Break up of the heavy electron at a quantum critical point

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The point at absolute zero where matter becomes unstable to new forms of order is called a quantum critical point (QCP). The quantum fluctuations between order and disorder\(^1\)–\(^5\) that develop at this point induce profound transformations in the finite temperature electronic properties of the material. Magnetic fields are ideal for tuning a material as close as possible to a QCP, where the most intense effects of criticality can be studied. A previous study\(^6\) on the heavy-electron material \(\text{YbRh}_2\text{Si}_2\) found that near a field-induced quantum critical point electrons move ever more slowly and scatter off one-another with ever increasing probability, as indicated by a divergence to infinity of the electron effective mass and cross-section. These studies could not shed light on whether these properties were an artifact of the applied field\(^7\)–\(^8\), or a more general feature of field-free QCPs. Here we report that when Germanium-doped \(\text{YbRh}_2\text{Si}_2\) is tuned away from a chemically induced quantum critical point by magnetic fields there is a universal behavior in the temperature dependence of the specific heat and resistivity: the characteristic kinetic energy of electrons is directly proportional to the strength of the applied field. We infer that all ballistic motion of electrons vanishes at a QCP, forming a new class of conductor in which individual
electrons decay into collective current carrying motions of the electron fluid.

Recent work\textsuperscript{6} on the heavy electron material YbRh\textsubscript{2}Si\textsubscript{2}\textsuperscript{9} has demonstrated that a magnetic field can be used to probe the heavy electron quantum critical point. This material exhibits a small antiferromagnetic (AFM) ordering temperature \( T_N = 70 \) mK (Fig. 1a) that is driven to zero by a critical magnetic field \( B_c = 0.66 \) T (if the field is applied parallel to the crystallographic \( c \)-axis, perpendicular to the easy magnetic plane)\textsuperscript{6}. For \( B > B_c \), a field-induced Landau Fermi Liquid (LFL) state characterized by \( \Delta \rho = AT^2 \) (where \( \Delta \rho (T) = \rho (T) - \rho_o \) is the temperature dependent part of the electrical resistivity) is established below some cross-over temperature \( T_0 (B) \) which grows linearly with field. The \( A \) coefficient, being proportional to the quasiparticle-quasiparticle scattering cross section was found to diverge as \( A(B) \propto 1/(B - B_c) \) for \( B \to B_c \). Comparative studies of the resistivity and the electronic specific heat \( C_e (T) = \gamma_o (B) T \) in the field ranges \( 0.5 \) T – \( 4 \) T (\( B \perp c \) with \( B_c = 0.06 \) T) and \( 2 \) T – \( 6 \) T (\( B \parallel c \)) revealed a field-independent ratio \( A/\gamma_o^2 \) slightly smaller than the empirical Kadowaki-Woods ratio\textsuperscript{13} that holds for LFL systems. This seemed to suggest a divergence of the effective quasiparticle (QP) mass as \( 1/(B - B_c)^{1/2} \) as \( B \to B_c \).

In this letter, we report the first-ever observation of the divergence of the QP mass at a QCP, established very close to \( B = 0 \).

By alloying YbRh\textsubscript{2}Si\textsubscript{2} with Germanium, using a nominal concentration \( x = 0.05 \), we have been able to fine-tune, in a new set of studies, the Néel temperature of this material and the critical field far closer to zero, to a point where, for the first time, we may reliably probe the zero-field transition using field-tuning. The phase diagram for a high-quality YbRh\textsubscript{2}(Si\textsubscript{0.95}Ge\textsubscript{0.05})\textsubscript{2} single crystal is shown in Fig. 1b. NFL behavior dominates over a funnel-shaped region of the \( T-B \) phase diagram down to the lowest accessible temperature of \( 20 \) mK. The critical field has been suppressed to as low as \( B_c = 0.027 \) T (\( B \perp c \)). As in the undoped material, there is a broad cross-over regime between the NFL and field polarized LFL regime with a mean cross-over temperature \( T_o \) that is seen to rise linearly with the field \( B \). Very weak AFM order develops in the \( x = 0.05 \) sample below \( T_N = 20 \) mK, as evidenced by the extremely weak anomaly in the electronic specific heat coefficient (Fig. 2a).
Past experience\textsuperscript{7,8} suggested that a finite field quantum critical point has properties which are qualitatively different to a zero field transition, shedding doubt on the reliability of these measurements as an indicator of the physics of a quantum phase transition at zero field. However, the zero–field properties of YbRh\textsubscript{2}(Si\textsubscript{1−x}Ge\textsubscript{x})\textsubscript{2} above $T \approx 70$ mK for the undoped ($x = 0$) and doped ($x = 0.05$) crystals are essentially identical (Fig. 2a), suggesting that by suppressing the critical field we are still probing the same quantum critical point. In both compounds, the ac-susceptibility follows a temperature dependence $\chi^{-1} \propto T^\alpha$ from 0.3 K to $\leq T \leq 1.5$ K, with $\alpha = 0.75$\textsuperscript{14}, and the coefficient of the electronic specific heat, $C_{el}(T)/T$, exhibits\textsuperscript{9} a logarithmic divergence between 0.3 K and 10 K. However, in the low-$T$ paramagnetic regime, i.e., $T_N < T \lesssim 0.3$ K, the ac-susceptibility follows a Curie-Weiss law (inset of Fig. 2a) with a Weiss temperature $\Theta_W \approx 0.3$ K, and a surprisingly large effective moment $\mu_{\text{eff}} \approx 1.4\mu_B$/Yb\textsuperscript{3+}, indicating the emergence of coupled, unquenched spins at the quantum critical point. The electronic specific heat coefficient, $C_{el}(T)/T$, exhibits a pronounced upturn below 0.3 K (Fig. 2a).

We now discuss the field dependence of the electronic specific heat in YbRh\textsubscript{2}(Si\textsubscript{0.95}Ge\textsubscript{0.05})\textsubscript{2} in more detail. In these measurements, magnetic fields were applied perpendicular to the crystallographic c-axis, within the easy magnetic plane (Fig. 2b). At fields above 0.1 T, $C_{el}/T$ is weakly temperature independent, as expected in a LFL\textsuperscript{15}. A weak maximum is observed in $C_{el}(T)/T$ at a characteristic temperature $T_o(B)$ which grows linearly with the field (inset of Fig. 3a), indicating that entropy is transferred from the low-temperature upturn to higher temperatures by the application of a field $B \geq B_c = 0.027$ T. As the field is lowered the temperature window over which $C_{el}(T, B)/T = \gamma_o(B)$ is constant shrinks towards zero and the zero-temperature $\gamma_o(B)$ diverges (Fig. 3a). For example, in a field of 0.05 T a constant value $\gamma_o(B) \approx 1.54(7)$ Jmol$^{-1}$K$^{-2}$ only develops below 40 mK. These results indicate the formation of a field-induced LFL state at a characteristic scale $T \lesssim T_o(B)$. As the window of LFL behavior is reduced towards zero, an ever increasing component of the zero-field upturn in the specific heat coefficient is revealed in the temperature dependence. This confirms that the major part of the upturn in the specific heat coefficient observed in zero field is electronic.
in character, and must be associated with the intrinsic specific heat at the QCP.

This conclusion is also supported by the electrical resistivity data which reveal a field-dependent cross-over from a $T$-linear resistivity at high temperatures, to quadratic behavior $\Delta \rho = A(B)T^2$ at sufficiently low temperatures. Most importantly, the data show that at low fields and temperatures, the same scale $T_o(b) \propto b$ (where $b = B - B_c$ is the deviation from the critical field) governs the cross-over from LFL to NFL behavior in both the thermodynamics and the resistivity. This can be quantitatively demonstrated by noting that the finite field transport and specific heat data collapse into a single set of scaling relations (see Fig. 3 inserts),

$$\frac{C_V}{T} = \frac{1}{b^{1/3}} \Phi \left( \frac{T}{T_o(b)} \right), \quad \frac{d\rho}{dT} = F \left( \frac{T}{T_o(b)} \right),$$

where $\Phi(x) \sim (\max(x, 1))^{-1/3}$ and $F(x) \sim x/\max(x, 1)$. The NFL physics is described by the $x \to \infty (T >> T_o(b))$ behavior of these equations, where $d\rho/dT$ is constant and $C_V/T \propto T^{-1/3}$. By contrast, the field-tuned LFL is described by the $x \to 0$ limit of these equations. Were there any residual pockets of LFL behavior that were left unaffected by the QCP, we would expect a residual quadratic component in the resistivity, and the data would not collapse in the observed fashion. We are thus led to believe that the break up of the LFL involves the entire Fermi surface.

From the second scaling relation in (1), we see that the $A$-coefficient of the $T^2$ term to the resistivity diverges roughly as $1/b$, a result that is consistent with earlier measurements on pure YbRh$_2$Si$_2$ carried out further away from the QCP coefficient $\gamma_o(b)$ grows as $b^{-1/3}$ (Fig. 2a). Notice that the field dependence at absolute zero temperature can be interchanged with the temperature dependence at $B = B_c$, but only in the upturn region. At high magnetic field deviations from the QCP, earlier measurements showed$^6$ that the Kad-owaki Woods ratio$^{13} K = A/\gamma_o^2$ is approximately constant. Closer to the QCP, where the scaling behavior is observed, $K = A/\gamma_o^2 \approx b^{-1/3}$ is found to contain a weak field dependence (Fig. 3b).

We now turn to discuss the broader implications of our measurements. The observed
divergence of both the $A$-coefficient and the coefficient $\gamma_o$ of the $T$-linear specific heat certainly rule out a 3D SDW scenario, which predicts that both quantities will remain finite at sufficiently low temperature in the approach to a zero-field QCP ($B \to B_c \to 0$), but it can be used to obtain still more insight into the underlying scattering mechanisms between the quasiparticles. In a 2D SDW scenario, the scattering amplitude between two heavy electrons is severely momentum dependent. When used to compute the transport relaxation rate, the SDW scenario leads to the result $A \propto 1/\kappa^2$, with $\kappa$ the inverse correlation length\(^\text{16}\). The observed divergence in $A(b)$ would require $\kappa^2 \propto b$. However, the fluctuations of the soft 2D spin fluctuations only produce a weak logarithmic renormalization in the heavy electron density of states, measured by the specific heat coefficient, $\gamma_o \propto \ln(1/\kappa)$. Thus the 2D SDW scenario predicts a weak divergence in the in–$T$ linear specific heat, but a strongly field dependent enhancement of the Kadowaki Woods ratio in the approach to a QCP ($b \to 0$), given by

$$\gamma_{SDW} \propto \ln(1/b) \quad \text{and} \quad K_{SDW} \propto \frac{1}{b \ln^2(b)}. \quad (2)$$

The strong violation of these predictions by our data, presented in Fig. 3a and 3b, rules out 2D spin fluctuations as the driving force behind the thermodynamics and the dominant source of scattering near the heavy electron QCP.

Taking a more general view, scaling behavior of the transport scattering rate tells us that the only scale entering into the density of states and the scattering amplitude is the single scale $T_o \propto b$ of the heavy electron fluid. A truly field–independent Kadowaki Woods ratio would indicate that the quasiparticle scattering amplitude has the form

$$A^* = T_F^* F[\{k_{in}\} \to \{k_{out}\}]. \quad (3)$$

The weak field dependence of the Kadowaki Woods ratio over a wide range of fields implies that the characteristic length scale of the most singular scattering amplitudes renormalizes more slowly in the approach to the QCP than expected in a SDW scenario.

Our data also provide some insight into the thermodynamics in the vicinity of the QCP.
By integrating the scaling form (1) for the specific heat over temperature, the entropy
\[ S(T) = \int_0^T dT' (C_{V}/T') \]
in the vicinity of the QCP can be described by the form
\[ S(T, B) = b^{1-\eta} S \left( \frac{T}{T_0(b)} \right) \quad (4) \]
where \( \eta = 1/3 \). The appearance of a field–dependent pre-factor in this equation forces
the entropy to vanish at the QCP, as required by the third law of thermodynamics. The
exponent in the pre-factor also determines the effective Fermi temperature
\[ T_F^*(b) \propto \gamma(b)^{-1} \propto T_0(b)^\eta \]
Thus the requirement that the entropy vanishes at the QCP (\( \eta < 1 \)) prevents a
direct proportionality between the Fermi temperature of the heavy LFL and the scale \( T_0(b) \)
governing the cross-over to NFL behavior. It follows that the Fermi temperature and cut-off
temperature \( T_0(b) \) must obey a relationship of the form
\[ T_F^*(b) = T_\Lambda \left( \frac{T_0(b)}{T_\Lambda} \right)^\eta \quad (5) \]
where \( T_\Lambda \) is an upper cut-off that we might identify with the single ion Kondo temperature
of the Yb\(^{3+} \) ions(\( \approx 25 \)K). Such a power law renormalization of the characteristic energy
scale would be expected in the presence of locally critical fluctuations that extend down
from \( T_\Lambda \) to the infra-red cut–off provided, in this case, by the magnetic field\(^{17} \).

In this respect, our results support the conclusions recently drawn from earlier measure-
ments on the quantum critical material \( CeCu_{6-x}Au_x \) (\( x = 0.1 \))\(^{18} \), and used in a recently
proposed theory for quantum criticality by Si \( et \) \( al. \)\(^4 \), suggesting that the most critical scat-
tering is neither three, two or even one dimensional, but local– as if the most critical fluc-
tuations in the underlying quantum phase transition are fundamentally “zero dimensional”
in character.

One of our most striking observations is that below \( T \approx 0.3 \) K where \( \chi(T) \) follows a Curie-
Weiss law, the electronic specific heat coefficient \( C_{el}(T)/T \) for both samples starts to deviate
towards larger values, separating away from the \( – \log T \) dependence that is valid\(^9 \) up to 10 K
(Fig. 2a). This “upturn” continues in the \( x = 0.05 \) sample down to approximately 20 mK, if
the critical field of 0.027 T (\( B \perp c \)) is applied. We ascribe this intrinsically electronic feature
to the critical fluctuations associated with the zero-field quantum phase transition that exists at a slightly larger Ge concentration. The unique temperature dependence of $C_\text{el}(T)/T$ for $T < 0.3$ K is disparate from the linear temperature dependence of the electrical resistivity which holds all the way from $\geq 10$ K to $T \approx 10$ mK. Since the former (thermodynamic) quantity probes the dominating local 4f (“spin”) part of the composite quasiparticles, while the latter (transport) quantity is sensitive to the itinerant conduction-electron (“charge”) part, one may view the observed disparity as a direct manifestation of the break up of the composite fermion in the approach to the QCP.
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YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$)$_2\n\text{B} \parallel c$

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FIG. 1. Evolution of $\varepsilon$, the exponent in $\Delta \rho(T) = [\rho(T) - \rho_o] \propto T^\varepsilon$, within the temperature–field phase diagram of YbRh$_2$(Si$_{1-x}$Ge$_x$)$_2$ single crystals. The non–Fermi–liquid (NFL) behavior, $\varepsilon = 1$ (yellow color), is found to occur at the lowest temperatures right at the quantum critical point (QCP), $B = B_c$ [a, $x = 0$, $B_c = 0.66$ T ($\parallel c$), residual resistivity $\rho_o = 1\mu\Omega\text{cm}$; b, $x = 0.05$, $B_c = 0.027$ T ($\perp c$), $\rho_o = 5\mu\Omega\text{cm}$] and in a largely extended field range at higher temperatures. For $B > B_c$, a broad cross–over regime from the NFL state to the field–induced heavy Landau Fermi–liquid (LFL) state (at lower temperature) is stated. The LFL state is characterized by $\Delta \rho(T) \propto T^\varepsilon$, $\varepsilon = 2$ (blue color). As shown in (a) the antiferromagnetically ordered phase of pure YbRh$_2$Si$_2$ below $T_N = 70$ mK and $B_c$ shows, owing to an extremely small ordered moment, the outward appearance of a heavy LFL state, too. Its phase boundary to the paramagnetic state is manifested by a rapid change in $\varepsilon$ from 2 to 1.

The low ordering temperature of pure YbRh$_2$Si$_2$ increases as external pressure is applied$^9$. The extrapolation of $T_N(p) \rightarrow 0$ yields a critical pressure $p_c = -0.3(1)$ GPa, reflecting that a small expansion of the unit cell volume, $V$, would tune $T_N \rightarrow 0$. This can be achieved by the substitution of Si by the isoelectronic, but larger, Ge$^{10}$. Studies of the electrical resistivity under pressure revealed a $T_N \propto (p + p_c)^n$ variation, with $n = 1.33$ for both compounds. The $T_N(p)$-dependences of the $x = 0$ and $x = 0.05$ crystals can be matched if all $x = 0.05$ data points are shifted by the same amount $\Delta p = -0.17(2)$ GPa to lower pressure$^{10}$, yielding $T_N = 20(5)$ mK. Using the bulk modulus $B_0 = 189$ GPa of YbRh$_2$Si$_2$$^{11}$, the small pressure shift of $\Delta p = -0.17(2)$ GPa is equivalent to a volume expansion of $\Delta V = 0.14(3)$ Å$^3$. This transforms into an effective Ge content $x_{\text{eff}} = 0.019(6)$, if the value $\Delta V/V = 7.65(78)$ % for the relative change of the unit-cell volume with Ge concentration in the solid-solution YbRh$_2$(Si$_{1-x}$Ge$_x$)$_2$ is used, with $V(x = 0) = 158.4(2)$ Å$^3$ and $V(x = 1) = 166.07(54)$ Å$^3$, cf. Ref.$^{12}$, in agreement with microprobe analysis$^{14}$. 

FIG. 2. Low–temperature electronic specific heat of YbRh$_2$(Si$_{1-x}$Ge$_x$)$_2$ single crystals as $C_{\text{el}}/T$ vs $T$ in semi–log plots at zero field and at low values of the applied magnetic field $B$. Insets show low–$T$ $B = 0$ ac–susceptibility as $\chi^{-1}$ vs $T$ (a) and magnetization as $M$ vs $B$ (b). $C_{\text{el}}$ is obtained by subtracting the nuclear quadrupolar contribution, $C_Q = \alpha_Q/T^2$ (with $\alpha_Q = 5.68 \times 10^{-6}$ JKmol$^{-1}$, calculated from recent Mössbauer results$^{11}$) (a) and, in addition, the nuclear Zeeman contribution $C_{\text{hf}} = \alpha(B)/T^2$ (b), from the raw data. Here, $\alpha(B)$ has been deduced by plotting $CT^2$ vs $T^3$. The magnetization, $M$ vs magnetic field $B$ (black points in the inset), is calculated via $(B_{\text{hf}} - B)/A$, with $A$ the hyperfine coupling constant for Yb in this compound and the hyperfine field $B_{\text{hf}} = \sqrt{(\alpha(B) - \alpha_Q)/\alpha_{\text{dip}}}$; $\alpha_{\text{dip}}$ represents the strength of the nuclear magnetic dipolar interaction and amounts to $7.58 \times 10^{-8}$ JKmol$^{-1}$T$^{-2}$, Ref.$^{19}$. With the assumption of $A = 120$ T/$\mu_B$, the data points agree perfectly with the measured magnetization curve at 40 mK (red line in the inset of (b)).

The $B = 0$ results shown in (a) reveal an upturn in $C_{\text{el}}(T)/T$ for paramagnetic YbRh$_2$(Si$_{1-x}$Ge$_x$)$_2$ ($x = 0, T_N = 70$ mK; $x = 0.05, T_N = 20$ mK) below $T = 0.3$ K. In the same temperature range the susceptibility $\chi(T)$ shows a Curie–Weiss law, $\chi^{-1} \propto (T - \Theta)$ [inset of (a)]. For both samples very similar values are found for the Weiss temperature, $\Theta \approx -0.3$ K, as well as for the large effective moment, $\mu_{\text{eff}} \approx 1.4\mu_B$/Yb$^{3+}$. For YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$)$_2$, entropy is shifted from low to higher temperatures when a magnetic field is applied (b). The cross–over temperature between the field–induced LFL state ($C_{\text{el}}(T)/T \approx \text{const.}$) and the NFL state at higher temperature is depicted by the position of the broad hump in $C_{\text{el}}(T)/T$ which shifts upwards linearly with the field, $B \leq 0.8$ T ($\perp c$).
FIG. 3. Field dependences of the Sommerfeld coefficient, $\gamma_o$, of the electronic specific heat (a) and of the ratio of the $A$-coefficient in the $T^2$ term of the electrical resistivity and $\gamma_o^2$ (b) for YbRh$_2$(Si$_{0.95}$Si$_{0.05}$)$_2$. Note that $\gamma_o$ and $A$ are proportional to the effective quasiparticle mass and the effective quasiparticle–quasiparticle scattering cross section, respectively. The magnetic field was applied perpendicular to the $c$–axis, and the applied field values are corrected, on the abscissae, by the value of the critical field, $B_c = 0.027$ T. $\gamma_o$–values in (a) were obtained from two different samples: Three independent measurements on sample #1 are displayed by closed symbols (circles, up and down triangles). The open symbols (diamonds) show the results of measurements on sample #2. As $B \to B_c$, $\gamma_o$ diverges $\propto (B - B_c)^{-0.33}$ (red line), i.e., much stronger than logarithmically (black dashed line). The symbols used in the semi–log plot $K = A/\gamma_o^2$ vs $(B - B_c)$ of (b) correspond to values for the electronic specific heat coefficient shown in (a). Half filled circles (squares) display data for which the $A$-coefficient of the electrical resistivity was determined by extrapolating (interpolating) $A(B - B_c)$ with respect to $(B - B_c)$. The half filled diamond represents a point for which the $\gamma_o$ value was obtained by interpolation. The black dashed line indicates $K_{SDW} \propto [(B - B_c)\ln^2(B - B_c)]^{-1}$ for the 2D SDW scenario$^{16}$. This is at strong variance from the (at $B - B_c < 0.3$ T) experimentally observed $K \propto (B - B_c)^{-1/3}$, arising from the stronger than logarithmic increase of $\gamma_o$ upon cooling (red line in a). For $(B - B_c) > 0.3$ T, $K$ becomes field independent within the error bars at a constant value of 5.4 $\mu\Omega\text{cm}\text{mol}^2\text{K}^2\text{J}^{-2}$ (green horizontal line in b). A similar high field behavior has been reported previously$^6$ on pure YbRh$_2$Si$_2$. Noteworthy, an almost identical value for $K$ was found. Insets show scaling behavior of the low–$T$ electronic specific heat where, according to equation (1), the ordinate is displaying $\Phi(B, T) = (B - B_c)^{0.33} C_{el}/T$, a, as well as of the temperature derivative of the electrical resistivity, $d\rho/dT$, b, as a function of $T/(B - B_c)$. 

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