by conducting some kind of scaled-spin-polarized calculations in which the internal magnetic field would be artificially bring down to some degree. As for the reason of such excessive spin-splitting, the following might be responsible: spin-polarized version of DFT takes into account self-consistently only the spin part of magnetism. But in actinides the orbital part of magnetism is comparable with its spin part. Orbital magnetic moments directed opposite to the spin moments, so it is natural to suppose that the inclusion of orbital magnetization self-consistently will decrease the magnetic splittings and, thus, bring the magnetic (and electronic) structure of actinides in better agreement with experiments. However, as far as we know, such functional has not yet been elaborated though some works exist, [25]. Under the circumstances, it seems to be useful a simplified model - the so called orbital polarization approach, proposed in [26].

Third object that has been under study in the present work is the compound PuNp. Its electronic and magnetic structures have been investigated and they are waiting to be compared with the structures of PuAm, which are going to be studied during next quarter.

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