

# Non-Universality of the Kadowaki-Woods Ratio in Layered Materials

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# Declaration

This thesis is an account of research undertaken between February and November of 2006 in the Department of Physics, School of Physical Sciences, Faculty of Engineering and Physical Sciences, University of Queensland, Brisbane, Australia under the supervision of Dr Ben Powell and Dr John Fjaerestad.

Except where acknowledged otherwise, the material presented here is, to the best of my knowledge, original and has not been submitted in whole or part for a degree in any university.

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Anthony Jacko  
November, 2006

# Acknowledgements

I would like to thank my supervisors Ben Powell and John Fjaerestad for their help and guidance throughout the year. Their patience and guidance were invaluable. I would also like to thank the Condensed Matter Group, and Ross McKenzie in particular, for their helpful comments on both the project and my presentations over the year. Many thanks to my friends and colleagues in Room 301, it's been a long and stressful year, and it was good to have other people going through the same thing. Finally, thanks to Mr Beans and Nestlé Condensed Coffee and Milk, I couldn't have done it without you.

# Abstract

The Kadowaki-Woods ratio is a ratio involving the electronic contributions to resistivity and heat capacity, and is seen experimentally to be universal within some classes of materials. We first derive the Kadowaki-Woods ratio for a weakly correlated system with a screened Coulomb interaction. We then use a phenomenological Fermi liquid theory and the analytical properties of the self energy to derive the Kadowaki-Woods ratio.

We show that the Kadowaki-Woods ratio is not universal. We also show that it is not renormalised by many-body interactions. In this way we can understand the apparent ‘universal’ value of the Kadowaki-Woods ratio for heavy fermion systems, and the different universal value for transition metals, as well as its non-universality in layered materials such as organic charge transfer salts and some transition metal oxides. This implies that the different values of the Kadowaki-Woods ratio are due to the non-interacting properties rather than to renormalisations.

“Whatever you do will be insignificant, but it is very important that you do it.”

- Mohandas Gandhi

# List of Frequently Used Symbols

- $A$  : Coefficient of the quadratic (in temperature) part of the resistivity.  
Due to electron-electron interactions.
- $A(\varepsilon, k)$  : The spectral density as a function of energy  $\varepsilon$  and wavevector  $k$ .
- $B$  : A dimensionless constant in the scattering rate.
- $C$  : Coefficient of the  $T^5$  part of the resistivity.  
Due to electron-phonon interactions.
- $D(\varepsilon)$  : Density of states at an energy  $\varepsilon$ .
- $D^*(\varepsilon)$  : Renormalised density of states at an energy  $\varepsilon$ .
- $f$  : Fermi distribution function.
- $G_R(\varepsilon, k)$  : The retarded Greens function (propagator), as a function of energy  $\varepsilon$  and wavevector  $k$ .
- $k_F$  : Fermi wavevector, corresponding to the highest occupied state.
- $\bar{k}_F$  : Approximate Fermi wavevector in quasi-2D.
- $k_{TF}$  : Thomas-Fermi wavevector, a characteristic inverse screening length.
- $m^o$  : Unrenormalised electron mass.
- $m^*$  : Effective quasi-particle mass.
- $n$  : Number density of electrons.
- $T_c$  : Superconducting critical temperature.
- $Y$  : Valence of the material (electrons per unit cell).
- $Z$  : Renormalisation factor, given by  $m/m^*$

- $\alpha$  : Cubic coefficient of the resistivity. Due to electron-phonon interactions.
- $\gamma$  : Linear (in temperature) coefficient of the specific heat.  
Due to electron-electron interactions.
- $\varepsilon_F$  : Fermi energy (energy of the highest occupied state at zero temperature).
- $\rho$  : Resistivity
- $\rho_o$  : Residual resistivity (temperature independent part) due to impurities.
- $\Sigma_R(\omega, k)$  : The retarded Self energy of energy  $\omega$  and wavevector  $k$ .
- $\sigma$  : Conductivity, inverse resistivity.
- $\tau$  : Quasiparticle scattering time (mean time between collisions).

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# Chapter 1

## Strongly Correlated Systems

In this chapter we discuss the properties of strongly correlated systems. Strongly correlated systems are systems in which there are large many-body interactions. The physical properties of such systems may be qualitatively different to those of non-interacting systems. In some systems however, they behave like a weakly-interacting system, and this can be understood in terms of Fermi liquid theory

### 1.1 Fermi Liquid Theory

Fermi liquid theory was developed by Landau in 1956 [1] and has been very successful in describing a range of fermionic systems. In this theory, originally developed for weakly interacting liquid  $^3\text{He}$  (a fermionic isotope of Helium), the interactions give rise to new fundamental excitations, called quasi-particles. These quasi-particles are weakly interacting versions of the fundamental excitations of the non-interacting system. Their properties are renormalised versions of the original particles properties. Landau's idea was that if you adiabatically turn on the interaction in a many-body system, the eigenstates should evolve smoothly into eigenstates of the interacting system. This would mean that there is a one-to-one correspondence between the non-interacting states and the new interacting eigenstates. Some properties of the state, such as charge and spin, are unchanged, while others, like the mass or magnetisation, are renormalised by the interactions. Perhaps the most common way to understand these new fundamental excitations is as quasi-particles. The idea of the quasi-particle is that each particle is dragging along (for an attractive interaction) a cloud of other particles, and it is this central particle plus the cloud that make up the quasi-particle. This cloud has the effect of shielding the central particle from interactions with

particles outside the cloud. Thus quasi-particles are in general weakly interacting. For a repulsive interaction like the Coloumb interaction, the same sort of thing occurs, but the cloud surrounding the central particle is a cloud of holes. In this picture it is easy to imagine that the quasi-particles should be heavier than a single non-interacting particle. In Landau Fermi liquid theory, interactions are parameterised in terms of symmetric and antisymmetric Fermi liquid coefficients  $F^s_n$  and  $F^a_n$  respectively [2]. Particular properties of the system are enhanced by particular coefficients, for example, the quasiparticle effective mass  $m^*$  depends on the zeroth order symmetric parameter  $m^* = m^o(1 + F^s_1)$  [2] where  $m^o$  is the non-ineracting mass. This mass renormalisation comes up very often, and has another notation

$$m^* = \frac{m^o}{Z}.$$

## 1.2 Heavy Fermion Systems

Heavy fermion systems are systems in which strong many-body interactions lead to a renormalised electron mass orders of magnitude larger than the bare electron mass. Despite these strong interactions, the Fermi liquid theory quasi-particle picture accurately captures the properties of these materials. This means that the renormalised particles (quasi-particles) are weakly interacting, so many properties are simply renormalised versions of the free electron gases properties. For example, they have a heat capacity that is linear in temperature, a resistivity with a quadratic temperature dependence, and a magnetic susceptibility that is inversely proportional to temperature for high temperatures, and constant at low temperatures. While many different systems are classified as heavy fermions, they have some chemistry in common, as well as a large effective mass. They tend to have two sources of electrons, one a partially filled conduction band, and the other a source of  $f$ - or  $d$ -electrons. These electrons are local magnetic moments, and there is an antiferromagnetic interaction between them and the conduction band [3].

The Kondo effect is a minimum of resistivity at a finite temperature. It is caused by scattering from magnetic impurities [4]. In scattering events involving a magnetic impurity, the electron interacts with the magnetic impurity antiferromagnetically, and they can flip spins [5]. Kondo showed that this interaction grows logarithmically as  $T \rightarrow 0$ . By considering the renormalisation group [6] it can be shown that at very low temperatures the large interaction will actually cause electrons to become bound to the magnetic impurities. These bound pairs act like non-magnetic scattering potentials.

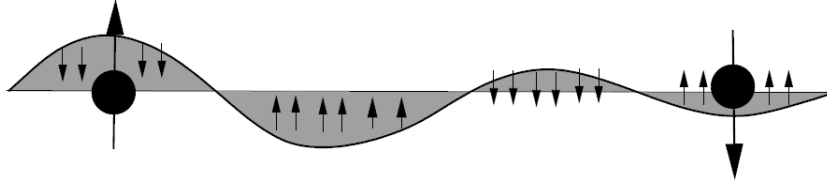


Figure 1.1: The RKKY interaction between magnetic moments (large circles) is mediated by the antiferromagnetic interaction with the conduction electrons (small arrows). Figure from Coleman [3].

Systems exhibiting the Kondo effect have a characteristic temperature, the Kondo temperature  $T_K$ , below which the resistivity starts to increase logarithmically, and then leads to antiferromagnetically bound states.

There is a competition between the usual Kondo effect and the mutual interaction of the magnetic moments, known as the RKKY interaction<sup>1</sup>. The RKKY interaction is an antiferromagnetic interaction mediated by the conduction electrons, as illustrated in Fig. 1.1.

If the Kondo energy is larger than the energy of the mutual interaction then the system will be a Fermi liquid, as in the heavy fermions, while if the mutual interaction of magnetic moments dominates, then the system will be an antiferromagnetic insulator [3]. In this way we can understand how these systems can be both strongly renormalised, as seen in their large effective masses, and still act like Fermi liquids.

### 1.3 Correlated Oxides

Correlated oxides are oxides that exhibit effects of strong correlations. The high- $T_c$  superconductors are correlated oxides, for example  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , where  $x$  measures the doping by strontium. Copper oxide layers are common to all high- $T_c$  superconductors. These are known as cuprates, and they require some doping to become superconductors. The undoped materials are antiferromagnetic insulators, despite having a partially filled band [7]. This is due to the strong interactions, and is known as the Mott insulator phase. Mott insulators occur when the interactions split the bands around the Fermi energy, resulting in a non-zero excitation energy. These correlated oxide materials become Fermi liquids as doping increases, and in between the Fermi liquid region and the antiferromagnetic insulator region they can

<sup>1</sup>Named for Ruderman, Kittel, Kasuya and Yosida.

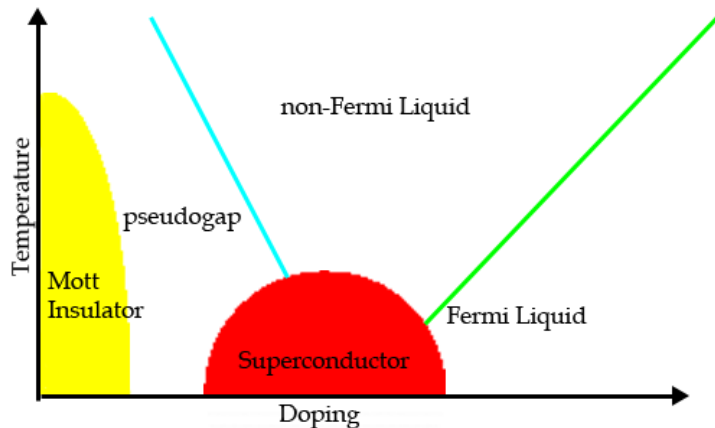


Figure 1.2: General phase diagram for cuprates in temperature and hole doping, showing the Mott insulator phase at low doping, the Fermi liquid at high doping, and a superconducting region in between. The pseudo-gap region is thought by some to be crossover region between the Kondo interaction and the RKKY interaction [3]. Based on a figure in [9]. This phase diagram is very similar to that of the organic charge transfer salts, with doping replaced by pressure.

superconduct [7]. This can be seen in the schematic phase diagram in Fig. 1.2. The non-Fermi liquid phase and pseudo-gap region between the Mott insulator and Fermi liquid phases is thought by some to be a crossover region where the Kondo interaction and RKKY interaction are of the same order of magnitude [3]. The most important point about this phase diagram is its similarity to that of the organic charge transfer salts, with pressure replacing doping on the horizontal axis [8].

## 1.4 Organic Charge Transfer Salts

Organic charge transfer salts are another class of systems with strong electron correlations. Like the cuprates, these systems tend to have reduced dimensions, i.e. coupled layers or chains. As in the cuprates, the band structure suggests that they should be metals but they are found to be antiferromagnetic Mott insulators. In fact, their phase diagrams have many common features, including a superconducting region and a Fermi liquid region, as well as the antiferromagnetic Mott insulator phase [8]. An important difference is that while in the cuprates the axes are doping and temperature, in the organics the axes are pressure and temperature. The similarities imply that increasing the pressure on the organic charge transfer salts has the same ef-

fect as doping, allowing lower energy excitations. While the superconducting critical temperatures of the organics (around  $10K$ ) may not be considered high on an absolute scale, the ratio of the Fermi temperature and the critical temperature is  $T_F/T_c \approx 100$  in both cuprates and organics.

## 1.5 The Wilson Ratio

Finding universal ratios in physics is an important probe of our understanding. To motivate our study of the Kadowaki-Woods ratio, we first examine another ratio which has been useful in developing our understanding of condensed matter systems. The Wilson ratio is the ratio of the Fermi-Liquid enhancements to the magnetic susceptibility at zero temperature  $\chi(0)$  and the linear specific heat coefficient  $\gamma$ , and was predicted by Wilson as a consequence of his Nobel prize winning work on the renormalisation group[6]. The ratio is defined as

$$R_W \equiv \frac{\chi(0)}{\chi_0} \frac{\gamma_0}{\gamma}$$

where  $\chi_0$  and  $\gamma_0$  are the non-interacting values for the magnetic susceptibility and specific heat, while  $\chi$  and  $\gamma$  are the Fermi liquid theory values. The low temperature expression for  $\chi$  is found to be [10]

$$\frac{\chi(0)}{\chi_0} = \frac{1 + F_1^s}{1 + F_0^a}$$

where  $F_0^a$  is the zeroth order anti-symmetric Landau Fermi liquid parameter. We also know that

$$\gamma = \frac{m^*}{m} \gamma_0$$

so the Wilson ratio is [11]

$$R_W = \frac{1}{1 + F_0^a}$$

showing dependence on only one of the Landau Fermi liquid parameters.  $F_0^a$  has a value of 1 away from ferromagnetic phases, and tends towards  $-1$  in the ferromagnetic phase [12], so the Wilson ratio diverges as the system approaches a ferromagnetic phases. Thus the non-universality of the Wilson ratio can be used as indication of proximity to a ferromagnetic phase.

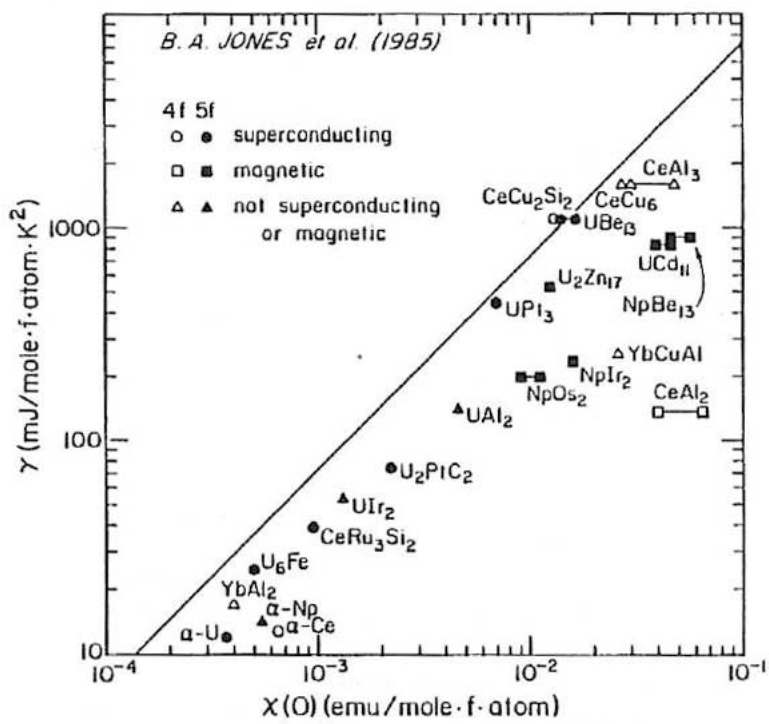


Figure 1.3: The Wilson ratio for many heavy fermion materials, taken from Jones *et al.* (1985).

# Chapter 2

## The Kadowaki-Woods Ratio

In this chapter we describe the Kadowaki-Woods ratio and its apparent universality in heavy fermion systems and transition metal compounds. We then discuss some early theories trying to explain why it is universal, as well as some more recent work considering dimensions, both of the quantity and of the material.

The Wiedemann-Franz law and Wilson ratio are two well understood ratios, but interesting new insights come from investigating the ratios that are not so well understood. The Kadowaki-Woods ratio, the ratio of electronic contributions to resistivity and specific heat, fits this description. The universality of the ratio of the electronic contribution to the temperature dependence of the resistivity,  $A$ ,

$$\rho = \rho_0 + AT^2 + CT^5 \tag{2.1}$$

and the square of the specific heat coefficient  $\gamma$ ,

$$c_V = \gamma T + \alpha T^3 \tag{2.2}$$

has been observed in several classes of metals. The  $T^5$  term  $C$  in  $\rho$  and the  $T^3$  term  $\alpha$  in  $c_V$  are phonon contributions (electron-phonon scattering in the case of the resistivity), and  $\rho_0$  is the residual resistivity. The universality of  $A/\gamma^2$  was first observed in the transition metals [13], and then later in heavy fermions, where it became known as the Kadowaki-Woods ratio [14]. The universality extends over several decades in both the transition metals and the heavy fermions.

## 2.1 Resistivity Due To Electron-Electron Scattering

The resistivity has a  $T^2$  temperature dependence due to electron-electron scattering. The reason for this is as follows. The resistivity (as derived in Section 4.1) is given by

$$\rho = \frac{m^*}{ne^2} \frac{1}{\tau},$$

where  $m^*$  is the effective electron mass,  $n$  is the number density of electrons, and  $\tau$  is the average time between electron-electron scattering events.

To determine the resistivity, the scattering time must be found. At zero temperature, two electron scattering involves an electron above the Fermi energy<sup>1</sup> scattering with one below. The exclusion principle requires that the resultant energies be above the Fermi energy ( $\varepsilon_F$ ). Conservation of energy requires that the resultant energies are below the energy of the higher energy electron ( $\varepsilon_1$ ). The energy difference  $\Delta\varepsilon = \varepsilon_1 - \varepsilon_F$  is proportional to the number of states between  $\varepsilon_1$  and  $\varepsilon_F$ , with some constant of proportionality  $\alpha$ .<sup>2</sup> If we choose one of the initial energies  $\varepsilon_1$  then there are  $\alpha\Delta\varepsilon$  choices for each of the resultant energies, and so  $(\alpha\Delta\varepsilon)^2$  combinations of resultant energies. Once these are picked, there is no choice for the initial energy of the other electron, within the Fermi sea. [15]

At finite temperature, there will be an additional contribution to the scattering rate. The width of the partially occupied region of the Fermi distribution function is proportional to the thermal energy  $k_B T$ . As above, this energy will be proportional to the number of extra electron energy states available to scatter to and from. Thus the thermal contribution to the scattering rate will depend on  $(k_B T)^2$ .

In 1981 Klipstein *et al* rejected the idea of the  $T^2$  resistivity coming from electron-electron scattering in  $\text{TiS}_2$  [16]. They instead proposed that electron-phonon scattering was responsible. According to them, electron-electron scattering cannot explain the observed large pressure dependence of the resistivity, and that there are no 'hole' states above the Fermi wavevector for electron-electron scattering to occur in.

Strack *et al* use this argument in the context of organic superconductors, saying that since these materials apparently do not follow the predicted electron-electron scattering behaviour as seen by their deviation from the

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<sup>1</sup>For the sake of argument let us say that it has been injected into the zero temperature system.

<sup>2</sup>The number of states is  $\alpha\Delta\varepsilon = \int_{\varepsilon_F}^{\varepsilon_1} D_{holes}(\varepsilon)d\varepsilon$  where  $D_{holes}$  is the density of states for holes, which at zero temperature is a constant above the Fermi energy and zero below.

Kadowaki-Woods ratio, then the dominant  $T^2$  mechanism could be electron-phonon [17]. The quasi-particle enhancements will affect electrons and phonons differently, meaning that the contributions to the  $T^2$  component of the resistivity will be scaled differently by the interactions. If the dominant low temperature scattering mechanism is electron-phonon, this could indicate that this is the pairing mechanism responsible for superconductivity.

This model is a modified Sommerfeld model, and as such neglects the periodic structure of the material. Further, we have assumed that electron-electron collisions can change the net current, and hence contribute to the resistivity. Without a lattice, electron-electron scattering conserves momentum and so will not change the net current. In a lattice, electron-electron scattering does not need to conserve crystal momentum. Momenta higher than that corresponding to the lattice constant  $a$ ,  $k_{max} = 2\pi/a$  are mapped onto momentum inside the first Brillouin zone. In this way there can be a change in crystal momentum (Bloch wavevector) and so a contribution to the resistivity. These are known as Umklapp scattering processes [15].

## 2.2 The Kadowaki-Woods Ratio in Transition Metals

It was observed by Rice in 1968 that the ratio now known as the Kadowaki-Woods ratio was universal in the elemental transition metals (metals where the outermost electron shell is a  $d$  shell). Rice proposed that electron-electron scattering as described by Baber [18] explains the universality of  $A/\gamma^2$  [13]. Baber's idea was to find the contribution of electron-electron scattering to the resistivity [18]. By considering a screened Coulomb interaction [19] a finite contribution to the resistance with a  $T^2$  dependence is found. This contribution is dependent on the square of the effective electron mass  $m^*$ , and so was expected to be more pronounced in transition metals than in normal metals. In normal metals the electron-electron scattering contribution is hundreds or thousands of times smaller than  $\rho_0$ , the temperature independent contribution to the resistivity (the residual resistivity), and thus is negligible [15]. At low temperatures its contribution is overwhelmed by  $\rho_0$ , while at higher temperatures the electron-phonon contributions dominate. In transition metals the enhanced electron mass means that this contribution is measurable, and it was found to agree with theory [18]. Rice proposed that if the scattering process is electron-electron scattering,  $A$  should depend on  $\gamma^2$  [13], because as has been noted previously  $\gamma/\gamma_0 = m^*/m$ . The ratio of these was observed in the transition metals to be universal, with an

experimentally determined value of  $(A/\gamma^2)_{TM} \simeq 4 \times 10^{-9} \Omega\text{m mole}^2\text{K}^2\text{J}^{-2}$ .

## 2.3 The Kadowaki-Woods Ratio in Heavy Fermions

In 1986 Kadowaki and Woods published their observation that the ratio  $A/\gamma^2$  is approximately constant in heavy fermion compounds [14]. The previous year, Sato *et al.* noticed that this ratio is universal in ‘dense Kondo systems’, systems like the heavy fermions with many magnetic moments as well as conduction electrons. They went no further than noting its existence and saying that the relationship is expected from Fermi liquid theory [20]. This ratio became known as the Kadowaki-Woods ratio. The term ‘heavy fermion’ refers to the large enhancement of the electron mass, and this is easily noticed in the enhancement of the specific heat. Since the dependence on the effective mass cancels in the Kadowaki-Woods ratio, it may be expected that these systems will follow the same universal behaviour as the transition metals. This implies that the coefficient of the resistivity could be larger than the bare value by six orders of magnitude, since mass enhancement factors of  $m^*/m = 1000$  are found in some heavy fermion systems.

Kadowaki and Woods pointed out that while  $A/\gamma^2$  showed the same sort of ‘universality’, the value of the ratio was about two orders of magnitude larger than for transition metals. Since the Kadowaki-Woods ratio is seen to be universal, and the mass enhancement varies by orders of magnitude, the mass enhancement must cancel in the ratio. In heavy fermion systems  $(A/\gamma^2)_{HF} \simeq 1 \times 10^{-7} \Omega\text{m mole}^2\text{K}^2\text{J}^{-2}$  [14]. This was thought at the time to be an effect of the large interactions present by definition in heavy fermion systems [14]. This explains the universality of the ratio in heavy fermion systems, but does not explain why it takes a different value to that of the transition metals. The universality of the Kadowaki-Woods ratio indicates that at low temperatures, the dominant mechanisms of scattering and thermal excitations are the same.

Possibly the first theoretical investigation of the Kadowaki-Woods ratio (submitted 3 months after the Kadowaki-Woods paper) was by Yamada and Yosida. They based their work on the periodic Anderson Hamiltonian applied to heavy fermions, and found the expected relationships between  $A$  and  $\gamma$  [21]. They concluded that the lattice is essential for a  $T^2$  resistivity. Yamada and Yosida stress that their results are only qualitative, but claim that their methodology could be applied to real systems with minimal changes. Unfortunately they did not address the issue of the difference between the Kadowaki-Woods ratio in heavy fermions and in transition metals.

Miyake, Matsuura and Varma [22] proposed that the disparity between

heavy fermions and transition metals can be explained through the conduction electrons self-energy. For a momentum independent self energy such as in a local Fermi liquid theory, the self energy and the mass enhancement are related by [23]

$$\frac{m^*}{m^o} = 1 - \lim_{\varepsilon \rightarrow 0} \operatorname{Re} \left( \frac{\partial \Sigma}{\partial \varepsilon} \right)$$

where  $\Sigma$  is the self energy and  $\varepsilon$  is the energy. In quantum field theory the self energy of an electron represents the effect of the electron interacting with its environment on its energy and lifetime [23]. They showed that while the real part of the derivative determines  $\gamma$  as seen above, the imaginary part of the self energy determines the resistivity coefficient  $A \propto \operatorname{Im}(\Sigma)$ . The idea Miyake *et al.* presented is that differences in the energy dependencies of  $\Sigma$  lead to the observed differences between transition metals and heavy fermions. This is because while the mass enhancements due to the energy dependence of the self energy cancel in the Kadowaki-Woods ratio, there is a deeper relationship between  $A$  and  $\gamma$  [10]. The real and imaginary parts of the self energy can be related by the Kramers-Kronig relations, and thus have a more fundamental relationship rather than just being linked by their renormalisation. The details of this are discussed in Chapter 6.

Miyake *et al.* explain the differences in observed Kadowaki-Woods ratios by saying it is not universal in general. They claim that in the limit that the frequency dependence is small, the Kadowaki-Woods ratio takes the value found in transition metals, while in the limit of a large frequency dependence, it takes the value found in the heavy fermion systems, and will lie inbetween these values for intermediate interactions [22]. Fig. 2.1 shows the Kadowaki-Woods ratio for heavy fermions and transition metals. They further note that a large residual resistivity can change the value of  $A/\gamma^2$ , shifting it from the universal value, and that intermediate values for the Kadowaki-Woods ratio are possible for intermediate frequency dependencies. Li *et al.* [24] noticed the very strongly renormalised conductivity and hence Kadowaki-Woods ratio in  $\text{Na}_{0.7}\text{CoO}_2$ , another layered correlated oxide. They suggest that this "unprecedented magnitude of electron-electron scattering" is due to either magnetic frustration, where the ground state is degenerate, or the proximity of a quantum critical point.

While this might explain why the transition metals and heavy fermions have different values, it does not explain the deviation of the organic superconductors, whose values of  $R_{KW}$  tend to be orders of magnitude above that of the heavy fermions, whose value is in turn greater than that of the transition metals. The results presented in Chapter 6 provide another interpretation of the observed non-universality of the Kadowaki-Woods ratio.

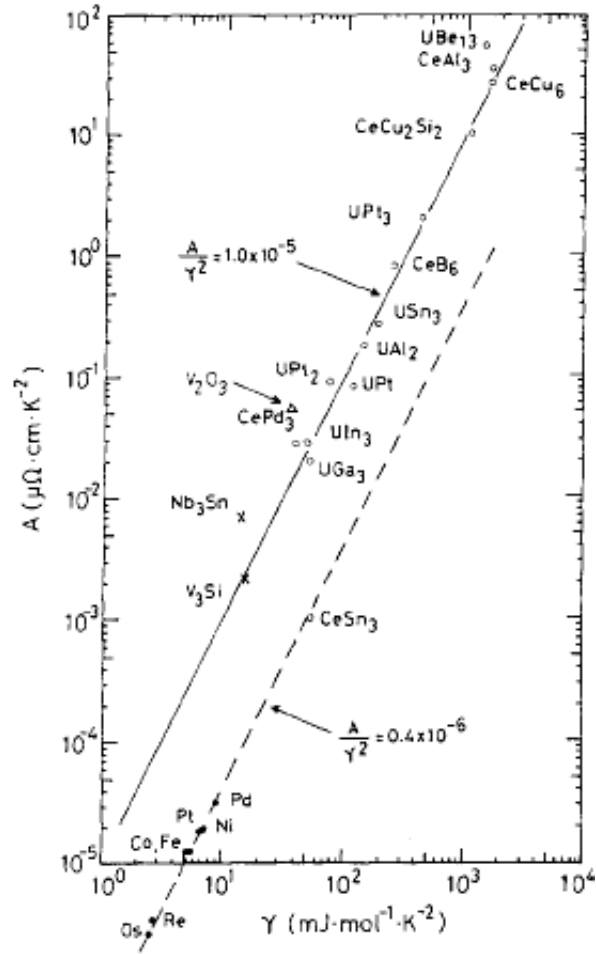


Figure 2.1: The Kadowaki-Woods ratio for heavy fermions (upper line) and transition metals (lower line), from Miyake *et al.* [22]. They explain the two ‘universal’ values as limiting cases of the interaction strength.

The derived Kadowaki-Woods ratio contains no renormalised quantities, but does contain non-interacting properties of the system.

## 2.4 Rescaling the Kadowaki-Woods Ratio in Correlated Oxides

A more recent attempt to understand the deviation of many materials from the two ‘universal’ Kadowaki-Woods ratios came from Hussey. He pointed out that the traditional way of calculating the ratio was comparing a resistivity in  $\Omega m$  with a heat capacity in  $J/mol$  [25]. His idea was to rescale the coefficient of the heat capacity to put it into S.I. units before trying to compare it with the coefficient of the resistivity. His idea was to convert  $\gamma$  to a volume quantity via Avogadro’s number, the unit cell volume, and the number of charge carriers per unit cell. It is an interesting coincidence that many of the heavy fermions have similar unit cell volumes, so this was not an issue earlier [25]. Hussey tried this method on a selection of correlated oxides, and found that this rescaling helped. He made further corrections by including the effects of multiple Fermi surfaces and carrier density, and the results of this rescaling can be seen in Fig. 2.2. Note that layered materials like strontium ruthenate ( $Sr_2RuO_4$ ) do not fit on the rescaled ‘universal’ Kadowaki-Woods ratio line. As will be seen in Chapter 4, this rescaling is also not sufficient to explain the deviation of the organic charge transfer salts from the ‘universal’ values, which is not surprising as they are also layered.

Another recent attempt at explaining the non-universality of the Kadowaki-Woods ratio was presented recently by Tsujii *et al.* [26]. While this result may explain some materials deviation from the heavy fermion value of the Kadowaki-Woods ratio, it results in a KWR lower than the heavy fermion value and so is not adequate in describing the deviation of the KWR in layered strongly correlated systems.

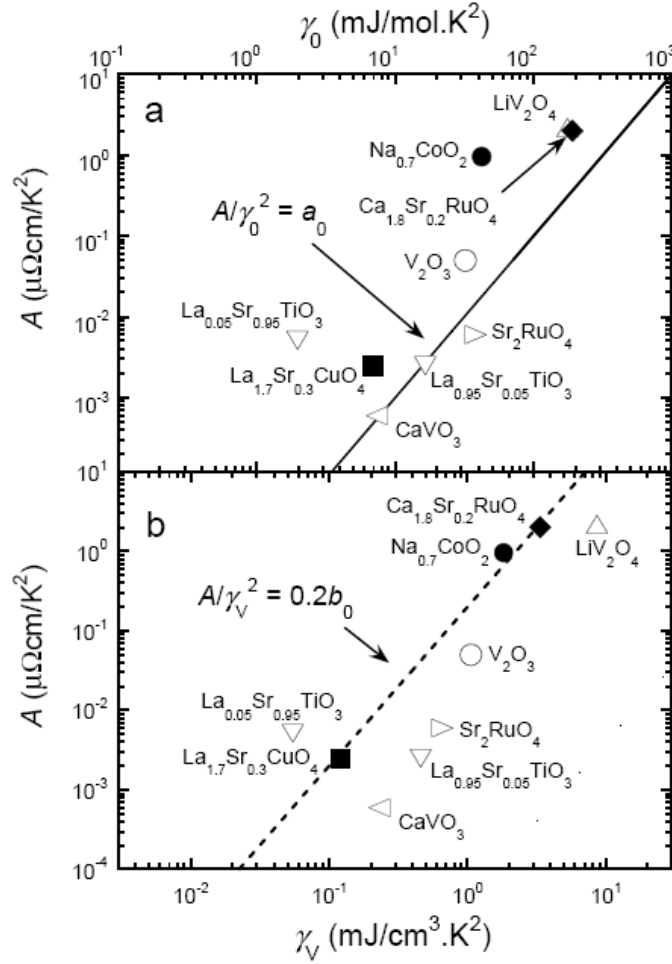


Figure 2.2: The Kadowaki-Woods ratio for a variety of correlated oxides, from Hussey [25]. The upper plot is the KWR in the traditional units, while in the lower plot all units have been converted to SI, and the solid lines are the respective values of the KWR in heavy fermions. Note that layered materials like strontium ruthenate ( $\text{Sr}_2\text{RuO}_4$ ) do not lie on the rescaled ‘universal’ Kadowaki-Woods ratio line.

# Chapter 3

## The Kadowaki-Woods Ratio in Organic Superconductors

In this chapter we discuss the Kadowaki-Woods ratio in organic charge transfer salts and other layered materials. The Kadowaki-Woods ratios of organic charge transfer salts do not seem to have any trend, varying by orders of magnitude, and all of them above the value for heavy fermions. This has been taken by some as a sign that electron-electron scattering is not the dominant scattering mechanism at low temperatures. Weger [27] stated that in quasi-2D materials, electron-phonon interactions could give rise to a  $T^2$  contribution to the resistivity. This contribution would not be effected by interactions in the same way as the electron-electron contribution and so there is no reason for the Kadowaki-Woods ratio to be a constant in these systems.

### 3.1 Rescaling The Kadowaki-Woods Ratio in Organic Superconductors

A striking thing about the Kadowaki-Woods ratio is its dimensions. It has units of  $\frac{K^2 \text{ mol}^2 \text{ s}}{\text{kg A}^2 \text{ m}}$ , comparing a molar value  $[\gamma] = \frac{J}{\text{mol K}^2}$  to a volume quantity  $[A] = \frac{\text{kg m}^3}{\text{s}^3 \text{ A}^2 \text{ K}^2}$ .<sup>1</sup> Hussey noted this in examining correlated oxides [25]. His entirely sensible idea was to convert  $\gamma$  to a volume quantity via Avogadro's number, the unit cell volume, and the number of molecules per unit cell. It is an interesting coincidence that many of the heavy fermions have similar unit cell volumes, so this was not an issue earlier [25]. This rescaling of  $\gamma$  succeeded in fitting some correlated oxides to the heavy-fermion value of the Kadowaki-Woods ratio, as seen in Fig. 2.2. In the organics however there

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<sup>1</sup>The notation  $[x]$  denotes the units of  $x$ .

is a wide range of unit cell volumes, so this effect could explain their broad range of values for  $R_{KW}$ . Fig. 3.1 shows the unscaled Kadowaki-Woods plot ( $\log A$  vs  $\log \gamma^2$ ) for a selection organic superconductors.

None of the ratios are particularly close to the predicted heavy fermion value. Fig. 3.2 shows the rescaled Kadowaki-Woods plot for the same materials. The correlated oxides and  $\text{Rb}_3\text{C}_{60}$  (the only relatively three dimensional material of the organics included) are close to the predicted value. It turns out that for the selection of materials given here, the ratio of the unit cell volume to units per unit cell is approximately constant. This is why the relative positions are basically unchanged (on the log-log scale) while the bulk moves along the  $\gamma$  axis. The other organics, which are all either quasi-2D or quasi-1D, are moved further away from the predicted value. This could indicate that the dimensionality of the systems is significant. It also makes the hypothesis of Miyake, Matsuura and Varma seem less likely, as they would expect all Kadowaki-Woods ratios to lie somewhere below the heavy fermion ratio. Stracks' prediction of electron-phonon interactions giving a  $T^2$  resistivity could still be true.

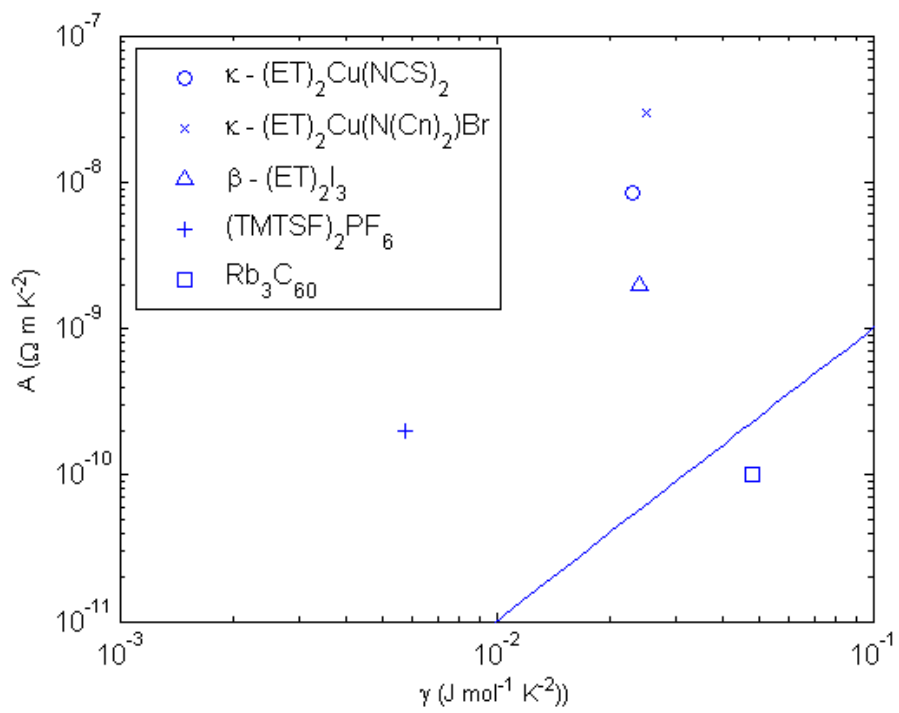


Figure 3.1: The Kadowaki-Woods plot for a selection of organic compounds in the 'usual' per mole units. The solid line is the 'universal' Kadowaki-Woods ratio in heavy fermions, with a value of  $R_{KW} \simeq 10^{-7} \Omega \text{ m mol}^2 \text{ K}^2 \text{ J}^{-2}$ .

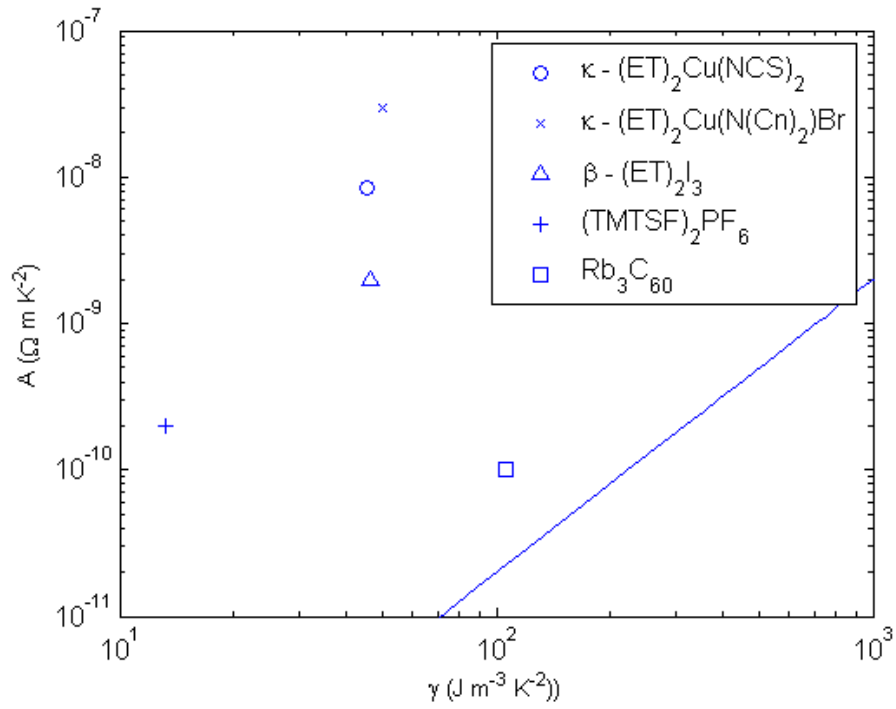


Figure 3.2: The Kadowaki-Woods plot for a selection of organic compounds in SI units, converted via unit cell volume and formula units per unit cell. The relative positions of the materials are the same as in Fig. 3.1 as for the selection of materials here the ratio of the unit cell volume to formula units per unit cell is approximately constant. The solid line corresponds to the value of the Kadowaki-Woods ratio in heavy fermions in SI units, with a value of  $R_{KW} \simeq 2 \times 10^{-15} \text{ K}^2 \text{ m}^5 \text{ s}^3 \text{ C}^{-2} \text{ kg}^{-1}$ .

# Chapter 4

## The Kadowaki-Woods Ratio in Weakly Correlated Metals

“Il est facile de voir que...”

- Pierre-Simon Laplace

In this chapter we use a simple model (appropriate for a weakly interacting metal) we calculate the Kadowaki-Woods Ratio in both three- and quasi-two-dimensional systems. The model is that of a free electron gas with interacting via a screened Coloumb potential (Thomas-Fermi screening).

### 4.1 Three Dimensional Materials

Ohms law for a three dimensional simple metal is derived and from this an expression for the Kadowaki-Woods ratio is found.

#### Conductivity in Three Dimensions

The first step to calculating the Kadowaki-Woods ratio is the derivation of the conductivity. To derive the conductivity we imagine applying an electric field to a metal in the  $x$  direction. This produces net current in that direction, which can be found by summing all the  $x$  components of the velocities in the first Brillouin zone, weighted by the Fermi distribution function  $f(k)$  and doubled to account for spin. The current density  $j$  is thus,

$$j = \frac{-2e}{(2\pi)^3} \int_{FBZ} d\mathbf{k} v_x(k) f(k), \quad (4.1)$$

where  $e$  is the magnitude of the electronic charge and  $v_x(k)$  the velocity in the  $x$  direction. In the relaxation time approximation<sup>1</sup>, the Fermi distribution function is approximated as

$$f(k) \simeq f_o(k) + \frac{e}{\hbar} \tau(k) \vec{E} \cdot \nabla_k f_o(k) \quad (4.2)$$

at low temperatures, where  $f_o(k)$  is the zero temperature distribution function,  $\tau(k)$  the scattering time and  $\vec{E}$  the applied electric field. This is simply the application of the Boltzmann transport equation [15].

Eq. (4.2) is inserted into Eq. (4.1) giving

$$j = \frac{-2e}{(2\pi)^3} \int d\mathbf{k} v_x(k) \left( f_o(k) + E_x \frac{e\tau(k)}{\hbar} \frac{\partial f_o}{\partial k_x} \right) \quad (4.3)$$

where we have taken  $\vec{E} = E_x \hat{x}$ . The first term in this expression vanishes<sup>2</sup> leaving:

$$j = \frac{-2e^2}{(2\pi)^3} \int d\mathbf{k} v_x(k) E_x \frac{\tau(k)}{\hbar} \frac{\partial f_o}{\partial k_x} \quad (4.4)$$

Now, using the chain rule we find the derivative of the distribution function with respect to the energy  $\varepsilon$

$$\frac{\partial f_o}{\partial k_x} = \frac{\partial f_o}{\partial \varepsilon} \frac{\partial \varepsilon}{\partial k_x} = \frac{\partial f_o}{\partial \varepsilon} \hbar v_x(k) \quad (4.5)$$

so we can write the current as

$$j = \frac{-2e^2}{(2\pi)^3} \int d\mathbf{k} v_x^2(k) E_x \tau(k) \frac{\partial f_o}{\partial \varepsilon} \quad (4.6)$$

and use the fact that the distribution  $f_o$  is approximately a step function at the Fermi energy  $\varepsilon_F$  near zero temperature ( $T \ll T_F$ ) so  $\frac{\partial f_o}{\partial \varepsilon} \simeq -\delta(\varepsilon - \varepsilon_F)$ . Further, we can expand the integral over  $\mathbf{k}$  into a surface integral over constant energy surfaces and an integral over  $\mathbf{k}$  perpendicular to those surfaces.

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<sup>1</sup>The relaxation time approximation is the approximation that the result of a collision does not depend on the nonequilibrium distribution function, and that collisions in equilibrium do not alter the equilibrium [15].

<sup>2</sup>Physically, we expect no current in the ground state. The Fermi distribution is symmetric ( $f_o(k) = f_o(-k)$ ), while the velocity ( $v_x(k) = -v_x(-k)$ ) is not, so the first term is anti-symmetric.

$$d\mathbf{k} = dS_\varepsilon dk_\perp = dS_\varepsilon \frac{d\varepsilon}{|\nabla_k \varepsilon|} = dS_\varepsilon \frac{d\varepsilon}{\hbar v(k)} \quad (4.7)$$

Now we can collapse the energy integral leaving only the surface integral

$$j = \frac{2e^2}{(2\pi)^3} \int dS_\varepsilon \frac{d\varepsilon}{\hbar v(k)} v_x^2(k) E_x \tau(k) \delta(\varepsilon - \varepsilon_F) \quad (4.8)$$

$$= \frac{2e^2 E_x}{\hbar (2\pi)^3} \int_{\varepsilon=\varepsilon_F} dS_\varepsilon \frac{v_x^2(k)}{v(k)} \tau(k) \quad (4.9)$$

where the wavevector  $k$  is itself a function of the energy  $k = k(\varepsilon)$ . We approximate the integrand as constant over the Fermi surface, so we can take its average value outside the integral using

$$\left\langle \frac{v_x^2(k)}{v(k)} \tau(k) \right\rangle_{\varepsilon_F} = \frac{v(k_F) \tau(k_F)}{3} \quad (4.10)$$

for Fermi surface geometries close to spheres. Using  $v(k_F) = \frac{\hbar k_F}{m^*}$ ,  $k_F^3 = 3\pi^2 n$  and  $\int_{\varepsilon=\varepsilon_F} dS_\varepsilon = 4\pi k_F^2$ , where  $m^*$  is the effective electron mass,  $k_F$  the Fermi wavevector and  $n$  the electron density, we find

$$j = E_x \frac{e^2 \tau(\varepsilon_F) n}{m^*} \quad (4.11)$$

arriving at Ohms law (and rewriting  $\tau(\varepsilon_F)$  as  $\tau$ ),

$$\sigma = \frac{j}{E} = \frac{ne^2 \tau(\varepsilon_F)}{m^*} \quad (4.12)$$

$$E = \rho j = \frac{m^*}{ne^2 \tau} j. \quad (4.13)$$

## Kadowaki-Woods Ratio in Three Dimensions

The Kadowaki-Woods Ratio is the ratio of the electronic contributions to resistivity  $A$  and heat capacity  $\gamma$ .

$$R_{KW} \equiv A/\gamma^2$$

The resistivity, as shown in Eq. (4.13), is given by

$$\rho = \frac{m^*}{ne^2 \tau}$$

so it is clear that the interesting temperature dependence that we are looking for will arise in  $\tau$

As discussed earlier, the electron electron scattering rate  $\frac{1}{\tau}$  should have the following form [15, 18, 28]:

$$\frac{1}{\tau} = f^2(\varepsilon_1 - \varepsilon_F)^2 + B \frac{(k_B T)^2}{\hbar \varepsilon_F} \quad (4.14)$$

Dimensional analysis leads to the coefficient  $\frac{1}{\hbar \varepsilon_F}$  of the temperature dependent term, as this is the only combination of the temperature independent quantities characteristic of an electron gas ( $k_F, m, \hbar$ ) that leave this expression with overall dimensions of inverse time. The coefficient  $B$  is a dimensionless number whose exact form will be determined later.

Eq. (4.14) can be written as

$$\frac{1}{\tau} = \frac{1}{\tau_1} + \frac{1}{\tau_2(T)} \quad (4.15)$$

where  $\frac{1}{\tau_1}$  is the zero temperature contribution and  $\frac{1}{\tau_2(T)}$  is the finite temperature contribution.

Substituting the energy dependent expression into the above expression for resistivity we find:

$$\begin{aligned} \rho &= \frac{m^*}{ne^2} \left( \frac{1}{\tau_1} + \frac{1}{\tau_2(T)} \right) \\ &= \frac{f^2 m^* (\varepsilon_1 - \varepsilon_F)^2}{ne^2} + \frac{B m^* (k_B T)^2}{ne^2 \hbar \varepsilon_F} \\ \rho &= \rho(\varepsilon) + \frac{2B m^{*2} k_B^2}{\hbar^3 k_F^2 e^2 n} T^2. \end{aligned} \quad (4.16)$$

Hence

$$A = \frac{2B m^{*2} k_B^2}{\hbar^3 k_F^2 e^2 n}. \quad (4.17)$$

We note that this result may seem dubious in that we want a temperature dependent resistivity yet have used the  $T \rightarrow 0$  limit of the distribution function in obtaining this expression. Really we have taken the limit  $T/T_F \rightarrow 0$ , which is certainly true in the region that the  $T^2$  resistivity is dominant. Nevertheless, there is a more rigorous derivation of the resistivity is in Chapter 6.

Next we look at the heat capacity's temperature dependence.

$$C_V = \gamma T + \alpha T^3 \quad (4.18)$$

The linear term is the electron-electron contribution and so  $\gamma$  is the term we are interested in. This term is scaled from the non-interacting term by a ratio of the effective mass to the free electron mass [15].

$$\gamma = \gamma_0 \frac{m^*}{m} = \frac{n\pi^2 k_B^2 m^*}{\hbar^2 k_F^2} \quad (4.19)$$

This equation for  $\gamma_0$  comes from the free electron gas expression found in the Sommerfeld model. Taking the ratio  $\frac{A}{\gamma^2}$  from Eqs (4.17) & (4.19) we find

$$\frac{A}{\gamma^2} = \frac{2\hbar}{\pi^4 e^2 k_B^2} \frac{B k_F^2}{n^3} \quad (4.20)$$

which is independent of the effective electron mass, but still dependent on the Fermi wavevector, the electron density, and the dimensionless quantity  $B$ .

Now all that remains is to find the coefficient  $B$  in Eq. (4.20). This is found by considering scattering due to a screened potential, in this case Thomas-Fermi screening.

Thomas-Fermi screening is one of many theories for describing how the Coloumb interaction in a many-body system can be screened. It is a semi-classical theory assuming long wavelength fluctuations to the total potential [15]. By considering the charge density induced by a charged test particle on an otherwise uniform background of charges, it is straightforward to find [15] that the total (screened) potential  $\phi(r)$  caused by the perturbing potential is

$$\phi(r) = \frac{q}{r} e^{-4\pi e^2 \frac{\partial n_o}{\partial \mu} r} \equiv \frac{q}{r} e^{-k_{TF} r} \quad (4.21)$$

where  $q$  is the charge of the test particle,  $n_o$  is the unperturbed charge density,  $\mu$  is the chemical potential and  $k_{TF}$  is the Thomas-Fermi wavevector. It is clear that the Thomas-Fermi wavevector is the inverse of a screening length.

According to Pines, the electron-electron scattering rate is given by [28]

$$\frac{1}{\tau} = v_f^o n \sigma_{\text{eff}} = v_f^o n \frac{4\pi}{k_{TF}^2} \left( \frac{k_B T}{\varepsilon_F} \right)^2 \quad (4.22)$$

where  $\sigma_{\text{eff}}$  is the effective scattering cross section,  $k_{TF}$  is the Thomas-Fermi wavevector and  $v_f^o$  indicates the non-interacting value of the Fermi velocity. The scattering cross section is found by considering screened (Thomas-Fermi), isotropic (s-wave) scattering. For  $T \ll T_f$  the Thomas-Fermi wavevector is given by [15]

$$k_{TF}^2 = \frac{m^o e^2}{\epsilon_o \hbar^2 \pi^2} k_F \quad (4.23)$$

where  $\epsilon_o$  is the permittivity of free space and  $m^o$  is the unrenormalised electron mass. In general, for  $T \ll T_F$  we have [15]

$$k_{TF}^2 = \frac{e^2}{\epsilon_o} D_o(\epsilon_F) \quad (4.24)$$

where  $D_o(\epsilon_F)$  is the unrenormalised density of states at the unrenormalised Fermi energy. Putting this into the form of Eq. (4.14), we have

$$\frac{1}{\tau} = \frac{4\pi v_F^o n \hbar (k_B T)^2}{k_{TF}^2 \epsilon_F^o \hbar \epsilon_F} \quad (4.25)$$

and it is clear that the first part is the coefficient  $B$ .

$$\begin{aligned} B &= \frac{4\pi v_F^o n \hbar}{k_{TF}^2 \epsilon_F^o} = \frac{8\pi n}{k_{TF}^2 k_F} \\ &= \frac{8\pi \hbar^2 \epsilon_o}{3m^o e^2} k_F \end{aligned} \quad (4.26)$$

Where we have used  $n = k_F^3/3\pi^2$  and  $\epsilon_F^o/v_F^o = \hbar k_F/2$ . The expression for  $B$  in Eq. (4.26) can now be inserted into Eq. (4.20), giving a final expression for the Kadowaki-Woods ratio in terms of universal constants, the effective electron mass and the inverse of the Fermi wavevector to the sixth power (Eq. (4.27)).

$$\begin{aligned} \frac{A}{\gamma^2} &= \frac{8\pi \hbar^2 \epsilon_o k_F^o}{3m^o e^2} \frac{2\hbar}{\pi^4 e^2 k_B^2} \frac{k_F^2}{n^3} \\ &= \frac{18\hbar^3 \epsilon_o}{m^o e^4 k_B^2} \frac{1}{k_F^6} \end{aligned} \quad (4.27)$$

This expression is clearly not universal, with a dependence on the Fermi wavevector  $k_F^6$ , despite the observation that it is universal in some classes of materials, namely transition metals and heavy fermion systems. It is reassuring to see that the Kadowaki-Woods ratio as derived for a simple metal does not depend on the effective mass, as we expected from initial considerations. In fact, this expression contains no renormalised quantities.

## 4.2 Quasi-Two Dimensional Materials

Most organic superconductors are not three dimensional. Many are instead quasi-two dimensional, having planes that the electrons can flow relatively

freely in and hop between (tight binding model). The nature of this hopping (coherent or incoherent) is unimportant for this discussion as either leads to the same expression for conductivity [29].

The electron dispersion relation in a quasi-two-dimensional material is

$$\varepsilon = \frac{\hbar^2}{2m^*}(k_x^2 + k_y^2) - 2t_c \cos ck_z \quad (4.28)$$

so the velocity is

$$v(\mathbf{k}) = \frac{1}{\hbar} \nabla_{\mathbf{k}} \varepsilon = \left( \frac{\hbar k_x}{m^*}, \frac{\hbar k_y}{m^*}, \frac{2ct_c}{\hbar} \sin ck_z \right). \quad (4.29)$$

where  $c$  is the interlayer spacing and  $t_c$  is the hopping integral. In this model, there are two directions for which we would like to know the conductivity. One is when the current is measured in the direction of the tight binding (perpendicular to the planes of the free electron gas), and the other is in the plane of the free electron gas.

## Conductivity in Quasi - Two Dimensions: Perpendicular to Planes

As in the three dimensional case we begin with the equation for the current, which can be found by summing all the velocities in the first Brillouin zone, weighted by the Fermi distribution function. The electric field is being applied in the  $z$  direction, which will be referred to as the perpendicular direction from now on, as it is perpendicular to the planes of the material. The  $y$  and  $x$  directions will be referred to as the parallel directions.

$$j_{\perp} = \frac{-2e}{(2\pi)^3} \int_{FBZ} v_{\perp}(k) g(k) \quad (4.30)$$

$$g(k) \simeq g_o(k) + \frac{e}{\hbar} \tau(k) E \cdot \nabla_{\mathbf{k}} g_o(k) \quad (4.31)$$

$$j_{\perp} = \frac{-2e}{(2\pi)^3} \int d\mathbf{k} v_{\perp}(k) \left( g_o(k) + E_{\perp} \frac{e\tau(k)}{\hbar} \frac{\partial g_o}{\partial k_{\perp}} \right) \quad (4.32)$$

The first term goes to zero as before. Using this we write the conductivity as

$$\sigma_{\perp} = \frac{-2e^2}{(2\pi)^3} \int d\mathbf{k} v_{\perp}(k) \frac{\tau(k)}{\hbar} \frac{\partial g_o}{\partial k_{\perp}}. \quad (4.33)$$

Using the chain rule and approximating the Fermi distribution to a step function we find

$$\frac{\partial g_o}{\partial k_\perp} = \frac{\partial g_o}{\partial \varepsilon} \frac{\partial \varepsilon}{\partial k_\perp} = \frac{\partial g_o}{\partial \varepsilon} \hbar v_\perp(k) = -\hbar v_\perp(k) \delta(\varepsilon_F - \varepsilon), \quad (4.34)$$

and substituting this into Eq. (4.33) we have

$$\sigma_\perp = \frac{2e^2}{(2\pi)^3} \int d\mathbf{k} v_\perp^2(k) \tau(k) \delta(\varepsilon_F - \varepsilon). \quad (4.35)$$

Converting the integral to cylindrical polar coordinates,

$$d\mathbf{k} = d^3k = k_\parallel dk_\parallel d\theta dk_\perp, \quad (4.36)$$

and performing the angular integral we find

$$\sigma_\perp = \frac{2e^2}{(2\pi)^2} \iint k_\parallel dk_\parallel dk_\perp v_\perp^2(k) \tau(k) \delta(\varepsilon_F - \varepsilon). \quad (4.37)$$

We can change the variable of the delta function to  $k_\perp$ , giving

$$\sigma_\perp = \frac{2e^2\tau}{(2\pi)^2} \iint k_\parallel dk_\parallel dk_\perp \sum_{q'(k_\parallel)} \frac{v_\perp^2(k)}{|\hbar v_\perp(q'(k_\parallel))|} \delta(k_\perp - q'(k_\parallel)). \quad (4.38)$$

where we have again assumed that the scattering time is approximately constant over the Fermi surface. The sum over  $q'(k_\parallel)$  is over the zeros of  $(\varepsilon_F - \varepsilon)$  at a given value of  $k_\parallel$ , and is given by

$$q' = \pm \frac{1}{c} \arccos \left( \frac{\hbar^2 k_\parallel^2}{4t_c m^*} - \frac{\varepsilon_F}{2t_c} \right). \quad (4.39)$$

We can simplify this by writing

$$\sigma_\perp = \frac{2e^2\tau}{(2\pi)^2} \iint k_\parallel dk_\parallel dk_\perp \sum_{q(k_\parallel)} \frac{v_\perp^2(k)}{|\hbar v_\perp(q(k_\parallel))|} (\delta(k_\perp - q(k_\parallel)) + \delta(k_\perp + q(k_\parallel))) \quad (4.40)$$

where the  $q(k_\parallel)$  are the positive  $q'(k_\parallel)$ , ie

$$q = \frac{1}{c} \arccos \left( \frac{\hbar^2 k_\parallel^2}{4t_c m^*} - \frac{\varepsilon_F}{2t_c} \right). \quad (4.41)$$

The two delta functions have equal contributions to  $|v_\perp(q(k_\parallel))|$ , so this can be rewritten further as

$$\sigma_\perp = \frac{4e^2\tau}{(2\pi)^2} \iint k_\parallel dk_\parallel dk_\perp \sum_{q(k_\parallel)} \frac{v_\perp^2(k)}{|\hbar v_\perp(q(k_\parallel))|} \delta(k_\perp - q(k_\parallel)). \quad (4.42)$$

Collapsing the delta functions we have

$$\sigma_{\perp} = \frac{4e^2\tau}{\hbar(2\pi)^2} \int k_{\parallel} dk_{\parallel} \sum_{q(k_{\parallel})} |v_{\perp}(q(k_{\parallel}))| \quad (4.43)$$

remembering that  $v_{\perp}$  is only a function of  $k_{\perp}$ . The range of the integral over  $k_{\parallel}$  is the whole first brilluain zone. The domain of the arccos function is  $(-1 : 1)$ , so using these two limits, we can constrain the integral over  $k_{\parallel}$  to the only region where solutions (zeros) exist,  $q \in [q(k_{\parallel}) = -1 : q(k_{\parallel}) = +1]$ . Physically this means coordinates  $(k_{\perp}, k_{\parallel})$  that are on the Fermi surface. Before we do this, however, it will be useful to insert the expression for  $q$  into our equation for  $v_{\perp}$ .

$$\begin{aligned} \sigma_{\perp} &= \frac{4e^2\tau}{\hbar(2\pi)^2} \int k_{\parallel} \left| \frac{2ct_c}{\hbar} \sin \left( \arccos \left( \frac{\hbar^2 k_{\parallel}^2}{4t_c m^*} - \frac{\varepsilon_F}{2t_c} \right) \right) \right| dk_{\parallel} \\ \sigma_{\perp} &= \frac{2e^2\tau ct_c}{\hbar^2 \pi^2} \int k_{\parallel} \sqrt{1 - \left( \frac{\hbar^2 k_{\parallel}^2}{4t_c m^*} - \frac{\varepsilon_F}{2t_c} \right)^2} dk_{\parallel} \end{aligned}$$

We now make a change of variable

$$\beta = \frac{\hbar^2 k_{\parallel}^2 - 2\varepsilon_F m^*}{4m^* t_c}, \quad d\beta = \frac{\hbar^2}{2m^* t_c} k_{\parallel} dk_{\parallel} \quad (4.44)$$

such that

$$\sigma_{\perp} = \frac{4e^2\tau ct_c^2 m^*}{\hbar^4 \pi^2} \int \sqrt{1 - \beta^2} d\beta. \quad (4.45)$$

We can now investigate the bounds of the integral.  $q(\beta) = \arccos(\beta)/c$ , so we confine  $\beta \in [-1 : 1]$ ,

$$\sigma_{\perp} = \frac{4e^2\tau ct_c^2 m^*}{\hbar^4 \pi^2} \int_{-1}^1 \sqrt{1 - \beta^2} d\beta. \quad (4.46)$$

Physically, the values of  $\beta$  outside of this range are regions where there are no solutions to Eq. (4.41), ie there are no states on the Fermi surface with the corresponding value of  $k_{\parallel}$ .

Performing the integral we have

$$\sigma_{\perp} = \frac{4e^2\tau ct_c^2 m^*}{\hbar^4 \pi^2} \left( \frac{\beta \sqrt{1 - \beta^2}}{2} + \frac{\arcsin \beta}{2} \right) \Bigg|_{-1}^1 \quad (4.47)$$

finally giving

$$\sigma_{\perp} = \frac{2e^2\tau ct_c^2 m^*}{\pi \hbar^4}. \quad (4.48)$$

## Kadowaki-Woods Ratio in Quasi - Two Dimensions: Perpendicular to Planes

From the result for conductivity in equation Eq. (4.48) and using the previous result in Eq. (4.14) for  $1/\tau$  we can find the Kadowaki-Woods ratio for a quasi-two dimensional material when the conductivity is measured in the direction perpendicular to the planes. The coefficient  $A_{\perp}$ , given in Eq. (4.49), is found as previously for the 3D case. The coefficient  $\gamma$  is as given in Eq. (4.19). However the term  $n$  in equation Eq. (4.19) is different in the quasi-2D case, taking the form  $n = \bar{k}_F^2/2\pi c$ , where  $\bar{k}_F$  is the average of the wavevectors on the Fermi wavevector.

$$A_{\perp} = \frac{Bk_B^2\pi\hbar^3}{2e^2ct_c^2m^*\epsilon_F} = \frac{Bk_B^2\pi\hbar}{ce^2t_c^2\bar{k}_F^2} \quad (4.49)$$

Thus the Kadowaki-Woods ratio for a quasi-two dimensional material with conductivity measured perpendicular to the materials planes can be found.

$$R_{KW}^{\perp} = \frac{A_{\perp}}{\gamma^2} = \frac{4Bc\hbar^5}{\pi(et_c\bar{k}_F k_B m^*)^2} \quad (4.50)$$

Once again the dimensionless parameter  $B$  must be found. In the same way as previously we take Eq. (4.25) but this time use the quasi 2D expressions for  $k_{TF}$  and  $n$ .

$$k_{TF}^2 = \frac{me^2}{2\pi c\epsilon_o\hbar^2}$$

$$n = \frac{\bar{k}_F^2}{2\pi c}$$

The dimensionless constant  $B$  is found to be

$$B = \frac{8\pi n}{k_{TF}^2\bar{k}_F}$$

$$= \frac{8\pi\epsilon_o\hbar^2}{m^o e^2}\bar{k}_F \quad (4.51)$$

where  $m^o$  is once again the unrenormalised electron mass. Inserting this expression into Eq. (4.50) we find that the Kadowaki-Woods ratio measured in the direction perpendicular to the planes of a quasi-two dimensional material is dependent on the materials interlayer spacing and hopping amplitude, as well as the effective electron mass and Fermi wavevector (Eq. (4.52)).

$$R_{KW}^{\perp} = \frac{32\epsilon_o\hbar^7}{m^o k_B^2 e^4} \frac{c}{\bar{k}_F t_c^2 m^{*2}} \quad (4.52)$$

This expression is again clearly not universal, depending not only on the effective mass and Fermi wavevector, but also on the hopping integral  $t_c^2$  and interlayer spacing  $c$ .

## Conductivity in Quasi - Two Dimensions: Parallel to Planes

The derivation of the conductivity parallel to the planes requires a simple modification to that for perpendicular conductivity. It is straightforward to show that

$$\sigma_{\parallel} = \frac{-2e^2}{(2\pi)^3} \int d\mathbf{k} v_{\parallel}(k) \frac{\tau(k)}{\hbar} \frac{\partial g_o}{\partial k_{\parallel}}, \quad (4.53)$$

in a similar manner to the derivation of Eq. (4.33).

$$\frac{\partial g_o}{\partial k_{\parallel}} = \frac{\partial g_o}{\partial \varepsilon} \frac{\partial \varepsilon}{\partial k_{\parallel}} = \frac{\partial g_o}{\partial \varepsilon} \hbar v_{\parallel}(k) = -\hbar v_{\parallel}(k) \delta(\varepsilon_F - \varepsilon) \quad (4.54)$$

As before, we approximate the Fermi distribution as a step function. Thus,

$$\sigma_{\parallel} = \frac{2e^2}{(2\pi)^3} \int d\mathbf{k} v_{\parallel}^2(k) \tau(k) \delta(\varepsilon_F - \varepsilon). \quad (4.55)$$

Again converting the integral to cylindrical polar coordinates,

$$d\mathbf{k} = d^3k = k_{\parallel} dk_{\parallel} d\theta dk_{\perp} \quad (4.56)$$

we find

$$\sigma_{\parallel} = \frac{2e^2 \tau \hbar^2}{(2\pi)^2 m^{*2}} \int k_{\parallel}^3 dk_{\parallel} dk_{\perp} \delta(\varepsilon_F - \varepsilon) \quad (4.57)$$

where we have performed the integral over  $\theta$ , written the velocity  $v_{\parallel}(k)$  explicitly and again assumed that the scattering rate is constant over the Fermi surface.

The delta function can be expressed as

$$\delta(\varepsilon_F - \varepsilon) = \sum_{q'} \frac{\delta(k_{\perp} - q')}{|\hbar v_{\perp}|(q')} \quad (4.58)$$

where

$$q' = \pm \frac{1}{c} \arccos \left( \frac{\hbar^2 k_{\parallel}^2}{4m^* t_c} - \frac{\varepsilon_F}{2t_c} \right). \quad (4.59)$$

We can rewrite this in terms of the positive values of  $q'$ , denoted  $q$ , as

$$\delta(\varepsilon_F - \varepsilon) = \sum_q \frac{\delta(k_\perp - q) + \delta(k_\perp + q)}{|\hbar v_\perp(q)|}. \quad (4.60)$$

The conductivity is now

$$\sigma_{\parallel} = \frac{2e^2\tau\hbar^2}{(2\pi)^2m^{*2}} \int k_{\parallel}^3 dk_{\parallel} dk_{\perp} \sum_q 2 \frac{\delta(k_{\perp} - q)}{|\hbar v_{\perp}(q)|} \quad (4.61)$$

which has been further simplified by noting that the two delta functions once again give equal contributions.

Finally, we write the explicit form of the velocity and make a change of variable such that

$$\begin{aligned} \sigma_{\parallel} &= \frac{4e^2\tau\hbar^2}{(2\pi)^2m^{*2}} \int k_{\parallel}^3 dk_{\parallel} dk_{\perp} \sum_q \left| \frac{\delta(k_{\perp} - q)}{2t_c c \sin\left(\arccos\left(\frac{\hbar^2 k_{\parallel}^2}{4m^{*2}t_c} - \frac{\varepsilon_F}{2t_c}\right)\right)} \right| \\ &= \frac{4e^2\tau t_c}{c\pi^2\hbar^2} \int_{-1}^1 d\beta \frac{\beta + \frac{\varepsilon_F}{2t_c}}{\sqrt{1 - \beta^2}} \\ &= \frac{4e^2\tau t_c}{c\pi^2\hbar^2} \frac{\pi\varepsilon_F}{2t_c} \\ &= \frac{2e^2\tau\varepsilon_F}{c\pi\hbar^2} \end{aligned} \quad (4.62)$$

where

$$\beta = \frac{\hbar^2 k_{\parallel}^2}{4m^{*2}t_c} - \frac{\varepsilon_F}{2t_c}, \quad d\beta = \frac{\hbar^2}{2m^{*2}t_c} k_{\parallel} dk_{\parallel}$$

and the limits of the integration are set by the possible inputs to the arccos function which correspond to values of  $k_{\parallel}$  on the Fermi surface, as before.

## Kadowaki-Woods Ratio in Quasi - Two Dimensions: Parallel to Planes

Using the conductivity found in Eq. (4.62) and repeating the procedure of the previous sections we find that the coefficient of the resistivity in quasi-two dimensions measured parallel to the planes is given by

$$A_{\parallel} = \frac{2\pi c B k_B^2 m^{*2}}{e^2 \hbar^3 k_F^4}, \quad (4.63)$$

where we have approximated  $\varepsilon_F = \hbar^2 \bar{k}_F^2 / 2m^*$ , where  $\bar{k}_F$  is the average of wavevectors on the Fermi surface.

The two dimensional expression for  $\gamma$  is

$$\gamma = \frac{\pi k_B^2 m^*}{2c\hbar^2}, \quad (4.64)$$

so the Kadowaki-Woods ratio in quasi-two dimensions in the parallel direction is

$$R_{KW}^{\parallel} = \frac{A_{\parallel}}{\gamma^2} = \frac{2^3 \hbar B c^3}{\pi e^2 k_B^2 \bar{k}_F^4} \quad (4.65)$$

The quantity  $B$  is as given in Eq. (4.51), as it depends only on the geometry of the Fermi surface. Inserting the expression for  $B$  into Eq. (4.65) gives

$$R_{KW}^{\parallel} = \frac{2^6 \epsilon_o \hbar^3 c^3}{m^o k_B^2 e^4 \bar{k}_F^3} \quad (4.66)$$

again dependent on the interlayer spacing and Fermi wavevector. As expected the expression for the Kadowaki-Woods ratio measured in the parallel direction (Eq. (4.66)) does not depend on the interlayer hopping integral. Unlike the expression for interlayer transport, this expression does not depend on the enhanced mass.

### 4.3 Tight Binding Model

The most general we use here is the general tight binding model, where each dimension has a different hopping energy, such that

$$\varepsilon(k) = -2(t_a \cos ak_x + t_b \cos bk_y + t_c \cos ck_z). \quad (4.67)$$

#### Conductivity in the Tight Binding Model

In finding the conductivity, we begin as before with an expression for the conductivity in terms of an integral over the first Brillouin zone

$$\sigma = \frac{2e^2 \cos \theta}{(2\pi)^3} \int d\mathbf{k} \tau(\mathbf{k}) v_x^2(\mathbf{k}) \delta(\varepsilon - \varepsilon_F). \quad (4.68)$$

We rewrite the integral in terms of surfaces of constant energy and an energy integral, as in Eq. (4.7) finding

$$\begin{aligned}
\sigma &= \frac{2e^2 \cos \theta}{(2\pi)^3} \iint dS_\varepsilon d\varepsilon \frac{1}{|\nabla_{\mathbf{k}} \varepsilon|} \tau(\mathbf{k}) v_x^2(\mathbf{k}) \delta(\varepsilon - \varepsilon_F) \\
&= e^2 \cos \theta \int d\varepsilon D(\varepsilon) \tau(\mathbf{k}) v_x^2(\mathbf{k}) \delta(\varepsilon - \varepsilon_F) \\
&= e^2 \tau D^* \langle v_x^2 \rangle \cos \theta
\end{aligned} \tag{4.69}$$

where  $D^*$  is the renormalised density of states at the Fermi energy, and

$$D^*(\varepsilon) = \int \frac{dS_\varepsilon}{(2\pi)^3} \frac{2}{|\nabla_{\mathbf{k}} \varepsilon|}, \tag{4.70}$$

again approximating  $\tau$  as constant over the Fermi surface.

## Kadowaki-Woods Ratio in the Tight Binding Model

Proceeding as before, we find that

$$A = \frac{B k_B^2}{\hbar \varepsilon_F e^2 D^* \langle v_x^2 \rangle \cos \theta} \tag{4.71}$$

and using

$$\gamma = \frac{\pi^2 k_B^2 N_A D^*}{3\hbar} \tag{4.72}$$

we have

$$R_{KW}^{3DTB} = \frac{9\hbar B}{\pi^4 N_A^2 k_B^2 e^2 \cos \theta D^{*3} \varepsilon_F \langle v_x^2 \rangle}. \tag{4.73}$$

This is independent of renormalisation, since there will be a factor of  $Z^3$  on the density of states, and a factor of  $Z^{-3}$  from the Fermi energy and mean velocity squared. We use the definition of  $k_{TF}$  in Eq. (4.24) to find

$$B = \frac{4\epsilon_o \pi \langle v^o \rangle n \hbar}{e^2 D_o \varepsilon_F^o} \tag{4.74}$$

so

$$R_{KW}^{3DTB} = \frac{36\hbar^2 \epsilon_o n}{e^4 \varepsilon_F^{o2} \pi^3 N_A^2 k_B^2 \cos \theta D_o^4} \frac{\langle v^o \rangle}{\langle v_x^2 \rangle} \tag{4.75}$$

using  $Z \varepsilon_F^o = \varepsilon_F$  and  $Z \langle v^o \rangle = \langle v \rangle$ . In the same manner as before, we could attempt to find  $\langle v_x^2 \rangle$ . The integral

$$\langle v_x^2 \rangle = \frac{\frac{4t_c c}{\hbar^2} \iint dk_x dk_y \sqrt{1 - \left( \frac{\varepsilon_F + 2t_a \cos ak_x + 2t_b \cos bk_y}{2t_c} \right)^2}}{\int d\mathbf{k} \delta(\varepsilon - \varepsilon_F)} \tag{4.76}$$

is non-trivial to solve analytically, but could be easily integrated numerically for a given set of parameters.

## 4.4 Comparison of The Effects of Dimensionality

The Kadowaki-Woods ratios derived in this chapter are

$$\begin{aligned}
R_{KW}^{3D} &= 18h^3 \frac{\epsilon_o}{m^o k_B^2 e^4} \frac{1}{\bar{k}_F^6} \\
R_{KW}^{2D_\perp} &= 32\hbar^7 \frac{\epsilon_o}{m^o k_B^2 e^4} \frac{c}{t_c^2 m^* \bar{k}_F} \\
R_{KW}^{2D_\parallel} &= 2^6 \hbar^3 \frac{\epsilon_o}{m^o k_B^2 e^4} \frac{c^3}{\bar{k}_F^3} \\
R_{KW}^{3DTB} &= \cos \theta \frac{36\hbar^2 \epsilon_o}{e^4 \pi^3 N_A^2 k_B^2} \frac{n \langle v^o \rangle}{\epsilon_F^o D_o^4 \langle v_x^o \rangle}
\end{aligned}$$

$R_{KW}^{2D_\perp}$  depends on the inverse of the Fermi wavevector ( $\bar{k}_F^{-1}$ ), while  $R_{KW}^{2D_\parallel}$  depends on its cube ( $\bar{k}_F^{-3}$ ), and the three dimensional expression (3D) contains  $\bar{k}_F^{-6}$ .  $R_{KW}^{3D}$  and  $R_{KW}^{2D_\parallel}$  are independent of the effective mass, while  $2D_\perp$  has a factors of  $m^*$ . This can be interpreted as the effect of the mass on the interlayer hopping. The hopping integral  $t_c$  only appears in  $2D_\perp$  as would be expected.  $2D_\perp$  depends on the interlayer spacing while  $2D_\parallel$  depends on its cube. The ratio of the perpendicular and parallel Kadowaki-Woods ratios is

$$\frac{R_{KW}^{2D_\perp}}{R_{KW}^{2D_\parallel}} = \left( \frac{1}{t_c} \frac{\hbar^2 k_F}{cm^*} \right)^2 = \left( \frac{E_{well}}{t_c} \right)^2 \quad (4.77)$$

where

$$E_{well} = \frac{\hbar^2 k_F}{cm^*}$$

which looks like the ratio of the energy associated with hopping between layers, and the energy of a particle in an infinite potential well, the size of which is given by  $\frac{k_f}{c} \simeq \frac{2\pi\alpha}{ac}$ , where  $\alpha$  is the filling ratio and  $a$  is the size of the planar ( $x_\parallel$ ) dimensions.

### Values in the Organics

In terms of doing an experiment, it is extremely difficult to measure only the conductivity in the in-plane (parallel) direction. The resistivity in the plane is

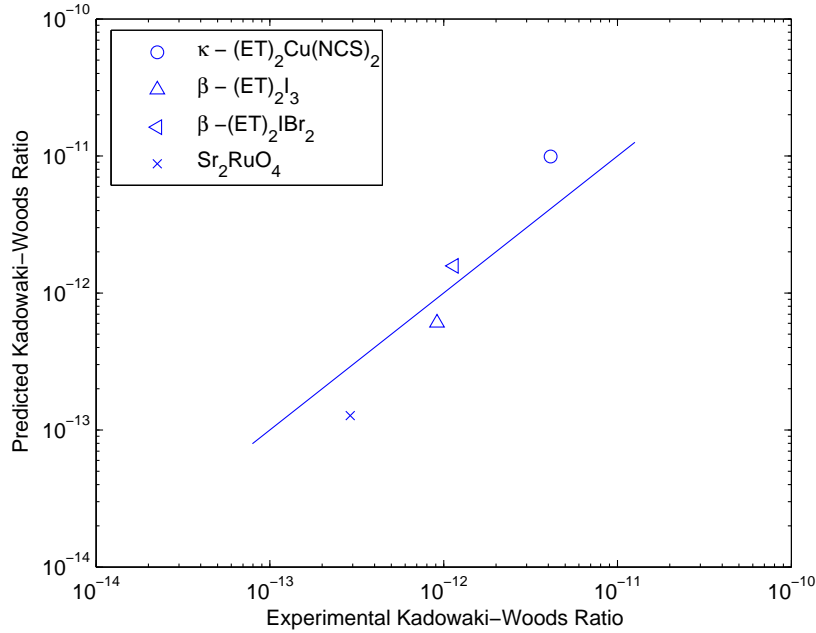


Figure 4.1: A comparison of the experimental values of the Kadovaki-Woods ratio for a range of quasi-two-dimensional materials and the values as calculated from the expression derived on the basis of a 2D free electron gas with a third tightly bound dimension (Eq. (4.52)). The solid line indicates a 1 : 1 correspondence between theory and experiment.

much larger than that perpendicular to the planes, so the in-plane resistivity will be washed out unless the contacts are connected to exactly the right planes of the sample. Since the interlayer spacing is on the order of  $10 \text{ \AA}$ , this is out of the reach. The perpendicular Kadovaki-Woods ratio  $R_{KW}^\perp$  has so far only been calculated for a small sample of quasi-two-dimensional materials which happen to be superconductors. As Fig. 4.1 shows, these results are very promising. The line in the figure indicates a 1 : 1 correspondence between theory and experiment.

## Chapter 5

# Quantum Many-Body Calculation of The Kadowaki-Woods Ratio: A Phenomenological Fermi Liquid Approach

“For those who want some proof that physicists are human, the proof is in the idiocy of all the different units which they use for measuring energy.”

- Richard Feynman

In this chapter we calculate the Kadowaki-Woods ratio using a phenomenological Fermi liquid approach. We use the Kramers-Kronig relation for the self energy to relate the heat capacity and resistivity, thus finding the Kadowaki-Woods ratio. As discussed previously, transition metals are found to have a universal Kadowaki-Woods ratio, while heavy fermion materials have a different, also universal ratio. Miyake, Matsuura and Varma [22] tried to explain these two values as limiting values of an interaction-dependent ratio that is not universal in general. Here we follow the derivation done by Miyake et al and investigate the resulting Kadowaki-Woods ratio for an anisotropic system. We find that the Kadowaki-Woods ratio is both not universal, and not renormalised by many body interactions.

## 5.1 The Self Energy

The self energy quantifies the effects of many body interactions on the propagation of quasi-particles. The retarded quasi-particle propagator is related to the retarded self energy  $\Sigma_R$  by Dyson's equation [2]

$$G_R(\omega, k) = \frac{1}{\omega - \varepsilon(k) - \Sigma_k(\omega, k)}. \quad (5.1)$$

In Fermi liquid theory there is an energy  $\omega_o$  such that for  $\omega \ll \omega_o$  the self energy can be expressed as [30]

$$\Sigma_R(\omega) = (1 - Z^{-1})\omega + iC\omega^2 \quad (5.2)$$

in the limit of  $T \rightarrow 0$  and for a system with no impurities. Note that the self-energy in a low temperature local Fermi liquid theory such as we are using is momentum independent.

For the purposes of this discussion, the self energy is probably best understood in terms of its effects on the quasi-particle properties. The derivative of the real part of the self energy with respect to energy  $\omega$  is related to the quasi-particle mass enhancement factor defined by  $m^*/m$ , and the imaginary part of the self energy is proportional to the quasi-particle scattering rate  $1/\tau$ . Using this information, we can write the retarded self-energy  $\Sigma_R$  as a function of the energy  $\omega$  as (in theorist's units)

$$\Sigma_R(\omega) = \Sigma'_R(\omega) + i\Sigma''_R(\omega) = \left(1 - \frac{m^*}{m}\right)\omega - i\frac{1}{2\tau(\omega)}. \quad (5.3)$$

The retarded self energy should tend towards zero in the high energy limit, so Miyake *et al.* [22] assume the following form for the imaginary part,

$$\Sigma''_R(\omega) = -\frac{1}{2\tau_o} - s\frac{\omega^2 + (\pi T)^2}{\omega_o^2}, \quad \text{for } |\omega^2 + (\pi T)^2| < \omega_o^2 \quad (5.4)$$

$$= -\left(\frac{1}{2\tau_o} + s\right) F\left(\sqrt{\frac{\omega^2 + (\pi T)^2}{\omega_o^2}}\right), \quad \text{for } |\omega^2 + (\pi T)^2| > \omega_o^2 \quad (5.5)$$

where  $s$  is a characteristic energy scale,  $T$  is temperature,  $\omega_o$  is a cut-off energy,  $\tau_o$  is the impurity scattering rate and  $F$  is a monotonic decreasing function with boundary conditions  $F(y) \rightarrow y^2$  as  $y \rightarrow 1$  and  $F(y) \rightarrow 0$  as  $y \rightarrow \infty$ . The form of Eq. (5.4) is known to be valid in the low energy, low impurity limit [30, 31] as it is clear that it reduces to Eq. (5.2).

## 5.2 The Kramers-Kronig Relations

This derivation closely follows that found in Landau & Lifshitz [32].

The Kramers-Kronig relation gives a direct correspondence between the real and imaginary parts of a response or correlation function.

In many physical systems we can determine the value of some quantity at a time  $t$  by looking at its linear response to some external driving function up to the time  $t$ . We say that the response  $K$  of  $y$  to some other function  $f$  is given by

$$y(t) = \int_{-\infty}^{\infty} K(t-t')f(t')dt'. \quad (5.6)$$

For  $f(t') = \delta(t')$  we have

$$y(t) = K(t) \quad (5.7)$$

thus it is clear that  $K$  is the response of  $y$  to a delta function perturbation. We change variable to find

$$y(t) = \int_{-\infty}^{\infty} K(\tau)f(t-\tau)d\tau \quad (5.8)$$

We expect the response to be causal, so we have  $K(\tau < 0) = 0$ . The Fourier transform pair for  $f$  is

$$f(t) = \int_{-\infty}^{\infty} f(\omega)e^{-i\omega t}d\omega, \quad f(\omega) = \int_{-\infty}^{\infty} \frac{1}{2\pi}f(t)e^{i\omega t}dt. \quad (5.9)$$

Thus we can write

$$\begin{aligned} y(t) &= \int_{-\infty}^{\infty} K(\tau) \int_{-\infty}^{\infty} f(\omega)e^{-i\omega(t-\tau)}d\omega d\tau \\ &= \int_{-\infty}^{\infty} \left( \int_{-\infty}^{\infty} K(\tau)e^{i\omega\tau}d\tau \right) f(\omega)e^{-i\omega t}d\omega \\ &= 2\pi \int_{-\infty}^{\infty} \chi(\omega)f(\omega)e^{-i\omega t}d\omega \end{aligned} \quad (5.10)$$

where  $\chi$  is defined as the Fourier transform of  $K$ , and thus

$$y(\omega) = \chi(\omega)f(\omega). \quad (5.11)$$

We can write  $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$ . The response  $K(t)$  is real, so

$$\chi^*(\omega) = \left( \int_{-\infty}^{\infty} K(\tau) e^{i\omega\tau} d\tau \right)^* \quad (5.12)$$

$$= \int_{-\infty}^{\infty} K^*(\tau) e^{-i\omega\tau} d\tau \quad (5.13)$$

$$= \int_{-\infty}^{\infty} K(\tau) e^{-i\omega\tau} d\tau \quad (5.14)$$

$$= \chi(-\omega), \quad (5.15)$$

thus

$$\chi^*(\omega) = (\chi'(\omega) + i\chi''(\omega))^* \quad (5.16)$$

$$= \chi'(\omega) - i\chi''(\omega) \quad (5.17)$$

$$= \chi(-\omega) \quad (5.18)$$

$$= \chi'(-\omega) + i\chi''(-\omega), \quad (5.19)$$

so  $\chi'(\omega)$  is even and  $\chi''(\omega)$  is odd.

If we let  $\chi$  be a function of a complex frequency  $\omega = \omega' + i\omega''$  we have

$$\chi(\omega) = \int_{-\infty}^{\infty} K(\tau) e^{i\omega'\tau - \omega''\tau} d\tau \quad (5.20)$$

$$\chi^*(\omega) = \int_{-\infty}^{\infty} K(\tau) e^{-i\omega'\tau - \omega''\tau} d\tau = \chi(-\omega^*). \quad (5.21)$$

We require that  $\omega'' > 0$ , otherwise the Fourier transform integral diverges.

The final stage of the derivation is a contour integral. Since  $\chi(\omega)$  is causal it has no poles in the upper-half of the complex plane, so we can write

$$\begin{aligned} \oint \frac{\chi(\tilde{\omega})}{\tilde{\omega} - \omega} d\tilde{\omega} &= 0 \\ &= \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi(\tilde{\omega})}{\tilde{\omega} - \omega} d\tilde{\omega} + \lim_{\varepsilon \rightarrow 0} \int_{\pi}^0 i\chi(\omega + \varepsilon e^{i\theta}) d\theta \end{aligned} \quad (5.22)$$

$$\therefore \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi'(\tilde{\omega}) + i\chi''(\tilde{\omega})}{\tilde{\omega} - \omega} d\tilde{\omega} = i\pi (\chi'(\omega) + i\chi''(\omega)) \quad (5.23)$$

where the first term of Eq. (5.22) is the Cauchy principal part and the second term is the infinitesimal semicircle above the pole at  $\omega$ , parameterised in terms of  $\theta$ . The final term to close the contour vanishes as usual as  $|\omega| \rightarrow \infty$ , as long as  $\chi(\omega)$  decays fast enough (Jordan's Lemma). Now all that remains

is to separate the real and imaginary parts of each side, thus arriving at the Kramers-Kronig relations

$$\chi''(\omega) = -\frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi'(\tilde{\omega})}{\tilde{\omega} - \omega} d\tilde{\omega} \quad (5.24)$$

and

$$\chi'(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi''(\tilde{\omega})}{\tilde{\omega} - \omega} d\tilde{\omega}. \quad (5.25)$$

Note that the only requirements needed to derive the Kramers-Kronig relations are that Jordan's Lemma is satisfied by the Fourier transform  $\chi$  of the response function, and that the response function  $K$  is causal.

### 5.3 Kramers-Kronig Transform for The Self-Energy

In 1961 Luttinger [33] proved that the self energy is analytic everywhere in the complex plane away from the real axis. This means that the Kramers-Kronig relation holds for the self energy, as physically  $\Sigma(\omega)$  should tend towards zero for large frequencies. His discussion is briefly summarised here.

The retarded propagator  $G_R(\omega, k)$  for a wavevector  $k$  at a frequency  $\omega$  is given in the Lehmann representation as [34]

$$G_R(\omega, k) = \int_{-\infty}^{\infty} d\varpi \frac{A(\varpi, k)}{\omega - \varpi} \quad (5.26)$$

where  $A(\omega, k)$  is the spectral density as a function of  $\varpi$  and  $k$ .

Letting  $\zeta \rightarrow \omega' + i\omega''$ , we have

$$\begin{aligned} G_R(\omega' + i\omega'', k) &= \int_{-\infty}^{\infty} d\varpi \frac{A(\varpi, k)}{\omega' + i\omega'' - \varpi} \\ &= \int_{-\infty}^{\infty} d\varpi \frac{A(\varpi, k)(\omega' - \varpi)}{(\omega' - \varpi)^2 + \omega''^2} - i\omega'' \int_{-\infty}^{\infty} d\varpi \frac{A(\varpi, k)}{(\omega' - \varpi)^2 + \omega''^2}. \end{aligned} \quad (5.27)$$

The imaginary part

$$\text{Im}[G_R(\omega' + i\omega'', k)] = -\omega'' \int_{-\infty}^{\infty} d\varpi \frac{A(\varpi, k)}{(\omega' - \varpi)^2 + \omega''^2} \quad (5.28)$$

is clearly only zero when  $\omega$ 's imaginary part  $\omega'' = 0$ , since both the spectral density and the denominator are positive definite.

The quasi-particle propagator is related to the retarded self energy  $\Sigma_R(\omega, k)$  by Dyson's equation [35]

$$G_R(\omega, k) = \frac{1}{G_R^o(\omega, k)^{-1} - \Sigma_R(\omega, k)} = \frac{1}{\omega - \varepsilon(k) - \Sigma_R(\omega, k)}, \quad (5.29)$$

where  $G^o(\omega, k)$  is the free particle propagator, so

$$\Sigma_R(\omega, k) = G_R^o(\omega, k)^{-1} - G_R(\omega, k)^{-1} = \omega - \varepsilon(k) - G_R(\omega, k)^{-1}. \quad (5.30)$$

$G_R(\omega, k)$  can only be zero when  $\omega''$  (and hence  $\text{Im}(G_R(\omega, k))$ ) is zero, which means it can only be zero on the real axis. This means that  $\Sigma_R(\omega, k)$  is analytic everywhere with the possible exception of on the real axis. By inserting an infinitesimal imaginary part we can shift any possible poles into the lower half plane and as such we can perform the Kramers-Kronig transform.

Since the Kramers-Kronig relation holds for the self energy, the definition of the imaginary part in Eqs (5.4) & (5.5) is sufficient to completely define  $\Sigma_R(\omega, k)$ , up to the full form of  $F(y)$ .

## 5.4 The Self-Energy Dependence of The Conductivity

Here we derive the conductivity in the 'pure' limit, the limit of a low impurity scattering rate.

We first derive the self energy dependence of the conductivity, and thus an equation for  $A(\Sigma'')$ . In the Kubo formalism, we can derive a quantum field theoretical analogue of the Boltzmann transport equation in terms of the self energy and vertex corrections [36]. In this treatment we neglect vertex corrections as they are expected to be small, as long as the electrons are not too localised [37]. We write the conductivity in the known form [23]

$$\sigma = \frac{\hbar^3 e^2}{3m^{\circ 2}} \int d\mathbf{k} \frac{k_x^2}{(2\pi)^3} \int \frac{d\varepsilon}{2\pi} A^2(\varepsilon, k) \left( \frac{-\partial f}{\partial \varepsilon} \right) \quad (5.31)$$

where  $A(\varepsilon, k)$  is the spectral density as a function of wavevector  $k$  and energy  $\varepsilon$ , and  $k_x$  is the component of the wavevector in the direction of the measurement. We know that the spectral density is related to the retarded Greens function by [23]

$$A(\varepsilon, k) = -2\text{Im}(G_R(\varepsilon, k)) \quad (5.32)$$

and the retarded Greens function is written as

$$\begin{aligned} G_R(\varepsilon, k) &= \frac{1}{\varepsilon - \varepsilon^o(k) - \Sigma'_R - i\Sigma''_R} \\ &= \frac{Z}{\varepsilon - Z\varepsilon^o(k) + ih/2\tau} \end{aligned} \quad (5.33)$$

so we have

$$A(\varepsilon, k) = \frac{-hZ/\tau}{(\varepsilon - Z\varepsilon^o(k))^2 + (h/2\tau)^2}. \quad (5.34)$$

It is clear that the spectral density has the form of a Lorentzian, and as such in the limit of a small scattering rate

$$\lim_{\Sigma''_R \rightarrow 0} A(\varepsilon, k) = \lim_{\Sigma''_R \rightarrow 0} \frac{-2\Sigma''_R Z^2}{(\varepsilon - Z\varepsilon^o(k))^2 + Z^2 \Sigma''_R{}^2} = -2\pi Z \delta(\varepsilon - Z\varepsilon^o(k)). \quad (5.35)$$

By considering

$$\lim_{\Gamma \rightarrow 0} \int_{-\infty}^{\infty} \frac{dx}{2\pi} \frac{2\Gamma}{x^2 + \Gamma^2} = 1, \quad (5.36)$$

we find that [23]

$$\lim_{\Gamma \rightarrow 0} \int_{-\infty}^{\infty} \frac{dx}{2\pi} \left( \frac{2\Gamma}{x^2 + \Gamma^2} \right)^2 = \lim_{\Gamma \rightarrow 0} \frac{1}{\Gamma} \quad (5.37)$$

and so we can write

$$\lim_{\Sigma''_R \rightarrow 0} A^2(\varepsilon, k) = \lim_{\Sigma''_R \rightarrow 0} \frac{-2\pi Z \delta(\varepsilon - Z\varepsilon^o(k))}{\Sigma''_R}. \quad (5.38)$$

Inserting this into Eq. (5.31) we find

$$\sigma = \frac{Z\hbar^3 e^2}{3m^{o2}} \int d\varepsilon \int d\mathbf{k} \delta(\varepsilon - Z\varepsilon^o(k)) \frac{k_x^2}{(2\pi)^3} \frac{1}{-\Sigma''_R} \left( \frac{-\partial f}{\partial \varepsilon} \right). \quad (5.39)$$

At this point, to simplify the calculation somewhat, we use our knowledge of the effect of the derivative of the Fermi distribution function at low temperatures. We expect it to pick out energies near the renormalised Fermi energy, so we thus approximate the result by removing the factor of  $k_x^2$  from the integrals, and averaging it over the Fermi surface. We denote this average by  $\langle k_x^2 \rangle$ . Further, we make the same approximation on the spectral density delta function, finding

$$\sigma = \frac{Z\hbar^3 e^2 \langle k_x^2 \rangle}{3m^{o2}} \int \frac{d\mathbf{k}}{(2\pi)^3} \delta(Z\varepsilon_F^o - Z\varepsilon^o(k)) \int d\varepsilon \frac{1}{-\Sigma''_R} \left( \frac{-\partial f}{\partial \varepsilon} \right). \quad (5.40)$$

The definition of the renormalised density of states at the renormalised Fermi energy  $\varepsilon_F^* = Z\varepsilon_F^o$  is

$$D^*(\varepsilon_F^*) \equiv \int \frac{d\mathbf{k}}{4\pi^3} \delta(Z\varepsilon_F^o - Z\varepsilon^o(k)) \quad (5.41)$$

$$\begin{aligned} &= \int \frac{d\mathbf{k}}{4\pi^3} \frac{\delta(\varepsilon_F^o - \varepsilon^o(k))}{Z} \\ &= \frac{D_o(\varepsilon_F^o)}{Z} \end{aligned} \quad (5.42)$$

Using this result we find

$$\begin{aligned} \sigma &= \frac{\hbar^3 e^2 \langle k_x^2 \rangle D_o(\varepsilon_F^o)}{6m^o2} \int d\varepsilon \frac{\left(\frac{-\partial f}{\partial \varepsilon}\right)}{-\Sigma_R''} \\ &= \frac{\hbar e^2 \langle v_x^2 \rangle m^{*2} D_o}{6m^o2} \int d\varepsilon \frac{\left(\frac{-\partial f}{\partial \varepsilon}\right)}{-\Sigma_R''} \\ &= \frac{\hbar e^2 \langle v_x^2 \rangle D_o}{3Z^2} \int d\varepsilon \frac{\left(\frac{-\partial f}{\partial \varepsilon}\right)}{-2\Sigma_R''(\varepsilon, T)} \end{aligned} \quad (5.43)$$

where we adopt the notation  $D_o$  for the unrenormalised density of states at the Fermi energy. The result in Eq. (5.43) has exactly the same form as that found in the work of Miyake et al [22].

At zero temperature the derivative of the distribution function with respect to energy becomes a delta function, so we have

$$\sigma(T=0) = \frac{\hbar e^2 \langle v_x^2 \rangle D_o}{3Z^2} \frac{1}{-2\Sigma_R''(0,0)} \quad (5.44)$$

where we have defined the Fermi energy  $\varepsilon_f = 0$ ,  $D_o$  is the unrenormalised density of states at the Fermi level as above, and  $\langle \dots \rangle$  indicates an average over the Fermi surface. So using Eq. (5.4) in SI units, we have for the zero temperature resistivity

$$\rho_o = \frac{-6Z^2 \Sigma_R''(0,0)}{\hbar e^2 \langle v_x^2 \rangle D_o} = \frac{6\pi}{e^2 \tau_o \langle v_x^2 \rangle D_o}. \quad (5.45)$$

For non-zero temperature we already know that

$$\sigma(T) = \frac{\hbar e^2 \langle v_x^2 \rangle D_o}{3Z^2} \int d\varepsilon \frac{\left(\frac{-\partial f}{\partial \varepsilon}\right)}{-2\Sigma_R''(\varepsilon, T)}. \quad (5.46)$$

Thus using Eq. (2.1) we can write

$$AT^2 \simeq \frac{3Z^2}{\hbar e^2 \langle v_x^2 \rangle D_o} \left( \left[ \int d\varepsilon \frac{\left(\frac{-\partial f_o}{\partial \varepsilon}\right)}{-2\Sigma_R''(\varepsilon, T)} \right]^{-1} + 2\Sigma_R''(0,0) \right). \quad (5.47)$$

We change variables to  $x = \varepsilon/k_B T$  (so the cut-off corresponding to  $\omega_o$  is  $x_o = \hbar\omega_o/k_B T$ ) and use Eq. (5.4) for  $\Sigma''$  within the cut off, finding

$$\aleph^{-1} \equiv \int_{-\infty}^{\infty} dx \frac{(-\frac{\partial f_o}{\partial x})}{-2\Sigma''_R(xk_B T, T)} \quad (5.48)$$

$$= \int_{-\infty}^{\infty} dx \frac{1}{4} \operatorname{sech}^2(x/2) \left[ \frac{\hbar}{\tau_o} + \frac{2s}{x_o^2} (x^2 + \pi^2) \right]^{-1} \quad (5.49)$$

$$= \frac{x_o^2}{8s} \int_{-\infty}^{\infty} dx \operatorname{sech}^2(x/2) \left[ \frac{\hbar x_o^2}{2s\tau_o} + x^2 + \pi^2 \right]^{-1}. \quad (5.50)$$

So we can write

$$AT^2 \simeq \frac{3Z^2}{\hbar e^2 \langle v_x^2 \rangle D_o} (\aleph + 2\Sigma''(0, 0)) \quad (5.51)$$

$$= \frac{3Z^2}{\hbar e^2 \langle v_x^2 \rangle D_o} \left( \left[ \frac{x_o^2}{8s} \int_{-\infty}^{\infty} dx \frac{\operatorname{sech}^2(x/2)}{\frac{\hbar x_o^2}{2s\tau_o} + x^2 + \pi^2} \right]^{-1} - \frac{\hbar}{\tau_o} \right). \quad (5.52)$$

We can now take the ‘pure’ limit by letting the residual resistivity approach zero, hence  $\tau_o \rightarrow \infty$ . We then evaluate the integral  $\aleph^{-1}$  numerically<sup>1</sup>, finding

$$\aleph^{-1} = \frac{x_o^2}{8s_o} \int_{-\infty}^{\infty} dx \frac{\operatorname{sech}^2(x/2)}{x^2 + \pi^2} \quad (5.53)$$

$$= \frac{x_o^2}{24s_o} \quad (5.54)$$

where the value of  $s$  in the pure limit is [22]

$$s_o = \frac{2n}{3\pi D_o} \quad (5.55)$$

and where  $n$  is the electron number density. We now have

$$\begin{aligned} A &= \frac{3Z^2 \aleph}{\hbar e^2 \langle v_x^2 \rangle D_o T^2} \\ &= \frac{72Z^2 s_o}{\hbar e^2 \langle v_x^2 \rangle D_o x_o^2 T^2} \\ &= \frac{48Z^2 n k_B^2}{\pi \hbar^3 e^2 \langle v_x^2 \rangle D_o^2 \omega_o^2}. \end{aligned} \quad (5.56)$$

<sup>1</sup>We used the numeric integration function in Mathematica, but any adaptive integration method will work by simply picking large but finite bounds for the integral, since this function dies off very quickly.

The expression for  $s_o$  for a 3D isotropic system reduces to

$$s_o = \frac{4\varepsilon_F}{9\pi}. \quad (5.57)$$

## 5.5 Kadowaki-Woods Ratio

Having found the coefficient of the resistivity, we now want to find the real part of the self energy in the pure limit, and hence the coefficient of the heat capacity. From the Kramers-Kronig transform, we know

$$\Sigma'(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\Sigma''(\omega')}{\omega' - \omega} d\omega' \quad (5.58)$$

$$\begin{aligned} -\Sigma'(\omega) &= \frac{s_o}{\pi\omega_o^2} \mathcal{P} \int_{-\omega_o}^{\omega_o} \frac{\omega'^2}{\omega' - \omega} d\omega' \\ &\quad + \frac{1}{\pi} \int_{\omega_o}^{\infty} \frac{s_o}{\omega' - \omega} F\left(\frac{\omega'}{\omega_o}\right) d\omega' \\ &\quad + \frac{1}{\pi} \int_{-\infty}^{-\omega_o} \frac{s_o}{\omega' - \omega} F\left(\frac{\omega'}{\omega_o}\right) d\omega' \end{aligned} \quad (5.59)$$

for  $|\omega| < \omega_o$  (meaning the pole is in the first term), where we have let  $T = 0$ . Dealing with the first term first, we have

$$\begin{aligned} I_1 &\equiv \frac{s_o}{\pi\omega_o^2} \mathcal{P} \int_{-\omega_o}^{\omega_o} \frac{\omega'^2}{\omega' - \omega} d\omega' \\ &= \frac{s_o}{\pi\omega_o^2} \lim_{\delta \rightarrow 0} \int_{\omega+\delta}^{\omega_o} \frac{\omega'^2}{\omega' - \omega} d\omega' \\ &\quad + \frac{s_o}{\pi\omega_o^2} \lim_{\delta \rightarrow 0} \int_{-\omega_o}^{\omega-\delta} \frac{\omega'^2}{\omega' - \omega} d\omega' \\ &= \frac{s_o}{\pi\omega_o^2} \lim_{\delta \rightarrow 0} \left[ (\omega^2 \ln(\omega' - \omega) + \omega\omega' + \omega'^2/2) \Big|_{\omega+\delta}^{\omega_o} \right. \\ &\quad \left. + (\omega^2 \ln(\omega' - \omega) + \omega\omega' + \omega'^2/2) \Big|_{-\omega_o}^{\omega-\delta} \right] \\ &= \frac{s_o}{\pi\omega_o^2} \left( 2\omega_o\omega + \omega^2 \left[ \ln\left(1 - \frac{\omega}{\omega_o}\right) - \ln\left(1 + \frac{\omega}{\omega_o}\right) \right] \right) \\ &\simeq \frac{2s_o}{\pi} \left( \frac{\omega}{\omega_o} - \frac{\omega^3}{\omega_o^3} \right). \end{aligned} \quad (5.60)$$

The other terms are given by

$$\begin{aligned}
I_2 &\equiv \frac{1}{\pi} \int_{\omega_o}^{\infty} \frac{s_o}{\omega' - \omega} F\left(\frac{\omega'}{\omega_o}\right) d\omega' \\
&\quad + \frac{1}{\pi} \int_{-\infty}^{-\omega_o} \frac{s_o}{\omega' - \omega} F\left(\frac{\omega'}{\omega_o}\right) d\omega' \\
&= \frac{1}{\pi} \int_1^{\infty} \frac{s_o}{y' - y} F(y') dy' \\
&\quad + \frac{1}{\pi} \int_{-\infty}^{-1} \frac{s_o}{y' - y} F(y') dy' \tag{5.61}
\end{aligned}$$

where we have changed variable such that  $y' = \omega'/\omega_o$  and  $y = \omega/\omega_o$ . We can use the binomial expansion to find

$$\begin{aligned}
I_2 &= \frac{1}{\pi} \int_1^{\infty} \frac{s_o}{y'(1 - y/y')} F(y') dy' \\
&\quad + \frac{1}{\pi} \int_{-\infty}^{-1} \frac{s_o}{y'(1 - y/y')} F(y') dy' \\
&= \frac{1}{\pi} \int_1^{\infty} \frac{s_o}{y'} F(y') \left(1 + \frac{y}{y'} + O\left(\frac{y}{y'}\right)^2 + \dots\right) dy' \\
&\quad + \frac{1}{\pi} \int_{-\infty}^{-1} \frac{s_o}{y'} F(y') \left(1 + \frac{y}{y'} + O\left(\frac{y}{y'}\right)^2 + \dots\right) dy' \tag{5.62}
\end{aligned}$$

Since  $F(y)$  is symmetric, terms of odd order in  $y'$  are zero. Since the remaining terms are symmetric, we have

$$\begin{aligned}
I_2 &= \frac{1}{\pi} \int_1^{\infty} \frac{s_o}{y'(1 - y/y')} F(y') dy' \\
&\quad + \frac{1}{\pi} \int_{-\infty}^{-1} \frac{s_o}{y'(1 - y/y')} F(y') dy' \\
&= \frac{2}{\pi} \int_1^{\infty} \frac{s_o y}{y'^2} F(y') + O\left(\frac{y}{y'}\right)^4 dy' \\
&= \frac{2s_o y}{\pi} \xi + \dots \\
&= \frac{2s_o \omega}{\pi \omega_o} \xi + \dots \tag{5.63}
\end{aligned}$$

where

$$\begin{aligned}\xi &\equiv \int_1^\infty F(y)/y^2 dy \\ &\leq \int_1^\infty 1/y^2 dy \\ &\leq 1.\end{aligned}$$

We expect  $F$  to die off slowly [22], so we expect  $\xi$  to be close to 1. Taking only the linear terms of  $I_1$  and  $I_2$ , we find that

$$\Sigma' \simeq -\frac{2s_o}{\pi} \frac{\omega}{\omega_o} (1 + \xi). \quad (5.64)$$

We can now write the linear coefficient of the heat capacity as

$$\begin{aligned}\gamma &= \gamma_o \left( 1 - \frac{\partial \Sigma'}{\partial \omega} \right) \\ &= \gamma_o \left( 1 + \frac{2s_o}{\pi \hbar \omega_o} (1 + \xi) \right)\end{aligned} \quad (5.65)$$

$$= \gamma_o \frac{2s_o}{\pi \hbar \omega_o} \left( 1 + \xi + \frac{\pi \hbar \omega_o}{2s_o} \right). \quad (5.66)$$

In the limit of  $\hbar \omega_o/s_o \ll 1$ ,

$$\gamma = \gamma_o \frac{2s_o}{\pi \hbar \omega_o} (1 + \xi). \quad (5.67)$$

Finally, using

$$\gamma_o = \frac{\pi^2}{3} k_B^2 D_o, \quad (5.68)$$

we have

$$\gamma = \frac{4n k_B^2}{9 \hbar \omega_o} (1 + \xi). \quad (5.69)$$

We can write the Kadowaki-Woods ratio

$$R_{KW} = \frac{3^5}{\pi \hbar k_B^2 e^2} \frac{Z^2}{n D_o^2 \langle v_x^2 \rangle (1 + \xi)^2} \quad (5.70)$$

where we have taken  $\hbar \omega_o/s_o \ll 1$ . The Kadowaki-Woods ratio is clearly not universal as it depends on the electron number density, the non-interacting density of states at the Fermi energy and most importantly in the case of anisotropic materials, the average of the square of the velocity in the direction of the conductivity measurement. It can be seen that this quantity

is not renormalised by the many-body interactions, since  $\hbar v = \nabla_k \varepsilon$ , the renormalised velocity is given by

$$\langle v_x^2 \rangle = \langle v_x^{o2} \rangle Z^2 \quad (5.71)$$

where  $v_x^o$  is the unrenormalised velocity. So we find

$$R_{KW} = \frac{3^5}{4\pi\hbar k_B^2 e^2} \frac{1}{n D_o^2 \langle v_x^{o2} \rangle}, \quad (5.72)$$

by approximating  $\xi \simeq 1$ , which constrains our choice of  $F(y)$  somewhat, but it is a reasonable constraint [22]. This result is valid for any metal that behaves like a Fermi liquid at low temperatures, and for which the impurity scattering rate is small. It is clear that this expression *is not renormalised by the many-body interactions*. While this expression is clearly not universal, it is reasonable to believe that it could appear universal in certain kinds of systems, particularly when plotted on a log-log scale. In this light it is not surprising that materials such as the organic superconductors do not have a universal Kadowaki-Woods ratio. Rather, the surprising thing is that it appears universal at all.

In a quasi-two dimensional material it is straightforward to find  $D_o$  and  $\langle v_x^{o2} \rangle$ . The density of states is

$$\begin{aligned} D_o &= \int \frac{d\mathbf{k}}{4\pi^3} \delta(\varepsilon - \varepsilon_F^o) \\ &= \int \frac{d\mathbf{k}}{4\pi^3} \delta\left(\frac{\hbar^2 k_{\parallel}}{2m^o} - 2t_c \cos(ck_{\perp}) - \varepsilon_F^o\right) \\ &= \frac{1}{\pi^2} \iint k_{\parallel} dk_{\parallel} dk_{\perp} \frac{\delta(k_{\perp} - q)}{|\hbar v_{\perp}(q)|} \\ &= \frac{1}{\pi^2} \int k_{\parallel} dk_{\parallel} \frac{1}{2t_c c \sqrt{1 - \left(\frac{\hbar^2 k_{\parallel}}{4t_c m^o} - \frac{\varepsilon_F^o}{2t_c}\right)^2}} \\ &= \frac{m^o}{c\pi^2 \hbar^2} \int_{-1}^1 d\beta (1 - \beta^2)^{-1/2} \\ &= \frac{m^o}{\pi c \hbar^2} \end{aligned} \quad (5.73)$$

where we have used  $\beta = \frac{\hbar^2 k_{\parallel}}{4t_c m^o} - \frac{\varepsilon_F^o}{2t_c}$  and  $q = \arccos(\beta)/c$  as in previous similar calculations, and  $t_c^o$  is the unrenormalised interlayer transport energy. The

averaged velocity is

$$\begin{aligned}
\langle v_x^{o2} \rangle &= \frac{\int \frac{d\mathbf{k}}{4\pi^3} v_x^{o2} \delta(\varepsilon - \varepsilon_F^o)}{\int \frac{d\mathbf{k}}{4\pi^3} \delta(\varepsilon - \varepsilon_F^o)} \\
&= \frac{\int \frac{d\mathbf{k}}{4\pi^3} v_x^{o2} \delta(\varepsilon - \varepsilon_F^o)}{D_o}
\end{aligned} \tag{5.74}$$

$$\begin{aligned}
\int \frac{d\mathbf{k}}{4\pi^3} v_x^{o2} \delta(\varepsilon - \varepsilon_F^o) &= \int \frac{k_{\parallel} dk_{\parallel}}{\hbar\pi^2} |v_x(q)| \\
&= \frac{4m^o c t_c^{o2}}{\hbar^4 \pi^2} \int_{-1}^1 \sqrt{1 - \beta^2} d\beta \\
&= \frac{2m^o c t_c^{o2}}{\hbar^4 \pi}
\end{aligned} \tag{5.75}$$

so we have

$$\begin{aligned}
\langle v_x^{o2} \rangle &= \frac{2m^o c t_c^{o2}}{\hbar^4 \pi} \frac{\pi c \hbar^2}{m^o} \\
&= \frac{2c^2 t_c^{o2}}{\hbar^2}.
\end{aligned} \tag{5.76}$$

In such a material the Kadowaki-Woods ratio is

$$R_{KW}^{2D1} = \frac{3^5 \pi \hbar^5}{8k_B^2 e^2 m^{o2}} \frac{1}{n t_c^{o2}}, \tag{5.77}$$

which is dependant only on the number density of charge carriers and the square of the unrenormalised interlayer transport energy.

For an isotropic three dimensional material such as the heavy fermions, we find

$$R_{KW}^{3D} = \frac{3^3 m^o}{2\pi \hbar k_B^2 e^2} \frac{\varepsilon_F^o}{n^3}. \tag{5.78}$$

This depends only on the unrenormalised Fermi energy and carrier density. Using the data in Appendix A we produced Fig. 5.1. As this figure shows, Eq. (5.78) predicts Kadowaki-Woods ratios in a variety of materials, over several orders of magnitude, to within a factor of 5 for all but one of these data points, CeCu<sub>2</sub>Si<sub>2</sub>.

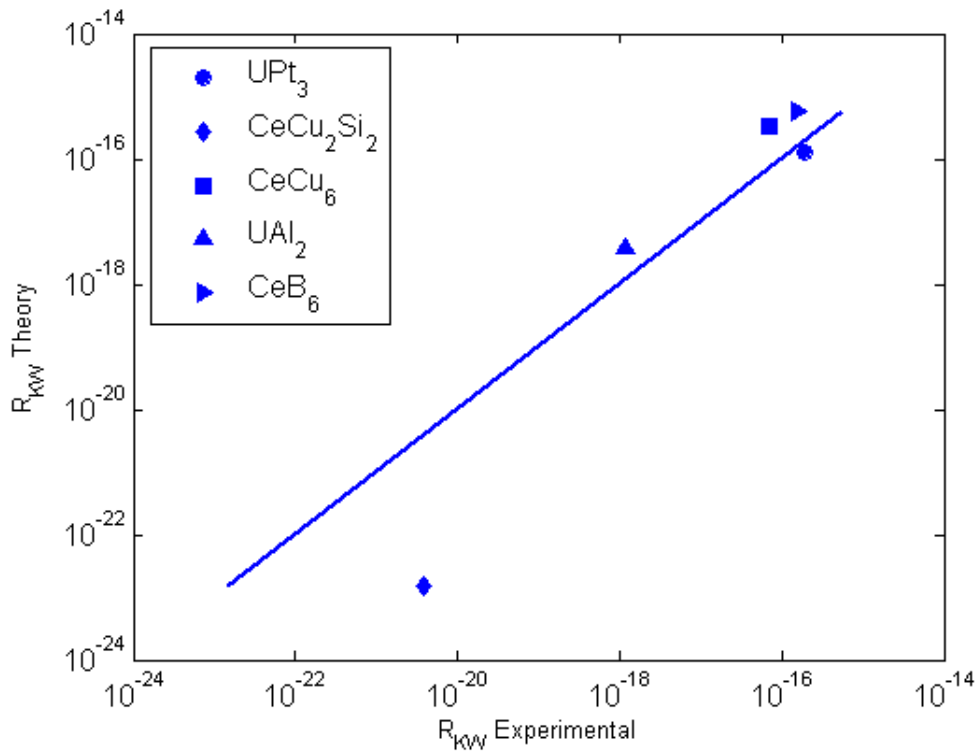


Figure 5.1: Experimental values versus theoretical predictions of the Kadowaki-Woods ratio using Eq. (5.78) in a variety of heavy fermion systems. The solid line is the line where theory perfectly matches experiment. The data used here can be found in Appendix A.

# Chapter 6

## Conclusions

“This is the end, my only friend, the end.”

- Jim Morrison

We have derived the Kadowaki-Woods ratio using both Boltzmann’s equation and the Kubo formula for the conductivity. Both these derivations indicated that the geometry of the system is a fundamental consideration when attempting to make predictions of the systems behaviour.

We first derived the Kadowaki-Woods ratio in a weakly interacting metal model for a variety of Fermi surface geometries. The results of these derivations are clearly dependent on the geometry, and in the case of the measurement of a quasi-two dimensional material perpendicular to its plane, dependent on renormalised properties.

We then derived the conductivity from the Kubo formalism, neglecting vertex corrections. We related this result to the heat capacity via the Kramers-Kronig relation and thus found a general expression for the Kadowaki-Woods ratio. This expression is found to be non-universal. It is also *not renormalised by the many-body interactions*. This is in opposition to the view of Miyake *et al.*, who claimed that the difference between the value of the Kadowaki-Woods ratio in heavy fermions and that in transition metals is due to limiting cases of the interaction strength. Our derivation indicates that the difference is rather due to the non-interacting properties.

While the results in heavy fermions are promising, further investigations with regard to the layered materials are needed for experimental confirmation.

# Appendix A

## Data for Heavy Fermion Materials

UPt <sub>3</sub>		
Quantity	Value	Reference
$A$	1.9 $\mu\Omega\text{ cm } K^{-2}$	[22]
$\gamma$	420 $mJ\text{ mol}^{-1} K^{-2}$	[38]
$V$	141 $\text{\AA}^3$	[39]
$Y$	2	[40]
$\varepsilon_F$	$2.62 \times 10^{-22}$ $J$	[41]
$m^*/m^o$	187	[39]
$R_{KW}^{exp}$	$1.94 \times 10^{-16}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-
$R_{KW}^{theor}$	$1.302 \times 10^{-16}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-

Table A.1: Data for UPt<sub>3</sub>.

CeB <sub>6</sub>		
Quantity	Value	Reference
$A$	0.7 $\mu\Omega\text{ cm } K^{-2}$	[22]
$\gamma$	250 $mJ\text{ mol}^{-1} K^{-2}$	[42]
$V$	71.0 $\text{\AA}^3$	[43]
$Y$	1.15	[43]
$\varepsilon_F$	$5.22 \times 10^{-20}$ $J$	[42]
$m^*/m^o$	6	[42]
$R_{KW}^{exp}$	$1.55 \times 10^{-16}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-
$R_{KW}^{theor}$	$5.59 \times 10^{-16}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-

Table A.2: Data for CeB<sub>6</sub>

CeCu <sub>6</sub>		
Quantity	Value	Reference
$A$	25 $\mu\Omega\text{ cm } K^{-2}$	[22]
$\gamma$	1500 $mJ\text{ mol}^{-1} K^{-2}$	[44]
$V$	420.6 $\text{\AA}^3$	[45]
$Y$	10	[46]
$\varepsilon_F$	$1.59 \times 10^{-21}$ $J$	[46]
$m^*/m^o$	380	[46]
$R_{KW}^{exp}$	$7.12 \times 10^{-17}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-
$R_{KW}^{theor}$	$3.41 \times 10^{-16}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-

Table A.3: Data for CeCu<sub>6</sub>

UAl <sub>2</sub>		
Quantity	Value	Reference
$A$	0.2 $\mu\Omega\text{ cm } K^{-2}$	[22]
$\gamma$	145 $mJ\text{ mol}^{-1} K^{-2}$	[47]
$V$	468.4 $\text{\AA}^3$	[48]
$Y$	80	[49]
$\varepsilon_F$	$1.43 \times 10^{-18}$ $J$	[50]
$m^*/m^o$	1.7	[51]
$R_{KW}^{exp}$	$1.18 \times 10^{-18}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-
$R_{KW}^{theor}$	$3.70 \times 10^{-18}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-

Table A.4: Data for UAl<sub>2</sub>

CeCu <sub>2</sub> Si <sub>2</sub>		
Quantity	Value	Reference
$A$	9.3 $\mu\Omega\text{ cm } K^{-2}$	[22]
$\gamma$	1000 $mJ\text{ mol}^{-1} K^{-2}$	[38]
$V$	167.7 $\text{\AA}^3$	[52]
$Y$	502 <sup>1</sup>	[52]
$\varepsilon_F$	$1.31 \times 10^{-22}$ $J$	[52]
$m^*/m^o$	400	[52]
$R_{KW}^{exp}$	$3.76 \times 10^{-21}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-
$R_{KW}^{theor}$	$1.48 \times 10^{-23}$ $K^2 m^5 s^3 kg^{-1} C^{-2}$	-

Table A.5: Data for CeCu<sub>2</sub>Si<sub>2</sub>

# Appendix B

## Data for Layered Materials

$\beta - (\text{ET})_2\text{I}_3$		
Quantity	Value	Reference
$\gamma$	24 $mJ \text{ mol}^{-1} K^{-2}$	[53]
$A$	$2 \times 10^{-7} \Omega \text{ cm } K^{-2}$	[54]
$V$	0.8559 $nm^3$	[55]
$Y$	1	[53]
$t_{\perp}$	$1.027 \times 10^{-22} J$	[56]
$k_F$	$3.40 \times 10^9 m^{-1}$	[57]
$c$	$1.53 \times 10^{-9} m$	[58]
$m^*/m^o$	3.9	[59]

Table B.1: Data for  $\beta - (\text{ET})_2\text{I}_3$

Sr <sub>2</sub> RuO <sub>4</sub>		
Quantity	Value	Reference
$\gamma$	37.5 $mJ \text{ mol}^{-1} K^{-2}$	[60]
$A$	$5.5 \times 10^{-6} \Omega \text{ cm } K^{-2}$	[60]
$V$	0.1908 $nm^3$	[60]
$Y$	(1.33, 0.91, 0.22)	[61]
$t_{\perp}$	$1.66 \times 10^{-23} J$	[61]
$k_F$	$(7.5, 6.2, 3) \times 10^9 m^{-1}$	[61]
$c$	$1.27 \times 10^{-9} m$	[60]
$m^*/m^o$	(16, 7, 3.3)	[61]

Table B.2: Data for Sr<sub>2</sub>RuO<sub>4</sub> with three Fermi surfaces.

$\kappa$ -(ET) <sub>2</sub> Cu(NCS) <sub>2</sub>		
Quantity	Value	Reference
$\gamma$	23 $mJ \text{ mol}^{-1} K^{-2}$	[62]
$A$	$8.5 \times 10^{-7} \Omega \text{ cm } K^{-2}$	[25]
$V$	1.688 $nm^3$	[63]
$Y$	2	[53]
$t_{\perp}$	$6.4 \times 10^{-24} J$	[64]
$k_F$	$(3.46, 1.35) \times 10^9 m^{-1}$	[65]
$c$	$1.31 \times 10^{-9} m$	[66]
$m^*/m^o$	(6.5, 3.5)	[67]

Table B.3: Data for  $\kappa$ -(ET)<sub>2</sub>Cu(NCS)<sub>2</sub> with two Fermi surfaces.

$\kappa$ - (ET) <sub>2</sub> Cu(N(CN) <sub>2</sub> )Br		
Quantity	Value	Reference
$\gamma$	25 $mJ \text{ mol}^{-1} K^{-2}$	[68]
$A$	$3 \times 10^{-6} \Omega \text{ cm } K^{-2}$	[25]
$V$	3.317 $nm^3$	[53]
$Y$	4	[53]

Table B.4: Data for  $\kappa$  - (ET)<sub>2</sub>Cu(N(CN)<sub>2</sub>)Br.

(TMTSF) <sub>2</sub> PF <sub>6</sub>		
Quantity	Value	Reference
$\gamma$	5.73 $mJ \text{ mol}^{-1} K^{-2}$	[69]
$A$	$2 \times 10^{-8} \Omega \text{ cm } K^{-2}$	[70]
$V$	0.714 $nm^3$	[71]
$Y$	1	[71]

Table B.5: Data for (TMTSF)<sub>2</sub>PF<sub>6</sub>.

Rb <sub>3</sub> C <sub>60</sub>		
Quantity	Value	Reference
$\gamma$	48 $mJ \text{ mol}^{-1} K^{-2}$	[72]
$A$	$1 \times 10^{-8} \Omega \text{ cm } K^{-2}$	[72]
$V$	3.0 $nm^3$	[53]
$Y$	4	[53]

Table B.6: Data for Rb<sub>3</sub>C<sub>60</sub>.

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