Kadowaki-Woods ratio for strongly coupled Fermi liquids

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On the basis of the Fermi liquid theory, the Kadowaki-Woods ratio $A/\gamma^2$ is evaluated by using a first principles band calculation for typical itinerant d- and f-electron systems. It is found as observed that the ratio for the d-electron systems is significantly smaller than that for the normal f systems, even without considering their relatively weak correlation. The difference in the ratio value comes from different characters of the Fermi surfaces. By comparing Pd and USn$_3$ as typical cases, we discuss the importance of the Fermi-surface dependence of the quasiparticle transport relaxation.

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It is widely known as a universal feature of heavy fermion systems that there holds the Kadowaki-Woods (KW) relation $A/\gamma^2 = 1 \times 10^{-5}$ $\mu\Omega$ cm (mol K$/mJ)^3$ between the electronic specific heat coefficient $\gamma$ of $C=\gamma T$ and the coefficient $A$ of the resistivity $\rho=AT^2$ in the clean and low temperature limit. According to the Fermi liquid theory, this is interpreted as an indication of the fact that $A$ is a smaller proportionality to quasiparticle mass enhancement due to strong electron correlation. On the other hand, transition metal systems have been reported before to obey a similar relation with a more than an order of magnitude smaller value of $A/\gamma^2$. In view of the observation that there seems to exist several types of systems in this regard, the recent finding by Tsujii et al. is quite impressive that many Yb-based compounds show the KW ratio as small as the transition metals. Kontani derived a simple model is usually adopted at the order of magnitude smaller value of $\rho_T$ in this regard, the recent finding by Tsujii et al. is quite impressive that many Yb-based compounds show the KW ratio as small as the transition metals. Kontani derived a simple model at the unstable state of trivalent Yb by applying the dynamical mean field approximation to a periodic Anderson model of an orbitally degenerate f-electron states coupled with a single conduction band.

To discuss the KW ratio $A/\gamma^2$ and the many-body mass enhancement effect, a simple model is usually adopted at the cost of neglecting material specific individual factors. In the present work, we are interested in such an effect as caused by a system-dependent factor, that is, the Fermi-surface dependence of quasiparticle current relaxation. The system should have a large enough Fermi surface relative to the Brillouin zone boundary in order for the quasiparticle current to dissipate effectively into an underlying lattice through mutual quasiparticle scatterings. In other words, the effectiveness of the transport relaxation may depend on the size and shape of the Fermi surface. To investigate this point definitely, we discuss the quasiparticle transport by taking into account the momentum dependence of quasiparticle scattering on the basis of realistic band structures. This has been hampered so far by a task required for not so simple Fermi surfaces of many band systems as could be simply modeled analytically. In terms of fairly realistic energy bands obtained from a first principles calculation, we evaluate those quantities which are not affected severely by the electron correlation effect. The theory in use is essentially within the phenomenological Fermi liquid theory described by renormalized quantities, and unlike a model calculation, no bare microscopic quantities appear explicitly. Schematic results using simple abstract models have been given before, in which a tight binding square lattice model and a two-band model are investigated.

For the ratio $A/\gamma^2$, we make use of the expression

$$\frac{A}{\gamma^2} \approx \frac{9\alpha F}{8\pi e^2} = 21.3\alpha Fa[\mu\Omega (\text{mol K}/mJ)^2],$$

which corresponds to Eq. (4.11) in Ref. 7 where we set $a=4$ $\AA$ for the lattice constant. In what follows, we substitute a calculated value for $a$. Below, we follow how to derive $\alpha F$, where $\alpha$ is a coupling constant and $F$ is a factor determined by the Fermi surface.

Following a microscopic analysis of the quasiparticle transport with vertex corrections properly taken into account, we may derive a phenomenological linearized Boltzmann equation. Generalizing the theory to take a many-band effect into account, in the low temperature $T\to 0$, we end up with the equation

$$u_{p\mu}^i = (\pi T)^2 \sum_{p'j,k} W_{p\mu}^{ij} \delta(p_j-p_{p'}+k) (l_{p\mu}^i + l_{p'\mu}^i - l_{p\mu+k\mu}^i - l_{p'-k\mu}^i),$$

where $u_{p\mu}^i$ and $\rho_{p\mu}^i = \delta(\mu-e_{p\mu})$ are the velocity component and the local density of state of the renormalized (mass-enhanced) quasiparticle with the crystal momentum $p$ in the $i$th band. The superscripts $i$ and $j$ are the band indices, while the subscript $\mu=x, y, z$ are Cartesian coordinates. In the right hand side of Eq. (2), the second to fourth terms in the parenthesis represent vertex corrections in the microscopic formulation. In terms of the solution $l_{p\mu}^i$, which physically represents stationary deviation of the Fermi surface in an applied electric field $E_\mu$, the conductivity is given by

$$\sigma = \sigma_\mu = 2e^2 \sum_{p, i} \rho_{p\mu}^i l_{p\mu}^i.$$ 

Equations (2) and (3) correspond to Eqs. (3.10) and (3.15) of Ref. 8, respectively. We may suppress the index $\mu=(x)$ in Eq. (3) as we discuss the cubic systems in what follows.

Instead of solving the simultaneous matrix equations [Eq. (2)] exactly, we use trial functions $l_{p\mu}^i$ as commonly applied in a variational principle formulation of the transport problems. Assuming
we obtain

$$\alpha F = \sum_{i,j} \alpha^{ij} c_{ij} \frac{\rho_i \rho_j}{(\rho^2 |v_i|)^2}$$

(4)

where

$$c_{ij} = \sum_{k_1,k_2,k_3,k_4} \rho_{k_1} \rho_{k_2} \rho_{k_3} \rho_{k_4} (e_{i+k_1} - e_{i+k_2} - e_{i+k_3} + e_{i+k_4})^2 / 4 \rho_i \rho_j$$

(5)

and

$$\rho |v_i| = \sum_{i,p} \rho_i^{(p)} |v_{i,p}^{(p)}|$$

(6)

We define coupling constants $\alpha^{ij} = \rho_i \rho_j (W^{ij}) / \pi$, where $\rho_i = \sum \rho^{(p)}_i$ is the density of states of the $i$th band at the Fermi level and $(W^{ij})$ denotes the quasi-particle scattering probability $W^{ij}_{pp^{'},k}$ averaged over the momenta $p$, $p'$, and $k$. As the double sum in Eq. (2), dominated by the Umklapp processes, covers a complicated shaped phase space out of the momentum sum as an averaged quantity. The total density of states $\rho = \sum \rho_i$ is substituted for $\gamma = 2 \pi^2 / 3$.

In heavy fermion systems, the momentum dependence of $W^{ij}_{pp^{'},k}$ could be generally neglected; for the quasiparticle scattering, $W^{ij}_{pp^{'},k}$ is primarily caused by strong on-site Coulomb repulsion $U$. Then, we can make an order of magnitude estimate of $\alpha^{ij}$ in terms of Landau parameters $F_{0}^{i,a}$ and $F_{0}^{a}$. For an anisotropic Fermi liquid, as in an isotropic case, one can derive that the charge and spin susceptibilities are given by $\chi^{c}_1 = 2 \rho_i / (1 + F_{0}^{c})$ and $\chi^{s}_1 = 2 \rho_i / (1 + F_{0}^{s})$, respectively. Thus, for the systems in which charge fluctuations are suppressed, $\chi^{c}_1 \rightarrow 0$, we obtain $F_{0}^{c} \gg 1$. On the other hand, in terms of $A_{0}^{a} = F_{0}^{a} / (1 + F_{0}^{a})$, one obtains a rough estimate of the coupling $\alpha^{cl} = \frac{1}{2} (A_{0}^{a} - A_{0}^{cl})^2 + \frac{1}{2} (A_{0}^{a} + A_{0}^{cl})^2$. Therefore, under the normal condition that the spin enhancement is moderate, $(1 + F_{0}^{s})^{-1} \sim 1$, $\alpha^{cl}$ should universally stay around a constant of an order of unity. This corresponds to the condition to make the Wilson ratio $R_{W} \approx 2$ in the impurity model.\cite{11,12} We discuss a normal state that the system is well away from critical instabilities, around which $A \gamma^2$ will be strongly enhanced at variance with experimental results under consideration.\cite{13,14} We evaluate $E$ numerically for $\alpha = \alpha^{cl} = 1$ to obtain $A / \gamma^2$ and investigate the Fermi-surface dependence.

It is noted that the factor $F$ is determined by the shape and extent of the Fermi surfaces relative to the Brillouin zone boundary. Microscopically, the mass enhancement due to the many-body effect is represented by the $\omega$ derivative of the electron self-energy $\Sigma(q, \omega)$ or by the renormalization factor $\rho^{(p)}_i = \rho^{(p)}_i - \rho^{(p)}_{i,p}$, where $\rho^{(p)}_{i,p}$ is a bare density of states. It is easily checked that the factor $\rho$ cancels in $F$ when $\rho^{(p)}_i$ is independent of $i$. Otherwise, in case that a dominant contribution to the resistivity comes from an electron-correlated main band, then the other bands may be neglected and $A / \gamma^2$ becomes independent of $\omega$ of the main band. As we see below numerically, it is found indeed that $F$ is dominated by a few scattering channels within a main band or two. Hence, we elaborate on a numerical estimate of $F$ on the basis of a realistic band calculation reproducing reliable Fermi surfaces of relevant bands, even if it may not take into account the local many-body correlation effects fully enough for the renormalized quantities such as $\rho_i$ and $\omega_i$ to be separately compared with experiments. As a matter of course, we must exclude the extreme case in which strong correlation modifies electron states around the Fermi level qualitatively from those of a band calculation. We apply our theory to those itinerant electron systems in which correlation strength is not negligible but not so strong.

To calculate $E$ for some typical cubic $d$ and $f$ itinerant electron systems in the $f$cc and $Cu_3Au$ structures, we have performed $ab initio$ band calculations within density functional theory using the plane wave pseudopotential code VASP with the Perdew-Wang 1991 generalized gradient approximation to the exchange correlation functional $E_{xc}$.\cite{14,15-17} By minimizing the total energy, we obtain the lattice constant $a$, which is accurate enough to be used in Eq. (1).

To evaluate $E$ numerically, we have to broaden the delta function $\rho^i_\mu = \delta(\mu - \epsilon^i_\mu)$ by $\Delta$ to pick up electron states around the Fermi level. The width $\Delta$ of the order of real temperature should be decreased as the number of the $k$ points is increased until we confirm to have a convergent result. For the number $L$ of subdivisions along reciprocal lattice vectors, band calculations are performed with $L_{band} \sim 50$, from which we obtain the band energies $\epsilon^i_\mu$ on the finer $k$ mesh of $L \sim 200$ by interpolation. As the fourfold $k$ sum in the numerator of Eq. (4), especially for the most important terms coming from the main $d$ or $f$ correlated bands, constitutes the most time consuming part of the calculation, we have to reduce the numerical task by some symmetry considerations not only on the cubic symmetry of the quasiparticle states but also on the relative directions of the four momentum vectors of the scattering quasiparticle states and the $x$ direction of the current flow. The reduction is particularly effective for the intraband scatterings $i \neq j$.

The calculated results are shown in Table I, where $F$ and $A / \gamma^2$ for $\alpha = \alpha^{cl} = 1$ are shown along with the lattice constant $a$, the number $N$ of metallic bands contributing to the resistivity, and $\rho |v_i|$ defined in Eq. (6). We find that our results

| Material      | $a$ (Å) | $\rho |v_i|$ | $F$ | $N$ | $A / \gamma^2$ |
|---------------|--------|-------------|-----|-----|----------------|
| USn$_3$      | 4.60   | 3.1         | 4.0 | 3   | 0.39           |
| UIn$_3$      | 4.61   | 4.9         | 1.6 | 3   | 0.16           |
| UGa$_3$      | 4.24   | 3.9         | 2.5 | 3   | 0.23           |
| Pd            | 3.86   | 7.4         | 0.23| 3   | 0.019          |
| Pt            | 3.91   | 8.4         | 0.15| 4   | 0.012          |

\(^{a}\)In unit of $\alpha = 1$.

\(^{b}\)In unit of $10^{-5}$ $\mu \Omega$ cm (mol K/mol J)$^2$.\n
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relevant bands, relative magnitudes of
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Among other things, the results indicate that different char-
are a few times smaller than those observed evenly, but the
accuracy of this order should not be taken seriously here.
Among other things, the results indicate that different char-
acters of the Fermi surfaces play an important role.
To show the relative contribution to the resistivity from
relevant bands, relative magnitudes of \( c_{ij} \) in the numerator of
Eq. (4) are shown for Pd and USn\(_3\) in Figs. 1 and 2, respec-
tively. For Pd, the contribution to \( F \) comes from the fourth to
sixth bands, among which dominant is the fifth hole band of
the 3d character. Similarly, the fifth band contributes majorly
not only to \( \rho \), i.e., \( \rho_5 = 5.4 \rho_3 = 12 \rho_0 \), but also to \( \rho |v_\parallel| \) in Eq.
(6). On the other hand, for USn\(_3\), while the 14th heavy elec-
tron band plays a central role, the 12th and 13th hole bands
also make non-negligible contributions through the interband
scatterings. Hence, as the first point to note, numerical im-
portance of the interband contributions makes \( F \) large in the
f-electron system. This is partly because \( \rho_i \) for \( i=12,13,14 \)
are comparable with each other, namely, \( \rho_{14} = 2 \rho_{13} = 3 \rho_{12} \).
Moreover, it is remarked that the large and nearly spherical
shapes of the Fermi surfaces are essential too. As the
second point to note, the importance of the Fermi-surface
geometry can be understood within a single band model by
comparing contribution from the main band. We find that

\[
\frac{c_{5,5}}{\rho_5^2} = 0.097 \quad \text{for Pd is an order of magnitude smaller than}
\frac{c_{14,14}}{\rho_{14}^2} = 0.93 \quad \text{for USn}_3.
\]

The difference comes from the different characters of the Fermi surfaces.

According to an elementary formula \( \sigma = e^2 \rho v^2 \tau = e^2 \rho v l \),
the conductivity \( \sigma \) depends on \( \rho v \) as well as \( l \). In this context,
the mean free path \( l \) is not a single-particle property deter-
mined by a lifetime of the particle state, but it is the transport
property which characterizes how efficiently the total electric
current decays into a lattice system, e.g., in our case,
through the mutual Umklapp scattering processes between
the current carriers. In particular, regardless of interaction,
electrons in free space will not have resistivity.\(^9\) Thus, to
evaluate the transport property \( l \) correctly, it is crucial to take
into account the momentum dependence of the scattering
states and the conservation modulo of the reciprocal lattice
vectors.

Note that \( \rho |v_\parallel| \) defined in Eq. (6) is related to the surface
area \( S \) of the Fermi surfaces, as \( \rho dS = Sdk_\parallel/(2\pi)^3 \). Hence,
\( \rho |v_\parallel| \) too is independent of the mass renormalization \( m \) as \( F \)
is, and for free electrons, we obtain \( \rho |v_\parallel| \approx k^2 \approx n^{3/2} \). One can
see a correlation between \( F \) and \( \rho |v_\parallel| \) in Table I. In fact, Pd
and Pt have twice as large \( \rho |v_\parallel| \) as the uranium compounds.
The difference cannot be simply explained by the difference
in the Fermi-surface volume \( n \). It is caused by the fact that
the \( f \)-electron systems have the nearly isotropic Fermi
surfaces, while the \( d \)-electron systems have complicated ones
with relatively large area compared to their total volume, as
indicated in Figs. 3 and 4. The different characters of the surfaces
affect not only the single-particle quantity \( \rho |v_\parallel| \) but also the transport property of the total current relaxation. As
the order of magnitude difference in \( F \) is not explained
merely by \( \rho |v_\parallel| \), we have to resort to the other factor, that is,
the transport property depending on the Fermi surfaces. It
originates from the detailed \( k \) dependence of the scattering
states, as represented in \( c_{ij} \), or by the phase space volume
available for all possible scattering channels under strict re-
strictions of energy and momentum conservations. Thus, our
quantitative analysis concludes the important effect on the
quasiparticle transport due to the shape and complexity of the
Fermi surfaces.
In summary, we evaluated the Kadowaki-Woods ratio \( A/\gamma^2 \) of some itinerant \( d \)- and \( f \)-electron systems numerically on the basis of the Fermi liquid theory using quasiparticle Fermi surfaces obtained by band calculations. In a single framework, we find that the \( d \)-electron systems have smaller ratio than the \( f \) systems, as observed, and among others, we pointed out an important effect to the transport coefficient \( A \) originating from a commonly neglected specific feature depending on the characters of the Fermi surfaces. The effect is not understood fully as a single-particle property of interacting systems, but we stress the importance of the phase space restriction due to momentum conservation in two-body scattering processes to dissipate a total electric current. In short, to realize effective dissipation, the system should have a large and regular shaped Fermi surface. In future, we will examine that the Fermi-surface dependent efficiency of mutual quasiparticle scatterings may depend on a type of transport current to be relaxed.

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18. In three-dimensional nearly ferromagnetic metals, e.g., for \( A^{RPA}_q = U/\kappa^2 + q^2 \) with \( \kappa^2 \approx 1 - U_{\chi_0} \ll 1 \), we obtain \( \rho^2(A^{RPA}_q)^2 \ll 1 \) for \( 0.04 \approx \kappa \ll 1 \).