

Comments on the Dense Kondo State

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The dense Kondo behaviour observed in Ce metal, its alloys and compounds is discussed by taking into account spin-orbit coupling and crystal field splitting. Especially, by using an expression for the Kondo temperature under a crystal field, we clarify the reason why the Kondo effect dominates the RKKY interaction.

§ 1. Introduction

The Ce metal, its alloys and compounds, such as CeAl₂, CeAl₃ and CeB₆, show the so-called dense Kondo behaviour at low temperatures. Typical phenomena in the dense Kondo system are logarithmic increase of the resistivity with decreasing temperature and strong enhancement of the T -linear term of the specific heat. These effects are observed also in the Yb system.

The usual Kondo systems such as CuMn contain magnetic atoms in very low concentration. At a little high concentration (~ 1 atomic percent), interaction effects between magnetic atoms are dominant and magnetic alloys show long range order or spin glass-like behaviour. However, dense Kondo systems show Kondo-like behaviour, though cerium ions exist in very high concentration. The purpose of this note is to clarify the reason why the Kondo effect dominates intersite interactions and also the reason why the dense Kondo behaviour is realized only in the systems containing Ce and Yb.

The Kondo effect in real metals has been discussed by many authors.^{1)~3)} Okada and Yosida¹⁾ discussed the Kondo effect for the case with transition metal impurities. They showed that the binding energy of the bound state for the core with orbital degeneracy can become large by exchanging orbital moments. For example, the binding energy, \tilde{E} , for d -shell with a single electron is given by

$$\tilde{E} = -D_0 \exp[-N/(2l+1)\rho|J|], \quad (1)$$

where $l=2$; $J(<0)$ is exchange coupling between conduction electron and localized d -electron. ρ is the density of state of the conduction band with width $2D_0$. N is the total number of atoms.

On the other hand, in the half-filled d -shell, orbital momenta are quenched owing to the strong Hund's coupling. The binding energy for this case is very small owing to the absence of orbital exchange, and is given by

$$\tilde{E} = -(2l+1)D_0 \exp[-(2l+1)N/\rho|J|]. \quad (2)$$

This result explains the very low Kondo temperature observed in CuMn system.

Ogawa and Yoshimori²⁾ (hereafter referred to as OY) discussed the effect of anisotropy energy on the ground state of Ce impurity in metals on the basis of Yosida theory.⁴⁾ Nozières and Blandin³⁾ gave a general consideration on the real systems, taking into account orbital structure of localized states, crystal field and spin-orbit splittings.

These theories¹⁾⁻³⁾ show the importance of taking into account real situations for individual cases in quantitative estimation of the Kondo temperature, T_K . It is Ce system with one f -electron and Yb system with one f -hole that shows the dense Kondo behaviour. Hereafter, we discuss the Ce system, since the same discussion can be done for the Yb system in a parallel way by exchanging electron and hole. The lowest state of spin-orbit splittings, $j=5/2$ for Ce^{3+} , splits into subgroups due to a crystal field. For example, it splits into Γ_7 doublet and Γ_8 quartet under a cubic crystal field.

Therefore, we consider the following Hamiltonian:²⁾

$$H = \sum_{k,M} \epsilon_k c_{kM}^\dagger c_{kM} - (J/2N) \sum_{\substack{MM' \\ kk'}} c_{kM}^\dagger c_{k'M'} (a_{M'}^\dagger a_M - b \delta_{MM'}) + \sum_M E_M a_M^\dagger a_M. \quad (3)$$

This Hamiltonian describes the crystal potential, E_M , in addition to the exchange term given by Coqblin and Schrieffer,⁵⁾ M being an eigenstate under a crystal field. c_{kM} and a_M are annihilation operators in the partial wave representation with respect to M for conduction electron with wave number k and localized electron, respectively. The energy level, E_M , is defined so as to satisfy the following condition:

$$\sum_M E_M = 0. \quad (4)$$

As we consider the trivalent Ce ion with one f -electron, we confine ourselves in the subspace

$$\sum_M a_M^\dagger a_M = 1. \quad (5)$$

The second term in the bracket of the exchange term is added to make the exchange term traceless.

In the next section, we discuss the binding energy of the bound state under a crystal field on the basis of Yosida theory,⁴⁾ following OY.²⁾ Then, we consider the Kondo temperature on the basis of scaling theory,^{3),6)} with emphasis on the crystal field dependence of T_K . Finally, we discuss the reason why the RKKY interaction is weak in our system.

§ 2. Binding energy of bound state under crystal field

Now, we consider the ground state energy of Hamiltonian (3), which has been discussed by Ogawa and Yoshimori.²⁾ In the absence of crystal field, the binding energy of the bound state, \tilde{E}_0 , is similar to (1) and is given by

$$\tilde{E}_0 = -D_0 \exp\{-2N/[\rho|J|(2j+1)]\}, \quad (6)$$

where $2j+1=6$ for $j=5/2$. Thus, we can see that the degeneracy of f -level plays an important role in increasing the binding energy,^{7),8)} corresponding to the orbital degenerate case in 3- d transition metals.¹⁾

However, a crystal field lifts the degeneracy. In usual case, crystal field splitting is larger than binding energy. Therefore, higher states seem not to give any essential effect in this system at low temperatures, and it seems to be a good approximation to take only the lowest state into account. However, this is not true for a moderately strong crystal field. It is the main purpose of this note to show an important role played by higher levels under crystal field.

The wave-function for the ground state of Hamiltonian (3) is written as^{2),4)}

$$\phi = \left[\sum_{1,M} \Gamma_1^M C_{1M} a_M^\dagger + \sum \Gamma_{123}^{MM'} C_{1M}^\dagger C_{2M} C_{3M'} a_{M'}^\dagger + \sum \Gamma_{12345}^{MM'M''} C_{1M}^\dagger C_{2M} C_{3M'}^\dagger C_{4M''} C_{5M''} a_{M''}^\dagger + \dots \right] |0\rangle, \quad (7)$$

where $\Gamma_1^M, \Gamma_{123}^{MM'} \dots$ are the amplitudes to be determined, and 1, 2, ... represent wave number $k_1, k_2 \dots$. $|0\rangle$ is the Fermi state.

Up to moderately strong crystal field, the integral equation for Γ_1^M is given by

$$(-\varepsilon_1 + E_M - \tilde{E}) \Gamma_1^M = \frac{J}{2N} \left[(2j+1)^{-1} \sum_2 \Gamma_2^M - \sum_{2M'} \Gamma_2^{M'} \right] + \frac{J}{2N} \sum_{2M'} \Gamma_2^{M'} K_{MM'} (-\varepsilon_1 - \varepsilon_2). \quad (8)$$

The ground state energy E is given by $E = \tilde{E} + \Delta E$, where

$$\Delta E = - \left(\frac{\rho J}{2N} \right)^2 4j(j+1)(2j+1)^{-1} 2D_0 \log 2 + \dots, \quad (9)$$

and \tilde{E} is determined by (8). The energy shift ΔE has E_M -dependent terms in addition to (9), but these terms can be neglected for the case with

$$\Delta = E_{\max} - E_{\min} \ll -\tilde{E}_0 / |J\rho / 2N|^{\nu_m/(2j+1)}, \quad (10)$$

where Δ is the total splitting due to the crystal field. ν_m is the degree of degeneracy of the lowest level and \tilde{E}_0 is given by (6).

The kernel of the integral equation has also E_M -dependence, but if

$$\log[\Delta / |\tilde{E}_0|] \ll 2N [(2j+1)\rho |J|]^{-1}, \quad (11)$$

it can be approximated by

$$K_{MM'}(\varepsilon) = (z^{-1} - 1)(2j+1)^{-1} [\delta_{MM'} \{1 + (2j+1)^{-2}\} - 2(2j+1)^{-1}], \quad (12)$$

$$z = 1 - (2j+1) \left(\frac{J\rho}{2N} \right) \log[(\varepsilon - \tilde{E}) / D_0].$$

By retaining the most divergent terms for E_M -independent terms and also for E_M -dependent terms, we obtain the eigenvalue equation,²⁾

$$\sum_M \log\{(E_M - \tilde{E}) / (-\tilde{E}_0)\} = 0. \quad (13)$$

The energy \tilde{E} determined by (13) contains a contribution from crystal field. An essentially anomalous part of the binding energy, \tilde{E}_a , is obtained by subtracting the average crystal field energy \bar{E}_a from \tilde{E} ,

$$\bar{E}_a = \sum_M E_M A_M, \quad (14)$$

$$\tilde{E}_e = \tilde{E} - \bar{E}_e = -(2j+1) / \sum_M (E_M - \tilde{E})^{-1}, \quad (15)$$

where A_M is a weight factor defined by $\langle \phi | a_M^\dagger a_M | \phi \rangle / \langle \phi | \phi \rangle$.

The eigenvalue equation (13) has been derived on the assumptions (10) and (11). However, this equation gives a correct limit of strong crystal field in the most divergent approximation,²⁾

$$\tilde{E} = -D_0 \exp[-2N/|J\rho\nu_m] + E_{\min}. \quad (16)$$

As shown later, strong crystal field means $\Delta \simeq D_0$. After the general discussion mentioned above, Ogawa and Yoshimori calculated the binding energy \tilde{E}_e for the case with a uniaxial anisotropy,

$$E_M = \Delta [M^2 - (35/12)] / 6, \quad (17)$$

$$M = \pm \frac{5}{2}, \pm \frac{3}{2}, \pm \frac{1}{2}. \quad (18)$$

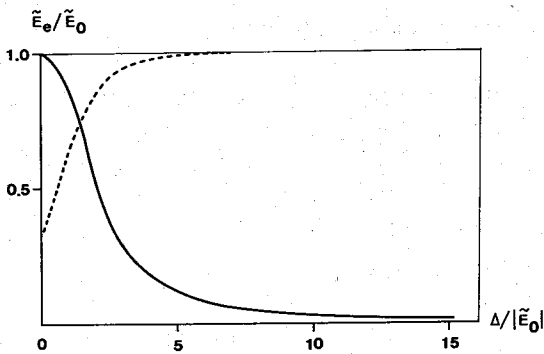


Fig. 1. The essential binding energy is shown as a function of the splitting, $\Delta = E_Q - E_D > 0$, due to the cubic crystal field. Here, we assumed that Γ_7 doublet is lower than Γ_8 quartet. The full line shows the binding energy \tilde{E}_e and the dotted line shows the weight factor of Γ_7 doublet in the ground state.

The numerical result is shown in their paper.²⁾

In the present paper, we assume a cubic crystal field and define crystal potential $E_D = -2\Delta/3$ for Γ_7 doublet and $E_Q = \Delta/3$ for Γ_8 quartet. Hereafter, we consider two cases depending on the sign of $\Delta = E_Q - E_D$.

Case 1. $E_Q > E_D, (\Delta > 0)$.

By inserting E_Q and E_D into (13) and (15), we obtain the binding energy. It is shown as a function of Δ in Fig. 1, together with the weight factor of the Γ_7 in the ground state. For a moderately strong crystal field ($D_0 \gg \Delta \gg -\tilde{E}_e$), the essential binding energy \tilde{E}_e is given by

$$\tilde{E}_{e1} \equiv \tilde{E}_0 \left(\frac{\tilde{E}_0}{\Delta} \right)^2. \quad (19)$$

For a very strong crystal field ($D_0 \sim \Delta$), the binding energy is given by

$$\tilde{E}_{e2} \equiv -D_0 \exp[-N/\rho|J|] = \tilde{E}_0 \left(\frac{\tilde{E}_0}{D_0} \right)^2. \quad (20)$$

The binding energy, \tilde{E}_{e1} of (19), in the intermediate case is rewritten by the use of \tilde{E}_{e2} of (20) as

$$\tilde{E}_{e1} = \left(\frac{D_0}{\Delta} \right)^2 \tilde{E}_{e2}. \quad (21)$$

Thus, we can see that higher states under a crystal field should not be neglected in order to obtain correct order of the binding energy, as far as $\Delta \ll D_0$ even if $\Delta \gg |\tilde{E}_0|$.

Case 2. $E_Q < E_D$, ($\Delta < 0$).

$$E_Q = -|\Delta|/3, \quad E_D = 2|\Delta|/3. \quad (22)$$

In a similar way to the case 1, \tilde{E}_e is obtained from (13) and (15). For a moderately strong crystal field, ($D_0 \gg |\Delta| \gg |\tilde{E}_e|$),

$$\tilde{E}_{e1} = \tilde{E}_0 (|\tilde{E}_0|/|\Delta|)^{1/2}. \quad (23)$$

For a very strong crystal field ($|\Delta| \sim D_0$),

$$\tilde{E}_{e2} = -D_0 \exp[-N/2(\rho|J|)] = \tilde{E}_0 \left(\frac{|\tilde{E}_0|}{D_0} \right)^{1/2}. \quad (24)$$

\tilde{E}_{e1} given by (23) is rewritten as

$$\tilde{E}_{e1} = \tilde{E}_{e2} (D_0/|\Delta|)^{1/2} = -\sqrt{\frac{D_0}{|\Delta|}} D_0 \exp[-N/2\rho|J|]. \quad (25)$$

§ 3. Scaling theory in the presence of crystal field

In the previous section, we have discussed the binding energy of the bound state. Here, we consider the Kondo temperature by using scaling theory.^{3),6)}

To make the scaling equation more general, we generalize the exchange part of Hamiltonian (3) as

$$\begin{aligned} H_{\text{ex}} = & - \sum_{\substack{MM' \\ kk'}} \frac{J_1}{2N} c_{kM}^\dagger c_{k'M'} a_{M'}^\dagger a_M - \sum_{\substack{mm' \\ kk'}} \frac{J_0}{2N} c_{km}^\dagger c_{k'm'} a_{m'}^\dagger a_m \\ & - \sum_{\substack{mm' \\ kk'}} \frac{J_2}{2N} (c_{kM}^\dagger c_{k'm} a_m^\dagger a_M + c_{km}^\dagger c_{k'M} a_M^\dagger a_m). \end{aligned} \quad (26)$$

Capital M represents a higher level state in a cubic crystal field, and m stands for a lower level state. The exchange interactions J_0 and J_1 work in the subspace of lower and higher levels, respectively and J_2 is that between two subspaces.

Following the poorman's derivation,⁶⁾ we obtain the following scaling equations:

$$\frac{d\tilde{J}_0}{dD} = \sum_m \frac{\tilde{J}_0^2}{D + E_m - z} + \sum_M \frac{\tilde{J}_2^2}{D + E_M - z}, \quad (27)$$

$$\frac{d\tilde{J}_1}{dD} = \sum_M \frac{\tilde{J}_1^2}{D + E_M - z} + \sum_m \frac{\tilde{J}_2^2}{D + E_m - z}, \quad (28)$$

$$\frac{d\tilde{J}_2}{dD} = \sum_M \frac{\tilde{J}_1 \tilde{J}_2}{D + E_M - z} + \sum_m \frac{\tilde{J}_0 \tilde{J}_2}{D + E_m - z}. \quad (29)$$

z is the total energy of the impurity in the crystalline field and the interacting electron.³⁾ Hereafter, we assume a cubic crystal field; $E_m = -2\Delta/3$ and $E_M = \Delta/3$ for $\Delta > 0$, and $E_m = \Delta/3$ and $E_M = -2\Delta/3$ for $\Delta < 0$.

Case 1. $\Delta > 0$, $z \simeq -2\Delta/3$.

$$\frac{d\tilde{J}_0}{dD} = \frac{2\tilde{J}_0^2}{D} + \frac{4\tilde{J}_2^2}{D + \Delta}, \quad (30)$$

$$\frac{d\tilde{J}_1}{dD} = \frac{4\tilde{J}_1^2}{D+\Delta} + \frac{2\tilde{J}_2^2}{D}, \tag{31}$$

$$\frac{d\tilde{J}_2}{dD} = \frac{4\tilde{J}_1\tilde{J}_2}{D+\Delta} + \frac{2\tilde{J}_0\tilde{J}_2}{D}. \tag{32}$$

If we assume $J_0=J_1=J_2=J$ and $\tilde{J}_0=\tilde{J}_1=\tilde{J}_2=\tilde{J}$, Eqs. (30)~(32) are reduced to the following equation:

$$\frac{d\tilde{J}}{dD} = \frac{2\tilde{J}^2}{D} + \frac{4\tilde{J}^2}{D+\Delta}. \tag{33}$$

This isotropic case is realized, when the localized f -level ϵ_f is low enough from the Fermi level,¹⁾ E_F , and Δ satisfies $U \gg |E_F - \epsilon_f| \gg |\Delta|$, U being the intra-Coulomb repulsion between f -electrons. The solution of (33) is given by

$$-\frac{1}{\tilde{J}} + \frac{2N}{\rho J} = 2 \log \frac{D}{D_0} + 4 \log \frac{D+\Delta}{D_0+\Delta}, \tag{34}$$

where D_0 and $\rho J/2N$ are given by an initial condition. For the case with $D_0 \gg \Delta$, \tilde{J} is given by

$$\tilde{J} = \frac{\rho J}{2N} \left[1 + \frac{\rho|J|}{N} \log(D/D_0) + \frac{2\rho|J|}{N} \log \frac{D+\Delta}{D_0} \right]^{-1}. \tag{35}$$

Since the Kondo temperature is defined as D giving rise to infinite coupling constant, T_K is determined by the following equation (in this paper we put $k_B=1$):

$$1 + \frac{\rho|J|}{N} \log \frac{T_K}{D_0} + \frac{2\rho|J|}{N} \log \frac{T_K+\Delta}{D_0} = 0, \tag{36}$$

$$T_K = D_0 e^{-N/\rho|J|} \times \left(\frac{D_0}{T_K+\Delta} \right)^2. \tag{37}$$

If $T_K \ll \Delta$, we obtain

$$T_K = D_0 e^{-N/\rho|J|} \left(\frac{D_0}{\Delta} \right)^2. \tag{38}$$

This is the same expression as (21).

Table I. Kondo temperature T_K for typical values of Δ and \tilde{J}_0 .

	T_K (K)	$ \Delta $ (K)	\tilde{J}_0	\tilde{J}_0^2
$\Delta > 0$ (Case 1)	50	0	0.062	0.004
	10	100	0.062	0.004
	0.001	∞	0.062	0.004
	10	∞	0.145	0.021
$\Delta < 0$ (Case 2)	20	0	0.054	0.003
	10	100	0.054	0.003
	1	∞	0.054	0.003
	10	∞	0.072	0.005

$$\tilde{J}_0 = |J\rho|/N, D_0 = 10^4 \text{K}.$$

Case 2. $\Delta < 0$, $z \simeq -|\Delta|/3$.

$$\frac{d\tilde{J}}{dD} = \frac{4\tilde{J}^2}{D} + \frac{2\tilde{J}^2}{D+|\Delta|}. \quad (39)$$

$$-\frac{1}{\tilde{J}} + \frac{2N}{\rho J} = 4 \log D/D_0 + 2 \log \frac{D+|\Delta|}{D_0+|\Delta|}, \quad (40)$$

$$\tilde{J} = \frac{\rho J}{2N} \left[1 + \frac{\rho|J|}{N} \log \frac{D+|\Delta|}{D_0} + 2 \frac{\rho|J|}{N} \log \frac{D}{D_0} \right]^{-1}. \quad (41)$$

T_K is given by

$$T_K = (D_0 / (T_K + |\Delta|))^{1/2} D_0 \exp[-N/2\rho|J|] \quad (42)$$

$$\simeq \left(\frac{D_0}{|\Delta|} \right)^{1/2} D_0 \exp[-N/2\rho|J|] \quad \text{for } T_K \ll |\Delta|. \quad (43)$$

This expression for T_K is the same as $|\tilde{E}_{e1}|$ in (25). The above results are shown in Table I for typical values of Δ and $\rho J/N = \tilde{J}_0$, with $D_0 = 10^4 \text{K}$.

§ 3. Conclusion and discussion

First, we assume that Γ_7 doublet is the ground state (case 1) and discuss the contribution of higher levels to the Kondo temperature. If we put $\Delta/T_K = 10$, we can see from Fig. 1 that the doublet state contributes to 94% of the ground state and the quartet to only 6%. If we use the same value for exchange coupling parameter and neglect mixing with higher levels, T_K or $|\tilde{E}_{e1}|$ decreases from 10 K to 10^{-3}K . This example shows that by mixing with higher levels in a few percents, the Kondo temperature increases drastically.

For case 2, in which Γ_8 quartet is the ground state, the Kondo temperature is high owing to the degeneracy of the ground state, similar to the case in the absence of crystal field.^{1),2),7)} Even in this case, T_K increases by one order of magnitude by mixing with Γ_7 doublet state (see Table I).

The role played by the higher levels for the case with $D_0 \gg \Delta \gg T_K$ can also be explained on the basis of the scaling theory. The coupling constant \tilde{J} is scaled with D decreasing from D_0 to $|\Delta|$ by

$$\frac{d\tilde{J}}{dD} = 6 \frac{\tilde{J}^2}{D}. \quad (44)$$

This is the same equation in the absence of crystal field splitting and increases the coupling constant rapidly. From (44) the coupling constant $\tilde{J} = \tilde{J}_i$ at $D = \Delta$ is given by

$$\tilde{J}_i = \frac{\rho J}{2N} \left[1 - \frac{3\rho|J|}{N} \log \frac{D_0}{\Delta} \right]^{-1}. \quad (45)$$

This coupling constant \tilde{J}_i is finite but larger than that scaled by using only lower states. Then, \tilde{J} is rescaled with D decreasing from Δ to T_K , starting from the enlarged initial coupling constant \tilde{J}_i . Thus, we have a large value of Kondo temperature.

The other magnetic rare earth metals than Ce and Yb have multiply occupied f -shell. For f -shell with plural electrons or holes, Hund's coupling is dominant and is of the same order as D_0 . Thus, Hund's coupling in f -shell reduces the degree of freedom of exchanging angular momentum and spin, to reduce remarkably the Kondo temperature.¹⁾ Thus,

we have shown the reason why the metallic system with Ce has a high Kondo temperature, although the exchange interaction is very weak.

Now, we discuss the reason why the RKKY interaction is weak in the Ce system. Though the RKKY interaction depends on the band structure in general, its strength is proportional to S^2 , S being the magnitude of the localized spin. Therefore, the RKKY interaction between cerium ions with a single f -electron is weak. For example, Gd metal with localized spin $S=7/2$ orders in the ferromagnetic state below 300K. Assuming that magnetic ordering temperature is scaled by S^2 , we obtain 6K as the critical temperature for Ce. This result is confirmed also by the following consideration. If we scale the paramagnetic Curie temperature by the de Genne factor $(g_j-1)^2 j(j+1)$, we obtain 3K for that of Ce^{3+} . These values for magnetic ordering temperature of Ce system are reasonable compared with observed ones. Moreover, if we take into account the Kondo effect, it lowers further the ordering temperature estimated above.

In conclusion, we have clarified the two reasons concerning the origin of dense Kondo state. One is that for the high Kondo temperature in spite of weak exchange interaction. The other is that for low ordering temperature due to weak RKKY interaction. By these two effects, the dense Kondo state is realized in Ce system. Though we have discussed the case of Ce, the same consideration can be applied to Yb with one hole in f -shell.

Finally, we would like to mention briefly the low temperature behavior of the dense Kondo system on the same standpoint we have taken in this paper. This standpoint is that the $4f$ level ϵ_f is deep enough, intra-Coulomb repulsion U is very large and s - f exchange model is well applied. This system shows the logarithmic increase of the resistivity with decreasing temperature, but since deep $4f$ levels are arranged on the periodic lattice, they act as the periodic potential to the conduction electrons. The periodicity makes scattering coherent. Therefore, the resistivity decreases through maximum at low temperatures and vanishes at the absolute zero unless magnetic order takes place.

In these low temperature regions, the conduction electrons will behave as heavy fermi liquid because they must keep localized $4f$ electrons in the Kondo states. This heavy fermion system can become superconducting if enough attraction exists between heavy fermions to overcome repulsion arising from the s - f exchange. In this case, strong repulsion U is ineffective since $4f$ -levels are singly occupied. Recently found superconductivity in the dense Kondo system, $CeCu_2Si_2$ ⁹⁾ could reasonably be understood in this way although detailed calculations are needed for quantitative understanding.

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