The thermoelectric power of Cerium or Ytterbium Kondo compounds : a review of theoretical models.

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Abstract

The experimental thermoelectric power (TEP) has been measured in many intermetallic Kondo compounds and several different shapes have been observed in Cerium or Ytterbium Kondo systems. However, most of the TEP curves present a peak at high temperatures with respect to the Kondo temperature T_k and generally another peak at low temperatures typically of order T_k. It is well established that the "high temperature" peaks are well explained by the Coqblin-Schrieffer model including crystal field (CF) and that they correspond to a fraction of the CF splitting. This model has been recently improved by considering the "poor man's scaling" theory which renormalizes the exchange integrals, at lower temperatures when approaching T_k. The situation is more complex at low temperatures below T_k in the Fermi liquid regime without CF excited states and the theoretical interpretation is more controversial. We will present firstly results obtained with the single-impurity Anderson model (SIAM), which treats Ce ions as impurities but neglects coherent scattering on different Ce sites. A positive (negative) peak is obtained for Cerium (Ytterbium) compounds and we can account for some TEP data which show two positive peaks separated by a negative minimum at intermediate temperature. Then, the compound $CeRu_2Ge_2$ undergoes a transition from a magnetic order to a non magnetic behavior under high pressure and we present a theoretical explanation of the TEP within the SIAM model in the non-crossing approximation, which gives finally a reasonable agreement with the TEP curves of $CeRu_2Ge_2$ at different pressures.

1. Introduction

The thermoelectric properties of intermetallic compounds with Ce or Yb ions with one f electron or one f-hole present several interesting characteristic features and we will make firstly here a brief summary of the different behaviours observed for the thermoelectric power (TEP) in such compounds. Then, we will make a review of the theoretical models developed to account for the TEP of such systems, by distiguishing between the so-called "high temperatures" and "low temperatures" regimes, i.e. above or below the Kondo temperature T_k. After these two sections, we will discuss in the last section the application of the single impurity Anderson Model (SIAM) treated in the non-crossing approximation (NCA) to the TEP of some Ce or Yb systems and its variation with applied pressure or with the relative concentration of the matrix in ternary or quaternary alloys.

At sufficiently high temperatures much above T_k, the TEP has very often a broad positive peak in Cerium Kondo compounds and a negative one in Ytterbium Kondo compounds, corresponding roughly to a fraction of the crystalline field (CF) effect splitting. These peaks were previously explained by the Coqblin-Schrieffer model (CSM) including CF effects [1] and we will present below an improved description of the TEP behaviour above T_k [2].

The experimental situation is more complex at low temperatures, where several different behaviours have been observed [3]. An extensive review of the experimental data can be found in one of our recent papers [2]. The first type of thermopower curve (a) is characterized by a broad positive peak at roughly 100K and by a deep negative minimum at lower temperatures, while the second type (b) for the thermopower is similar to the type (a), except that, at very low temperatures, the thermopower changes sign again and exhibits an additional positive peak. The type (a) has been observed in a few compounds such as $CeCu_2Ge_2$ [3] and $CeCu_2Si_2$ [4] at normal pressure.

The type (b) TEP has been observed in many compounds, especially if temperature reaches very low values. Type (b) has been observed in $CeAl_3$[5], $CePd_2Si_2$ [6], $Ce_xLa_{1-x}Pd_2Si_2$ [7], CePdGe [8] or CePtGe[8]. In fact, one can say that there is a continuous change between type (a) and type (b) and that for example a type (b) TEP is obtained by continuity from type (a) when measurements are extended at lower temperatures. Such a situation occurs for example in $CeAl_3$. The first experimental results [9] gave a pronounced positive maximum around 60 K, which was accounted for by the CSM model with CF effects [1]. Then, more recent experiments [5] gave a change of sign at roughly 8 K, a weak negative minimum at roughly 4 K, another change of sign at roughly 0.4 K and a small positive maximum at roughly 0.1 K. These features cannot be explained by the unrenormalized 3rd-order perturbation expansion of the CSM [1] but can be better accounted for by the calculations presented previously in detail in ref [2] and here. In some cases the positive low-temperature peak is canceled by superconducting or magnetic transitions. For example, in $CeCu_2Si_2$ [10], the positive low-temperature upturn does not appear in the zero field data and is seen only in an external magnetic field which suppresses the superconducting transition.We can conclude here that the type (a)-TEP is relatively rare and a type (b)-TEP is often obtained when measurements are performed down to very low temperatures.

The third type (c) describes then the TEP which is always positive and has a small positive low-temperature peak, in addition to a large positive high-temperature peak, but without change of sign between the two peaks. This behavior is observed in many Cerium Kondo systems, like for example $Ce_xLa_{1-x}Cu_6$ for small x [11], $Ce_xR_{1-x}Cu_2Si_2$ (with R=La,Y) [12] or CeNiGe [8]. The TEP of $CeRu_2Si_2$ single crystals is anisotropic and has a type (b) bebaviour for measurements along the c-direction and a type (c) behaviour for measurements perpendicular to it; in both cases, the slope of the TEP is always positive at very low temperatures [13]. In some cases, the high-temperature peak is very large and only a shoulder remains on the low-temperature side of that peak, as in $CeCu_6$ and $Ce_xLa_{1-x}Cu_6$ for large x values [11]; this behavior is called type (d).

The TEP of Cerium compounds has been extensively studied under high pressure in $CeCu_2Si_2$ [4], $CeCu_2Ge_2$ [3] and $CePd_2Si_2$ [3] compounds. Pressure reduces the large negative low-temperature peak and gives rise to a positive peak at low temperatures. Finally, at very high pressure, only a positive high-temperature peak with a more or less pronounced shoulder remains. Thus, the TEP of all the compounds mentioned above transforms under pressure from type (a) to type (b) and finally to type (c) or (d).

Doping affects also the shape of the TEP [7,12]. For example, replacing Y ions by larger Ce ions in dilute $Ce_xY_{1-x}Cu_2Si_2$ [12] expands the lattice and transforms S(T) from a single-peak (type (d)) to a two-peak structure (type (c)). A behavior like that is consistent with the '"chemical pressure effects", although doping might involve a charge transfer or change the character of the ground state and is more complex than just the hydrostatic pressure.

Indeed, there are a priori different types of TEP curves, but if the measurements are performed at low enough temperature and with sufficient accuracy, most of the experimental curves for the TEP in Ce Kondo systems show a positive initial slope, S(T)/T, at low temperatures. This low temperature positive peak is generally connected to the high temperature positive peak by a minimum which has very often a negative value.

Next, we discuss briefly the case of Ytterbium intermetallics, which exhibit, generally, a large negative TEP peak at high temperatures. A thermopower with a single negative peak without any additional structure, which mirrors the type (d) behavior of Ce systems, is found in $YbCu_2Si_2$ [14], Yb_2Cu_7 [15] or $YbAgCu_4$ [16]. A negative TEP with a large dip at high temperatures and a small dip at low temperatures is found in $YbPdCu_4$[16] and $Yb_2Rh_3Al_9$ [17] and mirrors the type (c) behavior of Ce systems. The two negative dips separated by a small positive TEP-maximum are found in $YbAuCu_4$ [16] and YbPtIn [18], which mirrors the type (b) behavior, while $YbAu_3$ and $YbAu_2$ [19] with a large negative dip at high temperatures and a positive peak at low temperatures mirror the type (a) behavior of Ce systems. However, the TEP of some Yb compounds like $YbAu_3$, $YbAu_2$, $YbPdCu_4$ or $Yb_2Rh_3Al_9$ become positive and slightly increasing at high temperatures above the negative high temperature extremum corresponding to CF effects. Thus, we can conclude that many Yb compounds have a negative TEP at low and high temperatures often separated by a maximum and that the TEP of Yb compounds and its dependence on pressure and doping present also some interesting features which are often mirroring the behavior of Ce compounds.

There is always a continuous interest in the measurements of the transport properties of heavy fermion systems and in particular of new compounds such as for example skutterudites, because the TEP and the figure of merit can reach large values in such compounds. The TEP can reach very large values, typically 50 to 100 μV/K in such Kondo Ce or Yb compounds and in general in strongly correlated electron systems. On the other hand, the figure of merit $Z= S^2/\kappa\rho$ can be also large in such systems, because the TEP S is large, the thermal conductivity κ is relatively small, but on the other hand the electrical resistivity ρ is generally large in metallic heavy fermion systems.

A full description of Ce and Yb compounds has been recently given in ref. 2. It is clear that the peaks observed at high temperatures in the TEP correlate in general with the magnetic electrical resistivity which is also passing through a maximum and decreasing logarithmically above it [20]. Such a behavior is clearly a good indication that the TEP properties in many strongly correlated electron systems are due to the exchange scattering of conduction electrons on the 4f states of rare-earth ions, as established since a long time. Theory should account for these different types of thermopower and can explain the temperature, pressure and doping dependence of the data.

We present here three theoretical calculations : first the renormalized perturbation theory, i.e. the classical Kondo perturbation theory but with a renormalized exchange integral for describing better the high temperature regime[2]; second the single impurity Anderson model (SIAM) with strong correlations for the low temperature regime[2], and third NCA (non crossing approximation) calculations within the SIAM for presenting an explanation for the pressure dependence of some Cerium compounds like $CeRu_2Ge_2$ [25].

The high-temperature theoretical model

The high-temperature properties of metallic systems with Ce and Yb ions are described by the Coqblin-Schrieffer Hamiltonian including the CF effects [20,21] :

$$H_{cs}=\sum_{\nu} E_{\nu} a_{\nu}^{+} a_{\nu} + \sum_{k\nu} \varepsilon_k c_{k\nu}^{+} c_{k\nu}$$

$$-J_0\sum_{k,k'}\sum_{\nu,\nu'}c^{+}_{k'\nu}c_{k\nu'}(a^{+}_{\nu'}a_{\nu}-\delta_{\nu\nu'}<n_{\nu}>)$$

$$+\sum_{k,k'}\sum_{\nu}\left(V_0-J_0<n_{\nu}>\right)c^{+}_{k'\nu}c_{k\nu} \qquad (1)$$

where all the symbols have their usual meaning [2,21].

The TEP had been previously computed using the Hamiltonian (1) and a peak corresponding to a fraction of the overall CF splitting Δ had been obtained [1]. The sign of the TEP at the peak value depends on the relative magnitude of J_0 and V_0, but the peak is generally positive in the case of Ce compounds and the model developed in ref. 1 accounts fairly well for the TEP of $CeAl_2$ or $CeAl_3$ or related compounds at sufficiently high temperatures. However, the previously derived TEP was decreasing with decreasing temperature T below the peak value down to a zero value at T=0, in disagreement with many experimental results on Ce compounds.

We will explain now the improvement recently reported in ref. 2 for still the high temperature regime, $T>T_K$. The TEP is calculated there by the renormalized perturbation theory (RPT), i.e. we renormalize the coupling constant of the standard 3rd-order perturbation expansion by the "poor man's scaling" [22]. In the case of two CF levels, with respective degeneracies m (for the ground state) and M, for the $4f^1$ or $4f^{13}$ configurations of Ce or Yb ions, the renormalized exchange integral J(T) is given by [22] :

$$\exp\left(-\frac{1}{\rho J(T)}\right)=\left(\frac{k_B T_k}{D}\right)^{m}\left(\frac{k_B T_k+\Delta}{D+\Delta}\right)^{M}$$

The TEP curves obtained in such a way present a maximum corresponding to a fraction of Δ, as previously obtained [1] and the effect of renormalization becomes important at low temperatures when temperature approaches T_k. Since the potential (non-spin-flip) scattering is not renormalized by scaling, the relative magnitude of J(T) changes with temperature and the TEP can now change its sign at low temperatures, i.e., below the maximum in Ce systems or below the minimum in Yb ones, but obviously still above T_k.

Figure 1 shows typical TEP curves for several values of the Kondo temperature : there is a sign change increasing with T_k, and given by $T_x=\alpha T_k$, where, for reasonable parameters, α lies between roughly 2.5 and 10. Thus, there is a large region in the parameter space of the renormalized CSM in which the TEP is negative for Ce systems (or positive for Yb systems). This point is a clear improvement of our present description [2] with respect to the previous one [1] and allows us to give a typical type (a) or (b) TEP curve and to better fit the experimental TEP curves of Ce compounds, as $CeAl_3$ or $CeCu_2Si_2$. Other plots, which can be found in ref. 2, have been done for different values the CF splitting Δ and of the potential scattering V_0. We can notice that the temperature T_x of the TEP sign change increases with T_k, decreases with $|V_0|$ and does not change with Δ. Finally, the RPT breaks down around T_k due to the crossover into the strong coupling regime and we present in the next section a model which is appropriate for the low temperature regime.

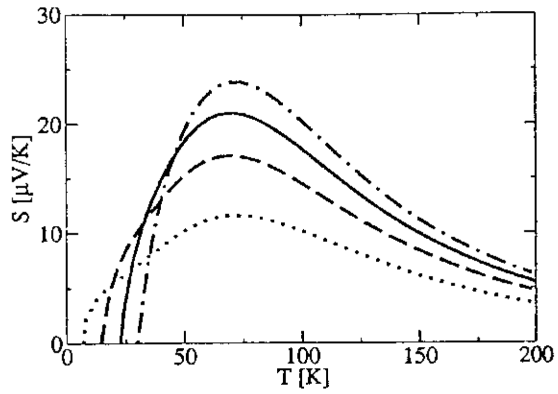

Fig 1. The thermopower plotted as a function of temperature for $\rho_0 V_0$ = - 0.35, Δ = 350 K, and four different values of T_k : dotted curve T_k = 2K; dashed curve T_k = 5K; solid curve T_k = 8K; dot-dashed curve T_k = 11K [2].

The low-temperature theoretical model

The ground state of the CSM is a Fermi Liquid (FL) with the local moment screened by the conduction states, and direct calculations of thermoelectric properties for $T<T_k$ would require a strong-coupling approach. In the general case, we can deal at low temperatures only with the ground state which is often a doublet. Thus, the properties of Ce or Yb systems below T_k can be described by the spin-1/2 Single Impurity Anderson Model (SIAM) in which the 4f ions vary between the zero-electron (hole) and the one-electron (hole) configuration. The low-T part of the TEP has been calculated within the FL approach [2] using the modified perturbation theory (MPT) [23] which gives fairly good results with respect to exact calculations [24]. The low-temperature properties of the CSM obtained in such a way follow the usual FL power laws and the TEP has the typical form $A(T/T_k)$, where A is of the order of $k_B/|e|$= 86.17 µV/K. The initial slope of S is positive for magnetic ions with less than one 4f electron (like Cerium) and negative for ions with less than one 4f hole (like Ytterbium). We also find that S attains a maximum (minimum) at about $T_k/2$ and changes sign at roughly T_k, where the crossover from the FL to the Local Moment regime takes place.

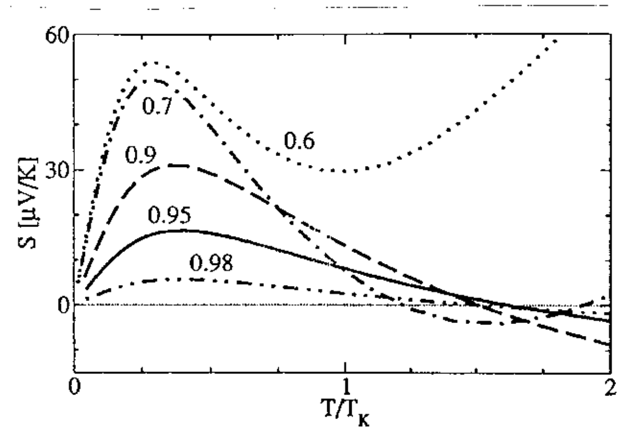

Fig. 2 The thermopower of the SIAM as a function of temperature, measured in units of T_k, shown for various numbers n_f of f electrons (indicated in the figure) and for a large Coulomb integral U = 0.5 W. The corresponding values of T_k /W are 0.011 [for $n_f(0)$ = 0.98] , 0.011, 0.012, 0.025 and 0.035 [for $n_f(0)$ = 0.6], respectively [2].

We have, therefore, derived theoretically the thermoelectric power of Ce and Yb alloys or compounds in the two limits. At high temperatures (i.e. $T > T_k$), we have used a renormalized perturbation theory of the CSM, assuming Kondo scattering on the full CF split multiplet, and at low temperatures (i.e. $T < T_k$), we have used the modified perturbation theory for the SIAM, assuming Kondo scattering on the lowest CF level only. The different results are plotted on Figures 1 and 2.

Starting from the low temperature regime, figure 2 gives positive peaks at a temperature typically of order $T_k/3$ or $T_k/2$ and then the TEP decreases and can become negative around T_k for some values of the different parameters. On the other side, for the high temperature regime, figure 1 gives, with increasing temperature, a TEP starting from zero at $T_x=\alpha T_k$ and increasing up to the maximum corresponding to a fraction of the crystal field splitting. We have no result for the TEP between these two regimes, but we can say that the TEP is always passing through a minimum but, depending on the shape of the low temperature curve and on the precise value of α, the TEP can either remain positive or have a negative minimum. These two solutions correspond to the experimental types (b) or (c) which are observed for the TEP of many Cerium compounds or alloys. It is important to note that the change of parameter values can also change the shape of the TEP. In a previous paper [26], we have shown that increasing the value of the term $|V_0|$ gives an important decrease of the sign change temperature T_x and consequently changes the TEP shape from type (b) to type (c). Our theoretical results have just to be sign-changed when going from the case of Cerium to the case of Ytterbium and the present model [2] yields two negative peaks with either a positive or a negative maximum between them for Ytterbium systems. This theoretical discussion allows to give a fairly reasonable explanation of the TEP in many Ce or Yb systems.

Our theoretical curves can account for a number of experimental curves observed for Cerium compounds. Curves with a negative minimum account for the experimental TEP curves of $CeAl_3$ and $CeCu_2Si_2$, as explained in the introduction. Moreover, the effect of pressure is well described by the change from a curve with a negative minimum to another curve without it and the disappearance of the negative interval under applied pressure has been observed in many Ce compounds such as $CeCu_2Si_2$, $CeCu_2Ge_2$ and $CePd_2Si_2$ compounds. Finally, we can remark that in general our theoretical model does not give a negative peak at low temperatures for Ce systems, except for a few cases with peculiar choices of parameters. Thus, our model cannot yield any (a) behavior, but, as we have remarked in the introduction, this behavior is rarely observed experimentally if the experiments are performed at sufficiently low temperatures or also if the eventual superconducting or magnetic behavior is suppressed or disregarded at very low temperatures.

NCA calculations of the TEP and its variation with pressure.

Then finally we have computed both the magnetic resistivity and the thermoelectric power for the single-site Anderson model by using NCA for deriving the spectral function. The starting point of this study was the transport properties of $CeRu_2Ge_2$ under very high pressure [3,25]. At low pressures, the TEP presents a weak peak around 10 K corresponding roughly to the Kondo temperature and a broader peak around 300 K corresponding to a fraction of the crystalline field splitting, separated by a negative minimum; the minimum becomes positive and weaker with increasing pressure and at very high pressures it remains only one broad peak at high temperatures.

We take the effective single-ion Anderson model, with a large crystal field splitting Δ between a ground state doublet and an excited quartet and with a strong k-f hybridization V. The parameter $\Gamma = \pi V^2/W$ measures the coupling between the 4f and a semi-elliptical conduction band of half-width W. The effect of pressure is complicate to be described, because several parameters are affected by pressure. Generally, pressure does not change Δ and W, but changes both the hybridization V and the relative position of the f-level with respect to the conduction band. It is assumed here that there is no relative change of the f-level energy compared to the center of the conduction band and that the only effect of pressure is to increase V and consequently Γ. Furthermore, Γ and Δ are typically of the same order of magnitude in $CeRu_2Ge_2$ and the influence of pressure is accounted for by an increase of the hybridization from $\Gamma < \Delta$ to $\Gamma > \Delta$. We write the electrical resistivity and the thermopower of the single-impurity Anderson model as a function of the correlation functions, then of the conduction electron scattering rate and finally of the 4f-electron spectral function A(ω). The function A(ω) has several peaks, one centered at –Δ, corresponding to the CF splitting and other ones close to the Fermi level which are caracteristic of the Kondo effect. Indeed, these peaks vary with temperature, pressure and the parameters of the model. All the detailed calculations and in particular the figures of the spectral functions are given in ref. 25.

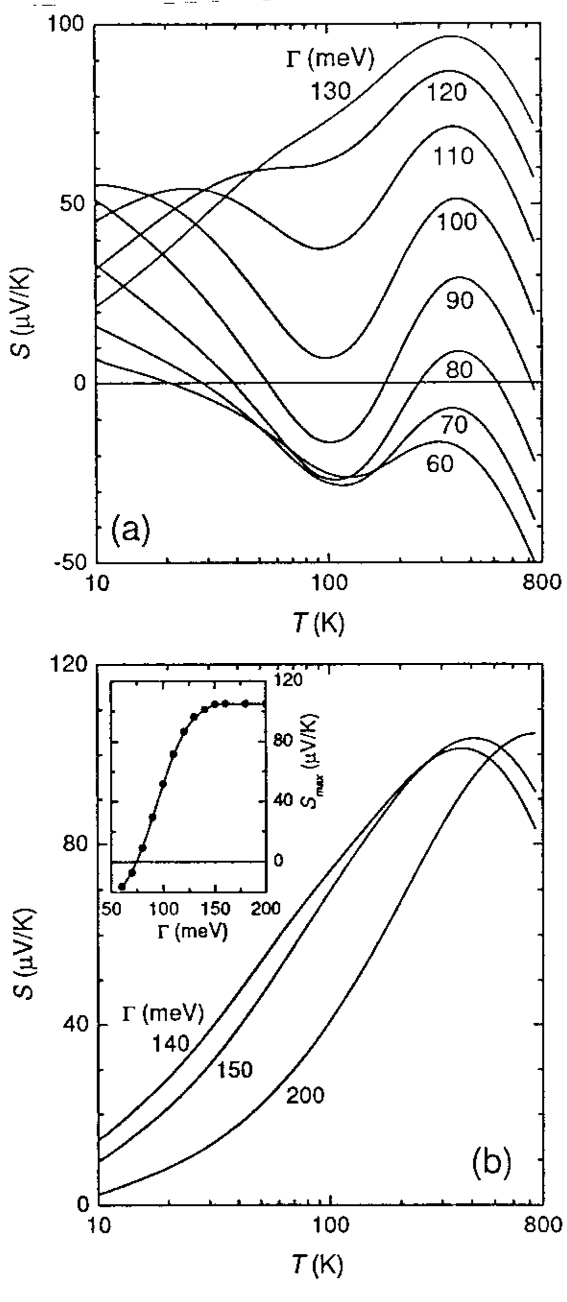

Fig. 3. Temperature dependence of the calculated thermopower S(T) for very small (a) and large (b) values of the hybridization Γ and Δ = 0.07 eV. The inset to (b) shows the Γ dependence of the amplitude S_{max} of the high temperature maximum of the thermopower [25].

We present on figure 3 the calculated TEP for a constant Δ = 0.07 ev and many values of the parameter Γ corresponding to a great variation of the hybridization V under pressure. There is always a high temperature peak at a roughly constant temperature typically of order 0.4 Δ, but the amplitude S_{max} of the TEP at this maximum varies rapidly with Γ, from a negative value for a small Γ value to a very large value for large Γ values. The low temperature regime is more complex : for small Γ, the TEP is passing through a negative minimum before coming back to a positive value at low temperatures; for intermediate Γ values, the TEP remains positive with two peaks and finally for large Γ values, the TEP remains positive and is continuously increasing up to the high temperature peak.

There is a reasonably good agreement between our theoretical results given in figure 3 and the TEP measurements on $CeRu_2Ge_2$: at low pressures, the TEP is almost zero, then it passes through a negative minimum and finally a positive high temperature peak ; for a larger pressure (typically 5.7 GPa), the TEP remains always positive with two peaks corresponding respectively to the Kondo one and the CF one; finally at very high pressures, there remains only one broad and large peak corresponding to both a large Kondo temperature and crystalline field effects. In conclusion, we can say that our model gives a fairly good explanation of the variation of the TEP of $CeRu_2Ge_2$ with pressure.

The NCA used to describe the pressure-induced features in $CeRu_2Ge_2$ is not restricted to this compound. Other ternary Cerium compounds like $CeCu_2Si_2$ or $CePd_2Si_2$ can also be described, if the parameters are adjusted appropriately. The method works also for Yb-compounds if we consider now a decrease of the coupling parameter Γ with pressure.

Conclusions

In conclusion, our present theoretical description yields a clear improvement with respect to the previous model of ref. 1 and in particular can provide a sign change around the Kondo temperature and consequently a negative TEP between two positive peaks in Ce compounds. For an eventual application, it is also interesting to notice firstly that the Kondo temperature varies very much but is generally quite low and second that the values of the TEP can reach very large values, typically of order 50 to 100 μV/K in such Kondo Ce or Yb compounds and in general in strongly correlated electron systems.

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