Multidimensional entropy landscape of quantum criticality

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The third law of thermodynamics states that the entropy of any system in equilibrium has to vanish at absolute zero temperature. At nonzero temperatures, on the other hand, matter is expected to accumulate entropy near a quantum critical point, where it undergoes a continuous transition from one ground state to another2,3. Here, we determine, based on general thermodynamic principles, the spatial-dimensional profile of the entropy $S$ near a quantum critical point and its steepest descent in the corresponding multidimensional stress space. We demonstrate this approach for the canonical quantum critical compound CeCu$_3$Au$_x$ near its onset of antiferromagnetic order. We are able to link the directional stress dependence of $S$ to the previously determined geometry of quantum critical fluctuations. Our demonstration of the multidimensional entropy landscape provides the foundation to understand how quantum criticality nucleates novel phases such as high-temperature superconductivity.

Quantum criticality arises near a second-order phase transition that is driven to zero temperature by competing interactions. For metallic systems, it provides a mechanism to generate new types of electron-derived excitations that are distinct from Landau’s Fermi liquid1. Because quantum fluctuations are enhanced when the dimensionality is reduced, quantum critical points (QCPs) often arise in anisotropic systems. The quantum critical fluctuations lead to unconventional scaling behaviour and the accumulation of entropy at very low $T$, thereby allowing unusual electronic excitations and new phases. The enhanced entropy $S$ upon approaching a QCP has been probed by measurements of the specific heat, and its dependence on pressure was studied by volume thermal expansion. The entropy landscape has been studied up to now using a single tuning parameter4. To understand how entropy evolves as the system traverses near a QCP, exploration of its profile in a multidimensional parameter space is needed.

Heavy-fermion systems represent prototype settings for QCPs induced by pressure. The latter tunes the hybridization of the almost localized 4f states with the conduction band, thereby tilting the balance in the competition between Ruderman–Kittel–Kasuya–Yosida (RKKY) and Kondo interactions. Previous experiments on quantum critical heavy-fermion systems5,6 focused on the volume expansivity $\alpha_V$ and volume Grüneisen ratio $\Gamma_V$. Both $\alpha_V/T$ and $\Gamma_V$ were found to diverge as $T \to 0$, indicating a diverging pressure dependence of $S$ and a vanishing entropy scale near the QCP as predicted7. Spatial anisotropy, a hallmark of many heavy-fermion systems, allows a QCP to be accessed with multiple tuning parameters.

Here we show that, for anisotropic systems, the directional dependence of the thermal expansivity provides a means to determine the spatial-dimensional profile of the thermodynamic singularities near a QCP. We establish a procedure to identify the combination of stresses that aims directly at the QCP and accomplishes the steepest change of the entropy $S$. We thereby can find the optimal way to approach the QCP and in principle directly link it with the geometry of the underlying quantum critical fluctuations.

We now specify the quantities of interest to our study. While the specific heat $C$ reveals the $T$ dependence of the entropy $S$, the linear thermal-expansion coefficients are related to its uniaxial pressure dependence: $\alpha_i = \partial \varepsilon_j / \partial T = -V^{-1} \partial S / \partial \sigma_j$. Here, $\varepsilon_i$ and $\sigma_j$ are strain and stress, respectively, along the principal crystallographic axes, for orthorhombic crystal structures: $i = a, b, c$ (see Supplementary Note). $V$ is the molar volume. If a system is dominated by a single energy scale $E^*$, as in a Fermi liquid, $C$ and $\alpha_i$ are proportional to each other. In this case, the proportionality factor, the Grüneisen ratio, related to the normalized stress dependence of $E^*$, $\Gamma_i(\sigma) = V\alpha_i(\sigma) / C = d \ln E^* / d \sigma_i$ is constant. The three components $\alpha_i$ are then proportional to each other—that is, their anisotropy is temperature independent, as $\Gamma_i / \Gamma_1 = \alpha_i / \alpha_1$ with $i, j = a, b, c$. The quantities define the volume expansivity $\alpha_V = \sum_i \alpha_i$ and volume Grüneisen ratio $\Gamma_V = \sum_i \Gamma_i$.

For our study of anisotropic quantum criticality, we choose the heavy-fermion compound CeCu$_3$Au$_x$, which is characterized by a strongly anisotropic structure with orthorhombic symmetry, space group Pnma (neglecting a minute monoclinic distortion, see Methods), and Ising-like magnetic anisotropy8. Consequently, all directional properties exhibit a considerable dependence on the crystal orientation. In addition, inelastic neutron scattering experiments9,10,11 provide evidence that the critical, incommensurate magnetic fluctuations at the QCP ($x_t \approx 0.1$) are of quasi-two-dimensional (2D) nature and form two sets of correlated planes that are spanned by $[0, 1, 0]$ and approximately $[0.73, 0.1, 0.68]$. Therefore, CeCu$_3$Au$_{0.1}$ constitutes an ideal platform to investigate the effect of anisotropic uniaxial pressures on quantum critical behaviour.

Figure 1a shows the 4f-electron contribution to the thermal-expansion coefficients $\alpha_i$ of CeCu$_3$Au$_{0.1}$ as a function of temperature in the range up to the Kondo temperature, $T_K \approx 6$ K. All $\alpha_i$ display characteristic features with slightly different positions in temperature. While $\alpha_c$ has a broad maximum at $\approx 1.5$ K, $\alpha_a$ and $\alpha_b$ show clear shoulders at $\approx 0.3$ K. In this $T$ range, $\alpha_a(\sigma)$ is negative while $\alpha_b(\sigma)$ and $\alpha_c(\sigma)$ are positive. The low-$T$ data below $\approx 1$ K cover the quantum critical regime. The data above 6 K change their anisotropy, reflecting the effect of the known crystalline-electric-field (CEF) excitations—that is, at $\approx 7$ meV, as found in inelastic neutron scattering and specific-heat measurements9.

In the quantum critical temperature regime, the linear thermal expansivities divided by temperature, $\alpha_i / T (i = a, b, c)$, are shown along each direction in Fig. 1b on a logarithmic $T$ scale. Each

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component $\alpha_i/T$ tends to diverge towards low temperatures, indicating non-Fermi-liquid behaviour. The same is also true for the volume thermal-expansion coefficient divided by temperature plotted in Fig. 1c. This should be contrasted to a Fermi liquid, where $\alpha_i/T$ is equal to $-V^{-1}dy_i/d\omega_i$, with the Sommerfeld coefficient $y_i = C/T$ expected to be constant at low $T$. The divergences of $\alpha_i/T$ versus $T$ for $T \rightarrow 0$ complement the well-known logarithmic divergence of $\gamma$ versus $T$ observed in the same compound, also shown in Fig. 1c. However, the divergences of $\alpha_i/T$ are much stronger than that of $y$, as predicted. Indeed, their ratio, the Grüneisen ratios $\gamma_i = V \alpha_i/C$, diverge for each direction $i = a, b, c$, as demonstrated in Fig. 2a. The same holds for the hydrostatic-pressure Grüneisen ratio $\gamma_p$ (see Fig. 2b).

We now turn to the analysis of the low-$T$ divergences. Because the Sommerfeld coefficient is best described by a logarithmic temperature dependence, we have fitted the linear thermal coefficients in terms of powers of logarithm. For all three directions, we find that the best representation of the data is given by $\alpha_i/T = a_i \ln^2(T_{0i}/T)$, with $a_i = -1.01(3), 1.92(3), 1.64(3) \times 10^{-6} \text{ K}^{-2}$, and $T_{0i} = 2.41(14), 2.43(7), 8.18(49) \text{ K}$ ($i = a, b, c$) (see solid lines in Fig. 1b). To obtain the best $\ln^2 T$ fit, a constant (Fermi-liquid like) contribution $a_{0i}/T = -0.60(8), 1.67(8)$, and $1.68(25) \times 10^{-6} \text{ K}^{-2}$ was assumed. The $T_{0i}$ are of similar magnitude to $T_{0i} = 6.33(7) \text{ K}$ extracted from $y = a \ln(T_{0i}/T)$ (ref. 10). $T_{0i}$ and $T_{0i}$ constitute cut-off temperatures for the quantum critical behaviour arising from the competition between Kondo and RKKY interactions and are, therefore, constrained to the Kondo energy scale. Thus, the different $T_{0i}$ might signal an anisotropy of the stress dependence of $T_K$ due to the shape of the $4f$ ground-state wavefunction. As $T_{0i}$ and $T_{0i}$ mark the upper limit of the non-Fermi-liquid regime, summing over $\ln^2(T_{0i}/T)$ with different $T_{0i}$ will at $T \ll T_{0i}$ still result in a $\ln^2 T$ dependence of $\alpha_i/T$, as indeed shown by the fit (solid line) in Fig. 1c. The fit parameters for $\alpha_i/T$ are $b = 2.70(5) \times 10^{-6} \text{ K}^{-2}$ and $T_{0i} = 4.69(22) \text{ K}$. Previous work on polycrystalline CeCu$_{6.5}$Au$_{0.5}$, which shows quantum critical behaviour in the specific heat closely resembling that of CeCu$_{6.5}$Au$_{0.5}$, reported a linear dependence of $\alpha_i/T$ versus $T$ for $0.07 \leq T \leq 0.5 \text{ K}$ (ref. 6).

The temperature dependence of our Grüneisen ratios $\gamma_i$ (Fig. 2), in line with the expectation, shows an approximate form of $\gamma_i = a_i \ln(T_{0i}/T)$. Because of $T_{0i} \neq T_0$, this dependence is only approximately observed at $T < 1 \text{ K}$. The deviation at higher $T$ is taken into account by adding a corrective term $2e_i + c_i/\ln(T_{0i}/T)$, with $c_i = \ln(T_{0i}/T_{0i})$.

To shed light on the origin of the logarithmic temperature dependence of $\gamma_i$, we first note that a divergent Grüneisen ratio is expected for any QCP: scaling dictates its $T$ dependence as $1/T^y$, where the exponent $y$ is the scaling dimension of the operator that tunes through the zero-temperature transition. For a spin-density-wave QCP, the expected exponent is $y = 1$, which is inconsistent with our measured Grüneisen ratio. In a local QCP involving a Kondo destruction, as arising in an Ising-anisotropic Kondo lattice appropriate for CeCu$_{6.5}$Au$_{0.5}$, the corresponding scaling dimension is $y = 0^+$ (see Methods), which is consistent with the observed logarithmic divergence of the Grüneisen ratio. We note that the model of critical spin fluctuations bootstrapped by energy fluctuations proposed $C/T \sim T^{-1/8}$ for the specific heat of a 2D QCP$^{12,13}$: since weak power-law and logarithmic $T$ dependencies are difficult to distinguish, a small-exponent power law is compatible with the $C/T$ data of CeCu$_{6.5}$Au$_{0.5}$; however, this same model predicted a $T^{-4}$ dependence for $\alpha_i/T$, which is much stronger than the observed $\ln^2 T$ dependence.

Regardless of the (model-dependent) analytical form of the divergence of the volume Grüneisen ratio, the natural question arises of how an anisotropic system such as CeCu$_{6.5}$Au$_{0.5}$ would respond to stress in an arbitrary direction. The above determination of $\alpha_i$ allows us to describe geometrically the components of the entropy derivatives with respect to stress $\partial S/\partial \sigma_i$ (Supplementary Note). For orthorhombic systems, the three normal stresses $\sigma_i$ are linearly independent of each other and span a Cartesian coordinate system. The steepest change of $S$ is given by the gradient that is formed by the components along each axis:

$$\nabla S = \left(\partial S/\partial \sigma_a, \partial S/\partial \sigma_b, \partial S/\partial \sigma_c\right)$$

This vector determines the stress combination that maximizes the entropy variation. The construction leads to an entropy landscape.
The dependence of $S$ on any arbitrary stress combination $\sigma_z$ can be expressed as the projection of $\nabla S$ onto the unit vector in the $\sigma_z$ direction:

$$\frac{\partial S}{\partial \sigma_z} = \nabla S \cdot \sigma_z / \sigma_z$$

This allows us to discuss the isotropic and anisotropic contributions to $\nabla S$, corresponding to the responses to the hydrostatic pressure and the so-called pure shear stresses, respectively (see Supplementary Note). The hydrostatic pressure is defined as $p = p \cdot (1, 1, 1)$, resulting in the well-known volume thermal expansion: $\alpha_T = -V^{-1} \nabla S \cdot p = -V^{-1} \nabla S \cdot \sum \sigma_i$.

The pure shear stresses are planar stresses, represented by combinations of two perpendicular uniaxial pressures of opposite sign, for example, $\sigma_{(ab)} = p \cdot (-1, 0, 1)$. As they are orthogonal to the hydrostatic pressure, $\sigma_{(ab)} \cdot p = 0$, they affect only anisotropic stress dependences. For an elastically homogeneous solid, their application results in the cancellation of the Poisson effect so that the shape of the solid is changed only in the $(ij)$ plane, while the volume remains constant (see Supplementary Note).

This leads us to analysing the responses to the pure shear stresses for an anisotropic system such as CeCu$_{5.9}$Au$_{0.1}$, which are not accessible by the hydrostatic-pressure or volume-dependent $\alpha_T$ and $\Gamma_V$. These stress combinations are proportional to the differences of the linear thermal-expansion coefficients along different, perpendicular directions: $\Delta S / \Delta \sigma_{(ij)} \propto \alpha_{(ij)} - \alpha_T$, where the hydrostatic, isotropic contributions $\alpha_T / 3$ cancel each other. Figure 4a shows the resulting differences divided by $T$. They exhibit contrasting scaling behaviour. Whereas $\alpha_{(ab)} / T$ and $\alpha_{(ab)} / T$ exhibit divergences with $\ln^2 T$ dependence, as do the single components and $\alpha_{(a)} / T$, $\alpha_{(b)} / T$ levels off below 1 K. Correspondingly, $\Gamma_{(ab)} = \Gamma_{(b)}$ becomes roughly constant at $T < 0.5$ K, thus seemingly pointing to a stability of the Fermi-liquid state for $T \to 0$ although the specific-heat coefficient $\gamma (T)$ clearly demonstrates non-Fermi liquid behaviour. This apparent dichotomy can be resolved by noting that none of the stress combinations perpendicular to $\nabla S$ (red circle in Fig. 3a) will tune the system to the QCP. In CeCu$_{5.9}$Au$_{0.1}$, it is accidental that one of the pure shear stresses is almost orthogonal to $\nabla S$. This is visualized by the fact that the direction of

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**Figure 3** | Entropy landscape of CeCu$_{5.9}$Au$_{0.1}$ at $T \approx 1$ K. **a.** Pictorial illustration of the anisotropic stress dependence of $S$. The red arrow represents the stress combination leading to the steepest slope $\nabla S$ aiming at the QCP. All perpendicular stress directions lie within the red plane and $\partial S / \partial \delta$ vanishes for these directions. The blue arrow indicates the hydrostatic pressure $p$. The blue plane perpendicular to $p$ represents the possible pure shear-stress combinations (thinner blue arrows for an orthorhombic Cartesian system $\sigma_{(ij)}$). As shown by the dashed red lines, $\nabla S$ has a hydrostatic component and one which is nearly parallel to $\sigma_{(a)}$. The pure shear stress $-\sigma_{(b)}$ is close to the intersection line between the two planes, and therefore approximately perpendicular to $\nabla S$. Accordingly, $\Gamma_{(b)}$ does not diverge at low $T$ as shown in Fig. 4. **b.** Two-dimensional analogue of the entropy landscape discussed for a given temperature. Tuning the system along directions orthogonal to the steepest slope $\nabla S$ (that is, following the contour lines) will leave the distance to the QCP unchanged.

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**Figure 4** | Pure shear stress dependences of CeCu$_{5.9}$Au$_{0.1}$ at $T = 1$ K. **a.** $\alpha_{(ij)} / T = \alpha_{(i)} / T - \alpha_{(j)} / T$ with $i \neq j = a, b, c$ as a function of $T$. The corresponding differences of the Grüneisen ratios $\Gamma_{(ij)} = \Gamma_i - \Gamma_j$ versus $T$. The solid lines are fits as described in the text.

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in the parameter space of directional stresses for each temperature. The result of this construction for CeCu$_{5.9}$Au$_{0.1}$ at $T = 1$ K is shown in Fig. 3a.

For a vanishingly small but nonzero temperature, the red arrow $\nabla S$ describes the steepest slope in the entropy landscape as the system is tuned towards the QCP, as illustrated for a 2D stress space $(\sigma_x, \sigma_y)$ in Fig. 3b. On the other hand, tuning along any direction perpendicular to $\nabla S$, marked by the red circle in Fig. 3a, will leave the distance to the QCP as a function of stress unaltered, and thus result in a vanishing critical contribution to the thermal expansion along this particular direction. From Fig. 1b we infer from the roughly constant ratios $\alpha_i / \alpha_j$ (i.e., $i = a, b, c$) that the direction $\nabla S$ is only weakly dependent on temperature. We note that exactly at the QCP, $\partial S / \partial \delta$ is strictly zero for any trajectory passing through the QCP at $\delta_c$. At $\delta_\Sigma$, $\partial S / \partial \delta$ changes its sign. Thus, our data indicate that the critical concentration $x_c$ is in fact a little larger than $x = 0.1$ for our sample.
the pure shear stress $\sigma_{yx}$ in stress space (that is, $(0, -1, 1)$) is close to the direction of the intersection line of the red and blue circles in Fig. 3a.

Knowing the entropy landscape of CeCu$_6$Au$_{0.1}$, we can ask how the anisotropic stress space connects with the geometry of the anisotropic quantum critical magnetic fluctuations determined by inelastic neutron scattering (the important differences between stress and strain for anisotropic systems are outlined in the Methods). To address this issue, we project the maximal stress shear stress applied to the $ac$ plane (see Fig. 3a), as can indeed be seen from Fig. 4b, showing $T_{(aa)}$ to be the largest $T_{ij}$. The application of this pure shear stress $\sigma_{yx}$ leads to a distortion of the $ca$ plane which is always normal to the planes of quantum critical fluctuations. This is, in fact, the only stress combination $\sigma_{yx}$/ $\sigma_{xy}$ which does not alter the distance between nearest-neighbour Ce atoms, but results in a tilt of the almost orthogonal fluctuating planes which does not alter the distance between nearest-neighbour fluctuations.

$\sigma_{yx}$ and $\sigma_{xy}$ would also be perpendicular to $p$. The components of this vector in the pure-shear-stress plane would be $\sigma_{yx}$ and $\sigma_{xy}$, with $a_x/a_y < 10\%$, as can be seen by the thinner red arrows in Fig. 3a. Physically, $\sigma_{yx}$ corresponds to a distortion of the $a-b$ plane (with $c = \text{const}$), and thus to a (smaller) change of the inclination angle between fluctuating planes. This merely reflects the fact that $\sigma_{yx}$ is not linearly independent. How these observations relate to the microscopic anisotropies of RKKY versus Kondo interactions requires a detailed determination of the electronic structure, which remains a challenge for the future.

Our determination of the entropy landscape in CeCu$_6$Au$_{0.1}$ explicitly demonstrates how entropy climbs as the system evolves towards its fully exposed quantum critical point in a multidimensional parameter space. The pronounced entropy enhancement renders quantum critical systems highly susceptible to the development of novel phases, such as unconventional superconductivity. This fundamentally promotes the understanding of superconductivity for heavy-fermion systems\textsuperscript{15}. Another prominent example along this line is the cuprates\textsuperscript{16}, where the entropy as a function of hole doping, $n_s$, peaks at the value where the $T_c(n_s)$ dome is maximum\textsuperscript{17}. This entropy maximum is found well above $T_c$ and, moreover, the anisotropy of the cuprates due to the quasi-2D electronic structure entails a strongly anisotropic stress dependence\textsuperscript{18}. Other pertinent systems with strong correlations include the iron pnictides and chalcogenides\textsuperscript{19}, and organic charge-transfer salts\textsuperscript{20}. In all these systems, novel phases emerge close to instabilities that are characterized by a strongly enhanced entropy in a phase space which is spanned up by multiple control parameters.

Methods

Methods, including statements of data availability and any associated accession codes and references, are available in the online version of this paper.

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Author contributions

K.G. and H.v.L. conceived the experiment, S.Z. and K.G. conducted the measurements, K.G. and H.v.L. performed the measurements, and K.G. and H.v.L. analyzed the data. All authors contributed to the writing of the manuscript.

Additional information

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Competing financial interests

The authors declare no competing financial interests.
Methods

Experimental set-up. The thermal-expansion measurements were carried out in a temperature range from 30 mK to room temperature by using a home-built dilatometer mounted in a dilution refrigerator (30 mK–90 K) and in a helium gas-flow cryostat (5–300 K). The raw thermal-expansion data were corrected for the expansion of the dilatometer by using measurements of Si and Cu single crystals. The investigated CeCu$_6$Au$_{1-x}$ single crystal was grown by the Czochralski method under a high-purity argon atmosphere, oriented with Laue X-ray diffraction, and spark-cut to yield a cube of approximately 3 mm edge length. For this length, the low-temperature resolution ($\Delta L/L$) of the measurements reached 10$^{-5}$. The measurements were performed along the three axes of the orthorhombic crystal structure (with orthorhombic axes notation), neglecting the very small monoclinic distortion setting in below $T_c$ ≈ 64 K with $\gamma_0 = 90.7^\circ$ for $T \ll T_c$ (refs 21–23). The 4f-electron contribution to the thermal expansivity was determined by subtracting the expansivities of an isostructural LaCu$_6$ single crystal$^{[1]}$.

Pressure versus volume—stress versus strain. Throughout the main part of this temperature range from 30 mK to room temperature by using a home-built experimental set-up.

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