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Transport properties of the $YbAl_3$ compound: On the energy scales of $YbAl_3$ from thermopower data

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ABSTRACT

We report thermopower and resistivity measurements of an $YbAl_3$ single crystal obtained by the "self-flux" method. Our data reveal Fermi liquid behavior up to $T_{FL} = 35$ K. The Kondo temperature, $T_K \approx 450$ K, was determined from the high temperature thermopower data. The characteristic temperature, T_0, extracted from the temperature slope of the thermopower in the Fermi liquid regime is higher than the T_K. We discuss a possible explanation of the obtained energy scales of $YbAl_3$.

1. Introduction

$YbAl_3$, a valence fluctuating compound, displays a power factor:

$$P = \frac{\alpha^2}{\rho} \quad (1)$$

even higher than Bi_2T_3 [1] (a semiconductor which is among the most commonly used materials today in thermoelectric devices). As a result, $YbAl_3$ has been the subject of much interest. However, in the literature only a few measurements of the thermopower, α, of $YbAl_3$ can be found. Thermopower is an essential property of thermoelectric materials and principally determines their thermoelectric quality (Eq. (1)). We have found only one reported thermopower measurement on a single crystal of $YbAl_3$ [2]. These measurements were performed down to only 10 K, insufficiently low to reveal some important physical characteristics of $YbAl_3$. Additionally, a long time ago, in 1974, the thermopower of $YbAl_3$ was reported on arc-melted ingots [3]. In this paper, we present for the first time the thermopower measured on a monocrystalline sample prepared by the "self-flux" method [4]. Various measurements and investigations on these $YbAl_3$ monocrystals have been reported elsewhere, but not the thermopower.

Using the monocrystalline samples, we expected to observe an increase in the power factor (1) due to a reduction in resistivity. At high temperatures, we have found a resistivity 30% lower than in polycrystalline samples [3] and hot press samples [2]. The resistivity at room temperature is about the same, 30 μΩ cm, as was measured on other monocrystalline samples [2]. At low temperatures, significant differences are found even we compare our results to those measured on monocrystalline samples [2] where the monocrystals were prepared by another method. Thus, our samples show better thermoelectric characteristics (see Eq. (1)) than those reported in Refs [2,3].

$YbAl_3$ is a potential thermoelectric material, but also, due to its high T_K, it is interesting for broader theoretical reasons: it provides a good opportunity for studying some unresolved questions in Kondo lattice systems. The quality of the samples and the precise thermopower measurements enable us to observe some curious features and interesting characteristics at lower temperatures. Hence, in what follows we devote more attention to discussing our experimental results consistent with recently obtained theoretical results on Kondo lattice problem rather than emphasizing the thermopower performance of $YbAl_3$.

2. On the energy scales of Kondo system

One of the main unresolved questions concerning a Kondo lattice system, besides the character of the underlying physics of the coherent heavy fermion state at low temperatures, is its energy

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scales. Historically, the problem of the energy scales existed for a long time for impurity Kondo systems. The low temperature logarithmic upturn in the resistivity of an AuFe alloy was noticed in 1934 [5] and was explained in 1958 by Kondo [6]. He attributed this behavior to the scattering of conduction electrons on the spin of the dissolved atom, which has an open shell (3d-, 4f-, or 5f-) in a non-magnetic matrix (like Au or, for example, $LuAl_3$). Theoretical solution for the resistivity in local moment (LM) regime, which describes this upturn in resistivity, is given by [6]:

$$\rho \propto -\ln\left(\frac{T}{T_K}\right) \tag{2}$$

where the characteristic temperature of the LM regime is called the Kondo temperature, T_K. The solution has an unphysical infinity at low temperatures. Approaching zero temperature, the Kondo interaction assumes the strong coupling limit and the spins of the dissolved ions become screened by conducting electrons thus forming dynamical Kondo singlets and, hence, the resistivity saturates. If on-site interaction, i.e., Kondo interaction, overwhelms inter-site or RKKY interaction ($T_K \gg T_{RKKY}$), the system can be described by mean-field theory in Landau's Fermi liquid picture. Thus Fermi liquid (FL) regime emerges below the LM region and can be evidenced by the resistivity in form:

$$\rho = \rho_0 - A\left(\frac{T}{T_0}\right)^2 \tag{3}$$

T_0 denotes the characteristic temperature of the Fermi, or, better, Landau liquid regime and can be extracted from the relations describing physical quantities in this region. While the resistivity follows the T^2 law, the magnetic susceptibility is constant, specific heat and thermopower are linear in T. The temperature dependences of these physical quantities are the same as for a system of free electrons, which is inherently a Fermi liquid system. The expression for thermopower in FL regime can be written in form:

$$\alpha = B\left(\frac{T}{T_0}\right) \tag{4}$$

In the free electron model:

$$B = \frac{\pi^2 k_B}{3e} \tag{5}$$

and the characteristic temperature T_0 is the Fermi temperature, T_F [7].

After Kondo's work in 1958, a key question was the relationship between T_K and T_0. According to Wilson's renormalization theory, T_0 is equal to T_K [8]. Consequently, behavior of the physical quantities of a Kondo impurity system is determined by only one scale: T_K (if $T_K \gg T_{RKKY}$), although one can distinguish two physically different regimes in Kondo impurity systems. Thus, T_K is between the upper temperature at which the Fermi liquid laws are valid (T_{FL}) and the temperature where the logarithmic term in resistivity begins. Hence, $T_K \approx T_{FL}$ (but $T_{FL} < T_K$), and, according to Wilson's theory, $T_K = T_0$ is valid for Kondo impurity systems.

In Kondo lattice systems, T_K again denotes the beginning of LM region. At the lowest temperatures, FL regime develops if $T_K \gg T_{RKKY}$. The physical quantities have the same temperature dependence as in the FL regime of Kondo impurity systems. Although the resistivity follows the T^2 dependence, it should be noted that FL regime in Kondo lattice systems is connected with coherency due to electronic scattering on the regular array of Kondo ions. Therefore, instead of saturation (Eq. (3)), the resistivity decreases tending towards very low values of ρ_0 but again with the T^2 dependence:

$$\rho = \rho_0 + A\left(\frac{T}{T_0}\right)^2 \tag{6}$$

For Kondo lattice systems, the same/similar question as for Kondo impurity systems arises: what is the characteristic temperature T_0 in the fully coherent state of a Kondo lattice system which is characterized by Fermi liquid behavior of physical quantities? Is it (A) T_K, as in Kondo impurity systems, or (B) T_{FL}, the highest temperature of FL region, or (C) something else. Many papers have discussed the relationship between T_K and T_{FL}; however, T_0 is often neglected or simply taken as T_{FL} [[17–20] and references therein].

Because the thermopower is sensitive to the shape of the density of electronic states, $g(\varepsilon)$, at the Fermi level E_F, it is possible to shed light on some of the above considerations by comparing thermopower data with recently published theoretical investigations on Kondo lattice systems.

3. A review of the investigations on $YbAl_3$

$YbAl_3$ is interesting for theoretical investigations of Kondo lattice systems because of its high Kondo temperature, T_K, but also due to its simplicity. As T_K increases, thermopower at higher temperatures in Kondo systems of 4f-ions typically evolves from a two-peak structure [9,10] towards one-peak behavior, which is a characteristic of intermediate valence, or valence fluctuating systems, like UAl_2 [11]. This means that crystal electrical field effects (CEF) diminish with increasing T_K and are not present in $YbAl_3$. Further, $YbAl_3$ crystallizes in a cubic unit cell and neither anisotropy nor dimensionality plays an important role in its behavior. $YbAl_3$ is a Kondo lattice compound because in each unit cell a Kondo ion Yb exists. $YbAl_3$ also displays moderately heavy fermion (HF) behavior. The hybridization between the f-electrons and conduction electrons is strong and charge fluctuations of the Yb 4f-electrons become energetically favorable resulting in a non-integral valence of Yb; hence, the term intermediate valence, or valence fluctuating system might be the best description of $YbAl_3$.

Experimental evidence of the strong hybridization is also found in the broad maximum at 125 K in the magnetic susceptibility, $\chi(T)$, and in the specific heat, $c_V(T)$ around 100 K, corresponding to a Kondo temperature $T_K \approx 650$ K [12]. Inelastic neutron scattering experiments show a broad Lorentzian spectrum centered at about 40 meV corresponding to a Kondo scale $T_K \approx 450$K [13,14]. In addition, these experiments show a narrow peak at about 30 meV associated with a hybridization gap that vanishes above 50 K.

Optical conductivity measurements at 7 K reveal a narrow Drude-like response corresponding to heavy quasiparticle masses, $m_{eff} = 25–30\, m_e$, and another mid-infrared (IR) peak at ~0.15–0.2 eV associated with the formation of a pseudo-gap or hybridization gap. Above 40 K, the Drude peak broadens and the mid-IR peak is suppressed, but below 40 K, the optical spectra do not change appreciably, indicating a fully coherent ground state is formed and persists up to $T_{FL} \approx 40$ K [15]. De Haas van Alphen experiment gives $m_{eff} = 14–23\, m_e$ [16].

Based on the temperature dependence of magnetic susceptibility, the authors of Ref. [12] concluded that two energy scales exist in $YbAl_3$ and that the high temperature scale is $T_K \approx 500$ K and the low temperature one is $T_0 = T_{FL} \approx 40$ K. (It should be noted that in Refs. [12–16] the authors use the notation T_{coh} for the temperature where the physical quantities obey the Fermi liquid characteristics). We note that their inferred value of T_0 for $YbAl_3$ is different from ours obtained from thermopower data.

4. Experimental

The thermopower was measured on an $YbAl_3$ monocrystal grown by the "self-flux" method [4]. From a single crystal with dimensions of about 10 mm × 10 mm × 10 mm, samples were cut to avoid twinning. The sample used for the thermopower measurement was 6 mm × 2 mm × 0.5 mm. A second measurement to validate the obtained result of the first sample was also performed. The main characteristics of the thermopower were the same for the second sample, but

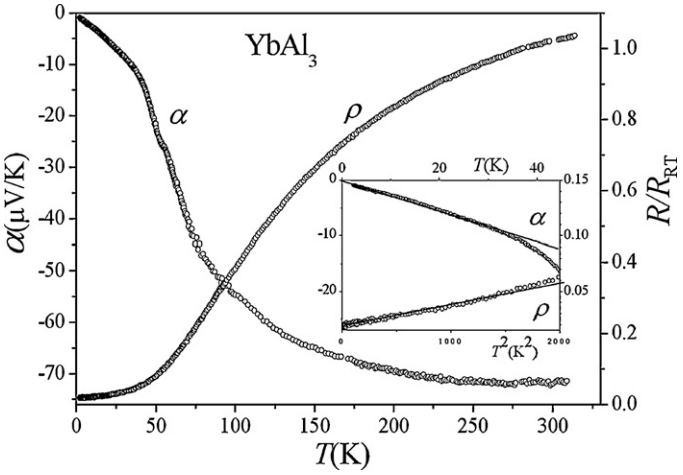

Fig. 1. The thermopower and the resistivity normalized to the room temperature value of $YbAl_3$ vs. temperature. Inset: The thermopower vs. temperature T (upper scale) and the resistivity vs. T^2 (bottom scale).

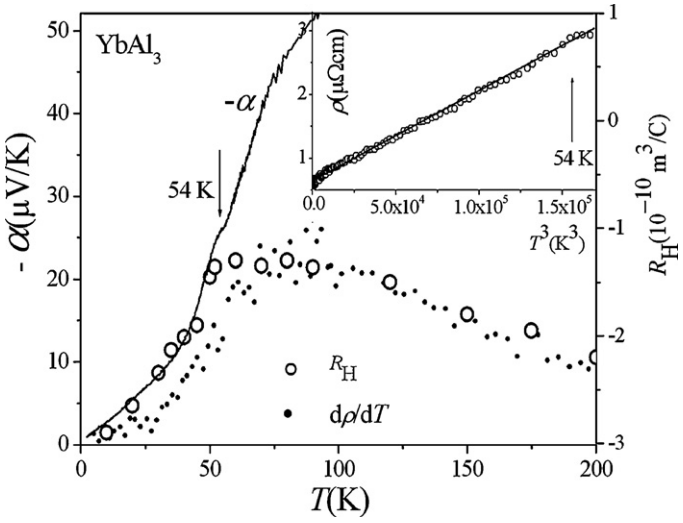

Fig. 2. The negative of the thermopower, $-\alpha$, the Hall constant, R_H [12] and the derivative of resistivity, $d\rho/dT$ (in arbitrary units), of $YbAl_3$ vs. temperature. The arrow shows the kink in the thermopower, consistent with the change of slope of R_H and the change of the slope of $d\rho/dT$ near 52 K. In the inset the resistivity is displayed vs. T^3.

with larger error bars, especially at high temperatures, due to the shortness of the second sample (4 mm). The sample for the resistivity measurement was as thin as possible, 6 mm × 0.5 mm × 0.3 mm, in order to maximize measured resistance. The measurements of the resistivity and thermopower were performed in the ⟨1 0 0⟩ direction of the (100) plane.

The room temperature value of the resistivity of our sample was $\rho_{RT} = 30.4\ \mu\Omega$ cm and the residual resistivity $\rho_0 = 0.54\ \mu\Omega$ cm. These values are similar to those reported in Ref. [12] and indicative of high sample quality. The differential method for thermopower measurements was used in an automated configuration. In contrast, in Ref. [3] the integral method, seldom used today, was applied. The thermocouple was Au7%Fe – chromel pair. The thermocouple ends were glued on the sample to the voltage wires as close as possible ensuring no electrical but good thermal contact using GE varnish. The temperature difference between the ends of the sample was roughly 0.1 T at the lowest temperatures slowly increasing to 0.015 T at 100 K and about 1.5 K above. The temperature difference in Refs. [1] and [2] was 4 K. Both the resistivity and thermopower were measured in reversing mode and in the same run. The resistivity was measured by the four point method and the copper current and voltage leads were attached on the (1 0 0) plane of the sample by silver paint as were the thermovoltage leads for the thermopower measurement.

5. Results

The temperature dependence of the thermopower of $YbAl_3$ is shown in Fig. 1 and is typical of IV compounds [11]. The thermopower is negative, consistent with the observed Hall effect [12]. The thermopower has an extreme value of −72 μV/K at 300 K, similar to Refs. [1–3]. This value is close to −86 μV/K, the maximum expected value of the thermopower of a metal [7].

In many reports, the position of the extreme of thermopower, T_{ext}, is taken as the Kondo temperature. However, a better estimate of T_K is $T_K \approx 1.2$–$1.8\ T_{ext}$, as shown in Ref. [9], based on calculation within the Anderson model [21]. By taking $T_K = 1.5T_{ext}$, we estimate $T_K = 450$ K. This value is comparable to estimates from various other physical quantities [12,14–16].

The resistivity is also displayed in Fig. 1 and suggests the onset of coherency, a characteristic of Kondo lattices, above room temperature, consistent with a T_K far above 300 K.

From the inset of Fig. 1, one sees that the thermopower of $YbAl_3$ shows linear dependence up to about 35 K, which is characteristic of thermopower in FL regime (Eq. (4)). The inset shows that the resistivity obeys Eq. (6) up to 35 K, consistent with Ref. [12].

As a test for HF character of a compound one takes the extended Kadowaki–Woods relation [22]:

$$\frac{A}{\gamma^2} = \frac{1 \cdot 10^{-5}}{\left[N(N-1)/2\right]} \tag{7}$$

A is the coefficient in (7), γ is linear temperature coefficient of specific heat at $T \to 0$ and $N = 2J + 1$ is the degeneracy. Our resistivity data give $A = 54.68 \times 10^{-5}\ \mu\Omega$ cm/K^2. Taking $\gamma = 40.65$ mJ/mol K^2 [12], one obtains $A/\gamma^2 = 0.034 \times 10^{-5}\ \mu\Omega$ cm/(mol K/mJ)^2. Assuming the degeneracy of the Yb^{3+} ion ($J = 7/2$) is $N = 2J + 1 = 8$, the right hand side of Eq. (6′) gives $0.036 \times 10^{-5}\ \mu\Omega$ cm/(molK/mJ)^2. This is very close to the experimental value indicating that the enhanced effective mass originates from quasiparticle–quasiparticle scattering.

A similar relation to Kadowaki–Woods exists for thermopower [23,24]:

$$q = \frac{\left|\alpha/T\right|}{\gamma_m} N_F\,|e| \tag{7′}$$

where α/T is the temperature slope of thermopower in FL regime, e is the charge of an electron and N_F is Faraday's number $N_F e = 9.64 \times 10^4$ C. Theoretically, the absolute value of the dimensionless quantity q should be between 2/3 and 1. Taking $\alpha/T = -0.28\ \mu$V/K^2 from our experiment, we obtain $q = 0.66$, consistent with the theoretical prediction.

Above the FL region, we found that the resistivity obeys the T^3 law (inset of Fig. 2). This behavior is similar to that found in the $Yb_xIn_{1-x}Cu_4$ ($x \le 0.3$) alloy system: at the lowest temperatures there is the T^2 dependence followed by T^3 behavior which persists in these alloys up to the valence transition [25].

An intriguing small kink from 52 K to 56 K appears in the thermopower. This feature was unexpected and we have verified its existence several times in both temperature increasing and decreasing mode of measurements. Interestingly, we note that at 52 K, the Hall coefficient, R_H, also has a drastic change showing a maximum between 52 and 75 K (Fig. 2). Such a behavior of R_H showing a maximum is seen in many heavy fermions and can be described above the maximum by [[26] and the references therein]:

$$R_H = R_O + 4\pi\chi(T)R_S \tag{a1}$$

and also in some cases by a similar anzatz:

$$R_H = R_O + 4\pi\chi(T)\rho_{mag}(T)r_S \tag{a2}$$

where R_O and R_S are temperature independent ordinary and extraordinary Hall coefficients and r_S a constant. Both relations suggest that R_H decreases when magnetic susceptibility χ and

magnetic resistivity ρ_{mag} decrease with increasing temperature. It happens about and above T_K where the coherence is completely absent. But in $YbAl_3$, the decrease of the R_H happens at much lower temperature than T_K and much lower then is the maximum in magnetic susceptibility; just above the maximum in R_H, the susceptibility and resistivity still increase. That means that the maximum in R_H is not connected with the reconstruction in the Fermi surface, but rather with the transition into the fully coherent state, at least in $YbAl_3$. This conclusion is strengthen by the behavior of the resistivity. It is known that resistiviy is the most sensitive physical quantity on coherence. In Fig. 2, it can be seen that the derivative of the resistivity has also a wide maximum just between 52 K and 75 K.

6. Discussion

The thermopower data presented here for $YbAl_3$ yield about the same value of the Kondo temperature, $T_K \approx 500$ K, and, together with the resistivity data, the same value for the Fermi liquid temperature, $T_{FL} \approx 40$ K, as was inferred from other physical quantities [12,14–16].

From the slope $\alpha/T = -0.28\ \mu V/K^2$ for T tending zero, one can estimate T_0. Landau theory allows us to use the relations of the free electron model for estimation. Eq. (4) together with Eq. (5) gives $T_0 = 1020$ K. This result for T_0 seems strange because $T_0 > T_K$. According to the picture coming from Kondo impurity systems, one would expect $T_0 = T_K$. One could argue that this result is due to the fact that we have taken constant B (5) coming from the free electron model, while in FL region of Kondo lattice system one has to assume existence of free quasiparticles but not free electrons. In any event, the use of the free electron formula for Kondo lattice systems in FL regime is typical.

The low temperature transport coefficients of the degenerate periodic SU(N) Anderson model have been calculated in the limit of infinite correlation between f-electrons, within the framework of dynamical mean-field theory [24]. The result for the constant B for $n_f \approx 1$ (Eq. (6) in Ref. [24]) can be written in form:

$$B_q = \frac{4\pi^2 k_B}{n_f e} \tag{8}$$

In the case of Yb^{3+}, there is one hole in the f state and hence $n_f = 1$. This result leads to a value of T_0 that is twelve times higher than that deduced from Eq. (5).

Further, using the free electron formula for specific heat, C_V [27]:

$$C_V/T = \gamma_e = \frac{1}{2}\pi^2 R\frac{Z}{T_0} \tag{9}$$

And taking $\gamma_e \equiv \gamma_m$ from ref. [12] and the number of valence electrons, $Z = 1$, one finds a characteristic temperature $T_0 = 1010$ K. The result deeply depends on Z. We could take $Z = 3$ as there is the Yb^{3+} ion in $YbAl_3$, but even if one would take $Z = 0.5$, the result is $T_0 > T_{FL}$.

In the case of valence fluctuating systems, in order to take the degeneracy of the ground state into account, one often takes Rajan's theoretical result [28]:

$$C_V/T = \gamma_e = \frac{N-1}{6T_0}\pi R \tag{10}$$

Taking the degeneracy $N = 8$, one obtains $T_0 = 748$ K. Thus, all of these estimations give $T_0 \gg T_{FL}$ and $T_0 > T_K$.

This result, as noted above, is counterintuitive and only one other compound displays the same characteristics: $YbInCu_4$ (as well as some alloys derived from $YbInCu_4$) [25,29,30]. $YbInCu_4$ is famous for its valence transition at $T_V = 42$ K with only a very small associated change of lattice parameter. The characteristic temperature of the low temperature phase, which is the HF phase, is about 480 K. The characteristic temperature of the high temperature phase, which is the LM phase, is 25 K [25,29], or less than 2 K according to Ref. [31]. Thus, according to our notations for $YbInCu_4$ it is: T_0 (480 K) > T_{HF} (42 K) > T_{FL}. Also: $T_0 > T_K$ (25 K or 2 K).

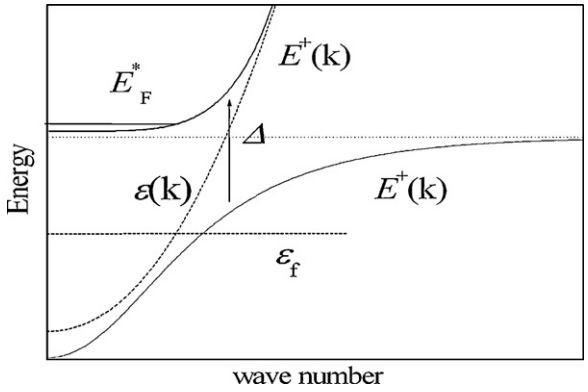

Fig. 3. Dispersion relation $E(k)$ of the hybridized f-level ε_f with conducting electrons represented by the s-like band $\varepsilon(k)$. The renormalized gap Δ is about 40 K.

In Figs. 1 and 2, one can notice that the sequence of the various temperature dependences of the resistivity is the same in $YbAl_3$ and in $YbAg_xIn_{1-x}Cu_4$ ($x \leq 0.275$) [25].

In the alloys of $YbAg_xIn_{1-x}Cu_4$ ($x \neq 0$), the transition is much smoother than in the compound, $x = 0$. This is best seen in the thermopower as a cusp (Figs. 3 and 4 in Ref. [25]) but not a jump (Fig. 1 in Ref. [25]). There is no cusp at the thermopower minimum of $YbAl_3$ as in $YbAg_xIn_{1-x}Cu_4$, but the kink at about 50 K (52 K–56 K) might be the temperature interval where the low and high temperature phases meet.

In what follows, we discuss some physical processes which could limit the coherence in $YbAl_3$, perhaps explaining why the fully coherent (FC) state and associated FL regime persists up to 35 K only, although the characteristic temperature of the FL regime, T_0, is much higher.

Successful fitting of the resistivity data above 35 K to the T^3 dependence (inset of Fig. 2) indicates the existence of strong electron–phonon interaction. Accordingly, one might speculate that phonons destroy FC state, which is characterized by FL behavior.

The Periodic Anderson (PA) Hamiltonian is supposed to give the best description of a Kondo lattice system [32]. The hybridization between conducting electrons and the Kondo ions in a Kondo lattice leads to a dispersion relation, $E(k)$, schematically shown in Fig. 3. The lower band, $E^-(k)$, is full and the renormalized Fermi energy or Fermi level, E_F^*, is at the bottom of the upper band $E^+(k)$. In $YbAl_3$, there are enough electrons from Al to fill the lower band. In other words, they fill the f-level of Yb. The remaining electrons are in the upper band. The electronic character of the band is responsible for the negative sign of the thermopower and the Hall constant. As already noted, the temperature interval in which coherence is inferred from our thermopower data is compatible with the recent optical conductivity measurements on $YbAl_3$. Optical conductivity reveals that the hybridization gap, Δ, and the Drude features do not change below 40 K [15], implying that the renormalized bands and, therewith, the coherence regime are fully developed below 40 K. Conversely, above 40 K thermal energy is sufficient to enable electronic transitions from the lower to upper band over the renormalized gap and these transitions might be the most probable reason for destroying coherence.

A recently published theoretical investigation of Kondo lattice systems by Burdin and Zlatić suggests the possibility of $T_0 > T_K$ and not just $T_0 = T_K$ and $T_0 < T_K$ [33]. These relations depend on the slope of the density of states at the Fermi level. For systems with

$T_0 > T_K$, this theory predicts an abrupt evolution from FL towards LM regime. In $YbAg_xIn_{1-x}Cu_4$ ($x \leq 0.275$) alloy system, the transition is realized by the valence transition. In $YbAl_3$, the transition is also much faster than in other Kondo lattice systems.

7. Summary

We have presented the thermopower of the valence fluctuating system $YbAl_3$, measured for the first time on monocrystals grown by the "self-flux" method [4,12,14–16]. From the high temperature data we have determined the Kondo temperature $T_K = 450$ K. The Fermi liquid region stretches up to $T_{FL} = 35$ K. From the temperature slope of the thermopower up to T_{FL}, we estimate $T_0 = 1020$ K. This result is unexpected because $T_0 > T_K$. The main novelty of the Burdin–Zlatić theory [33] is the fact that it permits all possibilities in HF systems: $T_0 < T_K$, $T_0 = T_K$ and, even, $T_0 > T_K$. According to the theory, this relationship between T_0 and T_K depends on the slope of the electronic density of states at the Fermi level, emphasizing the importance of thermopower measurements in such systems.

Similar to the behavior we observe in $YbAl_3$, $YbAg_xIn_{1-x}Cu_4$ also displays $T_0 > T_K$, or, in terms of Refs. [26,29,30], the characteristic temperature of the low temperature phase, T_L, is higher than the characteristic temperature of the low temperature phase, T_H. Among other similarities, the T^3 dependence in the resistivity after the T^2 one was found in both systems, which, as far as we know, has not been reported for other HF systems.

In conclusion, we have observed a HF system, $YbAl_3$, in addition to $YbInCu_4$, for which $T_0 > T_K > T_{FL}$. At the present, we are not able to distinguish among two possible explanations, the Burdin–Zlatić theory and strong electron–phonon interaction, which is responsible for $T_0 \neq T_{FL}$ in $YbAl_3$. But the best view at present perhaps is: the justification of the relation $T_0 > T_K$ one can find within the Burdin–Zlatić theory, and $T_0 \neq T_{FL}$, i.e., $T_{FL} < T_0$ we try to explain by strong electron–phonon interaction, which destroys the coherence state in $YbAl_3$. We think that our result is a good justification of the Burdin–Zlatić theory. Here we have presented that thermopower is a proper experimental technique to reveal the relationship among characteristic temperatures of a Kondo lattice system: T_0, T_K and T_{FL}.

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