Critical end-point and metamagnetic quantum criticality in \(\text{Sr}_3\text{Ru}_2\text{O}_7\)

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We report a metamagnetic critical point at \((B^*,T^*) \approx (5.1 \text{ T}, 1.1 \text{ K})\) for magnetic fields applied perpendicular to the (tetragonal) c-axis of \(\text{Sr}_3\text{Ru}_2\text{O}_7\). First-order behaviour well below \(T^*\) indicates that \((B^*,T^*)\) marks a critical end-point that terminates a line of first-order metamagnetic transitions. The absence of first-order behaviour in the metamagnetic transition with \(B||c\) confirms that the non-Fermi liquid behaviour for \(B||c\) is underpinned by a metamagnetic quantum critical end-point for which \(T^* \to 0\). Scaling behaviour of the resistivity under hydrostatic pressure yields the surprising result that although \(B^*\) increases rapidly with pressure, \(T^*\) has only a weak pressure dependence.

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The intense exploration of the perovskite ruthenates over the past few years has revealed a wide range of challenges to our understanding of the metallic state. This family includes the unconventional superconductor \(\text{Sr}_2\text{RuO}_4\) \(^1\) and the itinerant ferromagnet \(\text{SrRuO}_3\) \(^2,3\); Ca-doping \(\text{Sr}_2\text{RuO}_4\) at the Sr site even provides access to a metal-insulator transition \(^4\). We recently reported evidence of non-Fermi liquid behaviour in \(\text{Sr}_3\text{Ru}_2\text{O}_7\) in the vicinity of a metamagnetic transition which occurs at fields \(B_M(T)\) which depend on field orientation, with \(B_M(0) = 5.1 \text{ T for } B \perp c\), and \(B_M(0) = 7.8 \text{ T for } B||c\) \(^5,6\). There are two regimes of non-Fermi liquid behaviour. Above \(1 \text{ K}\) the metamagnetic transition broadens rapidly with temperature indicating a fall in the susceptibility for \(B \sim B_M\) and there is a quasi-linear temperature dependence of the resistivity over a large region of the \((B,T)\) plane centered on \(B_M\); also, in measurements with \(B||c\), a logarithmic divergence has been seen in the electronic specific heat, \(C(T)/T \sim -\ln T\) \(^7\). Below \(1 \text{ K}\) in contrast, non-Fermi liquid behaviour is confined to \(B \sim B_{M||}\); for other values and orientations of \(B\) the susceptibility and resistivity cross-over to conventional Fermi-liquid behaviour. The resistivity in particular shows \(T^2\) behaviour, but the range of temperature in which this is observed collapses as \(B \to B_{M||}\) while the coefficient \(A\) in \(\rho(T) = \rho_0 + AT^2\) becomes divergent. Concurrently, for \(T \to 0\) (and \(B||c\)) a novel manifestation of non-Fermi liquid behaviour appears as a power law in the resistivity that is higher than \(T^2\), being closer to \(T^4\) \(^8\). It has been speculated that the \(T^4\) behaviour signals the onset of a qualitatively new state, driven by divergent fluctuations and susceptibility associated with a so-called metamagnetic quantum critical end-point \(^9\).

Historically, metamagnetism refers to magnetic field induced phase transitions of local moment antiferromagnetic insulators. Instead, the itinerant metamagnetism addressed in this Letter is thought to arise from a magnetic field induced spin splitting of the Fermi surface. Metamagnetism of this kind has been detected in numerous metallic magnets; in some systems the rapid rise in the magnetisation \(M\) is continuous (‘metamagnetic-like’ behaviour \(^10,11\)) but in others the metamagnetic transition can be a true phase transition with a discontinuous jump in \(M\) as a function of \(B\) \(^1\). In a true metamagnetic transition the amplitude of the metamagnetic jump \(\Delta M\) decreases as \(T\) increases, vanishing at a critical point \((B^*,T^*)\) in the \((B,T)\) plane. Above \(T^*\) one sees crossover behaviour. The termination of the line of first-order transitions at an isolated critical end-point is analogous to the liquid–vapour co-existence curve. If the critical end-point were to occur at \(T^* = 0\) one would have a metamagnetic quantum critical end-point.

Non-Fermi liquid behaviour is often seen in systems with metamagnetic or metamagnetic-like transitions. For instance, \(\text{UCoAl}\) \(^12\) has a first order metamagnetic transition at \(B \sim 0.65 \text{ T}\), and a critical point at \((B^*,T^*) \sim (0.8 \text{ T}, 13 \text{ K})\) \(^13\). However, the reported non-Fermi liquid behaviour in the resistivity as \(T \to 0\) is assumed to arise from the proximity to a zero field ferromagnetic quantum critical point, rather than a metamagnetic one \(^14\). In \(\text{MnSi}\), though, the vicinity to itinerant metamagnetism at high pressures may be responsible for the observed non-Fermi liquid behaviour over a very large region of \(T\) and \(\rho\), but the quantum critical end-point
does not appear to play a special role. Regarding the heavy fermion metamagnetic systems, such as CeRu$_2$Si$_2$ and UPt$_3$, non-Fermi liquid properties have also been observed, but none of the heavy fermion systems studied to date have shown a first-order jump in $M$ for any combination of parameters. Thus the relevance of fluctuations associated with a quantum critical end-point cannot be unambiguously demonstrated.

In this Letter we report the presence of a line of first order metamagnetic phase transitions in Sr$_3$Ru$_2$O$_7$ terminating in a critical end-point for $B \perp c$. The absence of such first-order behaviour in the metamagnetic transition with $B \parallel c$ implies that $T^*$ vanishes at some intermediate angle, thus supporting the suggestion that a quantum critical end-point underlies the non-Fermi liquid behavior for $B \parallel c$. Moreover we investigate how the critical point evolves under pressure, showing that although $B^*$ rises rapidly with pressure, $T^*$ is comparatively unaffected. This unexpected pressure dependence of the critical end-point, inferred from the resistivity, shows that quantum criticality in Sr$_3$Ru$_2$O$_7$ is not consistent with the standard model of itinerant metamagnetism.

The single crystals of Sr$_3$Ru$_2$O$_7$ examined here were grown from high purity starting materials in an infrared image furnace in Kyoto. The high sample quality was confirmed by measurements of the d.c. magnetisation and resistivity ratio before and after pressurisation in a miniature Cu:Be clamp cell. The ambient pressure properties of the samples studied here correspond to those observed and reported for around 50 other crystals from the same batch.

A.c. susceptibility measurements were carried out on a 1 mm$^3$ sample in an 18 T cryomagnetic facility with a base temperature of 6 mK. A small modulation field of less than 0.01 T was applied at a frequency of 77 Hz. Au wires were Ag-epoxied onto the samples and then soldered to Cu-clad NbTi leads inside the pressure cell. A 1:4 methanol-ethanol mixture served as pressure transmitting medium which ensured that the pressure was highly isotropic at low $T$. The low temperature resistivity was measured in a commercial 12 T variable temperature cryomagnet with a $T$ vs $B$ measurement facility. Shown in Fig. 1 is the $M$ vs $T$ curve for a sample in an 18 T cryomagnetic facility with a $T$ vs $B$ measurement facility. The inset shows the position of the peak in the $(B, T)$ plane, with a solid line representing the first-order regime. Nevertheless, the magnitude of the peak, given in absolute units in Fig. 1, does not diverge as expected from a naive Landau-Ginzburg free energy for a metamagnetic transition, nor from a more sophisticated treatment more appropriate to a quantum critical metamagnetic transition. The simplest explanation we can suggest is that slight inhomogeneities in the sample smear out the transition, and indeed $T^*$ has shown some variation between samples though we believe that this may reflect in-plane anisotropy of the metamagnetic transition.

We turn next to the basal plane resistivity, used as a probe of the electronic behaviour. Shown in Fig. 2 is the normalised, isothermal magnetoresistance $\rho(B)/\rho(B = 0)$ measured at 1.5 K at several pressures up to 10 kbar, for both $B \parallel c$ and $B \perp c$. At the lowest $T$ a double peak structure may be resolved at the fields $B_{M,1}$ and $B_{M,2}$ for $B \perp c$. Together with $B_{M,2}$ (for $B \parallel c$) there are three identifiable anomalies. An extrapolation of these three transition fields to negative pressures, shown in Fig. 3, reveals that they all extrapolate to $B_{M,2} = 0$ at the same negative pressure $p_{c,\perp} = p_{c,||} = p_c \approx -14$ kbar. This suggests that at a hypothetical negative pressure $p_c$, there is a zero-field ferromagnetic transition. The existence of a zero-field quantum critical point at negative pressure is reinforced by the fall in $A$, the $T^2$ coefficient of resistivity, measured at $B = 0$, with increasing pressure (see Fig. 3, inset).

Our resistivity curves as a function of pressure are very reminiscent of those observed in CeRu$_2$Si$_2$, in which it was found that there is scaling of the form

$$M(H, T, p)/\mu_B = \psi(H/H_s(p), T/T_s(p))$$

where $H_s(p)$ and $T_s(p)$ are scaling parameters. We find similar scaling behaviour when we plot the relative change of the magnetoresistance $\Delta \rho = \rho(B) - \rho(B = 0)$, normalised by $\Delta \rho(B_{M,1})$, versus $B/B_{M,1}$ and $B/B_{M,2}$ ($B_{M,2}$ corresponding to the average of $B_{M,1}$ and $B_{M,2}$). This quantity is seen to evolve in a universal manner, implying that $\rho(B, T, p)$ scales with the critical field and may be described by a function $\rho(B, T, p)$. The inset of Fig. 1 shows the position of the peak in the $(B, T)$ plane, with a solid line representing the first-order regime. Nevertheless, the magnitude of the peak, given in absolute units in Fig. 1, does not diverge as expected from a naive Landau-Ginzburg free energy for a metamagnetic transition, nor from a more sophisticated treatment more appropriate to a quantum critical metamagnetic transition. The simplest explanation we can suggest is that slight inhomogeneities in the sample smear out the transition, and indeed $T^*$ has shown some variation between samples though we believe that this may reflect in-plane anisotropy of the metamagnetic transition.

In the standard model, pressure dependence is built in via one parameter only, $\chi(T = 0)$. For the case of Sr$_3$Ru$_2$O$_7$ both $B^*$ and $T^*$ are expected
to be equally strongly pressure dependent, in stark contrast with experimental observations. The actual strong pressure dependence of $B^*$ and the weak $p$ dependence of $T^*$ require instead an additional pressure dependence of the other parameters, such as the mode-mode coupling term or the frequency and momentum spread of the spin fluctuation spectrum. The required cancellation of $p$-dependencies for $T^*$ would be purely accidental. The non-divergence of $\chi$ at the critical point, combined with the pressure independence of $T^*$, suggests that the underlying physics of the metamagnetic transition in Sr$_3$Ru$_2$O$_7$ is beyond the standard sixth-order Ginzburg-Landau theory in which $M$ is the expansion parameter $[17]$. CeRu$_2$Si$_2$ might provide an interesting alternative model: there the metamagnetic-like transition is a crossover associated with the destruction of antiferromagnetic correlations $[24]$; moreover magnetovolume effects provide positive feedback to dramatically sharpen what would, at constant volume, be a rather broad crossover $[8, 25]$. If the magnetovolume coupling were just a little bit stronger it appears that the crossover could become first order.

Although bulk thermodynamic measurements show that Sr$_3$Ru$_2$O$_7$ is nearly ferromagnetic $[26]$, recent neutron scattering experiments have found that a fall in $\chi(T)$ below 16 K at $B = 0$T is associated with the growth of antiferromagnetic spin fluctuations $[27]$. Moreover lattice distortions in the form of tilts and rotations of the oxygen octahedra couple strongly to magnetism in the ruthenates $[28, 29, 30]$, and they are thought to be decisive in stabilising ferro- vs. antiferromagnetism $[31, 32]$. Sr$_3$Ru$_2$O$_7$ has a rotation-distortion of $\sim 7^\circ$ about the c-axis $[28, 29]$ that provides an obvious mechanism for positive magnetoelastic feedback for the metamagnetic transition. So it seems plausible that a drop in antiferromagnetic correlations, made discontinuous by positive feedback from magnetoelastic coupling, could produce the observed first-order metamagnetic behaviour in Sr$_3$Ru$_2$O$_7$.

This model might explain why $\chi$ does not diverge at the metamagnetic critical point, and critical fluctuations at finite $q$ would contribute to the large peak in the resistivity seen at $B_M$ in our resistivity curves. It is not clear that the CeRu$_6$Si$_2$ model explains the weak pressure dependence of $T^*$, but some experimental tests suggest themselves: in particular, neutron scattering could be used to track the field dependence of both antiferromagnetic correlations (as was done in CeRu$_2$Si$_2$ $[24]$) and the rotation angle of the octahedra through the metamagnetic transition.

In conclusion, we have found a metamagnetic critical end-point at finite temperature for fields applied in the basal plane $B \perp c$ of Sr$_3$Ru$_2$O$_7$, though the peak in the susceptibility at the critical point is much smaller than expected. We have in addition shown that the application of pressure drives the critical point up in field, but surprisingly does not produce a substantial shift of the critical temperature.

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FIG. 2: Normalised magnetoresistance for $B \parallel c$ (upper) and $B \perp c$ (lower) in Sr$_3$Ru$_2$O$_7$ for pressures up to 10 kbar (increasing pressure from left to right) at 2.5 K.

FIG. 3: Increase of the metamagnetic transition fields $B_{||}$, $B_{\perp 1}$ and $B_{\perp 2}$ in Sr$_3$Ru$_2$O$_7$ as a function of pressure, where $B_{\perp 1}$ corresponds to the lower field value of the double transition and $B_{\perp 2}$ the higher. In any case, all three critical fields extrapolate to a unique negative critical pressure $p_c \approx -14$ kbar.

FIG. 4: Normalised magnetoresistance (refer to text) as a function of the magnetic field scaled by the respective critical field $B_M$. Left: $B_M = (B_{\perp 1} + B_{\perp 2})/2$ is an average of the two critical fields for $B_{\perp c}$. Right: $B_M = B_{||}$.

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