Thermal transport in granular metals

I. S. Beloborodov[1], A. V. Lopatin[1], F. W. J. Hekking[2],
Rosario Fazio[3] and V. M. Vinokur[1]

[1] *Materials Science Division, Argonne National Laboratory - Argonne, IL 60439, USA*
[2] *Laboratoire de Physique et Modélisation des Milieux Condensés*
CNRS and Université Joseph Fourier - BP 166, 38042 Grenoble-cedex 9, France
[3] *NEST-INFM and Scuola Normale Superiore - I-56126 Pisa, Italy*

Abstract. – We study the electron thermal transport in granular metals at large tunnel
conductance between the grains, $g_{\rm T} \gg 1$, and not too low a temperature, $T > g_{\rm T}\delta$, where δ
is the mean energy level spacing for a single grain. Taking into account the electron-electron
interaction effects, we calculate the thermal conductivity and show that the Wiedemann-Franz
law is violated for granular metals. We find that interaction effects suppress the thermal
conductivity less than the electrical conductivity.

The electrical conductivity σ and the thermal conductivity κ of an ordinary metal are
related to each other via a universal relation [1] known as the Wiedemann-Franz law:

$$\kappa/\sigma = L_0 T, \tag{1}$$

where $L_0 = \pi^2/3e^2$ is the Lorentz number, T is the temperature and e is the electron charge
(we use units such that $k_{\rm B} = \hbar = 1$). The Wiedemann-Franz (WF) law holds as long as
scattering processes are (quasi-)elastic, *i.e.* only weakly energy dependent.

Equation (1) is a direct consequence of Fermi-liquid theory. This theory describes the
properties of a weakly disordered, interacting electron gas, provided that screening renders
the Coulomb interactions sufficiently weak and short-ranged. Under these conditions, the
low-lying excitations of the metal are non-interacting fermionic quasi-particles that carry
both charge and energy, and, as a consequence, the WF law is valid [2]. In other words, no
additional information is obtained from a measurement of the thermal conductivity that is
not already present in the electrical conductivity as long as interaction effects result only in
the renormalization of electron spectral parameters.

By now, it is well known that the simultaneous action of disorder and interactions changes
the behavior of metals at low energies [3]. Interference of diffusively scattered electron waves
weakens the screening properties of the electron gas, thereby leading to an energy-dependent
increase of the interaction strength. Therefore, in contrast to an ordinary Fermi liquid, both

transport and thermodynamic quantities acquire an additional non-trivial energy dependence. An example is the so-called zero-bias anomaly: close to the Fermi level, Coulomb effects suppress the tunneling density of states. Another example is the temperature-dependent interaction correction to the Drude conductivity.

Since the low-energy properties of disordered metals with interactions deviate from those of an ordinary Fermi liquid, one is, in view of the above, naturally led to the conclusion that the Wiedemann-Franz law (or deviations from it) may serve as an important probe uncovering the underlying microscopic nature of disordered metallic systems. Although this problem has been addressed by several authors [4–8] over the past years, it has not been solved completely as yet. Reference [4] finds that the WF law holds, whereas in refs. [5–8] deviations from the WF relation are found (although these papers do not agree on the precise form of the deviations). We refer the reader to refs. [7,8] for a detailed account of the present status of the field.

In this letter we investigate this issue from a somewhat different perspective considering thermal transport in granular metals. A granular metal consists of small metallic grains coupled via tunnel junctions [9]. The electron tunneling between grains induces randomness, while the small electrical capacitance associated with the tunnel-coupled grains induces Coulomb charging effects. The interest in the physics of granular metals is twofold. First, their properties are generic for a wealth of disordered strongly correlated systems. On the other hand, as the interaction strength and degree of disorder can be controlled by properly choosing the parameters of the tunnel junctions and the grains, these systems offer a unique experimentally tunable disordered interacting system.

Depending on the value of the dimensionless tunnel conductance g_T between the grains, we can distinguish a low-conductivity regime, corresponding to $g_T \ll 1$ and a metallic, highly conducting regime, where $g_T \gg 1$. Electric transport in the limit $g_T \ll 1$ has been studied extensively [10, 11] since the early work by Mott [12]. More recent work deals with the electronic properties of granular systems in the metallic regime [13–16].

In what follows we focus on the electron thermal transport in the metallic regime, where the dimensionless tunnel conductance is large, $g_T \gg 1$. We show that in the absence of interactions the Wiedemann-Franz law holds and the thermal conductivity is given by

$$\kappa_0 = L_0 \sigma_0 T, \tag{2}$$

where $\sigma_0 = 2e^2 g_T a^{2-d}$ is the classical Drude conductivity for a granular metal (including spin), a is the size of a single grain, and d is the dimensionality of the system.

We find further that interactions violate the Wiedemann-Franz law in granular metals. Specifically, our result for the thermal conductivity can be conveniently formulated in terms of the quantity

$$\delta\kappa = \kappa - L_0 T \sigma, \tag{3}$$

that reflects deviations from the Wiedemann-Franz law. Including interaction effects, $\delta\kappa$ in eq. (3) is given by

$$\delta\kappa = \begin{cases} \gamma T/a, & d = 3, \\ \dfrac{T}{3} \ln\left(\dfrac{g_T E_C}{T}\right), & d = 2, \end{cases} \tag{4}$$

where E_C is the charging energy of an isolated grain and $\gamma \approx 0.355$ is a numerical constant. Our main result (4) is valid at not too low temperatures, $T > g_T \delta$, where δ is the mean level spacing of the grains. At these relatively high temperatures [15], the electronic motion is coherent within the grains, but coherence does not extend to scales larger than the size of

a single grain. Under these conditions, the electric conductivity σ in the right-hand side of eq. (3) is given by the expression obtained in refs. [15, 16],

$$\sigma = \sigma_0\big[1 - (1/2\pi dg_\mathrm{T})\ln(g_\mathrm{T}E_\mathrm{C}/T)\big], \tag{5}$$

where the electric conductivity σ_0 was defined below eq. (2). Contrary to the electric conductivity of granular metals, σ, given by eq. (5), the thermal conductivity $\delta\kappa$ in eq. (4) is sensitive to the long-ranged character of Coulomb interaction and therefore depends on the dimensionality of the sample, even at temperatures $T > g_\mathrm{T}\delta$.

One can see from eqs. (3) and (4) that the thermal conductivity increases with temperature in both two and three dimensions. From eq. (4) it follows that for granular films both $\delta\kappa$ and the correction to the electric conductivity (second term in the right-hand side of eq. (5)) have the same logarithmic behavior such that in 2d the thermal conductivity can be written as

$$\kappa_{2\mathrm{d}} = \kappa_0 - \frac{(\pi - 2)}{6}\, T\ln\left(\frac{g_\mathrm{T}E_\mathrm{C}}{T}\right), \tag{6}$$

where κ_0 is defined by eq. (2).

Having summarized the main results, we now turn to the quantitative description of our model and derivation of eq. (4): We consider a d-dimensional array of metallic grains. The motion of electrons inside the grains is diffusive and they can tunnel between grains. Interaction effects stem from the long-range part of the Coulomb interaction, which is just the classical electrostatic charging energy of the grains. Such an array of weakly coupled metallic grains is described by the Hamiltonian $\hat{H} = \sum_i \hat{H}_i$, where the sum is taken over all grains in the system and

$$\hat{H}_i = \sum_k \xi_k a_{i,k}^\dagger a_{i,k} + \frac{e^2}{2}\sum_j \hat{n}_i\, C_{ij}^{-1}\, \hat{n}_j + \sum_{j\neq i, p, q} t_{ij}^{pq} a_{i,p}^\dagger a_{j,q}\,. \tag{7}$$

The first term in the right-hand side of eq. (7) describes free electrons in the i-th isolated disordered grain, $a_{i,k}^\dagger(a_{i,k})$ are the creation (annihilation) operators for an electron in the state k and $\xi_k = k^2/2m - \mu$ with μ being the chemical potential. The second term describes the charging energy with the capacitance matrix C_{ij} and the electron number operator $\hat{n}_i$ in the i-th grain. The last term represents the tunneling between the grains, where t_{ij}^{pq} is the tunnel matrix element corresponding to the points of contact of i-th and j-th grains.

The thermal conductivity of a granular metal is given by the analytical continuation of the Matsubara thermal current-current correlator. For granular metals the thermal current operator $\hat{J}_{ij}^h$ between the i-th and the j-th grain can be obtained as follows. The energy content of a given grain i changes as a function of time, such that

$$\mathrm{d}\hat{H}_i/\mathrm{d}t = i\big[\hat{H}, \hat{H}_i\big]. \tag{8}$$

Energy conservation requires that this energy flows to the other grains in the system. From a straightforward calculation of the commutator in eq. (8) we obtain $\hat{J}_{ij}^h$, the amount of energy flowing per unit time from grain i to a neighboring grain j. It can be written as

$$\hat{J}_{ij}^h = \hat{J}_{ij}^{(1)} + \hat{J}_{ij}^{(2)}, \tag{9a}$$

where $\hat{J}_{ij}^{(1)}$ is the heat current in the absence of electron-electron interaction effects,

$$\hat{J}_{ij}^{(1)} = \frac{i}{2}\sum_{p,q}(\xi_p + \xi_q)\, t_{ij}^{pq}\, a_{i,p}^\dagger a_{j,q} + \mathrm{H.c.}, \tag{9b}$$
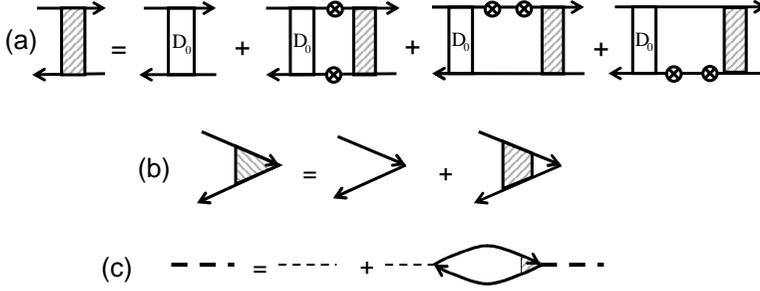
Fig. 1 – Diagrams representing: (a) the diffusion propagator of granular metals $D(\Omega_n, \boldsymbol{q})$ in eq. (11). The tunneling vertices are described by the circles; (b) interaction vertex renormalized by impurities and intergranular scattering; (c) the effective screened Coulomb interaction for granular metals $V(\Omega_n, \boldsymbol{q})$ in eq. (12).

while the second term, $\hat{J}_{ij}^{(2)}$, on the right-hand side of eq. (9a) appears due to Coulomb interaction effects:

$$\hat{J}_{ij}^{(2)} = -\frac{1}{4} \sum_l \left[\hat{n}_i C_{il}^{-1} \hat{J}_{lj} - \hat{n}_j C_{jl}^{-1} \hat{J}_{li} \right] + \text{H.c.} \tag{9c}$$

In eq. (9c) the operator $\hat{J}_{ij}$ is the electric current which is given by the following expression:

$$\hat{J}_{ij} = i \sum_{p,q} t_{ij}^{pq} \, a_{i,p}^{\dagger} a_{j,q} + \text{H.c.} \tag{9d}$$

For large tunnel conductance, the Matsubara thermal current-current correlator can be analyzed by virtue of the perturbation expansion in $1/g_{\text{T}}$, using the diagrammatic technique developed in refs. [13, 14] which we briefly outline here. The average electron Green function has the same form as that of a homogeneously disordered metal with self-energy $\Sigma(\omega_n) = -(i/2\tau) \text{sgn} \, \omega_n$, where ω_n is a fermionic Matsubara frequency. The elastic mean free time τ is mainly determined by the processes of scattering by impurities inside the grain. Intergranular scattering processes (tunneling) are included, assuming that tunneling elements are random Gaussian variables correlated according to

$$\left\langle t_{ij}^{p_1 p_2} \, t_{ji}^{p_3 p_4} \right\rangle = t^2 \delta_{p_1 p_4} \delta_{p_2 p_3} \, . \tag{10}$$

Here i and j are the indices of the neighboring grains and t is the tunnel amplitude, which is related to the dimensionless tunnel conductance, $g_{\text{T}} = 2\pi t^2/\delta^2$. Inclusion of intergranular scattering processes results in a small renormalization of the mean free time τ. The diffusive motion inside a single grain is given by the ladder diagram that results in the diffusion propagator $D_0^{-1} = \tau |\Omega_n|$, where Ω_n is a bosonic Matsubara frequency. The coordinate dependence in D_0 is neglected since in the regime under consideration all characteristic energies are less than the Thouless energy. The complete diffusion propagator is given by the ladder diagrams shown in fig. 1a resulting in the following expression:

$$D^{-1}(\Omega_n, \boldsymbol{q}) = \tau(|\Omega_n| + \varepsilon_{\boldsymbol{q}} \delta), \tag{11}$$

where $\varepsilon_{\boldsymbol{q}} = 2g_{\text{T}} \sum_{\boldsymbol{a}} (1 - \cos \boldsymbol{q} \boldsymbol{a})$ with $\boldsymbol{a}$ being the lattice vectors is the function of quasi-momentum $\boldsymbol{q}$ that appears due to the tunneling between the grains. The same ladder diagrams describe the renormalized interaction vertex in fig. 1b. This vertex is used to obtain the
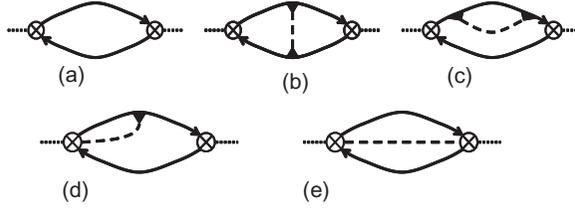
Fig. 2 – Diagrams describing the thermal conductivity of granular metals at temperatures $T > g_\mathrm{T}\delta$: the diagram (a) corresponds to κ_0 in eq. (1). Diagrams (b)-(e) represent first-order corrections in $1/g_\mathrm{T}$ to the thermal conductivity of granular metals due to electron-electron interaction. The solid lines denote the propagator of electrons, the dashed lines describe the effective screened electron-electron propagator and the black triangles describe the elastic interaction of electrons with impurities. The tunneling vertices are described by the circles.

polarization operator, that defines the effective dynamically screened Coulomb interaction for granular metals, fig. 1c,

$$V(\Omega_n, \boldsymbol{q}) = \left[\frac{C(\boldsymbol{q})}{e^2} + \frac{2\varepsilon_{\boldsymbol{q}}}{|\Omega_n| + \delta\varepsilon_{\boldsymbol{q}}} \right]^{-1}, \tag{12}$$

where $C(\boldsymbol{q})$ is the Fourier transform of the capacitance matrix.

In the absence of electron-electron interactions, the thermal conductivity is given by the diagram (a) in fig. 2. A direct calculation of this diagram confirms that the thermal conductivity is given by κ_0 in eq. (2) and thus that the WF law holds. At temperatures $T > g_\mathrm{T}\delta$, the first-order interaction corrections to the thermal conductivity are given by the diagrams (b)-(e) in fig. 2. We neglect the diagrams that can be obtained from (b)-(e) by insertion of an extra diffusion propagator since their contribution is small in the parameter $g_\mathrm{T}\delta/T$. The diagrams (b) and (c) in fig. 2 stem from contributions of the non-interacting part $\hat{J}_{ij}^{(1)}$ to the thermal current-current correlator. These diagrams are analogous to the ones considered in ref. [16] for the interaction correction $\delta\sigma$ to the conductivity of granular metals. In fact, we find that the contribution of diagrams (b) and (c) to $\delta\kappa$ in eq. (3) is cancelled exactly by the corresponding term $L_0 T \delta\sigma$. Thus, although these diagrams lead to corrections to both σ and κ, they do not lead to a violation of the WF law. Diagrams (d) and (e) contain the interacting part $\hat{J}_{ij}^{(2)}$ of the thermal current. There are no analogous diagrams for electric conductivity, hence any non-vanishing contribution from (d) and (e) inevitably leads to a violation of the WF law. The diagram (d) in fig. 2 turns out to be zero. Thus, among the diagrams (b)-(e) in fig. 2 only the diagram (e) contributes to $\delta\kappa$ in eq. (3) giving rise to the correction

$$\delta\kappa = -\frac{g_\mathrm{T} a^{2-d}}{2\pi T^2} \sum_{\boldsymbol{q}} \int \frac{\Omega \, \mathrm{d}\Omega}{\sinh^2(\Omega/2T)} \, \mathrm{Im}\, \tilde{V}_{\boldsymbol{q}}(\Omega), \tag{13a}$$

where the sum is taken over the quasi-momentum $\boldsymbol{q}$ and the potential $\tilde{V}_{\boldsymbol{q}}(\Omega)$ is given by the following expression:

$$\tilde{V}_{\boldsymbol{q}}(\Omega) = \frac{-i\Omega \left[1 + \cos(q_x a_x)\right] E_\mathrm{C}(\boldsymbol{q})}{-i\Omega + 4 E_\mathrm{C}(\boldsymbol{q}) \varepsilon_{\boldsymbol{q}}}. \tag{13b}$$

Here $E_\mathrm{C}(\boldsymbol{q}) = e^2/2C(\boldsymbol{q})$ is the charging energy. For the 3d case we simplify the right-hand side of eq. (13b) taking the limit $E_\mathrm{C}(\boldsymbol{q}) \to \infty$. Performing integration over Ω and summation over $\boldsymbol{q}$ in eq. (13a), we obtain our final result for the thermal conductivity (4) in the 3d case,

where the numerical coefficient

$$\gamma = \frac{\pi a^3}{6} \int \frac{\mathrm{d}^3 q}{(2\pi)^3} \frac{1 + \cos(q_x a_x)}{\sum_{\boldsymbol{a}} [1 - \cos(\boldsymbol{q}\boldsymbol{a})]} \approx 0.355.$$

For the 2d case, one can see that the main contribution to the integral over $\boldsymbol{q}$ comes from the region of small quasi-momenta $q \ll a^{-1}$, where one can use the asymptotic expression for the capacitance matrix $C(\boldsymbol{q}) = a^2 q/2\pi$. Performing the integrations in eq. (13a) with logarithmic accuracy, we obtain the final result for the correction to the thermal conductivity of 2d granular metals (4).

So far, we ignored the fact that electron-electron interactions also renormalize the chemical potential μ. Generally, this renormalization may affect the thermal conductance: the thermal current vertex, eq. (9b), as well as the electron Green functions depend on μ. To first order in the interactions, the renormalization of μ only leads to corrections to diagram (a) in fig. 2. As can be easily shown, for this diagram the renormalization of the two heat current vertices is exactly cancelled by the renormalization of the two electron propagators. Therefore, the renormalization of the chemical potential by the interactions does not affect our results in lowest order.

In this paper we only consider the electron contribution to thermal conductivity. As is well known, phonons will provide an independent, additional contribution to thermal transport. Although it is generally argued that phonons can be ignored at low temperatures, this point needs to be considered with care as we will now argue. Consider acoustic phonons that propagate with velocity c_{ph} in a three-dimensional impure host, characterized by a phonon scattering length l_{ph}. In this simple model, the phonon contribution to the thermal conductivity is given by $\kappa_{\mathrm{ph}} = T^3 l_{\mathrm{ph}}/c_{\mathrm{ph}}^2$. We see that $\kappa_{\mathrm{ph}} \sim \kappa_0$ at a temperature $T^* \sim \sqrt{g_{\mathrm{T}} c_{\mathrm{ph}}^2/l_{\mathrm{ph}} a}$. The electron contribution to thermal transport dominates only for $T < T^*$. In particular, if the phonon contribution is to be ignored in the temperature range relevant to us, we need to impose $g_{\mathrm{T}}\delta < T^*$. It is clearly a matter of system parameters whether or not this condition will be satisfied in a typical experiment. Avoiding the phonon contribution in a measurement of thermal conductivity thus implies constraints on the choice of materials as well as on sample fabrication.

In conclusion, we have investigated the electron thermal transport in granular metals in the limit of large tunneling conductance between the grains and temperatures $T > g_{\mathrm{T}}\delta$. We have calculated the thermal conductivity of granular metals and shown that the Wiedemann-Franz law in granular metals is violated if electron-electron interaction effects are taken into account. Our result can be expressed conveniently through the Lorentz number of the granular metal, $L = \kappa/\sigma T$. In the absence of interactions, the Lorentz number is a universal constant, $L = L_0 = \pi^2/3e^2$. In the presence of interactions, we find that $L = L_0 + \delta L$, where $\delta L = \mathrm{const}$ for a three-dimensional granular system, whereas $\delta L \sim \ln(g_{\mathrm{T}} E_{\mathrm{C}}/T)$ for a granular film. In both cases, $\delta L > 0$, indicating that interactions suppress electric conductivity more than the thermal conductivity.

Since interacting granular metals behave differently from homogeneous disordered metals, we cannot directly compare our results with the ones obtained in refs. [4–8]. Nevertheless, the structure of our result for thermal conductivity, as given by eqs. (3) and (4), is consistent only with that of the results of ref. [7, 8]. Specifically, we find two contributions of opposite sign related to interactions. One contribution is the interaction correction to electric conductivity, multiplied by TL_0. The other contribution leads to a violation of the WF law and is logarithmic in temperature for the 2d case, as in ref. [7]. Of course the prefactors are different, and a complete agreement can be expected only in the low-temperature limit, $T < g_{\mathrm{T}}\delta$, which is beyond the scope of this letter.

Experiments on thermal transport are important since they allow for a direct observation of deviations from ordinary Fermi-liquid behavior. However, measurements of the electron thermal conductivity of a mesoscopic conductor at low energy is still an experimental challenge. So far, only two-dimensional electron gas systems were studied [17]; more recent experiments aimed at achieving the accuracy that is necessary to observe deviations from the WF law [18]. We hope that our results that reveal deviations from Fermi-liquid behavior of granular metals will stimulate further experiments on thermal transport.

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