Generalized Kadowaki-Woods Relation
in Heavy Fermion Systems with Orbital Degeneracy

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(November 4, 2018)

We present a theoretical study of the Kadowaki-Woods relation in the orbitally degenerate periodic Anderson model. Based on Fermi liquid theory, we derive the generalized Kadowaki-Woods relation in the strong coupling limit, \( A^\gamma \sim 10^{-5} \frac{A^2}{N(N-1)} \left[ \mu\text{eVcm(mol-K/mJ)}^2 \right] \), where \( A \) is the coefficient of the \( T^2 \) term in the resistivity, \( \gamma \) is the \( T \)-linear specific heat coefficient, and \( N \) is the \( f \)-orbital degeneracy. This result naturally explains the remarkably smaller value of \( A^\gamma \) in various orbitally degenerate (mainly Yb-based) heavy Fermion systems, reported by Tsujii et al., J. Phys. Cond. Mat. 15 (2003) 1993.

Key Words: Kadowaki-Woods relation, orbitally degenerate periodic Anderson model, Fermi liquid theory

In Fermi liquid (FL) systems, the specific heat \( C \) and the resistivity \( \rho \) behave as \( C = \gamma T \) and \( \rho = \rho_0 + AT^2 \) at sufficiently low temperatures. Because \( \gamma \propto m^* \) and \( A \propto (m^*)^2 \) (\( m^* \) being the effective mass of quasiparticles) according to the FL theory, the ratio \( A^\gamma \) is expected to be independent of \( m^* \). In fact, many Ce- and U-based heavy Fermion (HF) systems follow a universal relation, \( A^\gamma \sim 1 \times 10^{-5} \left[ \mu\text{eVcm(mol-K/mJ)}^2 \right] \), which is called the Kadowaki-Woods (KW) relation [1]. Moreover, the KW relation holds experimentally even in the close vicinity of a magnetic quantum critical point under a magnetic field [5], the fact of which is consistent with theoretical analyses [6-8].

Recently, however, various Fermi liquid systems which does not follow the KW relation have been found experimentally. Especially, Tsujii et al. have revealed that \( A^\gamma \sim 0.4 \times 10^{-6} \left[ \mu\text{eVcm(mol-K/mJ)}^2 \right] \) in many Yb-based HF systems like YbCu\(_{4}\)Ag (\( \gamma = 240 \text{mJ/mole} \cdot \text{K}^2 \)), YbCu\(_{5-x}\)Ag\(_x\) (210 \( \sim \) 460 mJ/mole \( \cdot \) K\(^2\)), and others [2,9,10]. It is about 20~30 times smaller than the conventional KW ratio, although they are expected to be Fermi liquids. Thus, the violation of the KW relation should be a very important and fundamental subject on the FL theory. The authors of ref. [2] suggest that materials with smaller \( A^\gamma \) have almost fully degenerate ground states. In fact, the crystalline electric field (CEF) in Yb-based HF systems is in general smaller than that in Ce-based HF ones because Yb\(^{3+}\)-ion is smaller than Ce\(^{3+}\)-ion, known as lanthanoid contraction. Note that a smaller value of \( A^\gamma \) is also observed in Pd, Pt, Ni and Fe where \( \gamma \gg \gamma_{\text{band}} \) [11].

In the present work, we revisit the KW relation in HF systems based on the FL theory, by taking the \( f \)-orbital degeneracy into account. By applying the diagrammatic method developed in analyzing the impurity Anderson model [12-14], we succeed in deriving a generalized KW relation, eq. (12), which is valid in the strong coupling case where \( m^* \gg m_{\text{band}} \). By putting \( N = 8 \) (\( N \) being the orbital degeneracy) which corresponds to a free Yb\(^{3+}\)-ion, we obtain that \( A^\gamma \sim 0.36 \times 10^{-6} \left[ \mu\text{eVcm(mol-K/mJ)}^2 \right] \), which is consistent with experimental observations. This is the first theoretical derivation of the KW relation in orbitally degenerate systems.

As discussed in ref. [2], the violation of the KW relation in Yb-compounds cannot be ascribed to the impurity effect nor an accidental singularity of the band structure inherent to individual compounds. For this reason, we attack this universal issue based on a conventional orbitally degenerate periodic Anderson model (OD-PAM) with a single conduction band. Because of the strong \( L-S \) coupling, the \( f \)-electron state for Yb\(^{3+}\)-ion with \( 4f^{13} \) \((\text{Ce}^{3+}\)-ion with \( 4f^4 \)) is specified by the total angular momentum \( J = 7/2 \) \((J = 5/2)\) and its \( z \)-component \( M \). The degeneracy of \( f \)-orbital is \( N = 2J + 1 \).

Here, we study the following OD-PAM [15-17]:

\[
H = \sum_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \sum_{\mathbf{k}M} E_{f_{\mathbf{k}M}} f^\dagger_{\mathbf{k}M} f_{\mathbf{k}M} + \sum_{\mathbf{k}M\sigma} \left( V^*_{\mathbf{k}M\sigma} f^\dagger_{\mathbf{k}M} c_{\mathbf{k}\sigma} + \text{h.c.} \right) + \frac{U}{2} \sum_{iM \neq M'} n_{iM} f^\dagger_{iM} f_{iM'},
\]

where \( c^\dagger_{\mathbf{k}\sigma} \) \((f^\dagger_{\mathbf{k}M})\) is the creation operator of a \( c \)-electron \((f \)-electron) with spin \( \sigma = \pm \frac{1}{2} \) \((f \)-electron with \( M = J, J-1, \cdots, -J)\), \( n_{iM} = f^\dagger_{iM} f_{iM} \), \( \epsilon_k \) is the dispersion of the \( c \)-electrons, and \( E_f \) is the localized \( f \)-level energy. Here we assume \( E_f > \mu \) \((\mu \) being the chemical potential) commonly for Ce and Yb-based HF systems, by considering the particle-hole
transformation for \( \text{Yb}^{3+} \)-ion. The \( c \)-\( f \) mixing potential matrices are given by \( V_{kM}^{c} = V_{0} \cdot \delta_{M,\sigma} \) for non-orbital-degenerate case \((J = 1/2, N = 2)\), and it is given by \( V_{kM}^{f} = V_{0} \sqrt{4\pi /3} \sqrt{(J-1)^{2} / 2} \sigma_{f} \left( \theta_{k}, \varphi_{k} \right) \) for \( \text{Yb}^{3+} \) \((J = 7/2, N = 8)\), and \( V_{kM}^{f} = 2\pi V_{0} \sqrt{4\pi /3} \sqrt{(J-1)^{2} / 2} \sigma_{f} \left( \theta_{k}, \varphi_{k} \right) \) for \( \text{Ce}^{3+} \) \((J = 5/2, N = 6)\), where \( \gamma_{f}^{\mathrm{H}}(\theta_{k}, \varphi_{k}) \) is the spherical harmonics. We note that the relation \( \sum_{M=-J}^{J} |V_{kM}^{M}|^{2} = V_{0}^{2} \) is satisfied in each case.

The \( f \)-Green function of the present model, \( G_{kM,M'}^{f}(\epsilon) \), is given in ref. \[16\]. It is shown that \( G_{kM,M'}^{f}(\epsilon) \propto e^{i(M-M')\phi_{k}} \), so it vanishes except for \( M' = M \) after the summation taken over \( k \). Here we study the three-dimensional OD-PAM within the framework of the dynamical-mean-field-approximation (DMFA) where the \( d = \infty \) limit is taken systematically \[18,19\]. In the DMFA, the self-energy \( \Sigma(\omega) \) is constructed of local \( f \)-Green function(s), \( g(\omega) \equiv \frac{1}{N_{0}} \sum_{k,M} G_{kM,M}(\omega) \), which is given as \[16,17\]

\[
g(\omega) = \frac{2}{N} G^{f}(\omega) \left( \frac{1 - 2}{N} \right) G^{0}(\omega),
\]

\[
G^{f}(\omega) = \frac{1}{N_{0} \sum_{k} } \left( \frac{1}{G^{0}(\omega) - V_{0}^{2} / (\omega + \mu - \epsilon_{k})} \right)^{-1},
\]

\[
G^{0}(\omega) = (\omega + \mu - E_{f} - \Sigma(\omega))^{-1},
\]

where \( G^{0} \) represents the \( f \)-Green function without mixing with \( c \)-electrons. \( N_{0} \) is the number of sites. Note that \( g(\omega) \) is diagonal with respect to \( M \) and independent of \( M \). Below, we will utilize this fact to analyze the strong coupling case.

First, we discuss the charge susceptibility \( \chi_{c} \) and the specific heat coefficient \( \gamma \), which are given as

\[
\chi_{c} = \frac{1}{z_{\mu}} N \rho^{f}(0),
\]

\[
\gamma = \frac{\pi^{2}}{6} \frac{1}{z} N \rho^{f}(0),
\]

at zero temperature, where \( 1/z_{\mu} = 1 - \partial \Sigma(\epsilon)/\partial \epsilon |_{\epsilon=0}, \) \( 1/z_{c} = 1 - \partial \Sigma(\omega)/\partial \mu, \) and \( \rho^{f}(0) \equiv \text{Im}(\epsilon_{f}^{\phi} / \pi) \) is the \( f \)-electron density of states (DOS) per channel at the Fermi level; the total \( f \)-electron DOS is \( N \rho^{f}(0) \).

By shifting the frequencies of every closed loop in \( \Sigma(\omega) \) by \( \omega \), we obtain the identity \[3, 14\]

\[
\frac{1}{z} = \frac{1}{z_{\mu}} + \sum_{j,M'} \Gamma_{i,M,j,M'}(0,0)\rho^{f}(0),
\]

where \( i,j \) are site indices, and \( \Gamma_{i,M,j,M'}(\epsilon, \epsilon') \) is the full four-point vertex within the DMFA; see Fig. 1(a). In the strong coupling limit where \( U \) is sufficiently large, \( \gamma \) will be strongly enhanced whereas \( \chi_{c} \) is suppressed. This means that \( 1/z \approx 1/z_{\mu}, \) so \( 1/z \mu \) in eq.(7) can be dropped in the strong coupling case.

Here, we introduce a modified renormalization factor \( 1/z_{\mathrm{loc}} \) by dropping \( \Gamma_{i,M,j,M'} \) except for \( j = i \) in eq.(7), which we call the local approximation. We note that by adding the term \( U \sum_{i,M} n_{iM}^{f} n_{iM}^{f} = U \sum_{i,M} n_{iM}^{f} \) to eq.(1) virtually, we can neglect the Pauli principle in the Coulomb interaction in constructing diagrams \[14,16\]. This added term can be absorbed by shifting \( E_{f} \) in eq.(1) because it is a \( M \)-independent constant in the paramagnetic uniform state. Then, the identity \( \Gamma_{i,M,j,M'}(0,0) = (1 - \delta_{MM'})\Gamma_{\mathrm{loc}}(0,0) \) is easily recognized within the DMFA as shown in Fig. 1(b), where \( \Gamma_{\mathrm{loc}}(0,0) \) is the asymmetric local vertex composed of local Green functions and \( U \)'s. We can also check this identity order by order with respect to \( U \) \[14\]. Taking notice of the factor arises from the summation over \( M' \), we find that \( 1/z_{\mathrm{loc}} = (N - 1)\Gamma_{\mathrm{loc}}(0,0)\rho^{f}(0); \) see Fig. 1(c). As a result, the specific heat coefficient in the local approximation, \( \gamma_{\mathrm{loc}} \), is given as

\[
\gamma_{\mathrm{loc}} = \frac{\pi^{2}}{6} N(N - 1)\Gamma_{\mathrm{loc}}(0,0)\rho^{f}(0)^{2},
\]

when \( 1/z_{\mathrm{loc}} \gg 1 \). We comment that \( \gamma_{\mathrm{loc}} \approx \gamma \) is expected in usual paramagnetic heavy Fermion systems where magnetic fluctuations are not prominent because in such a case the term \( \Gamma_{i,M,j,M'}(0,0) \) with \( i \neq j \), which represents the inter-site magnetic correlations, will be small \[16\]. In fact, the universal KW relation \( A/\gamma^{2} \sim 1 \times 10^{-7} \mu\Omega cm[\text{mol-K}/\text{mJ}]^{2} \) in many \( \text{Ce} \) and \( \text{U} \)-based HF systems means the validity of the approximation introduced in the present work.

We comment that a similar strong coupling analysis was performed to derive the Wilson ratio \( W_{R} = (\chi/\gamma)(2\pi^{2}k_{B}^{2}/\mu_{\text{eff}}) \) in the present model, and the relation \( W_{R} \approx 1 - 1/(N - 1) \) is derived on condition that the inter-site magnetic-correlations are weak \[16\]. This result will be consistent with a smaller \( W_{R} \) \((W_{R} \lesssim 1)\) in \( \text{YbCu}_{5-x} \text{Ag}_{x} \), considering that \( \mu_{\text{eff}} \) will be slightly smaller than 4.54 \mu\text{B} \( \) for a \( \text{Yb}^{3+} \) ion due to a small but finite CEF \[9\].

Next, we analyze the imaginary part of the self-energy. Its \( T^{2}\)-term within the DMFA is give as \[3\]

\[
\text{Im} \Sigma(0) = \text{Im} \Sigma(0) = \frac{\pi(\pi T)^{2}}{2} (N - 1)\Gamma_{\mathrm{loc}}^{2}(0,0)\rho^{f}(0)^{3},
\]

as shown in Fig. 1(d). Using eq. (9), we derive the expression for \( A \) within the DMFA. According to the Kubo formula, the conductivity \( \sigma \) is given by

\[
\sigma = \frac{e^{2}}{N_{0}} \sum_{k} \left( \frac{\partial f}{\partial \epsilon} \right) \left| G_{k}^{f}(\epsilon) \right|^{2} \left( \frac{\partial \epsilon}{\partial k_{x}} \right)^{2},
\]

where \( f(\epsilon) = (\epsilon / \text{RT} + 1)^{-1} \) and \( G_{k}^{f}(\epsilon) = (\epsilon + \epsilon_{k} - V_{0}^{2} / (\epsilon + \mu - E_{f} - \Sigma(\epsilon)))^{-1} \) is the Green function for \( c \)-electrons. Note that vertex corrections for currents are dropped in eq. (10), which is allowed within the DMFA \[18,19\].
The coefficient $A \equiv \rho/T^2$ is given by eqs. (10) and (9). Assuming the spherical Fermi surface and using the relation $(N/2)\rho_f(0) = \rho''(0) V_0^2 / (\mu - E_f - \Sigma(0))^2 / \rho''(0)$ being the DOS for c-electron per spin, we obtain that

$$A = 3\pi^7 k_F^{-4} N(N - 1) \Gamma_{loc}(0,0) \rho_f(0)^4,$$

where $k_F$ is the Fermi momentum. The number of electrons per unit volume in the present model is given by $n = k_F^3/3\pi^2$. By assuming the free electron model for the conduction electrons and reviving $h$ and $k_B$, we obtain the following "generalized KW relation" in the strong coupling case:

$$A_{\gamma_{loc}}^{-2} = \frac{h}{c^2 k_B^2} \left( \frac{9(3\pi^2)^{-1/3}}{n^{2/3} a^3 N^2} \right) \frac{1}{\pi N(N - 1)} \approx 1 \times 10^{-5} \mu\Omega\mathrm{cm} \cdot \mathrm{mol} \cdot \mathrm{K}/\mathrm{mJ}^2,$$

where both $\Gamma$ and $\rho_f(0)$ are cancelled out. Here, we have used $h/e^2 = 2.6 \times 10^4 \Omega$, $k_B = 1.38 \times 10^{-23} \mathrm{JK}^{-1}$, and assumed that $n^{-1/3} \approx a = 1 \times 10^{-8} \mathrm{cm}$ (a being the lattice spacing).

According to eq. (12), $A_{\gamma_{loc}}^{-2} \approx 1 \times 10^{-5} \mu\Omega\mathrm{cm} \cdot \mathrm{mol} \cdot \mathrm{K}/\mathrm{mJ}^2$ for $N = 2$ ($J = 1/2$), which corresponds to the Kramers doublet ground state due to strong CEF. On the other hand, $A_{\gamma_{loc}}^{-2} \approx 0.36 \times 10^{-6} \mu\Omega\mathrm{cm} \cdot \mathrm{mol} \cdot \mathrm{K}/\mathrm{mJ}^2$ for $N = 8$ ($J = 7/2$), which corresponds to Yb-based HF systems with weak CEF. This result is consistent with the experiments reported in ref. [2].

In the next stage, we study the KW relation in the weak coupling region ($1/z \gtrsim 1$) using the second-order-perturbation-approximation (SOPA) with respect to $U$, both for $J = 1/2$ case and $J = 7/2$ case [17]. In the numerical calculation, we use the spherical Brillouin zone ($|k| \leq \pi$) for simplicity of the numerical calculation. We put $\epsilon_k = -4 + 8(k/\pi)^2$ (the bandwidth being 8), $E_f = -2.5$, $V_0 = 1.8$ and $a = 1.15$ ($n_f = 0.8$). Hereafter, we replace $E_f$ in eq. (1) with $E_f + 0.005\epsilon_k$ for the sake of convenience of numerical calculations. Figure 2 shows the total f-electron DOS, $N\rho_f(0)$, both for $U = 0$ and for $U = 2$ obtained by the SOPA at zero temperature. The non-interacting DOS for $J = 7/2$ coincides with that for $J = 1/2$ except for the sharp peak around $\epsilon = E_f \sim 0.5$ which is given by $G^{\theta}(0)$ in eq. (2). The bottom (top) of the hybridization gap is $\epsilon = 0.17 (1.84)$ for $U = 0$. We note again that $E_f > \mu$ in the present calculation by considering the particle-hole transformation for 4$f^{13}$-electrons in Yb$^{3+}$ ($J=7/2$). In the case of $U = 2$, the DOS around the Fermi level and $E_f$ level are renormalized within a smaller energy width due to the $\epsilon$-dependence of $\mathrm{Re}\Sigma(\epsilon)$ [17,20].

Figure 2 (c) shows the imaginary part of the self-energy for $U = 2$ obtained by the SOPA, which is given by

$$\mathrm{Im}\Sigma(\epsilon - i\delta) = \pi U^2 (N - 1) \int_0^\epsilon \! \! d\omega \int_{-\epsilon+c+\omega}^{i\epsilon+c+\omega} \! \! d\omega' \times \rho_f(\omega) \rho_f(\omega') \rho_f(\epsilon - \omega + \omega').$$

Thus, $\mathrm{Im}\Sigma_{J=7/2}(\epsilon)$ coincides with $(7/64)\mathrm{Im}\Sigma_{J=1/2}(\epsilon)$ for $|\epsilon| \lesssim |E_f - \mu| \sim 0.6$ within the SOPA, as shown in Fig. 2 (c). However, $\mathrm{Im}\Sigma_{J=7/2}(\epsilon)$ takes much larger value for $|\epsilon| \gtrsim 0.6$ by reflecting the huge weight of $G^{\theta}(0)$ around $E_f$. The real part of the self-energy is obtained from eq. (13) using the Cauchy integral. Figure 3 shows $A/\gamma^2$ as functions of $U^2$, where $A$ and $\gamma$ are obtained by the SOPA. $[A/\gamma^2]_{J=7/2} = (7/64)[A/\gamma^2]_{J=1/2}$ is realized within $O(U^2)$, which holds approximately in the weak coupling region where $U^2 \lesssim 0.2$. We find that $1/z_{J=1/2} = 1.25$ and $1/z_{J=7/2} = 1.13$ for $U = 2$. Taking the result by the SOPA as well as eq. (12) derived by the strong coupling analysis, we can naturally estimate that the ratio $A/\gamma^2$ for $J = 7/2$ is about one order smaller than that for $J = 1/2$ in any intermediate coupling case. A perturbation calculation up to $U^4$-order will be useful for a detailed study, which is a future problem.

Finally, we study the KW relation for the following SU(N)-PAM:

$$H = \sum_{k\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_{kM} E_{\text{f}} f_{kM}^\dagger f_{kM} + \sum_{kM} \left( f_{kM}^\dagger c_{kM} + \text{h.c.} \right) + \frac{U}{2} \sum_{\substack{iM \neq M' \nu}} n_{iM}^f n_{iM'}^f,$$

where $M = J, J - 1, \ldots, -J$ and $N = 2J + 1$. Although this model has been frequently analyzed by slave-boson 1/N-expansion method, it is less realistic than eq. (1) in that (i) both c- and f-bands have N-fold degeneracy and (ii) the c-f mixing is allowed only for electrons with equal $M$. Apparently, the Green function is diagonal with respect to $M$. The local f-Green function $g(\epsilon)$ is given by $G^{\theta}(\epsilon)$ in eq. (3), instead of eq. (2).

By performing the same analysis within the DMFA, it is shown that eqs.(8) and (9) are also valid in SU(N)-PAM in the strong coupling limit. On the other hand, the conductivity is given by eq. (10) times $N/2$. By using the relation $\rho_f(0) = \rho''(0) V_0^2 / (\mu - E_f - \Sigma(0))^2$ in SU(N)-PAM, we obtain that

$$A = 12\pi^7 k_F^{-4} N(N - 1) \Gamma_{loc}(0,0) \rho_f(0)^4.$$
In summary, we have studied the KW relation in HF systems with orbital degeneracy. By analyzing the OD-PAM, eq. (1), on the basis of the FL theory, we have derived a generalized KW relation in the strong coupling limit, eq. (12). The obtained result naturally explains the remarkably smaller value of $A\gamma^{-2}$ observed in various Yb-based orbitally degenerate HF systems reported in ref. [2]. A numerical analysis using the SOPA was also presented. Another generalized KW relation has been derived based on the SU(N)-PAM, eq. (16), which also tells that $A\gamma^{-2}$ becomes drastically smaller due to the orbital degeneracy. However, the SU(N)-PAM may be less realistic than the OD-PAM for larger $N$. The present simplified OD-PAM will be enough to understand a global aspect of the KW relation in Ce and Yb-based HF systems. It is an important future problem to study the effect of small but finite CEF splitting on the value of $A\gamma^{-2}$ by numerical methods.

Finally, we comment on the pressure dependence of $A$ in CeCu$_2$Ge$_2$ [21]: It suggests that the value of $A\gamma^{-2}$ decreases suddenly when the ground state degeneracy increases (i.e., $\Delta_{CEF} < T_K$) for $P>15$GPa. This interesting behavior will be explained within the framework of the present study. The generalized KW relation proposed in the present work is confirmed in various HF compounds with $N = 2 \sim 8$ [22]. Its importance will increase further as various new compounds with orbital degeneracy are discovered in future.

The author is grateful to K. Yamada, T. Saso, D. Vollhardt, Y. Yoshimura and N. Tsujii for useful comments and discussions.

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![FIG. 1.](image)

(a) a full four-point vertex. (b) a full local four-point vertex, which is given by $(1 - \delta_{MM'})\Gamma_{loc}$. $\Gamma_{loc}$ is an asymmetric local vertex. (c) expression for $1/Z_{loc} - 1$. (d) expression for $\text{Im}\Sigma$. In (c) and (d), the factor $(-N)$ originates from the summation over $M'$ and the Fermion loops.
FIG. 2. Total $f$-electron DOS ($N\rho^f(\epsilon)$) for (a) $U = 0$ and (b) $U = 2$. A renormalization is recognized in the case of $U = 2$. (c) $\text{Im}\Sigma(\epsilon - i\delta)$ given by SOPA ($U = 2$). $\epsilon = 0$ corresponds to the Fermi level.

FIG. 3. $A_{\gamma}^{-2}$ obtained by SOPA as functions of $U^2$. $A_{\gamma}^{-2}$ for $J = 7/2$ is about 10 times smaller than that for $J = 1/2$ in the weak coupling region.