Wiedemann-Franz law in scattering theory revisited

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The violation of Wiedemann-Franz (WF) law has been widely discussed in quantum transport experiments as an indication of deviation from Fermi-liquid behavior. The conventional form of WF law is only concerned with the transmission function at Fermi-level which, however, vanishes in many practical situations. We reinvestigate the WF law in noninteracting quantum systems with vanishing zero energy transmission and report a universal number $21/5$ as an upper bound of Lorenz ratio $\mathcal{R}$ in weakly energy-dependent scattering theory. We provide different experimental realizations for the observation of $\mathcal{R} = 21/5$ namely the transport setups with graphene, the multilevel quantum dot and double quantum dot. The reported universal Lorenz ratio paves an efficient way of experimentally obtaining the informations about the associated quantum interferences in the system. Our work also provides enough evidence which concludes that the violation of WF law does not necessarily imply the non-Fermi-liquid nature of underlying transport processes; equally, the Fermi-liquid transport characteristics cannot be concluded by an observed validation of WF law.

Rapid development of quantum technology has stimulated a plethora of quantum transport experiments [1]. Thermoelectric phenomena is one of the common transport measurements of interests in chemical potentials $\mu$. The left (L) and right (R) reservoirs are in equilibrium, separately, at temperatures $T$. The left ($L_h$) and right ($L_c$) current are expressed in terms of the transport integrals, respectively. The heat current $I_h$ and charge current $I_c$ flow across the impurity caused by the temperature gradient $\Delta T \equiv T_L - T_R$ and the mismatch of chemical potentials $\Delta \mu \equiv \mu_L - \mu_R$. The charge and the heat currents in the linear response theory are connected by the Onsagar relations [4, 5] which in atomic units read

$$\begin{pmatrix} I_c \\ I_h \end{pmatrix} = \begin{pmatrix} L_{11} & L_{12} \\ L_{21} & L_{22} \end{pmatrix} \begin{pmatrix} \Delta V \\ \Delta T \end{pmatrix} .$$

(1)

The Onsagar transport coefficients $L_{ij}$ in Eq. (1) provide all the thermoelctric measurements of interests in linear response regime [6]. To this end, we set the transport integrals relating the Onsagar coefficients

$$\mathcal{L}_n = \frac{1}{4T} \int_{-\infty}^{\infty} \frac{d\varepsilon}{\cosh^2 \left( \frac{\varepsilon}{2T} \right)} \mathcal{T}(\varepsilon, T), \quad n = 0, 1, 2. \quad \text{(2)}$$

Here $T$ is the reference temperature and $\mathcal{T}(\varepsilon, T)$ is the energy and temperature dependent spectral function (the transmission coefficient).

The transport coefficients characterizing the charge current are expressed in terms of the transport integrals, namely, $L_{11} = \mathcal{L}_0$ and $L_{12} = -\mathcal{L}_1 / T$ [7]. In addition $L_{12}$ and $L_{21}$ are related by the Onsagar reciprocity relation and the coefficient $L_{22}$ relates the thermal conductance $\mathcal{L}_0$ with the electrical conductance $\mathcal{L}_1$. The corresponding Lorenz ratio simply implies that the transmission function $\mathcal{T}(\varepsilon, T)$ of the transport mechanism responsible for heat and charge currents are fundamentally the same [6].

$$\mathcal{L}_n = \mathcal{L}_1 = \mathcal{L}_2 \equiv \frac{S_h}{T_0} T. \quad \text{(3)}$$

In addition, the Wiedemann-Franz (WF) law connects the electronic thermal conductance $\mathcal{K}$ to the electrical conductance $G$ in low temperature regime of a macroscopic sample by an universal constant, the Lorenz number $L_0$, defined as $L_0 \equiv \mathcal{K}/\mathcal{G}T = \pi^2/3$. The constant value of Lorenz number simply implies that the transport mechanisms responsible for heat and charge currents are fundamentally the same [6]. The possible deviation from WF law at the nano scale has been accounted for by considering the Lorenz ratio [8].

$$\mathcal{R} \equiv \frac{L(T)}{L_0} = \frac{3}{(\pi T)^2} \left[ \mathcal{L}_2 - \frac{\mathcal{L}_1}{\mathcal{L}_0} \right]^2 ,$$

(4)

the deviation of $\mathcal{R}$ from unity amounts the violation of WF law. Although the WF law is expected to be violated strongly at the nano scale, surprisingly it works quantitatively well for $T \to 0$ even for some interacting systems with both Fermi-liquid and non Fermi-liquid correlations such as the Kondo correlated systems [6, 9, 10]. This suggests that the Fermi-liquid nature of transport can not be concluded by the observed validation of WF law. In addition, it might be also possible that the quantum transport in Fermi-liquid regime (for both interacting and noninteracting systems) strongly violates the WF law. To explore this possibility in detail, we restrict ourselves by considering the noninteracting systems described by the scattering theory. To this end, we sketch briefly the main assumption behind the derivation of original WF law and provide the logical reason of relaxing such assumption which eventually results in the different Lorenz ratio from the conventional one. We consider the transmission function satisfying the condition [11, 12]

$$0 \leq \mathcal{T}(\varepsilon, T) \leq N ,$$

(5)
with $N$ being the number of transverse conduction modes. In addition, for the system modelled by the scattering theory the transmission function is merely energy dependent, the temperature comes solely from the Fermi-function $T(\varepsilon, T) = T(\varepsilon)$ [11]. The fundamental assumption of obtaining WF law at nano scale is to consider the smooth transmission function such that

$$T(\varepsilon) \simeq T_0 + \frac{\partial T(\varepsilon)}{\partial \varepsilon} \bigg|_{\varepsilon=0} \varepsilon + \cdots,$$  

(6)

where $T_0$ is the zero energy transmission function. Assuming the unitary condition $T(\varepsilon = 0) \rightarrow 1$, one can readily see that $\mathcal{L}_0 = 1$, $\mathcal{L}_2 = (\pi T)^2/3$ and $\mathcal{L}_1 \simeq 0$ satisfying the condition provided by WF law $L_0 = \pi^2/3$.

Toward the urge for enhancing thermoelectric performance, the concept of ideal energy filters with an energy-dependent transmission function $T(\varepsilon) \propto \Gamma(\varepsilon - \varepsilon_0)$ has been suggested, with $\varepsilon_0$ being the position of single level contributing the transport and $\Gamma$ is some energy scale of the system [13]. This delta function transmission has then realized being of not much practical application since this abruptly reduces the efficiency at maximum power [14]. As an overcome of this difficulty, two-level systems have been proposed where the quantum interference can substantially improve the maximum thermoelectric power and the efficiency at maximum power [15]. We note that the quantum interference between different conduction channels is quite common phenomena at the nano scale including the strongly correlated systems [16–18]. In the presence of two or more interfering transport channel, the fundamental assumption of WF law that $T(\varepsilon = 0) \rightarrow 1$ is completely violated, rather the destructive interference results in $T(\varepsilon = 0) \rightarrow 0$. In addition, the system might posses particle-hole (PH) symmetry on the top of level interference, that is $T(\varepsilon) = T(-\varepsilon)$. These two properties can also be observed in graphene since the associated unusual band structure [19–22].

For these two different practical cases presented above, the transport through the multi-level quantum dot (QD) or multi-QD and the graphene, the crude assumption of smooth transmission function given by Eq. (6) is not of practical use. In this case one has to rather go beyond the first order expansion of transmission coefficient (see the following section). The fundamental question of paramount importance, both theoretical and experimental interest, then would be what about the WF law for the systems possessing $T(\varepsilon = 0) \rightarrow 0$ and $T(\varepsilon) = T(-\varepsilon)$. More generally, what is the connection between the electronic thermal conductance and the electrical conductance for the systems with graphene like transmission?

To unveil the form of WF law with vanishing zero energy transmission, we consider a generic noninteracting system which is well described within the scattering theory. Since many experimental systems posses rather weak energy dependence of their transmission [1], we express the transmission function into the Taylor series in energy

$$T(\varepsilon) = T_0 + T_1 \frac{\varepsilon}{\Gamma} + T_2 \left(\frac{\varepsilon}{\Gamma}\right)^2 + \cdots,$$  

(7)

where $T_0$ is the zero energy transmission and $T_{1,2}$ are the expansion coefficients. The truncation of the series Eq. (7) at the second order is indeed the reasonable approximation for most of the practical situations unless the exotic situation with $T_{0,1,2} = 0$ is encountered. Although, the simultaneous vanishing of $T_0$, $T_1$ and $T_2$ is very unlikely to be the case of real experiment, we will revisit this case later and for now we truncate the series Eq. (7) at second order. The Lorenz ratio obtained from the Eq. (7) then reads

$$\mathcal{R} = \frac{3}{5} \left[ 7 - \frac{16T_0}{\pi^2 T^2 T_2 + 3T_0} + \frac{5\pi^2 T^2 T_2^2}{\pi^2 T^2 T_2 + 3T_0} \right] + \cdots,$$  

(8)

with $T \equiv T/\Gamma$. In addition we have used the Sommerfeld integrals $J_i = \frac{1}{\pi T} \int_{-\infty}^{\infty} d\varepsilon e^{i\varepsilon j} \cosh(\varepsilon / 2T)$ with $J_0 = 1$, $J_2 = (\pi T)^2/3$ and $J_4 = 7/15 \times (\pi T)^4$ and vanishing odd integrals. For single level transport where $T_0 \neq 0$, one can expand Eq. (8) in low temperature limit to get

$$\mathcal{R}|_{T_0 \neq 0} = 1 + (\pi T)^2 \left[ \frac{16 T_2}{15 T_0} - \frac{1}{3} \left( \frac{T_4}{T_0} \right)^2 \right] + \cdots,$$  

(9)

which immediately verifies the WF for $T \rightarrow 0$. For the systems possessing the special property $T_0 = T(\varepsilon \rightarrow 0) = 0$ we have, however, quite different form of Lorenz ratio [23]

$$\mathcal{R}|_{T_0 = 0} = 21/5 - S_0^2/L_0 + \cdots$$  

(10)

In scattering theory (or systems described by the Fermi-liquid paradigm) thermopower posses a linear temperature scaling behavior which gets vanishes for $T \rightarrow 0$. Therefore the zero temperature limit of Lorenz ratio reads

$$\mathcal{R}|_{T_0 = 0, T \rightarrow 0} = \frac{21}{5}, \quad L = \frac{7\pi^2}{5}.$$  

(11)

This results suggests that the presence of quantum interference or a system with rather special geometry with vanishing zero energy transmission strongly violates the WF law. In this limit the Lorenz ratio attains the temperature independent universal number 21/5.

The prediction of universal number 21/5 for the Lorenz ratio originated mainly form the assumption of vanishing zero energy transmission $T_0 = 0$. The contribution of $T_1$ to Lorenz ratio eventually vanishes even if the PH symmetry is explicitly broken by taking the limit of $T \rightarrow 0$ (thermopower possesses a linear scaling with temperature). Therefore the consideration of only non-vanishing element $T_2$ is the sole reason of observed universal Lorenz ratio. Although it is very unlikely that a real system possesses the property of $T_{0,1,2} \rightarrow 0$, it
A QD with two-levels $\varepsilon_i$ ($i=1,2$) is tunneled coupled to the left (L) and right (R) electronic reservoirs. The symbol $\Gamma_{\alpha i}$ ($\alpha=L, R$) stands for the coupling strength between the lead $\alpha$ and the energy level $\varepsilon_i$ in the QD. We assume that the tunneling does not mix the two levels which is valid in the noninteracting transport setups. Lower panel: The vanishing zero energy transmission function in multi-level QD with properly chosen coupling strengths $\Gamma_{\alpha i}$ and level positions $\varepsilon_i$.

is also worth of commenting on this rather exotic situation. In this case the Eq. (7) rather starts from the third order term in the energy expansion and the Lorenz ratio attains rather a big number of 465/49 at the limit of $T \to 0$. With these observations, one can conclude that there is no unique upper bound for the Lorenz ratio in scattering theory. Nevertheless, most of the experimental systems have reasonably weak energy dependence of their transmission and hence the predictions made in our work based on Eq. (7) have rather broad domain of applicability.

The graphene based transport would be one of the simplest example to experimentally verify the predicted universal number for Lorenz ratio [22]. In the following we further corroborate on the ubiquitouslyness of predicted Lorenz ratio in quantum experiments and study the more general behavior of Eq. (10). To this end, we consider a two-level quantum dot tunnel coupled to two conducting leads (reservoirs) as shown in Fig. 1. In addition, for the activation of quantum interference we assume that the two levels $\varepsilon_{1,2}$ couple with different parity to the leads and their coupling strengths differ by a factor $a^2$, $\Gamma_{L1} = \Gamma_{R1} = \Gamma$, $\Gamma_{L2} = \Gamma_{R2} = a^2\Gamma$ (see Ref. [14] for detail). In this case the transmission function $T_M(\varepsilon, \varepsilon_1, \varepsilon_2)$ reads

$$T_M(\varepsilon, \varepsilon_1, \varepsilon_2) = \Gamma^2 \left| \frac{1}{\varepsilon - \varepsilon_1 + i\Gamma} - \frac{a^2}{\varepsilon - \varepsilon_2 + ia^2\Gamma} \right|^2. \quad (12)$$

The zero energy transport will be nullified for a particular choice of $\varepsilon_2 = a^2\varepsilon_1$, that is $T_M(\varepsilon \to 0, \varepsilon_1, \varepsilon_2 \to a^2\varepsilon_1) \to 0$. For this case of $\varepsilon_2 = a^2\varepsilon_1$ we recast the transmission function into the form

$$T_M(\varepsilon, \varepsilon_0) = \frac{(a^2 - 1)^2 \varepsilon^2}{(1 + (\varepsilon - \varepsilon_0)^2)(a^4(1 + \varepsilon_0^2) - 2a^2\varepsilon_0 + \varepsilon^2)}, \quad (13)$$

where energies are expressed in the unit of $\Gamma$, that is $\varepsilon/\Gamma \equiv \varepsilon$ and $\varepsilon_1/\Gamma = \varepsilon_0/\Gamma \equiv \varepsilon_0$. For this transmission function, in the limit of $T \to 0$ we recover again the universal Lorenz ratio of $21/5$ irrespective of the parameters $\varepsilon_0$ and $a$. This can be easily seen by considering the above transmission shape at the strong coupling regime, $(\varepsilon, \varepsilon_0) \ll 1$, which within the lowest order expansion reads

$$T_M(\varepsilon) = (a^2 - 1)^2 \varepsilon^2/a^4 + \cdots \quad (14)$$

The violation of WF law with varying temperature for the two level system consider above is as shown in Fig. 2, which apparently reaches the universal number of $21/5$ for the Lorenz ratio at the limit of $T \to 0$.

For further discussion on the experimental verification of Lorenz ratio, we consider a double quantum dot setup with equal tunneling amplitudes $\Gamma$ and respective energy levels $\varepsilon_{1,2}$ as shown in Fig. 3. Tuning the system in the regime of $\varepsilon_1 = -\varepsilon_2 = \varepsilon_0$, the double quantum dot (DQD) transmission function $T_{DQD}(\varepsilon)$ reads [24–28]

$$T_{DQD}(\varepsilon) = \frac{1}{\sqrt{1 - \varepsilon_0^2}} \left[ \frac{\Omega^2}{\varepsilon^2 + \Omega^2} - \frac{\Omega^2}{\varepsilon^2 + \Omega'^2} \right], \quad \Omega_{\pm} = 1 \pm \sqrt{1 - \varepsilon_0^2},$$

where we expressed the energy in the unit of $\Gamma$ satisfying the condition $1 > \varepsilon_0/\Gamma \equiv \varepsilon_0$. Since the low energy expansion of the function $T_{DQD}(\varepsilon)$ has the simple form $T_{DQD}(\varepsilon) = 4(\varepsilon/\varepsilon_0)^2 + \cdots$, the universal number of $21/5$ for the Lorenz ratio is exactly recovered at $T \to 0$.

We note that one of the fundamental factor validating above discussions is also the quantum interference effects. Testing our predictions with a multi-level QD, therefore, only needs the proper tuning of parity factor $a$ as seen from the Eq. (13). Besides, for the study of quantum interferences in DQD setup, the relative phases of the tunneling amplitudes may represent an
Aharonov-Bohm flux [29]. The detail study of the WF law in DQD setup with Aharonov-Bohm (AB) geometry is left for the future work. It can be also the case that dot structure possesses the small area so that it does not really generate the AB-phase [28]. Given that, however, the destructive interference (anti-resonance) in DQD setup resulting in $T_0 \rightarrow 0$ needed to verify our predictions can be achieved by the appropriate choice of the gate voltage.

For the propose of strengthening our prediction, in the following we formulate the multi-level transport description using more general scattering matrix formulation. In two terminal transport description, the electron operators in the left and right electronic reservoirs in the parallel configuration. $\Gamma_\alpha i$ represents the coupling strength between the lead $\alpha$ and the $i$th QD. Lower panel: The energy dependence of the transmission function for the DQD setup in the upper panel which exhibits the anti-resonance provided the specific choice of parameters satisfying $\Gamma_\alpha i = \Gamma$ and $\epsilon_1 = -\epsilon_2 = \epsilon_0$.

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