Multiple metamagnetic quantum criticality in Sr$_2$Ru$_2$O$_7$

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Bilayer strontium ruthenate Sr$_2$Ru$_2$O$_7$ displays pronounced non-Fermi liquid behavior at magnetic fields around 8 T, applied perpendicular to the ruthenate planes, which previously has been associated with an itinerant metamagnetic quantum critical end point (QCEP). We focus on the magnetic Grüneisen parameter $\Gamma_H$, which is the most direct probe to characterize field-induced quantum criticality. We confirm quantum critical scaling due to a putative two-dimensional QCEP near 7.845(5) T, which is masked by two ordered phases A and B, identified previously by neutron scattering. In addition we find evidence for a QCEP at 7.53(2) T and determine the quantum critical regimes of both instabilities and the effect of their superposition.

Quantum criticality denotes critical behavior that is associated with continuous transformations of matter at zero temperature. Due to the absence of thermal fluctuations at $T = 0$ it is qualitatively different from classical criticality. In metals the unconventional excitation spectrum near a quantum critical point (QCP) causes the breakdown of Fermi liquid (FL) behavior and its intimate relation to exotic states, such as unconventional superconductivity, adds even more importance to this topic. To date, the influence of quantum critical magnetic excitations on electrons in a metal is far from being understood. For instance the applicability of the itinerant Hertz-Millis-Moriya theory on $f$-electron based Kondo lattice systems has been disproved by several experiments and alternative descriptions are not fully established yet. Quantum criticality related to itinerant metamagnetism is exceptional in the sense, that electronic degrees of freedom are irrelevant, and a quantitative application to experimental results should be possible.

The generic metamagnetic quantum critical end point (QCEP) arises from the suppression to $T = 0$ of the end point of a line of first-order metamagnetic transitions in temperature-field phase space by tuning e.g. composition, pressure or the magnetic field orientation. Metamagnetic QCEPs have been realized in the $f$-electron based compounds CeRu$_2$Si$_2$, UCoAl and Sr$_3$Ru$_2$O$_7$, as well as in $d$-electron Sr$_2$Ru$_2$O$_7$.

We focus on bilayer strontium ruthenate Sr$_2$Ru$_2$O$_7$. Magnetization of this compound along the tetragonal $c$-axis at low temperature exhibits three successive super-linear, i.e. metamagnetic, rises at $\mu_0H_{M1}=7.5$ T, $\mu_0H_{M2}=7.85$ T and $\mu_0H_{M3}=8.1$ T. The first one is a metamagnetic cross-over (M1). The second and third ones are first order metamagnetic transitions (M2 and M3), ending at critical temperatures of about 1 and 0.5 K, respectively. A line of second-order thermal phase transitions, connecting the critical end points of M2 and M3, has been discovered in electrical resistivity and thermodynamic experiments, which recently by neutron scattering has been identified as phase boundary of a spin-density-wave (SDW) "phase A" (see Fig. 1.). The lower and upper critical fields of SDW-A correspond respectively to $H_{M2}$ and $H_{M3}$. Additionally, another SDW "phase B" has been observed in between $H_{M3}$ and 8.3 T. The observed incommensurate ordering vectors in both SDW phases have been related to Fermi surface nesting. Magnetic susceptibility and magnetostriction vectors in both SDW phases were previously associated to a critical field close to $H_{M2}$. Outside the SDW phases A and B and not to close to the M1 crossover, thermal expansion obeys quantum critical scaling in accordance with the expectations for a two-dimensional (2D) metamagnetic QCEP near 7.845 T.

We solve this discrepancy by proving that Sr$_2$Ru$_2$O$_7$ displays two QCEPs at $\mu_0H_{c1} = 7.53(2)$ T and $\mu_0H_{c2} = 7.845(5)$ T, respectively. We determine regimes in phase space where either of the two QCEPs leads to scaling of the magnetic Grüneisen parameter. We also show where scaling fails due to the superposition of criticality from both instabilities. Multiple quantum criticality as origin for behavior that is incompatible with the generic predictions of QCPs can be of relevance for various material classes.

The magnetic Grüneisen parameter, $\Gamma_H = T^{-1}(dT/dH)_S$ measures the relative temperature change with magnetic field under adiabatic conditions, called adiabatic magnetocaloric effect. Due to the entropy accumulation near field-driven quantum criticality, generically this property is expected to obey (i) a sign change when tuning the field across the critical value, (ii) a divergence upon cooling (at constant field) within the quantum critical regime and (iii) universal scaling
Anomalies in isothermal specific heat coeﬃcient display more broadened symmetric peaks for the heat capacity are expected. Our measurements, however, display more broadened $C/T$ peaks on the high-field compared to the low-field sides. As discussed later, this may be related to a slight increase of the effective dimensionality of the critical ﬂuctuations at large fields.

The magnetic ﬁeld dependence of the 0.2 K data is in perfect agreement with previous data [8, 15], see SM [16]. As shown by the blue solid line in Fig. 2, the data are well described by $C/T \propto (H_c - H)^{-1/3}$, predicted for a 2D QCEP [3, 24] with critical ﬁeld close to $H_{M1}$ but signiﬁcantly smaller than $H_{M2}$. This indicates that the previously anticipated scenario with a single ﬁeld-tuned QCEP near $H_{M3}$ [8] is insuﬃcient.

The existence of two separate 2D metamagnetic QCEPs is evident from the analysis of the magnetic Grüneisen parameter $\Gamma_H$ given below. In contrast to the speciﬁc heat coeﬃcient, which has a substantial non-critical background, $\Gamma_H$ is more sensitive to quantum criticality because of a negligibly small non-critical contribution.

Figure 3 shows an isothermal scan of the magnetic Grüneisen parameter at 0.2 K. $\Gamma_H(H)$ increases by more than a factor 10 in between 6 to 7.5 T. For any ﬁeld-tuned QCP, the magnetic Grüneisen parameter versus ﬁeld must follow a linear dependence and crosses zero at the critical ﬁeld. As shown in the inset of Fig. 3, this universal dependence is indeed observed, yielding a critical ﬁeld very close to $H_{M1}$, which conﬁrms our heat capacity analysis.

At ﬁelds beyond $H_{M1}$ a cascade of further sign changes and anomalies is found in $\Gamma_H(H)$. They are associated with metamagnetic transitions $M2$ and $M3$ and respective the SDW phases A and B [12, 14], as well as (see the green arrows) an anomaly labeled "C" in the phase diagram of Fig. 1.
subsequently displays a negative divergence as $T \to 0$, related to the nearby QCEP1 (cf. Fig. 1).

We now turn to a quantitative comparison of our data with the theory of metamagnetic quantum criticality \cite{3, 24}. The latter predicts $\Gamma_H h \sim h^2/(T(4+2d)/3$ in the quantum critical and $\Gamma_H h = (3-d)/3$ in FL regime, where $d$ denotes the dimensionality and $h = \mu_0 (H - H_\Gamma)$. This leads to universal scaling in a plot of $\Gamma_H h$ vs $h^2/T^\epsilon$, where $\epsilon = (4+2d)/3 = 8/3$ for $d = 2$. Respective scaling behavior of our data is shown in Figure 5. Here we fixed the critical field to 7.845 T \cite{11}, which is the position of QCEP2. The data collapse over several orders of magnitude, similar as previously found for thermal expansion \cite{11}, proves quantum critical behavior and indicates the applicability of the itinerant theory. However, a close inspection provides further information \cite{16}. First, for fields below $H_{M2}$, scaling is cut-off near the crossover to the FL regime. This could be associated to the influence of QCEP1, as discussed above. Second, for fields $H > H_{M2}$ the data within the FL regime approach a saturation of $\Gamma_H h \approx 0.2$, which is smaller than the value 1/3 predicted for a QCEP with dimensionality $d = 2$ \cite{24} and may indicate that the effective dimensionality slightly increases at large fields. The value of 0.2 would correspond to $d_{eff} = 2.4$. Metamagnetism in Sr$_3$Ru$_2$O$_7$ is supposed to arise from van Hove singularities near the Fermi level \cite{26}. A change of the de Haas-van Alphen frequencies near 8 T has been ascribed to magnetic breakdown \cite{27}. This could explain the increase of the effective dimensionality of critical fluctuations, deduced from our scaling analysis.

The different regimes where the magnetic Grüneisen parameter displays scaling with respect to QCEP 1 and QCEP2 are indicated in Fig. 1. In between both regimes neither scaling works, because criticality from both instabilities is adding up (see SM \cite{16}). Next, we discuss the influence of the ordered phases A and B. In the approach of these phase transitions, $\Gamma_H$ data deviate from the expected quantum critical regime.
Our measurements of the magnetic Grüneisen parameter and specific heat coefficient establish the existence of two itinerant metamagnetic QCEPs in bilayer strontium ruthenate Sr$_3$Ru$_2$O$_7$ for magnetic fields applied parallel to the c-direction. QCEP1 appears at a metamagnetic crossover near 7.5 T while QCEP2, which has already previously been established, is located at 7.845 T. The phase diagram shown in Fig. 1 indicates the scaling regimes "QC1" and "QC2" determined from the magnetic Grüneisen parameter behavior (see also SM [16]). While "QC2" is largely extended at elevated temperatures, "QC1" is confined to a narrow regimes close to QCEP1. In between these scaling regimes, there exist a range in phase space, in which scaling fails due to the superposition of criticality from both instabilities. The phase diagram is even richer and contains also two SDW phases A and B [12] and some anomalous yet unidentified regime labeled "C". Likely, the observed complexity is related to the complicated electronic structure of this material [26]. The Fermi surface contains several pockets that could give rise to nesting and sheets near a van Hove singularity. From a general perspective, multiple quantum criticality may be of origin of anomalous behaviors in different material classes, including heavy-fermions and high-$T_c$ superconductors. The Grüneisen parameter is ideally suited to disentangle multiple quantum criticality.

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![Fig. 5](image-url) (Color online) Metamagnetic quantum critical scaling of the magnetic Grüneisen parameter in Sr$_3$Ru$_2$O$_7$. The y-axis displays $\Gamma_\mu H(T)$ while the x-axis shows $T^{8/3}/h$ with $h = \mu_0(H - H_c)$ and the critical field $H_c = 7.845$ T [11]. Panels (a) and (b) displays regimes below and above the critical field. Red and blue dotted lines indicate predicted asymptotic quantum critical and FL behavior for $d = 2$ metamagnetic QCEP [24]. In panel (a) data on the left of the black arrow have been excluded (for failed scaling see SM [16]).
In the main text, we are discussing the field dependence of our heat capacity data below 7.5 T. It shows a power-law divergence towards $H_{c1}$ with exponent 1/3 in accordance with the itinerant theory for a two-dimensional QCEP. However, previous data of the field dependence of the entropy increment, obtained by non-adiabatic magnetocaloric effect measurements in A.W. Rost et al., Science 325, 1360 (2009), as well as continuous field sweep a.c. heat capacity data at 0.25 K, see A.W. Rost et al., Phys. Stat. Sol. B 247, 513 (2010), had been described differently (see main text). Rost et al. have used the function $(H_{c2} - H)^{-1}$ (note the different exponent and different critical field) that would be highly incompatible with the prediction from the itinerant theory. It is therefore interesting to directly compare their data with ours. As shown in Figure 6 both data sets for $C/T$ (note that in the Fermi liquid regime $C/T$ is temperature independent) differ by less than 4 mJ/Ru-mol·K$^2$ equivalent to 2%. This nicely indicates the reproducibility of the thermodynamic results on Sr$_3$Ru$_2$O$_7$. Based on our magnetic Gruneisen analysis (cf. inset of Fig. 3 main text), the description of $C/T$ by $(H_{c1} - H)^{-1/3}$ in contrast to $(H_{c2} - H)^{-1}$ is appropriate.

**FIELD DEPENDENCE OF THE MAGNETIC GRÜNEISEN RATIO**

![Graph](image)

**FIG. 7.** Magnetic field dependence of magnetic Gruneisen parameter $\Gamma_H$ of Sr$_3$Ru$_2$O$_7$ at $T = 1$ K.

As shown in Fig. 3 in the main text, at 0.2 K the magnetic Gruneisen ratio displays very complicated behavior with several extrema and sign changes. Figure 7 displays respective results on Sr$_3$Ru$_2$O$_7$. Upon increasing magnetic field from 6 T, the Gruneisen parameter is negative and increases in absolute value. Near $H_{c1}$ it passes a local minimum and maximum and continues to diverge until a sharp minimum and sign change is reached very close to $H_{c2}$, beyond which $\Gamma_H$ is positive and decreases with increasing $H$. Since $\Gamma_H = -(dS/dH)/C$ the zero crossing of the magnetic Gruneisen ratio, which occurs

**SUPPLEMENTAL MATERIAL**

**FIELD DEPENDENCE OF THE SPECIFIC HEAT**

![Graph](image)

**FIG. 6.** Magnetic field dependence of specific heat divided by temperature of Sr$_3$Ru$_2$O$_7$. Open black circles display the data (see main text) measured at 0.2 K, while the solid blue line has been extracted from A.W. Rost et al., Phys. Stat Sol. B 247, 513 (2010). The latter data were obtained using an a.c. heat capacity technique during continuous magnetic field sweep at 0.25 K.
along a line above the QCEP2 (shown in Fig. 1 of the main text), indicates an accumulation of entropy. This is a generic signature of quantum criticality (cf. M. Garst and A. Roch, Phys. Rev. B 72, 205129 (2005)). At lower temperatures, the interplay of QCEP1 and QCEP2 results in a more complicated behavior of $\Gamma_H(H)$ displayed in Fig. 3 of the main text.

Next we focus on isothermal field data of $\Gamma_H(H)$ for large fields between 8.5 and 10 T shown in Figure 8. The colored arrows indicate anomalous behavior, leading to inflection points in the field dependence. The respective fields are temperature dependent, cf. regime "C" in the phase diagram Fig. 1 of the main text. This excludes quantum oscillations as origin. We note, that no clear signature in heat capacity has been found in this regime of phase space.

SCALING ANALYSIS FOR QCEP1

The phase diagram displayed in Fig. 1, main text, illustrates the range in $T$-$H$ parameter space where quantum critical scaling in the magnetic Grüneisen parameter with respect to the first (QC1) and second (QC2) quantum critical end point holds, respectively. It also shows a regime "QC1+QC2" where scaling fails because of the superposition of criticality from both instabilities. Furthermore scaling fails near the spin-density-wave states A and B and the yet unidentified regime C. In this section we focus on the scaling and its failure of $\Gamma_H$ due to QCEP1, while in the subsequent section respective analysis for QCEP2 is detailed.

As illustrated in Figs. 2 and 3 (inset) of the main text, at 0.2 K, specific heat and $\Gamma_H$ follow the field dependences in accordance with the scaling predictions for QCEP1. Here we analyze up to which temperature this scaling holds. For this purpose, Figure 9 shows a plot of $1/\Gamma_H$ vs $\mu_0 H$ at several elevated temperatures. Since for field tuned instabilities $\Gamma_H \sim (H_c - H)^{-1}$ with universal (temperature independent) pre-factor, such plot is best suited to determine the upper bound in temperature of the scaling regime. It is found, that all curves up to 0.6 K follow a universal dependence indicated by the blue line, while the data at 1 K follow a clearly different slope (incompatible with the expected universality). In addition they would extrapolate to a very different critical field, but the critical field should be temperature independent. Thus, scaling with respect to QCEP1 breaks down in between 0.6 and 1 K.

Next, we show scaling plots, similar to those of Fig. 5 in the main text, but now for the critical field of QCEP1, $H_{c1} = 7.53(2)$ T. Figure 10 (a) shows the scaling analysis using $H_{c1}$ for fields below the critical field, while part (b) displays data for $H > H_{c1}$. The latter case is restricted to a very narrow field interval, because in the approach of $H_{c2} = 7.845(5)$ T the magnetic Grüneisen parameter changes sign due to the influence of QCEP2. Apparently, the data do not collapse at all for $H > H_{c1}$, which is attributed to the influence of QCEP2. In addition for $H < H_{c1}$ a data collapse is only found at low temperatures, consistent with the above observation of a breakdown of scaling at $T > 0.6$ K.

SCALING ANALYSIS FOR QCEP2

We now turn to the determination of the range in $T$-$H$ parameter space where scaling with respect to QCEP2 works (cf. regime "QC2" in Fig. 1 main text). For this purpose, we re-
FIG. 10. Scaling analysis of $\Gamma_H$ for Sr$_3$Ru$_2$O$_7$ with respect to the QCEP1, using $h = \mu_0(H - H_{c1})$ for fields below (a) and above (b) the critical field $\mu_0 H_{c1} = 7.53(2)$ T.

To summarize, the magnetic Grüneisen ratio indicates multiple quantum criticality with a QCEP1 near $\mu_0 H_{c1} = 7.53(2)$ T and a QCEP2 near $\mu_0 H_{c2} = 7.845(5)$ T. Using scaling analysis with respect to both $H_{c1}$ and $H_{c2}$, we determined the regimes “QC1” and “QC2” in the phase diagram of Fig. 1, main text, as well as the regime labeled “QC1+QC2” in which latter scaling fails because of the superposition of critical behavior from both instabilities.