

Violation of the Wiedemann-Franz law in a large- N solution of the t - J model

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We show that the Wiedemann-Franz law, which holds for Landau Fermi liquids, breaks down in a large- n treatment of the t - J model. The calculated ratio of the in-plane thermal and electrical conductivities agrees quantitatively with experiments on the normal state of the electron-doped $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ ($x=0.15$) cuprate superconductor. The violation of the Wiedemann-Franz law in the uniform phase contrasts with other properties of the phase that are Fermi liquid like.

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A recent experiment that measured the electrical and thermal conductivities of the copper-oxide superconductor $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ in its normal state found striking deviations from the Wiedemann-Franz law.¹ Simply stated, the Wiedemann-Franz law says that fermion quasiparticles transport both electrical and heat currents, with the ratio of the heat conductivity κ to the electrical conductivity σ given by

$$\frac{\kappa}{\sigma T} = \frac{\pi^2}{3} \left(\frac{k_B}{e} \right)^2. \quad (1)$$

Ordinary Landau Fermi liquids respect the Wiedemann-Franz law, so deviations from it indicate the presence of non-Fermi liquid physics. The results of Ref. 1 are therefore broadly consistent with other experimental evidence that points to non-Fermi liquid behavior in the cuprate phase diagram.

One possible interpretation of the breakdown of the Wiedemann-Franz law is that the quasiparticle fractionalizes into separate carriers of charge and spin. To see what effects such fractionalization might have, consider first heat transport in a system of weakly interacting electrons, which could be viewed as a crude approximation to electrons in the highly overdoped region of the cuprate phase diagram. The electrons transport both charge and heat. The electrical conductivity is given by the Drude formula $\sigma = ne^2\tau/m$ and the thermal conductivity $\kappa \propto T$ because while each quasiparticle carries fixed charge e , it only carries an energy of order the temperature. Next consider a model for the undoped cuprates: the Néel ordered antiferromagnetic insulator with zero electrical conductivity. Phonons and spin waves transport heat, and as both excitations are bosonic in character with linear dispersions at low energy, each contributes similarly, yielding a thermal conductivity $\kappa \propto T^3$.

Leaving aside these ordinary states of matter, consider the case in which the spins in the insulator, instead of ordering, fractionalize into spinons with an extended Fermi surface and a nonzero density of states.^{2,3} Now spins transport heat in much the same way as charges do in the noninteracting electron system, with $\kappa \propto T$. The Lorenz ratio is infinite, and remains significantly larger than 1 upon doping with holes or electrons.

Just this scenario is predicted in a large- n treatment of the t - J model on the square lattice. We follow the approach of

Refs. 4–6. Implementing the single-occupancy constraint by introducing slave-boson operators b_i , the t - J model may be written

$$H = -t \sum_{\langle i,j \rangle} (c_i^\dagger b_i c_j b_j^\dagger + \text{H.c.}) + J \sum_{\langle i,j \rangle} \left(\vec{S}_i \cdot \vec{S}_j - \frac{1}{4} n_i n_j \right) + \frac{1}{2} \sum_{i \neq j} V(|\vec{r}_i - \vec{r}_j|) n_i n_j. \quad (2)$$

Number and spin operators are related to the electron creation and annihilation operators by $n_i \equiv c_i^\dagger c_i$ and $\vec{S}_i \equiv (1/2) c_i^\dagger \vec{\sigma}_i c_i$, where there is an implicit sum over pairs of raised and lowered Greek indices. The single-occupancy constraint is now holonomic, $b_i^\dagger b_i + c_i^\dagger c_i = 1$, with the physical meaning that only a hole, or a single electron, may occupy each site of the lattice. (In the following we generally refer to hole doping with the understanding that our calculations apply equally well to electron doped systems.) Included in Eq. (2) is the off-site Coulomb repulsion $V(r)$; however, in the uniform and staggered flux phases discussed below it plays no role other than to contribute an additive constant to the total energy.

Because spin-exchange involves no net flow of charge, electrical current only arises from the hopping term. The continuity equation relates the time rate of change of the occupancy on a given site to the lattice divergence of the current flowing into the site:

$$e^* \frac{dn_j(t)}{dt} = -i e^* [n_j, H] = - \sum_{\hat{e}=x,y} \frac{j_{j,j+\hat{e}}^e - j_{j-\hat{e},j}^e}{a}, \quad (3)$$

where a is the lattice spacing between copper atoms. Thus

$$j_{j,j+\hat{e}}^e = -i e^* t a^* (c_j^\dagger b_j c_{j+\hat{e}} b_{j+\hat{e}}^\dagger - \text{H.c.}) \quad (4)$$

is the electric current flowing from site j into site $j+\hat{e}$ along the link connecting the two sites. We emphasize that neither the spin-spin exchange interaction J nor the Coulomb interaction $V(r)$ appear in the expression for the electrical current. This result, which is a direct consequence of the gauge invariance of the spin-exchange and Coulomb interactions in

our microscopic calculation, contrasts with that obtained recently⁷ within the more phenomenological “*d*-density wave” picture.⁸

The heat current $j_{j,j+\hat{e}}^q$ can be found by taking the time derivative of the Hamiltonian density:

$$\frac{dh_j(t)}{dt} = - \sum_{\hat{e}=\hat{x},\hat{y}} \frac{j_{j,j+\hat{e}}^q - j_{j-\hat{e},j}^q}{a}, \quad (5)$$

where

$$h_j = \sum_{i=j\pm\hat{x},\hat{y}} (-tc_i^\dagger b_i c_{j\alpha} b_j^\dagger + \text{H.c.}) + J\vec{S}_i \cdot \vec{S}_j \quad (6)$$

and the sum is only over the four sites i that are nearest-neighbors of site j . We have dropped the $n_i n_j$ interaction terms. These do not contribute to the dc thermal conductivity because $\langle n_i \rangle$ remains unchanged in the presence of currents. However, in contrast to the electrical current, the heat current has contributions both from hopping, and from spin-exchange:

$$\begin{aligned} j_{j,j+\hat{e}}^q &= t*a*[c_j^\dagger{}^\alpha b_j \partial_t (c_{j+\hat{e}\alpha} b_{j+\hat{e}}^\dagger) + \text{H.c.}] \\ &+ \frac{J*a}{2} * [c_j^\dagger{}^\alpha (\partial_t c_{j+\hat{e}\alpha}) c_{j+\hat{e}}^\dagger{}^\beta c_{j\beta} \\ &+ c_j^\dagger{}^\alpha c_{j+\hat{e}\alpha} (\partial_t c_{j+\hat{e}}^\dagger{}^\beta) c_{j\beta}]. \end{aligned} \quad (7)$$

The model generalizes from the physical case of $n=2$ (up and down spins) to arbitrary (even integer) values of n by letting the Greek indices run over $1, \dots, n$. In the large- n limit the spin-spin interaction factorizes in the particle-hole channel. Formally this factorization is implemented via a Hubbard-Stratonovich transformation within the functional integral approach. Complex-valued mean fields along the bonds are introduced:

$$\chi_{ij} = \frac{J}{n} \langle c_i^\dagger{}^\alpha c_{j\alpha} \rangle. \quad (8)$$

The χ_{ij} fields function as the order parameter, and as they are spin-rotation invariant, there is no possibility of Néel or other spin order, and the mean-field Hamiltonian respects spin-rotational symmetry. Furthermore, at sufficiently low temperatures the (holon) boson fields condense,⁹ and we may make the replacement $b_i = b_i^\dagger = \sqrt{x}$, where x is the hole density. The mean-field Hamiltonian, which is exact in the $n \rightarrow \infty$ limit, then takes the form

$$H_{MF} = \sum_{\langle i,j \rangle} \left[\frac{n}{J} |\chi_{ij}|^2 - (tx + \chi_{ij})(c_j^\dagger{}^\alpha c_{i\alpha} + \text{H.c.}) \right]. \quad (9)$$

For parameters appropriate to the cuprates, $t=0.44$ eV (following Hybertsen *et al.*¹⁰) and $J=0.13$ eV (obtained by Singh *et al.*¹¹), the minimum energy configuration has χ_{ij} both real and constant when the doping exceeds $x>0.12$. There are no broken symmetries in this “uniform” phase. Upon suppressing dimerization with the addition of a biquadratic spin-exchange interaction,^{5,12} a staggered flux (SF)

phase with counter-circulating orbital currents^{4-6,13} occurs for $x<0.12$, that is, in the underdoped region of the phase diagram. (The biquadratic interaction which simultaneously exchanges four fermions disappears in the physical $n=2$ limit of up and down spins, and thus does not alter the physics of the t - J model.⁵) More realistic models include nonzero next- and third-nearest neighbor hopping amplitudes¹⁴ but these terms do not qualitatively affect the phase diagram or transport behavior.

The expressions for the two currents simplify in the large- n limit. As the boson operators may be replaced by the c -number \sqrt{x} , the electrical current becomes

$$j_{j,j+\hat{e}}^e = -ie*t*x*a*(c_j^\dagger{}^\alpha c_{j+\hat{e}\alpha} - \text{H.c.}). \quad (10)$$

Upon further replacing the fermion bilinear operator $c_i^\dagger{}^\alpha c_{j\alpha}$ with $(n/J)\chi$, the heat current also simplifies:¹⁵

$$j_{j,j+\hat{e}}^q = (t*x + \chi)*a*(c_j^\dagger{}^\alpha \partial_t c_{j+\hat{e}\alpha} + \text{H.c.}). \quad (11)$$

As the heat current differs only by the $(t*x + \chi)$ prefactor from that of a noninteracting tight-binding system, in the low-temperature limit the Lorenz ratio for in-plane transport is simply

$$\frac{\kappa}{\sigma T} = \frac{\pi^2}{3} \left(\frac{k_B}{e} \right)^2 * \left(\frac{tx + \chi}{tx} \right)^2. \quad (12)$$

Thus for any $\chi \neq 0$ the ratio differs from unity, indicating a breakdown of Fermi liquid theory. Note that all details of the scattering mechanisms cancel out in the ratio. Direct calculation of the two conductivities in linear response shows that the integrals over momentum have identical form. Only the frequency integrals differ; hence for static impurities in the weak-scattering limit the Lorenz ratio is given by Eq. (12). We note that while the order parameter χ is perturbed by the application of external electric fields or thermal gradients, the perturbation in χ does not alter the dc response.¹⁶

In the SF phase the χ fields are complex numbers,⁴⁻⁶ and the prefactor $(tx + \chi)^2$ should be replaced by $|tx + \chi|^2$. Specifically, for $\chi = |\chi| \exp(i\theta)$, with phase θ , the Lorenz ratio generalizes to¹⁷

$$\frac{L}{L_0} = \frac{(tx)^2 + |\chi|^2 + 2tx|\chi|\cos(\theta)}{(tx)^2}, \quad (13)$$

where $L \equiv \kappa/(\sigma T)$ and $L_0 \equiv (\pi^2/3)(k_B/e)^2 \approx 2.45 \times 10^{-8} \text{W}\Omega\text{K}^{-2}$ is the Lorenz number.

We turn now to a comparison of our predictions, Eqs. (12) and (13), with experiments. In Fig. 1 we plot the Lorenz ratio as a function of the doping. As expected, the ratio diverges as the insulating limit $x \rightarrow 0$ is approached because the spinons transport only heat, not charge. In the opposite limit of large doping $\chi \rightarrow 0$, and the ratio approaches unity. Landau Fermi liquid theory is recovered in the dilute limit of widely spaced electrons. We emphasize that the uniform phase with χ_{ij} constant and real does not break any symmetries. It exhibits weak pseudogap behavior because, according to the mean-field equations, $|\chi|$ increases slightly in size at low temperatures, which in turn increases the quasiparticle bandwidth

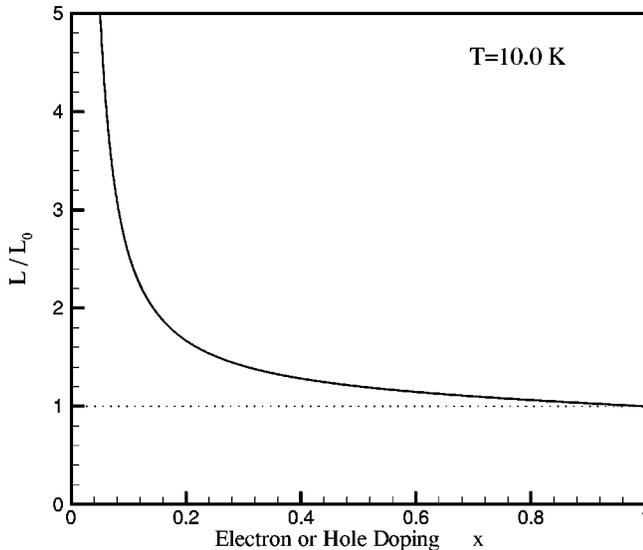


FIG. 1. The Lorenz ratio [Eqs. (12) and (13)] as a function of the doping for $t=0.44$ eV, $J=0.13$ eV. The ratio approaches unity in the dilute limit, $x \rightarrow 1$.

[see Eq. (9)] and decreases the density of states (DOS).¹⁸ For example, at a hole concentration of $x=0.15$, $|\chi|=0.024$ eV at $T=500$ K rising to $|\chi|=0.026$ eV at zero temperature; consequently the DOS drops by 2%. This contrasts with the strong pseudogap behavior of the SF phase which has a gap along most of the Fermi surface and which breaks time reversal invariance. In either phase, however, the fermionic quasiparticles are noninteracting in the $n \rightarrow \infty$ limit and hence behave as long-lived Landau quasiparticles such as those found in ordinary Fermi liquids.

We note that the Wiedemann-Franz law is strongly violated in s -wave superconducting states because while Cooper pairs carry charge, the condensate has no entropy. In a d -wave superconductor, quasiparticle excitations at the nodes result in a modified Wiedemann-Franz law.¹⁹ The violation that we find occurs in the normal state, and is a consequence of the spin-charge separation inherent in the large- n solution of the t - J model, and not of any incipient superconducting tendencies.

In Fig. 2 we plot the temperature dependence of the Lorenz ratio for three dopings at which transport experiments have been conducted: $x=0.06$, 0.15 , and 0.26 . For a single crystal of the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ material with hole doping $x=0.06$, the resistivity was measured upon suppressing the superconductivity by application of a 18 T magnetic field along the c axis.²⁰ The thermal conductivity was, however, measured in the superconducting state, so it is not possible to extract a real Lorenz ratio. Nevertheless it is intriguing that $L/L_0 \approx 5$ at low temperatures, based on the numbers appearing in the inset to Fig. 3 of Ref. 20. This compares reasonably well with the theoretical value of 4.1 seen in Fig. 2.

At optimal doping, experimentally available magnetic fields can only eliminate superconductivity in electron-doped compounds. In Ref. 1 a 13 T field was applied to $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ at $x=0.15$ to access the normal state. The measured ratio of $L/L_0 \approx 2$ found at temperatures above

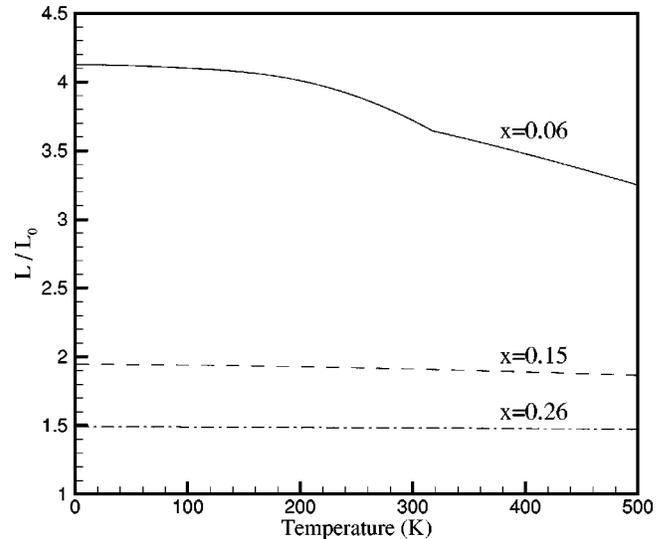


FIG. 2. The Lorenz ratio [Eqs. (12) and (13)] as a function of temperature at dopings $x=0.06$, 0.15 , and 0.26 for $t=0.44$ eV, $J=0.13$ eV. In the underdoped $x=0.06$ case there is a transition to the staggered flux phase at a temperature of approximately 300 K.

0.3 K is again in reasonable quantitative agreement with the theoretical value of 1.95. At temperatures below 0.18 K, however, the experimentally determined ratio drops rapidly below 1. We have no explanation for the observed behavior at the lowest temperatures.²¹

Finally, in the highly overdoped regime Proust *et al.*²² have studied the $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$ material at a hole concentration of $x \approx 0.26$. Superconductivity was suppressed in a 13 T field, and $L/L_0 = 0.99 \pm 0.01$ in good agreement with the Wiedemann-Franz law for Fermi liquids. The theoretical value of the ratio is 1.5. We speculate that the persistence of non-Fermi liquid behavior at large doping in the mean-field theory is an artifact of the large- n approximation. Finite- n corrections could possibly restore Fermi liquid behavior in the overdoped region. At large doping $|\chi|$ is small compared to the effective hole hopping amplitude tx , so fluctuations in χ may be expected to be relatively more important than at low doping.

In summary we have shown that the Wiedemann-Franz law is violated in a mean-field treatment of the t - J model. Our analysis, which holds for weak scattering, is exact in the $n \rightarrow \infty$ limit. The Lorenz ratio is significantly larger than 1 both in the uniform phase ($x > 0.12$) and in the SF phase ($x < 0.12$). The theoretical prediction is in reasonably good quantitative agreement with existing experimental measurements on the cuprate materials.

After this work was completed a paper by Kim and Carbotte (KC) appeared²³ that examined the Wiedemann-Franz law within the context of the phenomenological d -density wave picture. There are several differences between their work and ours. The main difference is that we study both the uniform phase which has no broken symmetries, and the SF or d -density phase with time-reversal breaking counter-circulating currents. We find that the Lorenz ratio is significantly larger than 1 in both phases. Furthermore, at low temperatures KC find only small deviations from the

Wiedemann-Franz law. This is due in part to the fact that their d -density order parameter (the analog of our χ_{ij}) was chosen to be purely imaginary [equivalent to setting $\theta = \pi/2$ in our Eq. (13)] and also because their kinetic energy is not rescaled by the slave-boson doping factor, x , as it is in our microscopic analysis of the t - J model. KC also find a

large temperature variation in the Lorenz ratio for the case of strong scattering because the quasiparticle lifetime has a strong frequency dependence.

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