Destruction of Kondo effect in cubic heavy fermion compound \( \text{Ce}_3\text{Pd}_{20}\text{Si}_6 \)

J. Custers\(^1\), K.-A. Lorenzer\(^1\), M. Müller\(^1\), A. Prokofiev\(^1\), A. Sidorenko\(^1\), H. Winkler\(^1\), A. M. Strydom\(^2\), Y. Shimura\(^3\), T. Sakakibara\(^3\), R. Yu\(^4\), Q. Si\(^4\), and S. Paschen\(^1\)^* 

\(^1\) Institute of Solid State Physics, Vienna University of Technology, 1040 Vienna, Austria  
\(^2\) Physics Department, University of Johannesburg, Auckland Park 2006, South Africa  
\(^3\) Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan  
\(^4\) Department of Physics and Astronomy, Rice University, Houston, TX 77005, USA  

* e-mail: paschen@ifp.tuwien.ac.at

How ground states of quantum matter transform between one another reveals deep insights into the mechanisms stabilizing them. Correspondingly, quantum phase transitions are explored in numerous materials classes, with heavy fermion compounds being among the most prominent ones. Recent studies in an anisotropic heavy fermion compound have shown that different types of transitions are induced by variations of chemical or external pressure\(^1\)–\(^3\), raising the question of the extent to which heavy fermion quantum criticality is universal. To make progress, it is essential to broaden both the materials basis and the microscopic parameter variety. Here, we identify a cubic heavy fermion material as exhibiting a field-induced quantum phase transition, and show how the material can be used to explore one extreme of the dimensionality axis. The transition between two different ordered phases is accompanied by an abrupt change of Fermi surface, reminiscent of what happens across the field-induced antiferromagnetic to paramagnetic transition in the anisotropic YbRh\(_2\)Si\(_2\). This finding leads to a materials-based global phase diagram – a precondition for a unified theoretical description.
Quantum phase transitions arise in matter at zero temperature due to competing interactions. When they are continuous, the associated quantum critical points (QCPs) give rise to collective excitations which influence the physical properties over a wide range of parameters. As such, they are being explored in a variety of electronic materials, ranging from high $T_c$ cuprates to insulating magnets and quantum Hall systems\(^4\).\(^5\).

Heavy fermion compounds are prototype materials to study quantum phase transitions. Their low energy scales allow to induce such transitions deliberately, by the variation of external parameters such as pressure or magnetic field. Microscopically, electrons in partially-filled $f$ shells behave as localized magnetic moments. They interact with conduction electrons through a Kondo exchange interaction, which favors a non-magnetic ground state that entangles the local moments and the spins of the conduction electrons. They also interact among themselves through an RKKY exchange interaction, which typically induces antiferromagnetic order. The best characterized QCPs occur in heavy fermion compounds with anisotropic structures. Examples include the monoclinic CeCu\(_{6-x}\)Au\(_x\) (ref.\(^6\)), and tetragonal CePd\(_2\)Si\(_2\) (ref.\(^7\)), YbRh\(_2\)Si\(_2\) (ref.\(^8\)) and CeMIn\(_5\) ($M=\text{Co, Rh}$)\(^9\).

It has been known that tuning external parameters changes the ratio of the Kondo coupling to the RKKY interaction. Recently, the importance of a second microscopic quantity has been suggested. This is the degree of quantum fluctuations of the local moments, parameterized by $G$: magnetic order weakens with increasing $G$, as it would with enhancing the Kondo coupling $J_K$. These two quantities define a two-dimensional parameter space, which allows the consideration of a global phase diagram\(^10\). This global phase diagram is most clearly specified via the energy scale $T^*$ associated with the breakdown of the Kondo entanglement between the local moments and conduction electrons. So far $T^*$ has only been identified in tetragonal YbRh\(_2\)Si\(_2\) (refs.\(^8\)\(^11\)\(^12\)). It is believed that this energy scale not only provides a general characterization of the heavy fermion quantum criticality but also underlies the non-Fermi liquid behaviour and anomalous dynamical scaling\(^12\)\(^13\)\(^14\) observed in these and related materials.

In order to explore the additional axis of the global phase diagram, we take advantage of the fact that enhancing spatial dimensionality reduces the quantum fluctuation parameter $G$. Therefore, it would be invaluable to study the extreme three-dimensional cubic heavy fermion compounds and compare their quantum critical behaviour with that of the tetragonal and other non-cubic materials.
Here, we do so in the cubic heavy fermion compound Ce$_3$Pd$_{20}$Si$_6$. We show that this material undergoes a quantum phase transition at a readily accessible magnetic field, and are able to identify the Kondo breakdown energy scale $T^*$. This scale vanishes inside the ordered part of the compound’s phase diagram, thereby providing the first clear evidence for a Kondo breakdown in the three-dimensional part of the global phase diagram. Furthermore, we show that this vanishing scale is the origin of the non-Fermi liquid behaviour observed previously in this compound (Supplementary Information).

In the cubic crystal structure of space group $Fm\bar{3}m$, the Ce atoms occupy two different crystallographic sites, 4$a$ (Ce1) and 8$c$ (Ce2), both with cubic point symmetry (Fig. 1a). This structure persists down to at least 40 mK, as shown by high-resolution neutron diffraction measurements. The magnetic susceptibility $\chi(T)$ is Curie-Weiss like above 100 K (Fig. 1b), with an effective moment close to the full moment (2.54 $\mu_B$/Ce) of the $J = 5/2$ spin-orbit ground state. A clear anomaly can be seen in $\chi(T)$ somewhat below $T_N$ (Fig. 1c). The electrical resistivity $\rho(T)$ is typical of heavy fermion compounds. $\Delta\rho$, the resistivity with the phonon-scattering contribution subtracted, shows a $-\ln(T)$ behaviour due to incoherent Kondo scattering at high temperatures. The maximum at about 20 K signals the onset of Kondo screening (Fig. 1d). Also the Hall coefficient shows the typical heavy fermion behaviour at high temperatures (Fig. 1e). Between room temperature and 50 K it is well described by $R_H(T) = R_0 + R_A(T)$, where $R_0$ represents a temperature independent normal Hall coefficient and $R_A(T)$ an intrinsic skew-scattering term. At low temperatures, $R_A$ becomes small and the measured Hall coefficient is dominated by the normal Hall component (Supplementary Information).

The specific heat $C(T)$ reveals that, in addition to the phase transition at $T_N$ that is also seen in $\chi(T)$, there is a second phase transition at $T_Q$ (Fig. 1f, ref. 17). The upper transition at $T_Q$ has been tentatively attributed to antiferro-quadrupolar order at the 8$c$ site, and the lower transition at $T_N$ to magnetic order — presumably antiferromagnetic order in analogy with Ce$_3$Pd$_{20}$Ge$_6$ (ref. 20). These assignments are consistent with the $\Gamma_8$ quartet and the $\Gamma_7$ doublet of the crystalline electric field split ground states of the Ce 4$f$ orbitals on the 8$c$ and 4$a$ sites, respectively. The temperature–field phase diagram is shown in Fig. 1g. $T_Q$ is initially enhanced by the applied field but is eventually reduced at larger fields. According to measurements on single crystals, magnetic field is able to completely suppress $T_Q$ (ref. 19), suggesting the presence of a QCP at fields above 10 T. Within the ordered region $T_Q(B) > 0$,
$T_N$ can be seen to decrease monotonically and vanish at about 0.9 T. This specifies a readily accessible QCP, thereby providing a rare opportunity to study quantum phase transitions in cubic heavy fermion materials.

It follows from general symmetry considerations that antiferro-quadrupolar ordering preserves the cubic symmetry of the lattice, as described in Supplementary Information. In addition, such considerations as well as microscopic calculations show that antiferro-quadrupolar order in the presence of magnetic field induces dipolar order, thereby influencing the antiferromagnetic correlations. The induced antiferromagnetic order, in turn, implies a magnetic-field tuning of the antiferro-quadrupolar transition temperature (Fig. S5 of Supplementary Information), which is compatible with the experimentally observed phase diagram (Fig. 1g).

Through the magnetic coupling between the 8c and 4a sites, the entire ordered region will contain both magnetic and quadrupolar orders. Finally, in the absence of the competition by the RKKY interactions, the ground state multiplets at both sites will be quenched by their Kondo couplings with the conduction electrons.

Studying the isothermal control parameter dependence of transport properties is a well established means to probe the quantum critical fluctuations near a QCP. Fig. 2a-c shows selected isotherms of the Hall resistivity $\rho_H$ as a function of the applied magnetic field $B$. At the lowest temperatures $\rho_H(B)$ shows two kinks (Fig. 2a). One of these persists as a broadened feature at temperatures above $T_N$ (Fig. 2b,c). To quantify these features we fit the data with crossover functions (Methods), shown as lines in Fig. 2a-c. At temperatures above $T_N$, this procedure identifies the crossover field $B^*$, as well as the full width at half maximum FWHM and the step height $\Delta A = |A_1 - A_2|$ of the crossover. Below $T_N$, the fitting characterizes in addition the crossover at the Néel transition. The fitted quantities are shown in Fig. 3a-c for the crossover at $B^*$ and in Fig. S3 of Supplementary Information for the crossover at $B_N$. Broadened kinks in $\rho_H(B)$ correspond to broadened steps in the differential Hall coefficient, defined as the field derivative of the Hall resistivity $d\rho_H/dB$. This is shown in Fig. 2d for the crossover at $B^*$.

The features in the Hall resistivity have their counterparts in the longitudinal and transverse magnetoresistance $\rho_l(B)$ and $\rho_t(B)$ (Fig. 2e,f). The $B^*$ crossover appears as a broadened step-like decrease of the resistivity with increasing field. Below $T_N$, the $B_N$ crossover is seen as an increase of the resistivity with field at small fields. The resistivity also contains a component that increases more gradually with field. This is identified as a background...
contribution (Methods). The resistivity data with the background and the anomaly at \( T_N \) subtracted are shown in Fig. 2g,h and are fitted by the same crossover functions that describe the differential Hall coefficient. The fit parameters are also shown in Fig. 3a-c and Fig. S3 of Supplementary Information.

Fig. 3 demonstrates our key conclusions. \( T^*(B) \) – which is equivalent to \( B^*(T) \) – defines a crossover scale that is distinct from any phase transition line, except in the zero temperature limit where it merges with \( T_N(B) \) at a common critical field of about 0.9 T (Fig. 3a). The \( T^*(B) \) scale exists both outside and within the ordered part of the phase diagram delimited by the upper ordering temperature \( T_Q(B) \). The FWHM of the crossover decreases with decreasing temperature, extrapolating to zero in the zero temperature limit as evidenced by the pure power-law behaviour of FWHM(\( T \)) (Fig. 3b). At the same time the step height \( \Delta A \) remains finite in the zero temperature limit (Fig. 3c). Because the Hall effect measures the response of the electronic excitations near the Fermi surface, the crossover at nonzero temperatures and the jump in the extrapolated zero-temperature limit are most naturally interpreted in terms of a collapse of the heavy fermion Fermi surface to a strongly reconstructed one. This implicates the \( T^* \) line as signifying a Kondo breakdown, which is tantamount to a localization of the \( f \) electrons.

The observation of the collapsing Kondo breakdown scale implies new quantum critical excitations which are neither of the Landau Fermi liquid nor of the spin density wave QCP type. Instead, electronic excitations over the entire Fermi surface are expected to have a non-Fermi liquid form. In fact, the electrical resistivity is linear in \( T \) and the electronic specific heat coefficient \( \Delta C(T)/T \) is logarithmic in \( T \) (refs. 17, 27, Supplementary Information). Both are defining characteristics of non-Fermi liquid behaviour which appears also in other materials with Kondo breakdown QCPs. At magnetic fields away from \( B^*(T = 0) \), a Fermi liquid \( T^2 \) temperature dependence of the electrical resistivity is recovered at low temperatures; measurements at several magnetic fields (up to 5 T) suggest that the corresponding temperature scale \( T_{FL} \) gradually decreases as \( B \) approaches \( B^*(T = 0) \).

A collapsing Kondo breakdown scale has been observed in YbRh\(_2\)Si\(_2\) (refs. 8, 11, 12). In that case the \( T^* \) line merges with the zero temperature boundary between paramagnetic and ordered phases, thereby signaling the destruction of the Kondo effect and concomitant reconstruction of the Fermi surface at the onset of magnetic order. However, in Ce\(_3\)Pd\(_{20}\)Si\(_6\) the \( T^* \) line enters an ordered phase at finite temperature. We interpret this distinction as
due to the different dimensionality of the two compounds.

Spatial dimensionality modifies the degree of fluctuations, including that of the quantum magnetism associated with the $f$ moments. This is illustrated by the two-parameter global phase diagram shown in Fig. 4. The horizontal axis marks the strength $J_K$ of the Kondo coupling between the local $f$ moments and the conduction electrons. It controls the degree of quantum fluctuations due to spin flip processes associated with the Kondo coupling. The vertical axis $G$ describes the degree of quantum fluctuations within the local moment component.

Going from the three-dimensional (3D) cubic limit to the decoupled 2D limit amounts to moving upwards along the vertical axis. The tetragonal structure of YbRh$_2$Si$_2$ suggests that it is close to the 2D limit, with enhanced $G$, making it natural to have the ordered to paramagnetic phase boundary coinciding with the Kondo collapse. The cubic structure of Ce$_3$Pd$_{20}$Si$_6$ implies a smaller $G$, placing it in the part of the global phase diagram where Kondo collapsing occurs inside the ordered part of the phase diagram. Here, the competition between the RKKY and Kondo couplings gives rise to a $T^*$ line which separates two ordered ground states. Note that, at finite temperature, the $T^*$ line is distinct from the ordering transition lines. In the zero-temperature limit, it separates a Kondo-screened order at $B > B^*$ and a Kondo-destroyed order at $B < B^*$. Through the field-induced mixing of the antiferro-quadrupolar and antiferromagnetic orderings (Sec. E of Supplementary Information), this corresponds to the region of the $G$–$J_K$ phase diagram in Fig. 4 where a Kondo-destruction QCP (brown line) occurs within the ordered part of the phase diagram, from a phase AF$_S$ to a phase AF$_L$.

Our results provide a way to think about other quantum critical heavy fermion metals, in line with recent theoretical considerations$^{3,10,28,29}$. CeIn$_3$ is another cubic system placing it in a similar part of the vertical axis as Ce$_3$Pd$_{20}$Si$_6$. There is some indication for the divergence of the effective quasiparticle mass inside the ordered part of its phase diagram$^{30}$ making it instructive to search for the $T^*$ scale in that system. The recently designed CeIn$_3$/LaIn$_3$ superlattice$^{31}$ amounts to an elegant reduction of the dimensionality towards the extreme 2D limit. Resistivity measurements have already provided evidence for a reduced strength of magnetic ordering. It will also be illuminating to explore the possibility of a Kondo breakdown. Finally, there are materials which have 2D lattices that host geometrical frustration such as the Shastry-Sutherland lattice in Ce$_2$Pt$_2$Pb (ref. $^{32}$). It is possible that
these materials have even higher $G$ making them promising candidates to shed light on the upper part of the global phase diagram.

Reconstruction of Fermi surface is also being extensively discussed in other electronic materials, including cuprate superconductors\(^{33}\). Typically, it is tied to antiferromagnetic ordering or other spontaneous symmetry breaking transitions. Here, in $\text{Ce}_3\text{Pd}_{20}\text{Si}_{6}$, we find Fermi-surface reconstructions both at the antiferromagnetic transition, the $T_N$ line, and away from it, at the $T^*$ line. While the former is smooth, the latter extrapolates to a jump of the Fermi surface in the zero-temperature limit. Our results amount to a rare demonstration of Fermi-surface reconstruction away from symmetry-breaking transitions. By extension, our findings highlight the emergence of novel electronic excitations through a mechanism other than spontaneous symmetry breaking, a notion that is of considerable current interest in a variety of settings including topological matters.

To summarize, we have observed an energy scale associated with the destruction of the Kondo effect and the concomitant $f$-electron localization in a cubic heavy fermion compound. This not only extends the materials basis for this effect to the 3D extreme but also unambiguously establishes that the origin of the $T^*$ scale lies in robust many body correlations, as opposed to materials specific band structure effects. Our findings suggest a materials based global phase diagram for heavy fermion systems, which not only highlights a rich variety of quantum critical points but also indicates an underlying universality. Given that quantum critical fluctuations represent an established route towards unconventional superconductivity, the insight we have gained will likely be important for the physics of high-temperature superconductors.

**Methods**

**Synthesis and sample selection.** The polycrystalline samples were prepared from high-purity elements (Ce 99.99%, Pd 99.998%, Si 99.9999%) by either ultra-high purity argon-arc or radio-frequency heating. Because of the excellent stoichiometry of these polycrystals they are of higher quality than the best available single crystals; this is evidenced by larger residual resistance ratios, sharper phase transition anomalies and higher transition temperatures in the polycrystal\(^{18,19,34}\). The phase transition temperature $T_N(B)$ is isotropic in field down to the lowest measured temperature\(^{19}\). We therefore chose these polycrystals for our investigation.

**Characterization.** The magnetotransport measurements were performed by a standard
4-point ac technique in an Oxford dilution refrigerator and, above 2 K, in a PPMS from Quantum Design. The magnetization measurements were performed by a capacitive technique at low temperatures and in a SQUID magnetometer of Cryogenic Ltd. above 2 K.

**Data analysis.** The crossover in the magnetoresistance at $B^*$ was fitted with the empirical crossover function

$$\rho(B) = A_2 - \frac{A_2 - A_1}{1 + (B/B^*)^p}$$

introduced in ref. 11, and the crossover at $T_N$ with the function

$$\rho(B) = A_2 - \frac{A_2 - A_1}{1 + e^{\frac{B - B_N}{\Delta}}}. \quad (2)$$

The latter function was chosen because it represents a single, symmetrically broadened step of height $\Delta A$ and width $w$ at the finite field $B_N$, that describes the data very well. The Hall resistivity was modeled with the integral over these fitting functions.

As discussed in the main text, a smooth overall increase of $\rho$ with only weak temperature dependence appears to be superimposed onto these two features. Measuring the magnetoresistance in both the longitudinal (field parallel to electrical current, Fig. 2e) and the transverse (field perpendicular to electrical current, Fig. 2f) configuration helps us to identify this latter as a background contribution due to normal magnetoresistance. It is featureless at the QCP and should be eliminated for the analysis of quantum criticality. We approximate the temperature dependent background by rescaling the background functions of the lowest temperature isotherms (grey lines in Fig. 2e,f) with the quadratic temperature dependence of the 15 T resistivity data observed at the lowest temperatures. After the subtraction of this background ($\rho_l,_{\text{back}}$ and $\rho_t,_{\text{back}}$) the crossover-related magnetoresistance approaches zero at high fields. An exemplary fit is shown in Fig. S2 of Supplementary Information.
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**Author contributions**

S.P. initiated the study. S.P. and Q.S. designed the research. A.St. and A.P. synthesized and characterized the material. J.C., K.L., M.M., and H.W. performed magnetotransport measurements, A.Si. and Y.S. magnetization measurements. T.S. led the low-temperature magnetization investigation. K.L., H.W., A.Si., and S.P. analyzed the data. R.Y. and Q.S. set up the theoretical framework and performed the calculations. S.P., Q.S., and R.Y. prepared the manuscript. All authors contributed to the discussion.

**Additional Information**

The authors declare that they have no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturematerials. Reprints and permission information is available online at [http://www.nature.com/reprints](http://www.nature.com/reprints) Correspondence and requests for materials should be addressed to S.P.
FIG. 1: Characteristics of the heavy fermion compound Ce₃Pd₂₀Si₆. a, Cubic crystal structure. b, Inverse volume susceptibility in SI units $1/\chi^{SI}$ in the linear response regime vs temperature $T$. A Curie-Weiss fit at high temperatures yields an effective moment $\mu = 2.35\mu_B$ per Ce atom and a paramagnetic Weiss temperature $\Theta = -3$ K. c, A maximum in $\chi^{SI}(T)$ somewhat below $T_N$ clearly reveals the Néel transition. Only a very weak feature can be discerned at the putative antiferro-quadrupolar transition at $T_Q$. d, Temperature-dependent electrical resistivity $\rho(T)$ and $\Delta\rho(T) = \rho(T) - \rho_{ph}(T)$, where the contribution due to phonon scattering $\rho_{ph}(T)$ is determined from the non-$f$ reference compound La₃Pd₂₀Si₆ as in ref.17. e, Temperature dependent Hall coefficient in the linear response regime $R_H(T)$, together with a fit according to the anomalous Hall effect model described in Supplementary Information, and its extrapolation to lower temperatures (dashed line). The normal Hall coefficient $R_0$, assumed as temperature independent in this model, is shown as grey line. f, Electronic contribution to the specific heat $\Delta C$ vs $T$ in an applied magnetic field of 0.5 T (from ref.17). The two anomalies at $T_N$ and $T_Q$ are clear signatures of second-order phase transitions. It is expected that their ordering wavevectors are different. As in ref.27 the transition temperatures are estimated by entropy balance constructions. g, Temperature-field phase diagram with the transition temperatures $T_N$ and $T_Q$, determined from transport and specific heat measurements, respectively.
FIG. 2: Magnetotransport across the quantum critical point of Ce$_3$Pd$_{20}$Si$_6$. a, Hall resistivity $\rho_H$ vs applied magnetic field $B$ at different temperatures below $T_N$. The solid lines represent fits of a crossover function (see Methods) to the data. b, c, Corresponding plots for intermediate and high temperatures. The crossover fields $B_N$ and $B^*$ of the fits are indicated in a-c. d, Field derivative of the fits of a-c, $d\rho_H/dB$, normalized to the step height $\Delta A$, vs normalized field $B/B^*$. The extrapolated zero-temperature form, a sharp step, is shown as grey line. e, Longitudinal magnetoresistance $\rho_l$ vs $B$ at different temperatures (80, 100, 125, 150, 175, 200, 250, 301, 350, 402, 500, 602, 700, 900 mK, 1.1, 1.3, 1.5, 1.7, 1.9, 2.9, 5.0 K). f, Corresponding plot for transverse magnetoresistance $\rho_t$ (89, 193, 300, 366, 486, 632, 743, 963 mK, 1.1, 1.3, 1.5, 1.7, 2.0, 3.0, 5.0 K). The grey curves in e and f represent the background (see Methods). g, Crossover component at $B^*$ (see Methods) of $\rho_l(B)$, normalized to the zero-field resistivity at the respective temperature, vs normalized field $B/B^*$. The data points are shown as dots, the fits as full lines. The grey line again represents the extrapolated zero-temperature form. h, Corresponding plot for $\rho_t(B)$.

FIG. 3: Characteristics of the Fermi surface collapse in Ce$_3$Pd$_{20}$Si$_6$. a, Temperature $T^*(B)$ of the crossover at $B^*(T)$, determined from the fits of $\rho_H(B)$, $\rho_t(B)$, and $\rho_l(B)$ in Fig. 2 plotted in the temperature-field phase diagram of Fig. 1g. b, Temperature dependence of the full width at half maximum, FWHM($T$), of the crossover. c, Temperature dependence of the step height, $\Delta A(T)$, of the crossover. The crosses refer to Hall resistivity data corrected for the anomalous Hall effect (Supplementary Information).

FIG. 4: Materials-based global phase diagram for heavy fermion compounds near antiferromagnetic instabilities. Magnetic frustration parameter $G$ (left) vs Kondo coupling $J_K$ at $T = 0$. Lines of quantum critical points separate antiferromagnetic (AF) from paramagnetic (P) regions (thick red line), and regions of small (S) and large (L) Fermi surface (brown line). The latter line represents quantum critical points accompanied by a Kondo breakdown. Dimensionality (right) helps to calibrate the placement of selected materials (CPS: Ce$_3$Pd$_{20}$Si$_6$, CeIn$_3$, a CeIn$_3$/LaIn$_3$ superlattice, YRS: YbRh$_2$Si$_2$, CCA: CeCu$_{6-x}$Au$_x$, CPP: Ce$_2$Pt$_2$Pb, marked on the $G$-axis by the ticks) along the vertical axis. The present work allows to elucidate the three-dimensional part of the phase diagram.
Figure 2
Figure 3
Figure 4