Universality in Heavy Fermion Systems with General Degeneracy

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We discuss the relation between the T^2 coefficient of electrical resistivity A and the T-linear specific-heat coefficient γ for heavy-fermion systems with general N, where N is the degeneracy of quasiparticles. A set of experimental data reveals that the Kadowaki-Woods relation, $A/\gamma^2 = 1 \times 10^{-5}\ \mu\Omega\,\mathrm{cm(K\,mol/mJ)^2}$, collapses remarkably for large-N systems, although this relation has been regarded to be commonly applicable to the Fermi liquids. Instead, based on the Fermi-liquid theory we propose a new relation, $\tilde{A}/\tilde{\gamma}^2 = 1 \times 10^{-5}$ with $\tilde{A} = A/\frac{1}{2}N(N-1)$ and $\tilde{\gamma} = \gamma/\frac{1}{2}N(N-1)$. This new relation exhibits an excellent agreement with the data for the whole range of degenerate heavy fermions.

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The Fermi-liquid theory [1] is the most fundamental one to understand the electronic state of metallic systems. This theory has achieved great success in describing not only the electronic properties of normal metals but also unusual properties of the strongly correlated electron systems [2,3] such as f-electron based heavy-fermion compounds [2–5] and d-electron based intermetallics and oxides [2,6]. In this theory, the effect of electron-electron interactions is involved in the effective mass of quasiparticles, m^*. This enables a very simple representation of physical properties: the electronic specific heat C and the electrical resistivity ρ are described as $C = \gamma T$ and $\rho = AT^2$ with $\gamma \propto m^*$ and $A \propto m^{*2}$. Such a temperature dependence has actually been observed in numerous kinds of metals. Moreover, this description implies that the ratio A/γ^2 does not depend on m^*, resulting in the universal value of A/γ^2. In fact, it has been revealed that many f-electron based systems show the universal behavior, $A/\gamma^2 = 1.0 \times 10^{-5}\ \mu\Omega\,\mathrm{cm(K\,mol/mJ)^2}$ [7], called the Kadowaki-Woods (KW) relation. The KW relation has therefore been accepted as the most essential relation showing the validity of the Fermi-liquid theory.

Recently, however, significant and systematic deviations from the relation have been observed in many heavy-fermion compounds, despite that they apparently show Fermi-liquid behavior at low temperature [8]. This class of compounds includes Yb-based compounds such as $YbCu_5$, $YbAgCu_4$, YbCuAl, $YbNi_2Ge_2$, $YbInCu_4$, and $YbAl_3$, and Ce-based compounds such as $CeNi_9Si_4$ [9] and $CeSn_3$. Notably, the deviations in these systems are almost "universal," $A/\gamma^2 \approx 0.4 \times 10^{-6}\ \mu\Omega\,\mathrm{cm(K\,mol/mJ)^2}$. This systematic and large deviation cannot be explained by specific characters of materials, such as carrier density, band structure, anisotropy, etc. Instead, there seems to exist a common physical origin. The origin of this deviation therefore raises an important issue for the generality of the Fermi-liquid theory.

Very recently, a theoretical work based on the Fermi-liquid theory has suggested [10] that the values of A/γ^2, so far considered to be unique and independent of materials, do depend on the number of degeneracy of quasiparticles N. For isolated atoms, N is defined as $N = 2J + 1$, with J the total angular momentum. In solids, N can vary due to the competition between the crystal-field splitting Δ and the Kondo temperature T_K. For $T_K < \Delta$, the low-temperature properties are basically explained by the $N = 2$ ($S = 1/2$) Kondo model, since most of the degeneracy is lost due to the large Δ [5,11,12]. For $T_K > \Delta$, on the contrary, the crystal-field splittings are covered by the large Kondo effect, and the degeneracy is almost preserved down to low temperatures. In this case, the theory [10] gives the failed universality of A/γ^2.

In this Letter, we make a quantitative comparison of the experimental data in Ref. [8] and several recent works with the theoretical results of Ref. [10]. The results display a beautiful agreement between experiments and theory. Furthermore, we propose an advanced relation for A and γ based on these results. Using $\tilde{A}$ and $\tilde{\gamma}$, the values of A and γ normalized by $\frac{1}{2}N(N-1)$, we show that these two values of heavy-fermion systems with general N are related by a very simple formula, $\tilde{A}/\tilde{\gamma}^2 = 1 \times 10^{-5}\ \mu\Omega\,\mathrm{cm(K\,mol/mJ)^2}$. This new relation, namely, the "grand-KW relation," will be an important waymark for the research of strongly correlated electron systems with degeneracies, and remarkably extends the validity of the Fermi-liquid theory.

At first, we briefly describe the theoretical results of Ref. [10]. For the case of the strong-coupling limit where $m^*/m \gg 1$ (m^* and m being the mass of heavy quasiparticles and free electrons, respectively), the orbitally degenerate periodic-Anderson (ODPA) model gives [13]

$$A = \frac{hk_B^2}{e^2}\frac{3\pi^6}{2k_F^4 a^3}N(N-1)\Gamma_{\mathrm{loc}}^2(0,0)\rho_f^4(0), \qquad (1)$$

$$\gamma = N_A k_B^2 \frac{\pi^2}{6} N(N-1)\Gamma_{loc}(0,0)\rho_f^2(0). \quad (2)$$

where h is the Plank constant, k_B is the Boltzmann constant, k_F is the Fermi momentum, and N_A is the Avogadro number. In addition, a is the unit-cell length, and $\rho_f(0)$ is the density of states per f orbit at the Fermi energy. $\Gamma_{loc}(0,0)$ represents the effective interaction between quasiparticles. Note that A and γ given in Eqs. (1) and (2) are not simply proportional to $N(N-1)$, because $\Gamma_{loc}(0,0)$ also depends on N. The value A/γ^2 is then deduced as [14]

$$\frac{A}{\gamma^2} = \frac{h}{e^2 k_B^2 N_A^2} \frac{9(3\pi^2)^{-1/3}}{n^{4/3}a^3} \frac{1}{\frac{1}{2}N(N-1)} \approx \frac{1\times10^{-5}}{\frac{1}{2}N(N-1)}\ \mu\Omega\,\mathrm{cm(K\,mol/mJ)^2}. \quad (3)$$

For the case of $N = 2$, this formula gives the KW relation. For general N, this gives a set of universal relations. This is shown in Fig. 1 as the solid lines for $N = 2$, 4, 6, and 8.

In Fig. 1, experimental data are also plotted after Refs. [7–9,15–18]. At first, one can see that many heavy-fermion systems such as $CeCu_6$ or $CeCu_2Si_2$ agree with the KW relation, i.e., the theoretical prediction for $N = 2$. This is consistent with the situation $T_K < \Delta$, which results in the low degeneracy of $N = 2$ [19]. Moreover, it is clear that many Yb- and Ce-based systems, which have deviated from the KW relation, agree well with the theoretical predictions for $N = 6$–8.

It should also be noted that the A/γ^2 of Eu- and Sm-based compounds agrees very well with the line for $N = 8$ and $N = 4$, respectively. These Eu compounds are considered to be intermediate valent between $Eu^{2+}(S = 7/2)$ and $Eu^{3+}(J = 0)$ [16]. The Fermi-liquid state of them is hence considered to be emerged out of the degeneracy $N = 2S + 1 = 8$. For the two Sm-based systems, the value of $N = 4$ has been assumed, since the lowest crystal-field level is considered to be a quartet [17,18]. These quantitative agreements of A/γ^2 with respective theoretical lines are evidence that the A/γ^2 of heavy-fermion systems are not specific to materials, but are commonly scaled by degeneracy.

In the following, we go forward to unify these relations into a single relation. If the value of N is determined experimentally, we can define the normalized coefficients $\tilde{A}$ and $\tilde{\gamma}$ from Eqs. (1) and (2) as

$$\tilde{A} = \frac{A}{\frac{1}{2}N(N-1)}, \qquad \tilde{\gamma} = \frac{\gamma}{\frac{1}{2}N(N-1)}.$$

Then $\tilde{A}/\tilde{\gamma}^2$ is obtained from Eq. (3) as

$$\tilde{A}/\tilde{\gamma}^2 \approx 1\times10^{-5}\ \mu\Omega\,\mathrm{cm(K\,mol/mJ)^2}. \quad (4)$$

This formula does not include any N dependence. Hence, this should be applicable to arbitrary N systems.

In Fig. 2, we plot $\tilde{A}$ and $\tilde{\gamma}$ of f-electron based systems. For uranium compounds, we have tentatively assumed

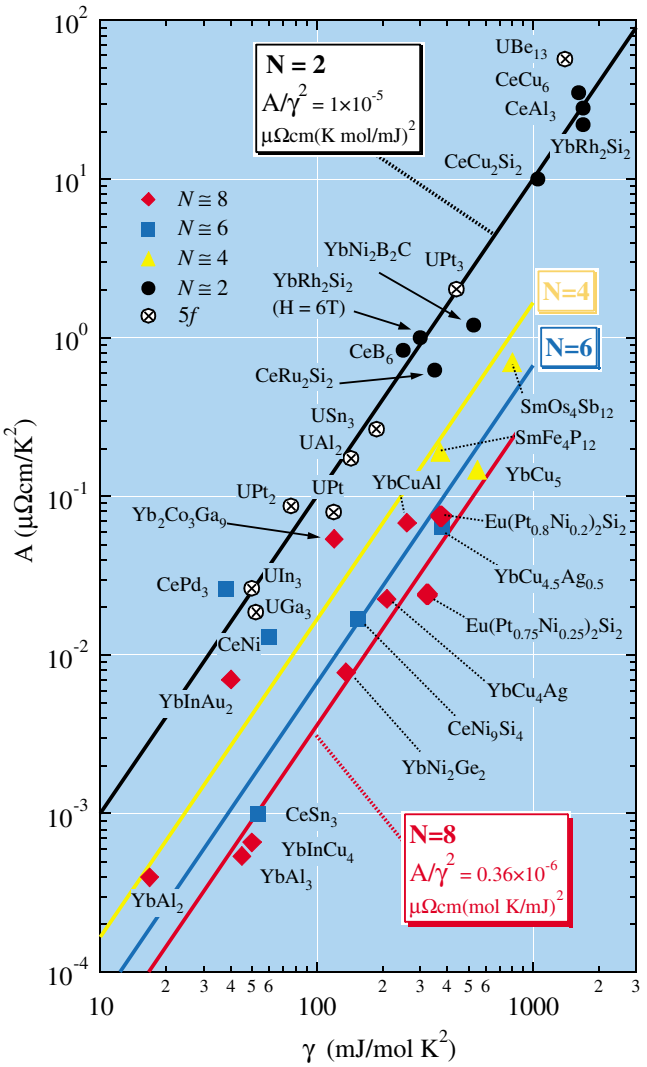

FIG. 1 (color). T^2 coefficient of electrical resistivity A vs T-linear coefficient of specific heat γ of heavy-fermion systems with various degeneracy. Experimental data are taken from Refs. [7–9,16–18]. The black line corresponds to the Kadowaki-Woods relation [7]. Other solid lines are the prediction from the orbitally degenerate periodic-Anderson model [10]. Colors of the symbols represent the degeneracy N probable for the systems: black, yellow, blue, and red indicate $N = 2$, 4, 6, and 8, respectively. The value of N of U compounds is not determined.

$N = 2$, which is discussed later. One can see that Eq. (4) scales $\tilde{A}$ and $\tilde{\gamma}$ universally for a wide range of materials. This fact shows the validity of the theoretical approach using the ODPA model and extends the validity of the Fermi-liquid theory. We stress that our new relation has the same form just as the original KW relation. The formula (4) may hence be called the grand-KW relation for general degeneracy.

Here it would be interesting to discuss to what extent this rule holds when the system is reached toward the quantum critical point (QCP). Even in the vicinity of the QCP, the Fermi-liquid state is realized at sufficiently low temperatures below a characteristic temperature (T_{coh} in the litera-

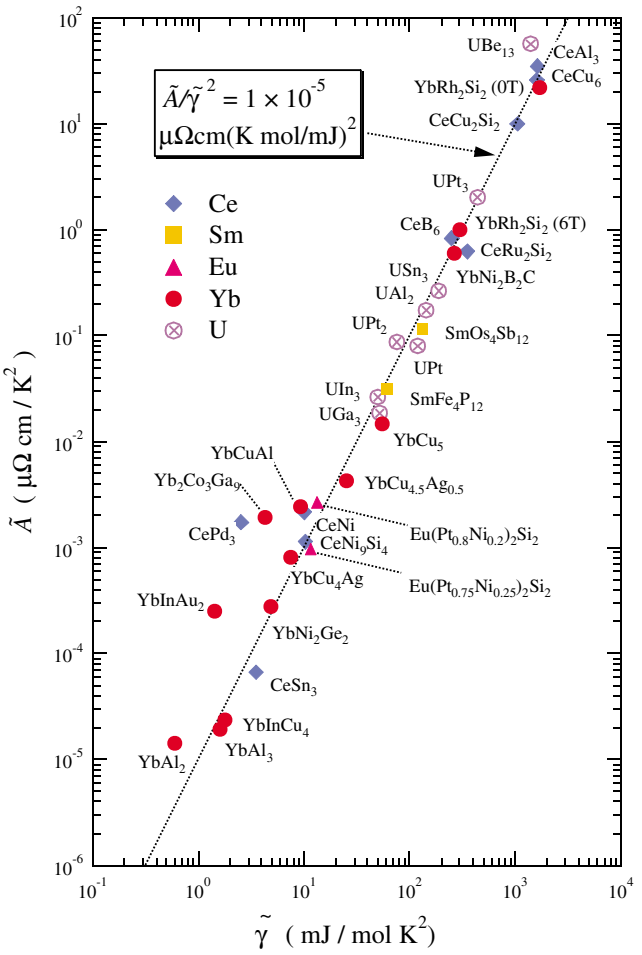

FIG. 2 (color). The plot of $\tilde{A}$ and $\tilde{\gamma}$ of heavy-fermion systems. $\tilde{A}$ and $\tilde{\gamma}$ are the divided values of A and γ by $\frac{1}{2}N(N-1)$, respectively. N of the U-based compounds is tentatively assumed to be 2. The dotted line represents the grand-KW relation (4) given in the text: $\tilde{A}/\tilde{\gamma}^2 = 1 \times 10^{-5}\ \mu\Omega\,\mathrm{cm(K\,mol/mJ)^2}$.

ture) as far as the system is in the magnetically disordered side. In this case, our theory yields that the value of $\tilde{A}/\tilde{\gamma}^2$ defined at the low-temperature limit ($T \to 0$) follows the relation (4) even in the vicinity of the QCP. This is because our theory is derived for the limit of $T \to 0$. It is also noticeable that the theoretical calculations of A/γ^2 for $N = 2$ based on the spin-fluctuation theory show that the ratio is approximately independent of the distance from the QCP [20,21]. In fact, the A/γ^2 of $YbRh_2Si_2$ and $CeInCo_5$, both of which are considered to be in the vicinity of QCP, are almost a constant as the external magnetic field is varied [22,23]. Meanwhile, for the case of "very" close to the QCP, where T_{coh} is quite low, a deviation from the universality may be observed, as is suggested theoretically [20] and experimentally on $YbRh_2(Si, Ge)_2$ [24]. This deviation, however, seems to occur in an extremely narrow condition where the Fermi-liquid description is probably not valid. Except for such extreme cases, the grand-KW relation is one of the common behavior of Fermi liquids, even in systems close to the QCP. Note that the f-orbital degeneracy will stabilize the Fermi-liquid state, because the N dependence of T_{K} ($\propto e^{-1/\rho N J_{\mathrm{K}}}$) will be much more prominent than that of T_{N} ($\propto N^2 J_{\mathrm{RKKY}}$). A large mass enhancement is realized with relatively higher T_{K} when $N > 2$.

There should, of course, exist exceptions. As is seen in the formula (3), the ratio A/γ^2 as well as that of $\tilde{A}/\tilde{\gamma}^2$ depends on the carrier concentration n, the wave number at the Fermi energy k_{F}, and so on. If one of these values is extremely different from the typical ones, $\tilde{A}/\tilde{\gamma}^2$ can deviate remarkably. Such an example is $CePd_3$. In Fig. 2, one can see that $CePd_3$ shows a large deviation from Eq. (4), though $CePd_3$ agrees well with the original KW relation [7]. This discrepancy results from the large degeneracy, $N = 6$ for $CePd_3$ [25]. It should be noted that $CePd_3$ has very small carrier concentration (0.3 electrons per f.u.) [26]. The A/γ^2 value is found to depend on n as proportional to $n^{-4/3}$ from Eq. (3) and also from other theoretical studies [2,20,21,27]. Taking this into consideration, the deviation of $CePd_3$ from the universal line is reasonable. A similar deviation is reported for the Kondo semiconductor CeNiSn [28]. An anomalously large A value ($54\ \mu\Omega\,\mathrm{cm/K^2}$) compared to its γ ($40\ \mathrm{mJ/mol\,K^2}$) has been attributed to its extremely low carrier concentration [28].

The compounds CeNi ($N = 6$) and YbCuAl ($N = 8$) also show slight deviations, possibly due to the error in the N estimations. For other exceptions, $YbInAu_2$ and $Yb_2Co_3Ga_9$, we have no explanation for the origin of deviation. Other causes, such as the multi-Fermi-surface effect [9], may have to be considered. In addition, strong anisotropy of the Fermi surface can cause deviation from the universal relation [2,29]. This effect would be, in general, more prominent in d-electron systems [30,31].

For U-based compounds, its degeneracy has been the subject of arguments. If the $5f$ electrons are well localized, N can be determined experimentally, as in the case of UPd_3 [32]. In most U compounds, however, it is considered that the $5f$ electrons have a more itinerant character than $4f$, since $5f$ orbitals are spatially more expanded. The definition of N in U compounds is therefore ambiguous. Here, one can see in Figs. 1 and 2 that those U compounds agree well with the theoretical prediction for $N = 2$. This can lead us to the possibility that the orbital degree of freedom is quenched and only the spin degree of freedom participates in the Fermi-liquid state in these $5f$ systems, similar to transition metals. Although the estimation of N from the A/γ^2 plot is not conclusive, this plot may serve as a hint to discuss the puzzling $5f$ electrons.

In addition, we note that the grand-KW relation is also powerful enough to describe the pressure dependent properties of heavy-fermion systems. In $CeCu_2Ge_2$ (or $YbNi_2Ge_2$), it is suggested that the value of A/γ^2 reduces (or increases) about 25 times at high pressures [33,34] probably due to the change of N by pressures. In our plot of $\tilde{A}$ and $\tilde{\gamma}$, these crossovers would be described on the

single scaling without breaking the universality. This situation may be hence ideal for the continuity principle of the Landau–Fermi-liquid theory [35]. Current interests in strongly correlated electron systems are extended to the orbitally degenerate cases. Hence, the grand-KW relation will be one of the most fundamental relations in Fermi-liquid systems. We also comment that the effect of the degeneracy must be taken into consideration in many other physical quanta, such as the anomalous Hall effect [36], etc. The analysis using the ODPA model will henceforth be indispensable.

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[15] For the values of N, we have employed the data listed in Ref. [8], most of which were determined by fitting the specific heat using the impurity-Kondo models. Although the real system is periodic, we believe that this estimation of N is reliable, because these fittings are performed for the data at relatively high temperatures, at which intersite effects are not prominent. However, it is difficult to determine the value of N explicitly when Δ (crystal-field splitting) and T_K are comparable.
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