Pristine quantum criticality in a Kondo semimetal

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The observation of quantum criticality in diverse classes of strongly correlated electron systems has been instrumental in establishing ordering principles, discovering new phases, and identifying the relevant degrees of freedom and interactions. At focus so far have been insulators and metals. Semimetals, which are of great current interest as candidate phases with nontrivial topology, are much less explored in experiments. Here we study the Kondo semimetal CeRu\textsubscript{4}Sn\textsubscript{6} by magnetic susceptibility, specific heat, and inelastic neutron scattering experiments. The power-law divergence of the magnetic Gr"unesien ratio reveals that, surprisingly, this compound is quantum critical without tuning. The dynamical energy over temperature scaling in the neutron response, seen throughout the Brillouin zone, as well as the temperature dependence of the static uniform susceptibility indicate that temperature is the only energy scale in the criticality. Such behavior, which has been associated with Kondo destruction quantum criticality in metallic systems, may well be generic in the semimetal setting.

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Quantum criticality is observed in many strongly correlated materials classes, with quantum spin systems, high-$T_c$ cuprate and iron pnictide superconductors, and heavy fermion metals being prominent examples. Among them, the quantum critical insulators are the best understood. For instance, in the insulating quantum magnet LiHoF$_4$ (Ref. 6), the experimentally detected quantum criticality is well described in terms of the Landau framework, i.e., by the critical fluctuations of the magnetic order parameter. In quantum critical metals, by contrast, the underlying physics is much richer. In some systems the Landau description works well, but in others it appears to fail. Theoretically, the charge carriers introduce new couplings to the order parameter or its underlying building blocks, which raises new possibilities for quantum criticality.

Here we explore the case of the Kondo semimetal CeRu$_4$Sn$_6$. Its noncentrosymmetric crystal structure (Fig. 1a), strong spin-orbit coupling associated with the three large Z elements, and the fact that the anisotropy in its electronic dispersion cannot be attributed to a nodal ground state wave function of the Ce$^{3+}4f^1$ electron have led to speculations that the material may be topologically nontrivial. Indeed, density functional theory (DFT) calculation within the local density approximation (LDA) plus the Gutzwiller scheme, predict the material to host Weyl nodes. If the nodal excitations persist in a full treatment of the Kondo effect, CeRu$_4$Sn$_6$ will be a Weyl-Kondo semimetal. Our comprehensive investigation of the magnetization, Gr"uneisen parameter, and inelastic neutron scattering as functions of temperature and magnetic field reveals that this material is quantum critical without tuning, behavior that is only rarely observed. This raises the exciting question as to whether a Weyl-Kondo semimetal phase may indeed be located nearby, nucleating out of the quantum critical fluctuations.

The semimetallic character of CeRu$_4$Sn$_6$ is evidenced by the weak temperature dependence of the electrical resistivity below about 30 K (Fig. 1b). We have also measured the temperature dependence of the specific heat $C_p$ (Fig. 1c) and the magnetization $M$, the latter with different magnetic fields $H$ applied along the two main crystallographic directions (perpendicular and parallel to $c$, Fig. 1d and e, respectively). The high-temperature anisotropy was recently shown to be due to single ion crystal field effects on the Ce$^{3+}4f^1$ electron. Deviations from this behavior below room temperature indicate a partial gapping of the electronic density of states and/or the onset of Kondo screening. Below 10 K, a strong field dependence is observed. The temperature dependence of the magnetic susceptibility $\chi = M/H|_{10\,\text{mT}}$ is not of simple Curie-Weiss type.
\[ \chi = C/(T - \Theta) \] but, instead, is well described by
\[
\frac{1}{\chi(T)} = \frac{1}{\chi(T = 0)} + \frac{(aT)^\alpha}{c}.
\]
with \( \alpha = 0.50 \pm 0.01 \) and \( \alpha = 0.78 \pm 0.03 \) for magnetic fields applied perpendicular and parallel to \( c \), respectively (Fig.1d,e insets). This is inconsistent with quantum criticality involving a Lorentzian fluctuation spectrum, where \( \alpha = 1 \) (Curie-Weiss law) is expected. The application of larger magnetic fields gradually restores Fermi liquid behavior, i.e., a temperature-independent low-temperature magnetization (Fig.1d,e main panels).

The magnetic Grüneisen ratio \( \Gamma_{\text{mag}} = -(\partial M/\partial T)/C_p \) is expected to diverge at any quantum critical point, as observed in a number of quantum critical heavy fermion metals. For CeRu\(_4\)Sn\(_6\), from the low-field (10 mT) magnetization and specific heat data presented above, we find \( \Gamma_{\text{mag}} \sim T^{-\epsilon} \) between about 0.4 and 4 K, with \( \epsilon = 1.43 \pm 0.07 \) and \( 1.62 \pm 0.11 \) for fields perpendicular and parallel to the \( c \) axis, respectively (Fig.2b,d), providing strong evidence for quantum criticality without tuning in CeRu\(_4\)Sn\(_6\).

The field and temperature scaling of the magnetization data underpins this assignment. In Fig.2c,d we plot \(-\partial(M/H)/\partial T \cdot H^\beta \) vs \( T/H^\gamma \) over more than two orders of magnitude in \( T/H^\gamma \), for fields between 10 mT and 0.5 T both perpendicular and parallel to \( c \). We find a good data collapse with the exponents \( \beta \) being equal to \( \alpha \) extracted from Fig.1d,e (insets) and \( \gamma = 0.35 \pm 0.02 \) and \( 0.43 \pm 0.02 \) for fields perpendicular and parallel to \( c \), respectively. The fitted exponents are internally consistent, as they satisfy a scaling relationship (see Supplementary Information). This kind of critical scaling, with a fractional exponent \( \alpha < 1 \), indicates that the system is at, or very close to, a beyond-Landau quantum critical point, and that magnetic field acts as a tuning parameter, with criticality at \( H = 0 \).

This is corroborated by our neutron scattering investigation presented next. The inelastic neutron scattering intensity is dominated by features broad in momentum space, with no apparent energy scale, setting the lower bound for a gap in the spin excitations to less than 0.1 meV. In Fig.3a-c we show the intensity distribution at 1.5 K, integrated from 0.2 meV to 1.2 meV, in the \((H0L)\), \((HK0)\), and \((HHL)\) planes, respectively. The energy profile along two high symmetry directions, \((00L)\) and \((H00)\), integrated over \( \pm 0.2 \) reciprocal lattice units (r.l.u.) perpendicular to these directions, is presented in Fig.3d,e. The strong peaks at \( H+K+L=\text{even} \) are tails of nonmagnetic elastic Bragg scattering. There is no apparent change in the momentum dependence with energy transfer, meaning that the \( Q \) and \( \omega \) dependence of the scattering cross section factorizes.
and there is no dispersion. This is clearly at odds with any description of the criticality in terms of
the fluctuations of an incipient symmetry-breaking order parameter. The broadness of the features
is especially pronounced along \((\frac{1}{2} + H \frac{1}{2} - H 0)\) (Fig. 3b) and, to a lesser extent, along the \((H00)\) direction for peak intensities at \(H + 0 + L = o d d\) (Fig. 3a) and symmetry-related directions where the scattering intensity simply follows the magnetic form factor of the \(Ce^{3+} f^1\) electrons. Scattering
that is momentum-independent along lines in the Brillouin zone is a sign of local (or Kondo de-
struction) quantum criticality \(^{12}\) and has also been observed in quantum critical \(CeCu_{5.9}Au_{0.1}\) (Ref. \(^{11}\)). The wavevector dependence of the scattering intensity away from these high-intensity lines is discussed below.

First, however, we investigate whether quantum critical scaling, as evidenced by the static uniform magnetic susceptibility \(\chi = (\partial M/\partial H)_{H \rightarrow 0} = \chi'(q = 0, \omega = 0, T)\) (Fig. 1d,e, insets), is also found in the dynamical spin susceptibility \(\chi(q, \omega, T)\). This would give rise to the form

\[
\chi(q, \omega, T) = \frac{c}{f(q) + (-i\hbar \omega + aT)^\alpha},
\]

where \(f(q)\) is an offset and \(\alpha = 1\) is expected for a Lorentzian fluctuation spectrum as prescribed
by the Landau order parameter description \(^{7,8}\), and a fractional exponent \(\alpha < 1\) points to its failure.
The dynamic structure factor \(S(q, \omega, T)\) measured by inelastic neutron scattering is related to the
imaginary part of \(\chi(q, \omega, T)\) which, near the ordering wave vector, should take the form

\[
\chi''(\omega, T) = \frac{1}{T^\alpha} \cdot g(h\omega/k_B T).
\]

We have determined \(S(\omega, T) \sim \chi''(\omega, T)\) by integrating the neutron scattering intensity for momentum transfers \(q\) about the \((100)\) wavevector, and plot it for temperatures between 0.1 K and 10 K, and energy transfers between 0.1 meV and 1.3 meV as \(S \cdot T^\alpha\) vs \(h\omega/k_B T\) in Fig. 4a. The best data collapse is found for \(\alpha = 0.6 \pm 0.1\) (minimum in \(\chi^2\), inset of Fig. 4a). This is consistent with the \(\alpha\) values determined from the static uniform magnetic susceptibility \((\alpha = 0.50 \pm 0.01\) for \(H \perp c\) and \(\alpha = 0.78 \pm 0.03\) for \(H \parallel c\)), in particular in view of the fact that the neutron scattering intensity is a directional average, taken around the \((100)\) wavevector, where spin fluctuations polarized along and perpendicular to \(c\) contribute in equal measures. Thus, also the inelastic neutron scattering data evidence quantum criticality beyond the long-wavelength fluctuations of an order parameter \(^{7,8}\), such as expected in the theory of Kondo destruction quantum criticality \(^{12}\).

Finally, we turn to the spatial profile of the critical fluctuations. To do so, we have fitted the
the experimental scattering intensity, corrected for the Ce form factor, in terms of Eqn. 2. The
resulting $f(q)$ in the (H0L) and (HK0) planes are shown in Fig.4b and c, respectively. For the rather featureless $(\frac{1}{2}+H \frac{1}{2}−H 0)$ “ridge”, the singular dynamical spin susceptibility discussed earlier corresponds to a vanishingly small $f(q)$ (see Fig.4c). Likewise, in the (H0L) plane, it is seen that $f(q)$ is vanishingly small for $H+0+L=\text{odd}$, where the dynamical spin susceptibility is also peaked. Along directions that move away from these ridges, the decreasing dynamical spin susceptibility corresponds to an increasing $f(q)$. We can understand these features of $f(q)$ in terms of the wavevector dependence of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction

$$J(q) = J_K^2 L(q, \omega = 0) = J_K^2 \sum_k \frac{f(\epsilon_k) - f(\epsilon_{k+q})}{\epsilon_{k+q} - \epsilon_k}.$$  (4)

Here, $J_K$ is the Kondo coupling strength and $L(q, \omega)$ the Lindhard function of the (uncorrelated) conduction electrons, which we have determined through LDA-DFT calculations, with one 4f electron per Ce$^{3+}$ ion placed in the ionic core. This “f-core” bandstructure, resembling that of LaRu$_4$Sn$_6$, characterizes the electronic structure of the spd conduction electrons that mediate the RKKY interaction between the 4f moments. The associated Fermi surface is shown in Fig.3f, from which we can identify nesting wavevectors that correspond to the $(\frac{1}{2}+H \frac{1}{2}−H 0)$ and $H+0+L=\text{odd}$ ridges. These can be seen as the broad planes perpendicular to the (00L) direction ($\Gamma$ to $M$) and between X points extending along (00L). At these wavevectors, collectively denoted as $Q$, the RKKY interaction is maximally antiferromagnetic. The Weiss temperature scale

$$\Theta(q) = J(q) - J(Q)$$  (5)

provides an understanding of the offset $f(q)$, when the latter is raised to the power $1/\alpha$. In other words, the neutron scattering intensity is maximal where the Weiss temperature $\Theta(q)$ has a minimum, implying that the RKKY interaction defines the $q$ space structure. Accordingly, the system may be near antiferromagnetic order mediated by the RKKY interaction, with an ordering wavevector at $H+K+L=\text{odd}$.

Beyond-Landau quantum criticality, as indicated by a fractional exponent $\alpha < 1$, has been documented in a handful of other heavy fermion compounds,\textsuperscript{11,12,26-28} Except for the case of the heavy fermion metal CeCu$_{5.9}$Au$_{0.1}$, a determination of $\alpha$ from inelastic neutron scattering—as provided here—has, however, remained elusive. Our discovery of beyond-Landau quantum criticality, in both the inelastic neutron response and the static uniform magnetic susceptibility, in a genuinely quantum critical semimetal implicates the generality of the phenomenon. Since this constitutes the first observation of this phenomenon in a semimetal, it will be important to
explore whether beyond-Landau quantum criticality is inherent to systems with reduced charge carrier concentration.

In heavy fermion systems, quantum criticality is generally observed at the $T = 0$ collapse of (antiferromagnetically) ordered phases. CeRu$_4$Sn$_6$, however, is quantum critical without tuning pressure, stoichiometry, or applied fields to a phase boundary. A natural question then arises: Is an antiferromagnetic phase nearby, as suggested by the minima of the Weiss temperature near possible antiferromagnetic ordering wavevectors, and can it be reached by tuning, for instance with pressure? And even more excitingly, do the quantum critical fluctuations give rise to new emergent phases, maybe unconventional superconductivity in analogy to $\beta$-YbAlB$_4$ (Ref. 29) or, in view of all the necessary conditions being fulfilled, even a Weyl-Kondo semimetal phase akin to that recently discovered in Ce$_3$Bi$_4$Pd$_3$ (refs 18–20)? This will require further experiments at lower temperatures and as function of tuning parameters, which we hope this work will stimulate.
FIG. 1: Characterization of CeRu$_4$Sn$_6$. (a) Unit cell with view along tetragonal direction $a = b$ (left), polyhedron around central Ce atom (center), and unit cell with view along $c$ of the noncentrosymmetric tetragonal $I42m$ crystal structure of CeRu$_4$Sn$_6$, with $a = 6.8810$ Å and $c = 9.7520$ Å (Refs. [30][31]). (b) Electrical resistivity vs temperature, with current within the tetragonal plane, evidencing the semimetallic character of CeRu$_4$Sn$_6$. (c) Low-temperature specific heat over temperature ratio, plotted on semi-logarithmic scales, evidencing a stronger than logarithmic divergence. (d) Magnetization over magnetic field, measured in different magnetic fields ($B = \mu_0H$) applied perpendicular the tetragonal $c$ axis, as function of temperature. The inset shows the inverse susceptibility, defined as $1/\chi = H/M|_{10\text{mT}}$, with a least-square fit with $1/\chi = 1/\chi_0 + c \cdot T^\alpha$ to the date below 10 K, with $\alpha = 0.50 \pm 0.01$. (e) Same as (d) for magnetic fields applied along $c$, with $\alpha = 0.78 \pm 0.03$. The fit was done below 3 K, where the noise in the data is small (note that $c$ is the hard direction, along which $\chi$ is much smaller).
FIG. 2: **Signatures of quantum criticality in thermodynamic properties of CeRu₄Sn₆.** (a) Magnetic Grüneisen ratio $\Gamma_{\text{mag}}$ vs temperature at $B = \mu_0 H = 10\, \text{mT}$, applied perpendicular to the $c$ axis, with a power law fit to the data (exponent $1.43 \pm 0.07$). (b) Same as a for magnetic fields applied along $c$, with exponent $1.62 \pm 0.11$. (c) Scaled negative temperature derivative of the magnetization over field ratio, $-\partial(M/H)/\partial T \cdot H^\beta$ vs scaled temperature, $T/H^\gamma$, for fields perpendicular to $c$, with exponents $\beta$ and $\gamma$ that collapse the data. (d) Same as c for magnetic fields applied along $c$. 
FIG. 3: Quasielastic neutron scattering from CeRu₄Sn₆. (a,b,c) Energy integrated neutron scattering intensity in the (H0L), (HK0), and (HHL) planes, respectively. Saturated intensity at H+K+L = even is residual Bragg intensity. Along (1/2+H 1/2−H0) and equivalent directions in b and, to a lesser extent, along (H00) centered at H+0+L = odd in a the scattering intensity is almost structureless. (d,e) Energy vs momentum along (00L) and (H00), respectively. The Q and ω dependence of the scattering cross section factorizes so that there is no dispersion and the signal continues below the resolution limit of the instrument. (f) “f-core” Fermi surface derived from LDA-DFT calculations.
FIG. 4: Dynamical scaling of CeRu$_4$Sn$_6$. (a) Scaled neutron scattering intensity, $S \cdot T^\alpha$, vs scaled energy transfer, $\hbar \omega / k_B T$. The inset shows the scaled $\chi^2_{\text{red}}$, which is minimal for $\alpha = 0.6$. (b,c) $f(q)$ (see Eqn. 2) in the (H0L) and (HK0) plane, respectively, obtained through a global fit of Eqn. 2 to the $q-E$ dependent data in Fig. 3 with a single consistent value of $\alpha$. The white areas correspond to blinded out data near the nuclear Bragg peaks, which are situated at $H+K+L = \text{even}$. Along $(\frac{1}{2}+H \frac{1}{2}-H0)$ in c and about (H01) in b, $f(q)$ is minimal, indicating maximally antiferromagnetic interactions (see text).
Methods
For our study, single crystals of CeRu$_4$Sn$_6$ were grown from self-flux, using the floating zone melting technique with optical heating as reported previously$^{32}$. The magnetic properties between 2 K and 300 K were measured in a SQUID magnetometer from Cryogenic Ltd., the data between 0.3 K and 2 K employed a $^3$He insert. High-temperature magnetization measurements up to 1000 K were performed with a vibrating sample magnetometer in a PPMS from Quantum Design. The $^3$He PPMS option was used for specific heat measurements in the temperature range from 0.3 K to 20 K.

Neutron scattering was performed with the MACS spectrometer at the NIST NCNR. For the momentum-space mapping, we co-aligned 3 g of single crystals and used incident energies from 3.7 meV to 5 meV and a fixed final energy of 3.7 meV, allowing for the use of Be and BeO filters before and after the sample, respectively, and providing an effective resolution better than 0.05 Å$^{-1}$ and 0.15 meV. For the $\omega/T$ scaling measurements, 2 g of single crystals were accommodated within the restrictions of a dilution refrigerator. We utilized a fixed final energy of 2.5 meV and incident energies up to 3.7 meV. A magnetic field of 10 mT was applied along (0K0) to avoid superconductivity in the aluminum sample mount and maintain good thermal contact.

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Supplementary Information

Scaling and exponent relationship

Here we derive a relationship between the scaling exponents, which we first summarize. The exponent $\alpha$ characterizes the temperature dependence of the magnetization, and is defined as the exponent in the power-law temperature dependence of the linear-response magnetic susceptibility

$$\chi(T) \sim \frac{1}{T^\alpha}.$$ (6)

We also introduce $\beta$ and $\gamma$, which describe the magnetic-field dependence of the nonlinear magnetization. The exponent $\gamma$ is defined by generalizing Eqn. 6 to the case when the magnetic field is not vanishingly small, namely by

$$\frac{M}{H} = \frac{1}{T^\alpha} f(T/H^\gamma)$$ (7)

which, when taking the limit $H \to 0$, recovers Eqn. 6. Additionally, the exponent $\beta$ is defined through the scaling of the temperature derivative of the magnetization as

$$\frac{\partial(M/H)}{\partial T} \times H^\beta = g(T/H^\gamma).$$ (8)

Based on scaling (dimensional analysis), Eqn. 7 implies that

$$\frac{\partial(M/H)}{\partial T} = \frac{1}{T^{1+\alpha}} f_1(T/H^\gamma),$$ (9)

which is equivalent to

$$\frac{\partial(M/H)}{\partial T} = \frac{1}{H^{\gamma(1+\alpha)}} g(T/H^\gamma).$$ (10)

Comparing Eqs. 8 and 10 gives rise to the relationship

$$\beta = \gamma(1 + \alpha)$$ (11)

among the three exponents.

We now apply this relationship to the exponents we have extracted for CeRu$_4$Sn$_6$. Consider first $H \perp c$. The fitted exponents are $\alpha = 0.5$, $\gamma = 0.35$, and $\beta = 0.5$. The product $\gamma(1 + \alpha) = 0.525$ is compatible with $\beta$ within about 5% (thus within the experimental error bars, see main text). For $H \parallel c$, the fitted exponents are $\alpha = 0.78$, $\gamma = 0.43$, and $\beta = 0.78$. The product $\gamma(1 + \alpha) = 0.765$ is consistent with $\beta$ within about 2%, again within experimental accuracy.

We note that we have assumed hyperscaling in the derivation of the scaling relationship. The hyperscaling is expected to be satisfied at interacting critical points. By contrast, Gaussian critical
points will contain dangerously irrelevant variables that may invalidate the hyperscaling for some observables. The fact that the fitted exponents obey the scaling relationship is consistent with the interacting nature of the quantum critical point that underlies the observations reported in the present work for CeRu$_4$Sn$_6$.

**Neutron Scattering**

Background from the sample environment was accounted for by subtraction of a linear combination of separate measurements of count rates for an empty sample can and a CYTOP-only standard (CYTOP is a fluorine-based epoxy that we used to secure the CeRu$_4$Sn$_6$ single crystals).

For incident energies below 3.7 meV, neutrons are incapable of undergoing Bragg diffraction with Al in and around the beam path, dramatically reducing the background associated with Bragg scattering from the split-coil magnet system.

Sample-out background scattering was subtracted to remove contamination at low scattering angles. To account for incoherent elastic nuclear scattering from the sample, a pseudovoigt function was fit to the high-Q incoherent elastic profile and scaled to the elastic line intensity with same magnitude of $Q$ as where our $E/T$ scaling fit was performed, at a point in the Brillouin zone where magnetic intensity is at a minimum. The data for $E/T$ scaling was collected at (100), and integrated $\pm 0.2 \text{ r.l.u.}$ along (H00) and (00L).

The modulation in intensity outside of the first zone by translation of a reciprocal lattice vector provides information on the microscopic origin of the scattering. The magnetic form factor, $F_{\text{mag}}$, modulates scattering intensity as $I(q + G) = I(q) \cdot |F_{\text{mag}}(q + G)|^2$ (Ref. 33). The response of unpolarized neutrons is then described by a linear combination of the components of the dynamic spin correlation function\(^{(33)}\) as

$$I(Q,\omega) = \sin^2 \theta \cdot S^{aa}(Q,\omega) + (1 - \sin^2 \theta \cos^2 \phi) S^{bb}(Q,\omega) + (1 - \sin^2 \theta \sin^2 \theta) S^{cc}(Q,\omega).$$

(12)

We find that $S^{aa} = S^{bb} = 2S^{cc}$ improves the correspondence with the observed intensity modulation. This modulation effectively accounts for a magnetic form factor which is the result of anisotropic hybridization and/or easy plane crystal field anisotropy. Previous field-dependent heat capacity measurements show a field-induced extra contribution below about 10 K, that is much larger for $H \perp c$ than for $H \parallel c$ (Ref. 32), consistent with our observation of a form factor and polarization factor which implies quasielastic fluctuations of easy-plane Ce 4$f$ electrons at low temperatures.