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Deviation from the Kadowaki–Woods relation in Yb-based intermediate-valence systems

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Abstract

The T^2-coefficient of the electrical resistivity, A, is compared with the electronic specific heat coefficient, γ, for a number of Yb-based compounds. It is revealed that many systems, including YbCuAl, $YbInCu_4$, $YbAl_3$, and $YbCu_5$, show A/γ^2 values close to 0.4×10^{-6} $\mu\Omega$ cm mol^2 K^2 mJ^{-2}, which are remarkably small compared to those obtained from an expression known as the Kadowaki–Woods relation: $A/\gamma^2 = 1.0 \times 10^{-5}$ $\mu\Omega$ cm mol^2 K^2 mJ^{-2}. Empirically, the compounds with the smaller A/γ^2 values appear to show weak intersite magnetic correlation and/or to have almost fully degenerate ($J = 5/2$ or 7/2) ground states.

1. Introduction

The low-temperature properties of many U- and Ce-based intermetallic compounds are well described as those of a Fermi-liquid state [1]. Here, the specific heat C and the electrical resistivity ρ as a function of temperature T vary as $C \propto \gamma T$ and $\rho \propto AT^2$, where γ and A are constant. The coefficients γ and A are related to the electron effective mass m^* as $\gamma \propto m^*$ and $A \propto (m^*)^2$, respectively. The ratio A/γ^2 therefore does not depend on m^*. In fact, Kadowaki and Woods [2] showed that many U- and Ce-based compounds show a universal relation, $A/\gamma^2 = 1.0 \times 10^{-5}$ $\mu\Omega$ cm mol^2 K^2 mJ^{-2}. Furthermore, this relation has proved to be applicable for d-electron systems with large γ—for example, A15-type intermetallics such as V_3Si [3], nearly ferromagnetic itinerant-electron systems such as YCo_2 [4], and metallic oxides close to the metal–insulator transition such as V_2O_3 under high pressure [3] and LiV_2O_4 [5]. These observations demonstrate that the Fermi-liquid state is a general feature for a wide range of correlated-electron systems.

Large γ has also been reported in a number of Yb-based intermetallics [6]. One naturally expects the Kadowaki–Woods relation, $A/\gamma^2 = 1.0 \times 10^{-5}$ $\mu\Omega$ cm mol^2 K^2 mJ^{-2},

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to also hold for these Yb compounds. For several Yb-based compounds, however, the A/γ^2 values have been reported to be remarkably small compared to those from the Kadowaki–Woods relation. For $YbCu_4Ag$ [7, 8] and $YbCu_{5-x}Ag_x$ [9, 10], the A/γ^2 values are 0.1–0.6 $\times 10^{-6}$ $\mu\Omega$ cm mol^2 K^2 mJ^{-2}, nearly two orders of magnitude smaller than that for the Kadowaki–Woods relation. It is also reported that the A/γ^2 values for $YbCu_{4.5}$ [11, 12] and $YbNi_2Ge_2$ [13] are close to this value. In addition, this value coincides well with the ratios for transition metals such as Re, Os, and Ni [3, 14].

This deviation of the A/γ^2 value from the Kadowaki–Woods relation poses quite an interesting issue to the Fermi-liquid theory, if it is a general phenomenon for a class of materials. However, there have been no systematic studies on the A/γ^2 values for Yb-based compounds. We have therefore investigated the A/γ^2 relation for several Yb-based compounds. We have measured the electrical resistivity for the Yb-based compounds $YbAl_2$, $YbInAu_2$, $YbInCu_4$, and YbCuAl. These systems are typical examples of Yb-based intermediate-valence or heavy-fermion compounds. We also have examined the A/γ^2 values for several other compounds using values reported in the literature.

2. Experimental details

Polycrystalline samples of $YbAl_2$ and YbCuAl have been prepared by argon arc melting and subsequent annealing. To compensate for the loss of Yb due to evaporation, a small excess of Yb was used in the synthesis. A polycrystalline sample of $YbInAu_2$ was prepared by melting the elements in a BN crucible placed in an evacuated silica tube, and slowly cooling. For $YbInCu_4$, a single-crystal sample was prepared by an In–Cu flux technique [15]. Powder x-ray diffraction shows that single-phased samples were obtained for all the compounds. The electrical resistivity was measured by a standard dc four-probe technique in the temperature range between 4.2 and 300 K.

3. Results

Figure 1 shows the electrical resistivity ρ as a function of temperature T. The residual resistivities ρ_0 of these samples are almost the same as or smaller than those reported in the literature [11, 15–19]. ρ for $YbInCu_4$ decreases abruptly at 40 K due to the valence transition of Yb [15]. The sharp transition indicates that our crystal is well ordered [20]. YbCuAl shows a relatively rapid decrease below approximately 20 K. This temperature roughly corresponds to the characteristic temperature T^*, below which the coherence between Kondo singlets develops and the system enters the Fermi-liquid regime [21]. For $YbInAu_2$, T^* is of the order of 70 K. For $YbAl_2$, ρ does not show a distinct temperature dependence up to 300 K. This indicates that T^* for $YbAl_2$ is above room temperature.

In figure 2, $\rho - \rho_0$ is plotted as a function of T^2. For YbCuAl, $YbInAu_2$, and $YbAl_2$, $\rho - \rho_0$ is described by the T^2-power law at low temperatures, indicating the evolution of the Fermi-liquid state. For $YbInCu_4$, $\rho - \rho_0$ deviates from the T^2-dependence above 20 K, and the data up to 30 K are well fitted with the correction of T^5-dependence, as is shown in figure 2(b). This suggests that the conventional electron–phonon scattering is important in $YbInCu_4$.

The values of the T^2-coefficient A have been obtained by fitting the data at low temperatures, and these are listed in table 1. In these A-values, errors of the order of 20% can exist due to the inaccuracy in estimating the sample dimension. For $YbAl_2$ and $YbInCu_4$, however, the error in A can be much larger because of the small value of A and the presence of the T^5-term.

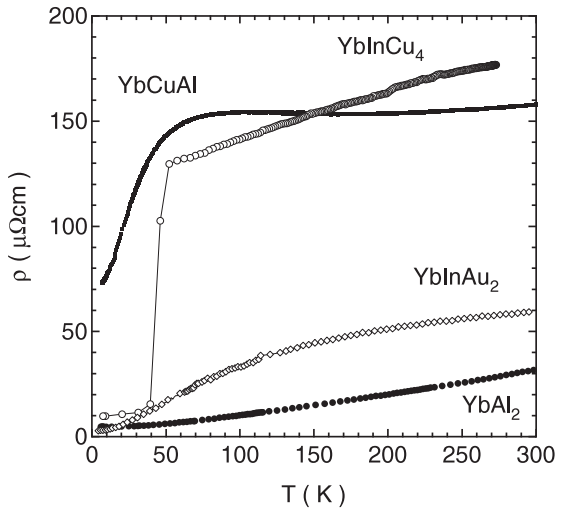

Figure 1. The electrical resistivity ρ for $YbAl_2$, $YbInAu_2$, YbCuAl polycrystals and flux-grown $YbInCu_4$ single crystal as a function of temperature T.

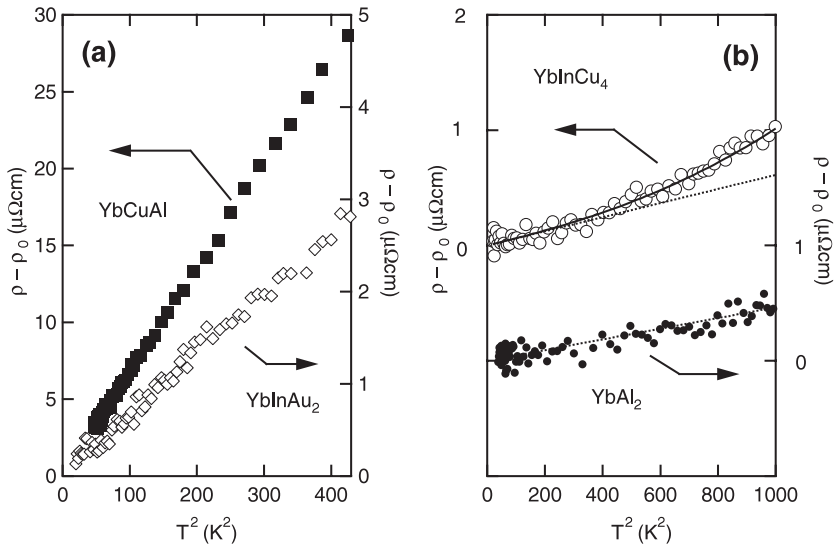

Figure 2. Electrical resistivity with the residual resistivity subtracted, $\rho - \rho_0$, as a function of the square of temperature, T^2. The dotted lines in (b) indicate the T^2-power law. The solid curve for $YbInCu_4$ is the result of a fitting by the sum of T^2- and T^5-terms.

The A-values are plotted against γ on the Kadowaki–Woods plot [2–4], as shown in figure 3. We also plot the A- and γ-values of several systems reported in the literature. The values of A and γ are listed in table 1.

In figure 3, one can see that the Kadowaki–Woods relation, $A/\gamma^2 = 1.0 \times 10^{-5}$ $\mu\Omega$ cm mol^2 K^2 mJ^{-2}, is almost preserved for $YbNi_2B_2C$, $YbRh_2Si_2$, $Yb_2Co_3Ga_9$, CeNi, and $YbInAu_2$. On the other hand, it is apparent that several systems show a significant deviation from the relation. In particular, there appears to exist another 'universal' relation: $A/\gamma^2 \simeq 0.4 \times 10^{-6}$ $\mu\Omega$ cm mol^2 K^2 mJ^{-2}, for $YbCu_{5-x}Ag_x$, $YbCu_{4.5}$, YbCuAl, $YbNi_2Ge_2$,

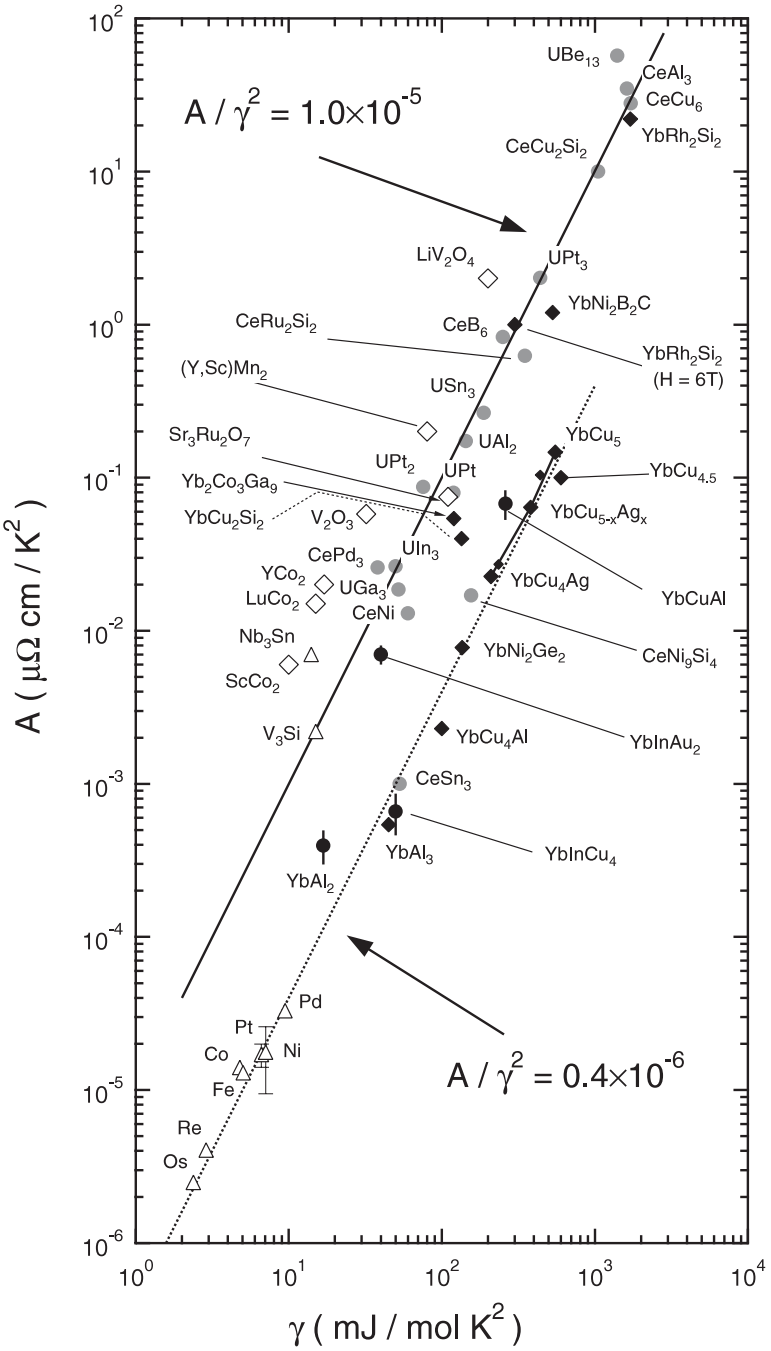

Figure 3. A plot of the T^2-coefficient of the electrical resistivity A versus the T-linear specific heat coefficient γ. Solid and dotted lines represent $A/\gamma^2 = 1.0 \times 10^{-5}$ and 0.4×10^{-6} $\mu\Omega$ cm mol^2 K^2 mJ^{-2}, respectively. Data for Yb compounds are represented by black filled symbols. Black filled circles (●) indicate the data of the present work. Grey filled symbols represent Ce- and U-based compounds. d-electron-based systems are indicated by open symbols. For references, see table 1.

Table 1. A list of the T^2-coefficients of the electrical resistivity A and the T-linear specific heat coefficient γ. The units of A, γ, and A/γ^2 are $\mu\Omega$ cm K^{-2}, mJ mol^{-1} K^{-2}, and 10^{-5} $\mu\Omega$ cm mol^2 K^2 mJ^{-2}, respectively. Here 'mol' means magnetic ion mole. The values of A given in this work are marked. The labels w, m, and s indicate that the magnetic correlation is weak, medium, and strong, respectively.

Compounds	A	γ	A/γ^2	Magnetic correlation	Ground state degeneracy
$YbCu_5$ [10]	0.15	550	0.05	w	4 [22]
$YbCu_{4.5}Ag_{0.5}$ [9]	0.064	380	0.04	w	6 [23]
$YbAgCu_4$ [9]	0.023	210	0.05	w	8 [24, 25]
$YbCu_{4.5}$ [6, 26]	0.1	600	0.03	w	
YbCuAl [6]	0.068[a]	260	0.1	w [27, 28]	8
$CeNi_9Si_4$ [29]	0.017	156	0.07	w	6
$YbNi_2Ge_2$ [13, 30]	0.0078	136	0.04		
$YbCu_4Al$ [31, 32]	0.0025	50–100	0.03–0.1		
$YbAl_3$ [6, 33]	5×10^{-4}	45	0.06	w [34]	8 [34]
$CeSn_3$ [2]			∼0.03	w	6 [35]
$YbInCu_4$ [36]	6×10^{-4} [a]	50	0.03	w [37]	8?
$YbAl_2$ [6]	4×10^{-4} [a]	17	0.14	w [34]	8 [34]
Pd, Pt [14]			0.04	s [38]	
$Yb_2Co_3Ga_9$ [39]	0.054	112	0.43	w [40]	8 [40]
$YbCu_2Si_2$ [6, 41]	∼0.04	135	∼0.2		
CeNi [42, 43]	0.013	60	0.36	m? [44]	
$YbInAu_2$ [45]	0.0070[a]	40	0.44		
$CeCu_6$ [2]			∼1.0	s [46]	2?
UPt_3 [2]			∼1.0	s [47]	
LiV_2O_4 [5]	2.0	200	5	s [48]	2
$YbRh_2Si_2$ (0 T) [49, 50]	22	1700	0.8	s	2?
$YbRh_2Si_2$ (6 T) [49, 50]	1.0	300	1.1	s [51]	2?
$YbNi_2B_2C$ [52]	1.2	530	0.43	m?	2–3
$CeRu_2Si_2$ [53]	0.62	350	0.51	s [46]	2?
$Sr_3Ru_2O_7$ [54]	0.075	110	0.6	s	3
$CePd_3$ [2]			2.5	w [55]	6 [55]
V_2O_3 [3]			4	s	3
YCo_2 [4]				s [56]	
(Y, Sc)Mn_2 [57]	0.2	80	3.1	s [58]	

[a] This work.

$YbAl_3$, $YbInCu_4$, $YbCu_4Al$, $CeNi_9Si_4$, and $CeSn_3$. The extrapolation of this line coincides well with the A/γ^2 value for transition metals such as Pd or Re [14]. As for $YbAl_2$ and $YbCu_2Si_2$, the A/γ^2 ratio appears to be situated between the two lines.

4. Discussion

We have shown in the preceding section that many Yb-based and several Ce-based compounds show values of A/γ^2 nearly two orders of magnitude smaller than that for the Kadowaki–Woods relation. In this section, we discuss possible origins of the deviation from the Kadowaki–Woods relation.

First, it must be remarked that the A-value depends not only on T_K but also on many other factors, including the anisotropy, carrier concentration, Fermi-surface topology, band structures, and site disorder. In particular, the effect of site disorder may be important as has

been pointed out by several authors, since the effect can suppress A through the Kondo-hole effect [11–13]. Here, the site disorder gives additional resistivity: $\rho^{\text{disorder}} = \rho_0 - BT^2$ [59], which results in the reduced T^2-coefficient of $A - B$. However, although this mechanism may be important in some systems, it is unlikely that the Kondo-hole effect can explain all the small A/γ^2 values, especially the almost universal value, $A/\gamma^2 = 0.4 \times 10^{-6}\,\mu\Omega\,\text{cm mol}^2\,\text{K}^2\,\text{mJ}^{-2}$. The Kondo-hole effect should result in a random distribution of the A/γ^2 values. Moreover, the additional term, $\rho^{\text{disorder}} = \rho_0 - BT^2$, has been observed only for a limited number of systems, such as $YbInAu_2$ under high pressures [11]. Thus, we believe that the Kondo-hole effect is not the crucial origin for most of the small A/γ^2 values. As for the other effects, such as those of the carrier concentration and band structures, although they must be taken into consideration, it is unlikely that they can explain the smaller 'universal' relation.

We next consider this phenomenon in relation to three mechanisms: (i) the single-body band effect, (ii) the intersite magnetic correlation, and (iii) the ground state degeneracy.

4.1. Single-body band effect

Miyake *et al* [3] have considered theoretically the effect of many-body correlations in heavy-fermion systems, where conduction electrons have a strong frequency dependence in the self-energies. They have suggested that the many-body effect can enhance the A/γ^2 value to about 25 times larger than that in the case of a single-body band, and this would explain the 25 times difference in A/γ^2 between the transition metals and the heavy-fermion systems.

A similar argument may be applicable for $YbAl_2$, for which γ is relatively small. In this compound, the many-body and the single-body effects can be comparable, which may reduce A/γ^2 to the same order as that in transition metals. On the other hand, for YbCuAl, $YbCu_4Ag$, $YbCu_5$, and $CeNi_9Si_4$, these systems exhibit much larger values of γ (100–600 mJ mol^{-1} K^{-2}) than those of the transition metals. This indicates that the DOS of the conduction band is enhanced through the many-body effect, i.e., Kondo-lattice formation.

Even in $YbAl_3$, for which the γ-value (45 mJ mol^{-1} K^{-2}) may not be so large, de Haas–van Alphen effect measurements have revealed that the Fermi surface of $YbAl_3$ is strongly renormalized due to the many-body effect of the localized 4f moment and the conduction electrons [60]. We therefore suppose that the single-body band effect is not dominant for these systems.

4.2. Intersite magnetic correlation

As is pointed out in [2], many U-based compounds on the Kadowaki–Woods line exhibit strong intersite magnetic correlation, including magnetic ordering in UPt, UGa_3, and UIn_3, and distinct spin fluctuations in UAl_2 and UPt_3. Magnetic correlation due to the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction is also considered to be important for Ce- and Yb-based compounds such as $CeAl_3$ [61, 62], $CeCu_2Si_2$ [63], CeB_6 [64], $CeCu_6$, $CeRu_2Si_2$ [46], and $YbRh_2Si_2$ [49, 50]. For d-electron systems such as LiV_2O_4 [48], YCo_2 [56], and $(Y, Sc)Mn_2$ [58], the presence of strong magnetic correlation has been demonstrated by means of neutron scattering or NMR. It is notable that these compounds include both nearly ferromagnetic (UAl_2 [65], YCo_2 [56], etc) and nearly antiferromagnetic systems ($CeCu_6$ [46], $CeCu_2Si_2$ [63], etc).

On the other hand, for the systems with the smaller A/γ^2 values, magnetic correlation does not appear to be important. Physical properties are well explained using a single-impurity model for $YbCu_5$ [10, 66], $YbCu_{5-x}Ag_x$ [9, 22, 23], $YbCu_4Ag$ [24, 25], YbCuAl [27], $CeNi_9Si_4$ [29], and $CeSn_3$ [35]. Single-impurity character is also demonstrated by neutron

scattering experiments for YbCuAl [28] and $YbInCu_4$ [37]. Therefore for these compounds, intersite magnetic correlation can be neglected. For $YbCu_{4.5}$, magnetic ordering is not observed even at 235 kbar down to 50 mK [26], suggesting weak RKKY interaction.

This remarkable contrast in strengths of magnetic interactions implies that there exist qualitatively different scattering mechanisms in the electrical resistivity. For the case with strong magnetic interactions, A and γ are enhanced both by the large density of state (DOS) at the Fermi energy (E_F) and by the strong spin fluctuations. In contrast, for the single-impurity-like case, the large A is solely attributable to the DOS at E_F. One may hence consider this difference to be a possible origin for the two 'universal' A/γ^2 relations.

However, this tendency is inconsistent with the theoretical calculations based on the spin-fluctuation theory [67, 68]. Takimoto *et al* [67] have shown that the value of A/γ^2 is almost constant from the pure Kondo regime (single-impurity limit) almost to the magnetic instability limit. The value of A/γ^2 that they estimated is close to the Kadowaki–Woods relation. A similar result is also reported by Continentino, who has shown that the A/γ^2 value is independent of the distance from the magnetic instability [68].

It should also be noted that there exist some exceptions from the above tendency. For $CePd_3$, intersite magnetic correlation is considered to be of minor importance [55]. For $YbInAu_2$ and Nb_3Sn, no evidence of strong magnetic correlation is reported. However, the A/γ^2 values for these systems are close to the Kadowaki–Woods relation. On the other hand, Pd and Pt show small A/γ^2 values, although they are well-known examples of nearly ferromagnetic metals [38]. Therefore, these findings may not justify the above scenario, in which strong intersite magnetic correlation enhances A/γ^2 values.

4.3. Ground state degeneracy

One may note that most of the systems with the smaller A/γ^2 values in figure 3 are classified as intermediate-valence compounds, where the crystal-field splitting, Δ_{CF}, is of minor importance, and the full degeneracy N for the Yb^{3+} or Ce^{3+} ion is almost preserved [69]. In fact, the physical properties of $YbCu_4Ag$, YbCuAl, $CeSn_3$, and $CeNi_9Si_4$ are explained by the impurity model for $N = 8$ (Yb^{3+}) and $N = 6$ (Ce^{3+}), as is noted in the preceding subsection. For $YbAl_3$ and $YbAl_2$, Shimizu *et al* [34] have suggested on the basis of ^{171}Yb NMR that the $N = 8$ ground state is well conserved in these compounds. Even in $YbCu_5$, which exhibits a large $\gamma = 550\,\mathrm{mJ\ mol^{-1}\ K^{-2}}$, Δ_{CF} is considered to be comparable with the Kondo temperature T_K, and the low-temperature properties are characterized by $N = 4$ [22, 23].

In contrast, systems such as $CeCu_6$, $CeAl_3$, and $CeCu_2Si_2$ are considered to be affected by crystal-field effects, which reduce the ground state degeneracy to $N = 2$ [69].

The difference in N is related to the magnitude of the magnetic moment which participates in the Fermi-liquid formation. It can therefore affect the A/γ^2 values. We note that the difference between $N \geqslant 4$ and $N \leqslant 3$ states can be crucial. Theoretical calculation predicts that the DOS has a peak situated above E_F for the case of $N \geqslant 4$, whereas the peak is positioned just at E_F for $N \leqslant 3$ [70]. Similarly, magnetic excitation spectra also show inelastic structure for large N [71, 72]. Hence, we suppose that in those Yb- and Ce-based intermediate-valence systems, their substantially degenerate states ($N \geqslant 4$) lead to the development of excitation spectra with qualitatively different shapes from those for the heavy-fermion systems with small N ($\leqslant 3$), resulting in the significant difference in the A/γ^2 values. There have been no theories regarding the N-dependence of the A/γ^2 value as far as we know. Theoretical investigation of the relation between N and A/γ^2 is desirable[4].

[4] Lawrence has tentatively suggested that A/γ^2 can be a decreasing function of N [75], if we assume that $A = \rho^{\mathrm{unit}}/T_K^2$, where ρ^{unit} is the unitarity limit described as $\rho^{\mathrm{unit}} \propto N \sin^2(\pi/N)$, and that $\gamma = (N-1)\pi k_B/6T_K$ [70].

However, some exceptions not covered by this understanding should be noted. For $Yb_2Co_3Ga_9$ [40], the full degeneracy for Yb^{3+} is considered to be preserved, though it exhibits the same order of A/γ^2 as that for the Kadowaki–Woods relation. In addition, it is unclear whether all the U-based compounds on the Kadowaki–Woods line can be understood in terms of $N \leqslant 3$ ground states. In these U compounds, the enhanced low-energy excitations due to intersite magnetic correlation may also contribute.

We also note that $CePd_3$ is again exceptional. This compound is a typical intermediate-valence compound. The full degeneracy $N = 6$ is considered to be preserved, and the intersite magnetic correlation is also of minor importance [55]. Nevertheless, this compound shows the same order of A/γ^2 value as heavy-fermion compounds. However, it should be remarked that the carrier concentration of $CePd_3$ is unusually small ($\sim$0.3 per unit cell) [73], and hence this compound would be close to the Kondo insulators. This situation prevents us from making a naive comparison for $CePd_3$.

Finally, an important suggestion has arisen experimentally, from electrical resistivity measurements under pressures. Knebel *et al* [13] have examined $AT_{\max}^2$ for $YbNi_2Ge_2$ under pressure, where $T_{\max}$ is the maximum temperature of the resistivity adhering to a T^2-power law, and should be proportional to γ^{-1}. The value of $AT_{\max}^2$ is therefore a measure of A/γ^2. They have found that $AT_{\max}^2$ increases by a factor of about 25 times under high pressures. Similar variation of $AT_{\max}^2$ under pressure has also been reported for $CeCu_2Si_2$ [74]. These phenomena are most probably related to either the change of the intersite magnetic correlation or that of the ground state degeneracy. Hence, performing similar experiments on much more systems may eventually reveal the underlying origin of the two 'universal' A/γ^2 relations.

In addition, the effect of disorder must be carefully taken into consideration. If the disorder had little effect on the host-material properties, its effect on the resistivity would be describable in terms of the Kondo-hole effect, which reduces A. In contrast, in most systems, disorder is considered to affect the whole range of electrical properties in the Kondo-lattice formation. This would reduce the characteristic temperature T^*, which would result in enhanced A-values. This may be the case in $YbInAu_2$, where site disorder readily occurs because of its crystal structure. Therefore, the A/γ^2 values in several systems may not be intrinsic. Experimental works using high-quality samples are particularly important.

5. Summary

We have measured the electrical resistivity and have examined the A/γ^2 values of $YbAl_2$, $YbInAu_2$, $YbInCu_4$, and YbCuAl. We have also plotted A versus γ for several systems reported on in the literature. The results reveal that the A/γ^2 values are not constant across the range of materials, but vary in the range of 0.4×10^{-6}–1.0×10^{-5} $\mu\Omega$ cm mol^2 K^2 mJ^{-2}. In particular, it has been found that there exists another 'universal' relation; $A/\gamma^2 = 0.4 \times 10^{-6}$ $\mu\Omega$ cm mol^2 K^2 mJ^{-2}, for a number of systems including YbCuAl, $YbAl_3$, $YbInCu_4$, $YbCu_4Al$ as well as $YbCu_5$, $YbCu_{4.5}$, $YbCu_4Ag$, $YbNi_2Ge_2$, $CeNi_9Si_4$, and $CeSn_3$. This value of A/γ^2 is about 25 times smaller than that known from the Kadowaki–Woods relation: $A/\gamma^2 = 1.0 \times 10^{-5}$ $\mu\Omega$ cm mol^2 K^2 mJ^{-2}.

To explain this deviation, several possible mechanisms have been discussed. The Kondo-hole effect or the single-body band effect may be occurring in some systems. However, these two mechanisms cannot explain the almost 'universal' nature of the smaller A/γ^2 values. Thus, we have considered the tendencies of these compounds empirically, and have focused on the intersite magnetic correlation and the ground state degeneracy.

As regards the former, we have pointed out that most of the systems with the smaller A/γ^2 value exhibit single-impurity-like behaviour. This suggests that the intersite magnetic interactions are negligible in these systems. In contrast, many systems on the Kadowaki–Woods line are known to show strong magnetic correlation. This tendency is valid for many compounds plotted in figure 3. However, this interpretation is inconsistent with the theoretical results based on the spin-fluctuation theory.

As regards the latter, it has been found that the ground state degeneracy N differs substantially between the systems with the smaller A/γ^2 values and those on the Kadowaki–Woods line. Almost all the systems with the smaller A/γ^2 values (except for d-electron systems) show a substantially degenerate ground state; i.e., $N \geqslant 4$. On the other hand, those on the Kadowaki–Woods line are considered to have the $N \leqslant 3$ state due to the crystal-field splitting. Since this difference can affect the magnitude of the magnetic moment as well as the shape of the excitation spectra, the A/γ^2 values may also vary depending on N.

For both the cases, i.e., the intersite magnetic correlation and the ground state degeneracy, our interpretations are only empirical. Hence, theoretical confirmations are strongly desired.

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