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Abstract

We investigate a Fermi surface effect on the ideal Lorenz ratio of an anisotropic Fermi liquid caused by the onset of Umklapp scatterings. After discussing simple models by way of illustration, we present numerical results for transition metals, and indicate a material with a simple Fermi surface like sodium cobaltite as a possible candidate to observe the effect.

Keywords: Lorenz number; Fermi liquid; Umklapp process; electron correlation; quasiparticle transport

1. Introduction

The discovery of large thermopower in Na_xCoO_2[1] has prompted investigations for related good thermoelectric (TE) materials and encouraged experimental and theoretical researches on TE properties of strongly correlated electron systems. In practical applications, one of the important material parameters is the figure of merit $ZT = S^2/(\kappa/\sigma T)$, defined by the Seebeck coefficient S, thermal conductivity κ, electrical conductivity σ, and absolute temperature T. While it is important to enhance S, it is equally important to suppress the Lorenz number L defined by $L = \kappa/\sigma T$[2]. However, the latter is usually supposed to be hampered by the Wiedemann-Franz (WF) law, according to which the Lorenz number L should be a universal constant, e.g., $L = \frac{\pi^2}{3}(k/e)^2$ for impure metals. In fact, normal metals which generally have low S are not suitable for good TE devices. Thus it is interesting to see if the limit set by the WF law or the constancy of L is overcome.

We investigate the material dependence of L for correlated paramagnets, i.e., anisotropic Fermi liquids of d and f electrons where electron-electron scatterings are dominant. One of the major interests in recent studies of correlated electrons is persistent non-Fermi liquid behavior[3]. Some unusual properties may be explained within the orthodox framework of Fermi liquid theory. Anisotropic deformation of an Fermi surface caused by Pomeranchuk instability is one of intriguing mechanisms[4, 5, 6]. It must be worthwhile discussing unconventional behavior of anisotropic Fermi liquids in other respect too. We discuss the *ideal* value L_0 of anisotropic Fermi liquids. The Lorenz number of pure transition metals has been observed experimentally[7, 8, 9, 10]. Theoretically, Herring had derived a 'universal' value for the Lorenz number of

Fermi liquids[11]. This problem had been addressed also specifically by treating *s-d* hybridization as perturbation, namely, by a two band model of conduction *s*-electrons scattering off localized *d*-electrons[12, 13]. At low temperatures, Herring's argument based on the Fermi liquid theory should be the proper approach. The constant value of Herring is obtained by neglecting momentum dependence of quasiparticle scatterings to relax thermal and electrical currents altogether. In this paper, we investigate the importance and effect of the momentum dependence on the basis of a formula which we derive theoretically.

In §2, on the basis of Fermi liquid theory, we derive an expression for the Lorenz number to be used in what follows. We introduce a dimensionless factor $\mathcal{L}$, by which we describe the material dependence. Qualitatively, it is not difficult to see how $\mathcal{L}$ depends on the Fermi surface (FS). Quantitatively, however, it is not easy to evaluate multiple integrals for $\mathcal{L}$ duly properly with enough accuracy. This is a striking contrast to ordinary transport calculations based on a relaxation time approximation. In §3.1, we present results for simple models to show the effect representatively. In §3.2, we present numerical results for cubic transition metals, and compare them with available experimental data. Last, in §3.3, we discuss a simple model of the sodium cobaltite to see that the effect may be realized. It is remarked that a qualitatively similar effect should be expected for the resistivity coefficient A of the T^2 term, as the effect is essentially caused by Umklapp processes responsible for finite electrical resistivity.

2. Fermi Liquid theory

2.1. General theory

Under a temperature gradient $\nabla_r T$ and an electric field $\boldsymbol{E}$, the transport equation for the distribution function $n_p(\boldsymbol{r}, t)$ of quasiparticle with electric charge e is given by

$$\frac{\partial n_p}{\partial t} + \nabla_{\boldsymbol{r}} n_p \cdot \nabla_{\boldsymbol{p}} \tilde{\varepsilon}_p + \nabla_{\boldsymbol{p}} n_p \cdot (e\boldsymbol{E} - \nabla_{\boldsymbol{r}} \tilde{\varepsilon}_p) = I(n_p). \quad (1)$$

Here $\tilde{\varepsilon}_p$ denotes *local* excitation energy of the quasiparticle with crystal momentum p, and it depends on the distribution function $n_p(\boldsymbol{r},t)$ implicitly through the Landau function $f_{pp'}$[14]. In the right-hand side, $I(n_p)$ is a collision integral due to intrinsic quasiparticle scatterings. To solve Eq. (1), we introduce the linear deviation $\delta\bar{n}_p(\boldsymbol{r})$ by

$$n_p(\boldsymbol{r}) = n_0(\tilde{\varepsilon}_p, T) + \delta\bar{n}_p(\boldsymbol{r}), \qquad (2)$$

where $n_0(\varepsilon, T) = 1/(\mathrm{e}^{(\varepsilon-\mu)/T} + 1)$ is the equilibrium distribution function at the temperature T. It is convenient to define φ_p by

$$\delta\bar{n}_p(\boldsymbol{r}) = \frac{\partial n_0}{\partial \varepsilon_p}\varphi_p, \qquad (3)$$

according to which φ_p is interpreted as local deformation of the FS caused by the external disturbances. To the linear order in φ_p, it is shown that the collision term $I(n_p) \equiv I[\varphi]$ is given by

$$TI[\varphi] = \sum_{p',k} W_k^{pp'} n_p n_{p'}(1 - n_{p-k})(1 - n_{p'+k})(\varphi_p +$$

$$\varphi_{p'} - \varphi_{p-k} - \varphi_{p'+k})\delta(\varepsilon_p + \varepsilon_{p'} - \varepsilon_{p-k} - \varepsilon_{p'+k}),$$

where $n_p = n_0(\varepsilon_p, T)$ and $W_k^{pp'}$ denotes the transition probability that quasiparticles with momenta p and p' are scattered into those with $p-k$ and $p'+k$. For the left-hand side in Eq. (1), we obtain

$$\nabla_{\boldsymbol{r}} n_p \cdot \nabla_{\boldsymbol{p}} \tilde{\varepsilon}_p = \left(\frac{\partial n_0}{\partial T}\nabla_{\boldsymbol{r}}T + \frac{\partial n_0}{\partial \tilde{\varepsilon}_p}\nabla_{\boldsymbol{r}}\tilde{\varepsilon}_p\right) \cdot \nabla_{\boldsymbol{p}}\tilde{\varepsilon}_p$$

$$= \frac{\partial n_0}{\partial T}\nabla_{\boldsymbol{r}}T \cdot \nabla_{\boldsymbol{p}}\tilde{\varepsilon}_p + \nabla_{\boldsymbol{p}} n_p \cdot \nabla_{\boldsymbol{r}}\tilde{\varepsilon}_p. \qquad (4)$$

The last term is canceled with the second term in the parenthesis in Eq. (1). We may set

$$\frac{\partial n_0}{\partial T} = -\frac{\varepsilon_p - \mu}{T}\frac{\partial n_0}{\partial \varepsilon_p}$$

and $\nabla_{\boldsymbol{p}}\tilde{\varepsilon}_p = \boldsymbol{v}_p$ in Eq. (4), and

$$\nabla_{\boldsymbol{p}} n_p = \frac{\partial n_p}{\partial \varepsilon_p}\boldsymbol{v}_p \qquad (5)$$

in Eq. (1). In a steady state, the first term of Eq. (1) vanishes. As a result, we obtain

$$\frac{\partial n_p}{\partial \varepsilon_p}\left(-\frac{\varepsilon_p - \mu}{T}\nabla T \cdot \boldsymbol{v}_p + e\boldsymbol{E} \cdot \boldsymbol{v}_p\right) = I[\varphi]. \qquad (6)$$

The electric and heat currents carried by quasiparticles are given by

$$\boldsymbol{J} = 2e\sum_p \boldsymbol{v}_p \frac{\partial n}{\partial \varepsilon_p}\varphi_p, \qquad (7)$$

and

$$\boldsymbol{J}_Q = 2\sum_p \boldsymbol{v}_p(\varepsilon_p - \mu)\frac{\partial n}{\partial \varepsilon_p}\varphi_p, \qquad (8)$$

respectively. The solution φ_p of the linear equation (6) is represented as a sum of the terms proportional to $\boldsymbol{E}$ and $\nabla_{\boldsymbol{r}}T$. By comparing Eqs. (7) and (8) with the phenomenological formula,

$$\boldsymbol{J} = \sigma\boldsymbol{E} - \sigma S\nabla T,$$
$$\boldsymbol{J}_Q = T\sigma S\boldsymbol{E} - \kappa_0\nabla T,$$

we get expressions for the transport coefficients σ, S, and κ_0, where κ_0 is the thermal conductivity at zero electric field. The thermal conductivity at zero current κ is given by $\kappa = \kappa_0 - T\sigma S^2$[2]. The difference between κ and κ_0 can be neglected at low temperatures. Although the above results expressed by the renormalized quantities ε_p, $\boldsymbol{v}_p$ and φ_p remind us of similar results for a weakly interacting Fermi gas, it should be remarked that the above results include many body effects not only through the renormalization of the individual quasiparticles, but also in the shift φ_p caused by the applied fields, which implicitly includes a collective Fermi liquid effect depending on the Landau function $f_{pp'}$[14, 15]. We do not present their explicit expressions, as they do not affect the results of our investigation.

For definiteness, assuming that the currents $\boldsymbol{J}$ and $\boldsymbol{J}_Q$ flow in the x-direction, we introduce the two functions l_1 and l_2 by

$$\varphi_p = -eE_x l_1 + \partial_x T l_2. \qquad (9)$$

Then we obtain the equations

$$-\frac{\partial n_p}{\partial \varepsilon_p}v_{px} = I[l_1], \qquad (10)$$

and

$$-\frac{\partial n_p}{\partial \varepsilon_p}t_p v_{px} = I[l_2], \qquad (11)$$

where $t_p = (\varepsilon_p - \mu)/T$. The transport coefficients are obtained by substituting Eq. (9) into Eqs. (7) and (8), for which we have to solve the integral equations, Eqs. (10) and (11).

2.2. Approximations

In the *isotropic* system of ^{3}He, the collision integral I is given by normal processes obeying the conservation of momentum, and it has been evaluated analytically by Abrikosov and Khalatnikov[16]. They evaluated thermal conductivity of the isotropic Fermi liquid by an elaborate approximation based on the geometry of the scattering processes. Similarly, we follow them to decouple the crystal momentum $\boldsymbol{p}$ into the radial (energy) component $t_p = (\varepsilon_p - \mu)/T$ and the perpendicular (angular) component $\bar{\Omega}_p$ to set $l_{i,p} = M_i(\bar{\Omega}_p)N_i(t_p)$ ($i = 1, 2$). Accordingly, the momentum sum is given by $\sum_p = T\int\rho\mathrm{d}t\int\mathrm{d}\bar{\Omega}_p$, where ρ represents the density of (quasiparticle) states (DOS) under the normalization condition $\int\mathrm{d}\bar{\Omega}_p = 1$. The FS dependence comes in through $M_i(\bar{\Omega}_p)$, for which equations are obtained by integrating out $N_i(t_p)$ over the energy variable t_p. To this end, however, we have to adopt a

different method, as we discuss an anisotropic Fermi liquid on a lattice where Umklapp processes should be treated on an equal footing with normal processes. To simplify the multiple sum in I, we replace $(1 - n(\varepsilon_{p'+k}))\delta(\varepsilon_p + \varepsilon_{p'} - \varepsilon_{p-k} - \varepsilon_{p'+k})$ with $(1 - n(\varepsilon_p + \varepsilon_{p'} - \varepsilon_{p-k}))\delta(\varepsilon_{p'+k} - \mu)$. This must be legitimate because significant contributions to the collision term should come from a thermal neighborhood of the FS. This will not affect our results essentially, as we discuss below at the end of §2.3. It is important for us that this approximation brings no specific FS dependence into our results.

First we note that we can reproduce an electrical conductivity formula derived and discussed previously microscopically [15, 17]. By setting $N_1(t_p)$ as a constant, we can evaluate analytically the energy integrals for the four terms of $I[l_1]$ in Eq. (10). As a result, we obtain

$$
\begin{aligned}
v_{px} &= \frac{\pi^2}{2}\rho^2 T^2 \int \mathrm{d}\bar\Omega_{p'} \int \mathrm{d}\bar\Omega_{p''} W^{pp'}_{p-p''}(M_{1,p} + M_{1,p'} \\
&\quad -M_{1,p''} - M_{1,p+p'-p''}) \times \delta(\varepsilon_{p+p'-p''} - \mu) \\
&= \frac{\pi^2 T^2}{2}\sum_{p',p''} W^{pp'}_{p-p''}(M_{1,p} + M_{1,p'} \\
&\quad -M_{1,p''} - M_{1,p+p'-p''})\rho_{p'}\rho_{p''}\rho_{p+p'-p''}, \quad (12)
\end{aligned}
$$

and

$$
\sigma = 2e^2\rho \int v_{px} M_1(\bar\Omega_p)\mathrm{d}\bar\Omega_p. \quad (13)
$$

In Eq. (12), we used $\rho_p = \delta(\varepsilon_p - \mu)$. For practical purpose, we assume the angular dependence $M_{1,p} \propto v_{px}$. This does not spoil the essential point discussed below, that is, normal and Umklapp processes are distinguished in terms of the function $M_{1,p} + M_{1,p'} - M_{1,p''} - M_{1,p+p'-p''}$ in Eq. (12), which should not vanish for the latter processes to give a finite electric resistivity. In effect, by a simple model calculation, we find that this approximation is numerically reliable[15]. Thus, from Eqs. (12) and (13), we obtain the resistivity coefficient A for the electrical resistivity $\sigma^{-1} = AT^2$[15],

$$
\begin{aligned}
A &= \frac{\pi^2}{4e^2\left(\sum_p \rho_p v_{px}^2\right)^2}\sum_{p,p',p''} W^{pp'}_{p-p''}\rho_p \rho_{p'}\rho_{p''}\rho_{p+p'-p''} \\
&\quad v_{px}(v_{px} + v_{p'x} - v_{p''x} - v_{p+p'-p''x}). \quad (14)
\end{aligned}
$$

Next we derive a corresponding expression for the thermal conductivity. This is more difficult because to this end we need to take into account the energy dependence of the solution. In this respect, a simple functional form for $N_2(t)$ is often assumed by noting that $N_2(t)$ should be an odd function of t. Following Herring[11], we substitute $N_2(t) \propto t/(t^2 + \pi^2)$, which holds valid in the low energy limit of our concern. Note that the energy dependence has been derived and discussed previously[16, 18]. Thus, by taking the t-derivative at $t_p = 0$ of Eq. (11), we obtain a similar equation,

$$
v_{px} = \rho^2 T^2 \int \mathrm{d}\bar\Omega_{p'} \int \mathrm{d}\bar\Omega_{p''} W^{pp'}_{p-p''}(M_{2,p} - c(M_{2,p'}
$$

$$
+M_{2,p''} + M_{2,p+p'-p''}))\delta(\varepsilon_{p+p'-p''} - \mu), \quad (15)
$$

where

$$
c = \int_0^\infty \mathrm{d}t \left(t \coth\frac{t}{2} - 2\right)\frac{2t/(t^2 + \pi^2)}{\sinh t} \simeq 0.162, (16)
$$

and

$$
\kappa_0 = 2\pi^2\rho T \int_{-\infty}^\infty \frac{\partial n}{\partial t}\frac{t^2 \mathrm{d}t}{t^2 + \pi^2}\int v_{px} M_2(\bar\Omega_p)\mathrm{d}\bar\Omega_p. \quad (17)
$$

From these, we obtain the thermal resistivity coefficient B of $\kappa_0^{-1} = BT$. It is remarked that the difference in the integrand kernels of Eqs. (12) and (15) stems from the fact that $N_2(t)$ for κ_0 must be an odd function, while $N_1(t)$ for σ is even. This is required to make the respective conductivities finite. Assuming $M_{2,p} \propto v_{px}$ as above, we obtain the ratio

$$
L_0 \equiv \frac{\kappa_0}{\sigma T} = \frac{\pi^2}{12}(12 - \pi^2)\left(\frac{k}{e}\right)^2 \mathcal{L}, \quad (18)
$$

where the Boltzmann constant k is written explicitly. The constant factor in Eq. (18) originates from the energy dependence, while the dimensionless factor $\mathcal{L}$ is determined by the directional dependence of the scatterings. For the former, our result differs by a factor of $\pi^2/12 \simeq 0.82$ from the previous result[11]. This is because we set $N_1 =$const. to recover our previous results for σ, which is exact in the sense of Fermi liquid theory. The dimensionless factor $\mathcal{L}$ in Eq. (18) is given by

$$
\begin{aligned}
\mathcal{L} &= \frac{A}{B} = \int \mathrm{d}\bar\Omega_1 \mathrm{d}\bar\Omega_2 \mathrm{d}\bar\Omega_3 W^{p_1 p_2}_{p_1-p_3} v_{p_1 x}(v_{p_1 x} + v_{p_2 x} \\
&\quad -v_{p_3 x} - v_{p_1+p_2-p_3,x})\delta(\varepsilon_{p_1+p_2-p_3} - \mu) \\
&\quad / \int \mathrm{d}\bar\Omega_1 \mathrm{d}\bar\Omega_2 \mathrm{d}\bar\Omega_3 W^{p_1 p_2}_{p_1-p_3} v_{p_1 x}(v_{p_1 x} - c(v_{p_2 x} \\
&\quad +v_{p_3 x} + v_{p_1+p_2-p_3,x}))\delta(\varepsilon_{p_1+p_2-p_3} - \mu) \\
&= \sum_{1,2,3} W^{p_1 p_2}_{p_1-p_3}\rho_{p_1}\rho_{p_2}\rho_{p_3}\rho_{p_1+p_2-p_3} v_{p_1 x}(v_{p_1 x} \\
&\quad +v_{p_2 x} - v_{p_3 x} - v_{p_1+p_2-p_3,x}) \\
&\quad / \sum_{1,2,3} W^{p_1 p_2}_{p_1-p_3}\rho_{p_1}\rho_{p_2}\rho_{p_3}\rho_{p_1+p_2-p_3} v_{p_1 x}(v_{p_1 x} \\
&\quad -c(v_{p_2 x} + v_{p_3 x} + v_{p_1+p_2-p_3,x})). \quad (19)
\end{aligned}
$$

2.3. General discussion

It is generally difficult to evaluate the multiple momentum sums in $\mathcal{L}$ duly properly with good accuracy. This is not only because the integrals cannot be deconvoluted into simpler forms but also because there occur the delta function singularities in the integrands. Before detailed calculations, let us see how $\mathcal{L}$ varies qualitatively.

First it is noted that we get a trivial constant result $\mathcal{L} = 1$ if we neglect all the vertex correction terms, i.e., the three terms following $v_{p_1 x}$ in the parentheses both in the denominator and the numerator of Eq. (19). Thus, to

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obtain a non-trivial result $\mathcal{L} \neq 1$, it is essential not to disregard the detailed momentum structure of relevant quasiparticle scatterings. In fact, $\mathcal{L}$ is determined by the shape and the size of FS. In a sense, it is a mathematical expression of the fact that thermal and electrical resistivities are controlled by different types of scattering processes. Normal processes are important for the thermal resistivity B, while the electrical A is caused by Umklapp processes. For the numerator, or the resistivity coefficient A, to take a finite value, there must exist at least a set of four momenta $\boldsymbol{p}_i$ ($i = 1, 2, 3, 4$) on the FS satisfying the Umklapp condition $\boldsymbol{p}_1 + \boldsymbol{p}_2 = \boldsymbol{p}_3 + \boldsymbol{p}_4 + \boldsymbol{G}$, where $\boldsymbol{G}(\neq 0)$ is a reciprocal lattice vector. This condition is not met when the FS is too small. The limit Fermi momentum is estimated by the equation $|\boldsymbol{p}_i| = |\boldsymbol{G}|/4$ which holds at a threshold where all the momenta are parallel or anti-parallel. In the vicinity of the threshold, it is expected that $\mathcal{L}$ should have a strong FS dependence. This is the main interest of our investigation. The differences in the effects of vertex corrections to electric and heat currents have been microscopically analyzed[19]. We argue that the Lorenz number is the ideal quantity to reveal the difference in the vertex corrections to the two different currents manifestly.

Although our primary interest is in the Lorenz ratio, it would be worthwhile to mention the Seebeck coefficient S. In the same manner as Eq. (13), we find

$$\sigma S = \frac{2\pi^2 e}{3} T \int \frac{\mathrm{d}(\rho v_{px})}{\mathrm{d}\varepsilon} M_1 \mathrm{d}\bar{\Omega}_p. \tag{20}$$

As σ depends on M_1, the effect of the angular dependence M_1 is canceled for the thermopower S. Thus it is concluded that S would be determined primarily by the energy derivative of quasiparticle DOS at the Fermi level. This is formally the same result as for free electrons, though in our case the electron mass m^* to which S is proportional is effectively enhanced by a many body effect. For instance, we obtain $S = \frac{2\pi^2}{3e} \frac{\rho'}{\rho} T = \frac{\pi^2}{3e} \frac{T}{\varepsilon_F}$ for the Fermi energy $\varepsilon_F = \frac{p_F^2}{2m^*}$ of a parabolic band in the isotropic system. Hence, to discuss the enhancement of S quantitatively, we have to make microscopic analysis on the one-electron property m^*. This is beyond the scope of this paper. Furthermore, in the vicinity of a ferromagnetic quantum critical point, we have to take into account paramagnon drag thermopower in addition to this diffuse term, as we discussed recently[20].

To conclude this section, we make a numerical estimate of the vertex-correction effect on the thermal resistivity coefficient B. The effect on B is quantitative but not qualitative. For definiteness, we focus ourselves to the isotropic Fermi liquid, for which one may consult the classical work by Abrikosov and Khalatnikov[16]. Their result is given by

$$B \propto \langle 2W(\theta, \phi)(1 - \cos\theta)/\cos(\theta/2) \rangle, \tag{21}$$

where θ and ϕ denote scattering angles between quasiparticles and the outer bracket denotes to take an average over them. Formally, this can be written as a sum of two terms, $B = B_{\mathrm{qp}} + B_{\mathrm{vc}}$. The former B_{qp} is a quasiparticle term which is formally obtained by using a quasiparticle lifetime in place of a proper transport lifetime. The latter B_{vc} denotes the vertex correction term. By close inspection, we find

$$B_{\mathrm{qp}} \propto \langle 3W/\cos(\theta/2) \rangle, \tag{22}$$

and

$$B_{\mathrm{vc}} \propto -\langle W(1 + 2\cos\theta)/\cos(\theta/2) \rangle, \tag{23}$$

with the same proportionality factor. By neglecting the angular dependence of W, we obtain $B_{\mathrm{vc}}/B = -1/8 \simeq -0.13$. On the other hand, the denominator of Eq. (19) gives $B_{\mathrm{qp}} \propto \langle W \rangle$ and $B_{\mathrm{vc}} \propto -c\langle W(1 + 2\cos\theta) \rangle$, so we get $B_{\mathrm{vc}}/B \simeq -0.16$ under the same condition. Thus we find that the correction effect is negative in sign and the relative magnitude is of the order of 0.1. In fact, we can check that this is actually the case from numerical results of the following sections. The above estimate indicates that inaccuracy of the approximations should be of the order of several per cent.

3. Results

3.1. Nearly free electron model

To show a singular dependence of $\mathcal{L}$ on the electron density, we begin with simple lattice models with an isotropic Fermi surface, for which the angular integrals in Eq. (19) are considerably simplified.

Microscopically, the scattering probability $W_k^{pp'}$ is given by the scattering amplitudes for quasiparticles with parallel and anti-parallel spins, $A_{\uparrow\uparrow,k}^{pp'}$ and $A_{\uparrow\downarrow,k}^{pp'}$, i.e., by

$$W_k^{pp'} = \frac{2\pi}{\hbar} \left(|A_{\uparrow\downarrow,k}^{pp'}|^2 + \frac{1}{2}|A_{\uparrow\uparrow,k}^{pp'}|^2 \right). \tag{24}$$

The momentum dependence of $W_k^{pp'}$ may become conspicuous in the vicinity of quantum critical points, where there occur quantum fluctuations localized in k space. To take this effect into account, we adopt

$$A_{\uparrow\uparrow,k}^{pp'} \propto A_{\uparrow\downarrow,k}^{pp'} \propto A_k^{pp'} = \frac{1}{1 + \xi^2 \gamma(\boldsymbol{k} - \boldsymbol{Q})}, \tag{25}$$

which is peaked at the wavevector of an incipient order $\boldsymbol{k} = \boldsymbol{Q}$. The parameter ξ is the correlation length of the fluctuations around $\boldsymbol{k} = \boldsymbol{Q}$. To the integrals, dominant contributions come from around the long wavelength limit $k \to 0$, so that we use $\gamma(\boldsymbol{k}) = \frac{1}{z} \sum_{\boldsymbol{d}} (1 - \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{d}})$ as a lattice periodic function to reproduce the proper behavior $\gamma(\boldsymbol{k}) \simeq k^2$ in this limit, where z is the coordination number of the lattice. The sum is taken over the nearest neighboring lattice vectors $\boldsymbol{d}$.

For a two-dimensional (2D) model with $\boldsymbol{d} = (\pm 1, 0), (0, \pm 1)$, and $z = 4$, we have to make a special remark. One finds that normal forward scatterings make the denominator B
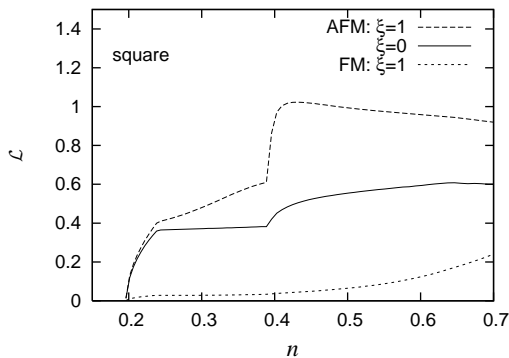
Figure 1: For a parabolic band on a square lattice, the dimensionless Lorenz factor $\mathcal{L}$ is shown as a function of the number of electrons per orbital n. The solid line is the result for the correlation length $\xi = 0$, i.e., without any fluctuation effect. The dashed line is the result for which the fluctuations with $\xi = 1$ around the ordering vector $\boldsymbol{Q} = (\pi, \pi)$ (AFM) are taken into account. The dotted line is for $\xi = 1$ and $\boldsymbol{Q} = (0, 0)$ (FM). In all cases, $\mathcal{L}$ vanishes at the threshold $n_{c,1} \simeq 0.2$.

Figure 2: The factor $\mathcal{L}(\equiv A/B)$ of a parabolic band on a simple cubic (sc) and a face centered cubic (fcc) lattice. In the inset, the electric resistivity coefficient $A \propto \sigma^{-1}/T^2$ and the thermal one B for the sc lattice are shown as a function of n.

Table 1: Calculated values of the ideal Lorenz ratio L_0 and the experimental values $L_{\exp}$.

	$\mathcal{L}$	L_0 (10^{-8}V^2/K^2)	$L_{\exp}$ (10^{-8}V^2/K^2)
Pd ($\xi = 0$)	1.0	1.3	
Pd ($\xi = 5$Å)	0.60	0.78	1.1 [7]
Ni	1.1	1.4	1.0 [8]
Fe	0.87	1.1	1.1 [9]
W	0.61	0.79	0.4 [10]

of Eq. (19) logarithmically divergent. This is caused by the same mechanism as it occurs for the inverse lifetime of quasiparticle, for which there occurs the sum for $c = 0$ of the denominator in Eq. (19)[21]. On the other side, the numerator A still remains finite, as noted by Fujimoto *et al.*[22]. To see a typical filling dependence of $\mathcal{L} = A/B$, we put a small decay rate Γ to quasiparticles. In a real system, such a cutoff must be provided either by three dimensionality or by impurities. In effect, the dependence on Γ is numerically very weak. In the nearly free electron approximation, we use $\varepsilon_p = p^2$ for the quasiparticle dispersion, for the result is independent of the quasiparticle mass.

In Fig. 1, we show $\mathcal{L}$ for the parabolic band on a square lattice as a function of the electron number n per orbital for $\Gamma = 0.02$. The solid line is the result for $W_k^{pp'}$=const., i.e., $\xi = 0$, which clearly indicates characteristic anomalies, as expected. The first onset of Umklapp processes involving the smallest reciprocal lattice vector $\boldsymbol{G} = (2\pi, 0)$ is noticed at $n_{c,1} = \pi/16 \simeq 0.20$, and the second one occurs at $n_{c,2} \simeq 0.39$ for $\boldsymbol{G} = (2\pi, 2\pi)$. The threshold fillings can be estimated straightforwardly, as they are geometrically determined by the given FS. The dashed line in Fig. 1 represents the result for the commensurate antiferromagnetic (AFM) fluctuations with $\boldsymbol{Q} = (\pi, \pi)$ and $\xi = 1$, for which a shoulder structure for $n > n_{c,2}$ is emphasized by the fluctuations. By contrast, the dotted line is the result for the ferromagnetic (FM) fluctuations with $\boldsymbol{Q} = (0, 0)$. In this case, $\mathcal{L}$ is suppressed. This is because scatterings with the fluctuations increase the relative weight of normal processes contributing to the thermal resistivity B.

Similarly, we show results for three dimensional (3D) systems. In Fig. 2, the lattice-structure dependence of $\mathcal{L}$ is presented for the parabolic band. The filled circles are the result for a simple cu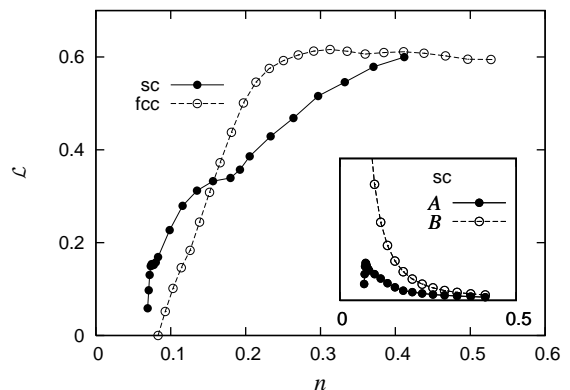bic (sc) lattice, while the open circles are the result for a face centered cubic (fcc) lattice. In both cases, we set $W_k^{pp'}$ as constant. As the numerical evaluation was time consuming, in Fig. 2, the calculated points are connected by the lines as a guide to the eye. The results are interpreted similarly as above. The first threshold of the sc lattice lies at $n_{c,1} \simeq 0.065$ for $\boldsymbol{G} = (2\pi, 0, 0)$, while for the fcc lattice we obtain $n_{c,1} \simeq 0.085$ with $\boldsymbol{G} = (2\pi, 2\pi, -2\pi)$. For the former, we observe the secondary kink at $n_{c,2} \simeq 0.19$ due to $\boldsymbol{G} = (2\pi, 2\pi, 0)$. For the latter, however, the corresponding structures expected at $n_{c,2} \simeq 0.13$ and $n_{c,3} \simeq 0.37$ for $\boldsymbol{G} = (4\pi, 4\pi, 0)$ and $(4\pi, 0, 0)$ respectively are not conspicuous. In the inset of Fig. 2, the resistivity coefficients A and B are shown, respectively, for the sc lattice. We find that the presence of the threshold $n_{c,1}$ is more easily anticipated from a relatively gradual n-dependence of $\mathcal{L}$ than from the electrical resistivity coefficient A. The latter changes abruptly around $n_{c,1}$.

To summarize, it was shown that the ideal Lorenz ratio of a Fermi liquid is not constant but can have a peculiar lattice structure dependence. In principle, it is not difficult to know where to expect the singular behavior. The singularity becomes less distinct in 3D systems than in 2D systems, so that it may be easily overlooked in 3D systems.

3.2. Transition metals

In practice, generally, we confront difficulties in the numerical evaluation of the ratio $\mathcal{L}$. By following the same

line as in the previous calculations[23], we manage to obtain results for some special materials with a highly symmetric lattice structure. By focusing ourselves on pure transition metals in cubic systems, we make use of band structures obtained from the first principles calculations by the Vienna Ab-initio Simulation Code (VASP)[25]. Then we apply Eq. (19) to the main band with the largest DOS, which is physically considered to make a dominant contribution to the resistivities. We make full use of the cubic symmetry in order to reduce the numerical task. The other subsidiary bands would have only a minor effect in the sense that they contribute lesser to the resistivities, not only because they have smaller weights of the DOS, but their Fermi surfaces are so small and/or detached from the zone boundary that Umklapp scatterings should become ineffective geometrically. In fact, this is in conformity with our previous results in a many band analysis[23]. For example, for palladium, we pick up the fifth band with an intricate open heavy d electron surface, which exhibits flat dispersion from X to W points above the Fermi level[24] (Fig. 3 of [23] corresponds to Fig. 4 of [24]). Large values observed for the resistivity coefficients A and B are primarily ascribed to this hole band, while the other bands, e.g, the sixth electron band, have only a minor effect[23, 24].

The results are shown in Table 1 with experimental results[7, 8, 9, 10]. We set $W_k^{pp'}$ as constant, with the exception of Pd, for which we present also the result with the ferromagnetic correlation length $\xi = 5\text{Å}$[26] taken into account according to Eq. (25). We observe a discernible role of the fluctuations. We find a small value for tungsten, which is qualitatively in agreement with an experimental report. This is caused by the fact that the relevant band allows a rather small number of Umklapp processes owing to its peculiarly necked FS[27]. Quantitatively, however, we need some remarks. To extract the electron terms for tungsten is more difficult than the other transition metals, because the resistivity coefficients A and B as well as a electronic specific heat coefficient γ are small. Wagner *et al.* have interestingly reported very small values of $L_{\exp}$ for several samples[10]. In Table 1, we cited $L_{\exp} = 0.4 \times 10^{-8}$ V^2/K^2 for the sample with the lowest residual resistivity $\rho_0 = 0.566 \times 10^{-10}\Omega$cm. They obtained the resistivity coefficients by least squares fitting for 1.5 - 6.0K[28]. They included a phonon term to fit the electrical resistivity, while no phonon term was observed correspondingly in the thermal resistivity. This is inconsistent theoretically. At low temperatures below 1.5K, Uher *et al.* found a consistent fit without phonon term, i.e., with $\rho = \rho_0 + AT^2$[28]. They observed that the electrical resistivity coefficient A is systematically modified from those of Wagner *et al.* This may hold true for the thermal resistivity, though not reported. Furthermore, they observed only a small deviation from Matthiessen's rule, i.e., no systematic variation of A with ρ_0[28]. In contrast, the thermal resistivity coefficient B tends to decrease with decreasing impurity content[28]. By plotting $L_{\exp}$ against ρ_0

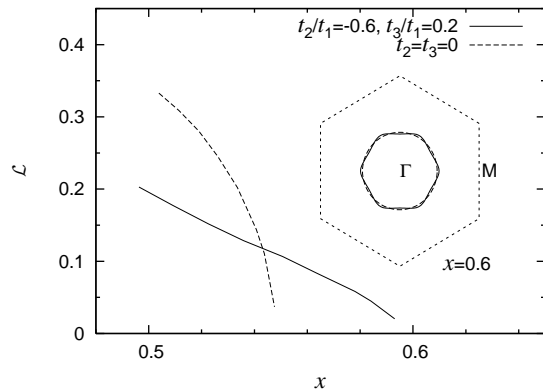Figure 3: $\mathcal{L}$ as a function of the filling x and the Fermi surface at $x = 0.6$ of a single band tight-binding model of Na$_x$CoO$_2$. A slight deformation of the Fermi surface due to the hopping integrals t_2 and t_3 has a strong effect on $\mathcal{L}$.

of the experimental data[10], we observe an apparent increase in $L_{\exp}$ as ρ_0 decreases past $1.0 \times 10^{-10}\Omega$ cm down to $0.5 \times 10^{-10}\Omega$ cm. Therefore, the ideal limit $L_{\exp}$ to be compared with our L_0 can be larger than that presented in Table 1. Furthermore, in general, it might be necessary to consider explicitly the extraneous effects due to impurities and so on. In this regard, Matthiessen's rule is usually assumed, according to which the electronic contribution to resistivity applies independently of and additively to the other scattering mechanisms. Even if the rule is violated, the qualitative predictions on the FS dependence should not lose validity. We plan to investigate the Matthiessen rule in a future work, according to which we find an interesting deviation of the rule caused by the thermal resistivity, though it is negligible in the context of this work (cf. Appendix).

3.3. Na$_x$CoO$_2$

Last, we discuss a simple model of Na$_x$CoO$_2$, which interests 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 Γ point with a slight hexagonal distortion bulging into the sides of the hexagonal Brillouin zone[29, 30, 31]. The FS suggests that the Umklapp threshold lies around $x_c \sim 0.6$. As noted above, even slight change in the shape of the FS around the threshold should have an appreciable effect on the doping dependence of quasiparticle transport properties. If we assume an undistorted parabolic band, we get an estimate of $x_c = 2n_{c,1} - 1 \simeq 0.55$. Experimentally, the T^2 dependence of the electrical resistivity has been observed up to $x \simeq 0.7$[32, 33], around which therefore it is necessary to take account of three dimensional lattice distortion[30, 31]. For simplicity, we neglect the three dimensional effect and incipient ferromagnetic in-plain correlations[34]. To examine how the factor $\mathcal{L}$ tends to vanish around a threshold $x \lesssim x_c$, we use a 2D tight-binding model on a triangular
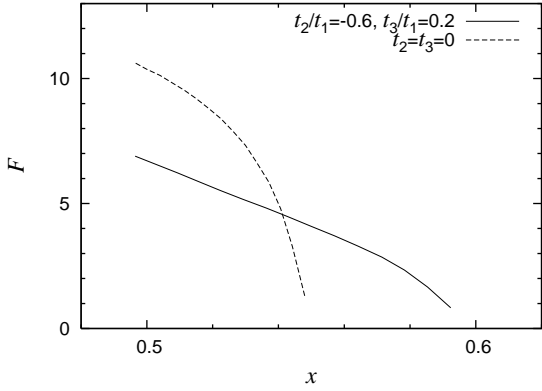
Figure 4: As in Fig. 3, we show the filling x dependences of the factor F, to which the Kadowaki-Woods ratio A/γ^2 is proportional.

lattice, in which up to third neighbor hopping integrals t_i ($i = 1, 2, 3$) are considered[35]. In Fig. 3, 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 $x = 0.6$. To compare with the dashed line for $t_2 = t_3 = 0$, the solid line shows the strong effect around $x \lesssim x_c$ caused by slight change of the FS due to t_2 and t_3. As the threshold value $x_c \sim 0.6$ should depend sensitively on portions of the FS closest to the Brillouin zone boundary, the value itself must not be taken too seriously quantitatively.

Unfortunately, the theoretical prediction is not directly compared with experiments, because normally the intrinsic electron term for the thermal resistivity ($\propto BT$) of the perovskite oxides is outweighed by the terms due to phonons and impurities[32, 33]. Therefore, we present the result for the resistivity coefficient A, as the FS dependence of the factor $\mathcal{L}$ is in good correspondence to that of the electrical resistivity. For the coefficient A, or the Kadowaki Woods (KW) ratio A/γ^2, the absolute value of the scattering amplitude is required. Under the strong coupling condition $\rho^2 W_{p-p''}^{pp'} \simeq \pi$[23], we obtain $A/\gamma^2 = 9F/16\pi e^2$ with

$$F = \frac{1}{\rho^4 \left(\sum_p \rho_p v_{px}^2 \right)^2} \sum_{1,2,3} \rho_{p_1} \rho_{p_2} \rho_{p_3} \rho_{p_3 - p_1 - p_2}$$
$$\times v_{p_{1x}}(v_{p_{1x}} + v_{p_{2x}} - v_{p_{3x}} - v_{p_1 + p_2 - p_3, x}) \qquad (26)$$

We display F as a function of x in Fig. 4, which looks similar to $\mathcal{L}$ in Fig. 3. The ratio A/γ^2 goes to zero as x approaches x_c, but it is of the order of a common value, $A/\gamma^2 \simeq 1 \times 10^{-5} \mu\Omega$ cm(mol K/mJ)^2. Li et al. have observed a strongly enhanced value at $x = 0.7$[32]. The T^2-dependence must be due to intrinsic quasiparticle scatterings. Nevertheless, it seems difficult to consider the deviation simply as the FS effect, and it is less likely to be caused by randomness or imperfection (Appendix). It may be due to enhancement of the scattering strength $\rho^2 W_{p-p''}^{pp'} \gg 1$ caused by a proximity to some sort of instability[32].

4. Summary

On the basis of anisotropic Fermi liquid theory, we investigated the ideal Lorenz ratio of correlated metals by taking due care of the momentum dependence of quasiparticle scatterings to relax electrical and thermal currents. It was shown explicitly that the ratio is not constant but can vary very conspicuously. The ratio can be made vanishingly small not only by quantum fluctuations but by the filling control across the thresholds of Umklapp scatterings. We derived a mathematical formula and applied it to cubic transition metals. Although it is generally not a simple matter to extract the ratio experimentally in practice, we discussed that the variable ratio may be corroborated in a correlated metal with a two-dimensional Fermi surface.

Appendix A. Impurity Effect

In the main text, we assumed the clean limit where extraneous effects due to impurities and imperfections are disregarded. In order to see that they do not affect our result essentially, we discuss electrical resistivity in the opposite limit where quasiparticle scatterings are dominated by the extrinsic elastic scatterings. For this purpose, to the right-hand side of Eq. (10), we add an impurity scattering term I_{imp} given by

$$I_{\text{imp}}[l_1] = \left(-\frac{\partial n_p}{\partial \varepsilon_p} \right) \frac{l_1}{\tau}, \qquad (A.1)$$

where a phenomenological relaxation time τ is introduced. In the dirty limit, the electron term $I[l_1]$ can be neglected altogether. Then we obtain $l_1 = l_1^0 \equiv v_{px}\tau$, as the zeroth order solution. This gives the residual resistivity $\rho_0 = \sigma_0^{-1}$. To obtain the correction term of the electronic origin, we look for a perturbative solution by setting $l_1 = l_1^0 + l_1^1$, where l_1^1 denotes the first order correction. To the first order, we get

$$I[l_1^0] + I_{\text{imp}}[l_1^1] = 0, \qquad (A.2)$$

from which we obtain l_1^1 explicitly. In terms of the solution $l_1 = l_1^0 + l_1^1$, we obtain the total conductivity $\sigma = \sigma_0 + \sigma_1$, where the electronic term σ_1 depends linearly on l_1^1 so that it gives the electronic resistivity proportional to T^2. Owing to $\sigma_0 \gg \sigma_1$ by assumption, the resistivity $\rho = \sigma^{-1}$ is given by

$$\rho \simeq \sigma_0^{-1} - \frac{\sigma_1}{\sigma_0^2} \equiv \rho_0 + AT^2. \qquad (A.3)$$

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For the resistivity coefficient A thus defined, it is straightforward to check that the same result is recovered as given in Eq. (14). Therefore, the formula used in the main text is valid in this case too. In effect, it is surely expected that the resistivity coefficient should not strongly depend on impurities. This is physically plausible, as it represents nothing but a specific example of Matthiessen's rule. The rule is customarily adopted also for thermal resistivity similarly. We examine a deviation from the rule in a future work by developing an exact solution of a transport equation.

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