I. INTRODUCTION

The discovery of large thermopower in Na$_2$CoO$_2$[1] has prompted quest for related good thermoelectric (TE) materials and encouraged experimental and theoretical research on TE properties of strongly correlated electron systems. In practical TE application, the important material parameter is the dimensionless figure of merit $ZT = S^2/(\kappa/\sigma T)$, where $S$ is the Seebeck coefficient, $\kappa$ the thermal conductivity, $\sigma$ the electrical conductivity, and $T$ the absolute temperature. Hence it is equally as important to enhance $S$ as it is to decrease the Lorenz number $L$ defined by $L = \kappa/\sigma T$. The latter however is usually hampered by the Wiedemann-Franz (WF) law, according to which $L$ should be a universal constant, e.g., $L = \pi^2/(3k/e)^2$ for impure metals. In fact, normal metals, which generally have low $S$, will not be good TE devices with $ZT > 1$, unless the WF law is overcome. It would thus be interesting to investigate a possible material dependence of $L$ to see if it is a controllable variable in principle.

We discuss the ideal value $L_i$ (which is simply denoted as $L$ below) of a correlated electron system on a rigid lattice without impurity in the low temperature limit for the purpose of elucidating its material specific dependence. Indeed, for pure transition metals, the Lorenz numbers have been observed to vary from metal to metal[3, 4, 5, 6, 7]. Theoretically, Herring had derived a universal constant[8], while the problem had also been addressed specifically by treating $s$-$d$ hybridization as perturbation, namely, by a two band model of conductive $s$ electrons scattering off localized $d$ states[9, 10]. Herring’s argument based on the Fermi liquid theory should be the proper approach to the problem at low temperatures. We derive a dimensionless factor (Eq. (10)) so as to modify his constant result, with which we investigate the Fermi surface (FS) dependence of $L$ for some typical cases concretely. After showing nontrivial results obtained for simple models, we give results of numerical evaluation of the Lorenz numbers of some transition metals. Lastly, we find it interesting to investigate a model of the sodium cobalt oxide Na$_2$CoO$_2$, because a nontrivial $x$ dependence of $L$ is expected theoretically owing to its simple cylindrical FS[11].

II. FERMI LIQUID THEORY

We begin with the linearized transport equation of a Fermi liquid under the temperature gradient $\nabla T$ and the electric field $E$,

$$
\frac{\partial n_p}{\partial \epsilon_p} \left(- \frac{\epsilon_p - \mu}{T} \nabla T \cdot v_p + eE \cdot v_p \right) = I[\varphi],
$$

where $n_p = n(\epsilon_p) = 1/(e^{(\epsilon_p - \mu)/T} + 1)$ is the Fermi distribution function, and $W_{pp'}$ represents the transition probability of quasiparticle scattering $p, p' \rightarrow p - k, p' + k$. In terms of the solution $\varphi_p$ of Eq. (1), the electric and heat currents carried by quasiparticles are respectively given by

$$
J = 2e \sum_p v_p \frac{\partial n_p}{\partial \epsilon_p} \varphi_p, \quad Q = 2 \sum_p v_p (\epsilon_p - \mu) \frac{\partial n_p}{\partial \epsilon_p} \varphi_p,
$$

where $\epsilon_p$ and $v_p$ are energy and velocity of quasiparticles, respectively. In comparison with the phenomenological formulae $J = \sigma E - \sigma S \nabla T$ and $Q = T \sigma S E - \kappa_0 \nabla T$, we obtain the transport coefficients $\sigma$, $S$, and $\kappa_0$, the thermal conductivity at zero electric field. The thermal conductivity at zero current, $\kappa$, is given by $\kappa = \kappa_0 - T \sigma S^2[2]$. Formally, the above results expressed in terms of the renormalized quantities $\epsilon_p$ and $v_p$ bear resemblance to those of a weakly interacting Fermi gas. Nevertheless, many body effects are included not only in the renormalization for the individual quasiparticle, but also in the field induced shift of the quasiparticle distribution $\varphi_p$, which implicitly includes a collective Fermi liquid effect depending on Landau parameters[12, 13]. We do not write down the explicit expressions for them as they are irrelevant for our purposes in what follows.

For definiteness, let us assume that the currents $J$ and $Q$ flow in the $x$-direction. Then, for the two functions $l_1$ and $l_2$ defined by $\varphi_p = -eE_x l_1 + \partial_x T l_2$, we obtain the equations

$$
- \frac{\partial n_p}{\partial \epsilon_p} v_{px} = I[l_1],
$$

where $\epsilon_p$ and $v_{px}$ are energy and velocity of quasiparticles, respectively.
and
\[ -\frac{\partial n_p}{\partial x_p} t_p v_{px} = I[l_2], \quad (3) \]

where \( t_p = (\varepsilon_p - \mu)/T \). For an anisotropic system, the collision integral \( I \) has been evaluated analytically by an elaborate approximation\[14\]. As we cannot assume the predominance of normal scattering processes in general cases of anisotropic Fermi liquids of our concern, we have to make approximations in another way. To simplify the multiple momentum sum in \( I \), we replace
\[ (1-n(\varepsilon_p' + k))\delta(\varepsilon_p + \varepsilon_p' - \varepsilon_p - k - \varepsilon_p + k) \]
with \( (1-n(\varepsilon_p + \varepsilon_p' - \varepsilon_p - k)^2 - \varepsilon_p + k) \), for significant contributions to the collision term should come from a thermal neighborhood of the FS in any case. Moreover, as in the isotropic case, to describe the momentum dependence of the solutions \( l_i \), we decouple the crystal momentum variable \( p \) into the radial (energy) direction \( t_p = (\varepsilon_p - \mu)/T \) and the perpendicular component \( \rho_{p'} \), and set \( l_i(p) = M_i(\Omega_p) N_i(t_p) \) \((i = 1, 2)\). Then the momentum sum is written as \( \sum_p = \int \rho d\Omega_p \), where \( \rho \) represents the density of quasiparticle states (DOS) under the normalization condition \( \int d\Omega_p = 1 \). By integrating over the energy variables, we obtain equations for \( M_i(\Omega_p) \).

First we note that we can reproduce the same conductivity formula as derived and discussed previously\[13, 15\]. By setting \( N_1(t_p) \) as a constant, the energy integrals which appear in the four terms in \( I[l_1] \) of Eq. (3) can be calculated analytically. As a result, we obtain

\[ v_{px} = \frac{\pi^2}{2} \rho^2 T^2 \int d\Omega_p \int d\Omega' \rho_{p'} W_{p-p'} v_{px}(M_{1,p} + M_{1,p'} - M_{1,p'} - M_{1,p+p'-p'}) \delta(\varepsilon_{p'+p'} - \mu) \]

\[ = \frac{\pi^2}{2} T^2 \sum_{p',p''} W_{p-p'}(M_{1,p} + M_{1,p'} - M_{1,p'} - M_{1,p+p'-p''}) \rho_{p'} \rho_{p''} \rho_{p+p'-p''}, \quad (4) \]

and

\[ \sigma = 2e^2 \rho \int v_{px} M_1(\Omega_p) d\Omega_p. \quad (5) \]

In Eq. (5), we used \( \rho_p = \delta(\varepsilon_p - \mu) \). Assuming the angular dependence \( M_{1,p} \propto v_{px} \), we obtain the resistivity coefficient

\[ A = \frac{\pi^2}{2} \sum_{p,p',p''} W_{p-p'}(M_{1,p} + M_{1,p'} - M_{1,p'} - M_{1,p+p'-p''}) \rho_{p'} \rho_{p''} \rho_{p+p'-p''} v_{px}(v_{px} + v_{p'x} - v_{p''x} - v_{p+p'-p''}), \]

\[ = \frac{4e^2}{\left( \sum_{p} \rho_p v_{px}^2 \right)^2} \]

for the electrical resistivity \( \sigma^{-1} = AT^2 \).

On the other hand, we have to make a further approximation for \( \kappa_0 \). By taking the \( t \)-derivative at \( t_p = 0 \) of Eq. (3), while adopting \( N_2(t) \propto t/(t^2 + \pi^2) \)[8], we finally obtain similar equations, namely,

\[ v_{px} = \rho^2 T^2 \int d\Omega_p \int d\Omega' \rho_{p'} W_{p-p'}(M_{2,p} - c(M_{2,p'} + M_{2,p+p'-p''}) \delta(\varepsilon_{p+p'-p''} - \mu), \]

where

\[ c = \int_0^\infty dt \left( \text{coth} \frac{t}{2} - 2 \right) \frac{2t/2 + \pi^2}{\sinh t} \approx 0.162, \]

and \( \kappa_0 = 2\pi^2 \rho T \int_{-\infty}^{\infty} \frac{\partial n}{\partial x_p} \int d\Omega_p M_2(\Omega_p) d\Omega_p \), from which we obtain the thermal resistivity \( \kappa_0^{-1} = BT \). It is remarked that the difference in the integrand kernels of Eqs. (4) and (7) stems from the fact that \( N_2(t) \) for \( \kappa_0 \) is an odd function, while \( N_1(t) \) for \( \sigma \) is even. To evaluate the Lorenz number \( L = \kappa/(\sigma T) \) concretely, assuming \( M_2,p \propto v_{px} \) as above, we obtain

\[ L < L_0 = \frac{\kappa_0}{\sigma T} = \frac{\pi^2}{12}(12 - \pi^2)\mathcal{L} \left( \frac{k}{e} \right)^2, \quad \]

where the Boltzmann constant \( k \) is written explicitly\[27\]. The constant prefactor in Eq. (8) is separated as it comes from the energy integral, and corresponds to the result first derived by Herring\[8\], though our result differs by \( \pi^2/12 \). In addition, the dimensionless factor \( \mathcal{L} \) in Eq. (8) originates from the directional dependence of scattering, and is given explicitly by
Note that one would find a trivial result $\mathcal{L} = 1$, a constant Lorenz ratio, if one neglects both of the three vertex correction terms following $v_{p_{i}x}$ in the parentheses of the denominator as well as the numerator of Eq. (9). To have a nontrivial effect $\mathcal{L} \neq 1$, it is essential not to disregard detailed momentum structure of the relevant quasiparticle scatterings on the FS. In fact, $\mathcal{L}$ quantifies the effect of FS geometry on the availability of phase space for the quasiparticle scatterings to relax the transport currents. Essentially, the above expression represents the fact that thermal and electrical resistivities are determined mainly by different types of scattering processes, that is, normal processes are important for the thermal resistivity $B$, while the electrical $A$ is caused by Umklapp processes. In fact, for the numerator, or the resistivity coefficient $A$, to take a finite value, there must exist at least a set of four momenta $p_{i}$ ($i = 1, 2, 3, 4$) on the FS satisfying the Umklapp condition $p_{1} + p_{2} = p_{3} + p_{4} + G$, where $G(\neq 0)$ is a reciprocal lattice vector. This is not met if the FS is too small. Indeed, there is a limit Fermi radius estimated by the relation $|p_{i}| = |G|/4$ which holds at the threshold where all the vectors are parallel or antiparallel. In the vicinity of the threshold, one should expect a strong FS dependence of $\mathcal{L}$. This is a matter of our concern in the following.

III. RESULTS

A. Simple Model

The factor $\mathcal{L}$ may be sensitive to the momentum dependence of the scattering probability $W_{k'}^p = \frac{2\pi}{\xi} \left( |A_{11,k'}^p|^2 + \frac{1}{2} |A_{11,k}^p|^2 \right)$, where $A_{11,k}^p$ and $A_{11,k'}^p$ are the scattering amplitudes for quasiparticles with parallel and antiparallel spins. In particular, the momentum dependence can give rise to a conspicuous effect in the vicinity of quantum critical points where there are quantum fluctuations localized in $k$ space. To take this into account, we may assume

$$A_{11,k}' \sim A_{11,k}^p \sim A_{k}^p = \frac{1}{1 + \xi^2 \gamma(k - Q)}$$

(11)

in order to represent the effect of fluctuations peaked around $k = Q$. In addition to the correlation length $\xi$, one may adopt $\gamma(k) \propto \sum_{d} [1 - e^{i k d}]$, where the sum is taken over the nearest neighboring lattice vectors $d$, as it gives a simple lattice periodic function to give $\gamma(k) \propto k^{2}$ in the long wavelength limit $k \to 0$.

To illustrate how $\mathcal{L}$ varies, let us first investigate a simple two-dimensional (2D) model. We should make a special remark, however. In a strictly 2D system, one will find that normal forward scatterings make the denominator $B$ of Eq. (9) logarithmically divergent, as so for the inverse lifetime of quasiparticle which corresponds to the denominator of Eq. (9) with $c = 0$ [17]. On the other side, the numerator $A$ still remains finite as noted by Fujimoto et al. [17]. In effect, the divergence is suppressed by a small decay rate $\Gamma$ assigned to the quasiparticle states, $\rho_p = \delta(\varepsilon_p - \mu)$. In a real system, such a cutoff must be provided by an inevitable effect of three dimensionality of the system or by a finite density of impurities. As the dependence on $\Gamma$ is logarithmic and weak numerically, here we present a typical behaviour assuming $\Gamma$ as a given constant for simplicity.

First we discuss the simplest case of a parabolic band on a square lattice. It is easy to show that the re-
result is independent of the quasiparticle mass, so that we may simply use $\varepsilon_p = p^2$. We obtain Fig. 1 for $\Gamma = 0.02$, which representatively shows $\mathcal{L}$ as a function of the electron number $n$ per orbital for three types of $W_{k}^{pp'}$ according to Eq. (11). As expected, characteristic anomalies are clearly observed. The solid line for $\xi = 0$ ($W_{k}^{pp'} =$const.) typically shows the first onset of the Umklapp processes involving the smallest reciprocal lattice vector $\mathbf{G} = (2\pi, 0, 0)$ at $n_{c,1} \approx \pi/16 \approx 0.20$, as well as the second one at $n_{c,2} \approx 0.39$ for $\mathbf{G} = (2\pi, 2\pi, 0)$. It is noted that the threshold fillings can be easily evaluated as they are geometrically determined by the given FS. The dashed line indicates that the latter structure for $n > n_{c,2}$ is particularly emphasized by the commensurate antiferromagnetic (AFM) fluctuations with $Q = (\pi, \pi)$. On the other hand, the ferromagnetic (FM) fluctuations with $Q = (0, 0)$ generally suppress $\mathcal{L}$, as they strengthen the relative weight of the normal processes contributing to the thermal resistivity $B$. Similarly, one may obtain results for three dimensional systems, in which no cutoff is required. In Fig. 2 we observe the lattice-structure dependence of $\mathcal{L}$ for the parabolic band in a simple cubic (sc) lattice and a face centered cubic (fcc) lattice with $W_{k}^{pp'} =$const. For the sc lattice the first threshold lies at $n_{c,1} \approx 0.065$ due to $\mathbf{G} = (2\pi, 0, 0)$, while it is at $n_{c,1} \approx 0.085$ with $\mathbf{G} = (2\pi, 2\pi, 0)$ for the fcc lattice. For the latter, the secondary kinks expected at $n_{c,2} \approx 0.13$ and $n_{c,3} \approx 0.37$ corresponding to $\mathbf{G} = (4\pi, 4\pi, 0)$ and $(4\pi, 0, 0)$, respectively, are not so conspicuous as that found clearly at $n_{c,2} \approx 0.19$ due to $\mathbf{G} = (2\pi, 2\pi, 0)$ for the sc lattice. In the inset of Fig. 2 the resistivity coefficients $A$ and $B$ are shown respectively for the sc lattice. We remark that the presence of the threshold $n_{c,1}$ may be more easily anticipated from a relatively gradual $n$-dependence of $\mathcal{L}$ than from the electrical resistivity coefficient $A$, which drops abruptly at $n_{c,1}$. The results exemplify that the ideal Lorenz ratio is not a constant number but shows the lattice structure dependence interestingly.

\begin{table}[h]
\centering
\caption{Calculated values for the ideal Lorenz ratio are compared with experiments.}
\begin{tabular}{|c|c|c|}
\hline
$\mathcal{L}$ & $L_0$ & $L_{exp}$ \\
$10^{-8} \text{V}^2/\text{K}^2$ & $10^{-8} \text{V}^2/\text{K}^2$ & \\
\hline
Pd ($\xi = 0$) & 1.0 & 1.3 \\
Pt ($\xi = 5\text{Å}$) & 0.60 & 0.78 & 1.1 [3] \\
Ni & 1.1 & 1.4 & 1.0 [4] \\
Pd & 1.0 & 1.3 & 0.1 [5] \\
Fe & 0.87 & 1.1 & 1.1 [6] \\
W & 0.61 & 0.79 & 0.2-0.4 [7] \\
\hline
\end{tabular}
\end{table}

\begin{figure}[h]
\centering
\includegraphics[width=0.4\textwidth]{fig2.png}
\caption{$\mathcal{L} \equiv A/B$ of a parabolic band on a sc lattice and a fcc lattice. In the inset, the resistivity coefficients $A \propto \sigma^{-1}/T^2$ and $B$ for the sc lattice are shown as a function of $n$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.4\textwidth]{fig3.png}
\caption{For a single band tight-binding model of Na$_2$CoO$_2$, $\mathcal{L}$ as a function of $x$ as well as the Fermi surfaces at $x = 0.6$ are shown to indicate the strong effect caused by a slight deformation due to the hopping integrals $t_2$ and $t_3$.}
\end{figure}

\section{B. Transition metals}

In principle, we can evaluate $\mathcal{L}$ for real materials though it would generally require a hard task numerically. Let us evaluate them for transition metals in the same manner as described elsewhere, i.e., from the result of a first principle band calculation, we pick up a main band with the largest DOS, for which we apply Eq. (11). Table I shows calculated values along with experimental results. For these to be evaluated, we regarded $W_{k}^{pp'}$ as a constant, except Pd for which presented also is the result with a paramagnon ferromagnetic correlation effect with the correlation length $\xi = 5\text{Å}$, which is taken into account as above in Eq. (11). Considering the approximations made to derive Eq. (11), for the scattering amplitudes and so on, we conclude that we could explain a small $L_{exp}$ of tungsten, among others. This must be primarily due to a peculiar FS. For platinum, however, the observed value is inexplicable by our single band result.
C. Na$_2$CoO$_2$

Lastly, we discuss a tight-binding model of Na$_2$CoO$_2$. Though this material has attracted much interest by its large thermopower, it is of particular interest for us because of its simple cylindrical FS. As a nearly 2D system on a triangular lattice, it has an almost circular hole surface centered at the $\Gamma$ point with a slight hexagonal distortion bulging into the sides of the hexagonal Brillouin zone\textsuperscript{[11, 21, 22]}. Therefore, it provides us with an unique case to realize the nontrivial effect as discussed above. Indeed the FS suggests that the Umklapp threshold must exist around $x_c \sim 0.6$, and one should expect that even a slight change in the shape of the FS around the threshold could have a striking effect on the doping dependence of the genuine quasiparticle transport properties and the ideal Lorenz factor. In fact, if we assume a undistorted parabolic band, it is straightforward to show that we should obtain $x_c = 2n_{\text{c}} - 1 \approx 0.55$. Nevertheless, experimentally, the $T^2$ dependence of the electrical resistivity has been observed up to $x \approx 0.7$\textsuperscript{[23, 24]}, around which, therefore, it must be necessary to take into account a three dimensional lattice distortion\textsuperscript{[21, 22]}. For definiteness and simplicity, we restrict ourselves to the doping regime where the three dimensional effect is irrelevant. We do not consider incipient ferromagnetic in-plane correlations\textsuperscript{[23]}, as they would not modify the result qualitatively. Below, we pay special attention to an expected nontrivial behavior that the factor $\mathcal{L}$ should tend to vanish as we approach a threshold $x \lesssim x_c$. We present results for a 2D tight-binding model on a triangular lattice, in which up to the third neighbor hopping integrals $t_i$ ($i = 1, 2, 3$) are considered as in Ref.\textsuperscript{[20]}, where Kuroki and Arita have discussed the thermopower and the electrical conductivity by a relaxation time approximation with a single time scale $\tau$. Here the approximation must be abandoned from the outset, since otherwise one would only obtain a trivial result of a constant value regardless of the FS, as mentioned above.

To see how $\mathcal{L}$ tends to vanish around $x \lesssim x_c$ qualitatively, we show Fig.\textsuperscript{3} in which the $x$-dependence of $\mathcal{L}$ around $x \lesssim x_c$ is shown for $t_1 > 0$ and $\Gamma/t_1 = 0.02$, along with the Fermi surface for $x = 0.6$. In comparison with the dashed line for $t_2 = t_3 = 0$, the solid line shows the strong effect around $x \lesssim x_c$ due to a slight modification of the FS caused by $t_2$ and $t_3$. In effect, as the threshold value $x_c \sim 0.6$ itself depends sensitively on portions of the FS closest to the Brillouin zone boundary, the result cannot be regarded as a quantitative prediction. Furthermore, unfortunately, it is not easy to compare this nontrivial prediction directly with experimental results, because the intrinsic electron term of the thermal resistivity ($\propto BT$) for the perovskite oxides has been completely outweighed by contributions due to phonons and impurities\textsuperscript{[23, 24]}. Those extrinsic terms have to be separated out properly to verify the nontrivial filling dependence.

Lastly, as the vanishment of the factor $\mathcal{L}$ essentially reflects that of the electrical resistivity, it would be interesting also to calculate the $x$ dependence of the resistivity coefficient $A$, or the Kadokawa Woods (KW) ratio $A/\gamma^2$. To evaluate the resistivity coefficients themselves, however, we need the absolute value of the scattering amplitude. Assuming the strong coupling $\rho^2 W_{p-p'} \approx \pi \mu kT$, e.g., then we obtain $A/\gamma^2 = 9F/16\pi e^2$ with

$$F = \frac{\sum_{i=1,2,3} \rho_i \rho_{i+1} \rho_{i+2} \rho_i - \rho_{i+1} - \rho_{i+2} \rho_i + \rho_{i+1} + \rho_{i+2} \rho_i}{\rho_2 (\sum_p \rho_p v_{px}^2)^2},$$

As in Fig.\textsuperscript{3} for $\mathcal{L}$, we show $F$ as a function of $x$ in Fig.\textsuperscript{4}. The ratio $A/\gamma^2$ also vanishes as $x$ approaches $x_c$, as expected. However, numerically, we find that it should remain of the order of a common value, $A/\gamma^2 \approx 1 \times 10^{-7} \mu \Omega \text{cm (mol K/m)}^2$. Li et al.\textsuperscript{[23]} have observed a strongly enhanced deviation from this standard value for $x = 0.7$, which should be well beyond the threshold $x_c$. It is difficult to regard the numerical deviation simply as a FS effect. Apart from taking into account a relevant three dimensional effect, one would have to assume an enhanced scattering $\rho^2 W_{p-p'}^p \gg 1$, which may be due to strong scatterings caused by a proximity to some sort of instability\textsuperscript{[22]}. 

IV. SUMMARY

In summary, on the basis of anisotropic Fermi liquid theory, we investigated the ideal Lorenz ratio for correlated metals by taking due care of the momentum dependence of transport relaxation processes due to mutual elastic scatterings between quasiparticles. It was shown explicitly that the ideal Lorenz ratio of a correlated electron system is not a constant, but may vary drastically in circumstances and even be made vanishingly small, not only by quantum fluctuations but by a filling control
around thresholds of Umklapp scattering channels. Although it might not be easy to extract the ideal Lorenz value in practice, theoretically we pointed out and discussed that such a nontrivial effect should be expected in such a simple single-band system as Na$_x$CoO$_2$.

The numerical calculations were carried out on Altix4700 at Shizuoka University Information Processing Center.

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[27] In the same manner, we get $\sigma S = \frac{2\pi^2 e^3 T}{3\hbar} \int \frac{d\mathbf{p}}{4\pi^2} M_1 d\Omega_p$. For the parabolic band, we obtain $S = \frac{2\pi^2 e^3 T}{3\hbar} = \frac{e^2 T}{3\hbar} \frac{e}{\rho}$, the same result as that for free electrons.