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Anisotropic Violation of the Wiedemann-Franz Law at a Quantum Critical Point

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A quantum critical point transforms the behavior of electrons so strongly that new phases of matter can emerge. The interactions at play are known to fall outside the scope of the standard model of metals, but a fundamental question remains: Is the basic concept of a quasiparticle—a fermion with renormalized mass—still valid in such systems? The Wiedemann-Franz law, which states that the ratio of heat and charge conductivities in a metal is a universal constant in the limit of zero temperature, is a robust consequence of Fermi-Dirac statistics. We report a violation of this law in the heavy-fermion metal $CeCoIn_5$ when tuned to its quantum critical point, depending on the direction of electron motion relative to the crystal lattice, which points to an anisotropic destruction of the Fermi surface.

Discovered in 1853, the Wiedemann-Franz (WF) law (*1*) has stood as a robust empirical property of metals, whereby the thermal conductivity κ of a sample is related to its electrical conductivity σ through a universal ratio. In 1927, Sommerfeld (*2*) used quantum mechanics, applying to electrons the new Fermi-Dirac statistics, to derive the following theoretical relation

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

where T is the absolute temperature, k_B is Boltzmann's constant and e is the charge of the electron. The extremely good agreement between the theoretical constant $L_0 \equiv \frac{\pi^2}{3}\left(\frac{k_B}{e}\right)^2$ and the empirical value played a pivotal role in establishing the quantum theory of solids. In 1957, Landau went on to show that, even in the presence of strong interactions, electrons in a metal can still be described as weakly interacting fermions ("quasiparticles") with renormalized mass (*3*). This is the essence of what became known as Fermi-liquid (FL) theory, the "standard model" of metals. In the limit of zero temperature, the WF law survived unchanged because it does not depend on mass. (Eq. 1 is only a law at $T \to 0$, as only in that limit is energy conserved in collisions.) It has since been shown that the WF law remains valid as $T \to 0$ for arbitrary strong scattering, disorder, and interactions (*4*). It is built into the fabric of matter, valid down to the quanta of conductance, respectively equal to $\frac{\pi^2}{3}\frac{k_B{}^2 T}{h}$ for heat and $\frac{e^2}{h}$ for charge (*5*).

In the past decade, however, departures from FL theory have been observed in *d*- and *f*-electron metals when tuned to a quantum critical point (QCP), a zero-temperature phase transition between distinct electronic ground states (*6*). These typically show up as an anomalous temperature dependence of properties at the QCP, for example, a specific heat coefficient that never saturates, growing as $C/T \sim \log(1/T)$ (*7*), and an electrical resistivity that grows linearly with T (*8*). Quantum criticality also appears to be linked to the emergence of exotic forms of superconductivity (*9–11*) and nematic (*12*) electronic states of matter.

To determine whether Landau quasiparticles survive at a QCP, we have measured the transport of heat and charge in $CeCoIn_5$, a heavy-fermion metal with a QCP tuned by magnetic field H. In its phase diagram (Fig. 1), the QCP is located on the border of superconductivity and marks the end of a FL regime at $H = H_c = 5.0$ T, where the electrical resistivity obeys the FL form $\rho = \rho_0 + AT^2$ (*13*). A power-law fit to the A coefficient yields $A \sim (H - H_c)^{-\alpha}$, with $\alpha \cong 4/3$ and $H_c = 5.0 \pm 0.1$ T (*13*). At H_c, C/T never saturates (*14*). The same phenomenology is found at the field-tuned QCP of $YbRh_2Si_2$ (with $\alpha \cong 1$) (*15*).

In Fig. 2, we show how the thermal and electrical resistivities in the $T = 0$ limit behave in $CeCoIn_5$ as the field is tuned toward H_c. These are extrapolations to $T = 0$ of the low-temperature thermal resistivity, defined as $w \equiv L_0 T/\kappa$, and electrical resistivity ρ, for current directions parallel ($J \parallel c$) and perpendicular ($J \perp c$) to the tetragonal axis of the crystal lattice. The raw data and their extrapolation are shown in detail in (*4*). For $H = 10$ T, far away from H_c, $w(T)$ and $\rho(T)$ converge as $T \to 0$ for both current directions. However, very close to the QCP, for $H = 5.3$ T, they only converge for in-plane transport. In other words, transport along the c axis violates the WF law, with w_c extrapolating to a distinctly larger value than ρ_c as $T \to 0$. In the supporting material (*4*), we show that extrapolations are not needed to conclude in a violation of the WF law, as the difference data, $w_c(T) - \rho_c(T)$ versus T, shows a rigid T-independent shift from field to field. The normalized Lorenz ratio, $\frac{L}{L_0} \equiv \frac{\kappa}{L_0 \sigma} \equiv \frac{\rho}{w}$, is also seen to approach unity at 10 T but not at 5.3 T.

Our observation of a violation of the WF law at a QCP is characterized by three distinctive

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features: (i) charge conduction is essentially unperturbed; (ii) heat conduction becomes less efficient, so that $L < L_0$; and (iii) the violation is qualitatively anisotropic, present in one direction and absent in the other.

The constancy of charge conduction distinguishes this from the two known instances of WF violation. The first occurs in the case of superconductivity, where σ immediately goes to infinity as H drops below H_{c2}, whereas κ drops gradually, so that $L/L_0 = 0$. The second instance occurs in the other limit (σ→0), realized in the crossover from a metal to an insulator. In this limit, a violation has been observed in cuprates as the Mott insulating state is approached (*4*). The case of $CeCoIn_5$ is neither one of superfluid condensation nor one of charge localization, but that of a good metal violating the WF law. The fact that it is a downward violation, L (T→0) < L_0, seems inconsistent with the possibility of neutral fermionic excitations such as those predicted to emerge at a heavy-fermion QCP (*16*). Instead, we will argue that the Fermi surface is destroyed, anisotropically.

In the T→0 limit, the WF law holds as long as there is a step in the Fermi distribution function, that is, as long as a sharp Fermi surface exists. This step is proportional to the renormalization parameter Z, the defining property of a Landau quasiparticle (*17*). In standard FL theory, Z is a measure of how strongly the quasiparticle mass m^* is enhanced by electron interactions, with $Z \sim 1/m^*$. The anisotropic violation seen in $CeCoIn_5$ thus suggests that a sharp Fermi surface does not exist in the c direction but does exist in the plane. (This is consistent with the observation of de Haas–van Alphen oscillations in $CeCoIn_5$ for $H \parallel c$ (*18*), because these result from coherent electron orbits in the plane.) In other words, Z must be a function of polar angle, $Z = Z(\theta)$, whereby $Z = 0$ over a region around the "poles" (c axis) and $Z > 0$ in a region around the "equator" (basal plane), which provides evidence of anisotropic zeros in the Z parameter of a metal due to a QCP.

An anisotropic destruction of the Fermi surface is reminiscent of what occurs in the pseudogap state of underdoped high-temperature superconductors, where photoemission studies have revealed a Fermi surface broken into small arcs (*19*), shrinking to points along "nodal" directions ($\varphi = \pi/4$) as T→0 (*20*). This angle-dependent destruction may be caused by strong antiferromagnetic (AF) correlations; it certainly is predominant at points connected by the AF ordering vector. By analogy, the uniaxial destruction of the Fermi surface in $CeCoIn_5$ may be caused by spin fluctuations with a uniaxial character, a scenario which is consistent with both the known fluctuation spectrum and the finite temperature properties discussed below.

Having focused on the $T = 0$ limit, we now examine how quantum criticality unfolds as a function of T. The electrical resistivity of $CeCoIn_5$ at the QCP is plotted up to 15 K (Fig. 3) for both current directions. ρ_c shows a purely linear T dependence, from 0.4 μΩ cm at 25 mK all the way to 40 μΩ cm at 16 K. This 100-fold increase in resistivity extends by one order of magnitude the range over which criticality has so far been observed to persist in any material, proving beyond doubt that the power law is an intrinsic property of electrons scattered by critical fluctuations. ρ_a is qualitatively different. Its linear T dependence is seen only above 4 K or so, crossing over to a $T^{3/2}$ dependence below ≅ 1K (*21*).

A comparison of heat and charge conductivities reveals information about the momentum dependence of inelastic scattering. This was discussed in detail in the context of our study of $CeRhIn_5$ (*22*), the antiferromagnetic cousin of $CeCoIn_5$, with a Néel ordering temperature T_N = 3.8 K. The main piece of information that can be extracted directly is the characteristic temperature T_{SF} of magnetic fluctuations, defined as the temperature above which the WF law is restored, i.e., $w(T) \cong \rho(T)$. (T_{SF} is the magnetic analog of the Debye temperature, the characteristic temperature for the scattering of electrons by phonons, Θ_D.) In $CeRhIn_5$, $T_{SF} \cong$ 8 K (*22*), in good agreement with the onset of AF correlations seen with neutron scattering. In $CeCoIn_5$, the same approach applied to in-plane transport yields a field-dependent T_{SF}, ≅ 4 K at H_c and rising to match that of $CeRhIn_5$ at high field (*21*). This allows us to understand the strange behavior of ρ_a. The linear-T regime above T_{SF} arises from fluctuations without preferred spatial correlations, effectively scattering electrons on the whole Fermi surface and making it uniformly "hot." (A T-linear resistivity is also found in conventional metals when $T > \Theta_D$.) Below T_{SF}, the emergence of AF correlations peaked at certain q vectors in the plane leads to "hot spots" and the higher $T^{3/2}$ power law at low T. The fact that ρ_c remains linear down to the lowest temperatures suggests that T_{SF}→0 in this case, that no interplane correlations build up, and that the Fermi surface remains hot over large regions (away from the plane). [In our phonon analog, this would imply Θ_D→0, a quantum melting of the three-dimensional (3D) solid into stacks of solid sheets separated by nonviscous liquid or gas.]

Whereas the $T = 0$ intercepts are different, the linear T dependence of the c-axis electrical resistivity is paralleled by the thermal resistivity (Fig. 3, inset): $w(T)$ is perfectly linear down to the lowest T. Not only is $w_c \sim T$ but also the slope of w_c is roughly equal to that of ρ_c—a confirmation that T_{SF} indeed vanishes in this direction. This indicates that the usual (1 – cosθ) vertex—which makes small-angle scattering ineffective in degrading a charge current—is not working in $CeCoIn_5$. It may be unimportant because inelastic scattering is dominated by large-q processes, as one would expect from AF fluctuations, or it may be inoperative for some special reason, as in the "Kondo breakdown" model (*23*).

In this instance of anisotropic quantum criticality, given that Z and T_{SF} exhibit the same anisotropy ($Z, T_{SF} = 0$ along the c axis and Z, $T_{SF} > 0$ in the basal plane), it is tempting to suggest that (i) a vanishing energy scale, Z→0, and a

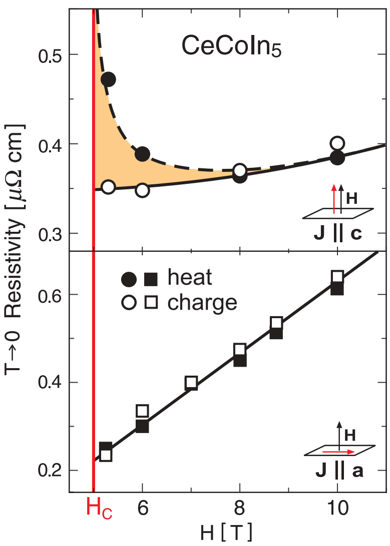

Fig. 2. Violation of the WF law. Residual resistivities (extrapolated to $T = 0$) as a function of magnetic field, for heat (solid symbols) and charge (open symbols) transport. For in-plane transport (**bottom**), the two resistivities track each other as a function of field, thereby obeying the WF law at all fields. For inter-plane transport (**top**), the electrical resistivity ρ_c is flat as $H \rightarrow H_c$, whereas the thermal resistivity increases, thereby causing a violation of the WF law at the QCP, with a Lorenz number $L < L_0$.

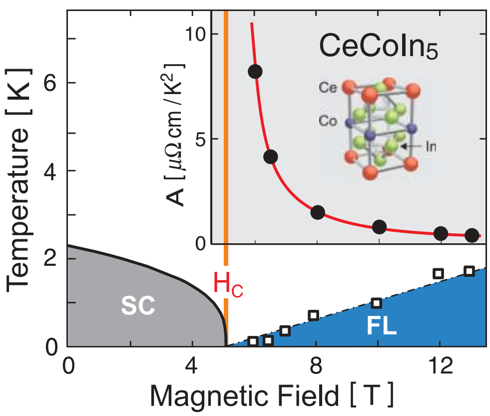

Fig. 1. Phase diagram of $CeCoIn_5$. Magnetic field-temperature phase diagram for a field perpendicular to the basal plane of the tetragonal crystal lattice (shown in inset), i.e., $H \parallel c$, as determined from in-plane resistivity measurements (*10*). The QCP is located at $H = H_c$ = 5.0 T (vertical red line). FL behavior, $\rho = \rho_0 + AT^2$, is obeyed in the blue wedge, ending at H_c. The coefficient A, proportional to the square of the electron effective mass, diverges at H_c as a power law. Below H_c, superconductivity (SC) sets in.

WF law violation are all related, and (ii) a good indicator for their joint occurrence is a linear-T resistivity. Returning to our comparison with cuprates, a similar connection between $\rho \sim T$ and $Z = 0$ appears to exist there as well. Indeed, a recent measurement of the (azimuthal) anisotropy of the in-plane scattering rate $\Gamma(\phi)$ in an overdoped cuprate (*24*) revealed that $\Gamma \sim T$ at $\phi = 0$, where the Fermi surface is eventually destroyed (at lower doping), and $\Gamma \sim T^2$ at $\phi = \pi/4$, where it survives.

It is instructive to compare our findings with the properties of other materials and theories of quantum criticality. A $T^{3/2}$ resistivity is observed in $CeIn_3$ near the pressure-tuned QCP where its AF order vanishes (*6*). $CeIn_3$ is the cubic parent compound of tetragonal $CeRhIn_5$ and, along with the increase in c/a ratio, the ordering temperature drops from $T_N = 10$ K in the former to $T_N = 3.8$ K in the latter. However, they still have comparable T_{SF} (assuming that in $CeIn_3$ $T_{SF} \cong T_N$). $CeCoIn_5$ encounters a further stretch of the c/a ratio, and long-range AF order is no longer stabilized. However, it can still be viewed as a layered version of $CeIn_3$, with similar in-plane correlations and scattering. In this sense, the $T^{3/2}$ dependence observed in $CeCoIn_5$ can be viewed as the result of antiferromagnetic fluctuations that are characteristic of the parent compound. Theoretically, a $T^{3/2}$ resistivity is expected for AF critical fluctuations in 3D from the so-called quantum spin density wave (SDW) model (*17*, *25*, *26*). In this scenario, critical scattering is peaked at "hot spots" connected by the AF wave vectors (*25*). As $T \rightarrow 0$, one would expect the Fermi surface to remain sharp everywhere else, and thus the WF law to prevail, as found here for in-plane currents.

A T-linear resistivity is observed at the composition-tuned QCP of $CeCu_{5.9}Au_{0.1}$ (*7*) and field-tuned QCP of $YbRh_2Si_2$ (*12*), where AF order is thought to disappear. [In these cases, the power law is linear in both high-symmetry directions (*15*, *27*).] The fact that a linear power is inconsistent with the SDW model for AF fluctuations in 3D prompted the proposal of a 2D version (*28*) and of an alternate theory, where critical scattering is local in space and therefore present at all wave vectors (*29*). These scenarios would lead to a more extreme breakdown of FL theory, because the Fermi surface is "hot" not only at certain specific spots but everywhere. It was argued in (*8*) that the specific heat data on Ge-doped $YbRh_2Si_2$, which shows a C/T that exceeds the $\log(1/T)$ dependence at low temperature, may be an indication of such enhanced breakdown. In $CeCoIn_5$, the fact that it is in the direction where $\rho \sim T$ that the WF law is violated is certainly consistent with this picture. Clearly, it would be interesting to test the WF law in $YbRh_2Si_2$.

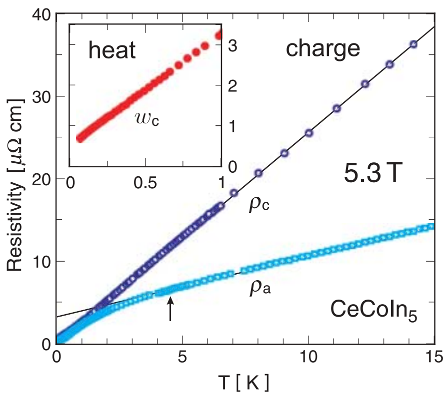

Fig. 3. Anisotropic quantum criticality. Electrical resistivity at the QCP (at $H = 5.3$, $T \cong H_c$) for in-plane (ρ_a) and inter-plane (ρ_c) current directions. ρ_c (T) remains linear over a 100-fold increase in magnitude. By contrast, ρ_a is linear only above a characteristic fluctuation temperature $T_{SF} \cong 4$ K (arrow) (*18*). (**Inset**) Thermal resistivity ($w_c \equiv L_0T/\kappa_c$) at the QCP, for inter-plane transport. w_c is perfectly linear down to the lowest temperature.

Bringing together our findings for $T \rightarrow 0$ and $T > 0$, a picture of qualitative anisotropy emerges, not present in either the SDW model or the local criticality model, at least in their current forms. The characteristic spin fluctuation temperature T_{SF} vanishes at the QCP for transport along the c axis but not in the plane. As a result, the breakdown of FL theory is extreme in the c direction: $\rho_c \sim T$ and $w_c \sim T$ down to the lowest temperatures and the $T = 0$ Fermi surface is blurred, that is, the quasiparticle Z parameter vanishes, in regions around the c-axis direction.

A possible origin for this anisotropic criticality is an anisotropic spin fluctuation spectrum. First, an AF instability is present in all three $CeMIn_5$ compounds (M = Co, Rh, Ir), as shown by the fact that magnetic ordering can be induced by Cd doping (*30*). Second, a magnetic field does tune the magnetism. In $CeRhIn_5$ under pressure (where it becomes in many ways more similar to $CeCoIn_5$, e.g., by developing superconductivity with the same T_c), a magnetic field stabilizes long-range magnetic order (*31*, *32*). In $CeCoIn_5$, it is the magnetic fluctuations that are tuned by a magnetic field (*21*), with T_{SF} starting at a value equivalent to that of $CeRhIn_5$ at high fields and then lowered to a minimum at H_c. Third, the AF fluctuations in $CeCoIn_5$ have strongly anisotropic character (*33*), with magnetic moments well coupled in-plane but weakly coupled interplane. This is consistent with the helical ordering of moments in $CeRhIn_5$, commensurate in-plane and incommensurate along the c axis. Therefore, it seems natural to link this uniaxial anisotropy with the observed anisotropy in T_{SF}, power laws, and $Z(\theta)$. What is not yet known is whether a scenario of AF critical fluctuations can indeed cause a violation of the WF law at $T \rightarrow 0$.

However, the AF scenario is not the only candidate for the anisotropic quantum criticality of $CeCoIn_5$. The "Kondo breakdown" model proposed recently (*23*, *34*), a type of deconfined QCP where the hybridization between conduction and f electrons goes to zero, captures some of the key signatures—absence of magnetic order in the phase diagram, strong anisotropy, multiple energy scales, and a T-linear behavior of both charge and heat resistivities. Proximity to a Pomeranchuk instability of the Fermi surface can also cause anisotropy in electronic liquids (*7*). Recent calculations show that the transport decay rate at such a QCP has a linear T dependence everywhere on the Fermi surface except at "cold" points, resulting in a $T^{3/2}$ dependence of the resistivity (*35*).

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