

Conditions for magnetism in Pu-based systems

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Abstract

The possibility of the 5f count variations and its impact on magnetic properties are discussed on the background of known data on diluted alloys, intermetallic compounds and monpnictides and monochalcogenides. Weakly paramagnetic character of δ -Pu is preserved when Pu is doped by Am, which expands the crystal lattice. Specific-heat data show that the γ -coefficient does not increase. Such behaviour can be understood on the basis of LSDA + U calculations, which yield invariably non-magnetic Pu state due to the proximity to the 5f⁶ state. It is also shown that the ground-state density of states should not be directly compared with photoelectron spectra, because the spectral density obtained from Hubbard-I calculations is in the narrow band regime of Pu dramatically different than the underlying density of states. We suggest that a general condition for Pu magnetism is some degree of hybridization with states of other elements in alloys and compounds, which allows for the reduction of the 5f count.

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1. Introduction

While light actinide elements are Pauli paramagnets with itinerant 5f states at the Fermi level, the localization for actinides behind Am leads to magnetic ordering analogous to lanthanides. The specific position of Pu just before the localization threshold should make it very sensitive to external variables, but various Pu allotropic phases have surprisingly nearly identical weak magnetic susceptibility, despite a large volume expansion exceeding 20% for the *fcc* δ -Pu comparing to monoclinic α -Pu. A susceptibility study over a broad temperature range covering all solid phases [1] reveals only a very weak temperature dependence and absolute values lower than $1 \times 10^{-8} \text{ m}^3/\text{mol}$. The situation does not change qualitatively if the *fcc* δ -Pu is stabilized at low temperatures by various dopants [2,3] and in fact the highest susceptibility was found for β -Pu [1] (monoclinic phase with the volume larger than α -Pu, but smaller than δ -Pu). This fact

suggests that magnetic features of Pu do not depend primarily on the overlap of the 5f functions between nearest neighbours.

This situation poses a problem for a theoretical description, because LDA or GGA calculations, which work reasonably well for a vast majority of metallic systems, indicate the formation of sizeable magnetic moments corresponding roughly to the 5f⁵ state in the expanded phases. Non-zero spin polarization not only appears whenever a spin-polarized solution is allowed, but is actually necessary to reproduce the large equilibrium volume. This situation led to a widespread opinion that some sort of static or slowly fluctuating moments indeed exist in δ -Pu, but they remained undetected (for overview see Ref. [4] and references therein). The original belief that the electronic structure of Pu inclines to the magnetic 5f⁵ configuration was based on the work of Johansson and Rosengren [5], who calculated that the fully localized 5f⁵ manifold should be energetically favourable over 5f⁶ in solids. Other arguments come from results of LDA/GGA calculations (see e.g., Ref. [6]), which indeed obtain the 5f count quite close to 5.0. Similarly, the mixed-level model (MLM) obtains for δ -Pu about one 5f electron in band states if assuming the 5f⁴ localized configuration [7]. On the other hand, there is no

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a priori reason why to assume only integral (or nearly integral) 5f counts in a system in which the 5f states are still band-like (albeit strongly correlated). In this respect it would be an equally non-physical constraint if we assumed integral 3d occupancy in Fe.

One possible solution of the lack of magnetism in δ -Pu was recently suggested by the *around mean field* LSDA + U calculations, which yield a non-magnetic ground state ($S_z = 0, L_z = 0$) and correct cohesion properties [8]. These calculations (as well as those of Ref. [9]) produce a band-like strongly correlated ground state for δ -Pu, in which the 5f manifold is hybridized with the valence band and the 5f count (5.44) is substantially higher than the originally expected value close to 5.0. Using the “atomic-like” language (which does not fully comply with the band character of the 5f state), one can think of the δ -Pu correlated ground state as approaching the non-magnetic 5f⁶ manifold. The calculations also indicate [10] that the magnetic moments are not formed, when the lattice is expanded by Am doping, or if the dimensionality is reduced. For pure Am, the LDA + U calculations [10] correctly reproduce the non-magnetic 5f⁶ state.

The aim of this contribution is to discuss several interesting cases as well as a broader systematics of occurrence of magnetism in Pu systems. The information is, as we believe, relevant for discussion of the nature of the 5f states in Pu systems.

2. Specific heat in the Pu–Am system

So as to explore the reaction of δ -Pu to the volume expansion, we have studied the Pu–Am system. The solid solution of Am in Pu stabilizes the *fcc* phase down to the lowest temperatures, expanding the volume (by 7% for 30% Am). If we assume that δ -Pu is a strongly correlated system at the verge of magnetic ordering, the expansion would most likely lead to a magnetic order or at least to the appearance of Pu local moments. However, no magnetic ordering appears when expanding the lattice (up to about 40% Am tested experimentally [3]) and magnetic susceptibility stays below $1 \times 10^{-8} \text{ m}^3/\text{mol}$ and finally decreases

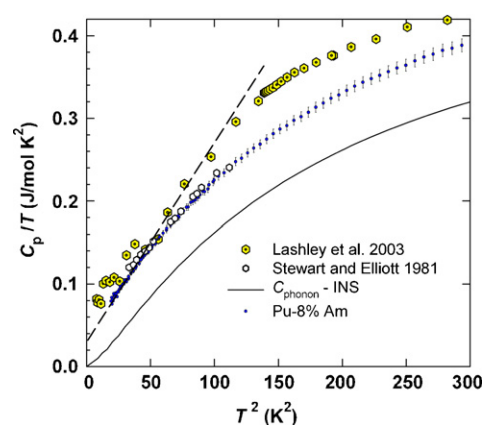


Fig. 1. Specific heat in the C_p/T vs. T^2 representation for Pu–8% Am, compared with data from Refs. [11,14] on Al-doped Pu. The dashed line represents a tentative linear extrapolation of the Pu–8% Am data, leading to $\gamma = 35 \text{ mJ/mol K}^2$. The full line is the phonon specific heat emulated from inelastic neutron spectra of Al doped δ -Pu. If this is used as a basis for extrapolation, $\gamma = 55 \text{ mJ/mol K}^2$ is obtained.

if related to mole Pu (the Van Vleck susceptibility of Am is higher than the susceptibility of δ -Pu.). An important issue is what happens with the enhanced γ -coefficient of the specific heat $\gamma = 64 \text{ mJ/mol K}^2$ [11]. If the primary reason for the elevated γ -coefficient in δ -Pu is a formation of a narrow quasiparticle band at the Fermi level E_F , it would be very sensitive to the doping or lattice expansion. On the contrary, there is no special reason for the γ -coefficient increase if local electron correlation effects are the main mechanism for the enhancement.

We have measured specific heat for the Pu samples with 8%, 15%, and 20% Am, all exhibiting the *fcc* structure. A strong self-heating limits the achievable temperature range, and only for 8% Am the full low- T analysis could be performed. We used a standard Quantum Design PPMS equipment, the data were corrected for the effect of self-heating (for details see Ref. [12]). Fig. 1 reveals that no enhancement of γ comparing with existing data on Al-doped δ -Pu takes place (Al induces a lattice contraction). Depending on the approximation of lattice contri-

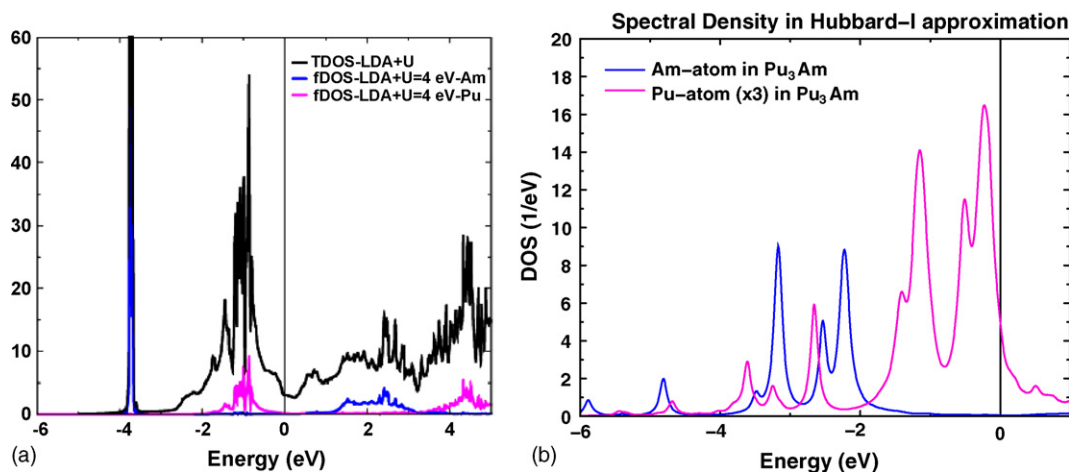


Fig. 2. (a) Density of states (states/eV) for $\text{Pu}_{0.75}\text{Am}_{0.25}$ showing the total density (TDOS) and partial f-densities for Am (sharp feature at -4 eV and broader feature at 2 eV) and Pu (occupied part around -1 eV , empty around 4 eV) obtained by LDA + U calculations. For additional details see Ref. [10]. (b) Spectral density for Pu and Am calculated in Hubbard-I (HIA) approximation starting from LDA + U ground state [A. Shick et al. unpublished (2006)].

bution, which is strongly non-Debye like, $\gamma \approx 35\text{--}55 \text{ mJ/mol K}^2$ has been obtained. For 15% Am, the γ -value is still by about 14 mJ/mol K^2 lower. Assuming that the contribution of Am states is negligible ($\gamma_{\text{Am}} = 2 \text{ mJ/mol K}^2$) we can evaluate γ per Pu that then even decreases for increasing Am content. These results suggest that the 5f states of Pu remain below E_F and many-body excitations are responsible for the γ -enhancement [13].

The reason for non-magnetic behaviour could be, as shown by LDA + U calculations (Fig. 2a), the strong spin–orbit interaction separating the nearly filled $5f_{5/2}$ band from the $5f_{7/2}$ band. The occupied part of the Pu 5f states corresponds to the number of 5f electrons $n_{5f} \approx 5.44$ for Pu (similar to δ -Pu), while the Am 5f states are very similar to those in the Am metal [10]. The main conclusion is that there is nearly no effect due to the expansion and Am dilution, since the Pu 5f states are located out from E_F in the ground state.

A direct comparison of the density of electronic states with the valence-band photoelectron spectra is not possible due to the dominance of final state effects. The spectral density obtained from DMFT-like Hubbard-I approximation (HIA) starting from LDA + U ground state shown in Fig. 2b displays close similarity to the valence-band photoelectron spectra (Fig. 3). A qualitative analysis is enough to realize from Fig. 3 that the spectra of Pu–Am alloys represent a weighted average of pure Pu and Am spectra. The same holds for the Pu 4f core-level spectra [15]. The strong similarity of the Pu spectral density calculated in HIA with DMFT calculations [13] for δ -Pu, where a high density

of quasiparticle states close to E_F occurs, explains the pattern observed in valence-band photoemission and brings arguments for the γ -enhancement, although the bare ground state DOS is clearly separated from E_F .

Unlike photoelectron spectra, magnetic properties are directly related to the ground state. If we admit that magnetic moments existing on the mean-field level (LDA + U) may be suppressed by many-body effects, the DMFT-type of theory would be still necessary. However, for other cases, the type of ground state should be linked to basic features of the electronic structure without invoking dynamic effects. An important question is therefore whether there is any systematic pattern of occurrence of magnetic order among Pu systems.

3. Pu ions diluted in a metallic matrix

A very interesting comparison of actinide (U, Np, Pu) atoms diluted in Pd was provided by Nellis and Brodsky [16,17]. U diluted in Pd is non-magnetic. A simple rigid-band model works surprisingly well assuming that the U 5f states contribute to the filling of the Pd 4d band for low concentrations. Once the 4d band is filled up (which happens at 8% U), U moments start to develop, which is manifest by, e.g., the Kondo effect in electrical resistivity. The 4d-band filling is reflected also by the fast decrease of γ (also clearly evidenced in the Th–Pd system). When the 5f states start to be occupied in U–Pd, γ starts to rise again. The complete depletion of the 5f states does not happen in the Np–Pd system, apparently due to the higher 5f count in Np compared to U. The Np doping leads to the fast suppression of the spin-fluctuation maximum in the susceptibility of Pd, but still contributes by a Curie term of $\approx 2.0 \mu_B/\text{Np}$ leading to an upturn at low T . Effective moments μ_{eff} exceeding $1.1 \mu_B/\text{Pu}$ were indicated also in the Pu–Pd system (doping up to 2% Pu). Although for higher concentrations the μ_{eff} values decrease, magnetic Pu ions clearly exist. The fast decrease of the susceptibility of the Pd matrix (much faster than in the Np–Pd system) has to be again interpreted as due to the 4d band filling. These facts support the idea that a certain reduction of the 5f count comparing to the Pu metal favours the Pu magnetic moment formation.

4. Pu in intermetallic compounds

By analogy to U, the importance of a minimum actinide spacing has been recognized as prominent precondition for the formation of magnetic moments [18]. Similar to U and Np, it is expected that for Pu systems also an additional hybridization with the valence states plays a role in the suppression of magnetism, when the large inter-plutonium spacing, $d_{\text{Pu–Pu}}$, and the small direct 5f–5f overlap would allow for magnetism. Is that all, or other reasons for magnetism suppression, besides the 5f–5f overlap and the 5f-ligand hybridization, can enter the game? Let us compare magnetic properties of U, Np, and Pu compounds, especially of the isostructural ones, i.e., with similar inter-actinide spacing and type of coordination. Generally, Np compounds exhibit (with few rare exceptions) a similar or stronger tendency to magnetism than their U counterparts. The reasons can be seen in a higher number of 5f electrons (about 4

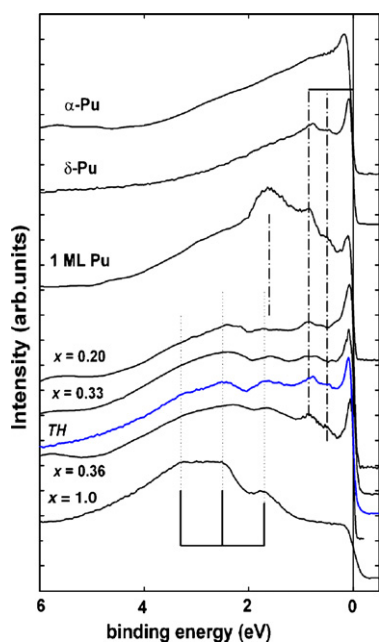


Fig. 3. UPS spectra with the HeII photoexcitation ($h\nu = 40.81 \text{ eV}$). The spectra of the $\text{Pu}_{1-x}\text{Am}_x$ layers marked by the respective x -values are compared with the spectra of pure α -Pu, δ -Pu, and 1 monolayer of Pu on Mg. The full lines mark the position of the 5f features in pure Am (at the bottom). The dash-dotted lines mark the positions of Pu-related lines. The spectrum labeled as TH, “theoretical” spectrum obtained by superposition of weighted (corresponding to the concentration of 33% Am) spectra of pure δ -Pu and Am, agrees well with the experimental spectrum for $x = 0.33$.

comparing to 3 or little less in typical U intermetallics), which makes the correlations in the *f* states more important and induces a certain contraction of the 5*f* orbitals, reducing the 5*f*–5*f* overlap for the same inter-actinide spacing. Continuing further to Pu, the tendency to magnetic order becomes much weaker and, excluding actinide Laves phases AnFe₂, which are driven by the 3*d*–3*d* coupling (PuFe₂ is ferromagnet with $T_C = 564$ K), the ordering temperatures of Pu compounds typically do not reach those of U or Np counterparts [19].

As to other Pu Laves phases with transition metals, most of them are weakly magnetic (PuMn₂, PuCo₂, PuNi₂, PuRu₂, PuIr₂). Particularly striking is the contrast of PuNi₂ with ferromagnetic UNi₂ and NpNi₂. Only PuPt₂ is ferromagnet ($T_C = 6$ K, ordered magnetic moment $\mu = 0.2 \mu_B/\text{f.u.}$) [19]. Somewhat higher ordering temperatures were found for equiatomic binaries PuSi ($T_C = 72$ K, μ_{eff} in the paramagnetic state $0.72 \mu_B$ [20], compared to $T_C = 120$ K in USi [19]) and PuPt ($T_N = 44$ K, compared to $T_C = 27$ K in UPt and $T_N = 27$ K in NpPt [19]). Important few magnetic cases also include PuPt₃ ($T_N = 40$ K, $\mu_{\text{eff}} = 1.3 \mu_B/\text{f.u.}$), PuPd₃ ($T_N = 24$ K, $\mu_{\text{eff}} = 1.0 \mu_B$) [19], and PuGa₃ ($T_C = 20$ K, $\mu = 0.2 \mu_B/\text{f.u.}$ or $T_N = 24$ K, depending on structure modification, both having $\mu_{\text{eff}} = 0.78 \mu_B$ [21]), the only exception being the antiferromagnetic PuRh₃ while URh₃ and NpRh₃ are non-magnetic spin fluctuators [19].

Easier comparison between U, Np, and Pu is possible for ternary compounds, as those (unlike binaries) form large isostructural groups but, unfortunately, for many compounds only the structure is known. Exceptions are PuNiGa and PuCoGa, which crystallize in the ZrNiAl structure type and are non-magnetic [22], whereas UNiGa, UCoGa and NpNiGa order magnetically. Remarkably, the discovery of superconductivity in PuCoGa₅ and PuRhGa₅ has initiated a broader study of the actinide “115” compounds. In the UTGa₅ group, magnetic order appears only for T = Ni, Pd, and Pt [23]. The particular cases of T = Co, Rh, Ir, which are very weak Pauli paramagnets, is clearly due to the 5*f*–*d* hybridization which is usually weak for the very late transition metals but gets stronger when moving to the left within each series [23]. The respective Np compounds exhibit different types of antiferromagnetic order (e.g., NpCoGa₅ has $T_N = 47$ K [24]). PuCoGa₅, PuRhGa₅, as well as PuNiGa₅ are non-magnetic, the first two have a superconducting ground state [25]. We can conclude that also in the “115” series the Pu compounds exhibit weaker magnetic properties than their Np and even U isotypes.

An interesting systematics can be drawn also from binaries of the rocksalt type, namely actinide monopnictides and monochalcogenides, the properties of which are well known. U and Np chalcogenides (AnS, AnSe, AnTe) are magnetic metals whereas Pu chalcogenides, despite their quite larger atomic volume, remain weakly paramagnetic semi-metals [26]. This is different from Pu pnictides (as PuSb) that are magnetic metals similar to U and Np pnictides. PuSb in particular is a very well documented case of magnetic Pu compound with extended set of microscopic information. Photoelectron spectroscopy deduced the 5*f* states out of the Fermi level and concluded their localization [27], while neutron diffraction identified a 5*f*⁵ ground state with ordered magnetic moments $0.75 \mu_B/\text{Pu}$ [28]. To test

whether a reduced 5*f* count would be obtained by the LDA + U calculations in such case, leading eventually to Pu magnetic moments, we performed the LDA + U calculations also on PuSb (both around-mean-field, AMF, and fully-localized-limit, FLL, LSDA + U). In case of AMF-LSDA + U, the hybridization with Sb electronic states leads to a lower 5*f* occupancy ($n_{5f} = 5.2$) than in, e.g., in δ -Pu and a total magnetic moment $0.76 \mu_B/\text{Pu}$ in good agreement with experiment. In case of FLL-LSDA + U, we obtained results which are similar to those of Ref. [9] yielding the magnetic moment of $0.87 \mu_B/\text{Pu}$ and 5*f* occupancy $n_{5f} = 5.0$. Our AMF-LSDA + U calculation for Pu chalcogenides (PuTe), which have one more conduction electron and less charge transfer from Pu towards the anion, gave the proper non-magnetic ground state. The FLL-LSDA + U calculations of Ref. [9] produce a magnetic solution in this case.

5. Discussion and conclusions

From the systematics of the occurrence of magnetic order or magnetic moments in diluted alloys we have tried to demonstrate that the occurrence of magnetism can be tuned not only by modifications of the Pu–Pu spacing and the 5*f*-ligand hybridization, which both destabilize magnetism in U and Np systems, but also by the tuning of the *f*-electron count in a limited range somewhere between 5*f*⁵ and 5*f*⁶. Although none of the Pu systems can be fully characterized as 5*f*⁶, already the proximity to 5*f*⁶ suppresses the magnetism in the framework of the LDA + U theory. This additional degree of freedom allows for magnetic moment formation only if the 5*f* count is reduced from the level of Pu metal. For δ -Pu, $n_{5f} \approx 5.4$ was found within the Pu atomic sphere and the state can be understood as 5*f*⁶ with isotropically distributed hole of 0.6 electron. Indeed, magnetic moment is formed typically in situations when a certain depletion of the 5*f* states can be expected due to a charge transfer, as in intermetallic compounds or alloys with strongly electronegative metals as Pd, Pt or Rh, or in partly ionic compounds as the pnictides. One should note that the 5*f*⁶ state in Am is very different from the hypothetical one in Pu. The calculations show that the 5*f*_{5/2} band in Am is below the lower edge of the conduction band and therefore only weakly hybridized while the states with the 5*f*_{5/2} character in Pu, although diving below E_F nearly completely, still hybridize with the non-*f* state and tend to form a band.

To corroborate our assumptions both experimental and theoretical effort, extending our knowledge and understanding of Pu compounds, is needed. As to experimental data, it would be tempting to conclude what is the 5*f* count on the basis of X-ray absorption (XAS) and electron energy-loss (EELS) spectroscopies following the deductions of Ref. [29]. Although the data were interpreted assuming the conventional $n_{5f} = 5$ value for Pu, it seems that these techniques do not have the capability to distinguish reliably precise absolute values of the 5*f* occupancy.

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