Application of the $S = 1$ underscreened Anderson lattice model to Kondo uranium and neptunium compounds

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Magnetic properties of uranium and neptunium compounds showing the coexistence of the Kondo screening effect and ferromagnetic order are investigated within the Anderson lattice Hamiltonian with a two-fold degenerate f level in each site, corresponding to $5f^2$ electronic configuration with $S = 1$ spins. A derivation of the Schrieffer-Wolff transformation is presented and the resulting Hamiltonian has an effective f-band term, in addition to the regular exchange Kondo interaction between the $S = 1$ f spins and the $s = 1/2$ spins of the conduction electrons. The resulting effective Kondo lattice model can describe both the Kondo regime and a weak delocalization of the $5f$ electrons. Within this model we compute the Kondo and Curie temperatures as a function of model parameters, namely the Kondo exchange interaction constant J_K, the magnetic intersite exchange interaction J_H, and the effective f bandwidth. We deduce, therefore, a phase diagram of the model which yields the coexistence of the Kondo effect and ferromagnetic ordering and also accounts for the pressure dependence of the Curie temperature of uranium compounds such as UTe.

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I. INTRODUCTION

In cerium, ytterbium, uranium, and other anomalous rare-earth and actinide compounds, the interplay between the Kondo effect and magnetism leads to the formation of various interesting phenomena which still attract a great deal of attention.[1–3] Both effects depend strongly on the hybridization between f and conduction electrons, which in turn significantly influence the level of localization of the f electrons. The extent of the localization is sensitive to various external parameters, such as temperature and pressure, but most importantly to the spatial extension of the orbitals. Actually, in the case of cerium compounds, $4f$ electrons are usually well localized, while in the case of uranium and other actinide compounds, $5f$ electrons can be either localized or itinerant, or in between, depending on the studied system. The nature of the electronic structure of actinide metals has been studied for a long time and extensively reviewed.[4–11]

The difference between the $4f$ and $5f$ electrons leads to different magnetic properties of rare-earth and actinide compounds.[12] In the case of cerium Kondo compounds, a competition between the Kondo effect on each Ce atom and the magnetic ordering of the Ce magnetic moments through the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction has been successfully described by the so-called Doniach diagram.[13] In this diagram, both the Néel temperature T_N (or the Curie temperature T_C) and the Kondo temperature T_K are obtained as functions of the Kondo exchange interaction constant J_K. In most of the Ce compounds, the magnetic ordering temperatures, T_N or T_C, are rather low, typically of the order of 5–10 K. With increasing pressure (i.e., with increasing J_K) ordering temperatures pass through a maximum and then tend toward zero at the quantum critical point, above which the systems exhibit nonmagnetic heavy fermion properties.

The situation in actinide compounds is more complex, and it is now established that some of them exhibit a coexistence of magnetic order and the Kondo effect. Indeed, this phenomenon has been observed in several uranium compounds, such as UTe,[14–16] $UCu_{0.9}Sb_2$,[17] and $UCo_{0.5}Sb_2$,[18] in which a ferromagnetic order with large Curie temperatures (equal, respectively, to $T_C = 102$ K, 113 K, and 64.5 K) and a logarithmic Kondo-type decrease of the resistivity above T_C has been experimentally detected. A similar behavior have been experimentally observed in the neptunium compounds $NpNiSi_2$[19] and Np_2PdGa_3,[20] with Curie temperatures equal to, respectively, $T_C = 51.5$ K and 62.5 K.

The origin of this fundamentally different behavior observed in uranium compounds lies in the fact that $5f$ electrons are generally less localized than $4f$ electrons and have a tendency toward partial delocalization, which in addition leads to a reduction of the magnetic moments. Moreover, $5f$ electrons often exhibit dual, localized and delocalized, behavior, and the experimental data do not clearly distinguish between a localized $5f^n$ configuration and a mixed-valence regime. Thus, it is always a challenge to decide which is the best framework for discussing actinide materials. For example, magnetic moments observed in UTe are substantially smaller than the free-ion values for either the $5f^2$ or the $5f^3$ configurations.[16] Similarly, in the series of uranium monochalcogenides, US lies closest to the itinerant side for the $5f$ electrons, USe is in the middle, and the $5f$ electrons are more localized in UTe, as evidenced by magnetization measurements.[16] Moreover, the Curie temperature of UTe passes through a maximum and then decreases with applied pressure, which is interpreted as a weak delocalization of the $5f$ electrons under pressure.[5,16] The dual nature of the $5f$ electrons has also been used by Zwicknagl *et al.*[8,9] to account for the behavior of some uranium compounds.

Another important difference between magnetic cerium and uranium compounds lies in the values of the $5f$-electron spins S, which are always larger than $1/2$ in uranium systems. (To avoid any confusion, in the following S always designates

the spin of the f-localized electrons and s the spin of the conduction electrons.) For spins S larger than $1/2$, the Kondo effect is more complex and depends on the number of screening channels; the underscreened Kondo impurity problem and more generally the multichannel impurity problem have been studied extensively[21–25] and solved exactly by the Bethe ansatz method.[26] It has been clearly established that the large spin S of the f electrons can be completely screened at $T = 0$ by spins of conduction electrons only if the number of screening channels n is equal to $2S$. If this is not the case, the problem is more complicated, as shown by Nozieres and Blandin.[21]

Here we are interested in the case of the underscreened Kondo effect, which occurs when n is smaller than $2S$; in this case, in contrast to the regular Kondo impurity case, the spin is only partially screened at low temperatures, and this leads to a reduced effective spin $S_{\rm eff} = S - n/2$. For a spin $S = 1$, this happens if there is only one active screening channel, i.e., when only one conduction band is present near the Fermi level. Indeed, for the systems studied here, this is an oversimplification: generally, all screening channels are not equivalent; thus, it is natural to consider that only one channel is coupled more strongly to the local spin. This channel will dominate the behavior, but other screening channels might play a role at lower temperatures, resulting in a two-stage Kondo screening with two Kondo temperatures.[27] Very recently, the underscreened impurity problem has been widely discussed in relation to experiments performed on quantum dots coupled to ferromagnetic leads,[28] and on molecular quantum dots (so-called molecular transistors).[29]

In a concentrated Kondo system, these reduced effective spins $S_{\rm eff} = S - n/2$ interact ferro- or antiferromagnetically through an RKKY exchange interaction, leading to magnetic ordering of the reduced moments. Thus, coexistence of magnetic ordering and the Kondo effect is expected to occur more easily in the underscreened case than in the standard $S = 1/2$ Kondo lattice.

The first attempt to describe the coexistence of ferromagnetism and the Kondo effect in uranium compounds was performed with the help of an underscreened Kondo lattice (UKL) model which considered localized f spins $S = 1$ to describe a $5f^2$ configuration of uranium ions.[30,31] This model describes the Kondo interaction, J_K, between localized $S = 1$ spins and $s = 1/2$ spins of conduction electrons, and an intersite ferromagnetic exchange interaction between the f spins, J_H. The mean-field treatment of the UKL model gives an analog of the Doniach phase diagram, and qualitatively accounts for the coexistence of ferromagnetism and the Kondo effect. However, this model is based on the assumption of localized $5f$ electrons. Of course, this assumption imposes a constraint on systems which can be described by the UKL, because, as we already discussed, many metallic actinides do not have fully localized electrons. To improve the model for these compounds, one needs to include in the UKL the possibility of $5f$-electron delocalization. This is the main goal of the present study.

This article is organized as follows: We start by considering, in Sec. II, the underscreened Anderson lattice (UAL) hamiltonian, in which the charge transfer is present from the beginning, and we transform it by using the Schrieffer-Wolff (SW) transformation[32] for $n^f_{\rm tot} = 2$, which allows for an effective f-band term, in addition to the exchange Kondo interaction between the $S = 1$ f spins and the $s = 1/2$ spins of the conduction electrons. Then, in Sec. III we present the mean-field treatment of the model, and in Sec. IV we compute the Curie and Kondo temperatures as a function of the different parameters, and in particular of the f bandwidth. We obtain new phase diagrams which could account for the pressure dependence of the Curie temperature in uranium systems such as UTe compound.

II. THE $S = 1$ SCHRIEFFER-WOLFF TRANSFORMATION

The standard model to describe the physics of the heavy fermion compounds is the periodic Anderson lattice model, whose Hamiltonian can be written as

$$H = H_c + H_V + H_f. \tag{1}$$

The first term describes a conduction c-electron band,

$$H_c = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}, \tag{2}$$

where $c^\dagger_{\mathbf{k}\sigma}$ creates a conduction quasiparticle with spin σ and momentum $\mathbf{k}$, and $\epsilon_{\mathbf{k}}$ is the energy of conduction electrons. The term H_f includes all local energy terms of the f electrons and is given by

$$\begin{aligned} H_f = \sum_{i\sigma\alpha} E^f n^f_{i\alpha\sigma} + \sum_i \big[& U\big(n^f_{i1\uparrow} n^f_{i1\downarrow} + n^f_{i2\uparrow} n^f_{i2\downarrow}\big) \\ & + U'\big(n^f_{i1\uparrow} n^f_{i2\downarrow} + n^f_{i1\downarrow} n^f_{i2\uparrow}\big) + (U' - J)\big(n^f_{i1\uparrow} n^f_{i2\uparrow} \\ & + n^f_{i1\downarrow} n^f_{i2\downarrow}\big) - J\big(f^\dagger_{i1\uparrow} f_{i1\downarrow} f^\dagger_{i2\downarrow} f_{i2\uparrow} + h.c.\big)\big], \end{aligned} \tag{3}$$

where E^f is the energy of the two-fold degenerate f level and $n^f_{i\alpha\sigma}$ is the number operator for f electrons on lattice site i, orbital α, and spin σ; U and U' are the Coulomb repulsions among electrons in the same and in the different orbitals, respectively; and J is the Hund's coupling constant. For two electrons per site, the ground state of H_f is the triplet state with $S = 1$. We assume here that this triplet state is much lower in energy than the singlet states. This is achieved if $U' - J$ is much smaller than $U' + J$ and than U. We study the SW transformation in this limit, assuming that two electrons on the same site can be coupled only in the $S = 1$ state. Both subsystems, localized f electrons and conduction band, are coupled via a hybridization term,

$$H_V = \sum_{i\mathbf{k}\sigma\alpha} (V_{\mathbf{k}\alpha} e^{i\mathbf{k}\cdot\mathbf{R}_i} c^\dagger_{\mathbf{k}\sigma} f_{i\alpha\sigma} + V^*_{\mathbf{k}\alpha} e^{-i\mathbf{k}\cdot\mathbf{R}_i} f^\dagger_{i\alpha\sigma} c_{\mathbf{k}\sigma}), \tag{4}$$

where $f^\dagger_{i\alpha\sigma}$ and $f_{i\alpha\sigma}$ are creation and annihilation operators for f electrons, carrying spin and orbital indexes σ and α ($\alpha = 1,2$), respectively.

In the SW transformation, the $f - c$ hybridization term is eliminated by a canonical transformation. Thus, using the classical method explained in Ref. 32, we start the procedure by writing the Hamiltonian as

$$H = H_0 + H_V, \tag{5}$$

where $H_0 = H_c + H_f$. Then, we look at the scattering of an initial state $|a\rangle$ to a final state $|b\rangle$ through an intermediate

state $|c\rangle$, where the states $|a\rangle$, $|b\rangle$, and $|c\rangle$ are eigenstates of H_0 and E_a, E_b, and E_c are their eigenvalues, respectively. The SW transformation consists in replacing the H_V term of the Hamiltonian by an effective interaction which is of second order in the hybridization parameter V of the starting Hamiltonian H_V. The detailed description of the calculations can be found in Refs. 1 and 32. The resulting effective Hamiltonian is given by

$$H \simeq H_0 + \tilde{H}, \tag{6}$$

where

$$\langle b|\tilde{H}|a\rangle = \frac{1}{2}\sum_c \langle b|H_V|c\rangle\langle c|H_V|a\rangle \left(\frac{1}{E_a - E_c} + \frac{1}{E_b - E_c}\right). \tag{7}$$

The SW transformation was initially performed for the case of one $4f$ electron in the $4f^1$ configuration. Here, we present a derivation of the SW transformation for the case of a $5f^2$ configuration, corresponding to an f spin $S = 1$ and to the so-called underscreened Kondo lattice model, where the $S = 1$ spins are coupled to a nondegenerate conduction band.

We study the SW transformation for two f electrons per site, allowing fluctuations of the number of f electrons between 1 and 2. Several interactions are generated by the SW transformation.[33] In the following we present the derivation of the most relevant terms, leading to both local and intersite effective interactions, namely the Kondo interaction and the effective hopping of f electrons.

A. The local effective interaction

First, we derive the $s - f$ exchange Hamiltonian for $S = 1$ localized f spins. The corresponding eigenstates of H_0 are, therefore, given by

$$\begin{aligned} |a\rangle &= c^\dagger_{\mathbf{k}\sigma'} f^\dagger_{i1\sigma} f^\dagger_{i2\sigma}|0\rangle, \\ |b\rangle &= \frac{c^\dagger_{\mathbf{k}\sigma}}{\sqrt{2}}(f^\dagger_{i1\uparrow} f^\dagger_{i2\downarrow} + f^\dagger_{i1\downarrow} f^\dagger_{i2\uparrow})|0\rangle \\ &= \frac{1}{\sqrt{2}}\sum_{\sigma'} c^\dagger_{\mathbf{k}\sigma} f^\dagger_{i1\sigma'} f^\dagger_{i2\bar{\sigma}'}|0\rangle, \\ |c\rangle &= c^\dagger_{\mathbf{k}\sigma} c^\dagger_{\mathbf{k}'\sigma'} f^\dagger_{i\alpha\sigma''}|0\rangle, \end{aligned} \tag{8}$$

and the corresponding eigenvalues are

$$\begin{aligned} E_a = E_b &= U' - J + 2E^f + \epsilon_{\mathbf{k}}, \\ E_c &= \epsilon_{\mathbf{k}} + \epsilon_{\mathbf{k}'} + E^f. \end{aligned} \tag{9}$$

The SW transformation leads to the standard Kondo-like $s - f$ exchange Hamiltonian, but here with f spins $S = 1$, and it is given by

$$\begin{aligned} H_K = \frac{1}{2}\sum_{i\mathbf{k}\mathbf{k}'} J_{\mathbf{k},\mathbf{k}'} \big[& c^\dagger_{\mathbf{k}'\uparrow} c_{\mathbf{k}\downarrow} S^-_i + c^\dagger_{\mathbf{k}'\downarrow} c_{\mathbf{k}\uparrow} S^+_i \\ & + (c^\dagger_{\mathbf{k}'\uparrow} c_{\mathbf{k}\uparrow} - c^\dagger_{\mathbf{k}'\downarrow} c_{\mathbf{k}\downarrow}) S^z_i \big], \end{aligned} \tag{10}$$

where the different components of the spin $S = 1$ read

$$\begin{aligned} S^+_i &= n^f_{i1} f^\dagger_{i2\uparrow} f_{i2\downarrow} + f^\dagger_{i1\uparrow} f_{i1\downarrow} n^f_{i2}, \\ S^-_i &= n^f_{i1} f^\dagger_{i2\downarrow} f_{i2\uparrow} + f^\dagger_{i1\downarrow} f_{i1\uparrow} n^f_{i2}, \\ S^z_i &= n^f_{i1\uparrow} n^f_{i2\uparrow} - n^f_{i1\downarrow} n^f_{i2\downarrow}, \\ n^f_{i\alpha} &= n^f_{i\alpha\uparrow} + n^f_{i\alpha\downarrow} \end{aligned} \tag{11}$$

and the corresponding exchange integral is

$$\begin{aligned} J_{\mathbf{k},\mathbf{k}'} = & -V^*_{\mathbf{k}\alpha} V_{\mathbf{k}'\alpha} e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i} \\ & \times \left(\frac{1}{U' - J + E^f - \epsilon_{\mathbf{k}'}} + \frac{1}{U' - J + E^f - \epsilon_{\mathbf{k}}}\right). \end{aligned} \tag{12}$$

This exchange integral $J_{\mathbf{k},\mathbf{k}'}$ can be easily simplified, because $\epsilon_{\mathbf{k}}$ can be restricted to values very close to the Fermi energy. Then the difference between $\mathbf{k}$ and $\mathbf{k}'$ can be neglected in the values of both $\epsilon_{\mathbf{k}}$ and $V_{\mathbf{k}\alpha}$. We also assume that the mixing parameter does not depend on the orbital index α. Thus, $J_{\mathbf{k},\mathbf{k}'}$ can be approximated in the following by

$$J_{\mathbf{k},\mathbf{k}'} \approx -\frac{2|V_{k_F}|^2}{U' - J + E^f - \mu} \equiv J_K, \tag{13}$$

where μ is the Fermi level and V_{k_F} is the value of V at the Fermi level. Eq. (13) gives the definition of the Kondo exchange interaction, J_K, that we will use in the following. It is also interesting to notice that the Kondo effect is large when the energy $U' - J + E^f$ is very close to the Fermi energy. In fact the denominator in Eq. (13) is the energy difference between the ground-state energy $U' - J + 2E^f + \epsilon_{\mathbf{k}}$ of two f electrons in $|a\rangle$ and $|b\rangle$ states and the energy of the intermediate state $|c\rangle$ with one f electron: $E^f + \epsilon_{\mathbf{k}} + \epsilon_{\mathbf{k}'}$.

B. The intersite effective interaction

Among the different terms emerging from the SW transformation, we consider in detail those that correspond to a nonlocal interaction involving two different sites i and j. In this case, the relevant initial and final states $|a\rangle$ and $|b\rangle$ are two-sites states with a total occupation of three f electrons, allowing charge fluctuations between sites i and j. Consequently, an effective f bandwidth is obtained in the second order in $V_{\mathbf{k}}$. Thus, we derive an effective band Hamiltonian H_W as the sum of three terms which arise from the SW transformation, where the sum over i and j refers to different sites:

$$H_W = H_{W1} + H_{W2} + H_{W3}, \tag{14}$$

where

$$\begin{aligned} H_{W1} = -\sum_{\mathbf{k}\alpha\sigma ij} & \frac{|V_{\mathbf{k}}|^2 e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)}}{U' - J + E^f - \epsilon_{\mathbf{k}}} (f^\dagger_{j\alpha\sigma} f^\dagger_{i1\sigma} f^\dagger_{i2\sigma} f_{j2\sigma} f_{j1\sigma} f_{i\alpha\sigma} \\ & - f^\dagger_{j\alpha\sigma} f^\dagger_{i1\sigma} f^\dagger_{i2\sigma} f_{j2\sigma} f_{j1\sigma} f_{i\bar{\alpha}\sigma} + h.c.), \end{aligned} \tag{15}$$

$$\begin{aligned} H_{W2} = -\frac{1}{2}\sum_{\mathbf{k}\alpha\sigma\sigma' ij} & \frac{|V_{\mathbf{k}}|^2 e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)}}{U' - J + E^f - \epsilon_{\mathbf{k}}} (f^\dagger_{j\alpha\bar{\sigma}} f^\dagger_{i1\sigma} f^\dagger_{i2\sigma} f_{j2\bar{\sigma}'} f_{j1\sigma'} \\ & \times f_{i\alpha\sigma} - f^\dagger_{j\alpha\bar{\sigma}} f^\dagger_{i1\sigma} f^\dagger_{i2\sigma} f_{j2\bar{\sigma}'} f_{j1\sigma'} f_{i\bar{\alpha}\sigma} + h.c.), \end{aligned} \tag{16}$$

$$\begin{aligned} H_{W3} = -\frac{1}{4}\sum_{\mathbf{k}\alpha\sigma\sigma'\sigma'' ij} & \frac{|V_{\mathbf{k}}|^2 e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)}}{U' - J + E^f - \epsilon_{\mathbf{k}}} (f^\dagger_{j\alpha\sigma} f^\dagger_{i1\sigma'} f^\dagger_{i2\bar{\sigma}'} f_{j2\bar{\sigma}''} \\ & \times f_{j1\sigma''} f_{i\alpha\sigma} - f^\dagger_{j\alpha\sigma} f^\dagger_{i1\sigma'} f^\dagger_{i2\bar{\sigma}'} f_{j2\bar{\sigma}''} f_{j1\sigma''} f_{i\bar{\alpha}\sigma} + h.c.). \end{aligned} \tag{17}$$

These three terms can be simplified in the mean-field approximation by introducing average occupation numbers $\langle n^f_{i\alpha\sigma}\rangle$. For simplicity, we will neglect all interorbital transfer terms. With these assumptions, we can see that the H_W terms can be considered as effective f-hopping terms between i and j sites with a spin-dependent hopping (see next section). If the $\mathbf{k}$ dependence of $V_\mathbf{k}$ is negligible, as is often assumed, one can deduce from Eqs. (15), (16), and (17) that there is no intersite hopping. Thus, the $\mathbf{k}$ dependence of $V_\mathbf{k}$ is at the origin of the effective f bandwidth and has to be taken into account. This $\mathbf{k}$ dependence is due to nonlocal hybridization between f and c electrons, which has no influence on the Kondo interaction but has to be taken into account for intersite interactions. In the following we write the coefficient which appears in the expression for H_W as

$$\frac{|V_\mathbf{k}|^2}{U' - J + E^f - \epsilon_\mathbf{k}} \approx -\frac{J_K}{2} g(\mathbf{k}), \tag{18}$$

where the function $g(\mathbf{k})$ includes the $\mathbf{k}$ dependence of $|V_\mathbf{k}|^2$, while the $\mathbf{k}$ dependence of $\epsilon_\mathbf{k}$ is not essential here, but can also be included.

Finally, the resulting transformed Hamiltonian contains two terms: H_K, which gives the Kondo exchange interaction for spin $S = 1$, and the important and new term H_W, which can be considered as an effective band term for the $5f$ electrons.

Thus, in addition to the Kondo interaction, we have derived here a term which leads to a finite f bandwidth; we will show that this newly introduced term gives a better description of uranium and actinide compounds where the $5f$ electrons are less localized than the $4f$ electrons in rare-earth compounds.

Besides this effective band term, there are other intersite interactions which lead to RKKY exchange, but they arise only to fourth order in hybridization $V_\mathbf{k}$. We do not compute all these terms, but, instead, we will introduce them phenomenologically as an additional intersite exchange parameter of the model, J_H.

III. THE MEAN-FIELD APPROACH

Combining all terms obtained in the preceding section, we can write the new effective Hamiltonian as

$$\mathcal{H} = H_c + H_K + H_W + \frac{1}{2} J_H \sum_{ij} \mathbf{S}_i \mathbf{S}_j. \tag{19}$$

The Heisenberg interaction J_H is considered here as a ferromagnetic exchange only between nearest neighbors. In fact RKKY interactions are long range and oscillating, but since our aim is to study the coexistence of ferromagnetism and the Kondo effect, we consider only ferromagnetic interactions. The mean-field approach has been previously described in Ref. 30. For the Kondo part H_K, it is based on a generalization of a functional integration approach described by Yoshimori and Sakurai for the single-impurity case[34] and by Lacroix and Cyrot for the $S = 1/2$ Kondo lattice case.[35,36]

Here we introduce the following mean-field parameters: the average occupation numbers $\langle n^f_{i\alpha\sigma}\rangle$, $\langle n^c_{i\sigma}\rangle$, and the "Kondo" parameter $\langle \lambda_{i\alpha\sigma}\rangle = \langle f^\dagger_{i\sigma} c_{i\alpha\sigma}\rangle$. We restrict ourselves to uniform solutions in which all these quantities are site and orbital independent, i.e., $\langle n^f_{j\alpha\sigma}\rangle = \langle n^f_{i\alpha\sigma}\rangle = n^f_\sigma$, $\langle n^c_{i\sigma}\rangle = n^c_\sigma$, and $\sum_\alpha \langle \lambda_{i\alpha\sigma}\rangle = \lambda_\sigma$.

Thus, H_W can be written as

$$H_W = \sum_{\mathbf{k}\sigma\alpha} \Gamma_{\mathbf{k}\sigma} f^\dagger_{\mathbf{k}\alpha\sigma} f_{\mathbf{k}\alpha\sigma}. \tag{20}$$

The effective band dispersion depends on spin but not on the orbital indices. Then,

$$\Gamma_{\mathbf{k}\sigma} = \Gamma_\sigma g(\mathbf{k}), \tag{21}$$

where

$$\Gamma_\sigma = -\frac{J_K}{2}\left[\left(n^f_\sigma\right)^2 + \frac{1}{2} n^f_\sigma n^f_{\bar\sigma} + \frac{1}{4}\left(n^f_{\bar\sigma}\right)^2\right] \tag{22}$$

and $g(\mathbf{k})$ is the dispersion relation for the f band. We assume, for simplicity, that the f-band dispersion is similar to the conduction electron dispersion, i.e., $g(\mathbf{k}) = P\epsilon_\mathbf{k} + P'$. We also assume that the f band should be narrower than the conduction band, so $P < 1$. The parameter P' can be included in the local energy E^f, and P is a multiplicative coefficient that can be absorbed in the definition of Γ_σ. Then we have

$$\Gamma_{\mathbf{k}\sigma} = \Gamma_\sigma g(\mathbf{k}) = A_\sigma \epsilon_\mathbf{k}, \tag{23}$$

with

$$A_\sigma = -\frac{J_K}{2} P\left[\left(n^f_\sigma\right)^2 + \frac{1}{2} n^f_\sigma n^f_{\bar\sigma} + \frac{1}{4}\left(n^f_{\bar\sigma}\right)^2\right]. \tag{24}$$

The total Hamiltonian can be now written in the mean-field approach as follows:

$$\begin{aligned}\mathcal{H} = &\sum_{\mathbf{k}\sigma} \epsilon^c_{\mathbf{k}\sigma} n^c_{\mathbf{k}\sigma} + \sum_{i\sigma\alpha} E^f_{0\sigma} n^f_{i\alpha\sigma} \\ &+ \sum_{\mathbf{k}\sigma\alpha} \Lambda_\sigma (c^\dagger_{\mathbf{k}\sigma} f_{\mathbf{k}\alpha\sigma} + h.c.) \\ &+ \sum_{\mathbf{k}\sigma} A_{\mathbf{k}\sigma} f^\dagger_{\mathbf{k}\alpha\sigma} f_{\mathbf{k}\alpha\sigma} + C,\end{aligned} \tag{25}$$

where we have

$$\begin{aligned}E^f_{0\sigma} = &E^f + U' n^f_{\bar\sigma} + (U' - J) n^f_\sigma + J_K \sigma m^c \\ &- \frac{J_K}{8}(\lambda_\uparrow + \lambda_\downarrow)^2 + J_H z \sigma M^f,\end{aligned} \tag{26}$$

$$\epsilon^c_{\mathbf{k}\sigma} = \epsilon_\mathbf{k} + \Delta_\sigma, \quad \Delta_\sigma = J_K \sigma M^f, \tag{27}$$

$$\Lambda_\sigma = -\frac{J_K}{4}(\lambda_\sigma + \lambda_{\bar\sigma}), \tag{28}$$

$$\begin{aligned}C = &-2U' N n^f_\uparrow n^f_\downarrow - (U' - J) N[(n^f_\uparrow)^2 + (n^f_\downarrow)^2] \\ &+ \frac{J_K}{2} N(\lambda_\uparrow + \lambda_\downarrow)^2 - \frac{J_H}{2} z N (M^f)^2 - J_K N m^c M^f,\end{aligned} \tag{29}$$

with $\sigma = \pm\frac{1}{2}$, $M^f = n^f_\uparrow - n^f_\downarrow$, and $m^c = \frac{1}{2}(n^c_\uparrow - n^c_\downarrow)$.

The diagonalization of the Hamiltonian gives one pure f band, $E^f_{\mathbf{k}\sigma}$, given by

$$E^f_{\mathbf{k}\sigma} = E^f_{0\sigma} + A_\sigma \epsilon_\mathbf{k}, \tag{30}$$

and two hybridized bands $E^\pm_{\mathbf{k}\sigma}$, given by

$$E^\pm_{\mathbf{k}\sigma} = \tfrac{1}{2}\left[\epsilon_\mathbf{k}(1 + A_\sigma) + E^f_{0\sigma} + \Delta_\sigma \pm S_{\mathbf{k}\sigma}\right], \tag{31}$$

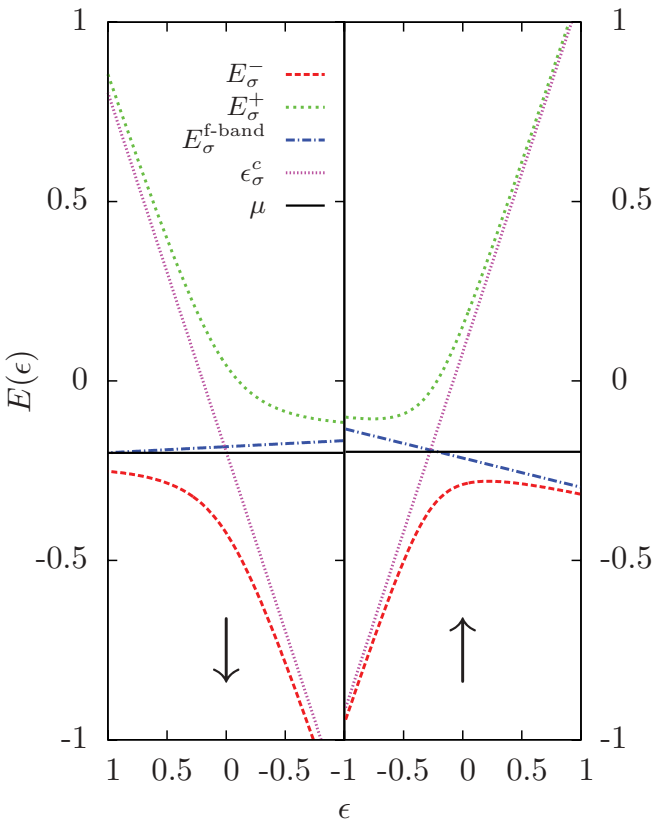

FIG. 1. (Color online) Band structure obtained in mean-field approximation, for giving the initial conduction band energy ϵ_σ^c (straight dotted line), the 3 f-bands E_σ^{f-band} (straight dashed-dotted line), E_σ^- and E_σ^+ (dotted lines), as explained in the text. The parameters used here are: $P = 0.12$, $J_K = 0.53$, $J_H = -0.01$, $n^c = 0.8$, and $n_{\text{tot}}^f = 2$.

with

$$S_{\mathbf{k}\sigma} = E_{\mathbf{k}\sigma}^+ - E_{\mathbf{k}\sigma}^- = \sqrt{\left[\epsilon_{\mathbf{k}}(1 - A_\sigma) - \left(E_{0\sigma}^f - \Delta_\sigma\right)\right]^2 + 8(\Lambda_\sigma)^2}. \quad (32)$$

In Fig. 1, we present a typical band structure resulting from the three bands $E_{\mathbf{k}\sigma}^f$ and $E_{\mathbf{k}\sigma}^\pm$ for $J_K = 0.53$ and $J_H = -0.01$. In all figures presented here, $z = 6$, where z is the number of nearest neighbors in a simple cubic lattice. In our calculations, the values of J_K and J_H are defined in units of the half-bandwidth D of the conduction band. One can see in Fig. 1 the important effect of the finite f bandwidth: The band structure is very different from that without any f bandwidth, used in Ref. 30.

IV. RESULTS AND CONCLUSIONS

In this section, we present numerical results obtained from this model, using the general method described in detail in Ref. 30: We derive the Green functions, and we calculate self-consistently the magnetization M^f for the f electrons, the magnetization m^c for the conduction electrons, and the two spin-dependent λ_σ parameters which describe the Kondo effect, by imposing constraints on the total number of f electrons and conduction electrons, $n_{\text{tot}}^f = 2$ and $n_{\text{tot}}^c = n^c$, respectively. Having solved the self-consistent equations, we study various properties of the model. The Curie and Kondo temperatures are defined, within this mean-field approach, as the temperatures at which respectively the magnetizations or the λ_σ parameters tend to zero.

As mentioned in the previous section, the half-f bandwidth derived from the SW transformation is spin dependent and is given by A_σ in Eq. (24). A_σ can be rewritten as

$$A_\sigma = -\frac{J_K P}{32}[7 + 3(M^f)^2 + 12\sigma M^f], \text{ where } \sigma = \pm\frac{1}{2} \quad (33)$$

indicating that the effective bandwidth and the magnetization are correlated. In fact it can be easily checked that the bandwidth for up spin increases with magnetization while it is the opposite for down spin; this is consistent with the double exchange process in the ferromagnetic phase, which favors itinerancy of the conduction electrons with spin parallel to the localized moment, because of intra-atomic Hund's coupling.

Here, however, we would like to explore the parameter dependence of the effective bandwidth including different possibilities for the relative variation of the Kondo coupling J_K and the f bandwidth W_f. In order to do that, we considered also the following definitions of W_f:

Case (a): a constant bandwidth: $W_f = \text{const}$.

Case (b): a bandwidth W_f proportional to the Kondo coupling constant, $W_f = QJ_K$; in this way we can take into account the effect of pressure on both the bandwidth and the Kondo coupling, since both are sensitive to the increase of hybridization under pressure.

Case (c): a bandwidth directly obtained from the SW transformation. From Eqs. (23) and (24), we get a spin-dependent bandwidth: $W_f = 2A_\sigma$.

In the following, all calculations are done assuming that the conduction band $\epsilon_{\mathbf{k}}$ has a width $2D$ and that its density of states is constant and equal to $1/2D$.

In Fig. 2, we present a plot of the temperature variation of the Kondo correlations $\lambda_\uparrow$ and $\lambda_\downarrow$, and also the f and c magnetizations, M^f and m^c, for cases (b) and (c). The parameters are $n^c = 0.8$ and $J_H = -0.01$. The upper plot is for case (b) with $Q = 0.12$, while the lower plot is for case (c) with $P = 0.12$.

The two magnetization curves clearly show a second-order magnetic phase transition at T_C, below which m^c and M^f are always antiparallel, as expected because J_K is an antiferromagnetic coupling. At low temperatures, the Kondo effect and ferromagnetism coexist, and, because of the breakdown of spin symmetry at T_C, $\lambda_\uparrow$ and $\lambda_\downarrow$ become slightly different in the ferromagnetic phase. We define the Kondo temperature as the temperature where $\lambda_\uparrow$ and $\lambda_\downarrow$ vanish. The fact that the Kondo parameter vanishes at a particular temperature is a well-known artifact of the mean-field approximation. Actually T_K is a crossover temperature, associated with the onset of Kondo screening. In all cases, the Kondo temperature T_K is larger than the Curie temperature T_C and we never found situations where $T_K < T_C$. Once ferromagnetism is established, the Kondo effect does not appear below T_C; it is blocked by the effective internal magnetic field.

To investigate the effect of the pressure on the Kondo and Curie temperatures, we computed these characteristic

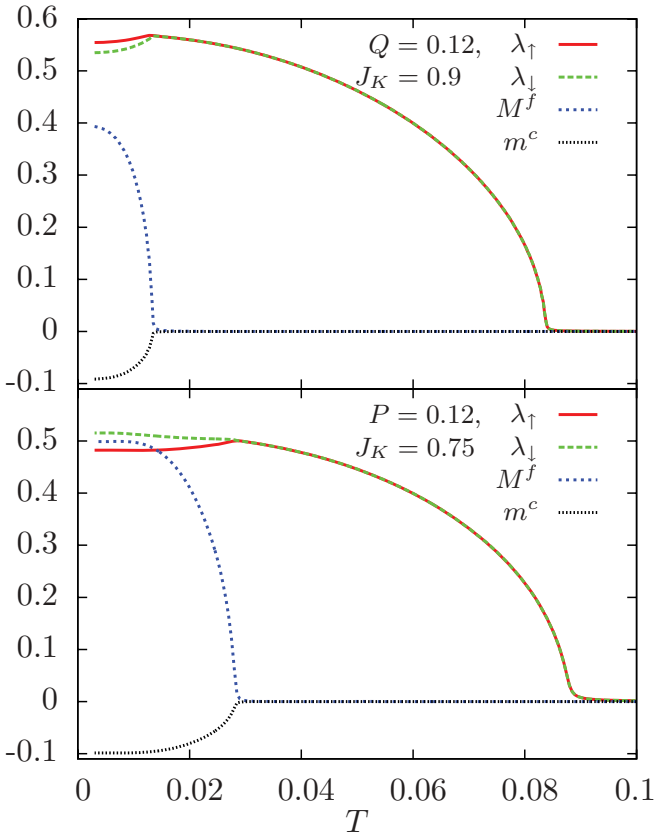

FIG. 2. (Color online) Magnetization of f electrons M^f (short dashed line), magnetization of c electrons m^c (dotted line), and Kondo parameters λ_σ (full and long dashed lines) for both spin directions for cases (b) and (c) (see text for definitions), with $J_H = -0.01$, $n^c = 0.8$, and $n^f_{\rm tot} = 2$.

temperatures for various values of J_K for fixed values of exchange interaction J_H and number of conduction electrons n_c. Figs. 3(a), 3(b), and 3(c) are obtained for the three different characterizations of the f bandwidth, cases (a), (b), and (c), respectively. We notice that the temperatures T_K and T_C are obtained as the temperatures at which the mean-field parameters (f and c magnetizations and the Kondo parameters λ) vanish. In the three cases we note that the Kondo temperature T_K becomes nonzero only above a critical value J_K^c which varies from case to case. In all cases, once nonzero, the Kondo temperature rapidly increases for larger values of J_K. The Curie temperature T_C is nonzero above a given J_K value in case (a), below a given J_K value for case (b), and is nonzero for all studied values of J_K for case (c). The reason for these different behaviors is easy to understand. In case (a) the f bandwidth is constant and the system needs a finite value of J_K to get magnetic ordering because f electrons are itinerant even for small values of J_K. In case (b) the f bandwidth increases linearly with J_K, so for low values of J_K the f electrons are localized and they are magnetic even for $J_K = 0$; thus as soon as J_K is different from zero, magnetic ordering occurs. With increasing J_K the f bandwidth also increases, and magnetism is destroyed because of the itinerant character of the f electrons. Finally, in case (c) the f bandwidth depends on both J_K and magnetization, and the dependence is different for up- and down-spin electrons; it can be seen in Fig. 3 that this complex dependence of the bandwidth leads to small variations of the Curie temperature, with a weak maximum. However, a crossing point, at which T_C and T_K are equal, is obtained in all cases.

Concerning the Kondo effect, in all three cases a peculiar behavior has been obtained for values of J_K just above this crossing point: At the temperature T_1 indicated in Fig. 3(a), 3(b), and 3(c) the Kondo parameters vanish, being nonzero only between T_1 and T_K. To better understand this behavior, in Fig. 4 we have plotted M^f, m^c, and $\lambda_\uparrow$ for $n^c = 0.8$ and $J_H = -0.01$, for values of J_K near the crossing of T_C and T_K, i.e., $J_K = 0.75$ for case (a), 0.8 for case (b), and 0.52 for case (c). It appears clearly that, with decreasing temperature, the Kondo effect occurs first, then there is a coexistence of the Kondo effect and ferromagnetism, and finally the Kondo effect disappears to yield only a strong ferromagnetism at extremely low temperatures. This behavior can be interpreted in the following way: The Kondo effect for a spin $S = 1$ cannot be complete, as explained in the introduction. Thus if exchange is large enough, the ordering of the remaining f moments occurs in the Kondo phase. However, at lower temperature, when these magnetic moments are large, they act as an internal magnetic field that destroys the Kondo effect. It should be pointed out that there is at present no experimental evidence

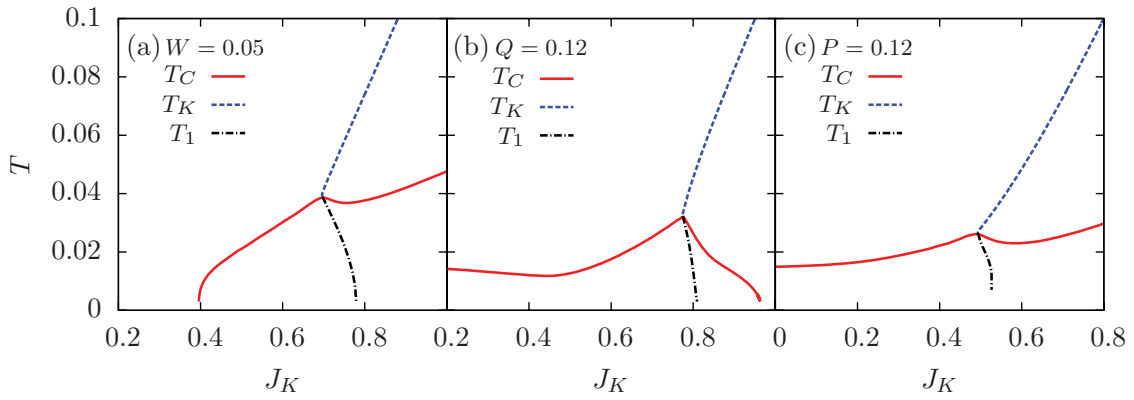

FIG. 3. (Color online) Curie temperature T_C (full line) and Kondo temperature T_K (dashed line) vs J_K for the three cases (a), (b), and (c), with $J_H = -0.01$, $n^c = 0.8$, and $n^f_{\rm tot} = 2$. For cases (a), (b), and (c), T_1 (dot-dashed line) is also shown (see text).

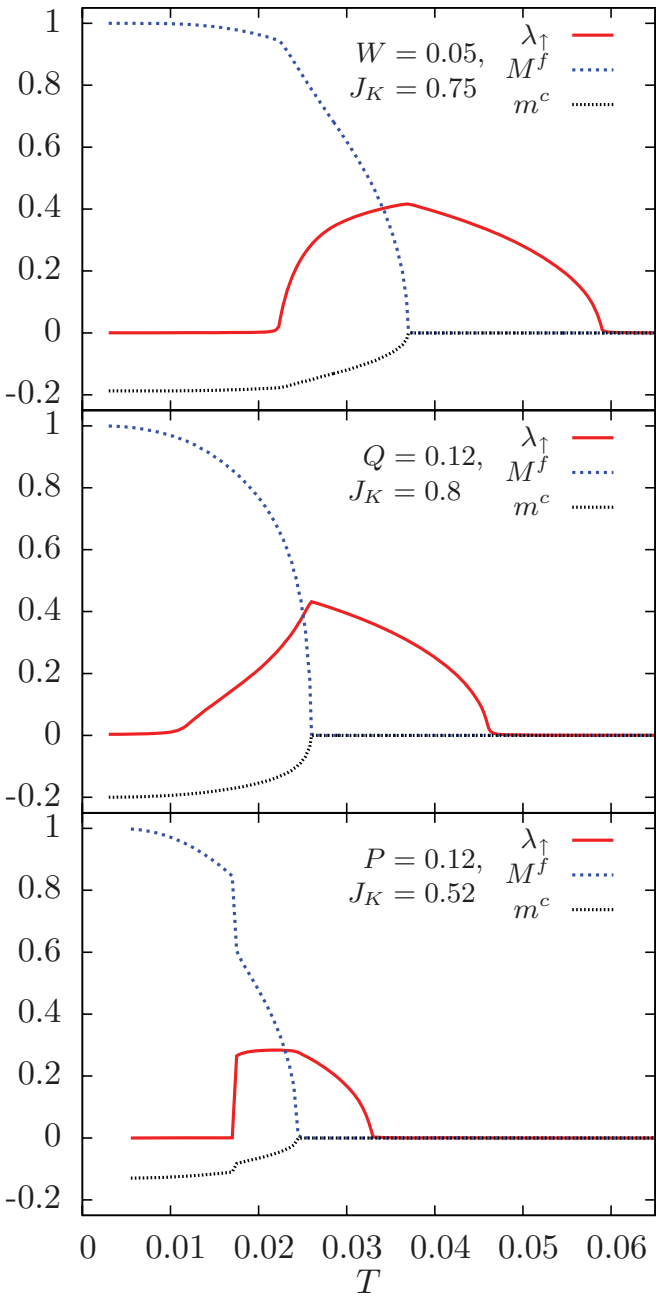

FIG. 4. (Color online) Magnetization of f electrons M^f (dashed line), magnetization of c electrons m^c (dotted line), and Kondo parameter $\lambda_\uparrow$ (full line) for cases (a), (b), and (c), with $J_H = -0.01$, $n^c = 0.8$, and $n^f_{\text{tot}} = 2$. Here $\lambda_\downarrow \approx \lambda_\uparrow$.

for or against such an effect in actinide compounds at very low temperature.

Another interesting result that can be pointed out is the decrease of the Curie temperature for large J_K above the intersection point, particularly in case (b), but also within a small range of values of J_K in case (c). This decrease can probably be considered as resulting from the "delocalization" of the $5f$ electrons. Let us also remark that J_K increases with increasing pressure and that Figs. 3(b) and 3(c) can give a description of the experimentally observed variation of T_C with pressure in UTe compound, which passes through a maximum and then decreases with applied pressure.[5,16]

To summarize, the present work improves upon the previous $S = 1$ UKL model of Ref. 30 by explicitly including the effect of a weak delocalization of the $5f$ electrons. Within this improved model, we have described new phenomena in the region where T_C and T_K are of the same order of magnitude: a possible disappearance of the Kondo effect at low temperature, which is a direct consequence of the underscreened Kondo effect, and a maximum of T_C as a function of J_K. It is worth noting that, in our model, the delocalization of the $5f$ electrons increases when J_K increases, i.e., when pressure is applied; then the magnetization decreases in the same way as the Curie temperature. Therefore, the change in the Curie temperature at large J_K is more influenced by delocalization than by competition between magnetism and the Kondo effect. This is a different result from the case of cerium compounds, where the magnetization is destroyed by the Kondo effect, i.e., by the screening of the magnetic moment. In the underscreened $S = 1$ Kondo lattice, because Kondo screening can never be complete, the Kondo effect alone does not destroy ferromagnetism.

To conclude, we have shown that our model includes two effects which are essential to describe the $5f$-electron compounds: the small delocalization of the $5f$ electrons and the $S = 1$ spins found in uranium or neptunium compounds. The first effect works against magnetism, while the second one favors magnetism. The competition between these two effects leads to complex phase diagrams which can improve the description of some actinide compounds and explain in particular the maximum of T_C observed experimentally in UTe compound with increasing pressure.

ACKNOWLEDGMENT

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