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Violation of the Wiedemann-Franz Law in a Single-Electron Transistor

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We study the influence of Coulomb interaction on the thermoelectric transport coefficients for a metallic single-electron transistor. By performing a perturbation expansion up to second order in the tunnel-barrier conductance, we include sequential and cotunneling processes as well as quantum fluctuations that renormalize the charging energy and the tunnel conductance. We find that Coulomb interaction leads to a strong violation of the Wiedemann-Franz law: the Lorenz ratio becomes gate-voltage dependent for sequential tunneling, and is increased by a factor 9/5 in the cotunneling regime. Finally, we suggest a measurement scheme for an experimental realization.

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Introduction.—Electron transport in conductors is accompanied by the transfer of both charge and heat (energy). Thermoelectric transport coefficients relate the charge and heat current, I_e and I_q, to applied voltage and temperature differences, ΔV and ΔT,

$$\begin{pmatrix} I_e \\ I_q \end{pmatrix} = \begin{pmatrix} G_V & G_T \\ M & K \end{pmatrix} \begin{pmatrix} \Delta V \\ \Delta T \end{pmatrix}. \tag{1}$$

The thermal conductance κ is defined by $I_q = \kappa \Delta T$ for $I_e = 0$, i.e., $\kappa = K - G_V T S^2$ where $S = -\Delta V/\Delta T = G_T/G_V$ denotes the thermopower. For macroscopic samples of ordinary metals, the Wiedemann-Franz law provides a universal relation between the two conductances by stating that the Lorenz ratio

$$L \equiv \frac{\kappa}{G_V T}, \tag{2}$$

is a constant given by the Lorenz number $L_0 = (\pi^2/3) \times (k_B/e)^2$. It is a consequence of Fermi-liquid theory, which is applicable when screening renders Coulomb interactions sufficiently weak. The Wiedemann-Franz law indicates that both charge and heat currents are supported by the same underlying scattering mechanisms with only weak energy dependence.

The situation is fundamentally different in mesoscopic systems in which level quantization and Coulomb interaction drastically affect transport. The thermopower has been measured in small dots with discrete level spectrum [1], chaotic dots [2], carbon nanotubes and molecules [3], and dots closer to the metallic (quasicontinuous) limit [4], and calculated for various mesoscopic systems [5,6]. Deviations from the Wiedemann-Franz law have been predicted for tunneling transport through quantum dots for weak coupling [7,8], in the Kondo regime [7,9], for open dots [10], and for granular metals [11].

In the present Letter we address the question of whether and how Coulomb interaction in a metallic single-electron transistor (SET) with weak tunnel couplings affects the Wiedemann-Franz law. The Coulomb interaction plays an important role for two reasons. First, the finite charging energy to add or remove an electron to or from the island suppresses some transport processes. This dramatically affects the charge and thermal conductance individually, but leaves Wiedemann-Franz law untouched since the same transport processes are suppressed for both electric and thermal conductance. Second, however, Coulomb interaction leads to a strong energy dependence of the scattering processes. This yields, in general, a violation of the Wiedemann-Franz law.

By performing a systematic perturbation expansion of all thermoelectric coefficients up to second order in the tunneling conductances, we calculate the effect of Coulomb interaction on the Lorenz ratio. We find that the latter is increased due to Coulomb interaction. In the low-temperature regime, the Lorenz ratio becomes gate-voltage dependent. For the sequential-tunneling contributions it remains L_0 only exactly on the resonance points where the charging-energy gap of the relevant transport process vanishes. With increased detuning the Lorenz ratio rises quadratically as a function of the charging-energy gap. Interestingly, we find that in the cotunneling regime, the Lorenz ratio becomes universal again, but by a factor 9/5 larger than L_0.

Model.—We model the metallic single-electron transistor by the Hamiltonian $H = H_L + H_R + H_I + H_{\rm ch} + H_T$. Here, $H_r = \sum_{kn} \epsilon^r_{kn} a^\dagger_{rkn} a_{rkn}$ with $r = L$, R and $H_I = \sum_{qn} \epsilon_{qn} c^\dagger_{qn} c_{qn}$ describe noninteracting electrons in the left and right lead and on the island, respectively [12]. Coulomb interaction on the island is described by the capacitance model $H_{\rm ch} = E_C(\hat{N} - n_x)^2$, where $E_C = e^2/(2C)$ defines the charging-energy scale with total island capacitance $C = C_L + C_R + C_g$, $\hat{N}$ is the number operator of excess charges on the island, and $en_x = C_L V_L + C_R V_R + C_g V_g$ is the "external charge" that is tunable by gate and bias voltage. To increase the number of electrons on the island from N to $N + 1$ one has to overcome the charging-energy gap $\Delta_N = \langle N+1|H_{\rm ch}|N+1\rangle - \langle N|H_{\rm ch}|N\rangle = E_C[1+2(N-n_x)]$. The resonance condition $\Delta_N = 0$, where the charging-energy gap vanishes, is fulfilled at half-integer values of n_x. Charge transfer processes are described by the tunneling Hamiltonian $H_T = \sum_{r=L,R}\sum_{kqn} T^r a^\dagger_{rkn} c_{qn} e^{-i\hat{\varphi}} + \text{H.c.}$, where we can assume the tunnel matrix elements T^r to be independent of the states k and q and channel index n, as they vary on the scale of the Fermi energy, which is much larger than all other relevant energy scales.

The tunnel-coupling strength for barrier $r = L, R$ is characterized by the dimensionless conductance $\alpha_0^r = \sum_n N_r(0)N_I(0)|T^r|^2 \ll 1$, where $N_{I/r}(0)$ are the density of states of the islands or leads at the Fermi level, and $\alpha_0 \equiv \sum_{r=L,R}\alpha_0^r$. The operator $e^{\pm i\hat{\varphi}}$ shifts the charge on the island by $\pm e$. In general, the electron temperatures of left lead, island, and right lead can all be different from each other and differ from the lattice temperature.

Thermoelectric coefficients.—Charge current $I_e^r = e\langle\sum_{kn} dN_{rkn}/dt\rangle$ and heat current $I_q^r = -\langle\sum_{kn}(\epsilon_{kr}^r - \mu_r)dN_{rkn}/dt\rangle$ leaving lead $r = L, R$ are given by

$$I_e^r = -\frac{ie}{\hbar}\int d\omega[\alpha_e^{r+}(\omega)C^>(\omega) + \alpha_e^{r-}(\omega)C^<(\omega)], \quad (3)$$

$$I_q^r = \frac{i}{\hbar}\int d\omega[\alpha_q^{r+}(\omega)C^>(\omega) + \alpha_q^{r-}(\omega)C^<(\omega)]. \quad (4)$$

They depend on the rate functions for charge and heat transport, respectively,

$$\alpha_e^{r\pm}(\omega) = \alpha_0^r\int_{-\infty}^{\infty} dE f_r^\pm(E+\omega)f^\mp(E), \quad (5)$$

$$\alpha_q^{r\pm}(\omega) = \alpha_0^r\int_{-\infty}^{\infty} dE(E+\omega-\mu_r)f_r^\pm(E+\omega)f^\mp(E), \quad (6)$$

as well as on the correlation functions $C^{\gtrless}(\omega)$ for the island charge. The Fermi function f is denoted by f^+, while $f^- = 1-f$. Applied temperature or voltage differences, $\Delta T = T_L - T_R$ and $\Delta V = V_L - V_R$, are accounted for by evaluating $f_r^\pm(E+\omega)$ at temperature T_r and voltage V_r, while $f^\mp(E)$ is taken at the island electron temperature T. Finally, we define $\alpha^\pm(\omega) = \sum_r \alpha_e^{r\pm}(\omega)$.

In the following we concentrate on the linear-response regime. Furthermore, we assume that the heat current is conserved; i.e., the heat current entering the island from one lead leaves the island to the other lead and the heat flux from the island electrons to lattice and substrate can be neglected. It is convenient to use current conservation $\sum_r I_{e/q}^r = 0$ to write the current as $I_{e/q} = (\alpha_0^R I_{e/q}^L - \alpha_0^L I_{e/q}^R)/(\alpha_0^L + \alpha_0^R)$, and then expand up to linear order in ΔV or ΔT. Then, only the *equilibrium* correlation functions $C^{\gtrless}(\omega)$, taken at $\Delta V = 0$ and $\Delta T = 0$, enter, which are related to the spectral density $A(\omega)$ for charge excitations on the island by $C^>(\omega) = -2\pi i[1-f(\omega)]A(\omega)$ and $C^<(\omega) = 2\pi i f(\omega)A(\omega)$.

We introduce dimensionless thermoelectric coefficients $g_V = G_V/G_{\rm as}$, $g_T = -(e/k_B)G_T/G_{\rm as}$, $m = -(e/k_BT)M/G_{\rm as}$, and $k = (e^2/k_B^2T)K/G_{\rm as}$, where $G_{\rm as} = 4\pi^2(e^2/h)\alpha_0^L\alpha_0^R/(\alpha_0^L+\alpha_0^R)$ is the classical charge conductance asymptotically reached in the high-temperature limit. The Lorenz ratio is $L = (k_B/e)^2[k/g_V - (g_T/g_V)^2]$. We, then, find

$$\begin{pmatrix} g_V & g_T \\ m & k \end{pmatrix} = \int d\omega \frac{\beta\omega/2}{\sinh\beta\omega}A(\omega)\begin{pmatrix} 1 & \frac{\beta\omega}{2} \\ \frac{\beta\omega}{2} & \frac{\pi^2+(\beta\omega)^2}{3} \end{pmatrix} \quad (7)$$

with the equilibrium spectral density $A(\omega)$.

Perturbation expansion.—We proceed by performing a systematic perturbation expansion of the spectral density in the tunnel conductance α_0, as was already done in Refs. [6,13], based on a diagrammatic real-time technique [14], to address charge conductance and thermopower, respectively. To write down the first two terms of the perturbation expansion we only have to specify the results for the correlation functions from Ref. [13] for vanishing voltage and temperature bias.

By sequential-tunneling processes only charge excitations at the resonances, $\omega = \Delta_N$, can be accessed. The zeroth-order contribution, hence, reads

$$A^{(0)}(\omega) = \sum_N (P_N + P_{N+1})\delta(\omega - \Delta_N), \quad (8)$$

where P_N are the equilibrium probabilities to find N charges on the island. The next-order contribution is given by $A^{(1)}(\omega) = \sum_{i=1}^3 A_i^{(1)}(\omega)$ with

$$A_1^{(1)}(\omega) = \sum_N [P_N\alpha(\omega) + P_{N-1}\alpha^+(\Delta_N + \Delta_{N+1} - \omega) + P_{N+1}\alpha^-(\Delta_N + \Delta_{N-1} - \omega)]\mathrm{Re}R_N(\omega)^2, \quad (9)$$

$$\begin{aligned} A_2^{(1)}(\omega) = &\sum_N (P_N + P_{N+1})\delta(\omega - \Delta_N) \\ &\times\Bigg[\partial(2\phi_N + \phi_{N+1} + \phi_{N-1}) - \frac{\phi_{N+1} - \phi_{N-1}}{E_C} \\ &+ \beta\sum_{N'} P_{N'}(\phi_{N'} - \phi_{N'-1}) \\ &- \beta\frac{P_N(\phi_N - \phi_{N-1}) + P_{N+1}(\phi_{N+1} - \phi_N)}{P_N + P_{N+1}}\Bigg], \end{aligned} \quad (10)$$

$$A_3^{(1)}(\omega) = -\sum_N (P_N + P_{N+1})\delta'(\omega - \Delta_N) \times[2\phi_N - \phi_{N+1} - \phi_{N-1}]. \quad (11)$$

Here we used the abbreviations $R_N(\omega) = 1/(\omega - \Delta_N + i0^+) - 1/(\omega - \Delta_{N-1} + i0^+)$ as well as $\phi_N = \alpha_0\Delta_N\mathrm{Re}\Psi(i\beta\Delta_N/2\pi)$, where $\Psi(x)$ is the digamma function, and $\partial\phi_N$ is a short notation for $\partial\phi_N/\partial\Delta_N$.

The first term describes two-electron cotunneling that describes the charge excitations away from resonance, $\omega \neq \Delta_N$. The second and third term are contributions on resonance, $\omega = \Delta_N$, that, in the low-temperature regime, are identified with corrections to sequential tunneling due to renormalization of the tunnel-coupling strength and the charging-energy gap, respectively.

Results.—Analytical results for the thermoelectric coefficients can be found from Eq. (7) with Eqs. (8)–(11). In Fig. 1 we show the resulting dimensionless thermoelectric coefficients g_V, $g_T = m$, k, and the Lorenz ratio L normalized by L_0, as a function of gate voltage for various temperatures, in Fig. 2 the temperature dependence of L/L_0 for various gate voltages. The tunnel coupling is chosen as $\alpha_0^{L/R} = 0.01$. Temperature and gate-voltage dependence of the Lorenz ratio can be elucidated by deriving analytical expressions for various limits. (i) In the high-temperature regime, $\beta E_C \ll 1$, Coulomb oscillations are washed out; i.e., there is no gate-voltage dependence. To calculate corrections to Wiedemann-Franz law in this regime, we expand the gate-voltage average of all thermoelectric coefficients in powers up to $(\beta E_C)^2$ to find

$$\frac{L}{L_0} = 1 + \frac{2}{\pi^2}\beta E_C - \frac{1 + 24\alpha_0}{3\pi^2}(\beta E_C)^2. \tag{12}$$

Deviations from Wiedemann-Franz law are visible before Coulomb oscillations set in, see Fig. 2, where for $k_B T \gtrsim E_C$ the curves coincide for all gate voltages while $L > L_0$. (ii) In the on-resonance low-temperature regime, $\beta E_C \gg 1$ but $\beta \Delta_N \ll 1$ for one N (say $N = 0$), transport is dominated by sequential tunneling and only the charge states 0 and 1 occur. The sequential-tunneling contribution then yields

$$\frac{L}{L_0} = 1 + (\beta\Delta_0)^2/(2\pi)^2, \tag{13}$$

in agreement with Ref. [8]. The Wiedemann-Franz law is only fulfilled for a vanishing charging-energy gap, $\Delta_0 = 0$, with corrections quadratic in $\beta\Delta_0$ away from resonance. These corrections indicate, that the contribution of each transported particle to the heat current scales with the charging-energy gap Δ_0 instead of temperature $k_B T$ as in bulk transport.

Higher-order corrections in α_0 lead to an increase of the Lorenz ratio. For $\Delta_0 = 0$ we find

$$\frac{L}{L_0} = \frac{1 + 4\alpha_0/3 - 2\alpha_0[\gamma + \ln(\beta E_C/\pi)]}{1 - 2\alpha_0[\gamma + \ln(\beta E_C/\pi)]}, \tag{14}$$

with Euler's constant $\gamma = 0.577\ldots$. The terms logarithmic in temperature are associated with the renormalization of the tunnel-coupling strength, that enter both g_V and k in the same way, so that it affects the Lorenz ratio only weakly. (iii) In the off resonance low-temperature regime, $\beta\Delta_N \gg 1$ for all N, transport is dominated by cotunneling. Expanding the thermoelectric coefficients up to quadratic order in temperature, and taking into account only Δ_0 and Δ_{-1} as the two lowest excitation energies, we find $g_V = \alpha_0(8\pi^2/3)E_C^2/(\beta\Delta_0\Delta_{-1})^2$, $g_T = m = 0$, and $k = 3\pi^2/5 g_V$. Because of weak energy dependence of the cotunneling scattering rate, proportionality between charge and heat conductance is recovered, however, with a different prefactor, as

$$\frac{L}{L_0} = \frac{9}{5}. \tag{15}$$

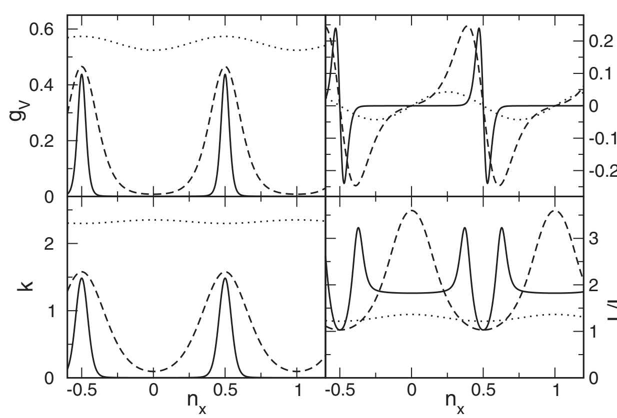

FIG. 1. Coulomb oscillations of thermoelectric coefficients and Lorenz ratio. For high temperatures ($k_B T = E_C/2$—dotted line) oscillations are washed out. In the sequential-tunneling regime ($k_B T = E_C/10$—dashed line) the Lorenz ratio is given by Eq. (13) around each resonance. For low temperatures ($k_B T = E_C/40$—solid line) the new universal Lorenz ratio $9/5 L_0$ is reached in the cotunneling regime.

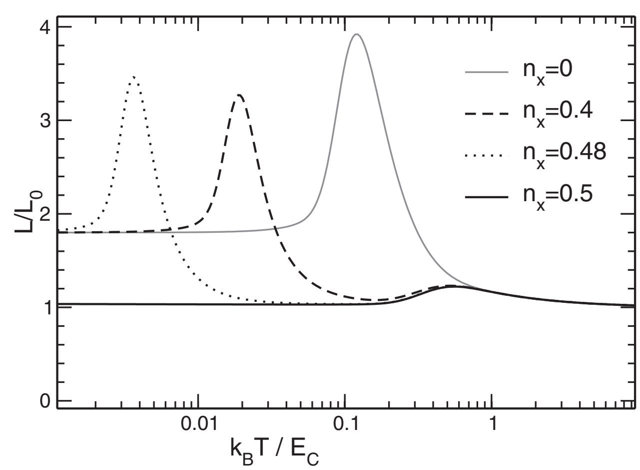

FIG. 2. Temperature dependence of Lorenz ratio for different gate voltages. Two maxima separate different tunneling regimes. The rise to the maxima starts at $k_B T \simeq E_C$ and $k_B T \simeq \Delta_0$, respectively. For $n_x = 0 \Leftrightarrow \Delta_0 = E_C$ the two maxima coincide, whereas at resonance ($n_x = 0.5 \Leftrightarrow \Delta_0 = 0$) the lower maximum is removed to the left.

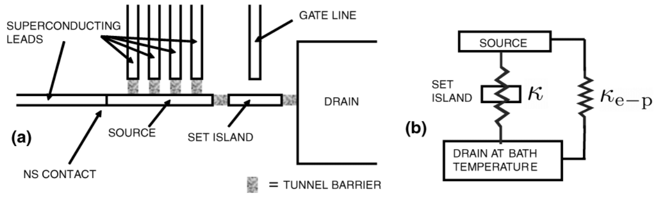

FIG. 3. A possible measurement scheme. In (a) the SET transistor is depicted in a configuration where the drain is thermalized at the bath temperature and the source is a small conductor whose temperature can be varied. In (b) we show a simple thermal model of the device.

Possible experimental realization.—A possible experimental configuration to measure the predicted effects is shown in Fig. 3(a). The whole structure can be fabricated by standard nanolithography and thin film metal deposition and with oxidized tunnel barriers in the SET. The normal metal source of the SET is connected to superconducting leads, some of which are tunnel coupled normal metal-insulator-superconductor (NIS) junctions and the rest are direct normal-metal-superconductor junctions (NS). Both the NIS junctions and the NS Andreev mirrors provide thermal isolation of the source [15]. Another role of the direct NS contacts is to suppress the charging energy of the source electrode while keeping it as small as possible to avoid coupling to phonons [16]. The NIS junctions are used for sensitive thermometry and for heating the source; in proper bias range they can also cool it [15]. In contrast to source electrode and SET island the drain is made large (wide and thick) to secure thermalization at the bath temperature. The NS contacts are used for characterizing the SET electrically by passing current and measuring voltage across. The structure appears rather simple; we note, however, that combining a normal metal SET with superconducting probes requires use of nonconventional material combinations. Figure 3(b) shows the relevant thermal model of the proposed setup. When applying either positive or negative heat flux into the source by a voltage on the NIS contacts, the dominant energy relaxation mechanism should be that discussed in this letter, i.e., electronic thermal conductance κ through the SET transistor. This requires $\kappa_{e\text{-}p} \ll \kappa$ for a small temperature bias of the source with respect to the drain at the bath temperature T_0. On the left-hand side of the inequality $\kappa_{e\text{-}p} = 5\Sigma \mathcal{V} T_0^4$ is the linearized thermal conductance by electron-phonon coupling from source to the lattice. Here $\Sigma \simeq 1 \times 10^9\ \mathrm{W\,K^{-5}\,m^{-3}}$ is a material specific constant [15] and $\mathcal{V}$ is the volume of the source, which can realistically be made as small as $1 \times 10^{-21}\ \mathrm{m}^3$ with a small number of probes attached. A similar argument applies to the choice of the size of the SET island. The inequality between the two heat conductances is satisfied at a realistic temperature of $T_0 = 30$ mK, in particular, near the resonance; deep in the cotunneling regime one may need to resort to the very different temperature and gate-voltage dependencies to distinguish contributions of κ from that of $\kappa_{e\text{-}p}$.

Conclusions.—We have theoretically investigated the influence of Coulomb interaction on thermoelectric transport coefficients for a metallic single-electron transistor and proposed a measurement scheme for experimental verification. We found strong violation of the Wiedemann-Franz law. For sequential tunneling the Wiedemann-Franz ratio depends quadratically on the charging-energy gap: the Wiedemann-Franz law is fulfilled only at the resonances, where the charging-energy gap vanishes. In the cotunneling regime the Lorenz ratio takes a new universal value of 9/5 of the Lorenz number.

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