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# Underscreened Kondo lattice model versus underscreened Anderson lattice model: Application to uranium compounds

B. Coqblin [a,*], J.R. Iglesias [b], N.B. Perkins [c], Acirete S. da R. Simoes [b], Christopher Thomas [b]

[a] L.P.S., UMR CNRS 8502, Université Paris-Sud, 91405 Orsay, France
[b] Instituto de Física UFRGS, 91501-970 Porto Alegre, Brazil
[c] UW – Madison, 1150 University Avenue, Madison, WI 53706-1390, USA

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ABSTRACT

We present here two theoretical models, the underscreened Kondo lattice (UKL) model and the underscreened Anderson lattice (UAL) model, which are appropriate models for a description of the coexistence of Kondo effect and ferromagnetic order experimentally observed in various uranium compounds. First we discuss a Kondo-ferromagnetic diagram (KFD), obtained in the framework of the UKL model with $S=1$ [N.B. Perkins, M.D. Nunez-Regueiro, J.R. Iglesias, B. Coqblin, Phys. Rev. B 76 (2007) 125101]. The KFD differs significantly from the analogous Doniach diagram for the regular Kondo lattice with $S=\frac{1}{2}$. While both the Kondo and Curie temperatures in KFD increase with an increase of the intrasite Kondo coupling, $J_K$, the Néel temperature in the Doniach diagram decreases with increase of $J_K$. Ferromagnetism is not suppressed by the Kondo interaction, but instead the coexistence between the two phenomena takes place. Then, we compare the obtained phase diagram with available experimental data. Indeed, the predicted behavior has been observed in some uranium compounds. However, there are certain difficulties in describing the pressure dependence of the Curie temperature of UTe, which firstly passes through a maximum and then decreases at higher pressures. This pressure dependence of the Curie temperature clearly indicates that the character of 5f electrons changes with pressure, becoming more and more itinerant. Then, the UKL model cannot capture the change of localization of the 5f electrons. In this case, the underscreened Anderson lattice (UAL) model, taking explicitly into account the possibility of the formation of a f-band, becomes more appropriate. It allows to treat the case of a non-integer number of f electrons and a finite f bandwidth. We present here the two-orbital UAL model and discuss some preliminary results.



## 1. Introduction

In the regular Kondo problem, the spin $S_f=\frac{1}{2}$ of the localized 4f electron (or hole) corresponding to the $4f^1$ (or $4f^{13}$) configuration is completely screened at very low temperatures by the spin $s_c=\frac{1}{2}$ of the conduction electrons. It is well known that, in the regular Kondo lattice, there is a competition between the Kondo effect and the magnetic order, resulting in the Doniach diagram [2], which separates Cerium (or Ytterbium) systems in the magnetically ordered ones and the non-magnetic heavy-fermion ones. This Kondo-magnetism competition in Cerium and Ytterbium compounds results in a relatively small ordering temperature, typically of order 5–10 K [3].

The situation of Uranium compounds appears to be different from that of Cerium systems. Uranium compounds like UTe [4,5], $UCu_{0.9}Sb_2$ [6] or $UCo_{0.5}Sb_2$ [7] exhibit a very different interplay between Kondo effect and magnetism, which leads to a ferromagnetic ordering at rather large Curie temperatures, equal respectively to $T_c=102$, 113 and 64.5 K for the above mentioned compounds. In addition, they also show a logarithmic Kondo-type decrease of the resistivity above $T_c$. A similar behavior has been recently observed in the Neptunium compound $NpNiSi_2$ [8] and in the d electron ferromagnetic UKL compound $Yb_{14}MnSb_{11}$ [9].

The appropriate description of the electronic structure of Uranium compounds is a challenging problem, which depends strongly on the level of localization of the 5f electrons, often being in a crossover region between localized and itinerant behavior. Consider Uranium monochalcogenides: the 5f electrons appear to be itinerant in US, more localized in UTe, and in intermediate regime in USe [5,10]. However, in many cases, the degree of localization of 5f electrons remains an open issue. As an example, there is an on-going controversy in UTe compound. Recent photoemission experiments on UTe have been interpreted as favoring itinerant magnetism [11], while previous results of ultraviolet photoemission spectroscopy [12] and X-ray photoemission spectroscopy [13] were interpreted in terms of localized

* Corresponding author. Tel.: +33 1 69156094; fax: +33 1 69156086.
*E-mail address:* coqblin@lps.u-psud.fr (B. Coqblin).

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electrons. In favor of the localized picture there are mainly the results from magnetic susceptibility experiments: magnetic moments, deduced from susceptibility, are close to the free ion values of Uranium, which implies that 5f electrons are relatively well localized in UTe [5,14].

The multichannel Kondo and Anderson Hamiltonians, for both the underscreened and overscreened cases, have been studied for a single impurity [15,16], but we have recently developed the underscreened Kondo lattice (UKL) model [1], in which two 5f electrons on each site are coupled to yield localized $S=1$ spins and we have successfully applied it to Uranium compounds in which 5f electrons are relatively well localized. In this model, the localized $S=1$ spins are coupled to a conduction band through a Kondo coupling, $J_K$, and interact among them ferromagnetically. However, clearly, the UKL model cannot be applied to Uranium compounds with less localized or itinerant 5f electrons. We suggest a new underscreened Anderson lattice (UAL) model, explicitly taking into account the possibility of f electron delocalization. This model allows to treat the case of a non-integer number of f electrons, with the occupation number $n_f = n_f^1 + n_f^2$ of two degenerate orbitals calculated as a function of the orbital f-band bandwidth, which is in any case much smaller than the conduction electron bandwidth, the hybridization $V$ between the f– and the conduction electrons and the Coulomb interaction $U$ between the f electrons.

## 2. The underscreened Kondo lattice model

The UKL model Hamiltonian is the following:

$$H = \sum_{\mathbf{k}\sigma}(\varepsilon_{\mathbf{k}} - \mu)c^{\dagger}_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma} + \sum_{i\sigma\alpha}E_o n^{f_\alpha}_{i\sigma} + J_K\sum_i \mathbf{S}_i\sigma_i + \frac{1}{2}J_H\sum_{ij}\mathbf{S}_i\mathbf{S}_j \qquad (1)$$

The first term represents the conduction band with energy $\varepsilon_{\mathbf{k}}$ and a constant density of states equal to $\frac{1}{2}D$ in the interval $[-D, D]$. The second term describes the energy of localized electrons at the energy $E_o$, which defines a parameter considered here as a fictitious chemical potential, i.e. a Lagrange multiplier for auxiliary f-fermions. The actual value of $E_o$ is fixed by a local constraint $n_f = \sum_{i\sigma\alpha} n^{f_\alpha}_{i\sigma} = 2$. The third term is the intrasite Kondo interaction described by the positive coupling constant $J_K(>0)$ and the fourth term is the ferromagnetic Heisenberg interaction $J_H<0$.

We studied the UKL model [1] in the mean field approximation by introducing the operators:

$$\hat{\lambda}_{i\sigma} = \sum_\alpha \hat{\lambda}^{\alpha}_{i\sigma} = \sum_\alpha \langle c^{+}_{i\sigma} f^{\alpha}_{i\sigma}\rangle \qquad (2)$$

and the magnetizations for both f and c electrons

$$M_i = S^z_i = \tfrac{1}{2}(n^f_{i\uparrow} - n^f_{i\downarrow}), \quad m_i = \sigma^z_i = \tfrac{1}{2}(n^c_{i\uparrow} - n^c_{i\downarrow}) \qquad (3)$$

The non-zero values of $\langle M\rangle$ and $\langle m\rangle$ describe the magnetic phase with a non-zero total magnetization, while a non-zero $\lambda_\sigma$ describes the Kondo effect and the formation of the heavy-fermion state. The detailed calculations can be found in Ref. [1]. Here we present only the plots of the Kondo temperature $T_K$ and of the Curie temperature $T_c$ as a function of the Kondo coupling constant $J_K$ for $J_H=-0.01$ and a number of conduction electrons $n_c = 0.8$. The Kondo temperature $T_K$ becomes finite only at the critical value $J_K^c \sim 0.65$ and then increases rapidly for larger values of $J_K$. On the other hand, the Curie temperature, $T_c$, is finite for all studied values of $J_K$. The two curves of $T_K$ and $T_c$ cross, slightly above $J_K^c$ and, for larger values of $J_K$, the Kondo temperature, $T_K$, is always larger than $T_c$. Indeed, the ferromagnetic order persists for all values of the ratio $J_K/J_H$, while the Kondo-ferromagnetism

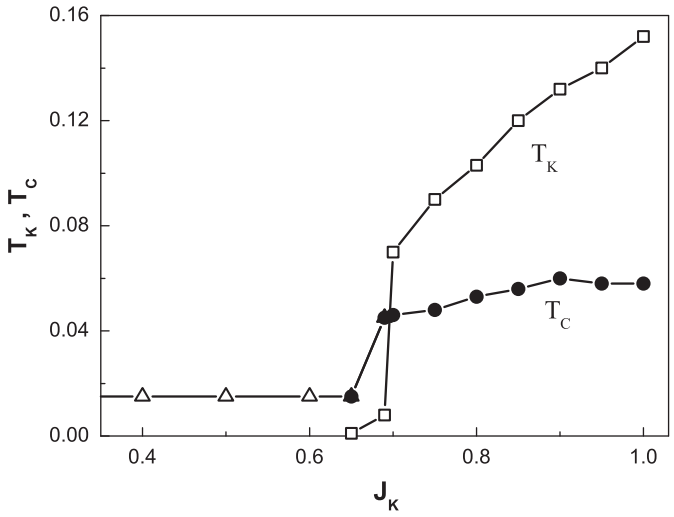


**Fig. 1.** Kondo-ferromagnetic phase diagram: plot of the Curie temperature $T_c$ and the Kondo temperature $T_K$ versus $J_K$ for $J_H=-0.01$ and $n_c=0.8$.

coexistence exists only for sufficiently large values of this ratio. The coexistence of both the Kondo effect and the ferromagnetic order has been indeed experimentally observed in various Uranium compounds, such as UTe, $UCu_{0.9}Sb_2$ or $UCo_{0.5}Sb_2$ and also in the new Neptunium $NpNiSi_2$ compound. We note that the "ferromagnetic-Kondo" diagram (KFD), shown in Fig. 1, is different from the Doniach diagram derived for the regular Kondo lattice model appropriate for Cerium or Ytterbium compounds, and represents really a new result for the study of magnetic actinide compounds.

## 3. The underscreened Anderson lattice model

However, the UKL model cannot address all the experimental observation in Uranium compounds. The study of the mono-chalcogenides UTe, USe and US, at very high pressures, revealed their very different variation of the Curie temperature with pressure [17,18]. Fig. 2 gives the experimental variation of the Curie temperature of UTe versus pressure, which presents an increase of $T_c$ up to a maximum at roughly 7 GPa and then a clear decrease with larger pressures [17,18]. On the opposite, the Curie temperature of US is continuously decreasing with pressure and $T_c$ of USe remains constant up to 10 GPa and decreases rapidly at higher pressures [17]. The initial increase of $T_c$ of UTe with pressure is clearly explained by the UKL model, since Fig. 1 shows an increase of $T_c$ with $J_K$ above the critical value $J_K^c$ and $J_K$ increases with pressure. On the other hand, the maximum and the decrease of $T_c$ at higher pressure cannot be understood in the UKL model, because a decrease of the Curie temperature is directly connected to a decrease of the 5f magnetization corresponding to a decrease of the number of 5f electrons and to an effective delocalization of 5f electrons. Similarly, the decrease of $T_c$ in US compound corresponds to a continuous delocalization of the 5f electrons, which are already itinerant at normal pressure. The case of USe is intermediate, with 5f electrons being in a crossover region between localized and itinerant behavior at normal pressure. There are several numerical studies available, among them ab initio band calculations by Sheng and Cooper [10], but the appropriate description of the electronic structure for uranium compounds at high pressures is a delicate issue.

In order to address the problem of interplay between magnetism and the Kondo effect in Uranium compounds with less localized behavior of f electron, the UKL model with a fixed

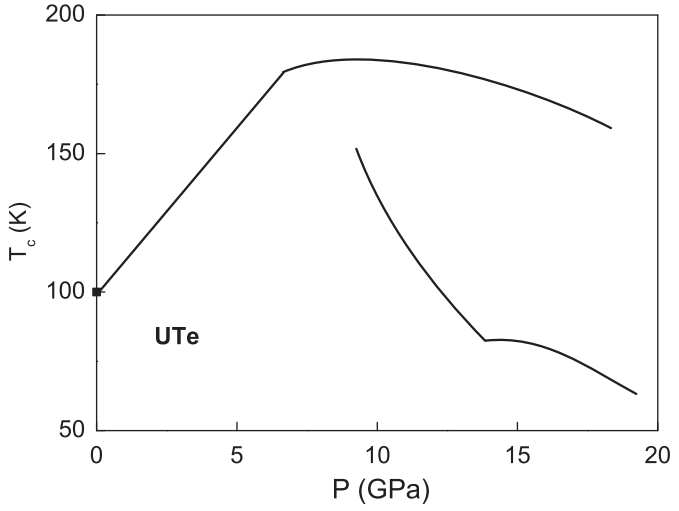


**Fig. 2.** Curie temperature $T_c$ of UTe compound versus pressure, according to Refs. [17] and [18].

total number $n_f = 2$ of 5f electrons has to be extended into a more general UAL model, which allows to treat the case of a non-integer number of f electrons. In the UAL model the occupation number $n_f = \sum_{\alpha=1,2} n_f^\alpha$ of the two degenerate orbitals can be calculated as a function of the orbital f bandwidth, of the hybridization $V$ between the f– and the conduction electrons and of the Coulomb interaction $U$ between the f electrons.

The UAL model Hamiltonian is the following:

$$H = \sum_{\mathbf{k}\sigma} \tilde{\varepsilon}_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \frac{1}{2} \sum_{ij\alpha\alpha'\sigma} E^\sigma_{ij} (f^{\alpha\dagger}_{i\sigma} f^{\alpha'}_{j\sigma} + h.c.)$$
$$+ \frac{V}{\sqrt{N}} \sum_{\mathbf{k}i\alpha\sigma} (e^{i\mathbf{k}\mathbf{R}_i} c^\dagger_{\mathbf{k}\sigma} f^\alpha_{i\sigma} + e^{-i\mathbf{k}\mathbf{R}_i} f^{\alpha\dagger}_{i\sigma} c_{\mathbf{k}\sigma}) + \sum_i [U(n^1_{i\uparrow} n^1_{i\downarrow} + n^2_{i\uparrow} n^2_{i\downarrow})$$
$$+ U'(n^1_{i\uparrow} n^2_{i\downarrow} + n^1_{i\downarrow} n^2_{i\uparrow}) + (U' - J)(n^1_{i\uparrow} n^2_{i\uparrow} + n^1_{i\downarrow} n^2_{i\downarrow})$$
$$- J(f^{1\dagger}_{i\uparrow} f^1_{i\downarrow} f^{2\dagger}_{i\downarrow} f^2_{j\uparrow} + h.c.)] \quad (4)$$

where $\tilde{\varepsilon}_{\mathbf{k}} = \varepsilon_{\mathbf{k}} - \mu$ is the energy of the conduction band defined with respect to the Fermi energy $\mu$; $\alpha$ and $\beta$ design the two f electrons. This is the usual UAL model, extended to the two-orbital case. We take here a description of the f-band to obtain a number of f electrons different from 2 and varying with the different parameters. We consider two Coulomb interactions $U$ and $U'$, the first between the two electrons with different spins on the same site inside the same orbital and the second one with different orbitals. If we want to exclude the singlet state, we should consider very high values of both $U$ and $U'$. We also take into account the exchange interactions between two f electrons on the same site, but we do not consider presently the ferromagnetic Heisenberg interaction corresponding to the last $J_H$ term of the UKL Hamiltonian [19].

We study the UAL model within the Green function (GF) technique [20,21]. From equations of motion, by straightforward but nevertheless tedious algebra, we obtain the expressions for the GFs up to fourth order, decoupling all higher terms in the non-crossing Born approximation. From equations of motion, we obtain the expressions for the Green functions, and construct the self-consistent system of equations.

We compute four mean field operators, i.e. the average magnetization $\langle M_i \rangle = \langle S^z_i \rangle$, $\langle m_i \rangle = \langle \sigma^z_i \rangle$ and the two operators $\langle c^+_{i\sigma} f^\alpha_{i\sigma} \rangle$, as in the UKL model. However, they are expressed now in higher order mean field averages.

Then we performed the calculations of the numbers $n_f$ and $n_c$ of f and conduction electrons, as in the UKL model [1]. But here, $n_f$ does not remain equal to 2 and can vary with the different parameters of the model, $E_{i\sigma}$, $V$ and $U$. The general effect of pressure is qualitatively described by decreasing $|E_{i\sigma}|$ and increasing the hybridization $V$.

In conclusion, the present UKL model explains the initial increase of the Curie temperature $T_c$ of UTe with pressure when the 5f electrons are still localized and we are presently developing the UAL model to account for the decrease of $T_c$ at higher pressures in UTe and with pressure in the other monochalcogenides, when the 5f electrons are more itinerant.

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