Thermal and electrical transport across a magnetic quantum critical point

Heike Pfau1, Stefanie Hartmann1, Ulrike Stockert1, Peijie Sun1, Stefan Lausberg1, Manuel Brando1, Sven Friedemann1, Cornelius Krellner1, Christoph Geibel1, Steffen Wirth1, Stefan Kirchner1,2, Elihu Abrahams3, Qimiao Si4 & Frank Steglich1

A quantum critical point (QCP) arises when a continuous transition between competing phases occurs at zero temperature. Collective excitations at magnetic QCPs give rise to metallic properties that strongly deviate from the expectations of Landau’s Fermi-liquid description, which is the standard theory of electron correlations in metals. Central to this theory is the notion of quasiparticles, electronic excitations that possess the quantum numbers of the non-interacting electrons. Here we report measurements of thermal and electrical transport across the field-induced magnetic QCP in the heavy-fermion compound YbRh2Si2 (refs 2, 3). We show that the ratio of the thermal to electrical conductivities at the zero-temperature limit obeys the Wiedemann–Franz law for magnetic fields above the critical field at which the QCP is attained. This is also expected for magnetic fields below the critical field, where weak antiferromagnetic order and a Fermi-liquid phase form below 0.07 K (at zero field). At the critical field, however, the low-temperature electrical conductivity exceeds the thermal conductivity by about 10 percent, suggestive of a non-Fermi-liquid ground state. This apparent violation of the Wiedemann–Franz law provides evidence for an unconventional type of QCP at which the fundamental concept of Landau quasiparticles no longer holds4–6. These results imply that Landau quasiparticles break up, and that the origin of this disintegration of the heavy quasiparticles—these insights could be relevant to understanding other deviations from Fermi-liquid behaviour frequently observed in various classes of correlated materials.

In metallic systems, continuously suppressing magnetic order gives rise to a QCP7 and leads to non-Fermi-liquid behaviour8–11. Whether quasiparticles persist near the QCP, however, is a fundamental open issue. An established means to probe the fate of the quasiparticles is to compare the thermal conductivity ($\kappa$) and the electrical conductivity ($\sigma$). If quasiparticles are well defined, the Wiedemann–Franz law specifies the zero-temperature ($T = 0$) value of the Lorenz number $L = \kappa/\sigma T$ to be $L_0 = (\pi k_B^2/3 e^2)$, where $k_B$ is Boltzmann’s constant and $e$ is the charge of an electron. Except for superconductors12, where the Lorenz ratio $L/L_0 = 0$, a violation of the Wiedemann–Franz law would constitute direct evidence for physics beyond the Fermi-liquid theory. $L/L_0$ becomes larger than one if there are additional carriers which contribute to the heat current but not to the charge current13. By contrast, $L/L_0 < 1$ at $T = 0$ implies a breakdown of Landau quasiparticles.

Heavy-fermion metals are prototype systems for antiferromagnetic QCPs. These rare-earth or actinide-based intermetallics contain both $f$-derived localized magnetic moments and itinerant (spd) conduction electrons, whose entanglement gives rise to the Kondo effect and the concomitant composite quasiparticles with huge effective mass. In these materials, two types of QCPs have been highlighted. The conventional spin-density-wave type relies on the fluctuations of the antiferromagnetic order parameter14–16. Here, the main part of the Fermi surface remains unaffected by the critical fluctuations, leaving the quasiparticles intact. In a related picture17, all states near the Fermi surface are influenced by quantum critical fluctuations. At an unconventional type of QCP, a breakdown of the Kondo entanglement dissociates all the heavy quasiparticles18. Neutron scattering and magnetic measurements19 in CeCu6−$_x$Au$_x$ as well as de Haas-van Alphen19 and thermodynamic and transport20 measurements in CeRhIn$_5$ have been interpreted in terms of a local Kondo-breakdown QCP21. In YbRh2Si2, the weak antiferromagnetic order is continuously suppressed by a tiny magnetic field22,23. Electrical transport and thermodynamic measurements have revealed multiple vanishing energy scales24 and a discontinuity of the Fermi surface across the QCP25,26. These materials provide a setting to characterize the quasiparticles near the antiferromagnetic QCP.

We focus on YbRh2Si2, in order to take advantage of the understanding of its Fermi surface25,26. Figure 1 displays the overall temperature–magnetic field ($T$–$B$) phase diagram (Fig. 1a) and the thermal conductivity below 12 K (Fig. 1b). At $B = 0$, the compound orders antiferromagnetically at the Neel temperature $T_N = 0.07$ K. Increasing $B$ to its critical value $B_c \approx 0.067$ T (with $B$ perpendicular to the $c$ axis, $\perp c$) continuously suppresses $T_N$ to zero, reaching the QCP. Below $T_{Q1}$, the paramagnetic phase at $B > B_c$ is a heavy Fermi liquid3, in which the Fermi surface is large as a result of the Kondo effect. However, in the antiferromagnetic phase ($B < B_c$), also a mass-enhanced Fermi liquid4, the Fermi surface is small, without incorporating the $f$ electrons25,26. The $T^*(B)$ line defines a crossover of the Fermi surface as a function of the control parameter $B$, and terminates at $B = B_c$ as $T \to 0$. On cooling, the field range of quantum critical behaviour shrinks to $B = B_c$ in the $T = 0$ limit, whereas a Fermi-liquid ground state exists on either side of the QCP. The QCP is clearly identified by an asymptotic ($T \to 0$) linear temperature dependence of the electrical resistivity, independent of sample quality21,22. In addition, the width of the $T^*$ crossover is proportional to $T$, extrapolating to a sharp jump of the Fermi surface at $T = 0$ (ref. 24). Although these measurements prove the existence of two different states on either side of the QCP, they leave open the nature of not only the electronic excitations in the quantum critical regime but also the dynamical processes underlying the Kondo breakdown.

A previous study of the thermal and electrical transport in the quasi-two-dimensional heavy-fermion metal CeCoIn5 (ref. 25), in which a magnetic QCP is suspected26 but not identified, found that the Wiedemann–Franz law is violated ($L/L_0 \approx 0.8$ as $T \to 0$) for $c$-axis transport but obeyed for in-plane transport. These results were discussed in terms of putative strongly anisotropic critical fluctuations, although how spin fluctuations may invalidate the Wiedemann–Franz law was left as an open question. Combined thermal and electrical transport has also been studied near the QCP of ZrZn2$_2$, which is considered a canonical ferromagnetic-spin-fluctuation system27.
Figure 1 | Phase diagram and thermal conductivity of YbRh$_2$Si$_2$. a, Temperature–magnetic field ($T$–$B$) phase diagram, indicating the antiferromagnetic phase (AF) boundary ($T_N$, solid line) and the crossovers between non-Fermi-liquid and Fermi-liquid (FL) regimes ($T_{FL}$, dashed line) as well as between small and large Fermi surfaces ($T^*$, double-dashed line). The crossover width at $T^*$ is proportional to temperature (red shaded region) (from ref. 24). The magnetic field, $B$, was applied within the basal tetragonal, easy magnetic plane, $\perp c$. Arrows indicate fields at which combined thermal and electrical transport measurements were performed (Fig. 2a–c). The Wiedemann–Franz law is strictly defined only in the $T = 0$ limit and is expected to describe the electronic transport of a Fermi liquid. This is illustrated in the low-$T$ transport properties of the field-induced paramagnetic phase, $B > B_c$ (Fig. 2c, d). It is also expected in the antiferromagnetic phase, $B < B_c$: here, at finite temperature the electronic thermal conductivity, $\kappa_{el}$, is masked by a contribution due to magnons, $\kappa_m$ (see text). However, as $T \to 0$, $\kappa_m$ vanishes such that the heat transport is purely electronic, and the Wiedemann–Franz law is valid. b, Thermal conductivity, $\kappa$, as a function of temperature, $T$, at zero field (red data points). The solid purple line displaying $\kappa_{WF}(T) = L_0T/\rho(T)$ was obtained under the assumption of the Wiedemann–Franz law to hold in the whole range of temperatures $T \leq 12$ K; here, $\rho(T)$ is the electrical resistivity and $L_0 = (\pi^2 k_b^2/3e^2)$ is Sommerfeld’s constant. The dashed blue line shows the phonon contribution $\kappa_{ph}(T)$, as discussed in the Supplementary Information. Inset, same data below $T = 0.1$ K.

Although the two transport quantities in ZrZn$_2$ have different temperature dependencies, with $L/L_0 < 1$, their extrapolated $T = 0$ limits satisfy the Wiedemann–Franz law.

YbRh$_2$Si$_2$ provides a unique opportunity to study the fate of Landau quasiparticles at QCPs beyond the spin-fluctuation description and, likewise, the nature of the critical fluctuations associated with the Kondo breakdown. The compound is also advantageous because superconductivity is absent down to at least 0.01 K (ref. 21), unlike the case of CeCoIn$_5$. This not only exposes the properties in the immediate vicinity of the antiferromagnetic QCP but also facilitates the characterization of the quasiparticles through the Lorenz ratio. YbRh$_2$Si$_2$ is a magnetically anisotropic metal; the possibility of quasi-two-dimensional transport necessitates the use of in-plane transport to probe any quasiparticle breakdown$^{25}$. The present study therefore focuses on the thermal and electrical transport within the tetragonal plane.

The thermal conductivity $\kappa(T)$ was measured between 0.025 K and 12 K and is shown in Fig. 1b for $B = 0$. For comparison, the electronic thermal conductivity calculated from the measured electrical resistivity $\rho(T)$ through the Wiedemann–Franz law, $\kappa_{WF}(T) = L_0T/\rho(T)$, is also presented. Above 4 K, $\kappa(T)$ exceeds $\kappa_{WF}(T)$ due to the contribution of phonons to the heat transport, $\kappa_{ph}(T)$ (see Supplementary Information). Below 4 K, $\kappa_{ph}(T)$ is suppressed, and $\kappa(T)$ becomes smaller than $\kappa_{WF}(T)$ down to about 0.035 K and somewhat larger at even lower temperatures (inset to Fig. 1b).

In order to investigate the Wiedemann–Franz law, we extrapolate the Lorenz ratio $L(T)/L_0 = \rho(T)/w(T)$ to $T = 0$ (here $w(T)$ is the thermal resistivity). Because a QCP is a singular point in the phase diagram, and given that there are temperature scales that vary as a function of the control parameter and vanish at the QCP, the combination of isofield and isothermal scans is crucial for the extrapolation (Supplementary Information section vi).

Figure 2 depicts the low-temperature behaviour of both the electrical resistivity $\rho(T)$ and thermal resistivity $w(T) = L_0T/\kappa(T)$ at zero field, $B = 0.06$ T $\approx B_c$ and $B > B_c$. Here $w(T)$ has the same unit as $\rho(T)$. Similar results at other magnetic fields are given in Supplementary Fig. 4. This comparison shows that $w(T)$ exceeds $\rho(T)$ over a wide range of temperature and field. Figures 3a, b and c, d display, respectively, the difference $w(T) - \rho(T)$ and the Lorenz ratio for the data shown in Fig. 2a–d. Corresponding plots for the data shown in Supplementary Fig. 4 are presented in Supplementary Fig. 5a–d.

Below $T = 0.15$ K, at $B \approx 0.6$ T, $w(T) > \rho(T)$ within the experimental resolution. This is illustrated for $B = 1$ T in Fig. 3b, which shows that $w(T) - \rho(T)$ approaches zero in this range of $T$ and $B$, and in Fig. 3d, which demonstrates that $L(T)/L_0 = 1$ within the experimental error. In this high-field range, both $\Delta \rho(T) = [\rho(T) - \rho_0] \propto T^2$ and $[w(T) - w_0] \propto T^2$ below the Fermi-liquid crossover temperature, marked by arrows in Fig. 2 and Supplementary Fig. 4. Here, $\rho_0$ and $w_0$ are the residual ($T \to 0$) electrical and thermal resistivities, which are identical within about 1%. These results establish the validity of the Wiedemann–Franz law in the Fermi-liquid phase for $B \approx 0.6$ T. For $0.2 T \approx B \approx 0.6$ T, the results shown in Fig. 2c and Supplementary Fig. 4d–f suggest similar Fermi-liquid behaviour at lower temperatures.

The system is in the quantum critical regime$^{21}$ at $B = 0$ and $T \gtrsim 0.1$ K, where $w(T) > \rho(T)$. Both $\rho(T)$ and $w(T)$ decrease linearly with temperature below about 0.3 K which allows extrapolation of the quantum critical behaviour of $\rho(T)$ and $w(T)$ to the $T = 0$ limit, giving
port, as concluded in ref. 29. Instead, the Fermi-liquid-type electronic thermal transport is not entirely due to electronic-quasiparticle transport. Below a critical behaviour (dashed lines in a and c) seems to occur at about 0.07 K, almost exactly where the corresponding feature becomes visible at $B = 0.06$ T, too. This is in striking contrast to Fig. 2a, showing that the deviation in the $w(T)$ data for $B = 0$ has set in already at $T = 0.1$ K. The reason for this seeming discrepancy lies in the pronounced drop of the electrical resistivity at $T_N \approx 0.07$ K (Fig. 2a). The extrapolation of the dashed lines in a and c to $T = 0$ demonstrates a violation of the Wiedemann–Franz law in a putative paramagnetic, non-Fermi-liquid ground state. This ground state is realized exactly at the critical magnetic field $B_c$, compare Fig. 1a. Error bars are derived from the standard deviation of the data in Fig. 2. Evolution of a shallow minimum in the isothermal 0.1 K $< T < 0.4$ K $L(B)/L_0$ dependence. Data at lower $T$ are not included because of the additional magnon heat transport at $B < B_c$ which will vanish as $T \rightarrow 0$. These minima are related to the $T^*(B)$ line of Fig. 1a, as indicated by the crossover fields (arrows) and widths (horizontal bars). Above $B^*(T) = B(T^*)$, $L_0$ values are consistent with $L_0/L_0$ values in Supplementary Fig. 10, implying in the $T = 0$ limit $L/L_0 = 1$ at $B \neq B_c$ and an abrupt dip at $B = B_c$. Error bars as in c and d.

Supplementary Information we demonstrate that the additional thermal conductivity is due to antiferromagnetic magnons; this magnon contribution will vanish in the $T = 0$ limit, as is inferred from the specific-heat data measured down to 0.018 K (Supplementary Information). Therefore, at $B = 0$ the Wiedemann–Franz law is expected to hold in the $T = 0$ limit.

At $B = 0.06$ T $\approx B_c$, $\rho(T)$ is linear below 0.12 K down to the lowest measured temperature, as is $w(T)$ below about 0.2 K (Fig. 2b). At $T \approx 0.07$ K, $w(T)$ shows a downturn which is similar to, though considerably weaker than, that at $B = 0$ which sets in at higher temperature (Fig. 2a). We interpret this feature as the contribution of overdamped magnons in the paramagnetic regime close to the QCP (Supplementary Information); as in the case of $B = 0$, this magnetic contribution is expected to vanish in the $T = 0$ limit. Extrapolating the linear-in-$T$ electrical resistivity and the electronic thermal resistivity, which is also linear in $T$ between 0.07 and 0.2 K, to $T = 0$ we find $(w_0 - \rho_0) > 0$ and $L(T \rightarrow 0)/L_0 < 1$, similar to the behaviour at $B = 0$ (Fig. 3a, c). Here, our extrapolation is taken near path C (Supplementary Fig. 8a).

These results provide an overall picture that can be placed in the context of the phase diagram of Fig. 1a. For fields sufficiently above the critical field $B_c$, the Wiedemann–Franz law is obeyed at low temperatures. At the same time, the data at $B = 0$ can be interpreted as validating the Wiedemann–Franz law in the $T = 0$ limit, that is, in the antiferromagnetic ground state. The validity of the Wiedemann–Franz law at magnetic fields away from $B_c$ and for sufficiently low temperatures is consistent with a field-induced continuous quantum phase transition between two Fermi liquids with, respectively, small and large Fermi surfaces, which has been inferred from magnetotransport and thermodynamic measurements. In contrast, the data in the paramagnetic quantum critical regime are extrapolated to a $T = 0$ limit that violates the Wiedemann–Franz law.

The isothermal field dependence, $L(B)/L_0$, further clarifies these results. This is given in Fig. 3e, which shows a shallow minimum near a field that tracks the $T^*(B)$ line in Fig. 1a. The minimum narrows as temperature is reduced and extrapolates, as $T \rightarrow 0$, to an abrupt dip at $B = B_c$ (Supplementary Information); the extrapolated $T = 0$ value at that point is about 0.9 (compare Fig. 3c). The systematic evolution of $L/L_0$ versus $B$ and $T$ provides evidence for the intrinsic nature of the apparent violation of the Wiedemann–Franz law in YbRh$_2$Si$_2$.

Our findings shed considerable new light on the dynamical electronic processes occurring at the QCP. Quasiparticles disintegrate at a
Kondo-breakdown QCP, as illustrated in Fig. 4. The large Fermi surface incorporates both the conduction electrons and delocalized \( e \) electrons, whereas the small Fermi surface involves only the conduction electrons. Because the quantum phase transition is continuous, this change of the Fermi surface must result from inelastic processes that operate near the QCP. Such dynamical processes must be electronic, extending to zero energy when the system is precisely at the QCP. Correspondingly, the quasiparticle residue of the large Fermi surface, \( Z_L \), and that of the small Fermi surface, \( Z_S \), must reach zero as the QCP is approached from the paramagnetic and antiferromagnetic sides, respectively. At the critical value of the control parameter, their values respectively at the large Fermi wavevector (\( k_F^L \)) and the small one (\( k_F^S \)) satisfy dynamical scaling:

\[
Z_L(k_F^L, T, \omega) = T^\alpha \varphi_L(\omega/T) \\
Z_S(k_F^S, T, \omega) = T^\beta \varphi_S(\omega/T)
\]

Here, \( \omega \) is the frequency, \( \alpha \) and \( \beta \) are scaling exponents, and \( \varphi_L \) and \( \varphi_S \) are scaling functions. These scaling forms of \( Z_L \) and \( Z_S \) capture the physics of the critical Kondo breakdown. This breakdown arises from the dynamical competition between RKKY (Ruderman–Kittel–Kasuya–Yosida) and Kondo interactions which, respectively, promote small and large Fermi surfaces. The resulting critical fluctuations between the small and large Fermi surfaces amount to quantum critical inelastic excitations. The vanishing quasiparticle weights, \( Z_L \) and \( Z_S \), imply that such quantum fluctuations and the concomitant fluctuating Fermi surfaces persist at the QCP, thereby making it natural for \( L/L_0 < 1 \) even in the quantum-critical region. The vanishing quasiparticle weights, \( Z_L \) and \( Z_S \), capture the physics of the critical Kondo breakdown. This breakdown arises from the dynamical competition between RKKY and Kondo interactions which, respectively, promote small and large Fermi surfaces. The resulting critical fluctuations between the small and large Fermi surfaces amount to quantum critical inelastic excitations. The vanishing quasiparticle weights, \( Z_L \) and \( Z_S \), imply that such quantum fluctuations and the concomitant fluctuating Fermi surfaces persist at the QCP, thereby making it natural for \( L/L_0 < 1 \) even in the quantum-critical region. The vanishing quasiparticle weights, \( Z_L \) and \( Z_S \), cancel out, where \( L \) and \( \alpha \) are the length and the cross-section of the sample, respectively (see Supplementary Information). Additional measurements of the electrical resistivity were performed on a second single crystal from the same batch, but with a different geometry factor (sample 2). As described in Supplementary Information, the measured resistivity values could perfectly be rescaled by a factor \( 1.25 \pm 0.03 \) and corrected by a difference in residual resistivity of \( 0.22 \mu \Omega \cdot \text{cm} \). Heat and charge currents as well as the magnetic field were applied within the basal tetragonal plane. However, we did not consider the distinction between the [100] and the [110] directions within the basal plane. The parallel orientation of the magnetic field, supplied by a superconducting solenoid, to the heat and charge flow allows us to neglect the contributions of transverse effects (Nernst and electrical/thermal Hall effects) in all measurements.

**METHODS SUMMARY**

The samples used in this work belong to the same batch and have been well characterized previously\(^ {1,2,4} \). Thermal and electrical transport coefficients were obtained from the same rectangular \((4.2 \times 0.5 \times 0.1 \text{ mm}^3)\) single crystal (sample 1) with the same contact geometry. This allows a reliable determination of the Lorenz ratio \( L(T)/L_0 = \rho(T)/T \), since the geometry factor \( L/A \) cancels out, where \( L \) and \( A \) are the length and the cross-section of the sample, respectively (see Supplementary Information). Additional measurements of the electrical resistivity were performed on a second single crystal from the same batch, but with a different geometry factor (sample 2). As described in Supplementary Information, the measured resistivity values could perfectly be rescaled by a factor \( 1.25 \pm 0.03 \) and corrected by a difference in residual resistivity of \( 0.22 \mu \Omega \cdot \text{cm} \). Heat and charge currents as well as the magnetic field were applied within the basal tetragonal plane. However, we did not consider the distinction between the [100] and the [110] directions within the basal plane. The parallel orientation of the magnetic field, supplied by a superconducting solenoid, to the heat and charge flow allows us to neglect the contributions of transverse effects (Nernst and electrical/thermal Hall effects) in all measurements.

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