


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 the 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 the 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 information 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.

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Rapid development of quantum technology has stimulated a plethora of quantum transport experiments [1]. Thermoelectric phenomena is one of the common transport measurements of nanoscale systems, such as in quantum dots, carbon nanotubes, and quantum point contacts [2,3].

A prototypical thermoelectric experiment at the nanoscale consists of a two-terminal device, namely, a quantum impurity tunnel coupled to two conducting reservoirs. The left (L) and right (R) reservoirs are in equilibrium, separately, at temperatures T_γ ($\gamma = L, R$) and chemical potentials μ_γ , 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 $\Delta V \equiv \mu_L - \mu_R$. The charge and the heat currents in the linear response theory are connected by the Onsager relations [4,5], which in atomic units read

$$\begin{pmatrix} I_c \\ I_h \end{pmatrix} = \begin{pmatrix} L_{11} & L_{12} \\ L_{21} & L_{22} \end{pmatrix} \begin{pmatrix} \Delta V \\ \Delta T \end{pmatrix}. \quad (1)$$

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

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

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 Onsager reciprocity relation, and the coefficient L_{22} relates the thermal conductance [1]. While the electrical

conductance is related with \mathcal{L}_0 alone, the thermopower is usually defined as $\mathcal{S}_{th} = \mathcal{L}_1/\mathcal{L}_0 T$. The thermal conductance \mathcal{K} reads

$$\mathcal{K} = \frac{1}{T} \left[\mathcal{L}_2 - \frac{\mathcal{L}_1^2}{\mathcal{L}_0} \right]. \quad (3)$$

In addition, the Wiedemann-Franz (WF) law connects the electronic thermal conductance \mathcal{K} to the electrical conductance G in the low-temperature regime of a macroscopic sample by a universal constant, the Lorenz number L_0 , defined as $L_0 \equiv \mathcal{K}/GT = \pi^2/3$. 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 nanoscale has been accounted for by considering the Lorenz ratio [8],

$$\mathcal{R} \equiv \frac{L(T)}{L_0} = \frac{3}{(\pi T)^2} \left[\frac{\mathcal{L}_2}{\mathcal{L}_0} - \left(\frac{\mathcal{L}_1}{\mathcal{L}_0} \right)^2 \right], \quad (4)$$

where the deviation of \mathcal{R} from unity amounts to the violation of WF law. Although the WF law is expected to be violated strongly at the nanoscale, surprisingly, it works quantitatively well for $T \rightarrow 0$ 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 cannot be concluded by the observed validation of WF law. In addition, it might be also possible that the quantum transport in the Fermi-liquid regime (for both interacting and noninteracting systems) strongly violates the WF law. To explore this possibility in detail, we restrict ourselves to 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 for relaxing such an

assumption, which eventually results in a different Lorenz ratio from the conventional one. We consider the transmission function satisfying the condition [11,12]

$$0 \leq \mathcal{T}(\varepsilon, T) \leq N, \quad (5)$$

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

$$\mathcal{T}(\varepsilon) \simeq \mathcal{T}_0 + \left. \frac{\partial \mathcal{T}(\varepsilon)}{\partial \varepsilon} \right|_{\varepsilon=0} \varepsilon + \dots, \quad (6)$$

where \mathcal{T}_0 is the zero-energy transmission function. Assuming the unitary condition $\mathcal{T}(\varepsilon = 0) \rightarrow 1$, one readily sees 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 $\mathcal{T}(\varepsilon) \propto \Gamma \delta(\varepsilon - \varepsilon_0)$ has been suggested, with ε_0 being the position of a single level contributing the transport and Γ is some energy scale of the system [13]. This δ function transmission has then been realized as not being useful for much practical application, since this abruptly reduces the efficiency at maximum power [14]. To overcome this difficulty, two-level systems have been proposed where the quantum interference can substantially improve the maximum thermoelectric power and efficiency at maximum power [15]. We note that the quantum interference between different conduction channels is a quite common phenomena at the nanoscale, including in strongly correlated systems [16–18]. In the presence of two or more interfering transport channels, the fundamental assumption of WF law that $\mathcal{T}(\varepsilon = 0) \rightarrow 1$ is completely violated, rather, the destructive interference results in $\mathcal{T}(\varepsilon = 0) \rightarrow 0$. In addition, the system might possess particle-hole (PH) symmetry on top of level interference, that is, $\mathcal{T}(\varepsilon) = \mathcal{T}(-\varepsilon)$. These two properties can also be observed in graphene with the associated unusual band structure [19–22].

For these two different practical cases presented above, the transport through the multilevel quantum dot (QD) or multi-QD and the graphene, the crude assumption of a 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 the transmission coefficient (see the following section). The fundamental question of paramount importance, of both theoretical and experimental interest, then would be what about the WF law for the systems possessing $\mathcal{T}(\varepsilon = 0) \rightarrow 0$ and $\mathcal{T}(\varepsilon) = \mathcal{T}(-\varepsilon)$. More generally, what is the connection between the electronic thermal conductance and the electrical conductance for the systems with graphenelike 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 possess rather weak energy dependence of their transmission [1], we express the

transmission function in the Taylor series in energy

$$\mathcal{T}(\varepsilon) = \mathcal{T}_0 + \mathcal{T}_1 \frac{\varepsilon}{\Gamma} + \mathcal{T}_2 \left(\frac{\varepsilon}{\Gamma} \right)^2 + \dots, \quad (7)$$

where \mathcal{T}_0 is the zero-energy transmission and $\mathcal{T}_{1,2}$ 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 $\mathcal{T}_{0,1,2} = 0$ is encountered. Although the simultaneous vanishing of \mathcal{T}_0 , \mathcal{T}_1 , and \mathcal{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 Eq. (7) then reads

$$\mathcal{R} = \frac{3}{5} \left[7 - \frac{16\mathcal{T}_0(\pi^2 \bar{T}^2 \mathcal{T}_2 + 3\mathcal{T}_0) + 5\pi^2 \bar{T}^2 \mathcal{T}_1^2}{(\pi^2 \bar{T}^2 \mathcal{T}_2 + 3\mathcal{T}_0)^2} \right] + \dots \quad (8)$$

with $\bar{T} \equiv T/\Gamma$. In addition, we have used the Sommerfeld integrals $\mathcal{I}_j \equiv \frac{1}{4T} \int_{-\infty}^{\infty} d\varepsilon \frac{\varepsilon^j}{\cosh^2(\varepsilon/2T)}$ with $\mathcal{I}_0 = 1$, $\mathcal{I}_2 = (\pi T)^2/3$, and $\mathcal{I}_4 = 7/15 \times (\pi T)^4$ and vanishing odd integrals. For single-level transport where $\mathcal{T}_0 \neq 0$, one can expand Eq. (8) in the low-temperature limit to get

$$\mathcal{R}|_{\mathcal{T}_0 \neq 0} = 1 + (\pi \bar{T})^2 \left[\frac{16}{15} \frac{\mathcal{T}_2}{\mathcal{T}_0} - \frac{1}{3} \left(\frac{\mathcal{T}_1}{\mathcal{T}_0} \right)^2 \right] + \dots, \quad (9)$$

which immediately verifies the WF for $T \rightarrow 0$. For the systems possessing the special property $\mathcal{T}_0 = \mathcal{T}(\varepsilon \rightarrow 0) = 0$, we have, however, a quite different form of Lorenz ratio [23]:

$$\mathcal{R}|_{\mathcal{T}_0=0} = 21/5 - \mathcal{S}_{\text{th}}^2/L_0 + \dots \quad (10)$$

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

$$\mathcal{R}|_{\mathcal{T}_0=0, T \rightarrow 0} = \frac{21}{5}, \quad L = \frac{7\pi^2}{5}. \quad (11)$$

This result 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 from the assumption of vanishing zero-energy transmission $\mathcal{T}_0 = 0$. The contribution of \mathcal{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 nonvanishing element \mathcal{T}_2 is the sole reason for the observed universal Lorenz ratio. Although it is very unlikely that a real system possesses the property of $\mathcal{T}_{0,1,2} \rightarrow 0$, it is also worth commenting on this rather exotic situation. In this case Eq. (7) rather starts from the third-order term in the energy expansion and the Lorenz ratio attains a rather a big number of 465/49 at the limit of $T \rightarrow 0$. 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

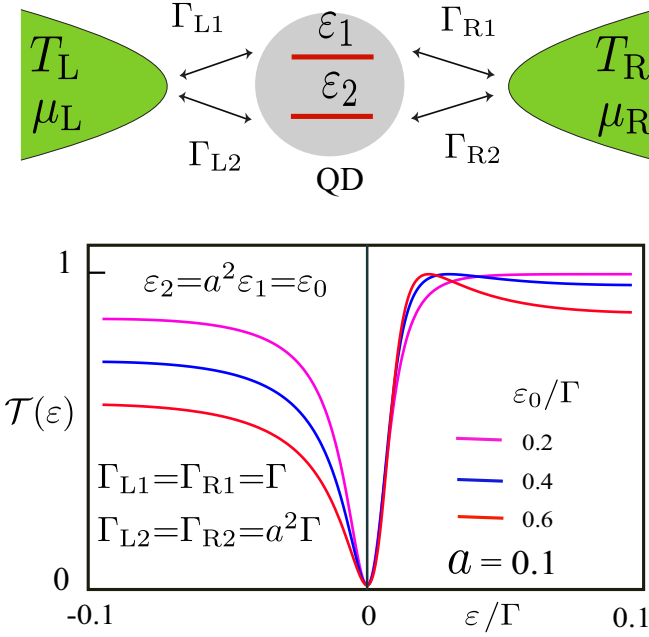


FIG. 1. Upper panel: The multilevel QD transport setup. A QD with two levels ε_i ($i=1, 2$) is tuned coupled to the left (L) and right (R) electronic reservoirs. The symbol $\Gamma_{\alpha i}$ ($\alpha=L, R$) stands for the coupling strength between the lead α and the energy level ε_i in the QD. We assume 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 multilevel QD with properly chosen coupling strengths $\Gamma_{\alpha i}$ and level positions ε_i .

predictions made in our work based on Eq. (7) have a rather broad domain of applicability.

Graphene-based transport would be one of the simplest examples to experimentally verify the predicted universal number for the Lorenz ratio [22]. In the following we further corroborate the ubiquitousness of the 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 details). In this case the transmission function $\mathcal{T}_M(\varepsilon, \varepsilon_1, \varepsilon_2)$ reads

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

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

$$\mathcal{T}_M(\tilde{\varepsilon}, \tilde{\varepsilon}_0) = \frac{(a^2 - 1)^2 \tilde{\varepsilon}^2}{[1 + (\tilde{\varepsilon} - \tilde{\varepsilon}_0)^2][a^4(1 + \tilde{\varepsilon}_0^2) - 2a^2\tilde{\varepsilon}\tilde{\varepsilon}_0 + \tilde{\varepsilon}^2]}, \quad (13)$$

where energies are expressed in the unit of Γ , that is, $\varepsilon/\Gamma \equiv \tilde{\varepsilon}$ and $\varepsilon_1/\Gamma = \varepsilon_0/\Gamma \equiv \tilde{\varepsilon}_0$. For this transmission function, in the limit of $T \rightarrow 0$ we recover again the universal Lorenz ratio

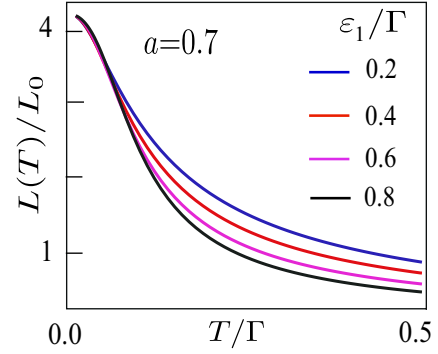


FIG. 2. The variation of Lorenz ratio $\mathcal{R} = L(T)/L_0$ with temperature T for given positions of energy level ε_1 in the two-level quantum transport setup at fixed parity parameter a [see Eq. (13) and following text for details].

of $21/5$ irrespective of the parameters $\tilde{\varepsilon}_0$ and a . This can be easily seen by considering the above transmission shape at the strong-coupling regime, $(\tilde{\varepsilon}, \tilde{\varepsilon}_0) \ll 1$, which within the lowest order expansion reads

$$\mathcal{T}_M(\tilde{\varepsilon}) = (a^2 - 1)^2 \tilde{\varepsilon}^2 / a^4 + \dots \quad (14)$$

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

For further discussion on the experimental verification of the Lorenz ratio, we consider a double-quantum-dot setup with equal tunneling amplitudes Γ 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 $\mathcal{T}_{DQD}(\varepsilon)$ reads [24–28]

$$\mathcal{T}_{DQD}(\tilde{\varepsilon}) = \frac{1}{\sqrt{1 - \tilde{\varepsilon}_0^2}} \left[\frac{\Omega_+^2}{\tilde{\varepsilon}^2 + \Omega_+^2} - \frac{\Omega_-^2}{\tilde{\varepsilon}^2 + \Omega_-^2} \right], \quad \Omega_{\pm} = 1 \pm \sqrt{1 - \tilde{\varepsilon}_0^2},$$

where we expressed the energy in the unit of Γ satisfying the condition $1 > \varepsilon_0/\Gamma \equiv \tilde{\varepsilon}_0$. Since the low-energy expansion of the function $\mathcal{T}_{DQD}(\tilde{\varepsilon})$ has the simple form $\mathcal{T}_{DQD}(\tilde{\varepsilon}) = 4(\tilde{\varepsilon}/\tilde{\varepsilon}_0)^2 + \dots$, the universal number of $21/5$ for the Lorenz ratio is exactly recovered at $T \rightarrow 0$.

We note that one of the fundamental factors validating the above discussions is also the quantum interference effects. Testing our predictions with a multilevel QD, therefore, only needs the proper tuning of parity factor a as seen from Eq. (13). Besides, for the study of quantum interferences in the DQD setup, the relative phases of the tunneling amplitudes may represent an Aharonov-Bohm flux [29]. The detailed study of WF law in the DQD setup with Aharonov-Bohm (AB) geometry is left for future work. It can be also the case that the dot structure possesses the small area so that it does not really generate the AB phase [28]. Given that, however, the destructive interference (antiresonance) in the DQD setup resulting in $\mathcal{T}_0 \rightarrow 0$ needed to verify our predictions can be achieved by the appropriate choice of gate voltage.

For the propose of strengthening our prediction, in the following we formulate the multilevel transport description using a more general scattering matrix formulation. In the

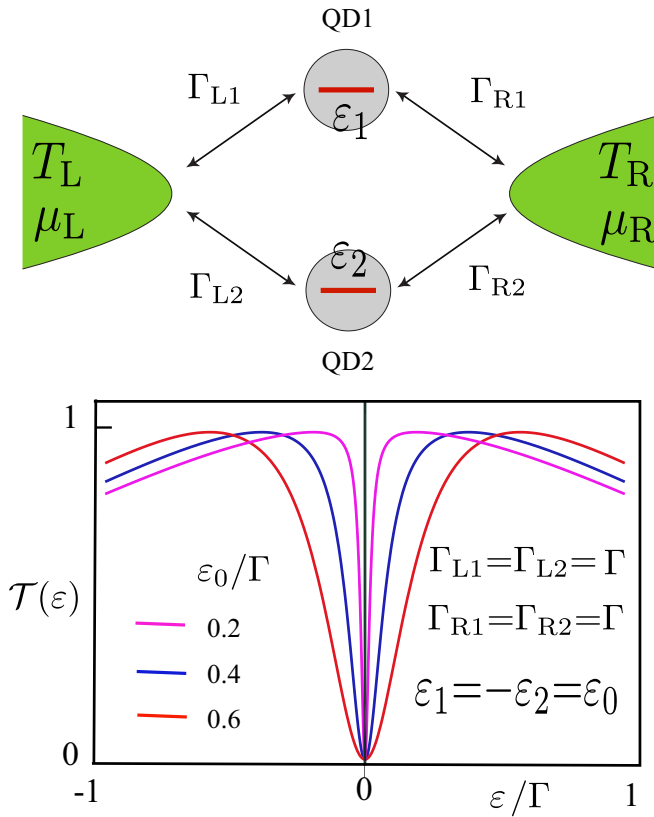


FIG. 3. Upper panel: The DQD transport setup. Two QDs (QD1 and QD2) each with single energy level ε_i ($i = 1, 2$) are tunnel coupled to the left and right electronic reservoirs in the parallel configuration. $\Gamma_{\alpha i}$ represents the coupling strength between the lead α and the i th QD. Lower panel: The energy dependence of the transmission function for the DQD setup in the upper panel, which exhibits the antiresonance, provided the specific choice of parameters satisfy $\Gamma_{\alpha i} = \Gamma$ and $\varepsilon_1 = -\varepsilon_2 = \varepsilon_0$.

two-terminal transport description, the electron operators in the left and right leads $c_{L,R}$ can always be rotated using the Glazman-Raikh rotation [30] (we consider the symmetric lead-dot coupling):

$$\begin{pmatrix} b_e \\ b_o \end{pmatrix} = \mathbb{U} \begin{pmatrix} c_L \\ c_R \end{pmatrix}, \quad \mathbb{U} \equiv \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}. \quad (15)$$

For the case of conventional single-level transport, the transformation equation, Eq. (15), effectively decouples the operators b_0 from the impurity degrees of freedom [31,32]. Multilevel setups, however, result in the coupling of both operators $b_{e,o}$ with quantum dot degrees of freedom [17]. For simplicity we consider just two levels, and both of them are close to the resonance scattering. The resonance phenomena are generally described in terms of scattering phase shifts $\delta_{e,o}$ occurring in interfering channels. Since both channels b_o and b_e are close to the resonance values, the phase shift occurring in the respective channels is accounted for by the scattering matrix S_{tot} :

$$S_{\text{diag}} = \begin{pmatrix} e^{2i\delta_e} & 0 \\ 0 & e^{2i\delta_o} \end{pmatrix}, \quad S_{\text{tot}} = \mathbb{U}^\dagger S_{\text{diag}} \mathbb{U}. \quad (16)$$

For the purely scattering effects, the transmission function can then be obtained as [17]

$$\mathcal{T}(\varepsilon) = |S_{\text{tot}}|^2 = \sin^2(\delta_e - \delta_o). \quad (17)$$

The energy dependence of the phase shifts $\delta_{e,o}$ comes from the expansion with some arbitrary constants $\alpha_{e,o}$ and $\beta_{e,o}$,

$$\delta_{e,o}(\varepsilon) = \delta_0 + \alpha_{e,o}(\varepsilon/\Gamma) + \beta_{e,o}(\varepsilon/\Gamma)^2 + \dots, \quad (18)$$

where δ_0 is the zero-energy phase shift, which can be set at the same level for all interfering channels [33]. Therefore, the low-energy form of the transmission coefficient reads

$$\mathcal{T}(\varepsilon) = (\alpha_e - \alpha_o)^2 (\varepsilon/\Gamma)^2. \quad (19)$$

The transmission coefficient expressed in Eq. (19) is very generic, which always attains the universal value $21/5$ of the Lorenz ratio. We note that the limiting case of $\alpha_e \rightarrow \alpha_o$ represents the complete destructive interferences between the channels participating in the transport process. At this limiting case the transport is completely blocked. In the absence of channel mixing (noninteracting systems), Eq. (17) is an exact result, showing, in a unified way, the importance of quantum interferences in generic transport experiments.

In conclusion, we reported an upper bound of the Lorenz ratio in the systems possessing weak energy-dependent transmission. In particular, we investigated the quantum transport through the noninteracting systems with vanishing zero-energy transmission where the main hypothesis of conventional WF law must be relaxed. The vanishing of zero-energy transmission has been explored for many systems of experimental interest and found to be associated either with quantum interference effects or the internal structure of the system. In this case of vanishing zero-energy transmission, the Lorenz ratio attains a universal number of $\mathcal{R} = 21/5$ at $T \rightarrow 0$, which is significantly higher than that predicted within conventional WF law. This universal number therefore provides an experimental way of characterizing the interference phenomena in noninteracting nanoscale devices. We observed that there exists no finite upper bound for the Lorenz ratio in scattering theory. Nevertheless, for many experimental systems the Lorenz ratio predicted in this work might even serve as an upper bound. Our prediction can easily be verified in graphene in the absence of screening effects and considering only the long-range impurity. In addition, we propose the noninteracting multilevel-quantum-dot and double-quantum-dot setups as the ideal candidates to test our predictions. Our work provides significant evidence which concludes that the violations of WF law do not necessarily imply a non-Fermi-liquid nature of the underlying transport phenomena; equally, Fermi-liquid transport characteristics cannot be concluded by an observed validation of WF law.

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