Violation of the Wiedemann-Franz Law in Hydrodynamic Electron Liquids

Alessandro Principi* and Giovanni Vignale
Department of Physics and Astronomy, University of Missouri, Columbia, Missouri 65211. USA
(Received 16 June 2014; revised manuscript received 16 January 2015; published 31 July 2015)

The Wiedemann-Franz law, connecting the electronic thermal conductivity to the electrical conductivity of a disordered metal, is generally found to be well satisfied even when electron-electron (e-e) interactions are strong. In ultraclean conductors in the hydrodynamic regime, however, large deviations from the standard form of the law are expected, due to the fact that e-e interactions affect the two conductivities in radically different ways. Thus, the standard Wiedemann-Franz ratio between the thermal and the electric conductivity is reduced by a factor \(1 + \tau_\text{th}/\tau_\text{ee}\), where \(1/\tau\) is the momentum relaxation rate and \(\tau_\text{ee}\) is the relaxation time of the thermal current due to e-e collisions. Here we study the density and temperature dependence of \(1/\tau_\text{th}\) of two-dimensional electron liquids. We show that at low temperature \(1/\tau_\text{th}\) is \(8/5\) of the quasiparticle decay rate; remarkably, the same result is found in doped graphene and in conventional electron liquids in parabolic bands.

DOI: 10.1103/PhysRevLett.115.056603
PACS numbers: 72.80.Vp, 65.80.Ck, 72.20.Pa

Introduction.—Hydrodynamics [1] offers a natural framework to deal with thermal transport in clean, strongly interacting many-particle systems. When a local quasiequilibrium state is established, a fluid can be characterized by five slowly varying time-dependent variables: the density \(n(r,t)\), the drift velocity \(v(r,t)\), and the local temperature \(T(r,t)\). Their time evolution is determined by the continuity equations for particle number and energy density, and by the Navier-Stokes equation. These equations in turn are controlled by a handful of transport coefficients: the bulk and shear viscosities and the thermal conductivity. Recently, it has been conjectured [2] that this hydrodynamic regime of the electron liquid can be realized in high-quality graphene, a monolayer of carbon atoms packed in a two-dimensional (2D) honeycomb lattice [3,4], in a broad range of temperatures and carrier densities. Recent experiments [5–7] have reported measurements of the electronic thermal conductivity of (suspended or supported) doped graphene. Its order of magnitude is found to be comparable to the extremely high phononic contribution [8], or even dominant at sufficiently low temperature.

The electronic thermal current \(J^{(Q)}\) is precisely defined as the temperature \(T\) times the entropy current carried by a high-mobility electron gas. Equivalently [9], \(J^{(Q)} = J^{(E)} - \mu J^{(N)}\), where \(J^{(E)}\) is the energy current, \(J^{(N)}\) is the particle current, and \(\mu\) is the chemical potential. The thermal current is related to the gradient of the temperature by the Fourier law \(J^{(Q)} = -\kappa T\) at zero particle current [9]. Here \(\kappa\) is the thermal conductivity.

The electronic thermal and charge (\(\sigma_e\)) dc conductivities of an electron gas in the diffusive regime [9,10] are connected by the Wiedemann-Franz (WF) law [11,12],

\[
\frac{\sigma_{\text{th}}}{\sigma_e T} = \frac{\pi^2 k_B^2}{3e^2} \equiv L_0,
\]

where \(L_0\)—the so-called “Lorenz number”—is a universal constant, independent of material parameters. This elegant statement reflects the fact that a single set of carriers (electrons) transports both the charge and the thermal energy, and that the scattering mechanism (mainly electron-impurity scattering at low temperature) affects both thermal and charge conductivities in the same way. The standard derivation of the WF law [9,13] ignores electron-electron (e-e) interactions, which can, in principle, change the value of the WF ratio by affecting the two conductivities in different ways. At a finite frequency \(\omega\), the conductivities can be expressed as

\[
\sigma_e(\omega) = \frac{Q_e D_e}{i\omega + 1/\tau_e},
\]

where \(\ell = c\) for the charge conductivity, \(\ell = \text{th}\) for the thermal conductivity, \(Q_e = e^2\), and \(D_e = \pi^2 k_B^2 T/3\). Here \(\tau_e\) and \(\tau_{\text{th}}\) are the relaxation times of charge and thermal currents, respectively, and \(D_e\) and \(D_{\text{th}}\) are the corresponding “Drude weights.” Electron-electron interactions can modify the WF ratio by creating a difference between (i) the relaxation times \(\tau_e\) and \(\tau_{\text{th}}\) and/or (ii) the Drude weights. In general, the “amended” WF relation follows immediately from Eq. (2), and reads

\[
\frac{\sigma_{\text{th}}}{\sigma_e T} = \frac{\pi^2 k_B^2 D_{\text{th}}}{3 e^2 D_e} \frac{\tau_{\text{th}}}{\tau_e} \equiv L.
\]

In the diffusive regime [14–19], \(\tau_{\text{th}}\) and \(\tau_e\) are nearly identical and are controlled by momentum-nonconserving processes. This, combined with the absence of any renormalization of the Drude weights [14,15] (see, however, below) led to the conclusion that the WF law remains valid also in the presence of e-e interactions.
In the hydrodynamic regime, which, by definition, is the regime in which the e-e scattering time is much shorter than the electron-impurity or electron-phonon scattering times [20], a large difference can appear between the charge-current and thermal-current relaxation times, with the former being much larger than the latter. This happens because charge currents can be relaxed only by momentum-nonconserving processes, while the thermal relaxation rate also has a contribution from e-e interactions (hereafter called $\tau_{ee}^{\mathrm{rel}}$) [21–23]. Thus, if $\tau$ is the momentum relaxation time, $1/\tau = 1/\tau_{e} + 1/\tau_{ee}^{\mathrm{rel}}$, and $1/\tau_{ee}^{\mathrm{rel}} \gg 1/\tau_{e}$. Below we prove that $\tau_{ee}^{\mathrm{rel}} = \tau$; at low temperature, $5/8$ of the quasiparticle lifetime $\tau_{qp}^{\mathrm{rel}}$. Since the latter is always finite, we conclude that the thermal ratio, renormalized by $\tau_{th}/\tau_{e} = (1 + 1/\tau_{ee}^{\mathrm{rel}})^{-1}$, can become arbitrarily small in the hydrodynamic limit. Even if the hydrodynamic regime is not fully established, the existence of two comparable relaxation rates $1/\tau$ and $1/\tau_{ee}^{\mathrm{rel}}$ will lead to significant and observable deviations from the standard WF law. Further, the thermal conductivity is predicted to have a broad maximum as a function of temperature when $\tau_{th} = \tau$; observation of this maximum will provide unequivocal confirmation of our theory.

Perhaps the most surprising outcome of our study is the realization that the violation of the WF law does not depend on the details of the quasiparticle energy dispersion: it is a universal feature of the hydrodynamic regime of Fermi liquids. For an ordinary Galilean-invariant electron gas, the difference between the relaxation time of the charge current and that of the thermal current arises trivially from the fact that the latter is affected by electron-electron collisions, while the former is not, due to momentum conservation. For electrons in doped graphene the reasoning is more subtle but equally compelling. Indeed, as shown in Ref. [2], at low temperature the particle current $J^{(N)}$ is essentially carried by the quasiparticles around the Fermi surface that have momentum of magnitude $|k| \sim k_F$, with small variations of order $T$. To the extent that variations in the magnitude of $k$ can be neglected, the velocity of a quasiparticle, $v \approx k/|k|$, can be approximated as $k/k_F$, which is simply proportional to its momentum $k$. This implies that the total particle current (alias charge current) can be split into two components [24]: a larger one proportional to the total momentum and a smaller one proportional to the temperature $T$. While the former is conserved in electron-electron collisions, the latter is not. This means that the second component is relaxed by electron-electron interactions and, in the absence of a driving field, decays to zero over time. (Note that the nonconservation of the particle current does not contradict the conservation of the particle number, which is of course satisfied.) The conserved component of the particle current dominates at low temperature, and the charge conductivity $\sigma_{th}$ is, therefore, infinite [2,10]. Contrary to $J^{(N)}$, the energy current $J^{(E)}$ of a system with linear energy dispersion is proportional to the momentum; therefore, it is strictly conserved in e-e collisions. Thus, the conductivity associated with energy currents would diverge. However, the thermal current is not $J^{(E)}$ but $J^{(E)} - \mu J^{(N)}$, and we find that $J^{(E)}$ exactly cancels the conserved part of $\mu J^{(N)}$. What survives is the relatively small part of $\mu J^{(N)}$ that is not conserved (and can be relaxed) by e-e collisions [24]. Thus, the thermal current is fully susceptible to decay from electron-electron interactions in doped graphene, and the associated conductivity $\sigma_{th}$ is, therefore, finite [25]. The only difference between doped graphene and the 2DEG is that in the former the collisions affect $J^{(N)}$, while in the latter they affect $J^{(E)}$; the net result for the thermal current is the same [24]. In what follows we apply the theory to doped graphene [3,4], which appears to be particularly promising for the realization and detection of the hydrodynamic regime [2]. However, we emphasize that our results are also valid for Galilean-invariant 2DEGs [26].

The difference between quasiparticle scattering rates in 2DEGs and in graphene does cause quantitative changes in the quasiparticle lifetime $\tau_{qp}^{\mathrm{rel}}$; nonetheless, the low-temperature relation $\tau_{th}^{\mathrm{rel}} = 5\tau_{qp}^{\mathrm{rel}}/8$ is dictated by angular symmetry and remains the same in both cases.

**Model and calculations.**—Particles in graphene are described by the massless Dirac fermion Hamiltonian (for each of the $N_f = 4$ spin and valley flavors; hereafter, $\hbar = 1$) [3,4]

$$\hat{H} = \sum_{k,\lambda} \varepsilon_{k,\lambda} \hat{\psi}^\dagger_{k,\lambda} \hat{\psi}_{k,\lambda} + \frac{1}{2} \sum_{q} v_q (\hat{n}_q \hat{n}_{-q} - \hat{n}_0),$$

where $\psi_{k,\lambda} (\psi^\dagger_{k,\lambda})$ destroys (creates) a particle with momentum $k$ in band $\lambda = \pm$, $\varepsilon_{k,\lambda} = \lambda v_F k$ is the band energy, and $v_F$ is the Fermi velocity. $v_q = (2\pi e^2)/(eq)$ is the nonrelativistic Coulomb interaction, $e$ is the dielectric constant of the environment, and $\hat{n}_q$ is the density operator. The Fermi energy is $E_F = \pm h \nu_F k_F$ (+ for electrons, − for holes), where $k_F = \sqrt{4\pi n/N_f}$ is the Fermi wave vector, and $n$ is the excess carrier density. Because of the particle-hole symmetry of the model, we consider exclusively n-type doping.

Within the linear-response regime, the thermal conductivity is defined in terms of the thermal-current linear response function $\chi_{J^{(E)}J^{(E)}}(q,\omega)$ as

$$\sigma_{th} = \lim_{\omega \to 0} \frac{i}{\omega T} \chi_{J^{(E)}J^{(E)}}(q = 0, \omega),$$

where $\alpha, \beta = x, y$. The calculation of $\sigma_{th}$ closely parallels that of the charge conductivity performed in Ref. [2]. Therefore, in what follows we briefly summarize the main conceptual steps; we provide more technical details in the Supplemental Material [24].

Figure 1 summarizes the diagrammatic calculation of $\chi_{J^{(E)}J^{(E)}}(q,\omega)$, given by the diagram of Fig. 1(a).
solid lines represent Green’s functions dressed by the GW self-energy insertions of Fig. 1(b). The dressed current vertex [solid dot in Fig. 1(a)] is [14–19]

\[
\Lambda^{(0)}_{\alpha\beta}(k_-, \theta, k_+, \theta, \omega_m) = (\epsilon_n + \omega_m/2)J^{\pi}_{\alpha\beta} k_{\theta}. \tag{6}
\]

where \(\epsilon_n\) and \(\omega_m\) are, respectively, fermionic and bosonic Matsubara frequencies [10], \(k_{\pm} = k \pm q/2\), and \(J^{\pi}_{\alpha\beta}\) is the matrix element of the number-current operator [24]. The dressed current vertex \(\Lambda\), represented as a triangle in Fig. 1, is determined by solving the self-consistent Bethe-Salpeter equation [panel (c)] [10].

\[
\sigma_{\text{th}}(\omega) = \frac{\pi^2 k_B^2 T}{3} \frac{\mathcal{D}_{\text{th}}^{(0)}}{-i\omega + 8/(5\tau_{\text{ep}})}. \tag{7}
\]

Notice that the Drude weight remains unrenormalized at this level of approximation. At low temperature [27,28]

\[
\frac{1}{\tau_{\text{ep}}^{\text{th}}} \rightarrow -\frac{4\pi}{3 N_f} (\frac{k_B T}{e_F}) \ln\left(\frac{\zeta k_B T}{e_F}\right), \tag{8}
\]

with \(\zeta = \pi/\sqrt{5}\) [24]. A similar logarithmic reduction of the thermal conductivity was found in Ref. [26] for the case of a Galilean-invariant 2DEG. Note that, at low temperature, \(\tau_{\text{ep}}^{\text{th}}\) is independent of the e-e coupling constant \(\alpha_{ee}\) [Eq. (8)]. In this regime, the dominant contribution to \(\tau_{\text{ep}}^{\text{th}}\) is due to the collinear scattering of quasiparticles, whose phase space diverges [29]. The same phase-space divergence, however, strongly enhances the screening of e-e interactions [30] (the effective interaction is, therefore, independent of the coupling constant). The two effects cancel, and, to the leading nonvanishing order, \(1/\tau_{\text{ep}}^{\text{th}}\) shows only a weak (logarithmic) enhancement.

**Thermal conductivity.**—In Fig. 2 we plot the interacting WF ratio \(L\) [Eq. (3)], in units of \(L_0\) [Eq. (1)], as a function

\[
T=300 \text{ K} \quad \alpha_{ee}=0.5 \quad \alpha_{ee}=0.9 \quad \alpha_{ee}=2.2
\]

\[
n [10^{12} \text{ cm}^{-2}] \quad \alpha_{ee}=0.5 \quad \alpha_{ee}=0.9 \quad \alpha_{ee}=2.2
\]

\[
T [\text{K}] \quad \alpha_{ee}=0.5 \quad \alpha_{ee}=0.9 \quad \alpha_{ee}=2.2
\]

\[
\text{Fig. 2 (color online). Panel (a) shows the renormalized WF ratio } L \text{ as defined in Eq. (3), in units of } L_0 \text{ [Eq. (1)], plotted as a function of the density } n \text{ (in units of } 10^{12} \text{ cm}^{-2}\text{). We fixed the temperature } T = 300 \text{ K, the ratio } \mathcal{D}_{\text{th}}/\mathcal{D}_0 = 1, \text{ and the momentum relaxation time } \tau \text{ to reproduce the electron mobility } \mu_e = 10^5 \text{ cm}^2/(\text{V s}). \text{ We show three curves for different values of the dimensionless coupling constant of e-e interactions, i.e., } \alpha_{ee} = 0.5 \text{ (solid line), } \alpha_{ee} = 0.9 \text{ (short-dashed line), and } \alpha_{ee} = 2.2 \text{ (long-dashed line). Panel (b) is the same as in panel (a) but shown as a function of temperature (in units of K) and for a fixed excess carrier density } n = 10^{12} \text{ cm}^{-2}\text{ (corresponding to a Fermi temperature } T_F \sim 1300 \text{ K). The weak } \alpha_{ee} \text{ dependence of the curves is due to subleading corrections to } \tau_{\text{ep}}^{\text{th}}, \text{ beyond the leading order of Eq. (8).}
\]

\[
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\]
of the carrier density at a fixed temperature $T = 300$ K [Fig. 2(a)] and as a function of temperature for a fixed carrier density $n = 10^{12}$ cm$^{-2}$ [panel (b)], corresponding to a Fermi temperature $T_F \sim 1300$ K. The momentum relaxation time $\tau$ is set to reproduce the electron mobility $\mu_e = 10,000$ cm$^2$/Vs. Even though this value is typical of samples close to the diffusive regime ($\tau \lesssim \tau_{th}^*$), the renormalization of the WF ratio is still remarkable.

In Fig. 3 we show the thermal conductivity of graphene as a function of the temperature for different values of the carrier density. Its downturn marks the onset of the hydrodynamic regime of the electron liquid. Indeed, $\sigma_{th} = L_0\sigma_e T / (1 + \tau / \tau_{th}^{*e})$, and $1/\tau_{th}^{*e}$ grows as $-T^2 \ln(T)$ for $k_BT \ll \epsilon_F$. The initial linear-in-$T$ (diffusive) behavior is replaced, as the temperature increases, by a slowly decreasing function of $T$. The position of the maximum of $\sigma_{th}$ can be determined from the analytical approximation of our formula, it is found, for any given density, as the point where the dashed line intercepts the solid line.

The renormalization of the thermal Drude weight.—The collisionless kinetic equation for the distribution function of quasiparticles in the presence of a small temperature gradient that oscillates in time and space with a frequency $\omega$ and wave vector $q$ is [10,24,33]

$$\langle q \cdot v^*_k - \omega \rangle n_{1,k} - n_0' (\xi_k^* v^*_k) \cdot \left[ q \sum_{k'} f_{k,k'} n_{1,k'} + \xi_k^* \nabla_T T \right] = 0,$$

where $n_{1,k} \equiv n_{1,k}(q, \omega)$ is the nonequilibrium correction to the distribution function, $\xi_k^*$ is the quasiparticle velocity measured from the chemical potential $\mu^*$, and $v^*_k = \nabla_k \xi_k^*$ is the quasiparticle velocity. $f_{k,k'}$ is the Landau interaction function [10], while $n_0'(\xi_k^*)$ is the derivative of the Fermi distribution function with respect to $\xi_k^*$. To $O(q^2/\omega^2)$, Eq. (9) is solved by the ansatz $n_{1,k}(q, \omega) = -n_0' (\xi_k^* A_k(q, \omega) \xi_k^*)$, where $A_k(q, \omega)$ is to be determined. The first term in the square brackets of Eq. (9) vanishes at order $T^2$, due to the cancellation of contributions from opposite sides of the Fermi surface (the $\xi_k^*$ factor is antisymmetric) [10,24,32]. Therefore, $A_k(q, \omega) = (q \cdot v^*_k) (\xi_k^* \nabla_T T / (\omega T))$. The induced variation of the entropy is $[34] \delta S(q, \omega) = \sum_k n_{1,k} \xi_k^* (\xi_k^* \nabla_T T)$. Since $\delta S(q, \omega) = (\pi^2 k_B^2 T^3 / 3 \omega^2)$ we find $\sigma_{th} = N_j k_F v^*_F / (4\pi \hbar)$. Many-body effects enter only through the renormalized Fermi velocity $v^*_F$. On the other hand [35,36], $\sigma_{th} = N_j k_F v^*_F (1 + F_1) / (4\pi \hbar)$, where $F_1$ is the first spin-symmetric Landau parameter [10]. Thus, at odds with [14,15], the WF ratio is further renormalized by a factor $\sigma_{th} / \sigma_c = (1 + F_1)^{-1}$. In both 2DEG and graphene, $1 + F_1 \sim 1$ in a broad range of values of the strength of $e-e$ interactions [10,36]. This leaves $\tau_{th}/\tau_c$ as the main factor controlling the value of the WF ratio.

Summary and conclusions.—According to the Wiedemann-Franz law the charge ($\sigma_c$) and thermal ($\sigma_{th}$) conductivities of a Fermi liquid satisfy the relation $\sigma_{th} = L_0 \sigma_e T$, where the Lorenz number $L_0 = \pi^2 k_B^2 / (3e^2)$. As they affect the two conductivities in different ways, $e-e$ interactions can dramatically change the value of the WF ratio. In Ref. [2] we showed that at low temperature they do not contribute to the relaxation of charge currents. Therefore, $\sigma_c$ diverges as the momentum relaxation time $\tau \to \infty$. On the contrary, the thermal conductivity—Eq. (7)—is always finite as long as $T \neq 0$. This result manifestly violates the WF law: the WF ratio is renormalized by a factor $R = (1 + \tau / \tau_{th}^{*e})^{-1}$.

Even though the violation of the WF law is a universal feature of the hydrodynamic regime of the electron liquid, we applied our theory to doped graphene, which is particularly promising for the realization of this regime. For micrometer-sized samples and typical doping concentrations, hydrodynamics is expected to be applicable in a temperature window between 50 and 300 K [2]. We showed that $R$ is quite small even for samples close to the diffusive regime. For example, for an electron density $n \sim 10^{12}$ cm$^{-2}$, a mobility $\mu_e \sim 100,000$ cm$^2$/Vs, and $T = 300$ K, $R \sim 1/2$. In high-quality sheets, which reach mobilities of the order of $\sim 10^5 - 10^6$ cm$^2$/Vs, the violation is expected to be much stronger.

The factor $R$ is a decreasing function of the temperature. Therefore, $\sigma_{th} = TR \sigma_c$, for finite $\tau$, is a nonmonotonic function of $T$ (Fig. 3). At low temperature, where the WF law is applicable, it is a linear function of $T$. As $T$ increases, $\tau_{th}^{*e} \lesssim \tau$ and $\sigma_{th}$ starts decreasing with the temperature. Eventually, electron-phonon interactions reverse this behavior [31]. Thus, $\sigma_{th}$ shows first a maximum, when the hydrodynamic regime is established, and then a minimum, when it is no longer in force. Both features can be
experimentally tested and constitute evidence for hydrodynamic thermal conduction.

This work was supported in part by DOE Grant No. DE-FG02-05ER46203 and by a Research Board Grant at the University of Missouri.

*principia@missouri.edu

[34] When \( J^{(N)} = 0 \), the thermal and energy current coincide. One might, therefore, expect \( J^{(0)} \) to be conserved (in graphene), and \( \sigma_n \) to be infinite. However, to maintain \( J^{(N)} = 0 \), an electric field (which opposes the particle flow) must inevitably build up. This in turn breaks the conservation of the energy current (i.e., of the momentum); therefore, \( J^{(E)} = -\sigma_n \nabla \cdot \vec{J} \).
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