Large Violation of the Wiedemann-Franz Law in Luttinger Liquids

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We show that in weakly disordered Luttinger liquids close to a commensurate filling the ratio of thermal conductivity κ and electrical conductivity σ can deviate strongly from the Wiedemann-Franz law valid for Fermi liquids scattering from impurities. In the regime where the umklapp scattering rate Γ_U is much larger than the impurity scattering rate Γ_{imp}, the Lorenz number $L = \kappa/(\sigma T)$ rapidly changes from very large values $L \sim \Gamma_U/\Gamma_{\mathrm{imp}} \gg 1$ at the commensurate point to very small values $L \sim \Gamma_{\mathrm{imp}}/\Gamma_U \ll 1$ for a slightly doped system. This surprising behavior is a consequence of approximate symmetries existing even in the presence of strong umklapp scattering.

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In a Fermi liquid, a quasiparticle carries charge e and has an energy of the order of $k_B T$. These basic properties are reflected in the Wiedemann-Franz (WF) law [1,2]: The ratio of the thermal conductivity divided by the temperature T and the electrical conductivity, the so-called Lorenz number

$$L = \frac{\kappa}{\sigma T} = \frac{\pi^2 k_B^2}{3e^2} = L_0, \tag{1}$$

takes a universal value L_0. The WF law $L = L_0$ is valid and routinely observed in the low-T regime of Fermi liquids where impurity scattering dominates.

Deviations from the WF law, $L/L_0 \neq 1$, in the low-T regime, which have, e.g., been reported for high-temperature superconductors [3] or close to quantum-critical points [4], are regarded as evidence that the low-energy excitations cannot be viewed as electronic quasiparticles. But even if a description of thermal and electric transport in terms of Fermi liquid quasiparticles is possible, the WF law will not be valid if inelastic scattering processes dominate which in general relax heat and charge currents differently. Typically, these corrections to L/L_0 are of the order of 1 and not very large [5,6].

Large violations of the WF law usually reflect a dramatic change of the excitation spectrum associated with the opening of a gap. For example, in a Mott insulator σ is exponentially small while heat can still efficiently be transported by spin fluctuations. The opposite case occurs in a superconductor where $\sigma = \infty$ while κ remains finite at finite T due to thermally excited quasiparticles.

In this Letter, we show that small changes in the doping can trigger enormous changes of the Lorenz number L in Luttinger liquids in situations where the umklapp scattering rate Γ_U is larger than the impurity scattering rate $\Gamma_{\mathrm{imp}} \ll \Gamma_U$; see Fig. 1. This happens even in regimes where umklapp scattering does *not* open a charge gap. This peculiar behavior can be traced back to the presence of approximate symmetries of the clean system which affect charge and heat current in a completely different way. This has to be contrasted with a situation where impurity scattering provides the dominant relaxation mechanism for both heat and charge currents. For this case Li and Orignac [5] have shown that only violations of order 1 of the WF law exist.

When investigating the thermal or electrical conductivity of low-dimensional systems, it is important to account for the role of symmetries and conservation laws even if these are only approximate. For example, in integrable one-dimensional models, conductivities are usually infinite at finite T [7] as the conservation laws protect the currents

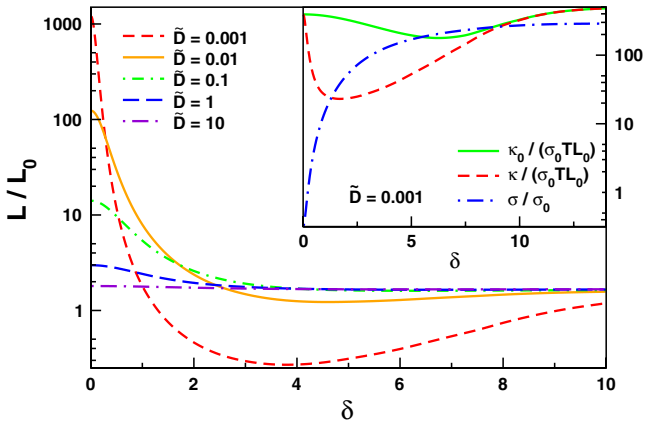

FIG. 1 (color online). Lorenz number L/L_0 (1) as a function of doping $\delta\nu$ away from 1/3 filling [$\delta = 3v_c G\delta\nu/(\pi T)$], using the variables of Eq. (10) (for $K_c = 0.6$, $K_s = 0.8$, $v_s/v_c = 0.5$). If disorder dominates $\tilde{D} \gtrsim 1$, L/L_0 is of order 1 and doping independent. For a clean system with $\tilde{D} \ll 1$, the WF law is strongly violated. A pronounced peak of height $1/\tilde{D}$ and width $\sqrt{\tilde{D}}$ at the commensurate filling is followed by a pronounced minimum. Inset: δ dependence of $\kappa_0/[TL_0\sigma_0(T)]$, $\kappa/[TL_0\sigma_0(T)]$, and $\sigma/\sigma_0(T)$ for $\tilde{D} = 0.001$, $\sigma_0(T) = (v_c^2 a^{2n_c-3}/g^2)(v_c/aT)^\beta$ with $\beta = K_s n_s^2 + K_c n_c^2 - 3$.

from decaying. Small perturbations render the conductivity finite but still large [8]. Below, we demonstrate the implications on the thermoelectric effects.

We consider a weakly disordered one-dimensional (1D) metal described by a single band with the filling $\nu = \nu_0 + \delta\nu$ and the electron density 2ν, where $\nu_0 = m_c/n_c$ with integers m_c and n_c is a commensurate filling. The low-energy Hamiltonian is given [9] by

$$\begin{aligned} H &= H_{\mathrm{LL}} + H_U + H_{\mathrm{imp}}, \\ H_{\mathrm{LL}} &= \int \frac{dx}{2\pi} \sum_{i=c,s} v_i \left(K_i (\partial_x \theta_i)^2 + \frac{1}{K_i} (\partial_x \phi_i)^2 \right), \\ H_U &= \frac{g}{(2\pi a)^{n_c}} \int dx e^{i\sqrt{2}[n_c \phi_c(x) + n_s \phi_s(x)]} e^{-i\Delta k x} + \mathrm{H.c.}, \\ H_{\mathrm{imp}} &= \frac{1}{\pi a} \int dx \eta(x) \{ e^{i\sqrt{2}\phi_c(x)} \cos[\sqrt{2}\phi_s(x)] + \mathrm{H.c.} \}, \end{aligned} \quad (2)$$

where H_{LL} is the usual Luttinger liquid Hamiltonian expressed in terms of spin (s) and charge (c) densities $\partial_x \phi_{c,s}$ and their conjugate variable $\partial_x \theta_{c,s}$ with $[\phi_{c,s}(x), \partial_{x'} \theta_{c,s}(x')] = i\pi\delta(x - x')$. H_U is the dominant umklapp scattering process where $\Delta k = 2n_c k_F - m_c G = n_c G \delta\nu$ (with $G = \frac{2\pi}{a}$) is proportional to the deviation from commensurate filling and $n_s = 0, 1$ for even and odd n_c, respectively. The term H_{imp} with a Gaussian correlated impurity potential $\langle \eta(x)\eta(x') \rangle = D\delta(x - x')$ describes a weak backscattering due to disorder.

Even in the presence of umklapp scattering, an approximate symmetry closely related to momentum conservation exists [10]. The so-called pseudomomentum

$$\tilde{P} = P_t - \frac{m_c G}{2n_c}(N_R - N_L) = P + \frac{\Delta k}{2n_c}(N_R - N_L) \quad (3)$$

[where $N_{R(L)}$ is the number of right (left) movers] commutes with $H_{\mathrm{LL}} + H_U$ (even if effects like band curvature or a weak three-dimensional coupling are added [10,11]). Here P_t is the crystal momentum, and $P = P_t - k_F(N_R - N_L)$ measures the momentum relative to the two Fermi points.

Because of the pseudomomentum conservation, even a strong umklapp scattering may not be sufficient to relax the heat and charge currents. To capture this, one needs a transport theory which properly accounts for the role of conservation laws and the associated vertex corrections. For the nonlinear interaction describing umklapp scattering in Luttinger liquids, the memory matrix approach to transport [12] is to our knowledge the only available method, especially as there are presently no numerical methods to calculate conductivities at finite but low T. As discussed in Ref. [13], this method allows one to calculate lower bounds to σ and κ in the perturbative regime and gives precise results as long as the relevant slow modes are included in the calculation. It was shown to capture prominent features of observable transport phenomena, e.g., magnetothermal transport in spin chains [14].

The first step to set up the memory matrix formalism is to list a number of relevant operators J_i which in our case includes the electrical current $J_1 = J_c = v_c K_c (N_R - N_L)$, the heat current $J_2 = J_h = -\sum_{i=c,s} \int v_i^2 \partial_x \phi_i \partial_x \theta_i$, and the momentum operator $J_3 = P = -\sum_{i=c,s} \int \partial_x \phi_i \partial_x \theta_i$. To leading order in H_U and H_{imp}, the matrix of conductivities is then obtained from

$$\hat{\sigma} = \hat{\chi} \hat{M}^{-1} \hat{\chi}, \qquad M_{ij} = \lim_{\omega \to 0} \frac{\mathrm{Im}\langle \partial_t J_i; \partial_t J_j \rangle_\omega}{\omega}, \quad (4)$$

with the 3×3 memory matrix $\hat{M} = \hat{M}_U + \hat{M}_{\mathrm{imp}}$. As the time derivatives $\partial_t J_i = i[H, J_i]$ are already linear in the weak perturbations g_U and η, the correlators are evaluated with respect to H_{LL}. $\hat{\chi}$ is the matrix of static susceptibilities $\chi_{ij} = \langle J_i; J_j \rangle_{\omega=0}$ with

$$\hat{\chi} \approx \frac{\pi T^2}{3} \begin{pmatrix} \frac{6 v_c K_c}{\pi^2 T^2} & 0 & 0 \\ 0 & v_c + v_s & \frac{1}{v_c} + \frac{1}{v_s} \\ 0 & \frac{1}{v_c} + \frac{1}{v_s} & \frac{1}{v_c^3} + \frac{1}{v_s^3} \end{pmatrix}. \quad (5)$$

The umklapp contribution to Eq. (4) is given by

$$\frac{\hat{M}_U}{c_U \Gamma_U} \approx \begin{pmatrix} \frac{2 v_c^2 n_c^2 K_c^2 F_{00}}{\pi T^2} & \frac{v_c n_c K_c F_3}{\Delta k} & \frac{-v_c n_c K_c \Delta k F_{00}}{\pi T^2} \\ \frac{v_c n_c K_c F_3}{\Delta k} & -v_c^2 F_4/2 & F_3/2 \\ \frac{-v_c n_c K_c \Delta k F_{00}}{\pi T^2} & F_3/2 & \frac{\Delta k^2 F_{00}}{2\pi T^2} \end{pmatrix}, \quad (6)$$

where $c_U = \frac{(\pi)^{K_c n_c^2 + K_s n_s^2 - 1}}{(2\pi)^{2n_c - 1}} (\frac{v_c}{v_s})^{K_s n_s^2}$ and $\Gamma_U = \frac{g^2}{a^{2n_c - 1}} \times (\frac{aT}{v_c})^{K_c n_c^2 + K_s n_s^2 - 1}$. F_{mn} are the dimensionless functions

$$\begin{aligned} F_{mn} &= 2 \int dx dt\, t e^{i\delta x} [\partial_x^m f_c(x,t)][\partial_x^n f_s(x,t)], \\ f_c(x,t) &= [\sinh(x + it) \sinh(x - it)]^{-(K_c n_c^2/2)}, \\ f_s(x,t) &= [\sinh(x v_c/v_s + it) \sinh(x v_c/v_s - it)]^{-(K_s n_s^2/2)}, \\ F_3 &= \pi \{ F_{20} + (v_s/v_c)^2 F_{02} + [1 + (v_s/v_c)^2] F_{11} \}, \\ F_4 &= \pi [F_{20} + (v_s/v_c)^4 F_{02} + 2(v_s/v_c)^2 F_{11}], \end{aligned} \quad (7)$$

which depend on doping and T via $\delta = v_c \Delta k/(\pi T)$. Note that $\hat{M}_U$ has a vanishing eigenvalue reflecting that $[H_U, \tilde{P}] = 0$. The disorder contribution is given by

$$\frac{\hat{M}_{\mathrm{imp}}}{c_{\mathrm{imp}} \Gamma_{\mathrm{imp}}} \approx \begin{pmatrix} (\frac{4 K_c v_c}{2\pi T})^2 & 0 & 0 \\ 0 & v_c v_s \tilde{K} & \frac{K_t^2}{1 + K_t} \\ 0 & \frac{K_t^2}{1 + K_t} & \frac{(\frac{K_c}{v_c^2} + \frac{K_s}{v_s^2}) K_t}{1 + K_t} \end{pmatrix}, \quad (8)$$

where $c_{\mathrm{imp}} = \frac{(2\pi)^{K_t - 1}}{2} (\frac{v_c}{v_s})^{K_s} \frac{\Gamma^2(K_t/2)}{\Gamma(K_t)}$, $\Gamma_{\mathrm{imp}} = \frac{D}{a^2} (\frac{aT}{v_c})^{K_t}$, $K_t = K_c + K_s$, and $\tilde{K} = \frac{(K_c v_c^2 + K_s v_s^2) K_t}{v_c v_s (1 + K_t)}$. Finally, σ, κ, and L of Eq. (1) are obtained from

$$\sigma = \hat{\sigma}_{11}, \qquad \kappa = \kappa_0 - T S^2 \sigma = \frac{1}{T} \left(\hat{\sigma}_{22} - \frac{\hat{\sigma}_{21}^2}{\hat{\sigma}_{11}} \right). \quad (9)$$

It should be noted that κ is measured experimentally in a setup where the charge current vanishes, resulting in the thermoelectric counter terms of Eq. (9). $S = \hat{\sigma}_{21}/(T\hat{\sigma}_{11})$ is the thermopower.

For given Luttinger liquid parameters $v_{c,s}$ and $K_{c,s}$, the Lorenz number depends only on two dimensionless quantities, describing the ratio of renormalized disorder strength and umklapp scattering and the doping:

$$\tilde{D} = \frac{\Gamma_{\mathrm{imp}}}{\Gamma_U} = \frac{Da^{2n_c-3}}{g^2(aT/v_c)^\gamma}, \qquad \delta = \frac{v_c \Delta k}{\pi T}, \tag{10}$$

with $\gamma = (n_c^2 - 1)K_c + (n_s^2 - 1)K_s - 1$. Figure 1 shows the striking doping dependence of σ, κ, and the Lorenz number L/L_0 for the filling 1/3 ($n_c = 3$, $n_s = 1$). For large effective disorder $\tilde{D} \gtrsim 1$, L/L_0 is of order 1 and there is essentially no doping dependence. For $\tilde{D} \ll 1$, one obtains instead a huge and sharp peak of height $1/\tilde{D}$ and width $\sqrt{\tilde{D}}$ followed by a wider dip located at $\delta \sim 1$, where the minimum scales as $\tilde{D}$.

This behavior can be understood by investigating the relation of the currents J_h and J_c to the approximately conserved $\tilde{P}$ [Eq. (3)]. From the continuity equation, one can show [11] that the cross susceptibility of J_c and $\tilde{P}$ is (up to exponentially small corrections) given by the doping $\delta\nu$ away from the commensurable point

$$\chi_{J_c\tilde{P}} = 2\delta\nu \approx \frac{\Delta k \hat{\chi}_{11}}{2n_c K_c v_c} + \hat{\chi}_{31} \tag{11}$$

while $\chi_{J_h\tilde{P}} \sim T^2 > 0$. $\chi_{J_i\tilde{P}}$ measures the "overlap" of the current and the conserved operator. A vanishing χ implies that the operators are orthogonal to each other; i.e., the current is *not* protected by the conservation law and can decay rapidly by umklapp processes. Therefore, at the commensurate point where $\chi_{J_c\tilde{P}} = 0$, J_c can decay by umklapp processes, while J_h is protected. Indeed, as shown in the inset in Fig. 1, at $\delta = 0$ one obtains $\sigma \sim 1/\Gamma_U$ small, but $\kappa \sim 1/\Gamma_{\mathrm{imp}}$, resulting in $L/L_0 \sim 1/\tilde{D}$ in the clean limit $\tilde{D} \ll 1$.

For finite doping, $\chi_{J_c\tilde{P}} = \delta\nu > 0$ and therefore $\sigma \sim (\delta\nu)^2/\Gamma_{\mathrm{imp}}$ grows rapidly until it becomes of the same order as the heat conductivity in the *absence* of electrothermal correction κ_0/T. In this regime, the leading contribution to κ/T, however, of order $1/\Gamma_{\mathrm{imp}}$ is exactly canceled by the thermoelectric counter terms in Eq. (9). The physical origin of this cancellation is that κ is measured under the boundary condition $J_c = 0$. As the component of J_c perpendicular to $\tilde{P}$ decays rapidly by umklapp, J_c and $\tilde{P}$ become almost parallel for small $\tilde{D}$ implying that effectively the heat conductivity measurement is performed under the boundary condition of vanishing $\tilde{P}$. Therefore κ becomes of order $1/\Gamma_U$, and $L/L_0 \sim \tilde{D}$. For neutral liquids a related effect is well known: While mass currents do not decay due to momentum conservation, the heat conductivity measured under the boundary condition of vanishing mass currents remains finite (this situation is more transparent as momentum and mass current are proportional to each other, while this is not the case for J_c and $\tilde{P}$). Finally, for $\delta \gg 1$ the umklapp scattering is exponentially suppressed, both σ and κ/T are of order $1/\Gamma_{\mathrm{imp}}$, and $L/L_0 \sim 1$ [5].

In Fig. 2, the T dependence of the WF ratio, σ, and κ are shown using the appropriate dimensionless variables

$$\tilde{\delta} = \frac{\delta}{\tilde{D}^{1/\gamma}}, \qquad \tilde{T} = \frac{T}{T_D}, \qquad T_D \equiv \frac{v_c}{a}\left(\frac{Da^{2n_c-3}}{g^2}\right)^{1/\gamma}. \tag{12}$$

Upon lowering T, the disorder close to 1/3 filling becomes more and more important, $\tilde{D}$ grows, and L/L_0 becomes of order 1 for low T. As explained above, for vanishing doping $\tilde{\delta} = 0$, σ is much smaller than κ/T as long as umklapp scattering dominates. For finite doping, umklapp scattering is exponentially suppressed at low T (see inset in Fig. 2). However, when it sets in ($\tilde{T} > 1$), it leads to a larger suppression of κ/T compared to σ due to the partial cancellations from thermoelectric corrections.

While the theoretical analysis of the problem described above is most transparent for the filling close to 1/3, it is useful to study a case with direct experimental realizations. One possible candidate is the quarter-filled quasi-1D Bechgaard salt $(TMTSF)_2PF_6$ [15], where the anisotropy of the kinetic energy (t_a:t_b:t_c = 250:20:1 meV) allows a Luttinger liquid description for $T \gtrsim 100$ K. Two extra complications arise at quarter filling: First, in the absence of disorder the effective low-energy model $H_{\mathrm{LL}} + H_U$ becomes the integrable sine-Gordon model, which formally has an infinite number of conservation laws on top of the pseudomomentum. For an analysis of transport, one has to identify the leading corrections which break integrability (see Ref. [8]). Second, for $H_{\mathrm{LL}} + H_U$ there is a strict separation of charge and spin degrees of freedom, the latter being not affected by umklapp scattering. We therefore have to take band curvature [16] into account, which couples spin and charge and breaks integrability:

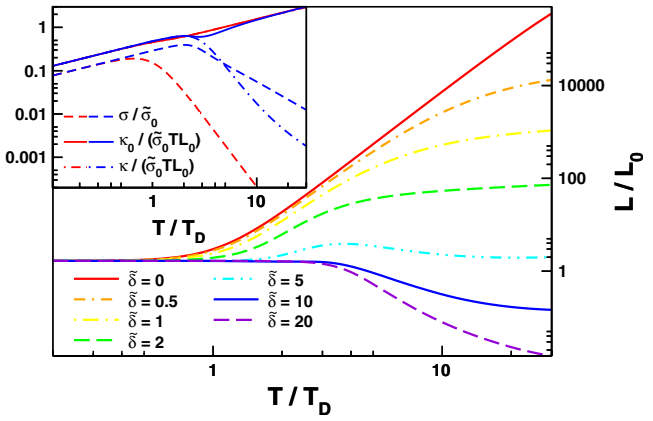

FIG. 2 (color online). T dependence of the Lorenz number for various dopings close to 1/3 filling using (12) (parameters as in Fig. 1). At low T, disorder always dominates resulting in a T-independent L/L_0 of order 1. At the commensurate point, $L/L_0 \sim 1/\tilde{D}$. Inset: $\kappa_0(T)/[TL_0\tilde{\sigma}_0(D)]$, $\kappa(T)/[TL_0\tilde{\sigma}_0(D)]$, and $\sigma(T)/\tilde{\sigma}_0(D)$ for $\tilde{\delta} = 0$ (red line) and $\tilde{\delta} = 10$ (blue line). Here $\tilde{\sigma}_0(D) = [Da^{2n_c-3}/g^2]^\alpha v_c^2/D$ with $\alpha = (2 - K_c - K_s)/\gamma$.

$$H_{\mathrm{BC}} = -\frac{1}{6\sqrt{2}m}\int[\partial_x\phi_c^3 + 6\partial_x\phi_s\partial_x\theta_s\partial_x\theta_c + 3\partial_x\phi_c(\partial_x\phi_s^2 + \partial_x\theta_s^2 + \partial_x\theta_c^2)] - \delta\mu\int\partial_x\phi_c. \quad (13)$$

Here we have added an extra T-dependent chemical potential $\delta\mu = \frac{T^2\pi^2}{12m}[\frac{1}{v_c^2}(K_c + K_c^{-1}) + \frac{1}{v_s^2}(K_s + K_s^{-1})]$ to account for the T-independent particle density 2ν in a 3D crystal. To leading order in $1/m$, corrections to $\hat{\chi}$ arise only for $\chi_{12} = \chi_{21} \approx \frac{\pi T^2}{3m}(1/v_c + 1/v_s)$ and $\chi_{13} = \chi_{31} \approx \frac{\pi T^2}{3m}(1/v_c^3 + 1/v_s^3)$. As both $N_R - N_L$ and P commute with H_{BC}, only $\hat{M}_{22}$ gets an extra contribution: $\hat{M}_{22}^{\mathrm{BC}} = \frac{\pi^8 T^5}{128m^2 v_s^4 v_c^4}K_c(K_s^{-2} + K_s^2 - 2)\int t\,\mathrm{Im}\{[4\cosh^2(x+it)+2]\times \sinh(x+it)^{-4}\sinh(xv_c/v_s + it)^{-2}\sinh(xv_c/v_s - it)^{-2}\}$. As $J_c \to J_c + P/m$, σ is given by $\sigma = \hat{\sigma}_{11} + 2\hat{\sigma}_{13}/m + \hat{\sigma}_{33}/m^2$ (the corresponding correction to J_h is subleading and therefore omitted).

An example for the expected doping and T dependencies is shown in Fig. 3 for a filling close to $1/4$ using parameters consistent with existing resistivity data for $(\mathrm{TMTSF})_2\mathrm{PF}_6$ [15]. Both $\rho(T)$ and $\sigma(\omega)$ in this system can be explained [15] by umklapp scattering in a $1/4$-filled Luttinger liquid with $K_c \approx 0.22$ leading to $\rho \sim g^2 T^{16K_c - 3}$ (i.e., $\sigma \sim T^{-0.56}$; see Fig. 3) along the chain. Other parameters like K_s, v_s, m, and, most importantly, disorder strength D are not known experimentally. The absence of any visible disorder contribution to $\rho(T)$ in the Luttinger liquid regime $T \gtrsim 100$ K allows us to estimate crudely $D \ll 0.0005$ in units of g^2/a^{2n_c-3}. Our results shown in Fig. 3 strongly suggest that a large violation of the WF law (after subtraction of the phonon contribution not discussed here) should be observable in Bechgaard salts and similar materials.

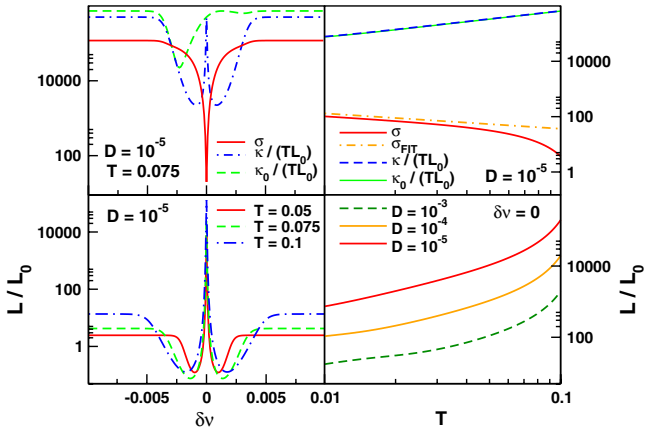

FIG. 3 (color online). Lorenz number (lower curves), κ, and σ (upper curves) for a system close to $1/4$ filling ($\sigma_{\mathrm{FIT}} = 10T^{-0.56}$ is the fit to σ), where $K_c = 0.22$ (chosen to be compatible with Ref. [15]), $K_s = 0.8$, $v_s/v_c = 1/2$, $g = 0.1v_c a^{n_c-3/2}$, $n_c = 4$, $n_s = 0$, and $m = 1/v_c a$; D is in units of $g^2/(a^{2n_c-3})$, and T (in units of v_c/a) is in the experimentally accessible regime.

Qualitatively, the doping dependence of L/L_0 for $1/4$ and $1/3$ filling are similar. The WF ratio L/L_0 shows a pronounced sharp peak of height $1/\tilde{D}$ followed by a dip for $v_c\Delta k \sim T$. T dependencies might differ in the two cases due to the different T dependence of $\tilde{D}$: Whether $1/\tilde{D}$ grows or shrinks upon lowering T depends on K_c and K_s. However, the most prominent T dependence arises from the fact that umklapp scattering is effectively switched off at lowest T for $\delta\nu > 0$, resulting in $L \sim L_0$.

We expect that the strong violation of the WF law in regimes where umklapp scattering is large compared to disorder will not only occur for the strictly 1D systems discussed here but even if a weak interchain tunneling (as in the case of Bechgaard salts) is taken into account, as a small modulation of the 1D bands does not affect the structure of approximate conservation laws; see [11]. Besides the disparate behavior of κ/T and σ, an interesting finding of our study is the importance of thermoelectric corrections for the slightly doped system. In the regime where L/L_0 gets very small due to a partial cancellation of κ_0 and $TS^2\sigma$, the dimensionless thermoelectric figure of merit $ZT = T\sigma S^2/\kappa_0$, which measures the efficiency of a thermoelectric element for power generation or refrigeration, becomes 1, a remarkably large value [17].

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