Supplementary Materials for

Evidence for a pressure-induced antiferromagnetic quantum critical point in intermediate-valence UTe$_2$

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Section S1: Landau theory for the relationship between the specific heat jumps of the two superconducting transitions

The point group of UTe2 is D$_{2h}$, which only admits one-dimensional irreducible representations (irreps). The spin-triplet nature of superconductivity in UTe2 implies that the order parameters should be parity-odd, and hence transform as ungerade irreducible representations. This fact, combined with the field-trained Kerr measurements of Hayes et al. (7), leads to the conclusion that either $\psi_1$ transforms as B$_{3u}$ and $\psi_2$ transforms as B$_{2u}$ or $\psi_1$ transforms as B$_{1u}$ and $\psi_2$ as A$_u$. In either case, their product transforms as B$_{1g}$. These considerations, combined with the superconducting U(1) symmetry, implies that several terms coupling $\psi_1$ and $\psi_2$ are allowed in the free energy. Explicitly, $\psi\psi_2$ and $\psi\psi_2^*$ (and their complex conjugates) transform as B$_{1g}$. The squares of these quantities will therefore appear in the free energy, where we note that invariance under U(1) enforces $\psi_1^2\psi_2^2$ and $\psi_1^*\psi_2^2$ to have the same coefficient. This leads to the free energy presented in the main text. In fact, the same form of the free energy holds for any $\psi_1$ and $\psi_2$ that transform as different one-dimensional irreps of D$_{2h}$.

Thus, hereafter we consider two superconducting order parameters that can be fine-tuned (e.g., via pressure) to have the same transition temperature T$_c$. Parameterizing $\psi_1$ and $\psi_2$ as a magnitude and a phase, the Landau free-energy expansion is then:

$$\mathcal{F} = \alpha(T - T_c)\psi_1^2 + \alpha\kappa(T - T_c)\psi_2^2 + \beta_1\psi_1^4 + \beta_2\psi_2^4 + 2g_1\psi_1^2\psi_2^2\cos 2\phi + g_2\psi_1^2\psi_2^2 + \epsilon(\psi_2^2 - \psi_1^2) \tag{1}$$

where $\psi_1$ and $\psi_2$ are the magnitudes of the two superconducting complex order parameters and $\phi$ is the relative phase between them. Here, $\alpha$, $\kappa$ are positive Landau coefficients. When the parameter $\epsilon$ is fine-tuned to zero, the two transitions take place at the same temperature T$_c$. In the phase diagram of UTe2, this happens at $P^* \approx 0.2$ GPa. Therefore, we can expand $\epsilon \propto P - P^*$, such that $\epsilon < 0$ ($P < P^*$) favors the state $\psi_2$ and $\epsilon > 0$ ($P > P^*$) favors $\psi_1$. Note that this formalism does not specify the particular irreducible representations associated with $\psi_i$.

The Kerr measurements performed in Ref. (7) impose restrictions on the Landau parameters, since they identified, below T$_c$, a state in which both $\psi_1$ and $\psi_2$ are simultaneously non-zero and time-reversal symmetry is broken, indicative of $\phi = \pm \pi / 2$. This result implies $g_1 > 0$. We stress that the choice of $\phi$ (and hence of $g_1$) is an assumption based on the available experimental data. If $g_1 < 0$ a different phase would be preferred. Minimization of the free energy with respect to $\phi$ for $g_1 > 0$ leads to:

$$\mathcal{F} = \alpha(T - T_c)\psi_1^2 + \alpha(T - T_c)\psi_2^2 + \beta_1\psi_1^4 + \beta_2\psi_2^4 - 2g_1\psi_1^2\psi_2^2 + \epsilon(\psi_2^2 - \psi_1^2) \tag{2}$$
with \( g \equiv g_1 - g_2 / 2 \). Moreover, for the two states to coexist microscopically, it must follow that \( \beta_1 \beta_2 - g^2 > 0 \). Finally, we impose \( \beta_1, \beta_2 > 0 \) to ensure the stability of the Landau functional.

Within a mean-field approach, for \( \epsilon = 0 \), there is only one transition at \( T_c \) to a state where both \( \psi_1 \) and \( \psi_2 \) coexist microscopically, with a relative phase of \( \pm \pi / 2 \). The specific heat jump across this transition is:

\[
\frac{\Delta C}{T_c} = \frac{\alpha^2 \beta_1 + \kappa(2g + \beta_1 \kappa)}{2} \beta_1 \beta_2 - g^2
\]

For \( \epsilon > 0 \), \( \psi_1 \) condenses first at \( T_{c,1} = T_c + \epsilon / \alpha \), followed by a secondary condensation of \( \psi_2 \) at:

\[
T_{c,2} = T_{c,1} - \epsilon \frac{\beta_1 (1 + \kappa)}{\alpha(g + \beta_1 \kappa)}
\]

Note that time-reversal symmetry is only broken below \( T_{c,2} \). Calculating the specific heat, we obtain two jumps at the two transitions:

\[
\frac{\Delta C_1}{T_{c,1}} = \frac{\alpha^2}{2 \beta_1}
\]

\[
\frac{\Delta C_2}{T_{c,2}} = \frac{\alpha^2 (g + \beta_1 \kappa)^2}{2 \beta_1 (\beta_1 \beta_2 - g^2)}
\]

For \( \epsilon < 0 \), \( \psi_2 \) condenses first at \( T_{c,2} = T_c + |\epsilon| / (\alpha \kappa) \) followed by the condensation of \( \psi_1 \) at \( T_{c,1} \) given by:

\[
T_{c,1} = T_{c,2} - |\epsilon| \frac{\beta_2 (1 + \kappa)}{\alpha \kappa (\beta_2 + g \kappa)}
\]

The specific heat jumps at these two transitions are given by:

\[
\frac{\Delta C_2}{T_{c,2}} = \frac{\alpha^2 \kappa^2}{2 \beta_2}
\]

\[
\frac{\Delta C_1}{T_{c,1}} = \frac{\alpha^2 (\beta_2 + g \kappa)^2}{2 \beta_2 (\beta_1 \beta_2 - g^2)}
\]

Note that \( \Delta C_1 \) depends on \( \epsilon \), since \( T_{c,1} \) depends on \( \epsilon \). However, \( \Delta C_1 / T_{c,1} \) is independent of \( \epsilon \).

As a result, we find the following relationship valid for any \( \epsilon \):

\[
\frac{\Delta C}{T_c} = \frac{\Delta C_1}{T_{c,1}} + \frac{\Delta C_2}{T_{c,2}}
\]
Therefore, the jump divided by T_c at the simultaneous transition is equal to the sum of the jumps (each divided by their respective transition temperature) when the transitions are split. In contrast, the sum of the two jumps is not the same as the jump of the simultaneous transition:

$$\Delta C_1 + \Delta C_2 = \Delta C + \epsilon \frac{\alpha(g - g_\kappa + \beta_1\kappa - \beta_2)}{2(\beta_1\beta_2 - g^2)}$$

Interestingly, if \( \psi_1 \) and \( \psi_2 \) belonged to the same two-dimensional irrep, the last term would vanish since \( \kappa = 1 \) and \( \beta_1 = \beta_2 \). Although two-dimensional irreps are not supported by the D_{2h} point group of UTe_2, this general result does provide an interesting criterion to distinguish whether coincident superconducting transitions arise from a single two-dimensional irrep or two one-dimensional irreps, which is an ongoing discussion for Sr_2RuO_4.

Although our ac calorimetry measurements do not provide quantitative values for the specific heat jumps, the results shown in Fig. 1(D) of the main text suggest that \( \Delta C_1 / T_{c,1} \) and \( \Delta C_2 / T_{c,2} \) (and thus their sum) depend only weakly on pressure for \( P < 0.7 \) GPa. This is consistent with the main assumption of our model, namely, that the main effect of pressure near the degeneracy point \( P^* \approx 0.2 \) GPa is to shift the transition temperatures of the two superconducting states, without significantly affecting the quartic coefficients of the Landau free-energy expansion in Eq. (1). We note that time-reversal symmetry breaking has only been observed at \( P = 0 \). However, the fact that mainly the quadratic coefficients change for pressures \( P < 0.7 \) GPa suggests that time-reversal symmetry breaking takes place over this pressure range. However, we cannot rule out that time-reversal symmetry is restored at higher pressures \( P \geq 1 \) GPa, where, as discussed below, it appears that the quartic coefficients are also impacted by the increasing pressure and \( T_{c,2} \) becomes very close to zero.

The variation of \( \Delta C_1 / T_{c,1} \) with pressure, already incipient at \( P = 0.6 \) GPa and more pronounced for \( P \geq 1 \) GPa, is an indication that the impact of pressure on the quartic coefficients of Eq. (1) becomes more significant in this pressure range. Indeed, from Eq. (5), it is clear that a coefficient \( \beta_1 \) that increases with \( \epsilon \) can cause a suppression of \( \Delta C_1 / T_{c,1} \) but an enhancement of \( \Delta C_2 / T_{c,2} \) depending on the values of the other Landau coefficients (the same argument would hold for Eq. (8) with \( \beta_1 \) replaced by \( \beta_2 \)). Of course, there are several allowed couplings that are quartic in \( \psi \), but linear in \( \epsilon \), e.g. \( \epsilon \psi_1^2\psi_2^2 \), \( \epsilon(\psi_1^4 + \psi_2^4) \), and \( \epsilon(\psi_1^4 - \psi_2^4) \). While determining which of these terms is the most relevant is not possible with the current data, the dependence of \( \Delta C_1 / T_{c,1} \) with pressure indicates that the type of coexistence between the two superconducting states may be different at high pressures as compared to ambient pressure, as discussed above.
Figure S1. Magnetic susceptibility times temperature ($\chi T$) versus temperature at ambient pressure. Adapted from Ref. (2). The downward curvature at low temperatures is characteristic of dominant antiferromagnetic correlations even at ambient pressure.
Figure S2. Coherence temperature. (A) Resistivity versus temperature as a function of pressure up to 250 K. As pressure is increased a clear peak is observed in resistivity versus temperature that indicates the temperature for Kondo coherence. (B) The Kondo coherence temperature versus pressure. As pressure is increased, the Kondo coherence is suppressed to lower temperature.
Figure S3. Uranium L3 absorption edge fits. The uranium L3 XANES was modelled using an arctan step function combined with a gaussian peak. The orange line is the combined fit function and overlaps with the data (blue). The red line represents the component of the fit from the arctangent step function, and the green line is the component from the Gaussian peak. Initially, US$_2$ was believed to be a reasonable U$^{4+}$ reference. Our broadened XANES spectrum, however, suggests that US$_2$ might be hybridized (31) and does not serve as a good U$^{4+}$ reference in comparison to UF$_4$. 
Figure S4. Ac calorimetry at 1.66 GPa. Ac calorimetry at 1.66 GPa showing $T_{m2}$ at 3 K and $T_{m1}$ at 7.3 K.