Metamagnetic quantum criticality in $Sr_3Ru_2O_7$ studied by thermal expansion

P. Gegenwart[(1)], F. Weickert[(1)], M. Garst[(2)], R.S. Perry[(3,4,5)], Y. Maeno[(4,5)]
[(1)] *Max-Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany*
[(2)] *Theoretical Physics Institute, University of Minnesota, Minneapolis, Minnesota 55455, USA*
[(3)] *School of Physics and Astronomy, University of St. Andrews, Fife KY16 9SS, Scotland*
[(4)] *International Innovation Center, Kyoto University, Japan*
[(5)] *Department of Physics, Kyoto University, Japan*
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We report low-temperature thermal expansion measurements on the bilayer ruthenate $Sr_3Ru_2O_7$ as a function of magnetic field applied perpendicular to the Ruthenium-oxide planes. The field-dependence of the c-axis expansion coefficient indicates the accumulation of entropy close to 8 Tesla, related to an underlying quantum critical point. The latter is masked by two first-order metamagnetic transitions which bound a regime of enhanced entropy. Outside this region the singular thermal expansion behavior is compatible with the predictions of the itinerant theory for a two-dimensional metamagnetic quantum critical end point.

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Quantum phase transitions (QPTs) in itinerant electron systems are of extensive current interest in condensed matter physics because they are not only at the origin of unusual finite temperature properties but also promote the formation of new states of matter like unconventional superconductivity in heavy fermion systems [1]. Dilatometric studies are especially suitable to investigate quantum criticality since they directly probe the sensitivity of thermodynamics resulting from the QPT being susceptible to pressure-tuning. In particular, near any pressure-tuned quantum critical point (QCP) the thermal expansion, which at constant pressure quantifies the temperature dependence of the sample volume, is more singular than the specific heat [2, 3]. Below, we use thermal expansion to study a certain type of QCP associated with metamagnetism in metals. Generally, metamagnetism describes the sudden rise of the magnetization $M(H)$ as a function of the applied magnetic field H. A line of first-order metamagnetic transitions in the magnetic field–temperature plane has an end point (H^*, T^*) beyond which the transition becomes a continuous crossover. An especially interesting situation now arises when the temperature T^* can be tuned to zero giving rise to an isolated quantum critical end point (QCEP) [4].

The material that initiated the interest in QCEPs is the ruthenate $Sr_3Ru_2O_7$ [5], which crystallizes in the bilayered version of the perovskite structure. Its metamagnetic transition conforms to the picture of a field-induced Stoner transition given the absence of a dramatic change of the Fermi surface [6] and its highly enhanced Sommerfeld-Wilson ratio. $Sr_3Ru_2O_7$ shows an almost isotropic zero-field susceptibility at low temperatures, and metamagnetism occurs both between 5 and 6 T for fields in the Ruthenium-oxide planes as well as near 7.8 T for $H \parallel c$ [5]. A detailed study on single crystals with a residual resistivity of about 3 $\mu\Omega$cm has revealed that the temperature, T^*, of the critical end point of a line of first-order metamagnetic transitions can indeed be tuned by changing the angle of the applied magnetic field from 1.25 K for fields perpendicular to the c-axis towards zero for $H \parallel c$ [7]. For this field orientation, non-Fermi liquid behavior occurs related to a QCEP close to 8 T [5]. However, the electrical resistivity at fields very close to the metamagnetic region has revealed additional features: In a new generation of high-quality $Sr_3Ru_2O_7$ single crystals with a residual resistivity as low as 0.4 $\mu\Omega$cm [8] three peaks in the susceptibility and magnetostriction have been observed below 1 K. The first one at 7.5 T marks a cross-over, whereas the two latter ones at 7.85 and 8.07 T are first-order metamagnetic transitions. They confine a low-temperature ($T \leq 1$ K) regime in which the residual electrical resistivity is strongly enhanced indicative of strong elastic scattering and which disappears by rotating the field for more than 10 degrees away from $H \parallel c$ [9]. It has been proposed [9] that this behavior might result from the formation of a symmetry-broken phase characterized by a spin-dependent Pomeranchuk deformation of the Fermi surface. Alternatively, a scenario of phase separation leading to charge inhomogeneities within the bounded regime has been discussed [10].

In this Letter, we report a c-axis thermal expansion study on $Sr_3Ru_2O_7$ at temperatures down to 100 mK and mT field steps close to the metamagnetic regime for $H \parallel c$. We observe quantum critical behavior compatible with the predictions for a two-dimensional (2D) metamagnetic QCEP [4]. Distinct anomalies in thermal expansion and electrical resistivity at the bounded regime are discussed.

The thermal expansion measurements were performed on a high-quality single crystal with 1.7 mm length along the c-axis, grown by floating zone technique [8]. Length changes along the c-axis have been detected by utilizing a high-resolution capacitive dilatometer in a dilution refrigerator. Below, we use nominal values for the field applied with aid of a 20 T superconducting magnet. For comparison with corresponding susceptibility measure-

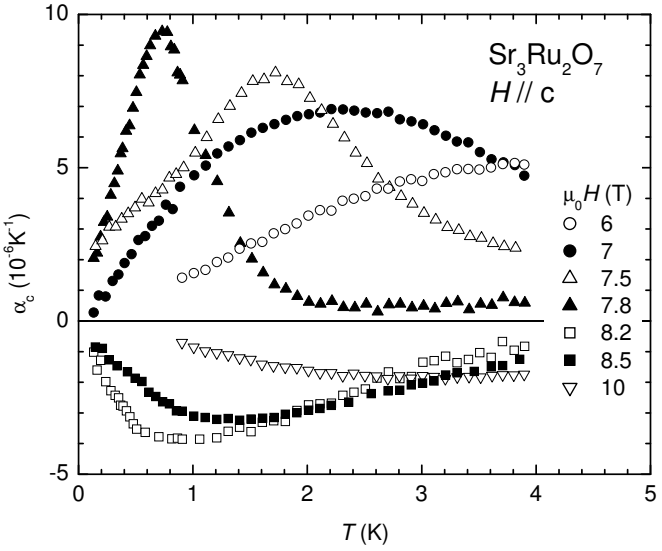

FIG. 1: Linear c-axis thermal expansion coefficient of $Sr_3Ru_2O_7$ as α_c vs T at various magnetic fields $H \parallel c$ indicated by different symbols.

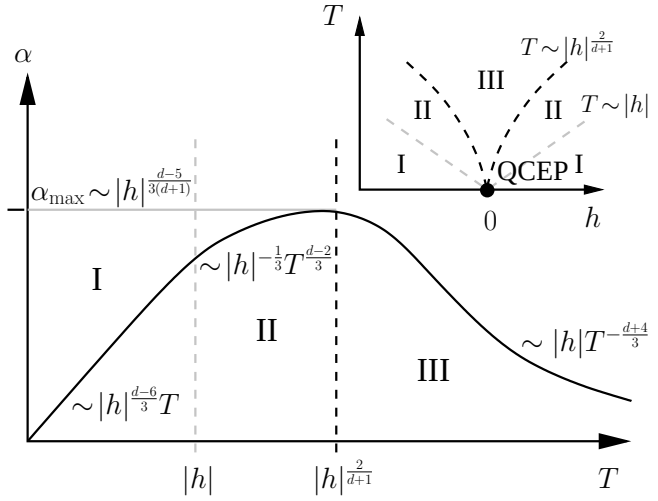

FIG. 2: Behaviour of the thermal expansion, α, in d dimensions near the metamagnetic QCEP of (1), as a function of temperature, T, and distance to the critical field, $h \propto H - H_c$. As explained in the text, $\alpha(-h) = -\alpha(h)$. Note that the intermediate regime II shrinks to zero with h. In $d = 2$ additional logarithmic corrections are present; II: $\alpha \sim |h|^{-1/3} \log \frac{T}{|h|}$, III: $\alpha \sim h/(T^2 \log \frac{1}{T})$.

ments [9] they need to be offset by 0.037 T. The linear thermal expansion coefficient $\alpha_c = d(\Delta L_c/L_c)/dT$ is obtained by calculating the slope of the relative c-axis length change in temperature intervals of 40 mK. Additionally, the electrical resistivity of a small plate-like piece from the same batch has been measured using a low-frequency four-point ac-method. The residual resistivity ratio of this sample equals 150.

Fig. 1 shows the temperature dependence of α_c in different fields between 6 and 10 T for temperatures up to 4 K. Strikingly, the thermal expansion changes sign across the metamagnetic cross-over. This was also observed near the metamagnetic cross-over in the heavy-fermion compound $CeRu_2Si_2$ [11]. Moreover, as the critical field H_c is approached the pronounced peak in α_c increases, narrows and shifts towards lower temperatures.

The characteristic shape of the thermal expansion as a function of T and H can be understood within the scenario of a QCEP. As outlined in Ref. [4], near the critical magnetic field, H_c, thermodynamics is governed by the longitudinal fluctuations of the magnetic polarization of the Fermi sea described by an Ising field ψ with the action

$$\mathcal{S}[\psi] = \int \frac{d^dk}{(2\pi)^d} \frac{1}{\beta} \sum_{\omega_n} \frac{1}{2} \left[r_0 + k^2 + \frac{|\omega_n|}{k} \right] |\psi(\omega_n, k)|^2 + \int_0^\beta d\tau \int d^dx \left[\frac{u}{4!} \psi^4(\tau, x) - h\psi(\tau, x) \right] . \quad (1)$$

When the system is tuned to its QCEP, $r_0 \approx 0$, which will be assumed in the following, these fluctuations are very soft near the metamagnetic transition resulting in strong thermodynamic signatures. The deviation of the magnetic field from its critical value, $h \propto H - H_c$, then acts as the control parameter of metamagnetism.

The leading contributions to the thermal expansion, $\alpha = V_m^{-1}(dV/dT)_{p,H} = -V_m^{-1}(dS/dp)_{T,H}$ (V_m: molar volume) will generically derive from the pressure dependence of the most relevant coupling, i.e. h or, equivalently, the critical magnetic field, $H_c = H_c(p)$. Derivatives of the free energy with respect to H and p then probe the same thermodynamic information, which would naturally explain the close correspondence between magnetostriction and susceptibility observed in $Sr_3Ru_2O_7$ [9]. For the critical part of the thermal expansion we get the relation $\alpha = \Omega V_m^{-1}(\partial S/\partial H)_T$, where $\Omega \equiv (dH_c/dp)_{H=H_c} \approx 5.6$ T/Gpa [12] is a measure for the (linear) hydrostatic pressure dependence of H_c. The corresponding relation also holds for the c-axis thermal expansion and the c-axis uniaxial pressure dependence of the critical field, which will explain the observed sign change of α_c across the metamagnetic transition, see Fig. 1. Indeed, as detailed in [13] a change of sign of the thermal expansion is a generic phenomenon near QCPs. The soft fluctuations associated with the $T = 0$ critical point enhance the entropy at finite T; the entropy will therefore increase when H_c is approached, which is reflected in a sign change of $\alpha \propto (\partial S/\partial H)_T$ across the transition. In particular, the accumulation point of entropy is identified by a vanishing thermal expansion.

Within the theory (1) this accumulation of entropy is expected exactly at $H = H_c$ ensured by the (emergent) reflection symmetry $\psi \to -\psi$ at $h = 0$, leading to the property $\alpha(-h) = -\alpha(h)$ near the QCEP. The behaviour of the thermal expansion deriving from (1) is summarized in Fig. 2. If the system is tuned away from

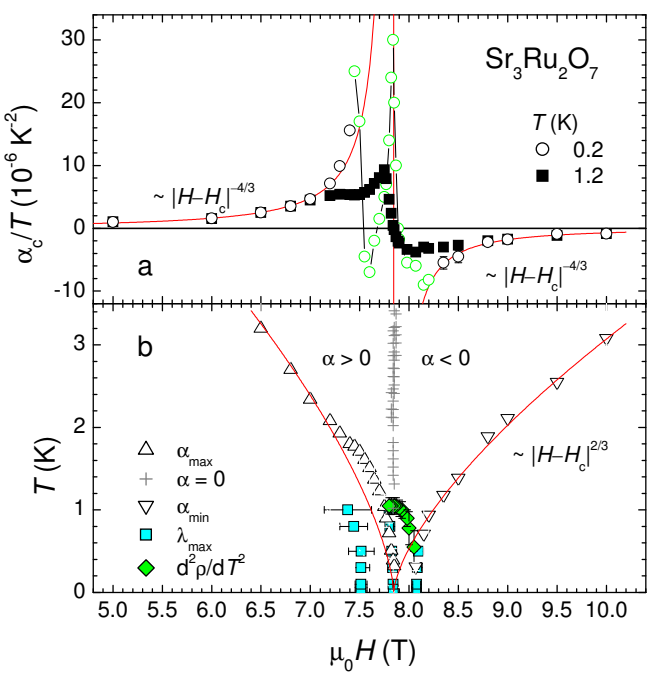

FIG. 3: (color online) (a): Field dependence of the thermal expansion coefficient of $Sr_3Ru_2O_7$ as α_c/T vs $\mu_0 H$ at 0.2 K (open circles) and 1.2 K (closed squares). The green open circles indicate data in the non-Fermi liquid regime, $\partial^2\alpha_c/\partial T^2 \neq 0$. The red solid lines show a fit $|H - H_c|^{-4/3}$ with $\mu_0 H_c = 7.845$ T. (b): Corresponding (T, H) diagram. Triangles and crosses show positions of thermal expansion extrema and zeros, respectively. Squares and diamonds indicate positions of peaks in magnetostriction and in $d^2\rho/dT^2$, respectively, see also Fig. 5.

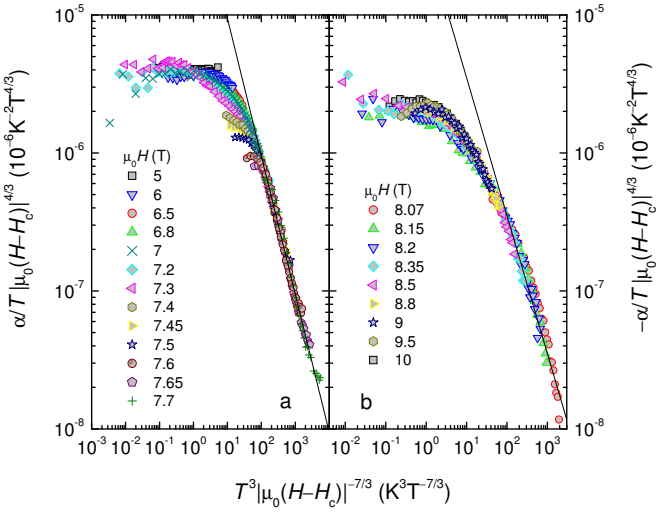

FIG. 4: (color online) Linear c-axis thermal expansion coefficient of $Sr_3Ru_2O_7$ rescaled as $\alpha_c/T \times |\mu_0(H - H_c)|^{4/3}$ vs $T^3 \times |\mu_0(H - H_c)|^{-7/3}$ with $\mu_0 H_c = 7.845$ T (on a double-log scale) for low-field (a) as well as high-field (b) data. For 7.4 T $\leq \mu_0 H \leq 8.07$ T data for temperatures below 1.1 K have been omitted. Lines indicate $\alpha_c \propto |\mu_0(H - H_c)|T^{-2}$.

the metamagnetic transition, $h \neq 0$, the field ψ attains a non-zero expectation value. At low temperature its response is non-linear, $\psi \propto |h|^{1/\delta}$, with the mean-field exponent $\delta = 3$. Thermodynamics is determined by the fluctuations around this mean-field solution giving rise to regimes I and II. The cross-over I/II is characterized by incipient deviation from Fermi-liquid behaviour. At elevated temperatures however thermal fluctuations will stabilize the mean-field potential resulting in a weaker, linear response, $\psi \propto h$. In this linear response regime III the behaviour of α is captured by renormalized mean-field theory giving a maximum of $|\alpha|$ at the crossover II/III followed by a steep decline at higher T.

For comparison with experiment the magnetic field dependence of the thermal expansion coefficient, α_c/T, is shown in Fig. 3a. At elevated temperatures (1.2 K) the behavior resembles that observed near the metamagnetic cross-over in $CeRu_2Si_2$ [11] with a sign change of α_c/T occuring at $\mu_0 H_c = (7.845 \pm 0.005)$ T. Using this value for the critical field, we can describe the field dependence inside the Fermi liquid regime by $\alpha_c/T \propto |H - H_c|^{-\epsilon}$ with $\epsilon = 1.35 \pm 0.1$, in agreement with the expectation for two-dimensional (2D) spin fluctuations ($\epsilon = 4/3$ red solid lines in Fig. 3a). Two-dimensional spin fluctuations were also observed in inelastic neutron scattering [14] at zero magnetic field which revealed that they are confined to the RuO_2 bilayers of the crystal structure. Close to the metamagnetic region, the low-T thermal expansion in Fig. 3a shows complicated behavior with three sign changes in a narrow field interval, which are attributed to the fine structure near H_c mentioned in the introduction. Outside this field region, the zeros of α are independent of H, see Fig. 3b. The positions of thermal expansion extrema follow a $|H - H_c|^{2/3}$ dependence as expected in $d = 2$ (red line in Fig. 3b). However, at lowest temperatures they do not coincide at a *single* (quantum critical end) point at $T = 0$ but merge with the two lines of first-order transitions. The presence of these first-order transitions might also be at the origin of the missing symmetry $\alpha(-h) = -\alpha(h)$ already apparent in Fig. 1.

The scaling-plot in Fig. 4 analyzes the full shape of α_c in more detail; however, omitting data at temperatures $T \leq 1.1$ K for fields very close to H_c. The thermal expansion has been rescaled such that a data collapse for the low- and high temperature asymptotes, regime I and III, should occur for a 2D QCEP. Note that the presence of the intermediate regime II prevents a full collapse onto a single scaling curve and deviations in this regime are even theoretically expected. The scaling behaviour of the raw data is remarkable given the uncertainties associated with a possible small linear in T background (see $\mu_0 H = 7.8$ T in Fig. 1). Moreover, in the low-field region, $H < H_c$, the additional anomaly at 7.5 T leads to distortions at low temperatures. The lines indicate the high-T asymptotes expected in $d = 2$. Whereas in the high-field

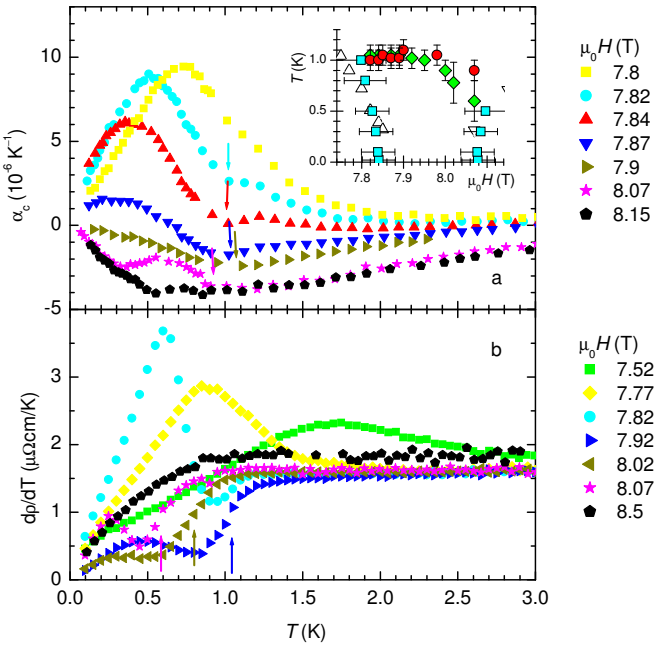

FIG. 5: (color online) (a): Temperature dependence of the thermal expansion coefficient α_c (a) and resistivity derivative $d\rho/dT$ (b) of $Sr_3Ru_2O_7$ at various magnetic fields $H \parallel c$. Arrows indicate anomalies associated with the entry into the bounded state. The inset displays the (T,H) phase diagram (cf. Fig. 3b) for the region very close to the metamagnetic transitions. Red circles, and green diamonds mark positions of arrows in (a) and (b), respectively.

region the fit is not compulsory, the behaviour in the low-field region is convincingly described by two-dimensional metamagnetic fluctuations. Note however that a recent NMR study [15] indicates that the full fluctuation spectrum near the critical field might also contain in addition an antiferromagnetic component.

At last, we concentrate on the field regime near 8 T between the two first-order transitions [8] (squares in inset of Fig. 5a) where at low temperatures an enhanced residual resistivity was observed [9]. As shown in Fig. 5 this regime in the (T,H) diagram is bounded in temperature by a cross-over line connecting the two first-order transitions where the thermal expansion and especially the electrical resistivity show anomalies. Here, the resistivity derivative drops sharply (cf. arrows in Fig. 5); the inflection points determined from the maxima in $d^2\rho(T)/T^2$ define the positions of the diamonds in the phase diagram shown in the inset. A discontinuous change in thermal expansion, expected when crossing a phase transition cannot be resolved. By contrast, only weak minima are observed, whose positions agree well with the cross-over in the resistivity and signatures in the dc-magnetization $M(T)$ found previously [9].

Because the lines of first-order transitions have slopes pointing away from the bounded state, it is possible to fine-tune the field such that this region subsequently is first entered and then left upon cooling. This leads to distinct anomalies in both the thermal expansion and the electrical resistivity observed, e.g., at a field $\mu_0 H = 8.07$ T that are absent at larger fields when the new state is not entered. The shape of the bounded region has interesting consequences for its entropic properties. From the Clausius-Clapeyron relation follows that the entropy increases when this state is entered upon crossing the first order transitions at constant temperature. A possible explanation for the enhanced entropy might be that the soft fluctuations are not completely cutoff at the crossover but rather survive within the bounded regime. Note that the formation of the bounded state at $T \approx 1$ K leads only to a small dip in the thermal expansion (Fig. 5a), and a pronounced peak characteristic for the presence of soft fluctuations also develops within the bounded region below 1 K at a magnetic field of e.g. 7.84 T. The observation of an enhanced entropy is also consistent with the NMR results [15] which showed that quantum critical fluctuations near the metamagnetic field persist within the bounded region down to lowest temperatures.

These results place additional constraints on the possible nature of the bounded region. Its enhanced entropy as well as the absence of phase transition signatures in the thermal expansion at the crossovers near 1 K are difficult to reconcile with a symmetry-breaking mechanism for the phase formation as proposed in [9] but it cannot be excluded. If the incorporation of fluctuation effects in a phase separation scenario [10] is able to explain the experimental signatures as e.g. the shape of the bounded region remains to be seen. Thus, further theoretical and experimental work is needed for clarification.

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