

# Magnetism and Kondo effect in strongly correlated electron systems

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We present here a review on three important problems of the Kondo lattice theory. First, the normal Kondo lattice effect has been extensively studied and accounts for many properties of cerium or ytterbium systems. Then, we present here a new theoretical study of the underscreened Kondo lattice problem, which can well account for the ferromagnetic - Kondo coexistence observed in some uranium compounds such as UTe. Finally, we discuss the spin glass-Kondo competition and the resulting phase diagrams showing spin glass, Kondo and magnetically ordered phases observed in disordered heavy fermion cerium alloys such as  $\text{CeCu}_x\text{Ni}_{1-x}$  alloys.

(Received April 1, 2008; accepted June 30, 2008)

**Keywords:** Kondo-effect, Strongly correlated electron systems, UTe,  $\text{CeCu}_x\text{Ni}_{1-x}$  alloys

## 1. Introduction

The physics of strongly correlated electron systems originates primarily from the presence of an inner d or f shell in one of the atoms embedded in the considered system. The d electrons in transition elements and the 4f or 5f electrons in rare-earths or actinides are generally well localized and correlated. The correlations tend firstly to favorize the existence of magnetism which is due to open inner d or f shells, as observed in pure Iron, Cobalt or Nickel in the 3d series, in the rare-earth metals and in actinide metals after Americium. Magnetism is also observed in many ionic, insulating or metallic systems containing the preceding magnetic elements.

At present, the generic term of strongly correlated electron systems is mostly reserved to systems, generally metallic, where there exists a strong interaction or hybridization between the inner d or f electrons and the conduction electrons. This type of physics has started from the experimental observation of the resistivity minimum in dilute alloys such as CuFe, CuMn, LaCe or YCe, the Friedel sum rule and the theoretical concept of " virtual bound state" [1], the derivation of the Anderson model [2] and the explanation in 1964 by Kondo [3] of the preceding resistivity minimum. At present, after the exact solution of the Kondo effect at low temperatures for a single impurity [4], there are many topics involved in the physics of strongly correlated electron systems.

First of all, the main topic concerns the so-called "heavy fermion" behaviour observed now in many cerium or other anomalous rare-earth compounds, after its first observation in  $\text{CeAl}_3$  [5]. In the "Kondo lattice", there

exists a strong competition between the Kondo single impurity behaviour and the magnetic ordering which has been firstly well described by the Doniach diagram [6], as we will discuss later on.

The metal-insulator transition, introduced by the outstanding work of Mott [7], and the study of compounds such as manganites are also a very important part of this type of physics. The discovery of superconductivity in  $\text{CeCu}_2\text{Si}_2$  compound by Steglich et al. [8] and then in some cerium compounds at high pressure and uranium compounds at normal pressure is also a very outstanding result. Strong correlations have been also proposed to be at the origin of the high-T<sub>c</sub> superconductivity which was discovered by Bednorz and Muller in 1986 [9], but this question is still not answered at present. Finally, the study of the physics of strongly correlated electron systems has considerably contributed to the development of new experimental techniques, such as high accuracy photoemission or related experiments, neutron scattering, NMR or muon spectroscopy, as well as all kinds of magnetic and transport measurements at extremely low temperatures and high pressures, as well as under very large magnetic fields.

It is well established that an anomalous behaviour is observed in many cerium, ytterbium, uranium or other anomalous rare-earth (Europium, Samarium, Praseodymium and Thulium) and actinide (Neptunium and Plutonium) systems. It is important to notice here that there are different anomalous behaviours observed in such systems, depending on the numbers of 4f electrons.

A very clear example of such different behaviours is provided by the well known and well studied phase

diagram of Cerium metal. At room temperature and normal pressure, Cerium metal is in the  $\gamma$  phase which is almost trivalent with a number of 4f electrons very close to 1 and is magnetic with a magnetization corresponding to the  $4f^1$  configuration. When pressure is applied at room temperature, Cerium undergoes a first order transition from this  $\gamma$  phase to the  $\alpha$  phase which is no more magnetic and has an "intermediate valence" with a smaller number of 4f electrons [10]. The phase diagram of Cerium presents also a critical point at higher temperature and pressure and Cerium metal becomes even superconducting at very low temperatures and high pressures. The first theoretical explanation [11] of this very peculiar phase diagram was based on the "promotion model" in which the number  $N_f$  of 4f electrons is almost 1 in the magnetic  $\gamma$  phase and decreases suddenly to a smaller and intermediate value in the non magnetic  $\alpha$  phase (or equivalently the "valence" defined here by  $4-N_f$  increases at the  $\gamma-\alpha$  phase transition). There have been many interesting other theoretical explanations of the Cerium phase diagram based on the promotion model [12] or the Mott transition [13] or the "Kondo collapse" model [14,15], which uses a mean-field approximation with an average operator to describe the Kondo correlation. In this model, the effective Kondo temperature is taken to be much larger in the  $\alpha$ -phase than in the  $\gamma$ -phase and the effective valence equal to the number of conduction electrons remains relatively small, of order 3.3, in  $\alpha$ -Ce even at very high pressures above the  $\gamma-\alpha$  transition. The  $\gamma-\alpha$  transition has been also treated more recently by band-structure calculations [16,17] or DMFT ones [18,19] and studied by many experiments, in particular by recent photoemission and resonant x-ray spectroscopy experiments [20,21]. The variation of the number  $N_f$  is much smaller at the  $\gamma-\alpha$  phase transition in these models than in the promotion model, but the decrease of  $N_f$  is still there.

Thus, the phase diagram of Cerium can be considered as a very good example of the difference between the case with an almost integer  $N_f$  value and the "intermediate valence" case. Moreover, the number of 4f electrons is often not an integer and varies with the external pressure or the matrix concentration, like in  $Eu(Pd_{1-x}Au_x)_2Si_2$  alloys [22] or  $YbInCu_4$  [23].

On the other hand, the "Kondo effect", which has been observed in many cerium or other anomalous rare-earth alloys or compounds, assumes a valence or a number of f-electrons close to an integer, but however the anomalous behaviour of the Kondo effect is due to the strong d-f hybridization. Thus, in conclusion, we can say that the anomalous behaviours observed in rare-earth or actinide strongly correlated systems can be either "intermediate valence" or "Kondo" ones.

In the present review, we will discuss only the second case in the case of a lattice. Thus, we will present firstly some main results on the Kondo lattice problem, then new

results on the Underscreened Kondo Lattice (UKL) model applied to some uranium compounds and finally we will discuss the competition between the Kondo effect and the spin glass in disordered cerium alloys.

## 2. The Kondo effect and the properties of cerium compounds

As we have said in the introduction, Kondo [3] explains in 1964 the decrease of the magnetic resistivity in  $\text{Log}T$  and, therefore, the minimum of the total electrical resistivity, by a perturbation calculation performed on the spin exchange Hamiltonian given by  $J_K s_c S_f$ . The  $\text{Log}T$  decrease of the resistivity at low temperatures has been observed in many magnetic dilute alloys such as  $\text{CuFe}$ ,  $\text{LaCe}$ ,  $\text{AuYb}$ , and also in many compounds with cerium or ytterbium or also with other anomalous rare-earths such as  $\text{PrSn}_3$  or  $\text{TmS}$  and it can be considered as the "signature" of the Kondo effect. Then, later on, an exact solution of the single impurity Kondo effect has been theoretically determined by both renormalization group technique [4] and Bethe ansatz approach [24, 25] and it results that the localized magnetic moment  $S_f=1/2$  is completely screened by the conduction electron spins  $s_c=1/2$  at  $T=0$ . At the same time, a heavy fermion behavior, characterized at very low temperatures by very large values of the electronic specific heat constant and of the coefficient of the  $T^2$  law of the electrical resistivity, has been observed in many cerium systems starting with the case of  $\text{CeAl}_3$  [5].

The single-impurity Kondo effect is valid in compounds at sufficiently high temperatures with respect to the Kondo temperature  $T_K$ , but it is no more valid at very low temperatures. In the case of a lattice, there is a strong competition between the Kondo effect and the magnetic order in many compounds or concentrated alloys containing rare-earths. This strong interplay between the Kondo effect on each site and the magnetic ordering arising from the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between rare-earth atoms at different lattice sites has been well described by the Doniach diagram [6], which gives the variation of the ordering temperature and of the Kondo temperature with increasing antiferromagnetic-like intrasite exchange interaction  $J_K$  between localized spins and conduction electron spins. The resulting ordering temperature  $T_N$  in the lattice is increasing initially with increasing  $\rho J_K$ , then passes through a maximum and tends to zero at a critical value  $\rho J_K^c$  corresponding to a "Quantum Critical Point" (QCP) in the Doniach diagram, as shown in figure 1. Such a behavior of  $T_N$  has been experimentally observed with increasing pressure in many cerium compounds, such as  $\text{CeAl}_2$  [26] or  $\text{CeRh}_2\text{Si}_2$  [27]. A similar effect has been observed in  $\text{YbCu}_2\text{Si}_2$  [28] or in other ytterbium

compounds, where the Néel temperature starts from zero at a given pressure and increases rapidly with pressure, since pressure has an opposite effect in ytterbium compounds than in cerium compounds.

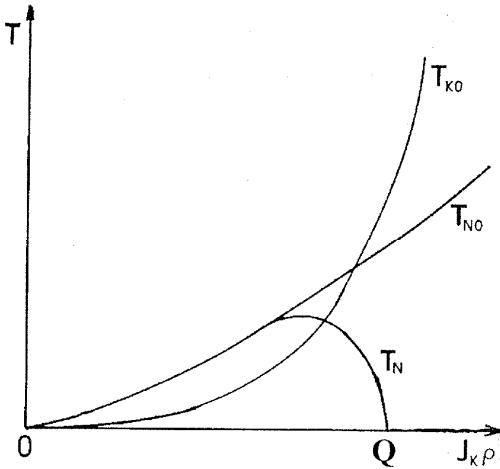


Fig. 1. The Doniach diagram:  $T_{NO}$ ,  $T_{K0}$  and  $T_N$  are respectively the single-impurity ordering temperature, the single-impurity Kondo temperature and the lattice ordering temperature.  $Q$  designs the Quantum Critical Point.

The Doniach diagram presents also the great advantage of separating cerium compounds in two categories: the first one includes cerium compounds which have a mixed Kondo-magnetic character corresponding to  $J_K$  values below the critical value of the QCP; for example,  $CeAl_2$  has together a Néel temperature of 3.85 K and a relatively large electronic constant  $\gamma$  of the specific heat equal to  $135 \text{ mJ/molK}^2$ . The second category corresponds to cerium compounds which do not order magnetically at very low temperatures and present a huge Kondo or heavy fermion character; for example,  $CeAl_3$ , which does not order magnetically even at very low temperatures, has an enormous value of  $\gamma=1600 \text{ mJ/molK}^2$ . On the other hand, the magnetic order can be antiferromagnetic, ferromagnetic in a few cases, or even a disordered one like spin glass. Detailed reviews and references can be found in refs. [29, 30, 31]. However, most of these compounds order generally in an antiferromagnetic order with a Néel temperature generally smaller than 10 K. There are a very few compounds which have both a Kondo character and a ferromagnetic order with a low Curie temperature; a recently studied compound is  $CeRuPO$  which orders ferromagnetically at  $T_c=15 \text{ K}$  and has still a relatively weak Kondo effect [32]. The ordering temperatures remain always relatively small in cerium or ytterbium Kondo

systems and we insist here on this point which will be different in the next studied case of the undercreened Kondo lattice.

The different properties of the Kondo systems have been extensively studied from both experimental [33] and theoretical sides. In particular the influence of both the Kondo effect and the crystalline field (CF) effect has been computed within the Coqblin-Schrieffer Hamiltonian [34] and has been observed experimentally in many cerium compounds [35]. It is well known that the magnetic resistivity of cerium compounds has a maximum at a temperature corresponding to the crystal field splitting  $\Delta$  and decreases as  $\text{Log}T$  above it and below it with different  $\text{Log}T$ -slopes. This behavior above the crystal field splitting, which has been firstly observed in  $CeAl_2$  and  $CeAl_3$  [35], has been observed in many other compounds and is considered now as the "signature" of the Kondo effect in cerium and ytterbium compounds, but also in alloys and compounds with Pr, Sm, Eu and Tm, such as  $PrSn_3$  [36] and  $TmS$  [37].

There is at present a great interest in the study of the thermal transport properties (thermoelectric power, thermal conductivity) of strongly correlated electron systems. The thermoelectric power of cerium compounds has been also computed in the same Kondo model and undergoes a maximum at a temperature of order a fraction of the CF splitting, typically  $\Delta/3$  [38]. The thermoelectric power of  $CeAl_3$  presents a positive maximum at roughly 60 K, in good agreement with the theoretical calculations. However, when temperature decreases, the thermoelectric power presents a negative minimum followed by a second positive maximum at 0.1 K [39]. Then, the theoretical calculation of the thermopower has been performed at low temperatures below the Kondo temperature  $T_K$  and a positive (negative) peak has been obtained in some cerium (ytterbium) compounds at a temperature typically of order  $T_K/2$  [40]. The thermoelectric power has been measured in many cerium compounds and several behaviors have been observed: the thermoelectric power of cerium compounds varies from a curve with two positive peaks separated by a negative minimum to a curve with also two positive peaks separated by a positive minimum and finally to a positive curve with only a positive peak at high temperatures [41]. A good example is provided by the variation with pressure of the thermoelectric power of the compound  $CeRu_2Ge_2$  [42, 43]. The thermal conductivity has been also computed in the same theoretical model for temperatures larger than  $T_K$  [44] and a careful study of the thermal conductivity of several anomalous rare-earth compounds shows clearly a good agreement with the Kondo and CF model [45].

Table 1. Some examples of heavy fermion compounds.

Compound	Crystal Structure	CF (in K)	T <sub>N</sub> (in K)	$\gamma(mJ/molK^2)$
<i>CeAl</i> <sub>3</sub>	Hexagonal	60-90	-	1600
<i>CeCu<sub>2</sub>Si<sub>2</sub></i>	Tetragonal	140-360	-	1000
<i>CeCu</i> <sub>6</sub>	Orthorhombic	100-240	-	1500
<i>CeRu<sub>2</sub>Si<sub>2</sub></i>	Tetragonal	220	-	350
<i>CeInCu</i> <sub>2</sub>	Cubic ( $\Gamma_7$ )	90	-	1200
<i>CeCu<sub>4</sub>Ga</i>	Hexagonal	100	-	1800
<i>CeAl</i> <sub>2</sub>	Cubic ( $\Gamma_7$ )	100	3.85	135
<i>CeB</i> <sub>6</sub>	Cubic ( $\Gamma_8$ )	500	3.2	300
<i>CeRh<sub>2</sub>Si<sub>2</sub></i>	Tetragonal	150	36	23
<i>Ce<sub>3</sub>Al<sub>11</sub></i>	Orthorhombic	100	6.2Ferromagnetic $T_c$	120
<i>CeIn</i> <sub>3</sub>	Cubic ( $\Gamma_7$ )	100	10	140
<i>CeAl<sub>2</sub>Ga<sub>2</sub></i>	Tetragonal	65-120	8.5	80
<i>CeCu</i> <sub>2</sub>	Orthorhombic	200	3.5	82
<i>CeCu<sub>2</sub>Ge<sub>2</sub></i>	Tetragonal	200	4.15	100
<i>Ce<sub>2</sub>Sn<sub>5</sub></i>	Orthorhombic	70-155	2.9	380
<i>YbCu<sub>4</sub>Ag</i>	Cubic	45	-	245
<i>YbBiPt</i>	Cubic	-	-	8000
<i>YbNi<sub>2</sub>B<sub>2</sub>C</i>	Tetragonal	40-200	-	530
<i>YbCu<sub>2</sub>Si<sub>2</sub></i>	Tetragonal	216	-	135
<i>YbNiAl</i>	Hexagonal	35	2.9	350
<b>Error!</b>	Hexagonal	-	6	500
<i>UPd<sub>2</sub>Al<sub>3</sub></i>	Hexagonal	-	14.3	150
<i>UNi<sub>2</sub>Al<sub>3</sub></i>	Hexagonal	-	4.6	120
<i>NpSn</i> <sub>3</sub>	Cubic	-	9.5	240

Several problems of strongly correlated electron systems are extensively studied at present, including the Non Fermi Liquid (NFL) behaviors or the occurrence of superconductivity close to the Quantum Critical point (QCP). In particular, the spin fluctuation approach [46] and a disorder-driven model [47] have been used to account theoretically for different NFL behaviours observed experimentally in cerium or uranium systems, as reviewed in refs. [31,48]. On the other hand, superconductivity has been observed around the high pressure QCP in several cerium compounds or at normal pressure in some uranium compounds like *URu<sub>2</sub>Si<sub>2</sub> [49].*

### 3. The Kondo lattice problem and band filling effects

The Kondo lattice model has been extensively studied, as given in different reviews [29, 31]. The Kondo-lattice

model has been firstly studied with only the intrasite Kondo interaction by different methods, the mean-field approximation [50], the renormalization group technique for the one-dimension lattice [51, 52] or the scaling theory [53]. The influence of the number  $n_c$  of conduction electrons and in particular the disappearance of the Kondo effect with  $n_c$ , called the "exhaustion" principle, have been firstly described by Nozieres [54, 55]. The Anderson-lattice model has been also studied [56, 57] and it is interesting to note that taking into account lattice effects is really important to account for some experiments, as for example, photoemission experiments [58, 59].

Now, we describe in more detail our study of the Kondo-lattice model within the mean-field approximation [60]. We have introduced a Kondo-lattice model with both an intrasite Kondo exchange interaction and an intersite magnetic exchange interaction between neighbouring localized spins. Then, in all these methods used to describe

both the Kondo effect and either short-range magnetic correlations or a given magnetic order (which could be an antiferromagnetic, a spin glass ...), we use two types of operators, describing the Kondo correlations or the magnetic correlations. In our first study [61], we have used a mean-field approximation with two correlators,  $\lambda$ , describing the intrasite Kondo correlation, and  $\Gamma$ , representing an intersite correlation between two neighboring moments.

Thus, the proposed Hamiltonian of the system is, therefore, given by:

$$H = \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} n_{\mathbf{k}\sigma}^c + \sum_{i\sigma} E_0 n_{i\sigma}^f + J_K \sum_i \mathbf{s}_i \cdot \mathbf{S}_i + J_H \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where  $\varepsilon_{\mathbf{k}}$  is the energy of the conduction band,  $J_K$  ( $>0$ ) is the Kondo coupling between the localized spin  $\mathbf{S}_i$  and the spin  $\mathbf{s}_i$  of a conduction electron at the same site and  $J_H$  is the interaction between nearest-neighboring localized spins. We consider here the case of choosing  $J_H$  to be positive, implying that intersite interactions are antiferromagnetic, as it is the case of most Cerium compounds, as shown in table 1.

In order to discuss the Kondo effect and magnetic correlations, we define the two correlation operators:

$$\lambda = \langle c_{i\sigma}^+ f_{i\sigma} \rangle, \quad \Gamma = \langle c_{i\sigma}^+ f_{i\sigma} \rangle \quad (2)$$

where  $\lambda$  describes the intrasite Kondo correlation and  $\Gamma$  represents an intersite correlation between two neighboring atoms. We have then performed an extended mean-field approximation, introduced by Coleman and Andrei [62] and presented in full detail in Ref. [60, 61]

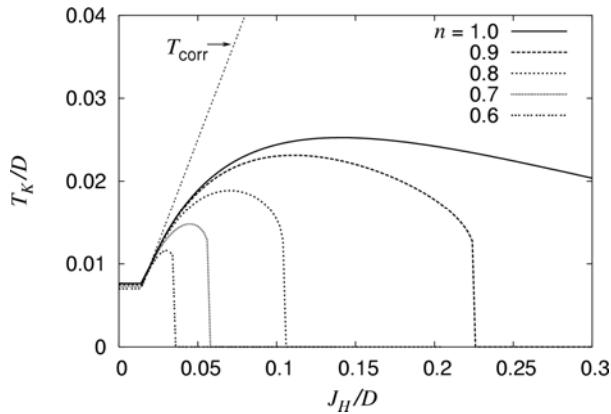


Fig. 2. Plot of the Kondo temperature  $T_K$  versus  $J_H$  for  $J_K/D=0.4$  and several values of the number  $n$  of conduction electrons. The correlation temperature  $T_{\text{corr}}$  is also shown [30].

Without entering details which can be found in refs. [30, 60], we present the results of the Kondo-lattice problem at finite temperatures, for the general case  $n_c < 1$ . The number of  $f$  electrons  $n_f$  is always taken equal to 1. The values of  $\lambda$  and  $\Gamma$  are determined by self-consistently solving the equations or by minimizing the free energy [60]. In our mean-field approximation,  $T_K$  and  $T_{\text{cor}}$  are defined as the temperatures at which respectively  $\lambda$  and  $\Gamma$  become zero.

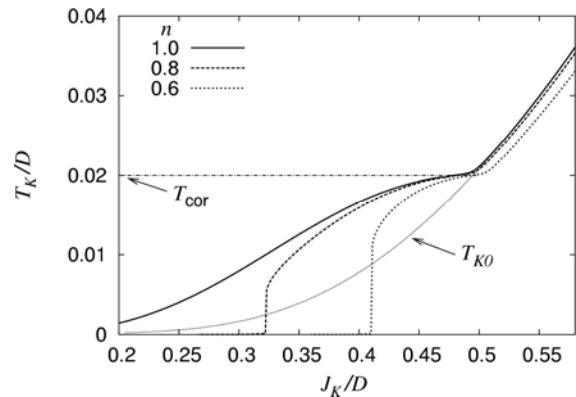


Fig. 3. Plot of the Kondo temperature  $T_K$  versus  $J_K$  for  $J_H/D=0.04$  and several values of the number  $n$  of conduction electrons. The correlation temperature  $T_{\text{corr}}$  and the single-impurity Kondo temperature  $T_{K0}$  are also shown [30].

Fig. 2 gives the curves of the Kondo temperature  $T_K$  versus  $J_H$  for a given  $J_K$  value and several values of the conduction band-filling  $n_c$ . We see clearly that  $T_K$  first increases and then decreases with  $J_H$  for fixed  $n_c$ , dropping abruptly to zero at some critical value of  $J_H$ . On the other hand, for a given  $J_H$ ,  $T_K$  decreases rapidly as  $n_c$  departs from half filling. We have also plotted  $T_{\text{corr}}$  which is linear with  $J_H$ , independently of the considered value of  $n_c$  [60, 61].

Fig. 3 gives  $T_K$  as a function of  $J_K$  for  $J_H/D=0.04$  and representative values of  $n_c$ , as well as the correlation temperature  $T_{\text{corr}}$ , which signals the onset of short-range magnetic correlations when the temperature is lowered at fixed  $J_K$ . For comparison, we plot the single-impurity Kondo temperature  $T_{K0}$ , which varies exponentially with  $J_K$  and is weakly dependent on  $n_c$  near half-filling.

Figs. 2 and 3 show some interesting results of our model. First, one can see the occurrence of short-range magnetic correlations above the Kondo temperature, in good agreement with experiment [60,63]. But the most noticeable feature of Figs. 2 and 3 is the almost catastrophic suppression of the Kondo effect with increasing intersite coupling, and the enhancement of this behavior as the band-filling factor is reduced. We also see that the Kondo effect disappears when  $n_c$  decreases below a critical value, which can be interpreted as a manifestation of the “exhaustion” principle introduced by Nozieres [54].

In the limit  $J_H=0$  and  $J_K/D \ll 1$ , we obtain :

$$T_K = C_0 D \sqrt{n_c(2-n_c)} \exp\left(-\frac{1}{\rho J_K}\right), \quad (3)$$

where  $C_0$  is a numerical constant:  $C_0=1.1337$ . Equation (3) gives  $T_K$  depending on the band filling as  $\sqrt{n_c(2-n_c)}$ , which corresponds to a  $\sqrt{n_c}$  dependence when  $n_c \rightarrow 0$ , as also found by Burdin [55].

We have shown that the temperature  $T_{\text{cor}}$  for the occurrence of short-range magnetic correlations lies above the Kondo temperature  $T_K$  for large  $J_H$  values, while  $T_{\text{cor}}=T_K$  for small  $J_H$  values. The theoretical result giving a temperature  $T_{\text{cor}}$  larger than  $T_K$  has been observed by neutron scattering experiments in some Cerium compounds [63].

Finally, it has been established, for  $n_c=1$  [61] and then for  $n_c < 1$  [60], that the Kondo temperature for the lattice can be significantly different and in fact generally smaller than the single-impurity one. The interpretation of the experimental results is not easy, but it was suggested experimentally in  $\text{CeRh}_2\text{Si}_2$  [27],  $\text{CeRu}_2\text{Ge}_2$  [64, 65] and  $\text{Ce}_2\text{Rh}_3\text{Ge}_5$  [66] that the Kondo temperature is smaller than the single impurity Kondo temperature  $T_{K0}$  and does not present an exponential variation as expected for  $T_{K0}$ .

On the other hand, it was observed that the lattice Kondo temperature can have the same behaviour as  $T_{K0}$  in many cerium and all known ytterbium compounds. For the case of ytterbium compounds, we can argue that the 4f electrons are more localized in Ytterbium than in Cerium, which results in smaller  $J_H$  values for Yb and this idea can explain the fact that the Kondo temperature of Yb compounds has the same behavior as the single impurity one. Further experiments are needed to better understand the conditions yielding a Kondo temperature for the lattice different from the single-impurity one. This interesting issue has also been addressed by different theoretical approaches of both the Kondo lattice and the Periodic Anderson model [55, 56, 67].

#### 4. The Underscreened Kondo lattice applied to uranium compounds

Then, we will discuss here a recent work on the underscreened Kondo lattice. Detailed results can be found in refs. [67, 68]. By definition, the underscreened Kondo model corresponds to the situation where the  $S_f$  localized spins are larger than the  $s_c=1/2$  spins and cannot be completely screened at  $T=0$ . An exact solution has been derived for the undercreened and overscreened single impurity [69], but in fact the undercreened Kondo lattice (UKL) has not been very often studied. Indeed, there is no exact solution for the Kondo lattice and we will present here a mean field solution which is able to describe the physics of some Uranium compounds.

In fact, the situation of Uranium alloys and compounds is complex and quite different from that of rare-earth systems, because the 5f electrons are much less localized than the 4f electrons of rare-earths, as seen for example in the actinide series where magnetism occurs only for Curium in the middle of the series. It is well known that Uranium compounds can present a great variety of different behaviours, from magnetic ones to non magnetic ones, with in particular many actinide metals or compounds which have been well accounted for by a spin fluctuation description of the 5f electrons [70].

We will discuss here the case of some uranium compounds, like  $\text{UTe}$  [71, 72],  $\text{UCu}_{0.9}\text{Sb}_2$  [73] or  $\text{UCo}_{0.5}\text{Sb}_2$  [74] which present a ferromagnetic ordering at very large Curie temperatures (equal respectively to  $T_c = 102\text{K}$ ,  $113\text{K}$  and  $64.5\text{K}$ ) and show also a LogT Kondo-type decrease of the resistivity above  $T_c$ .

The 5f electrons in uranium compounds are in a crossover region between localized and itinerant behavior. It is often difficult to decide, on the basis of the experimental data, between a local Kondo behavior corresponding to a  $5f^n$  configuration and a mixed-valence situation. One example is provided by uranium monochalcogenides:  $\text{US}$  lies closest to the itinerant side for the 5f-electrons,  $\text{USe}$  is in the middle and  $\text{UTe}$  is the closest to the localized side [75, 76, 77]. Recent photoemission experiments on  $\text{UTe}$  have been interpreted as favoring itinerant magnetism [78], but the magnetic moments deduced from magnetic susceptibility experiments in this compound are close to the free ion values of uranium, which implies that the 5f electrons are relatively well localized in  $\text{UTe}$  [72, 75]. Moreover, the dual nature of the 5f electrons, assuming two localized 5f electrons and one delocalized one, has been considered by Zwicknagl et al. [79, 80] who have obtained by band calculations a mass enhancement factor in good agreement with experiment in  $\text{UPt}_3$  and  $\text{UPd}_2\text{Al}_3$  and by Schoenes et al. [75, 76] who have carefully analyzed the variation of the localization of the 5f-electrons with concentration and pressure in diluted  $\text{US}$  and  $\text{UTe}$ . The electronic structure of uranium and plutonium monochalcogenides has been also studied by DMFT calculations [81]. So, the appropriate

description of the electronic structure for uranium compounds is a challenging problem and depends strongly on the considered system. Here we restrain ourselves to the study of the UKL model applied to uranium compounds such as  $UTe$ , when the uranium ions are relatively well localized and can be correctly described within a  $5f^2$  configuration, in which the two  $5f$ -electrons are bound into a spin  $S=1$  [82].

In order to describe the underscreened Kondo lattice, we use the following Hamiltonian [67, 68]:

$$H = \sum_{ks} (e_k - m) c_{ks}^\dagger c_{ks} + \sum_{isa} E_a n_{is}^{f_a} + J_K \sum_i S_i \sigma_i + \frac{1}{2} J_H \sum_{ij} S_i S_j \quad (4)$$

The third term is the intrasite Kondo interaction described by the coupling constant  $J_K$  and the fourth term is the intersite magnetic interaction described by  $J_H$ . The localized spins are here  $S=1$  which come from the two electrons describing the configuration  $f^2$  and the  $\alpha=1,2$  notation corresponds to these two electrons [82].

We have recently developed the underscreened Kondo lattice (UKL) model starting from the Hamiltonian (4) with two  $5f$  electrons [67, 68] and we have computed in the mean field approximation the magnetizations of the localized and conduction electrons and the Kondo correlations (taken here as  $\langle f_{i\alpha\sigma}^+ c_{i\sigma} \rangle$ ) for the two spin directions.

First, we present in Fig. 4 the temperature variation of the Kondo correlations  $\lambda_\uparrow$  and  $\lambda_\downarrow$ , and of the  $f$  and  $c$  magnetizations  $M$  and  $m$ . The parameters used in the calculation of Fig. 4 are:  $J_K=0.8$ ,  $J_H=-0.01$  and  $n_c=0.8$ . The two magnetization curves show clearly the existence of a continuous phase transition at the Curie temperature,  $T_C$ , corresponding to the disappearance of the ferromagnetic order. At low temperatures we observe the coexistence of a ferromagnetic order and Kondo behavior. The strength of the Kondo effect is the highest at  $T_C$ , since  $\lambda_\sigma$  grows and passes through a maximum at  $T_C$ , when the magnetization disappears. The parameters  $\lambda_\downarrow$  and  $\lambda_\uparrow$  are slightly different in the magnetic region but they coincide at  $T_C$ , when the spin symmetry is restored. For  $T>T_C$  the system exhibits only a Kondo behavior ( $\lambda_\sigma \neq 0$ ,  $M=0$  and  $m=0$ ). Finally, we define the temperature at which  $\lambda_\sigma$  vanishes as the Kondo temperature,  $T_K$ . The fact that  $\lambda$  vanishes at a particular temperature, instead of slowly decaying to zero, is a well known artifact of the mean-field approximation.

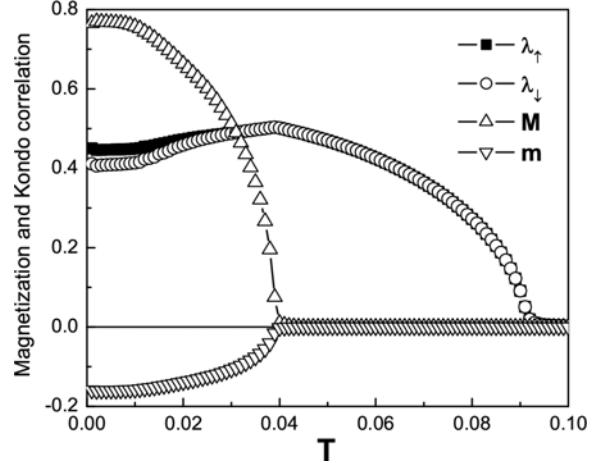


Fig. 4. Plot of  $\lambda_\uparrow$ ,  $\lambda_\downarrow$ ,  $M$  and  $m$  as a function of temperature for  $J_K=0.8$ ,  $J_H=-0.01$ ,  $n_c=0.8$ . At low temperatures we observe the coexistence of the ferromagnetic order and the Kondo effect.

For a large  $J_K$  value, we have obtained a Kondo-ferromagnetic state at very low temperatures below the Curie temperature  $T_c$ , then a Kondo state between  $T_c$  and the Kondo temperature  $T_K$  and finally a paramagnetic state above  $T_K$ . Then, we have derived the phase diagram giving  $T_c$  and  $T_K$  versus  $J_K$  and we have shown that  $T_K$  is increasing abruptly above a critical value  $J_K^c$ , while  $T_c$  is always non zero but increases slowly above  $J_K^c$  and remains there smaller than  $T_K$ . This "ferromagnetic-Kondo" diagram, shown in figure 5, is completely different from the Doniach diagram derived for cerium compounds.

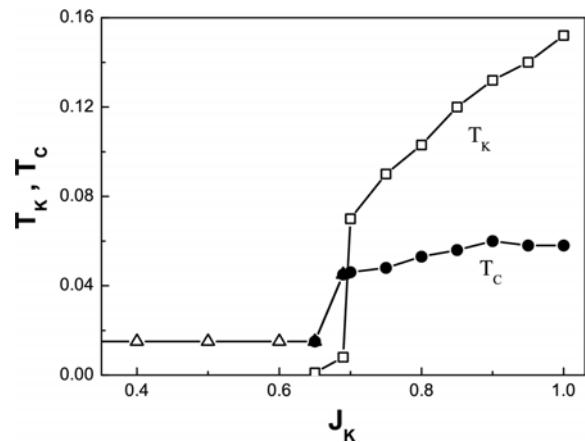


Fig. 5. The "ferromagnetic-Kondo" diagram : Plot of the Curie temperature  $T_c$  and of the Kondo temperature  $T_K$  versus  $J_K$  for  $J_H=-0.01$  and  $n_c=0.8$ .

Such a Kondo-ferromagnetism coexistence has been recently observed in other uranium compounds. We would like to mention  $UNiSi_2$  with a Curie temperature  $T_C=95K$  [83, 84, 85],  $UCo_{0.6}Ni_{0.4}Si_2$  with  $T_C=62K$  [86] and  $URu_{2-x}Re_xSi_2$  compounds where  $T_C$  increases rapidly with concentration  $x$  [87, 88, 89]. In these compounds  $T_C$  increases rapidly with  $x$  and there are clear evidences of the coexistence between the ferromagnetic order and a Non-Fermi-Liquid behavior [89]. Our UKL model can finally be applied to the case of the recently observed neptunium compound  $NpNiSi_2$ , which becomes ferromagnetic at  $T_C=51.5K$  and presents a Kondo behavior [90]. In the neptunium based compounds, 5f electrons are relatively well localized and the magnetic moment of Np can be described by a localized spin larger than  $S=1/2$ , corresponding again to the underscreened case.

However, possible extensions of the UKL model would be very interesting, in order to describe the coexistence between the ferromagnetic order and a Non-Fermi-Liquid behaviour, as observed in  $URu_{2-x}Re_xSi_2$  compounds [89] or the existence of a decrease of  $T_c$  versus  $J_K$  down to eventually a quantum critical point.

Finally, the question of the localization of the 5f electrons worths to be still discussed. The 5f electrons are well localized in UTe and itinerant in US, while they are intermediate in USe [77].  $T_c$  is initially increasing with pressure in UTe, in good agreement with the present UKL model, while it decreases at high pressure in UTe and decreases always with pressure in US; this pressure decrease of  $T_c$  is certainly connected with the delocalization of the 5f electrons and a complete treatment of the underscreened Anderson lattice model would be necessary to describe firstly the Kondo regime as in the UKL model and then a continuous transition to an intermediate valence regime.

## 5. The spin glass - Kondo interaction

The interaction between the Kondo effect and the spin glass in disordered cerium or uranium disordered alloys is a subject which is still well studied from both experimental and theoretical sides. Phase diagrams showing a Kondo phase, a spin glass one and a (ferro or antiferro) magnetic phase have been observed in disordered Cerium alloys like  $CeNi_{1-x}Cu_x$  [91, 92] or  $Ce_2Au_{1-x}Co_xSi_3$  [93] and in some disordered Uranium alloys such as  $UCu_{5-x}Pd_x$  [94] or  $U_{1-x}La_xPd_2Al_3$  [95].

The disordered alloys  $CeNi_{1-x}Cu_x$  and in particular the complexity of their low temperature phases have been extensively studied. A phase diagram has been firstly derived from magnetization and ac magnetic susceptibility measurements [92] : for  $x$  varying from  $x=0.7$  to  $x=0.2$ , when temperature decreases, one obtains firstly a spin

glass phase below the spin-glass freezing temperature  $T_f$  and then a ferromagnetic phase below the Curie temperature  $T_c$ ; with decreasing  $x$ ,  $T_f$  increases and  $T_c$  decreases and disappears at roughly  $x=0.2$ . Below roughly  $x=0.1$ , there is a Kondo non magnetic state and above  $x=0.8$ , there is an antiferromagnetic phase.

Then, many experiments have been performed on the  $CeNi_{1-x}Cu_x$  alloys, including neutron diffraction, muon spin relaxtion spectroscopy ( $\mu SR$ ), magnetization and magnetic susceptibility and specific heat at very low temperatures [96]. Small angle neutron scattering experiments as well as the occurrence of staircaselike hysteresis cycles at very low temperature (100mK) have made evidence for the existence of magnetic clusters of approximately 20 angstroms below the cluster-glass freezing temperature  $T_f$  in the range  $0.7 < x < 0.2$ . These recent experiments have shown that a long-range ferromagnetic order is observed below  $T_f$  without any indication of a sharp transition at a Curie temperature. These observations support a phenomenological model where a percolative process connects both magnetic states and in fact a cluster-glass exists always below  $T_f$  with a percolative process of magnetic clusters, the size of which increases with decreasing temperature. Recent theoretical simulations can also reproduce satisfactorily the experimental staircaselike hysteresis cycles observed at very low temperatures [97].

In fact, the other studied cases involving a spin glass phase are different from each other. For example, the phase diagram of the  $Ce_2Au_{1-x}Co_xSi_3$  presents at low temperatures with increasing  $x$  first an antiferromagnetic phase, then a spin glass phase and finally a non magnetic Kondo phase [93].

The Hamiltonian describing this situation is given by :

$$H = \sum_{k,\sigma} \varepsilon_k^c n_{k\sigma}^c + \sum_{i,\sigma} \varepsilon_0^f n_{i\sigma}^f + H_{SG} + J_K \sum_i [\hat{S}_{f,i}^+ \hat{S}_i^- + \hat{S}_{f,i}^- \hat{S}_i^+] \quad (5)$$

where the sum is over the  $N$  sites of a lattice.

The term  $H_{SG}$  corresponds to the intersite interaction between localized spins:

$$H_{SG} = \sum_{ij} J_{ij} \hat{S}_{fi}^z \hat{S}_{fj}^z \quad (6)$$

We have studied in detail the Hamiltonian (5) with the same mean field approximation as before to describe the Kondo interaction. But here there is a new term (6) which describes the spin glass character and which is given by a quantum Ising interaction between the z-components of the localized spins.

The description of the spin glass is rather difficult and we have studied several cases. Firstly, we have taken the Sherrington-Kirkpatrick model [98] where the exchange intersite integrals are assumed to be ramdomly distributed

with a gaussian distribution. The first model assumes a zero mean value of the gaussian distribution in order to study only the Kondo-spin glass interaction and we have, therefore, obtained there a phase diagram showing first a spin glass phase and then a Kondo phase with increasing  $J_K$  [99]. Then, in order to obtain a more complex phase diagram with a ferromagnetic [100] or an antiferromagnetic [101] phase occurring at low temperatures for smaller  $J_K$  values, we have taken the same model but with a non zero mean value  $J_0$  of the gaussian distribution. Thus, it results that the Kondo phase obtained for large  $J_K$  values is still there, but that there is a competition between the spin glass phase and the magnetic phase for smaller  $J_K$  values, which is interesting to account for some experimental data. However, in the ferromagnetic case, when  $J_K$  is typically of order  $J_0$ , we have obtained with decreasing temperature successively a ferromagnetic phase, then a mixed ferromagnetic-spin glass phase and finally a spin glass phase. The evidence of a spin glass phase at very low temperatures is in disagreement with experimental results previously described for  $CeNi_{1-x}Cu_x$  alloys.

Then, the previous description with a zero mean value which describes the spin glass-Kondo transition has been improved by adding a transverse field proportional to the  $x$ -component of the localized spin, in order to derive a quantum critical point for the magnetic phase [102]. Thus, we add here to the Hamiltonian (5) the following term:

$$H_T = 2\Gamma \sum_i \hat{S}_{fi}^x \quad (7)$$

Detailed calculations and results can be found in reference [102] and here we just present an extension of the previous calculation, where the Hamiltonian (5) is taken with both the terms (6) and (7) and where we take a non zero mean value in order to consider also the antiferromagnetic ordering in addition to the Kondo and spin glass effects [103]. We have taken here a  $\Gamma$  term varying in  $(J_K)^2$ , in order to simulate the RKKY interaction, as firstly done in ref. [61].

Fig. 6 shows two figures, i.e. on one side the experimental phase diagram of  $Ce_2Au_{1-x}Co_xSi_3$  alloys [93] and on the other side the resulting theoretical phase diagram obtained with reasonable parameters, as given in the figure caption. Thus, figure 6 shows a good agreement between experiment and theory, if we make the crude assumption that  $J_K$  varies roughly in a proportional way with the concentration  $x$  of cobalt.

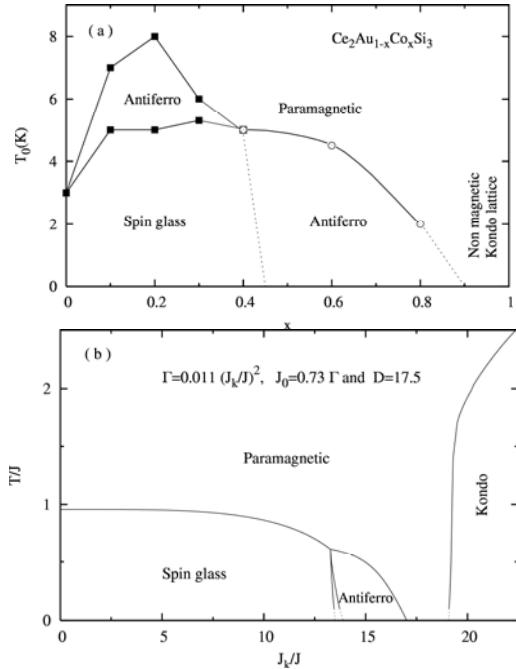


Fig. 6. Comparison between experimental and theoretical phase diagrams: the upper figure is an experimental phase diagram of  $Ce_2Au_{1-x}Co_xSi_3$  alloys [93]; the lower figure is a theoretical phase diagram  $T/J$  versus  $J_K/J$  for the relations  $\Gamma=0.011(J_K/J)^2$  and  $J_0/J=0.73\Gamma/J$ , where the dotted lines are the extrapolations carried for lower temperature [103].

On the other hand, we have recently studied the Kondo-spin glass-ferromagnetism coexistence in disordered cerium alloys by another new approach which introduces a generalization of the Mattis model [104] and which represents an interpolation between ferromagnetism and a highly disordered spin glass. The detailed calculations can be found in ref. [105] and here we start from the following simple version of the exchange integrals between spins on different sites  $i$  and  $j$ , given by :

$$J_{ij} = \frac{1}{N} \sum_{\mu} J_i^{\mu} \xi_i^{\mu} \xi_j^{\mu}, \quad (8)$$

where the  $\xi_i^{\mu} = \pm 1$  ( $\mu = 1, 2, \dots, p$ ;  $i = 1, 2, \dots, N$ ) are independent random distributed variables, with an equal probability for  $\xi_i^{\mu}$  to be equal to +1 or -1. For the classical Ising model, if  $p=1$ , the original Mattis model [104] is recovered. However, if  $p=N$  with the  $N^2$  random variables  $\xi_i^{\mu}$  having a zero mean and a variance one, in the limit of  $N$  large,  $J_{ij}$  tends to a Gaussian variable with a zero mean and a variance  $N^{-1/2}J$  as in the Sherrington-Kirkpatrick model [98]. Therefore, we can consider this model as an

interpolation between ferromagnetism and highly disordered spin glass. The critical parameter is here the ratio  $a=p/N$ , which gives an estimation of the relative importance of the ferromagnetic and spin glass phases for small  $J_K$  values; for large  $J_K$  values, the Kondo phase is always present.

We just present these results on Fig. 7 for a value  $a=0.04$ . We obtain a spin glass phase, then a mixed ferromagnetic-spin glass phase and finally a ferromagnetic phase with decreasing temperature for a relatively small  $J_K/J$  ratio. Figure 7 accounts for the experimental phase diagram of  $CeCu_{1-x}Ni_x$  disordered alloys where Kondo, spin glass and magnetically ordered phases are observed. This last approach gives an improved description of this phase diagram with the occurrence of the ferromagnetic phase below the spin glass phase at the lowest temperatures.

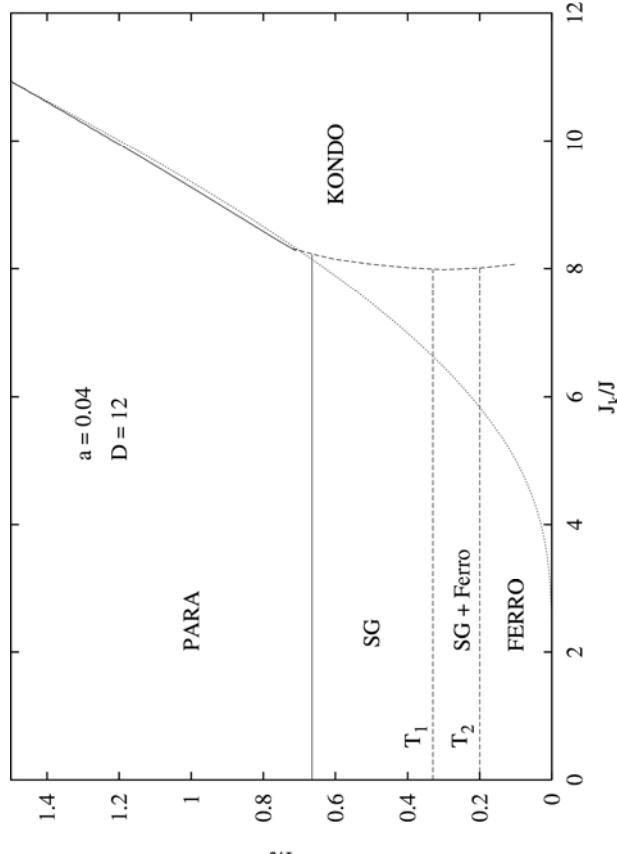


Fig. 7. The Kondo-spin glass-ferromagnetic diagram versus  $J_K/J$  for  $D=12$  and  $a=0.04$ .

The experimental phase diagram of  $CeCu_{1-x}Ni_x$  is in fact very complicated: for example, a very careful experimental study of  $CeCu_{0.6}Ni_{0.4}$  yields a percolative transition with decreasing temperature from a cluster-glass state with ferromagnetic correlations to a disordered ferromagnetic state [96] and recent theoretical simulations can reproduce satisfactorily the experimental situation

[97]. But, the results presented in figure 7 and in a more detailed way in ref.[105] give an improved description of the phase diagram of  $CeCu_{1-x}Ni_x$  alloys and we can conclude that the more "local" description given by the equation (8) seems to be more adequate here than the "average" description. However, the percolative transition observed in these alloys can probably be better treated by numerical simulations to describe the different ferromagnetic clusters and further work is necessary to have this better approach.

## 6. Conclusions

We have discussed the Kondo lattice problem for cerium or other anomalous rare-earth systems and also uranium compounds. In cerium (or ytterbium) Kondo compounds described by the configuration  $4f^1$  (or  $4f^{13}$ ), there exists a strong competition between the long range magnetic order and the Kondo effect and one result is that the eventual ordering temperatures are relatively small. On the opposite, in uranium compounds or eventually in a neptunium one, the configuration corresponds to  $5f^n$  with  $n=2$  or larger, and we have developed the Underscreened Kondo Lattice model, which can account for the observed coexistence between ferromagnetic order (with relatively large Curie temperatures) and Kondo behavior observed in uranium compounds like  $UTe$ . We have also discussed the interplay between the Kondo effect, the spin glass and the magnetic order (antiferromagnetic or ferromagnetic), which can be applied to disordered cerium systems such as  $CeCu_xNi_{1-x}$  or  $Ce_2Au_{1-x}Co_xSi_3$  alloys and a theoretical improvement has been recently obtained by considering a generalization of the Mattis model.

## Acknowledgements

B. C. thanks Professor Andrei Galatanu and the European COST Action P16 (Emergent Behaviour in Correlated Matter) for their strong support. This work was partially supported by the Brazilian agencies CNPq, CAPES and FAPERGS.

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