

Supplementary Material for

The Dominant Role of Critical Valence Fluctuations on High T_c Superconductivity in Heavy Fermions

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Table S1

Figs. S1 to S14

References [72-76] cited in the Supplementary Material only

Supplementary Table S1

compound	$\rho_0(p = 0)$ [$\mu\Omega\text{cm}$]	ρ_0 (maximum T_c) [$\mu\Omega\text{cm}$]	T_{cr} [K]	maximum T_c [K]	ρ -data first published	T_{cr} from
CeCu ₂ Si ₂	~ 5	25.5(2.0)	-13(4)	1.84	[42]	Fig. S4
CeCu ₂ Si ₂	0.4	5.7(0.2)	-8(3)	2.33	[8]	[8]
CeCu ₂ Si ₂	see Fig. S5	8.5(0.2)	-3.7(7)	2.44	Fig. S5	Fig. S5
CeCu ₂ Ge ₂	~ 2	26(3)	13.6(6.0)	1.47	[72]	Fig. S6
CeAg ₂ Si ₂	5.5	21(2)	12.7(6.0)	1.25	[52]	[52]
CeAu ₂ Si ₂	1.8	9.8(0.2)	-14(6)	2.33	[18]	[18]
CeAu ₂ Si ₂	2.5	20(2)	-13(6)	1.40	[27]	[27]
CeAu ₂ Si ₂	12	43(1)	-25(8)	0.66	[26]	[26]
CePd ₂ Si ₂	~ 4	1.93(0.05)	-44(12)	0.40*	[56]	Fig. S7
CePd ₂ Si ₂	2.6	1.83(0.05)	-49(15)	0.34*	[56]	Fig. S8
CePd ₂ Si ₂	1.25	1.1(0.05)	-48(12)	0.441	[19]	Fig. S9
CePd ₂ Si ₂	0.6	0.44(0.05)	-55(15)	0.325	[19]	Fig. S10
CeCu ₆	12.4	-- [†]	-17(9)	-- [†]	[34]	Fig. S11
CeCu ₅ Au	28.9	33(0.5)	-6.5(1.5)	0.11**	[75]	Fig. S12
CeRhIn ₅	$0.2^{T=2\text{ K}}$	2.8(0.1)	-9(3)	2.26	[17]	[17]
CeRhIn ₅	0.011	1.1(0.05)	-8(3)	2.25	[17]	[17]
CeIrIn ₅	$1.7^{T=1.5\text{ K}}$	0.37(0.02)	-28(12)	1	Fig. S13	Fig. S13

Table S1. Experimental values of $\rho_0(p = 0)$, ρ_0 taken at the pressure of maximum T_c , T_{cr} , and maximum T_c of several Ce-HF samples. Note that “maximum T_c ” refers to the maximum value of the bulk-superconducting transition temperature as function of pressure in a given sample, except for two CePd₂Si₂* samples and CeCu₅Au**. In general, bulk superconductivity coincides with zero resistivity, in agreement with ac heat capacity or magnetic susceptibility signals [8,18,51,73]. Resistivity ρ was measured in the basal plane or along the a-axis of the tetragonal structure, except for CeCu₆ and CeCu₅Au, where ρ was investigated for the b-axis of the orthorhombic structure. * T_c from resistivity transition offset criterion, where ρ does not completely drop to zero [56]. ** T_c from resistivity transition onset criterion [75]. [†] By matching the (p - T) phase diagrams of CeCu₆ and CeCu₅Au, hypothetical SC in CeCu₆ corresponds to negative pressure $p \approx -0.5$ GPa.

Supplementary Figure S1

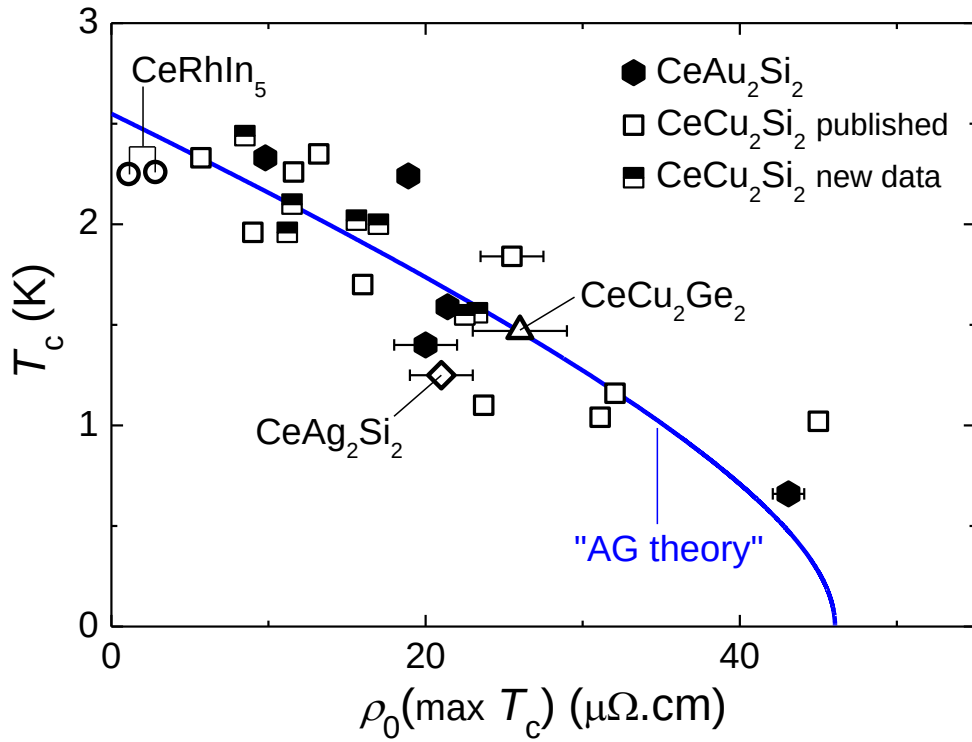


Fig. S1. Superconducting T_c vs. residual resistivity ρ_0 of the CeCu_2Si_2 -family and CeRhIn_5 (ρ_0 taken at the pressure of maximum T_c). All data points are from independent pressure experiments done in Geneva. Data for CeRhIn_5 from [17], CeCu_2Ge_2 from [72], and CeAg_2Si_2 from [52]. Filled hexagons refer to published data of CeAu_2Si_2 [18,26,27,51]. Empty cubes refer to published data of CeCu_2Si_2 [7,8,36,42,44,73,74]. Half-filled cubes refer to new data of CeCu_2Si_2 .

Supplementary Figure S2

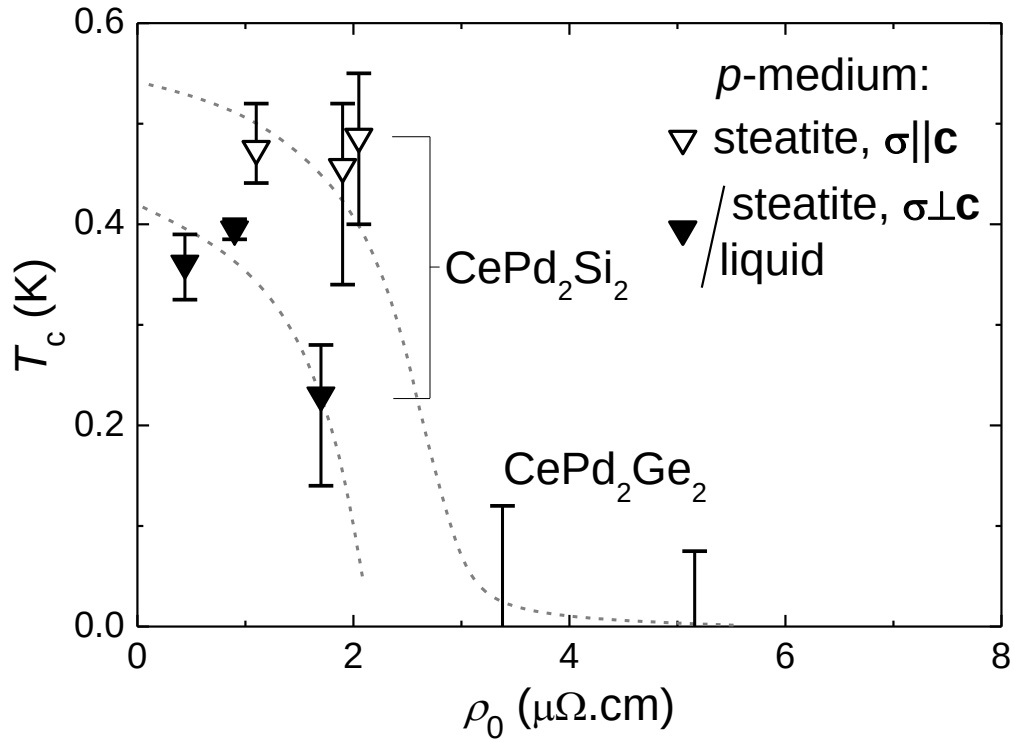


Fig. S2. Superconducting T_c vs. residual resistivity ρ_0 of CePd_2Si_2 [19,43,55,56] and CePd_2Ge_2 [76] (ρ_0 taken at the pressure of maximal T_c). Triangle symbols indicate the T_c defined at the 50% drop of ρ . The onset and end of the resistive superconducting transition are indicated by vertical bars. The dashed lines are guides to the eyes. An enhanced T_c due to strain effects [19] is observed in soft-solid steatite pressure medium with $\sigma \parallel \mathbf{c}$ compared to liquid pressure medium or steatite with $\sigma \perp \mathbf{c}$, and two data sets with qualitatively similar behavior can be distinguished.

Supplementary Figure S3

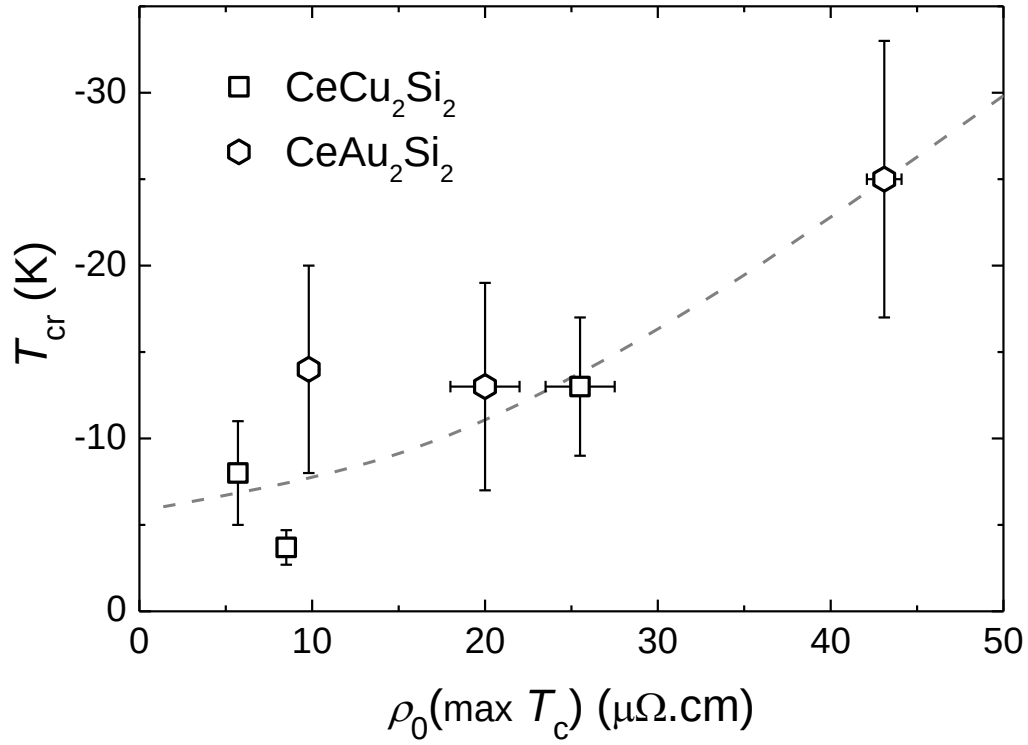


Fig. S3. Critical temperature T_{cr} vs. residual resistivity ρ_0 of CeCu_2Si_2 and CeAu_2Si_2 (ρ_0 taken at the pressure of maximal T_c). As qualitatively predicted by CVF theory [23], the two parameters ρ_0 and T_{cr} are correlated. Disorder induces an additional smearing of the VCO-induced resistivity collapse resulting in reduced experimental T_{cr} value.

Supplementary Figure S4

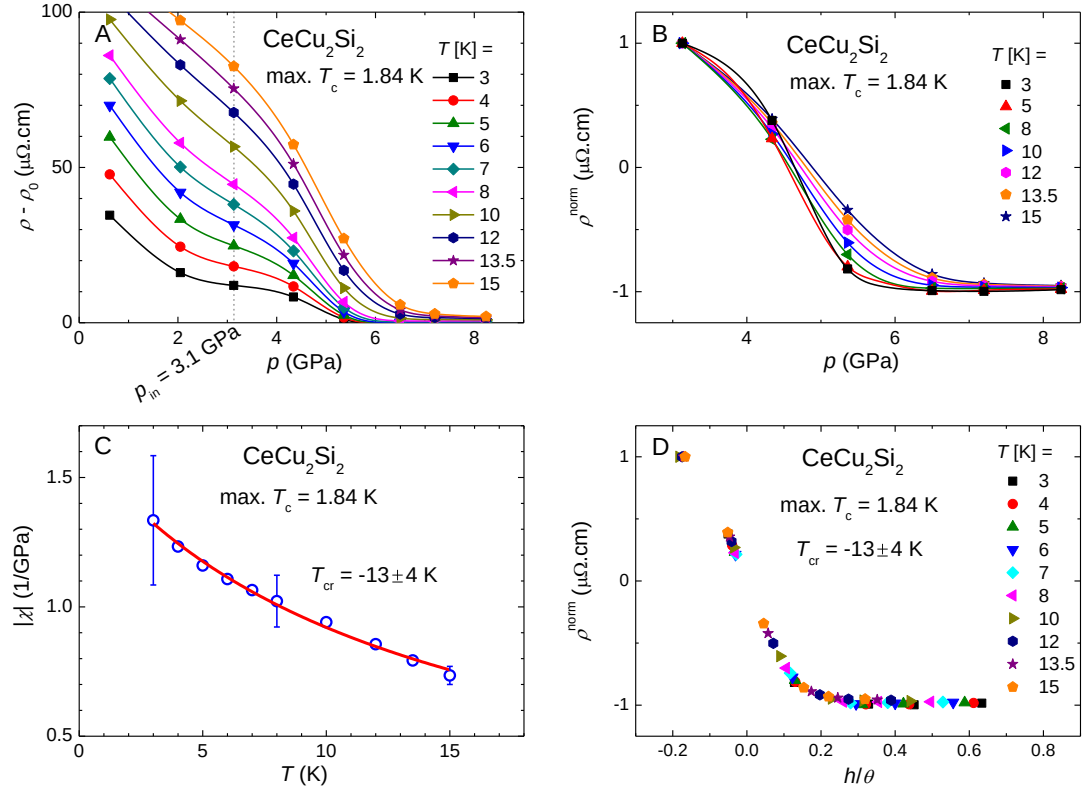


Fig. S4. Resistivity scaling analysis of a CeCu₂Si₂ sample with $T_c = 1.84$ K. $\rho(T)$ -data first published in [42]. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 3 up to 15 K. (B) Normalized resistivity ρ^{norm} vs. p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p\text{VCO}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -13 \pm 4$ K. Error bars on χ are shown for representative data points. (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ .

Supplementary Figure S5

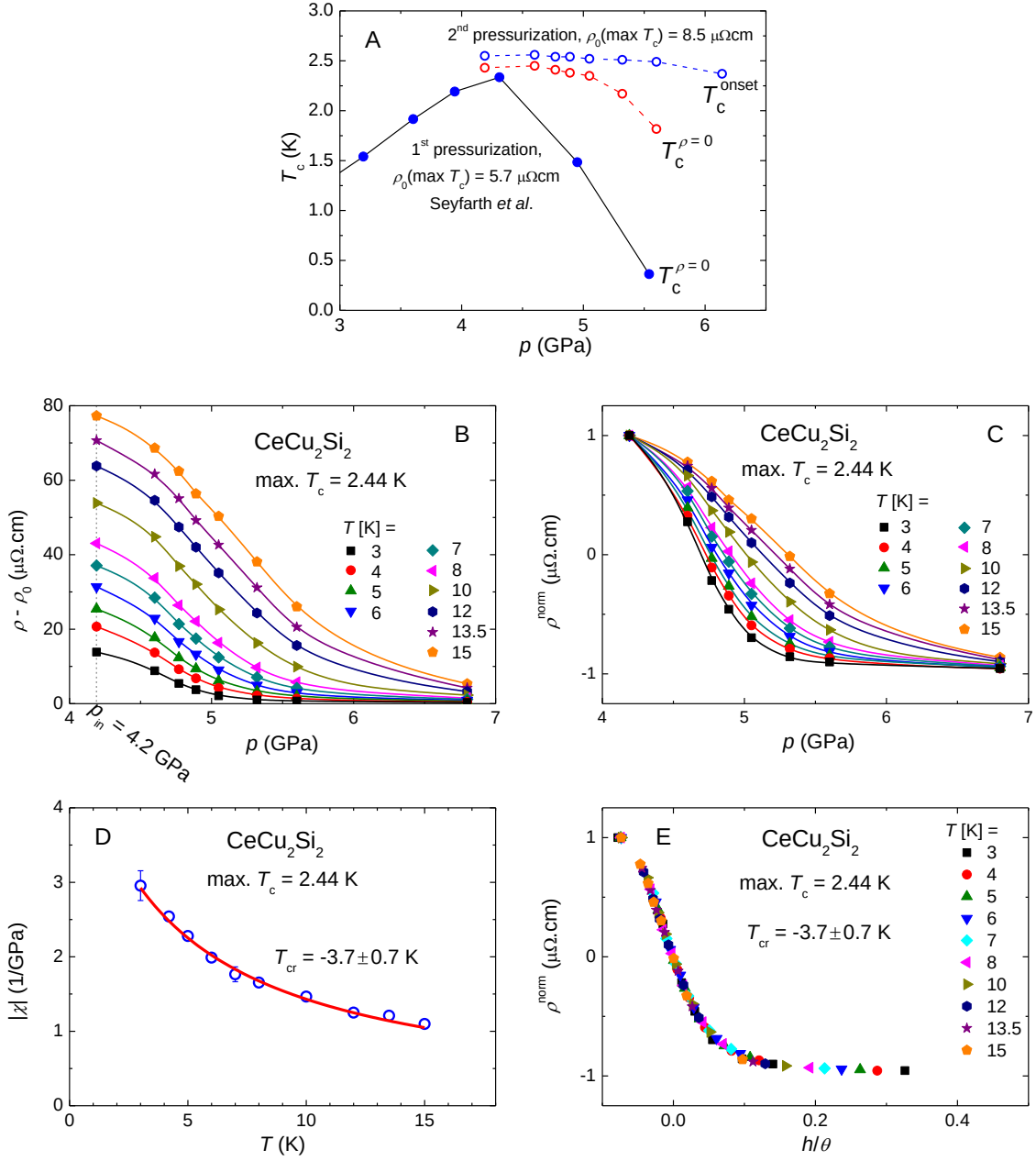


Fig. S5. Resistivity scaling analysis of a CeCu_2Si_2 sample with $T_c = 2.44$ K (record value!). Unpublished data of the second pressure cycle of Seyfarth *et al.* [8]. After a partial depressurization down to about 4 GPa, pressure was increased again by small steps for a better investigation of the VCO. (A) T_c vs. pressure of the first and second pressure cycles. Interestingly, the maximal T_c of the second cycle was a bit higher and the SC dome shifted up by about 0.5 GPa associated to a 50% increase in ρ_0 . (B) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 3 up to 15 K. (C) Normalized resistivity ρ^{norm} vs p . (D) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p\text{VCO}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -3.7 \pm 0.7$ K. Error bars on χ are shown for representative data points. (E) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ . With a high pressure-run density in the VCO regime, these data give the most accurate T_{cr} value.

Supplementary Figure S6

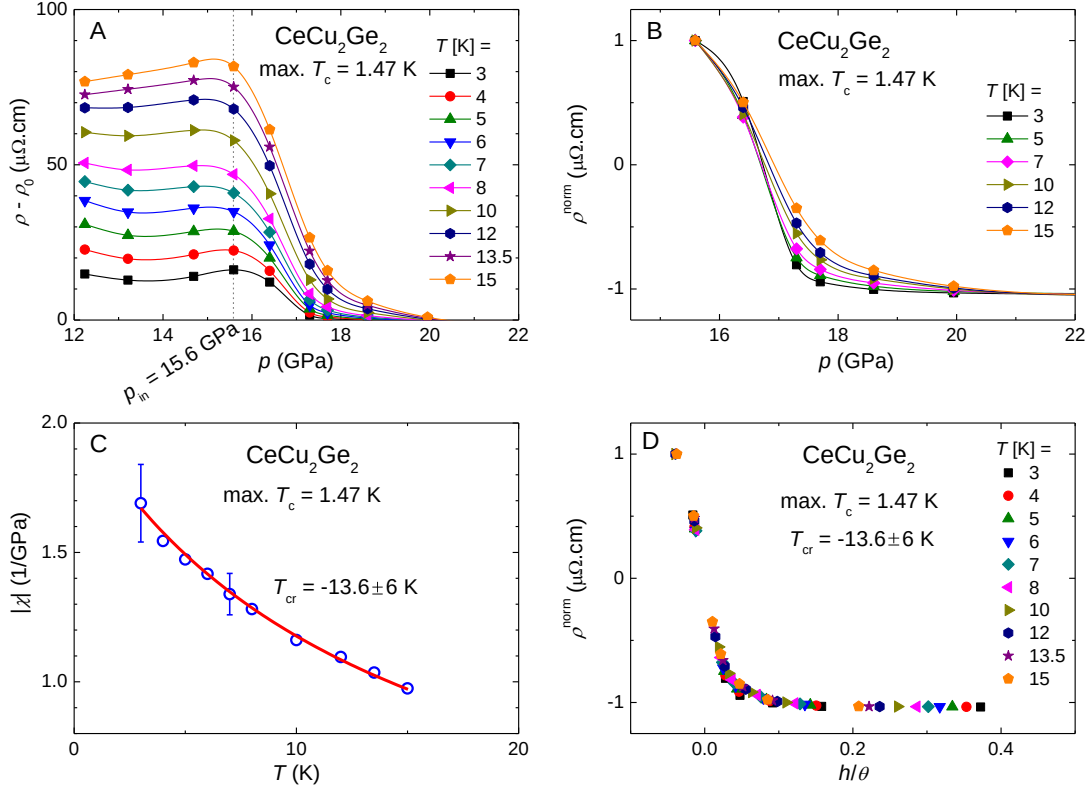


Fig. S6. Resistivity scaling analysis of a CeCu_2Ge_2 sample with $T_c = 1.47$ K. $\rho(T)$ -data first published in [72]. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 3 up to 15 K. (B) Normalized resistivity ρ^{norm} vs. p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p_{\text{VCO}}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -13.6 \pm 6$ K. Error bars on χ are shown for representative data points. (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ .

Comment on scaling analysis of CePd₂Si₂:

From the “raw” resistivity isotherms ρ^* (obtained in steatite p -medium) it is obvious that the resistivity collapse is slower than in the other compounds (see main text Fig. 2). Thus, the CEP should be at more negative temperature, which would imply a slower change of the slope χ as a function of T , and a bigger error on T_{cr} extracted by fitting with $\chi \sim 1/(T - T_{\text{cr}})$. Fortunately in CePd₂Si₂, the relatively high value $T_1^{\text{max}} \sim 100$ K at the pressure of the VCO allows to extend the analysis to higher temperature. By fitting $\chi(T)$ over an extended T -scale, the error of T_{cr} is reduced. Very robust scaling analysis is obtained for four CePd₂Si₂ samples (see Figs. S7-S10).

With a CEP at very negative temperature in CePd₂Si₂, the decrease of ρ^* is extended over a larger pressure scale compared to e.g. CeCu₂Si₂. For $T > T_N$, the decrease of ρ^* sets in already at pressures well below p_c . We have chosen p_{in} with the condition $p_{\text{in}} > p_c$ to go as low as possible in temperature to get the closest to the CEP. We have verified that the conditions $p_{\text{in}} < p_c$ and $T > T_N$ give similar results, with T_{cr} values within the error bar.

Supplementary Figure S7

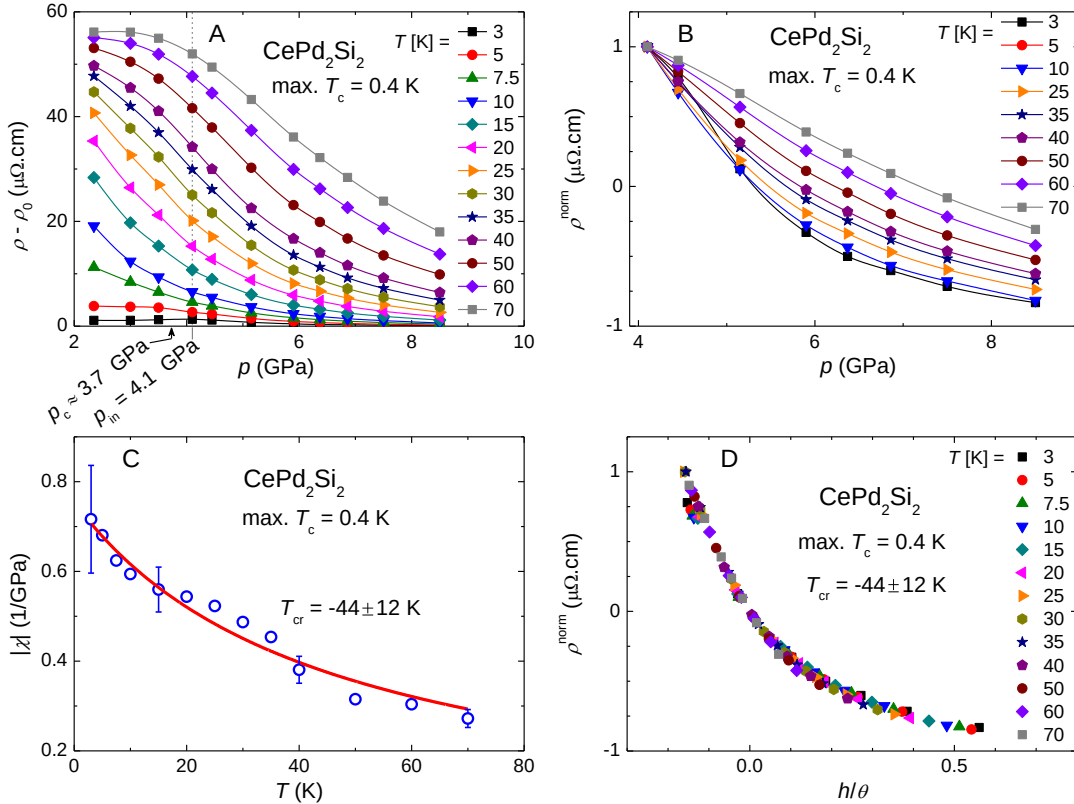


Fig. S7. Resistivity scaling analysis of a CePd₂Si₂ sample with $T_c = 0.4$ K. $\rho(T)$ -data first published in [56]. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 3 up to 70 K. (B) Normalized resistivity ρ^{norm} vs. p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p_{\text{VCO}}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -44 \pm 12$ K. Error bars on χ are shown for representative data points. (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ .

Supplementary Figure S8

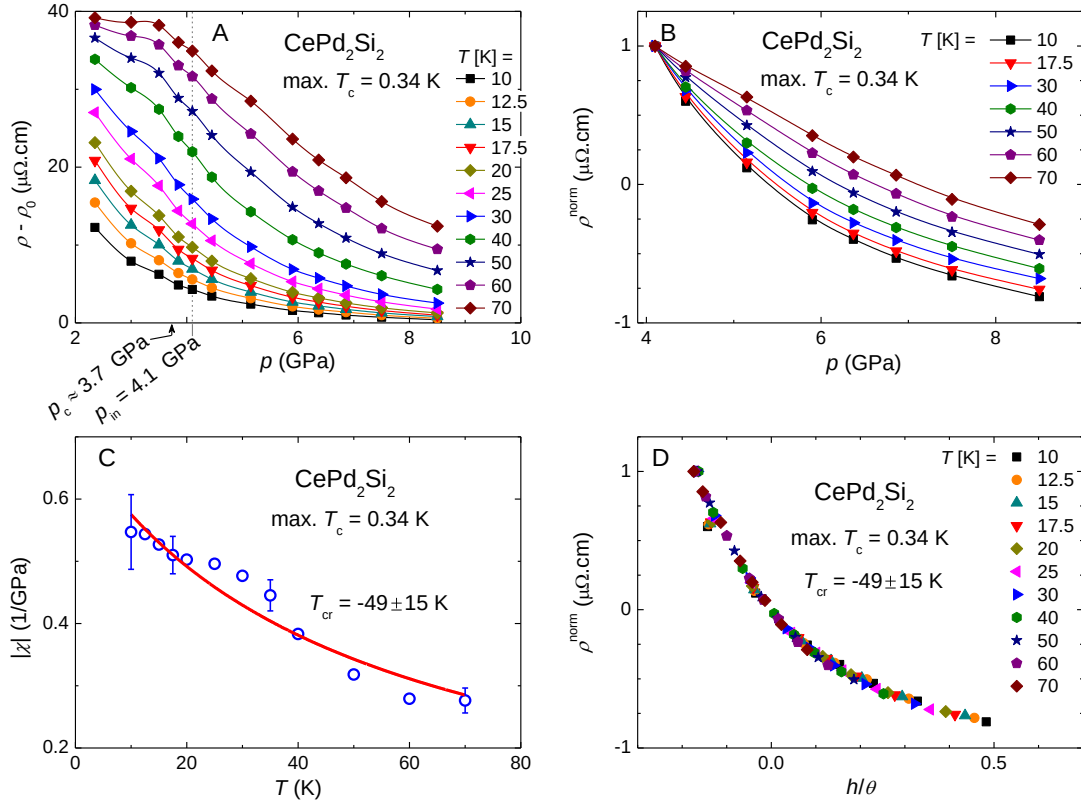


Fig. S8. Resistivity scaling analysis of a CePd_2Si_2 sample with $T_c = 0.34$ K. $\rho(T)$ -data first published in [56]. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 10 up to 70 K. (B) Normalized resistivity ρ^{norm} vs p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p_{\text{VCO}}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -49 \pm 15$ K. Error bars on χ are shown for representative data points. (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ .

Supplementary Figure S9

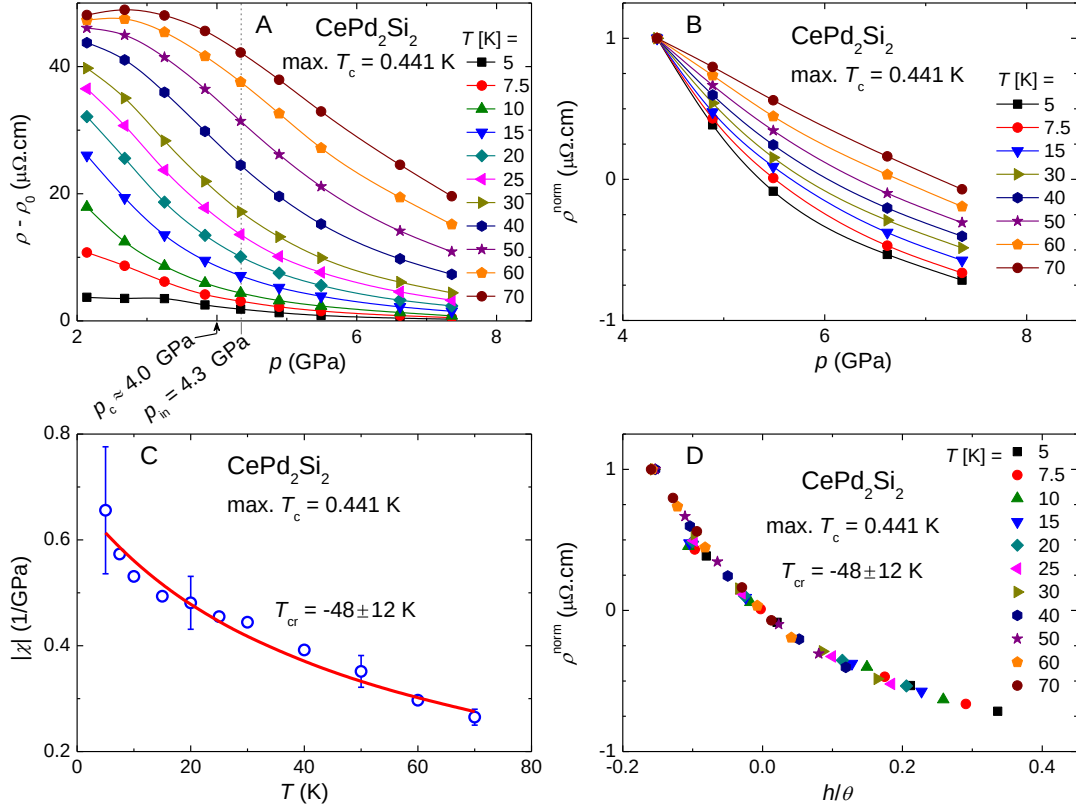


Fig. S9. Resistivity scaling analysis of a CePd₂Si₂ sample with $T_c = 0.441$ K. $\rho(T)$ -data first published in [19]. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 5 up to 70 K. (B) Normalized resistivity ρ^{norm} vs. p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p_{\text{VCO}}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -48 \pm 12$ K. Error bars on χ are shown for representative data points. (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ .

Supplementary Figure S10

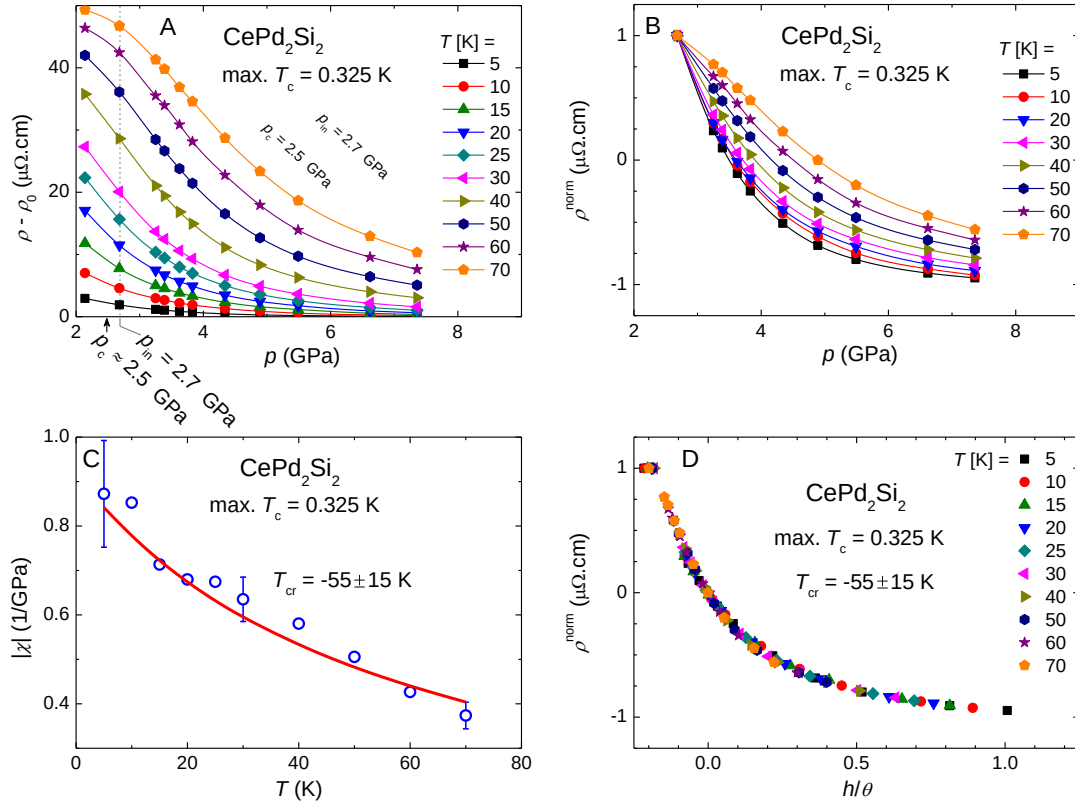


Fig. S10. Resistivity scaling analysis of a CePd_2Si_2 sample with $T_c = 0.325$ K. $\rho(T)$ -data first published in [19]. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 5 up to 70 K. (B) Normalized resistivity ρ^{norm} vs. p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p_{\text{VCO}}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -55 \pm 15$ K. Error bars on χ are shown for representative data points. (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ .

Supplementary Figure S11

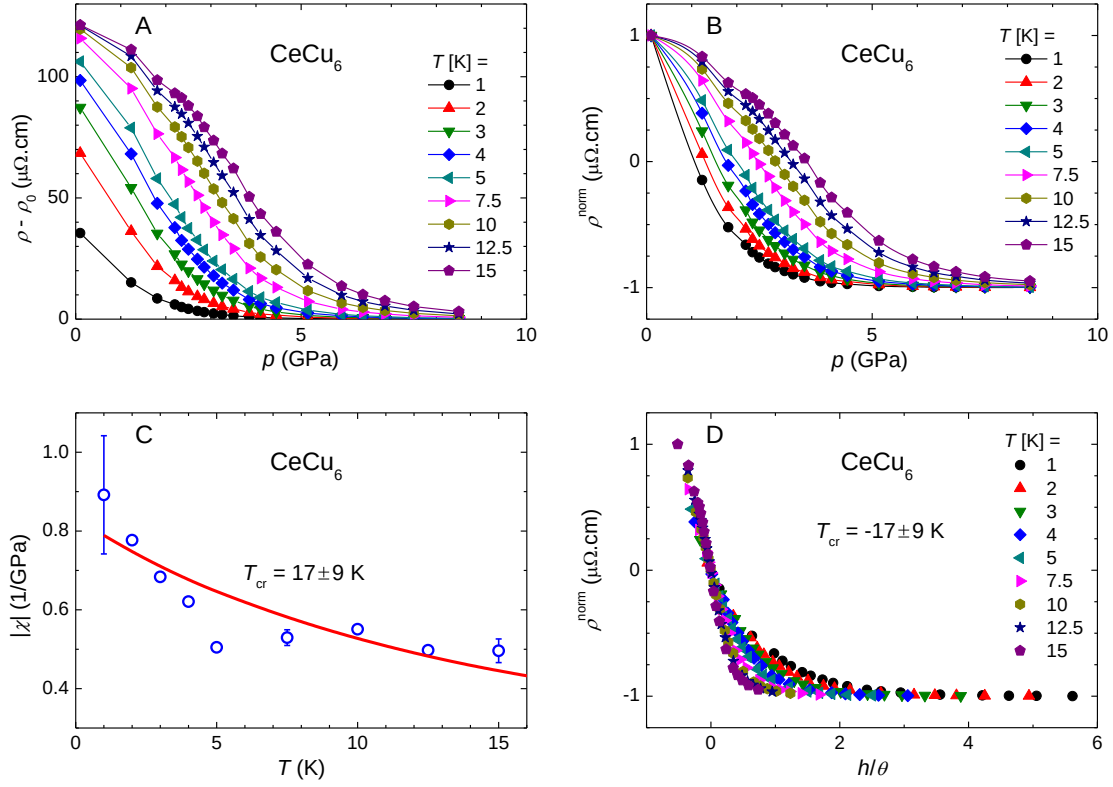


Fig. S11. Resistivity scaling analysis of a CeCu₆ sample. $\rho(T)$ -data obtained in steatite p -medium first published in [34]. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 1 to 15 K. Looking on the “raw” resistivity data ρ^* , we suspect that measurement errors for pressures up to 2 GPa are most likely due to the combined effect of error on p and bad thermalization of the sample (fast cooldown). (B) Normalized resistivity ρ^{norm} vs p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p_{\text{VCO}}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -17 \pm 9$ K. Error bars on χ are shown for representative data points. The bad overlap of fit and data is presumably due to the measurement errors noted before. Also, a low pressure-run density at the beginning of the pressurization results in a high error on χ . (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ . The curves do not collapse on a single scaling function. For now, it is not clear if this is intrinsic to CeCu₆ or due to the measurement errors only. A change of regime ascribed to the crystal field effect [34] may interfere.

Supplementary Figure S12

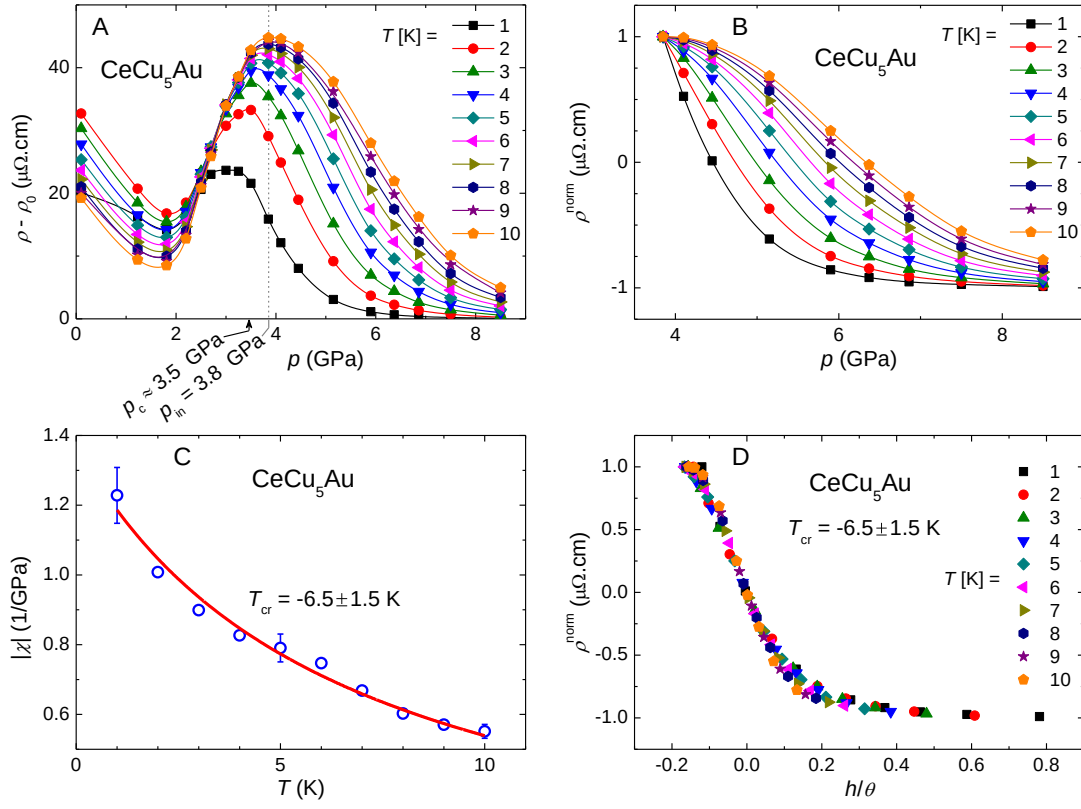


Fig. S12. Resistivity scaling analysis of a CeCu₅Au sample. $\rho(T)$ -data first published in [75]. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 1 up to 10 K. Note that the scaling analysis is limited to $T \leq 10$ K because of a low $T_1^{\text{max}} \sim 25$ K at the VCO. (B) Normalized resistivity ρ^{norm} vs. p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p\text{VCO}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -6.5 \pm 1.5$ K. Error bars on χ are shown for representative data points. (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ .

Supplementary Figure S13

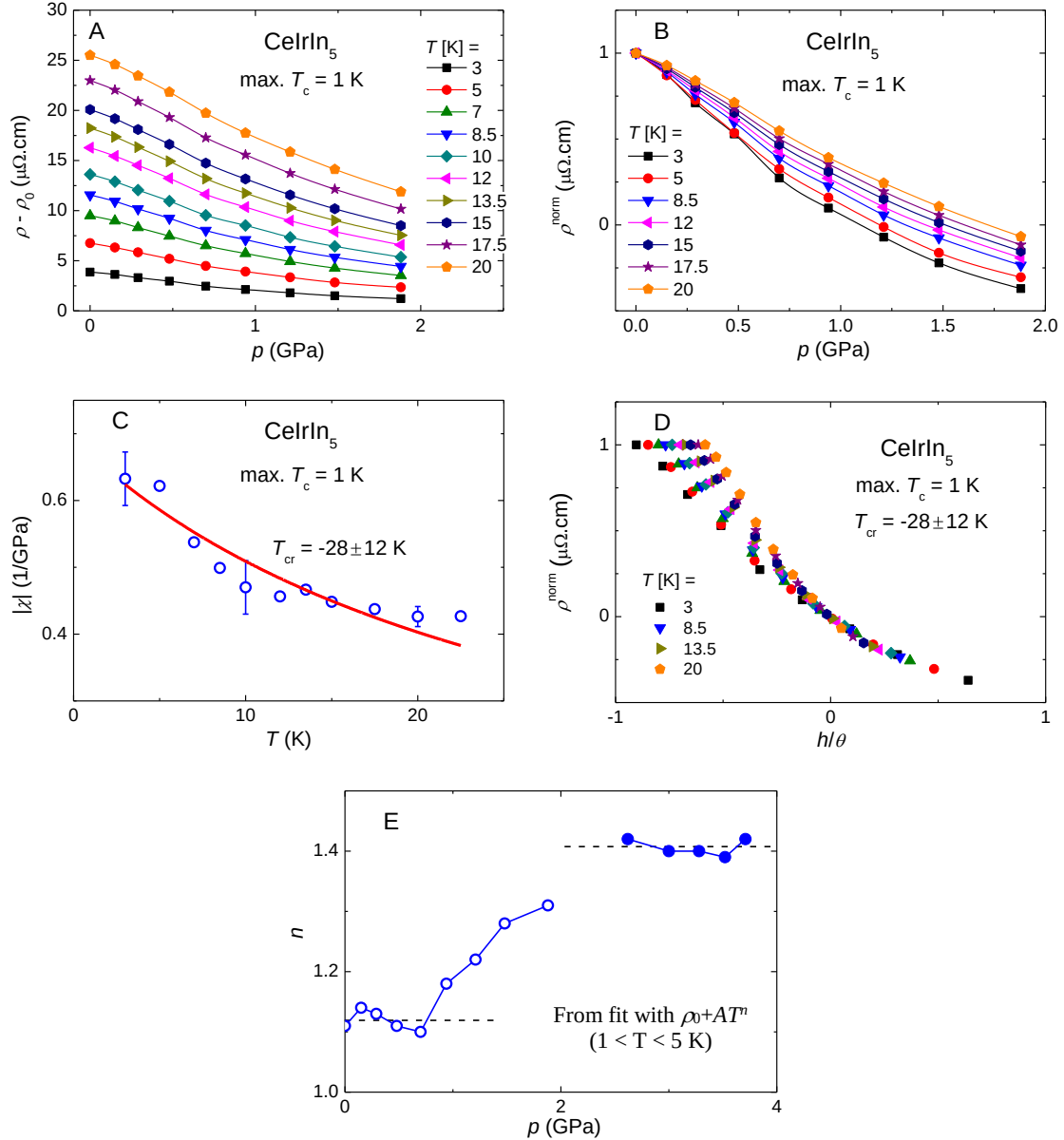


Fig. S13. Resistivity scaling analysis of a CeIrIn₅ sample. (A) Resistivity isotherms $\rho^* = \rho - \rho_0$ vs. pressure p at temperatures T from 3 up to 20 K. (B) Normalized resistivity ρ^{norm} vs. p . (C) Slope $\chi = (d\rho^{\text{norm}}/dp)_{p_{\text{VCO}}}$ vs. T . The red line represents a fit to the data with $\chi \sim 1/(T - T_{\text{cr}})$. The fit gives $T_{\text{cr}} = -28 \pm 12$ K. Error bars on χ are shown for representative data points. (D) Normalized resistivity ρ^{norm} vs. the generalized distance h/θ . The ρ^* isotherms collapse on a single curve only for $h/\theta > 0$. (E) Exponent n versus pressure p , obtained by fitting low-temperature resistivity $\rho(T)$ with $\rho_0 + AT^n$. Open symbols: data corresponding to Fig. S13A, full symbols: preliminary data from a new high-pressure experiment. The behavior of $n(p)$ hints to a pressure-induced change of regime, which may explain why the isotherms do not collapse for $h/\theta < 0$.

Supplementary Figure S14

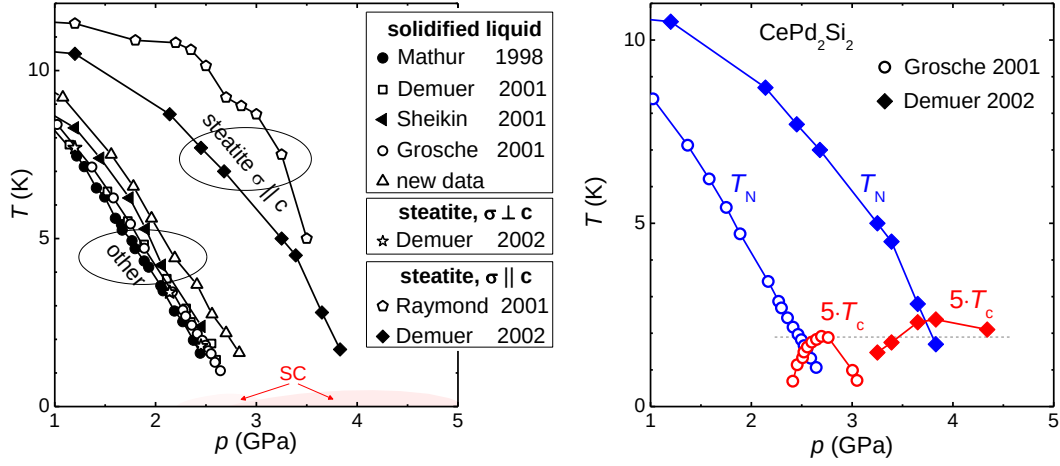


Fig. S14. Pressure-temperature phase diagram of CePd_2Si_2 samples measured in “solidified-liquid” [3,43,54,55] and “soft-solid-steatite” [19,56] pressure-transmitting medium (“solidified-liquid” = He, pentane, Daphne oil). Left-panel: all known data of the T_N transition line. A linear decrease of T_N occurs in “solidified-liquid” or steatite with stress $\sigma \perp c$. A more rapid vanishing of T_N occurs in steatite with stress $\sigma \parallel c$. The corresponding superconducting phases are represented schematically. Right panel: Phase diagram of two samples of similar quality as indicated by their residual resistivity $\rho_0 \approx 1.1 \mu\Omega\text{cm}$. A steeper collapse of the antiferromagnetic transition temperature T_N seems to favor higher superconducting T_c . Noteworthy, Demuer *et al.* [19] have observed a strain-driven enhancement of T_c . Two samples have been measured simultaneously in steatite with $\sigma \perp c$ and $\sigma \parallel c$, respectively. Despite lower sample quality, a higher T_c was found for $\sigma \parallel c$.

References

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