

Study of electron-electron scattering and the thermal resistivity of alkali metals.

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Abstract: In this paper the current understanding of the electron-electron scattering contribution to the temperature dependent part of the thermal resistivity (W_{ee}) of the alkali metals is reviewed. These systems are modelled by a uniform electron gas, with the electron mass modified to take account of band structure effects and the electron-electron interaction modified to take account of the screening of this interaction by the core electrons. A simple approximation for the scattering function based on the Landau parameters has been used. By Comparing predicted values of W_{ee} with experiment in the alkali metals, where the model is most appropriate, we conclude that the approximate scattering function is sufficiently accurate. Predicted values of W_{ee} in most simple metals are much smaller than in the alkalis. Estimates of the contribution from electron-electron scattering to the electrical resistivity based on the same scattering function have also been calculated for the alkali metals.

Key words: band structure, core electrons, electrons gas, Landau parameter, scattering function, thermal resistivity.

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I. Introduction

In recent years there has been considerable interest in the influence of electron-electron scattering on the transport properties of metals. Most attention has focused on the T^2 contribution to the electrical resistivity, ρ_{ee} , at low temperatures [Poddar and Kumar (2013)[1] Kumar et al (2015)[2], Poddar and Choudhary (1995)[3], Thakur and Poddar (1976)[4], Potter and Morgan (1979)[5], Poddar and Choudhary (1998)[6], Ruthruff et al (1978)[7], Black (1978)[8], and Lawrence (1976)[9]. The analysis of this contribution to ρ_{ee} is complicated even for simple metals by the critical importance of Umklapp scattering. From a theoretical point of view the electron-electron scattering contribution to the thermal resistivity (W_{ee}), which is proportional to T , is a simpler quantity to study. Since fewer approximations are required for its evaluation, it may be a more appropriate testing ground for theory. Moreover, as pointed out by Laubitz (1970)[10] in his study of the transport properties of the noble metals, the presence of W_{ee} leads to negative deviations of the Lorentz function $L(T) = \rho(T)/\{TW(T)\}$ from its classical value Lo [Ziman(1960)[11]] at temperatures above the Debye temperature. As a result simultaneous measurement of $\rho(T)$ and $W(T)$ at high temperatures can provide accurate values for W_{ee} . In this way W_{ee} has been measured for the alkali metals [Cook et al [(1972)[12], Cook (1979a)[13], Cook and Laubitz (1978)[14], Cook (1979b)[15] and Cook (1979c)[16]]. The existence of reliable experimental information makes clear tests of approximations for the electron-electron scattering function possible, especially for the alkali metals where the complicating influence of band structure effects is minimal.

A previous theoretical treatment of W_{ee} has been given by Kukkonen and Wilkins (1979)[17]. We shall compare the predictions of the two theories and comment on the connection between the two approximations for the scattering function.

II. Materials and Methods

The quasiparticle transport equation for the thermal conductivity of Fermi liquids has been solved independently by Brooker and Sykes (1968)[18] and by Jensen et al (1968)[19]. The expression for the thermal resistivity is,

$$W_{ee} = [3m^*4T / 8 \pi^2 h^6 k_F^3 H(\lambda)] \langle \omega(\theta, \Phi) (1 - \cos\theta) / \cos(\theta/2) \rangle \quad (1)$$

Where

$$H(\lambda) = \frac{3-\lambda}{4} \sum_{n=0}^{\infty} \frac{(4n+5)}{(n+1)(2n+1)[(n+1)(2n+3)-\lambda]} \quad (2)$$

$$\lambda = \langle \omega(\theta, \Phi) (1 + 2\cos\theta) / \cos(\theta/2) \rangle / \langle \omega(\theta, \Phi) / \cos(\theta/2) \rangle \quad (3)$$

$$\omega(\theta, \Phi) = \frac{1}{2} \omega_{\uparrow\downarrow}(\theta, \Phi) + \frac{1}{4} \omega_{\uparrow\uparrow}(\theta, \Phi) \quad (4)$$

$\omega_{\uparrow\downarrow}(\theta, \Phi)$ and $\omega_{\uparrow\uparrow}(\theta, \Phi)$ are the transition probabilities for parallel and opposite spin scattering respectively, and $\langle \omega(\theta, \Phi) / \cos(\theta/2) \rangle$ denotes an average over solid angle. The transition probabilities are related to the four-point scattering function Γ , by the equation.

$$w_{\uparrow\sigma_2}(\theta, \Phi) = (2\pi/\hbar)/z^2(K_F)\Gamma^x(p_1 \uparrow, p_2\sigma_2, p_1 + q \uparrow, p_2 - q\sigma_2)^2 \quad (5)$$

[Pines and Nozieres (1966)[20], Kukkonen (1975) [21]. All the momenta in equation (5) are on the Fermi surface and it is implicitly assumed that Γ^x is to be evaluated on the energy shell. The angle θ of equations (1), (3) and (4) is the angle between the initial momenta of the two quasiparticles p_1 and p_2 while Φ is the angle through which the relative momentum of the quasiparticles is scattered [Pines and Nozieres (1966)][20]. The perturbation expansion of Γ has been discussed by Nozieres (1964)[22], Kukkonen and Wilkins (1979)[17], have evaluated $\omega(\theta, \Phi)$ by approximating a subsum of the perturbation theory expansion for Γ . This is a very fundamental approach but in view of the complexity of Γ and the lack of any small parameter in the perturbation theory it is not possible to justify rigorously and particular approximation. On the other hand the forward scattering limit ($q=0=\Phi$) is related to the Landau parameters of a charged fermi liquid by

$$Z^2(k_f)\Gamma^\infty(p_1 \uparrow, p_2\sigma_2; p_1 \uparrow, p_2\sigma_2) = \frac{\pi^2 \hbar^2}{m^* k_F} \left[1 \pm \frac{F_0^a}{1+F_0^a} + \sum_{l=1}^{\infty} \left(\frac{F_l^a}{1+\frac{F_l^a}{2l+1}} \pm \frac{F_l^a}{1+\frac{F_l^a}{2l+1}} \right) \right] P_l(\widehat{p}_1 \cdot \widehat{p}_2) \quad (6)$$

[Nozieres (1964)[22], Brinkmann et al (1968) [23]] where the definitions given by Pines and Nozieres (1966)[20] have been used for the Landou parameters and the + and - signs are for $\sigma_2 = \uparrow$ and $\sigma_2 = \downarrow$ respectively. Using equation (6) in equations (5) and (1) and defining the dimensionless transition probability $\tilde{\omega}(\theta, \Phi)$ by

$$\omega(\theta, \Phi) \equiv (2\pi/\hbar) [\pi^2 \hbar^2 / (m^* k_F)]^2 \tilde{\omega}(\theta, \Phi) \quad (7)$$

yields

$$w_{ee} = [2.622 \times 10^{-2} m^* r_N^5 T/H(\lambda)] \langle \tilde{\omega}(\theta, \Phi)(1 - \cos\theta)/\cos(\theta/2) \rangle (cmKmw^{-1}) \quad (8a)$$

$$\tilde{\omega}_{\uparrow\sigma}(\theta, \Phi) = |\tilde{A}_{\uparrow\sigma}(\theta, \Phi)|^2 \quad (8b)$$

$$\tilde{A}_{\uparrow\uparrow}(\theta, \Phi = 0) = 1 + \frac{F_0^a}{1+F_0^a} + \sum_{l=1}^{\infty} \left[\left(\frac{F_l^a}{1+\frac{F_l^a}{2l+1}} + \frac{F_l^a}{1+\frac{F_l^a}{2l+1}} \right) P_l(\cos\theta) \right] \quad (8c)$$

$$\tilde{A}_{\uparrow\downarrow}(\theta, \Phi = 0) = 1 - \frac{F_0^a}{1+F_0^a} + \sum_{l=1}^{\infty} \left[\left(\frac{F_l^a}{1+\frac{F_l^a}{2l+1}} - \frac{F_l^a}{1+\frac{F_l^a}{2l+1}} \right) P_l(\cos\theta) \right] \quad (8d)$$

Where m^* in equation (8a) is to be given in units of the free-electron mass and $r_N = (\frac{4}{3} \pi n a_0^3)^{-\frac{1}{3}}$ is the usual metallic conduction-electron density parameter. Since $\tilde{\omega} \approx 1$ and $H(\lambda) \approx \frac{1}{2}$, equation (8a) exhibits the critical qualitative features of the trend of W_{ee} values among the alkali metals. The m^{*2} dependence will give rise to a significant band effect. More critical is the r_N^5 ($n^{-5/3}$) dependence which also plays an important role in the electron-electron scattering contribution to the electrical resistivity [Lawrence and Wilkins (1973)[24]]. It is clear from the outset that W_{ee} will be much higher in the low-density alkali metals than it is in the simple metals of groups 1B, 2B, 3A and 4A.

The Landau parameters of equations (6) and (8) can be evaluated from perturbation theory [Hedin (1965)[25], Rice (1965)[26]] with more confidence than the full scattering function and there is agreement on their approximate values for the electron-gas system. Moreover, as has been pointed out by Dy and Pethick (1969)[27], for He^3 , if we assume that only s and p waves contribute to the q dependence of the scattering, the scattering amplitudes are completely specified, by a symmetry argument, by their $\Phi=0$ values. Making this assumption yields

$$\tilde{A}_{\uparrow\uparrow}(\theta, \Phi) = \tilde{A}_{\uparrow\uparrow}(\theta, \Phi = 0) \cos \Phi \quad (9a)$$

$$\tilde{A}_{\uparrow\downarrow}(\theta, \Phi) = \frac{1}{2} (2\tilde{A}_{\uparrow\downarrow}(\theta, \Phi = 0) - \tilde{A}_{\uparrow\uparrow}(\theta, \Phi = 0) + \tilde{A}_{\uparrow\uparrow}(\theta, \Phi = 0) \cos \Phi) \quad (9b)$$

[We take the values at $\Phi=0$ from equations (8c) and (8d)]. This approximation has proved useful in applications to He^3 ; a test of its accuracy for a charged fermion system is presented.

There remains the problem of approximating the Landau parameters for a simple metal. We start from a model Hamiltonian $\tilde{H} = (\tilde{T}/m_b) + (\tilde{U}/\epsilon)$ where \tilde{T} and \tilde{U} are respectively the Kinetic and interaction-energy operators of the uniform-electron gas system and m_b is in units of the free electron mass. In general m_b will differ from unity because of band effects while $\epsilon = 1 + 4 \pi N \alpha$ where N is the number density of the ions and α is the polarisability of the individual ion cores [Perdew and Wilkins (1973)[28], Vosko and Perdew (1975)[29], Kukkonen and Wilkins (1979)[17]. The Landau parameters are related to the product of the quasiparticle Fermi-level density of states and functional derivatives of the self energy with respect to quasiparticle distribution functions. It follows from an examination of the perturbation-theory expressions for the self energy that the Landau parameters for the system with Hamiltonian \tilde{H} are given by

$$F_l^i(r_a) = F_{l(h)}^i \left(\frac{m_b r_a}{\epsilon} \right) \tag{10}$$

$$m^* = m_b \left[1 + \left(\frac{F_1^a}{3} \right) \right] = m_b \left\{ 1 + \left[F_{l(h)}^a \left(\frac{m_b r_a}{\epsilon} \right) / 3 \right] \right\} \tag{11}$$

III. Results and Discussion

For the range of density appropriate to the alkali metals the sum over l in equations (8b) and (8c) seems to converge rapidly, and we truncate it at $l = 1$. F_0^a and F_1^b were evaluated using

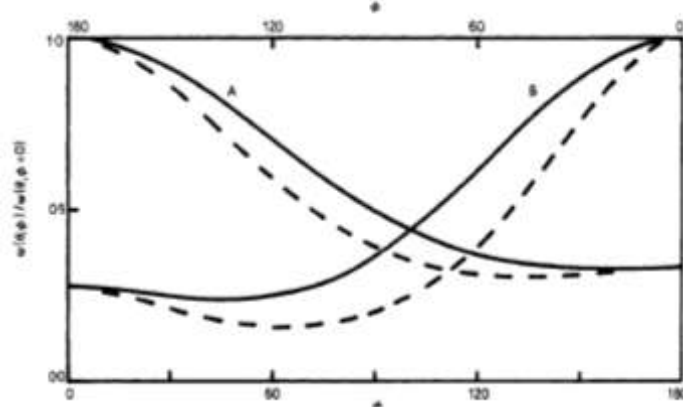


Fig. 1

Figure: 1 Comparison of $\tilde{\omega}_{\text{DP}}(\theta, \phi)$ in the Born Thomas Fermi approximation as a function of ϕ for $x=1$ ($r_s=3.01$). The two sets of curves are for $\theta = 90^\circ$ (curves A, scale on lower axis) and $\theta = 180^\circ$ (curves B, scale on upper axis). For both, the exact dependence on ϕ is indicated by a broken curve while the sp approximation to this dependence is indicated by a full curve and the quantity plotted is the ratio of $\tilde{\omega}(\theta, \phi)$ to the exact value at $\phi = 0$.

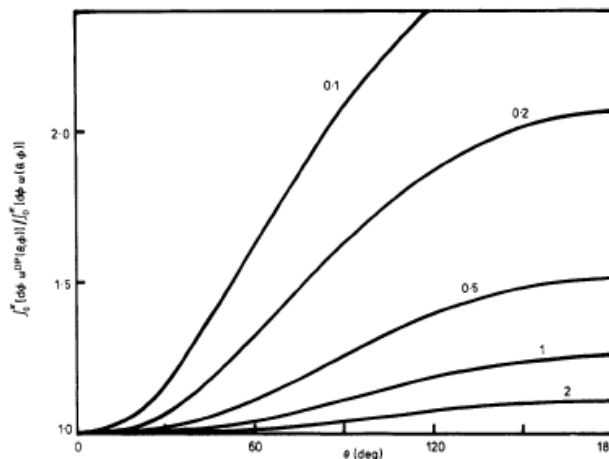


Figure. 2. Comparison of $\tilde{\omega}_{\text{DP}}(\theta, \phi)$ and $\tilde{\omega}(\theta, \phi)$ in the Born Thomas-fermi approximation as a function of θ for several densities x (values marked on curves), for each value of x (where $r_s \approx 3.01x$) the ratio of the average of $\tilde{\omega}_{\text{DP}}(\theta, \phi)$ over ϕ to the average of $\tilde{\omega}(\theta, \phi)$ over ϕ is presented as a function of θ

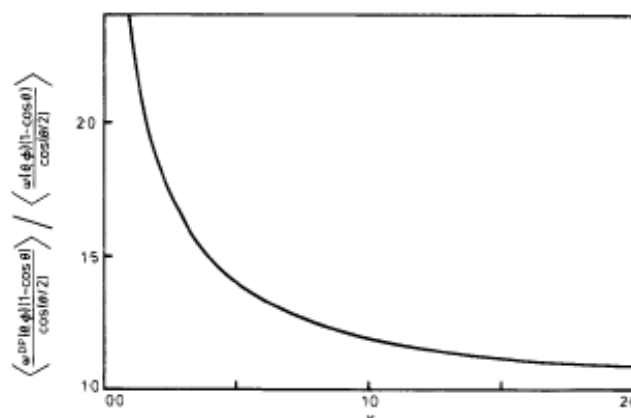


Figure : 3. The relative error produced in $\langle \tilde{\omega}(\theta, \phi) (1-\cos\theta)/\cos(\theta/2) \rangle$ by using the Dy- Pethick approximation for the ‘Born Thomas-fermi’ $\tilde{\omega}(\theta, \phi)$. It is noted that the error decreases rapidly with decreasing electron density and is $\leq 20\%$ in the alkali-metal density regime ($x \geq 1$).

equation (10) and the electron-gas calculation by Hedin (1965)[25] while F_1^a was fixed by the Pauli-principle requirement.

$$\tilde{\omega} = \uparrow\uparrow (\theta=0, \Phi=0) = 0 \tag{12}$$

[Brinkman et al. (1968)][23]. It is instructive to exhibit the sensitivity of the results to the model parameters m_b and ϵ , and we do so in figure (4) for K. The important point is that the predicted values of W_{ee} are rather insensitive to ϵ and depend on m_b mainly through the m_b factor in the quasiparticle mass [equation (11)]. This can be understood in terms of the behaviour of the Landau parameters as a function of density : as shown in figure (5) according to the calculation by Hedin they are almost constant in the range $r_a = 3.6$. For the purpose of comparison the Landau parameters obtained in the random-phase approximation (RPA) as calculated by Shastry (1978)[30] are also shown.

The calculated values of W_{ee} for the alkali metals are compared with experiment in Table 1. The values of m_b were taken from the calculation by Mac Donand and Vosko (1976)[31]. It should be noted that recent bandstructure calculations of the Fermi-level density of states in the alkali metals agree to within $\sim 1^a_0$ [Ching and Calla way (1974)[32], (1975)[33], Moruzzi et al 1978[34] Wilk et al (1979)[35]] and so these values have a high degree of confidence. It is frequently claimed that band structure calculations already include many-body corrections to the bare mass through their exchange correlation potentials. We do not believe that this is true since for a uniform system the band structure mass reduces to the bare-electron mass and not to the quasiparticle mass. The relation of the band structure mass to the quasiparticle mass in a real metal has been discussed in more detail by Mac Donald (1979)[36]. The values chosen for ϵ were those deduced from optical-reflectance measurements [Whang et al. (1971)[37] (1972a)[38], (1972b)[39]] for K, Rb and Cs. The semi-empirical value given by Mayer and Mayer (1933)[40], has been used for Li and the value for Na was taken from the work by Tessmann et al (1953)[41].

Table 1.

Electron-electron scattering contributions to the thermal and electrical resistivities in the alkali metals. $W_{ee}^{DMR}(\Delta)$ and $\rho_{ee}(\Delta)$ denote the contributions to the resistivities with band effects included. $W_{ee}^{DMR}(\Delta)$ has been corrected to account for the interference between electron-electron and electron-phonon scattering mechanisms as discussed in the text.

Metal	m_b	ϵ	Δ	$\frac{W_{ee}(\Delta=0)}{T}$ (cm MW ⁻¹)	$\frac{W_{ee}(\Delta)}{T}$ (10 ⁻¹⁴ ΩcmK ²)	$\frac{\rho_{ee}(\Delta)}{T^2}$ (10 ⁻¹⁴ ΩcmK ⁻²)	$\frac{w_{ee}^{DMR}(\Delta)}{T}$ (cm MW ⁻¹)	$\frac{w_{ee}^{DMR}(\Delta)}{T} - \frac{\rho_{ee}(\Delta)}{L_0 T^2}$ (cm MW ⁻¹)
Li ($r_b=3.25$)	1.54	1.02	0.08	73	75	12	100	Theory 95 Experiment —
Na($r_b=3.94$)	1.04	1.08	0.02	86	87	04	110	110 110±60 ^a
K ($r_b=4.87$)	1.08	1.15	0.04	280	290	20	350	340 270±50 ^b
Rb ($r_b=5.20$)	1.10	1.25	0.06	400	410	50	490	470 350±50 ^c
Cs ($r_b=5.63$)	1.30	1.35	0.09	730	730	140	900	830 800± ^d *

^aCook et al (1972) [12]

^cCook (1979a) [13]

^bCook (1979b) [15]

^dCook (1979c) [16]

* This is a preliminary number and no reliable error bars have been established to date.

Two corrections to the theoretical result should be included before comparing with experiment. The first type of correction we refer to as band Corrections and treat by following the analysis given by Lawrence and Wilkins (1973)[24]. It follows from their theory that, to a good approximation

$$w_{ee}(\Delta) = w_{ee}(\Delta = 0) \left[1 + \left(\frac{3\Delta}{8} \right) \right] \left[1 + \left(\frac{\Delta}{13} \right) \right] \left[1 + \left(\frac{3\Delta}{22} \right) \right]^{-1} \quad (13)$$

and

$$\rho_{ee}(\Delta) = 0.8L_0T w_{ee}(\Delta = 0)\Delta \left[1 + \left(\frac{\Delta}{10.4} \right) \right]^{-1} \quad (14)$$

Where $W_{ee}(\Delta = 0)$ is the thermal resistivity in the absence of band effects and Δ referred to as the ‘fractional Umklapp scattering’ in part reflects the variation of the magnitude of the quasiparticle velocity over the Fermi surface. Lawrence and Wilkins give an approximate formula for Δ which for the alkali metals becomes.

$$\Delta \sim 1.9 (V_{110}/\epsilon_f) [\tan^{-1}(3.09 V_{110}/\epsilon_f) - \tan^{-1}(0.38V_{110}/\epsilon_f)] \quad (15)$$

Where ϵ_f is the Fermi energy and V_{110} is a pseudopotential coefficient for the first Bragg plane. We have used non-local pseudopotentials calculated by the method given by Rasolt and Taylor (1975)[42] in evaluating V_{110} . [The pseudopotential parameters for Na are given by Cohen et al (1976)[43], these for Li and K by Dagens et al (1975)[44] and those for Rb and Cs by Poddar & Shekhar[45]. The corrected values of W_{ee} and the values of ρ_{ee} obtained by this procedure are listed in Table 1. Because of the Crudeness of equation (3.4), which is based on a two-OPW model and a local pseudopotential, the values for ρ_{ee} are rough estimates only; however they may be of use in guiding the search for such terms in the low-temperature electrical resistivity of simple metals.

The second correction required for the comparison of theory and experiment accounts for deviations from Matthiesen’s rule arising from the interference of electron-electron and electron-phonon scattering mechanisms. C.R. Leavens (1973) has shown that the apparent coefficient of T in the thermal resistivity above the Debye temperature should be augmented by approximately 20% by DMR provided that the electron-phonon scattering can be treated as elastic at these temperatures and that W_{ee} is much less than the electron-phonon contribution. This conclusion is consistent with the analysis by Lawrence and Wilkins (1973)[24] who also indicate that the DMR for ρ_{ee} should be much smaller.

Table- 2

Comparison of the thermal resistivities, spin susceptibilities and quasiparticle masses implied by two approximations for the electron-gas Landau parameters within the model outlined in the text. The superscript H refers to the Landau parameters calculated by Hedin (1965)[25], while the superscript RPA refers to the Landau parameters in the random-phase approximation.

Metal	W_{ee}^H/T (cmMW ⁻¹)	W^{RPA}/T (cm MW ⁻¹)	m^{*H}/m	m^{*RPA}/m	m^{*exp}/m^a	Z^H/Z_0	Z^{RPA}/Z_0	Z^{exp}/Z_0^b
Li	70	120	1.53	1.69	1.55±0.15	2.40	2.84	2.45→3.0
Na	86	97	1.02	1.10	1.08±0.04	1.60	1.69	1.64±0.02
K	280	430	1.07	1.17	1.11±0.03	1.69	1.92	1.70±0.01
Rb	400	620	1.09	1.19	1.10±0.04	1.72	1.95	1.72±0.01
Cs	730	1490	1.29	1.46	1.23±0.06	1.98	2.56	2.24±0.06

^a The values were obtained by removing the mass enhancements recommended by Grimvall (1976)[46] from the specific heat masses given by Kittel (1971)[47].

^b Mac Donald and Vosko (1976)[31] for references and further discussion.

The experimental quantity with which we compare is $W/T-\rho/(L_0T^2)$ for $T \gg \theta_D$ which can be identified with $W_{ee}/T- \rho_{ee}/(L_0T^2)$ since the phonon contribution drops out [Laubitz (1970)[10]. The agreement between theory and experiment in good, indicating that both the Dy-Pethick approximation and the model for including crystalline effects (m_b and ϵ) and adequate. The sensitivity of these results to the approximation for the electron-gas Landau parameters is illustrated in Table-2. Where the W_{ee} in the absence of band effects, obtained with the RPA Landau parameters. The larger values for the RPA Landau parameters lead to larger values for W_{ee} . The values of the spin susceptibility, Z and the quasiparticle mass implied by our model together with the Hedin and the RPA Landau parameters are also shown in Table- 2. The Hedin Landau parameters are generally more consistent with experiment especially for the alkali metals Na, K and Rb for which our model is most appropriate calculations of Z using the density functional formulism, which includes crystalline influences more accurately than our simple model, also strongly support Landau parameters similar to those calculated by Hedin [Wilk et al. (1979)[35]]. Although Hedin’s calculation does include approximately a class of diagrams in the perturbation theory for the self-energy additional to those included in the RPA it cannot be rigorously justified at alkali-metal densities. We do not see any fundamental reason for the Hedin calculation to be superior to others and the apparent superiority of its numerical predictions may be largely fortuitous.

IV. Conclusions

Comparison of theory and experiment for the contribution arising from electron-electron scattering to the thermal resistivity of alkali metals, W_{ee} shows that the simple approximation used for the quasiparticle scattering function is adequate. The electron – electron scattering contribution to the electrical resistivity ρ_{ee} is estimated only approximately because of the crude formula used for the fractional Umklapp scattering Δ . (We believe that the values of Δ in Table-1 are too high). An accurate calculation of Δ is certainly feasible, although it would require mostly numerical work. We believe that when combined with the approximate scattering function presented here and the theory given by Lawrence and Wilkins (1973)[24] such a value of Δ would produce very reliable estimates of ρ_{ee} . This is especially desirable in understanding the low temperature electrical resistivity of alkali metals because of the great difficulty of isolating ρ_{ee} experimentally. In conclusion we believe that in view of the great difficulty of developing a first principles microscopic theory for the present approach is a useful alternative for predicting the influence of electron-electron scattering on the transport properties of alkali metals. In particular by using an exact expression for the forward-scattering limit it favours exactly a Cancellation between contributions to Γ which is very important in low-density systems and which may be difficult to retain when making approximations to the very complicated perturbation theory expression for Γ .

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