The S=1 Underscreened Anderson Lattice model for Uranium compounds

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Abstract. Magnetic properties of uranium and neptunium compounds showing coexistence of the Kondo effect and ferromagnetic order are investigated within the degenerate Anderson Lattice Hamiltonian, describing a 5f2 electronic configuration with S = 1 spins. Through the Schrieffer-Wolff transformation, both an exchange Kondo interaction for the S = 1 f-spins and an effective f-band term are obtained, allowing to describe the coexistence of Kondo effect and ferromagnetic ordering and a weak delocalization of the 5f-electrons. We calculate the Kondo and Curie temperatures and we can account for the pressure dependence of the Curie temperature of UTe.

1. INTRODUCTION

The interplay between Kondo effect and magnetism is very important in cerium, ytterbium, uranium or other anomalous rare-earth and actinide systems [1, 2]. In contrast to 4f-electrons, the 5f-electrons in actinide compounds can be either localized or itinerant or in-between, depending on the studied system [3, 4, 5, 6, 7].

The difference between the 4f- and 5f-electrons leads to different magnetic properties. In the case of cerium Kondo compounds, the competition between the Kondo effect on each Ce atom and the magnetic ordering is described by the "Doniach diagram" [8], which gives rather low ordering temperatures, typically of order 5 to 10 K. On the other hand, some uranium compounds, like UTe [9], UCu0.9Sb2 [10] or UC0.5Sb2 [11] present a ferromagnetic order with large Curie temperatures (equal respectively to 102 K, 113 K and 64.5 K) and also exhibit a logarithmic Kondo-type decrease of the resistivity above TC. Recently, a similar behavior has been observed in two neptunium compounds NpNiSi2 [12] and Np2PdGa3 [13], with TC equal to respectively 51.5 K and 62.5 K.

Thus, it is clear that Kondo effect and ferromagnetic order coexist in some uranium and neptunium compounds. However, the localization of the 5f-electrons is a difficult question and in fact the values of the magnetic moments observed for example in UTe are substantially smaller than the free-ion values for either the 5f2 or the 5f3 configurations [9]. On the same side, in the series of uranium monochalcogenides, US lies closest to the itinerant side for the 5f-electrons, US is in the middle and the 5f-electrons are more localized in UTe, as evidenced by magnetization measurements [9]. Moreover, the Curie temperature of UTe is passing through a maximum and is then decreasing with applied pressure, which is interpreted.
as a weak delocalization of the 5f-electrons under pressure [4, 9]. The dual nature of the 5f-electrons, assuming two localized 5f-electrons and one delocalized one, has been also introduced to account for the properties of some uranium compounds [5].

The coexistence between the Kondo effect and the ferromagnetism in some uranium compounds was described by the Underscreened Kondo Lattice (UKL) model which considers localized f-spins \( S = 1 \) without any f-band width [14]. Here we will present a recent work in which a finite 5f-bandwidth is introduced within the Underscreened Anderson Lattice (UAL) model, in order to describe a weak delocalization of the 5f-electrons and to account for the pressure dependence of the Curie temperature of UTe [15, 16]. In contrast with \( S = 1/2 \) case, Kondo screening in Kondo \( S = 1 \) is not complete [17] and this allows coexistence of Kondo effect and magnetism.

2. THE EFFECTIVE \( S = 1 \) HAMILTONIAN.
We start here from the UAL Hamiltonian with two f-electrons per site (in different orbitals \( \alpha = 1, 2 \)) forming a \( S = 1 \) spin. The UAL model, which is explicitly given in Ref. [15], is composed of a conduction band of energy \( \epsilon_k \), a two-fold degenerate f-level at the energy \( E_f \), a \( c - f \) hybridization term, and Coulomb interactions between f-electrons: \( U \) and \( U' \) among electrons in respectively the same and different orbitals and Hund’s coupling \( J \). Then, we use the Schrieffer-Wolff (SW) transformation [18] and we obtain an effective Hamiltonian:

\[
H = H_c + H_{cf} + H_W.
\]

The first term describes the conduction electrons, the second one is the usual Kondo exchange Hamiltonian, but here with f-spins \( S = 1 \), and it is given by:

\[
H_{cf} = \frac{1}{2} \sum_{i \sigma k k'} J_K \left[ c_{k' \sigma}^\dagger c_{k \sigma} S_i^+ + c_{k' \sigma}^\dagger c_{k \sigma} S_i^- + (c_{k' \sigma}^\dagger c_{k' \sigma} - c_{k \sigma}^\dagger c_{k \sigma}) S_i^z \right]
\]

with the different components of the spin \( S = 1 \) given by: \( S_i^\pm = n_{i11} n_{i21} - n_{i11} n_{i21} \), \( S_i^z = n_{i11} f_{i11}^\dagger f_{i11} + f_{i21}^\dagger f_{i21} - 1 \), and a similar expression for \( S_i^- \). \( J_K \) is the usual Kondo interaction integral. The third term \( H_W \) of the new derived Hamiltonian yields an effective f-band term, with a bandwidth proportional to \( J_K \). Detailed calculations can be found in Refs [15] and [16].

Besides these two terms \( H_{cf} \) and \( H_W \), we add an exchange interaction \( J_H \) between the localized spins to account for the effective intersite ferromagnetic exchange. In fact, the exchange interaction \( J_K \) can in principle yield both the Kondo effect and magnetic order, but the addition of the exchange interaction \( J_H \) is necessary to really describe the magnetic order, as already well established theoretically [2, 14, 19]. Here \( J_H \) is considered as a ferromagnetic interaction between nearest neighbor f-spins.

Thus, the total mean field Hamiltonian is:

\[
H_{tot} = \sum_{k \sigma} \epsilon_k n_{k \sigma} + \sum_{i \sigma \alpha} E_{i \sigma} n_{i \sigma \alpha} + \sum_{k \alpha \sigma} \Lambda_{\sigma}(c_{k \sigma}^\dagger f_{k \alpha \sigma} + h.c.) + \sum_{k \alpha \sigma} A_{k \sigma} f_{k \sigma}^\dagger f_{k \sigma} + C
\]

The effective f-hopping term \( A_{k \sigma} \) is given by:

\[
A_{k \sigma} = -\epsilon_k J_K \left[ (n_{\sigma}^f)^2 + \frac{1}{2} n_{\sigma}^c n_{\sigma}^f + \frac{1}{4} (n_{\sigma}^f)^2 \right],
\]

while the effective \( c - f \) hybridization is equal to \( \Lambda_{\sigma} = -\frac{J_K}{2}(\lambda_{\sigma} + \lambda_{\sigma}) \), with \( \epsilon_{k \sigma}^c = \epsilon_k + \Delta_{\sigma} \) and \( \Delta_{\sigma} = J_K \sigma M_f \).

\( E_{i \sigma} \) is the effective position of the f-level given by:

\[
E_{i \sigma} = \epsilon_i - \frac{1}{2} \sum_{k \sigma} A_{k \sigma} n_{i \sigma \alpha} - \sum_{k \sigma} \frac{1}{2} (c_{k \sigma}^\dagger c_{k \sigma} + f_{k \sigma}^\dagger f_{k \sigma})
\]
Figure 1. Plots of $T_C$, $T_K$ and $T_1$ versus $J_K$ for the three cases (a), (b) and (c), with $J_H = -0.01$, $n^c = 0.8$ and $n_{\text{tot}}^f = 2$ and the values of the different parameters given in the figures (all the temperatures and the energies are expressed in units of $D$).

\[ E_{0\sigma}^f = E_f^0 + U'n_{\bar{\sigma}}^f + (U' - J)n_{\sigma}^f + J_K\sigma m^c - \frac{J_K}{8}(\lambda_{\uparrow} + \lambda_{\downarrow})^2 + J_H z\sigma M_f. \]

and $C$ is equal to:

\[ C = -2U'Nn_{\sigma}^f + (U' - J)N[(n_{\sigma}^f)^2 + (n_{\bar{\sigma}}^f)^2] + \frac{J_K}{2}N(\lambda_{\uparrow} + \lambda_{\downarrow})^2 - \frac{J_H}{2}zN(M_f)^2 - J_KN m^c M_f, \]

with $\sigma = \pm \frac{1}{2}$, $M_f = n_{\uparrow}^f - n_{\downarrow}^f$ and $m^c = \frac{1}{2}(n_{\uparrow}^c - n_{\downarrow}^c)$.

$n_{i\alpha\sigma}^f$ is the number of $f$-electrons per site, spin and orbital $\alpha$, $n_{\sigma}^f = <n_{i\alpha\sigma}^f>$ and $n^c$ the total number of conduction electrons. The $f$ and $c$ magnetizations $M_f = n_{\uparrow}^f - n_{\downarrow}^f$, $m^c = \frac{1}{2}(n_{\uparrow}^c - n_{\downarrow}^c)$ and Kondo parameter $\lambda_{\sigma} = \langle c_{i\sigma}^\dagger f_{i\sigma} \rangle$ should be calculated selfconsistently [14, 16].

The diagonalization of the Hamiltonian gives one pure $f$-band with dispersion given by:

\[ E_{k\sigma}^{\text{band}} = E_{0\sigma}^f + A_{k\sigma}, \]

and two hybridized $f-c$ bands $E_{k\sigma}^{\pm}$ given by:

\[ E_{k\sigma}^{\pm} = \frac{1}{2}[\epsilon_k + A_{k\sigma} + E_{0\sigma}^f + \Delta_{\sigma} \pm S_{k\sigma}], \]

with:

\[ S_{k\sigma} = \sqrt{[\epsilon_k - A_{k\sigma} - E_{0\sigma}^f + \Delta_{\sigma}]^2 + 8(A_{\sigma})^2}. \]

3. RESULTS and CONCLUSIONS.

We present here the main results obtained by the present calculations. The Kondo ($T_K$) or Curie ($T_C$) temperatures are defined in the mean field approach as the temperatures at which respectively the $\lambda_{\sigma}$ parameters or the magnetizations vanish. $T_K$ depends essentially on $J_K$,
Figure 2. $M_f$, $m_c$ and $\lambda_\sigma$ versus temperature (expressed in units of $D$) for the case (b), with $Q = 0.12$, $J_H = -0.01$, $n^c = 0.8$, $n^f_{tot} = 2$, and respectively two values of $J_K = 0.9$ (Figure 2a) and $J_K = 0.8$ (Figure 2b).

while $T_C$ depends obviously on both $J_K$ and $J_H$. We consider a constant density of states for the conduction electron band with a band width $2D$, and in the following all energies and temperatures are expressed in units of $D$.

Our basic interest is here to study the effect of the finite $f$-band width $W$ on the magnetic properties. Thus, we have considered the three following cases:
- case (a) : $W$ is taken constant,
- case (b) : $W$ is taken proportional to the exchange Kondo integral, i.e. $W = Q J_K$,
- case (c) : $W$ is given by our present derivation of the SW transformation, i.e. $W = 2P A_{k\sigma}/\epsilon_k$.

Figure 1 gives both $T_K$ and $T_C$, as a function of the Kondo exchange integral $J_K$, for the three cases (a), (b) and (c) defined above, with respectively the following parameters: $W = 0.05$, $Q = 0.12$ and $P = 0.17$. For small $J_K$ values, there is no Kondo effect and $T_C$ increases continuously with $J_K$, while for large $J_K$ values, $T_K$ increases rapidly with $J_K$. Two effects have to be pointed out: (1) in some cases $T_C$ decreases for large $J_K$; (2) there is a ”peculiar behavior” of $T_K$ for the two cases (a) and (b): in a certain $J_K$ range, $\lambda$ is non-zero only between two temperatures $T_K$ and $T_1$ and vanishes below $T_1$.

Then, we present on Figure 2 two plots of $M_f$, $m_c$ and $\lambda_\sigma$ versus temperature for $n^c = 0.8$, $J_H = -0.01$ and for the case (b) with $Q = 0.12$, but for two different values of $J_K$ equal to 0.9 (Figure 2a) and 0.8 (Figure 2b). Figure 2a shows clearly that both the Kondo effect and ferromagnetism exist together for $J_K = 0.9$ down to the lowest temperatures, with $T_K$ larger than $T_C$. Figure 2b corresponding to the ”peculiar behavior” shows that for $J_K = 0.8$, with decreasing temperature, Kondo effect occurs at $T_K$, then there is a coexistence of Kondo and ferromagnetism, and finally the Kondo effect disappears at $T_1$ to yield only a strong ferromagnetism at very low temperatures. This behavior can be interpreted in the following way: Kondo effect for a spin $S = 1$ cannot be complete when there is only one screening channel [17]. Thus if exchange is large enough, the ordering of the remaining $f$-moments occurs in the Kondo phase. However, at lower temperature, when these ordered magnetic moments are large, they act as an internal magnetic field and Kondo effect is destroyed by this effective magnetic field. One should notice that there is at present no experimental evidence in favor or in contrast of such an effect in actinide compounds at very low temperatures.

Finally, the decrease of the Curie temperature observed in the cases (b) and (c) of Figure 1 for large $J_K$ values above the intersection point with $T_K$ can probably be considered as resulting from a possible effect of ”delocalization” of the $5f$-electrons. Let us also remark that $J_K$ increases with increasing pressure and that the two figures corresponding to the cases (b) and
(c) can give a good description of the experimentally observed variation of $T_C$ with pressure in UTe compound, which is passing through a maximum and then decreasing with applied pressure [4, 9].

We can conclude that the present calculation improves the previous $S = 1$ UKL model of Ref. [14]. The model described in Ref. [14] started from a localized description of the $5f$-electrons in the $5f^2$ configuration, while here we started from an Anderson Hamiltonian and derived by the Schrieffer-Wolff transformation a new Hamiltonian with a non-zero $f$-band width, in addition to the usual Kondo exchange term. This new Hamiltonian can describe the onset of the "delocalization" of the $5f$-electrons and the new curves giving a maximum of $T_C$ as a function of $J_K$ can account for the pressure dependence of $T_C$ in UTe. Thus, the present work yields an improvement with respect to the UKL model of Ref. [14] for the description of the $5f$-electrons in some uranium compounds.

References

[1] Hewson A C, The Kondo problem to Heavy Fermions, Cambridge University Press (1993).
[2] Coqblin B, AIP Conference Proceedings, volume 846, pp. 3-93 (2006).
[3] Coqblin B, Iglesias-Sicardi J R and Jullien R 1978 Contemporary Physics 19 327
[4] Sheng Q G and Cooper B R 1996 J. Magn. Magn. Mater. 164 335
[5] Zwicknagl G, Yaresko A N and Fulde P 2003 Phys. Rev. B 68 052508
[6] Moore K T and van der Laan G 2009 Rev. Mod. Phys. 81 235
[7] Santini P, Carretta S, Amoretti G, Caciuffo R, Magnani N and Lander G H 2009 Rev. Mod. Phys. 81 807
[8] Doniach S 1976 Proc. of the "Int. Conf. on Valence Instabilities and Related Narrow-band Phenomena" ed R D Parks (Plenum Press) p 168
[9] Schoenes J, Vogt O, Lohle J, Hulliger F and Mattenberger K 1996 Phys. Rev. B 53 14987
[10] Bukowski Z, Troc R, Stepien-Damm J, Sulkowski C and Tran V H 2005 J. Alloys and Compounds 403 65
[11] Tran V H, Troc R, Bukowski Z, Badurski D and Sulkowski C 2005 Phys. Rev. B 71 094428
[12] Colineau E, Wastin F, Sanchez J P and Rebizant J 2006 J. Phys.: Cond. Matter 20 075207
[13] Tran V H, Griveau J C, Eloiirdi R, Muller W and Colineau E March 2010, presented at the 40 emes Journees des Actinides, Geneva, Switzerland
[14] Perkins N B, Nunez-Regueiro M D, Coqblin B and Iglesias J R 2007 Phys. Rev. B 76 125101.
[15] Thomas C, Simoes A S R, Lacroix C, Iglesias J R and Coqblin B 2009 Physica B 404 3008.
[16] Thomas C, Simoes A S R, Iglesias J R, Lacroix C, Perkins N B and Coqblin B, to be published
[17] Nozieres P and Blandin A 1980 J, de Physique 41 193
[18] Schrieffer J R and Wolff P A 1966 Phys. Rev. 149 491
[19] Iglesias J R, Lacroix C and Coqblin B 1997 Phys. Rev. B 56 11820