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Origin of the pressure dependence of the Kondo temperature in Ce- and Yb-based heavy-fermion compounds

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Abstract

We find that the unusual pressure dependence of the Kondo temperature in Ybbased heavy-fermion (HF) compounds can be explained by the competition between two mechanisms of coupling between f electrons and the lattice. An increase of the hybridization between f and conduction electron states under pressure results in the increase of the Kondo temperature while the suppression of valency fluctuations of rare-earth ions under pressure leads to the decrease of the Kondo temperature. In contrast to Yb compounds, in Ce compounds pressure enhances both the hybridization and valency fluctuations. As a result the Kondo temperature increases. We compare the theory with available experimental data and find a qualitative and quantitative agreement.

1. Introduction

For the last two decades Yb and Ce heavy-fermion (HF) compounds have attracted much attention because of their particular low-temperature electronic properties which are formed due to the strong Kondo effect. The compounds demonstrate similar low-temperature magnetic, thermal and transport properties [1, 2]. That is why it is especially astonishing that the pressure dependences of the Kondo temperature T_k in these compounds have opposite characters. T_k in all Ce heavy fermion compounds increases with increasing pressure P. The Grüneisen parameter $\Omega = -d \ln T_k/d \ln V$ is positive and large. For example, $\Omega = 115$ for CeCu₆ and $\Omega = 160$ for CeAl₃ [2]. The Kondo temperature T_k of Yb compounds decreases with increasing pressure P from the ambient pressure. Ω is negative and of the order of a few tens. This type of behaviour takes place in Yb₂Ni₂Al [3], YbCu₂Si₂ [4], YbRh₂Si₂ [5], YbCu_{4.5} [6], YbInAg₄ [7], Yb₂Fe₃Si₅ [8] and many other Yb-based compounds; see also [9, 10]. An unusual pressure dependence of the temperature of the resistance maximum T_m was observed in Yb₂Ni₂Al [3] and YbCu₂Si₂ [4] where the temperature T_m achieves a broad minimum at a pressure P_m of the

order of 10 GPa and then starts to increase slightly at *P* above P_m . Such a pressure dependence and the negative Grüneisen parameter Ω in the Yb heavy-fermion compounds constitutes a challenge to the conventional theory which predicts that Ω must be positive due to the increase of the hybridization strength between localized f states and conduction band states when the volume of the system decreases.

In order to explain the negative Grüneisen parameter Ω in the mixed valency compounds $YbIn_{1-x}Ag_xCu_4$ an additional mechanism of the interaction between f electrons and the lattice was recently proposed in [11]. This interaction takes into account lattice deformations produced by fluctuations of the ionic radius of rare-earth ions which accompany valency fluctuations of the ions. In the present paper, we will show that this mechanism of electron–lattice interaction allows us to explain the negative Grüneisen parameter Ω and the pressure dependence of the Kondo temperature observed in $YbCu_2Si_2$ [4] and others Yb heavy-fermion compounds. It can also be partly responsible for the large Grüneisen parameter Ω in Ce compounds.

2. Kondo effect under pressure

Let us consider fluctuations of the valency of Ce and Yb ions in HF compounds. Ce³⁺ ions in the ground state have the configuration 4f¹ with the magnetic moment j = 5/2. The first excited nonmagnetic state has the configuration 4f⁰. Thus, Ce ions fluctuate mainly between the 3+ and 4+ valency states. Yb³⁺ ions in the ground state have the configuration 4f¹³ with the magnetic moment j = 7/2 produced by one hole in the 4f shell. In the first excited state Yb³⁺ ions have the configuration 4f¹⁴ which corresponds to the completely filled 4f shell. Thus, Yb ions fluctuate mainly between 3+ and 2+ valency states. This picture is confirmed by various experimental methods. In accordance with the photoemission spectra (PES) measurements [12], neither 4f¹² nor 4f¹³ ²F_{5/2} contributes to the low-energy properties of Yb ions. Due to valency fluctuations, the valency ν of Yb and Ce ions deviates from 3+, i.e. $\nu = 3 + \Delta \nu$, where $\Delta \nu$ is negative for Yb and positive for Ce ions.

It is important to note the following peculiarity of the valency fluctuations between the configurations $4f^{13}$ and $4f^{14}$ of Yb ions. When a Yb ion transits from the ground state Yb³⁺ into the excited state Yb²⁺, the number of f electrons increases from 13 to 14. The Coulomb repulsion between the electrons on the f shell is enhanced. As a result the radius of the f shell with 14 electrons and, in turn, the ionic radius of the Yb ion are increased. The increase of the ionic radius produces a local pressure on neighbouring atoms which brings about a local volume extension. The microscopic origin of the effect is related with the Coulomb repulsion between f electrons and electron shells of nearest neighbouring atoms. It is clear that applying pressure also influences the valency fluctuations between the configurations $4f^{13}$ and $4f^{14}$. Recently this mechanism was discussed as a mechanism of the unusually high compressibility of ytterbium metal [13]. A similar mechanism was also discussed in [14].

Another situation takes place for Ce ions. If a Ce ion is excited from the ground state configuration $4f^1$ into the excited configuration $4f^0$, then the ionic radius of the Ce ion decreases. This leads to a local pressure that produces a local lattice shrinking.

Due to this interaction a change of the valency of f ions by a value $\Delta \nu$ produces a lattice strain. The energy of the interaction can be written as follows [11]:

$$E_{\rm fl} = N_{\rm f} D_{ij} e_{ji} \Delta \nu \tag{1}$$

where e_{ij} is a strain tensor, i, j = x, y, z. N_f is the total number of f ions in the lattice. For a cubic lattice the energy tensor D_{ij} takes the simple form $D_{ij} = \delta_{ij}d_f$. In this case the interaction energy E_{fl} takes the form $E_{fl} = d_f e_B \Delta v$ where $e_B = e_{xx} + e_{yy} + e_{zz}$ is the bulk strain which is related to a change of the volume \mathcal{V} of the system by the value $\Delta \mathcal{V}$, $e_B = \Delta \mathcal{V}/\mathcal{V}$. A simple

physical meaning of the energy (1) may be given: $E_{\rm fl}$ is equal to the work $P\Delta V$ produced by an internal pressure $P = N_{\rm f} d_{\rm f} \Delta v / V$ when changing the volume of the system by a value ΔV . Below we assume that the coupling constant D is positive. In Yb-based heavy-fermion compounds the valency change Δv is negative. According to equation (1), the system gains in energy $E_{\rm fl} < 0$ due to the interaction (1) when the volume of the system increases, i.e. at $e_{\rm B} > 0$. In Ce-based heavy-fermion compounds Δv is positive. Then, there is a gain in energy if the volume of the system decreases, i.e. at $e_{\rm B} < 0$.

There is also a conventional mechanism of the interaction between f electrons and the lattice which is related to a dependence of the overlap between the wavefunctions of the f states and conduction band states on a strain. This hybridization is characterized by a parameter

$$V = V_0 \exp(-re_{\rm B}) \tag{2}$$

where r > 0. V increases if $e_B < 0$. Both the interaction (1) and (2) must be taken into account when studying the influence of pressure on the Kondo effect.

We will study the dependence of the Kondo temperature on pressure in the framework of the single-impurity Anderson model. We add the electron–lattice coupling equation (1) to the Anderson Hamiltonian, taking into account that the parameter Δv must be replaced by a corresponding operator [11]. In the case of Ce compounds we have $\Delta v = 1 - \hat{n}_f$, where \hat{n}_f is the operator of the number of electrons on the 4f shell. For Yb compounds we put $\Delta v = \hat{n}_h - 1$, where \hat{n}_h is the operator of the number of holes on the 4f shell. As a result we get that the bare energy E_{0f} of the f-level is renormalized as follows:

$$E_{\rm f} = E_{\rm 0f} - \eta d_{\rm f} e_{\rm B} \tag{3}$$

where $\eta = 1$ for Ce ions and $\eta = -1$ for Yb ions.

The Kondo temperature T_k has been calculated within the Bethe ansatz for the *N*-fold degenerate Anderson model in the infinite-*U* limit [15, 16]:

$$T_{\rm k} = D\Gamma(1+1/N) \left(\frac{N\Gamma}{\pi D}\right)^{1/N} \exp\left[-\frac{\pi(\mu - E_{\rm f})}{N\Gamma}\right].$$
(4)

Here, μ is the chemical potential, the parameter $\Gamma \equiv \pi |V|^2 \rho_F$ characterizes the broadening of the f level, ρ_F is the density of states (DOS) in the conduction band at the Fermi surface, and *D* is the half-width of the conduction band. The equation (4) corresponds to the case $N\Gamma \ll \mu - E_f \ll D$. The energy of the antiferromagnetic coupling *J* between spins of conduction and localized f electrons is equal to

$$J = \frac{|V|^2}{\mu - E_{\rm f}} = \frac{\Gamma}{\pi (\mu - E_{\rm f})\rho_{\rm F}}.$$
(5)

Applying hydrostatic pressure *P* produces a bulk strain

$$e_{\rm B} = -P/c_{\rm B} < 0 \tag{6}$$

where $c_{\rm B}$ is the bulk modulus. Substitution of this relationship into equations (2)–(5) gives the pressure dependence of $T_{\rm k}$ and J.

Using equation (4), we find the Grüneisen parameter:

$$\Omega \equiv -\frac{\mathrm{d}\ln T_{\mathrm{k}}}{\mathrm{d}\ln \mathcal{V}} = -\frac{\mathrm{d}\ln T_{\mathrm{k}}}{\mathrm{d}e_{\mathrm{B}}}\bigg|_{e_{\mathrm{B}}=0} = \frac{2r}{N} + \frac{\pi}{N\Gamma_{0}}(\eta d_{\mathrm{f}} + 2r(\mu - E_{\mathrm{0f}})) \tag{7}$$

where $\Gamma_0 \equiv \Gamma(e_B = 0)$. This equation shows that in Ce compounds the electron-lattice interaction (1) gives an additional positive contribution to Ω because $\eta = 1$. Besides, this contribution may be sufficiently large if $d_f \gg \Gamma_0$. The conventional mechanism (2) also gives

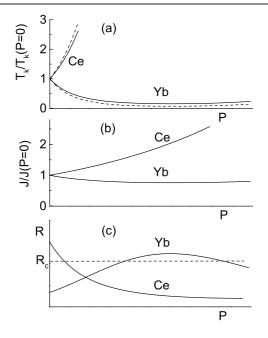


Figure 1. Schematic pressure dependence of the normalized Kondo temperature T_k , the normalized exchange coupling J and the ratio $R = T_{\text{RKKY}}/T_k$ of the RKKY energy scale T_{RKKY} to T_k for Ce and Yb heavy-fermion compounds. The dashed curves in panel (a) demonstrate the influence of the crystal-field splitting on $T_k(P)$ for the case $\Delta_{\text{CF}} = T_k(\Delta_{\text{CF}} = 0)/3$. The dotted curve in panel (c) shows a critical value R_c of the ratio when the quantum phase transition (QPT) occurs.

a large positive contribution to Ω at $(\mu - E_{0f})/\Gamma_0 \gg 1$. These two contributions may explain the very large value of Ω observed in many Ce-based heavy-fermion compounds.

Another situation takes place in Yb compounds where $\eta = -1$ and the Grüneisen parameter Ω is negative at $d_f > 2r(\Gamma_0/\pi + \mu - E_f)$.

The schematic behaviour of $T_k(P)$ and J(P) determined by equations (4) and (5) is shown in figures 1(a) and (b), respectively. In Ce compounds T_k and J increase monotonically with increasing pressure. In Yb compounds with $\Omega < 0$ these energy scales decrease at small P. Then they demonstrate a broad minimum at P_{\min} and start to increase at $P > P_{\min}$.

The Kondo scale T_0 of the Anderson model with spin–orbit splitting Δ_{SO} between the six-fold degenerate ground state multiplet and the eight-fold degenerate excited spin–orbit multiplet may be estimated within a factor of order unity by the expression [17]

$$T_0 \sim D\left(\frac{D}{\Delta_{\rm SO} + T_0}\right)^{8/6} \exp\left[-\frac{\pi(\mu - E_{\rm f})}{6\Gamma}\right].$$
(8)

The crystal-field splitting of the ground state multiplet also influences the Kondo scale. The Kondo temperature T_k corresponding to the crystal-field splitting of the ground state quartet into two doublets may be estimated by the equation [18, 19]

$$T_{\rm k} = T_{\rm k}^2 (\Delta_{\rm CF} = 0) / (\Delta_{\rm CF} + T_{\rm k}).$$
 (9)

Similar results have been derived in [20].

In the case of a pressure-independent crystal-field splitting $\Delta_{CF} = T_k(P = 0)/3$ the schematic dependence of T_k on pressure is represented in figures 1(a) and (b) for Ce and Yb compounds. One can see that in this case the crystal-field as well as the spin–orbit splitting

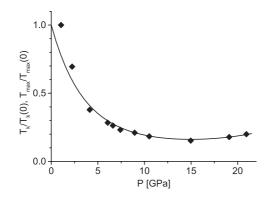


Figure 2. Pressure dependence of the normalized Kondo temperature $T_k/T_k(P = 0)$ calculated from equations (4)–(6) (solid curve) for the parameters given in the text. The full diamonds represent the experimental data [4] for the pressure dependence of the normalized high-temperature resistance maximum $[T_{\text{max}}/T_{\text{max}}(P = 0)]$ of YbCu₂Si₂.

does not change qualitatively the pressure dependence of the Kondo scale in comparison to the case $\Delta_{SO} = \Delta_{CF} = 0$.

3. Change of valency under pressure

Let us consider the influence of pressure on the valency ν of Yb and Ce ions. The Anderson model has interesting regimes parameterized by the f-level broadening Γ . The Kondo regime with almost integer valency of rare-earth ions takes place at $N\Gamma/(\mu - E_f) \ll 1$. The smaller the ratio the closer the valency to the integer value +3. The mixed valency regime with a remarkable deviation of ν from the integer value takes place at $N\Gamma/(\mu - E_f) \sim 1$. According to equation (5), the value of $N\Gamma/(\mu - E_f)$ is proportional to $J\rho_F$. In Yb compounds the value decreases with increasing pressure because the antiferromagnetic coupling J decreases, in accordance with the results represented in figure 1(b). The pressure dependence of the DOS ρ_F is usually weak in comparison with J(P) and may be neglected in our consideration. Therefore, the pressure suppresses valency fluctuations and moves Yb ions towards the localized state Yb³⁺. This result agrees with the data [4] for YbCu₂Si₂ and other Yb compounds. In Ce compounds, applying pressure increases the value of $N\Gamma/(\mu - E_f)$, see figure 1(b), and, in turn, enhances the valency fluctuations. Therefore, applying pressure moves Ce ions from the integer value of $N = 10^{-1} (\mu - E_f)^{-1}$.

4. Comparison with experiment

The pressure dependence of the Kondo temperature may be estimated from resistivity measurements. In heavy-fermion compounds the Kondo effect is responsible for the resistivity peak observed at a certain temperature T_{max} . This phenomenon is caused by the Abrikosov–Suhl resonance [21, 22] at temperature $T = T_k$. In the case when the resistivity maximum is a result of the Kondo effect alone one can assume that the observed temperature T_{max} is proportional to T_k , i.e. $T_{\text{max}}(P) \sim T_k(P)$ (see, for example, [1, 23]). Detailed resistivity measurements in YbCu₂Si₂ were performed in [4] in a wide range of pressures and temperatures. The pressure dependence $T_{\text{max}}(P)$ taken from [4] is plotted in figure 2 together with our theoretical calculations of $T_k(P)$ from equations (4) and (9). One can see that the theory reproduces well the observed pressure dependence of $T_{\text{max}}(P)$ in a wide range of pressures. Our theoretical estimation of the Grüneisen parameter $\Omega = -38$ is slightly larger than the experimental value $\Omega = -27$ [4]. In our calculations we neglected the pressure dependence of crystal-field splitting Δ_{CF} and assumed that Δ_{CF} compares to the value of $T_{\text{k}}(P = 0)$ [24]. We also used the following parameters: $(\mu - E_{\text{f0}})/\Gamma_0 = 4.5$, $d_f/\Gamma_0 = 70$, r = 3.1, $c_{\text{B}} = 168$ GPa [4]. Using a typical value $\Gamma_0 \sim 0.06$ eV gives a reasonable value $\mu - E_{\text{f0}} \sim 0.27$ eV (see, for example, data [12, 25, 26] for various Yb-based compounds) and $d_f \sim 4.2$ eV. This value of the parameter d_f corresponds to a characteristic pressure $P_0 \equiv n_{\text{f}}d_{\text{f}} = 7.1$ GPa which is produced by Yb ions if they are excited from the ground state $4f^{13}$ into the excited state $4f^{12}$ with a larger ionic radius. Here $n_f \equiv N_f/\mathcal{V}$ is the concentration of Yb ions.

The theoretical estimation of Ω for YbInCu₄ was also made in [11] and gave $\Omega = -31$, in good agreement with experimental value $\Omega = -30.6$ [27].

Our calculations represented in figure 2 demonstrate that the account of the electron lattice coupling (1) leads to the pressure dependence $T_k(P)$ which reproduces well the pressure behaviour $T_{max}(P)$ in YbCu₂Si₂ [4]. Recently a similar pressure dependence of the resistivity maximum $T_{max}(P)$ was also observed in YbRh₂Si₂ [5].

However, the electron–lattice coupling (1) is not the only mechanism which may be responsible for the observed pressure behaviour. Magnetic RKKY interactions may also play an important role, especially in the case when T_k becomes small and close to the RKKY energy scale $T_{\text{RKKY}} \propto \rho_F J^2$. The pressure dependence of crystal-field splitting is one more mechanism that may influence $T_k(P)$. This situation probably takes place at high pressures [3–5].

Thus, under a high pressure the RKKY interaction and crystal-field splitting may also influence the pressure dependence of the resistivity maximum. In this case a dependence of the resistivity maximum on pressure is a result of a combined effect of the Kondo effect, RKKY interaction and crystal-field splitting. In order to take into account this effect our model needs further improvements.

5. Influence of pressure on the phase diagram

The RKKY interaction between localized magnetic moments of rare-earth ions favours a magnetically ordered ground state. According to Doniach's approach [29], if $T_{\rm RKKY} \gg T_{\rm k}$, then the Kondo screening is suppressed by the RKKY interaction and a long-range magnetic order in the system of stable localized magnetic moments may appear. In the case $T_{\rm RKKY} \ll T_{\rm k}$, the Kondo effect leads to the formation of a heavy-fermion ground state. Some HF compounds reveal a quantum phase transition (QPT) [28] which can be interpreted as a result of a strong competition between the Kondo effect and the RKKY interaction. In accordance with this physical picture one can assume that the QPT occurs at a certain critical value R_c of the ratio $R \equiv T_{\rm RKKY}/T_{\rm k}$. A region R(P) close to $R_{\rm c}$ is the region of the strongest competition and interplay between magnetism and the Kondo effect. The parameter R can be tuned either by the choice of the material [30] or by pressure P; see, for example, [3-5, 8, 31-33] and references therein. The measurements [3, 8, 31–33] have demonstrated that in Ce heavyfermion compounds the application of pressure decreases R and can move the systems from the state with stable localized moments to the heavy-fermion state through the QPT. Another behaviour was observed in Yb heavy-fermion compounds where the application of pressure increases R and can move the compounds from the heavy-fermion state to the state with stable localized moments [3–5].

Let us consider the pressure dependence of R in the framework of our approach. In figure 1(c) we plot the schematic pressure dependence of the parameter R calculated from

equations (2)–(6) for Ce and Yb compounds. In Ce compounds, applying pressure decreases R as T_{RKKY} increases slower than T_k with increasing P. If at zero pressure we have $R(0) > R_c$, then the system is in the regime with stable magnetic moments. With increasing pressure the parameter R(P) decreases monotonically, and crosses the QPT point $R(P_c) = R_c$ at a critical pressure P_c . In Yb compounds with $\Omega < 0$, the pressure behaviour of R(P) has another character. At low pressure the parameter R(P) increases with increasing P. Therefore, pressure moves Yb systems from the Kondo regime towards a regime with stable magnetic moments. In the case $R(P = 0) < R_c$ and max $R(P) > R_c$, pressure moves the system to the QPT. Such a behaviour was observed, for example, in Yb₂Ni₂Al [3] and YbRh₂Si₂ [5]. At a certain pressure P_m the parameter R(P) achieves a broad maximum and then slowly decreases with increasing P. If max $R(P) < R_c$ then the theory predicts that the QPT does not occur, and the system will stay in the Kondo regime even at high pressure.

Above we have discussed the behaviour of T_k , J and T_{RKKY} under pressure. Unfortunately, this is not enough for a comprehensive determination of the phase diagram of an HF system. Apart from these parameters it is necessary to know the crystal structure and others characteristics. Nevertheless, we believe that the obtained results may be useful for understanding the effect of pressure on the phase diagram. In particular, the fact that in Yb-based HF compounds these parameters achieve a broad minimum at a high pressure $P_{\rm m}$ may explain the weak change of the phase diagram in a wide range of P around $P_{\rm m}$. From this point of view one can understand qualitatively the phase diagram of YbRh₂Si₂ in the (P, T) plane [5]. This compound is interesting due to its proximity to the QPT at the ambient pressure. The pressure investigations of YbRh₂Si₂ [5] revealed a broad region of a lowmoment (LM) magnetic state which exists up to very high pressures $P \approx 11$ GPa. In figure 3 we schematically compare the (P, T)-phase diagrams of YbRh₂Si₂ [5] with the most known Ce-based compound $CeCu_{6-x}Au_x$ [30, 34]. As evident from this figure one finds entirely different types of behaviour: while in the case of YbRh₂Si₂ an LM magnetic state exists in a broad pressure range, no such stable LM magnetic state has been observed in $CeCu_{6-x}Au_x$ and also in other Ce-based HF systems so far. We believe that the existence of this stable magnetic state with a weak pressure-dependent critical temperature may be understood within our approach if we assume that in this compound the parameter max R(P) is slightly larger than $R(P = 0) \approx R_c$. In this case the theory predicts that the parameter R(P) as well as T_k and T_{RKKY} will have a weak pressure dependence in a wide range of P > 0; see figure 1.

6. Conclusions

In conclusion, in the present paper we have analysed and compared the pressure dependence of the Kondo scale T_k , the energy of antiferromagnetic coupling J and the valency of rare-earth ions in Yb- and Ce-based heavy-fermion (HF) compounds. It was shown that the difference in the response on pressure in these compounds can be explained by account of the influence of pressure on both the hybridization and the ionic radius of the rare-earth ions. We demonstrated that it is the latter mechanism that may be responsible for a negative Grüneisen parameter $\Omega = -d \ln T_k/d \ln \nu$ observed in Yb-based HF compounds. In contrast to Yb compounds, this mechanism gives a positive contribution to the Grüneisen parameter of Ce compounds. We have found that in Yb compounds the Kondo scale T_k and the coupling constant J reveal a nonmonotonic pressure dependence with a broad minimum at a high pressure. We have shown that this result explains the unusual behaviour of the resistivity under pressure observed in Yb₂Ni₂Al [3], YbCu₂Si₂ [4] and YbRh₂Si₂ [5]. We have also demonstrated that the proposed mechanism can explain the observed difference in the influence of pressure on the competition between the RKKY interactions and the Kondo effect in Yb and Ce compounds. The theory

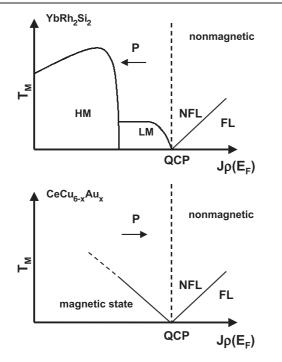


Figure 3. Schematic phase diagrams of YbRh₂Si₂ and CeCu_{6-x}Au_x. Here LM and HM denote low-moment and high-moment magnetic states, respectively [5]. The schematic phase diagram of the metallic alloys CeCu_{6-x}Au_x is deduced from the papers [30, 34] in which the temperature behaviour of the metallic alloys has been studied at different *x* [30] and applied pressures [34]. Note that a partial substitution of Cu ions by Au ions results in a negative internal pressure.

has been compared with available experimental data and qualitative and quantitative agreement has been found.

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