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Competition between Kondo and RKKY exchange couplings in $Pu_{1-x}Am_x$ alloys: Density functional theory with static Hartree-Fock and dynamic Hubbard-I approximations

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To clarify the role of the Kondo effect in screening local magnetic moments of plutonium 5*f* electrons as well as its competition to the RKKY interactions, we use a combination of density functional theory with static Hartree-Fock and dynamic Hubbard-I approximations to calculate the strength of both the Kondo exchange, J_K, and of the RKKY exchange, J_{RKKY}, couplings for $Pu_{1-x}Am_x$ system as a function of x. We find that within the range $0 \leq x \leq 1/2$, J_K increases despite that the atomic volume gets larger with the Am doping due to unexpected enhancement of hybridization between *f* and conduction electrons in the vicinity of the Fermi level. At the same time, the RKKY exchange is shown to reduce smoothly with increasing x. Our results imply that the Kondo effect should be robust against the increase in interatomic spacing of this alloy, which places this system away from quantum critical behavior.

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Magnetic behavior of metallic plutonium is a topic of hot debates in the current literature.[1,2] Naively one expects that Pu *f* shell filled with five electrons carries a total (spin +orbital) momentum, which depending on various spin-orbit coupling schemes and crystal-field effects should result in some nonzero values of magnetic moment. This has been confirmed by many state-of-the-art electronic structure calculations[3–6] based on density functional theory (DFT) in its local-density and generalized gradient approximations (LDA and GGA)[7] and by methods such as LDA+U (Refs. 8 and 9) allowing to incorporate effects of on-site correlation energy U using static Hartree-Fock-type approximations for the *f*-electron self-energies.[10] Experimentally, however, none of the six Pu crystallographic allotropes shows local-moment formation: their spin susceptibilities are temperature independent and display Pauli-like behavior;[11] specific-heat measurements indicate absence of magnetic entropy;[12] muon experiments did not detect the moment with the accuracy of $10^{-3}\mu_B$;[13] combinations of neutron elastic-scattering[14] and inelastic-scattering[15] data show no convincing evidence in existence of magnetic moments either.

The apparent discrepancy between theory and experiment can be attributed to miscounting the number of *f* electrons and can assume the true configuration to be f^6 as it was pointed using a variant of the LDA+U calculation[16] with a different prescription to determine the position of the *f* band (so-called choice of double-counting potential). However, this would produce a completely inert *f* shell and no large effective masses for the Fermi-surface electrons, which contradicts sharply with specific-heat data exhibiting an enhanced Sommerfeld coefficient.[1] Many-body based calculations[17] based on a combination of density functional and dynamical mean field[18] theories [so-called LDA +DMFT (Ref. 19)] have recently highlighted another effect: the Kondo coupling of Pu *f* electrons, which fluctuate between f^5 or f^6 atomiclike states with the conduction band made of *sd* electrons producing the Kondo singlet.

To understand this puzzle, one can try to increase Pu atomic volume in order to reduce the effect of hybridization and thus to decrease the value of the Kondo coupling J_K. As a result, if the Kondo screening mechanism is in play one expects that at some critical interatomic distance the local moment would eventually show up. Indeed most recent studies[20] of stretched plutonium lattice based on the LDA +DMFT calculations with continuous time quantum Monte Carlo (CT-QMC) method for the solution of impurity problem[21] have detected that at volumes of the order of 30% larger than the volume of the fcc δ phase the temperature behavior of spin susceptibilities turns from Pauli-like to Curie-like. To simulate this stretch experimentally, it has been suggested to dilute plutonium with americium.[22] However, after $Pu_{1-x}Am_x$ alloys have been made, studies of magnetic susceptibility, electrical resistivity, and photoelectron spectroscopy[23–25] indicated that the character of the 5*f* states does not vary with the Am doping. This came into a contradiction with the DFT based study of this system.[26]

We thus see that deducing the behavior of the Kondo exchange energy as a function of x is an interesting theoretical problem, which may shed a light on the description of the recent experimental results in the Pu-Am alloy. As a minimal model applicable for this description is the model of the Kondo lattice, it is also interesting to understand an approximate location of this system in the Doniach phase diagram[27] where the competition between Kondo and RKKY interactions may lead to exciting phenomena of quantum criticality and exotic superconductivity. This, for example, is seen in a different class of Pu-based 115 materials such as $PuCoGa_5$ and $PuRhGa_5$.[28]

In the present work, both the Kondo coupling strength, J_K, and the hypothetical magnetic interaction strength, J_{RKKY}, are calculated and compared against each other as a function of x using supercell based electronic structure framework. To deduce J_K, we measure the hybridization between the *f* level and the Fermi-surface *sd* electrons. To deduce J_{RKKY}, we use a linear-response approach[29] based on magnetic force theorem.[30] The *f*-electron self-energies are approximated by their atomic Hartree-Fock values using the LDA+U method but the results are checked against the LDA+DMFT calcu-

lations with the self-energies extracted by exact diagonalizing many-body atomic Hamiltonians.[31] The main finding of our work is an unexpected increase in J_K with Am doping due to a particular behavior of the hybridization function in the vicinity of the Fermi level and simultaneous decrease in J_{RKKY}. The latter is expected as we scale up the interatomic distances of the lattice. Our calculation shows that J_K always remains larger than J_{RKKY} assuming that the Kondo effect is robust against the increase in atomic volume of $Pu_{1-x}Am_x$, which upon doping transforms from the Kondo lattice to the diluted impurity limit. It also places this system into the heavy fermion region of the Doniach phase diagram away from the quantum critical behavior.

Our calculations are performed using the full potential linearized muffin-tin orbital (LMTO) method including relativistic effect of spin-orbit coupling.[32] An effective $U=4.5$ eV describing the on-site Coulomb repulsion among the $5f$ electrons is used while the other Slater integrals [$F^{(2)}$, $F^{(4)}$, and $F^{(6)}$] are computed from atomic physics and are subsequently rescaled to 80% of their values to account for the effect of screening.[33] Those numbers are known to give a reasonable description of the electronic structure for both Pu and Pu-Am system.[34–36] To simulate the effect of alloying in our Rapid Communication, we used supercells with four atoms corresponding to $x=0,\frac{1}{4},\frac{1}{2},\frac{3}{4},1$ in $Pu_{1-x}Am_x$ phase diagram. However the results of $x\geq\frac{3}{4}$ cannot be taken into account for J_{RKKY} because it corresponds to a ferromagnetic (FM) phase ($x=\frac{3}{4}$) and dilute impurity limit ($x\rightarrow 1$). We have also utilized experimental lattice parameters for various x.[37] Our calculated electronic structures and local magnetic moments are found to be consistent with the previous studies:[8,9,16,25,35] the $5f$ states of Am are well localized and reside in their $5f^6$ configuration, corresponding to the filled 5/2 shell; Pu $5f$ states are not fully occupied and found in configuration with 5.4 electrons, where the hole resides in the Γ_8 crystal-field level of the 5/2 shell. Around the Fermi energy the *sd* conduction electrons prevail through which Pu local moments can interact with each other via the RKKY mechanism and get screened via the Kondo effect.

In order to estimate the strength of the Kondo coupling we calculate the hybridization function between the $5f$ and the conduction states, $\Delta_{\alpha\beta}(\omega)$, which is generally expressed via the local Green's function for the f electrons as follows:[19,38,39]

$$\Delta_{\alpha\beta}(\omega)=(\omega-\epsilon_\alpha)\delta_{\alpha\beta}-G^{-1}_{\alpha\beta}(\omega)+\Sigma_{\alpha\beta}(\omega), \tag{1}$$

where

$$G_{\alpha\beta}(\omega)=\sum_{\mathbf{k}}[\omega\hat{I}-\hat{H}^{\mathbf{k}}-\Delta\hat{\Sigma}(\omega)]^{-1}_{\alpha\beta}. \tag{2}$$

Here $\hat{H}_{\mathbf{k}}$ is the single-particle LDA Hamiltonian matrix while $\Delta\hat{\Sigma}(\omega)$ is the self-energy correction that appeared in the f-electron block only with the double-counting potential after Ref. 10 was subtracted. The impurity levels ϵ_α can be found straightforwardly from $\hat{H}^{\mathbf{k}}$. The effects of nonorthogonal LMTO basis sets used in our calculation can be also taken into account.[19] Here we utilize the static Hartree-Fock approximation for the self-energy by forcing antiferromagnetically ordered state. This is done to keep the same level of accuracy with our subsequent evaluations of the RKKY interactions. We have benchmarked these calculations against the Hubbard-I approximation[31] assuming magnetically disordered solutions and find similar results. Both methods do not assume DMFT self-consistency with respect to the hybridization function and are reduced to the self-consistent determination of charge densities similar to the Kohn-Sham procedure in DFT. The imaginary part of the hybridization function taken at zero frequency determines the strength of the Kondo exchange according to a simple estimate,[40]

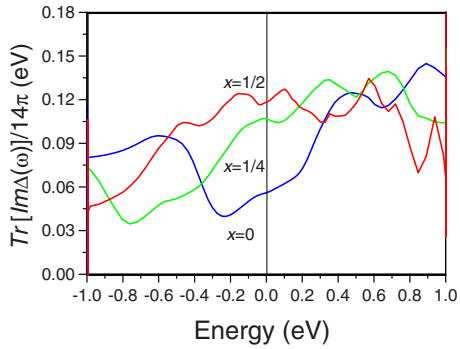

FIG. 1. (Color online) Calculated hybridization function $\mathrm{Tr}\{\mathrm{Im}\,\Delta(\omega)\}/N_d\pi$ around the Fermi level for $Pu_{1-x}Am_x$. Lines correspond to the Am doping with $x=0$, 1/4, and 1/2.

$$J_K=\frac{\mathrm{Tr}\{\mathrm{Im}\,\Delta(0)\}}{\pi N_d N(0)}\frac{U}{\epsilon_f(\epsilon_f+U)}, \tag{3}$$

where N_d is the corresponding degeneracy of the model, $N(0)$ is the density of states at the Fermi level, and $\epsilon_f=\mathrm{Tr}\{\hat{\epsilon}\}/N_d$. Thus we see that all the parameters in this expression can be evaluated in our calculation where we find that the average position of the impurity level $\epsilon_f\approx 1$ eV, the total density of states $N(0)\approx 1.5$ states/[eV atom] at the Fermi level are weakly dependent functions of Am concentration, and the trend in J_K is mainly determined by the behavior of Im $\Delta(0)$.

Figure 1 illustrates our calculated behavior of $\mathrm{Tr}\{\mathrm{Im}\,\Delta(\omega)\}/N_d\pi$ for frequencies around the Fermi level and for doping levels $x\leq\frac{1}{2}$ assuming full degeneracy $N_d=14$. Three various lines correspond to the dopings with $x=0$, 1/4, and 1/2. The following conclusions can be derived: First, all curves look very similar although there is an almost rigid shift of the order of 0.4 eV, which separates the calculated Im $\Delta(\omega)$ for various x. Second, we monitor the overall trend of decreasing the hybridization between the f and conduction electrons with the Am doping as one can trace the maximum of each plot. This is easily understood since interatomic distances get larger. However, importantly that for all doping ratios, Im $\Delta(\omega)$ has a valley and a hilltop at the right-hand side of the valley. For pure Pu, the Fermi level is located close to the dip and it gradually climbs up to the hilltop as x increases. This results in an unexpected increase in hybridization for $\omega=0$: $\mathrm{Tr}\{\mathrm{Im}\,\Delta(0)\}/N_d\pi=0.05$ eV for $x=0$; but jumps to 0.10 eV for $x=\frac{1}{4}$. Accordingly, J_K also increases because $N(0)$ remains approximately the same for all x val-

ues. If one sets $N_d=14$ in Eq. (3), then for pure Pu, J_K is 340 K but becomes 660 K for $x=\frac{1}{4}$ and further raises to 770 K for $x=\frac{1}{2}$. Then J_K decreases but is still sufficiently large as x approaches the dilute impurity limit. One thus concludes that the Kondo screening is the robust effect upon the Am doping, which would prevent the Pu moment to appear at all x.

As the description in terms of the Kondo lattice Hamiltonian may be relevant for $Pu_{1-x}Am_x$ system, its properties should be controlled by the competitions between the Kondo and RKKY exchange interactions, which—according to the Doniach phase diagram—depending on the precise value of J_K may lead to either weakly coupled magnetically ordered local-moment state or to the Kondo screened heavy fermion state in the strong coupling limit. It can even put the system in the vicinity of quantum critical point where exotic superconductivity is believed to occur. In the approximation when only a single conduction band hybridizes with the f level, J_{RKKY} scales simply as $J_K^2N(0)$ as seen by using the second-order perturbation theory for the Coqblin-Schrieffer Hamiltonian.[41] It may therefore be expected at first glance that both J_{RKKY} and J_K should behave similarly upon doping. However, in realistic situations detailed electronic structure of the material matters as various interband transitions contribute to exchange processes and this simple trend may be violated.

In order to estimate the strength of the magnetic interaction between localized $5f$ states that appeared while mapping the Pu sublattice onto the Heisenberg (pseudo) spin Hamiltonian, $H=\Sigma J_{RR'}\mathbf{S}_R\cdot\mathbf{S}_{R'}$, we utilize the magnetic force theorem within a rigid spin perturbation method.[30] In this framework, $J_{RR'}$ is given as a second-order derivative of the total energy induced by the rotations of magnetic moments at sites R and R', which can be found by calculating the following spin-susceptibility-type integral:

$$J_{RR'}^{\alpha\beta}=\frac{\partial^2 E}{\partial\phi_{\alpha R}\,\partial\phi_{\beta R'}}$$

$$=\sum_{\mathbf{q}}\sum_{\mathrm{k}jj'}\frac{f_{\mathbf{k}j}-f_{\mathbf{k}+qj'}}{\varepsilon_{\mathbf{k}j}-\varepsilon_{\mathbf{k}+qj'}}\langle\mathbf{k}j|[\sigma\times\mathbf{B}_R]_\alpha|\mathbf{k}+\mathbf{q}j'\rangle$$

$$\times\langle\mathbf{k}+\mathbf{q}j'|[\sigma\times\mathbf{B}_{R'}]_\beta|\mathbf{k}j\rangle e^{i\mathbf{q}\cdot(\mathbf{R}-\mathbf{R}')},\qquad(4)$$

Here $f_{\mathbf{k}j}$, σ, and $\mathbf{B}_R$ are the Fermi function, Pauli spin matrix, and the effective magnetic field at atom R, respectively. The latter is given by the difference in the electronic self-energies for spin-up and spin-down electrons. Since the LDA+U method is employed to recover antiferromagnetically ordered state, those became frequency independent matrices, and the evaluation of interatomic exchange interactions is straightforward.[29] In practical calculations using the supercells the total number of nearest-neighboring Js are different for different doping ratios, and we take average values after calculating all possible nearest Js.

We find that our calculated J_{RKKY} exhibits a trend opposite to J_K. It decreases as x increases as it is evident from Fig. 2. The J_{RKKY} in pure Pu ($x=0$) is smaller than J_K (134 K), which is reasonable in the sense that it is set by the scale $J_K^2N(0)$ and that from the experimental standpoint there is no

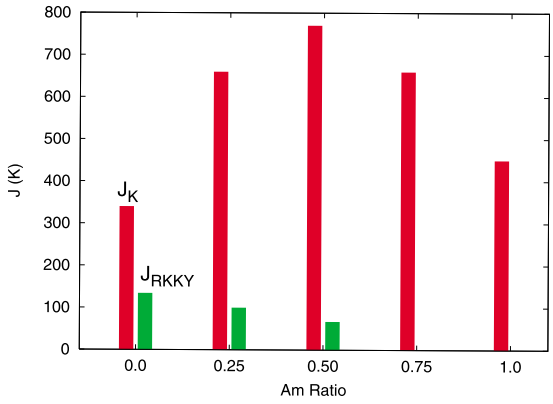

FIG. 2. (Color online) The calculated J_K and J_{RKKY} for various dopings of $Pu_{1-x}Am_x$ alloy.

local moment in Pu due to the Kondo screening. However, the behavior is quite different from a simple trend that $J_{\mathrm{RKKY}}\sim J_K^2N(0)$ as at $x=\frac{1}{4}$, $J_{\mathrm{RKKY}}=100$ K, and it becomes 67 K at $x=\frac{1}{2}$. This must be due to interband transitions presented in Eq. (4). The positive sign of J_{RKKY} refers to the AFM order by the convention in Eq. (4) and it is consistent with the assumed AFM ground state.

The comparison of these two quantities, J_K and J_{RKKY}, provides us with a clear picture for the magnetic properties of $Pu_{1-x}Am_x$. It follows that J_K is always larger than J_{RKKY} up to $x=\frac{1}{2}$, which covers up the whole range of the experiments up to now.[23–25] So, if the Kondo screening works for Pu, it should also work for the alloy. Moreover, the trend is quite suggestive as we approach the dilute limit. While we cannot extract the value of J_{RKKY} for $x\geq\frac{3}{4}$ (for $x=\frac{3}{4}$ there is only 1 Pu atom left in our supercell producing FM solution), the overall trend for J_{RKKY} to decrease is expected as the inter-Pu distances increase. It is therefore clear that J_{RKKY} would decrease further as x approaches to unity. The behavior of J_K for large x is controlled by Im $\Delta(0)$ as the Fermi level reaches the vicinity of the top point of the hybridization function as seen in Fig. 1. Therefore it is beginning to decrease slightly at values of $x\geq\frac{1}{2}$. Nevertheless, even in the dilute impurity limit—where the conduction bands are essentially made of Am sd electrons—our calculated J_K does not drop sharply as seen from Fig. 2. All this implies that the Pu-Am system is far from the quantum critical behavior and resides in the heavy fermion state.

There are possible sources of errors in our estimates. First, the calculated J_{RKKY} may be overestimated by the static approximation, such as LDA+U. This, in particular, was found in the previous studies[29,42] of transition-metal oxides where the calculated exchange interactions depending on the level of approximation for the self-energy can be further reduced by about 10%–30%. Second, the use of more refined impurity solvers and corresponding effects of the DMFT self-consistency will change our estimated values of J_K although this effect is not expected to be large due to (i) generally small values in the f-electron hybridization function and (ii) the position of the f level at around 1 eV, which is pretty far from the Fermi energy preventing the extreme sensitivity of the Kondo temperature. In fact, most recent

LDA+DMFT studies of this system based on the CT-QMC method have confirmed these conclusions.[43]

In summary, using a combination of density functional theory with self-energy corrections for the $5f$ electrons, we performed the estimates of the Kondo and RKKY exchange couplings for the whole range of dopings in $Pu_{1-x}Am_x$ alloy. It was found that J_K and J_{RKKY} exhibit opposite trends: within the range $0 \leq x \leq \frac{1}{2}$, the J_K increases—which is attributed to the details in the behavior of the hybridization function near the Fermi level—while J_{RKKY} is found to decrease as interatomic distances get larger with doping. Comparison between these two values assumes absence of quantum criticality in this system and provides a clear picture of the robust Kondo effect as the origin of its nonmagnetic behavior reported in recent experiments.

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