Phase Bifurcation and Quantum Fluctuations in Sr$_3$Ru$_2$O$_7$

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The bilayer ruthenate Sr$_3$Ru$_2$O$_7$ has been cited as a textbook example of itinerant metamagnetic quantum criticality. However, recent studies of the ultra-pure system have revealed striking anomalies in magnetism and transport in the vicinity of the quantum critical point. Drawing on fresh experimental data, we show that the complex phase behavior reported here can be fully accommodated within the framework of a simple Landau theory. We discuss the potential physical mechanisms that underpin the phenomenology, and assess the capacity of the ruthenate system to realize quantum tricritical behavior.

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In recent years, the field of itinerant electron metamagnetism has seen a resurgence of interest, much of it connected with quantum criticality. The phenomenon of metamagnetism can be thought as a magnetic equivalent of a liquid-gas transition with the role of pressure $P$ and density being played by the magnetic field $H$ and magnetization. At this level, the $(H, T)$ phase diagram of a metamagnet translates to the well-known $(P, T)$ phase diagram of the liquid-gas system; a first order phase boundary terminating in a critical point. However, in contrast to the liquid-gas system, the metamagnet plays host to a crystalline lattice, the “source” of the itinerant electrons. It is this difference that is responsible for the interesting new phenomena that can be observed in this system. Firstly, the coupling of electrons to the magnetic field breaks the spatial symmetry of the lattice and modifies the electronic orbitals. Such effects play a fundamental role in shaping the effective interaction between the electrons. This field sensitivity can be used to tune the critical end-point of the first order transition to zero temperature and, thereby, realize a quantum critical point. Secondly, as we will show below, the coupling of the itinerant electron system to the lattice may, by itself, induce striking changes in the phase diagram.

The existence of metamagnetic quantum critical points (QCPs) was demonstrated in Sr$_3$Ru$_2$O$_7$ where it was shown that the field angle $\theta$ (measured with respect to the $ab$-plane) acts as a tuning parameter, allowing the construction of an $(H, T, \theta)$ phase diagram for metamagnetism and quantum criticality. The metamagnetic transition tracks a first order line in the $(H, \theta)$ plane which terminates at a QCP when the energy/temperature scale associated with the metamagnetic transition is tuned to zero. Recently, intriguing evidence has been reported for a new mechanism by which quantum critical behaviour may breakdown in samples of extremely clean Sr$_3$Ru$_2$O$_7$. These new effects are strongly dependent on purity and are seen only in the best crystals with residual resistivity $\rho_0 < 1 \mu\Omega\text{cm}$. So far, this new behaviour has been studied mainly with the applied field oriented along the crystallographic $c$-axis, where the single metamagnetic QCP seen in lower purity samples is replaced by two first order metamagnetic transition lines at approximately 7.8T and 8.1T, each of which terminates in a finite temperature critical point with $T_c < 1$K (see Fig. 1). These lines enclose a region of anomalous transport and thermodynamic properties which extends up to a temperature scale of ca. 1K. A clear correlation exists between features in magnetic susceptibility and magnetostriction, demonstrating a strong magnetostructural coupling in Sr$_3$Ru$_2$O$_7$.

![Image](https://example.com/image.png)

**FIG. 1:** Measurements of the imaginary part of the a.c. susceptibility ($\chi''$) for $H \parallel c$ ($\theta = 90^\circ$) as a function of field for different temperatures. By correlating maxima in $\chi''$ with peaks in $\chi'$ (not shown), the magnetic phase diagram can be inferred. The peaks in $\chi''$ are due to dissipation associated with the crossing of a first order phase boundary. The phase boundaries and critical points inferred from the data at this field orientation are shown in red.

Building on the preliminary angle-dependent resistivity data reported in Ref. 2, the aim of this letter is two-fold: Firstly, we report measurements of resistivity $\rho$ and a.c. magnetic susceptibility $\chi$ which confirm and extend the earlier $c$-axis data revealing an intricate phase diagram where the line of first order metamagnetic transitions in the $(H, \theta)$ plane appears to bifurcate into the two transitions observed for $H \parallel c$. Secondly, we will show...
that this complex phase behaviour can be accommodated within the framework of a Landau phenomenology which ascribes the bifurcation to a “symmetry-broken” tricritical point structure. We discuss how the phenomenology, combined with the anomalous resistivity behaviour in the bifurcated region, places strong constraints on the physical mechanisms active in the Sr₃Ru₂O₇ system. Further, we address the potential experimental signatures of quantum tricritical behaviour.

![Image](67x418 to 290x637)

FIG. 2: Experimental phase diagram of ultra-pure crystals of Sr₃Ru₂O₇ as inferred from a.c. magnetic susceptibility data measured at 89Hz using similar techniques to those explained in detail in Ref. [4]. The planes record the loci of peaks in $\chi''$ (cf. Fig. 4), while the thick black lines show absolute maxima in $\chi'$. The vertical lines and dots identify the data which have been interpolated to construct the figure. At each angle the field was swept through the metamagnetic region at fixed temperatures ranging from 50 mK to 1.4K in steps of 100 mK. The inset shows the phase diagram associated with the Landau theory of $\chi$ with the parameter $s$ and orientation chosen to match the geometry of experiment (see main text).

The new data reported here are based upon detailed studies of the angular dependence of a.c. magnetic susceptibility ($\chi$) and resistivity ($\rho$). By correlating peaks in $\chi''(H,T)$ with absolute maxima in $\chi'(H,T)$, the loci of first order metamagnetic transitions and their critical end-points can be traced (see sample data in Fig. 1). Fig. 2 shows the detailed phase diagram inferred from a sequence of measurements taken at different angles $\theta$ on high purity single crystals with $\rho_0 < 0.7\mu\Omega cm$. In less pure samples, the temperature of the end-point is shown to fall monotonically with increasing angle, and is depressed to below 100 mK at angles $\theta \gtrsim 80^\circ$ [4]. Here, in the purer samples, one can see that the dependence is non-monotonic, with the critical line rising slightly in temperature for large $\theta$ [10]. At the same time, a second surface of first order transitions emerges, with an endpoint that rises with $\theta$. The complementary study of $\rho$, shown in Fig. 3 confirms that these first order transitions enclose a region of anomalously high $\rho$ when $H$ is aligned very close to the $c$-axis [3, 4]. Even when $\theta < 85^\circ$, where the anomaly in the $\rho$ is weak, one can identify two distinct ridges bifurcating from a single ridge at an angle of $\theta \approx 60^\circ$, a result consistent with that inferred from a $T = 100$ mK section through the magnetic phase diagram [14].

![Image](434x567 to 468x568)

FIG. 3: Resistivity data recorded at a fixed temperature $T = 100$ mK taken from the same range of field $H$ and angle $\theta$ as that used in Fig. 2.

Although the magnetic phase diagram is rich, the detailed bifurcation structure can be accommodated within the framework of a Landau functional which involves the simplest generalization of the canonical theory: At the mean-field level, in the vicinity of a conventional metamagnetic critical point, the Landau free energy can be expanded as $\beta F_0 = hm + \frac{r}{2}m^2 + \frac{s}{3}m^3 + \frac{u}{4}m^4 + \frac{1}{6}m^6$, where $m$ denotes the deviation of the magnetisation density from its value $M_s$ at the critical point $h^* = r^* = 0$. The parameters $r$ and $h$ (themselves functions of $T$, $H$, and $\theta$) span, respectively, directions parallel and perpendicular to the line of first order transitions, and $u > 0$. (The presence of a large external field in the metamagnet makes the system effectively uniaxial.) However, if the sign of the interaction $u$ is reversed, one is compelled to consider the generalization,

$$\beta F[m] = hm + \frac{r}{2}m^2 + \frac{s}{3}m^3 + \frac{u}{4}m^4 + \frac{1}{6}m^6,$$

where the presence of the cubic term in $m$ reflects the fact that, in the metamagnetic system, only the quintic term may be removed by rescaling.

To understand how the phase behaviour is recovered from [11], it is instructive to consider first a “symmetric” theory with $s = 0$. In this case, a change in the sign of $u$ leads to tricritical phenomena [12, 13]. As shown in Fig. 4, the phase diagram is characterised by a bifurcation of the critical line at the tricritical
point: \( h^* = u^* = r^* = 0 \). For \( u > 0 \), the critical line bounds a plane of first order transitions while, for \( u < 0 \), two critical lines bound first order planes which coalesce into a single plane along a line of degeneracy. The trajectories of the bifurcated critical lines for \( u < 0 \) are given by \( h^*(u) = \pm 6u^2(3u/10)^{1/2}/25 \), \( r^*(u) = 9u^2/20 \) while the line of degeneracy follows a trajectory \( h_{de} = 0 \), \( r_{de} = 3u^2/16 \). Restoring the cubic contribution, the point of bifurcation becomes ‘dislocated’ such that the second line of critical points emerges from the plane of first order transitions at a point \( P: u_{TP} = -(4s)^{1/3}, h_{TP} = su_{TP}/4 \), \( r_{de} = 3u^2/16 \) while, away from the region of bifurcation, an expansion in \( s \) shows the critical lines to asymptote to the trajectories \( h^*(u, s) \approx h^*(u) + 2us/5 \), \( r^*(u, s) \approx r^*(u) \pm s\sqrt{-6u}/5 \) (see Fig. 4b). The corresponding line of degeneracy follows the trajectory \( h_{de} = su/4 \), \( r_{de} = 3u^2/16 \).

When compared with the bifurcation structure of the measured phase diagram, the correspondence with the Landau theory is striking: The bifurcation in the experimental system is consistent with the second line of critical points emanating from a point \( P \) located at some small negative temperature and rising through the \( T = 0 \) plane at an angle of ca. 80° (see Fig. 2 inset). Moreover, the primary line of critical points shows an upturn which is also predicted by the Landau theory.

\[
\beta F[m, \psi] = \beta F_0 + \gamma (M_s + m)^2 \psi + \beta F_{\psi}[\psi].
\]

Crucially, when integrated out, fluctuations of the field \( \psi \) impart a negative contribution to the quartic interaction of \( m: u \rightarrow u - 4 \gamma^2 (\psi^2) \). What physical mechanisms could give rise to such a coupling?

In fact, there are relatively few choices available whose symmetry allows the simple coupling to magnetization given by (2). Correlations of the magnetostriction data with magnetic susceptibility reported in the \( \theta = 90^\circ \) system suggest an association of the field \( \psi \) with the lattice strain. (Indeed, this was the coupling originally considered in Refs. [15, 16, 17] .) However, a mechanism driven solely by harmonic fluctuations of the lattice sits uncomfortably with the observed disorder dependence of the bifurcation; the bifurcation appears only in the pure system and is quenched by tiny amounts of disorder, while the effect of disorder upon lattice fluctuations is likely to be weak. This difficulty may be resolved by drawing upon the physical origin of metamagnetism: By effecting an increased magnetic polarization, the Fermi level can be positioned in a region of high density of states (DoS). By combining metamagnetism with a weak structural transition or, potentially, an interaction-driven Fermi surface distortion, the system may take energetic advantage of singular features in the local (in \( k \)-space) DoS (similar to a Jahn-Teller or Peierls distortion in an insulator). Approaching the critical point of the undistorted system, a lattice or Fermi surface distortion could split the peak in the DoS and thereby advance the transition. Elastic scattering of electrons from impurities would smear out the features in the DoS that provide the energetic drive and so quench the bifurcation. Note that, due to the restrictions of symmetry, the Landau theory will take precisely the same form whether one chooses to identify \( \psi \) with the lattice strain or with the size of a Fermi surface distortion; indeed, structural distortions are inevitably accompanied by Fermi surface distortions.

As well as capturing both the observed features in magnetostriction and the quenching of the bifurcation by disorder, such a mechanism affords a natural explanation for the resistance anomaly: A degeneracy of stable lattice configurations or Fermi surface distortions would be accompanied by the nucleation of ordered domains. Once the (diverging) magnetic correlation length exceeds the domain size, the resistivity will become controlled by tiny amounts of disorder and rising through the \( T = 0 \) plane at an angle of ca. 80° (see Fig. 2 inset). Moreover, the primary line of critical points shows an upturn which is also predicted by the Landau theory.

To go beyond the level of phenomenology, it is necessary to identify physical mechanisms which may be responsible for reversing the sign of \( u \). Although one cannot rule out idiosyncrasies of the electron band structure influencing the coefficients of the Landau expansion, such effects taken alone would be unlikely to explain the extreme sensitivity to disorder and the anomalous resistivity dependence observed in the bifurcated region. However, the Landau coefficients may be adjusted indirectly by coupling the magnetization density to some auxiliary field (12, 16, 17).
ment of a tricritical point is accompanied by a substantial softening of classical and quantum fluctuations. In the present case, where \( s \neq 0 \), when the temperature exceeds the 'energy scales' of the bifurcation region \( \Theta \) (ca. 1 K), the fluctuations will remain characteristic of a quantum tricritical point (see below). As the temperature is reduced, the system will pass through two further regimes of behaviour: At the lowest temperatures, all magnetic fluctuations will be gapped. Between these high and low temperature extremes, the system will pass through a crossover regime where the behaviour will be determined by the proximity to the finite temperature critical points.

To address the behavior at higher temperatures, where fluctuations are controlled by a quantum tricritical point, one can employ an extended Hertz-Millis action \([20]\),

\[
S = \frac{1}{2} \int \frac{d\mathbf{q}}{(2\pi)^3} \frac{d\omega}{2\pi} \left( \rho_0 + |q|^2 + \frac{|\pi|}{\Gamma_\mathbf{q}} \right) |m(\mathbf{q},\omega)|^2 + \int d\mathbf{r} d\tau \left( \rho_m(\mathbf{r},\tau)^4 + \rho_0 m(\mathbf{r},\tau)^6 \right),
\]

where \( \rho_0 < 0 \), \( \rho_0 > 0 \) and \( \Gamma_\mathbf{q} = v|\mathbf{q}|. \) To leading order in the bare parameters \( \rho_0 \), \( \rho_0 \), and \( \rho_0 \), the influence of fluctuations can be incorporated by following a self-consistent renormalization procedure \([21]\) from which one obtains

\[
\begin{align*}
\langle \rho(T) \rangle &= \rho_0 + 12\rho_0 (m^2) + 9\rho_0 (m^2)^2 \\
\langle u(T) \rangle &= u_0 + 15v_0 (m^2) \\
\langle v(T) \rangle &= v_0.
\end{align*}
\]

Here the averages \( \langle \cdots \rangle \) are calculated self-consistently with the renormalized action. To leading order, one need retain only \( \rho(T) \), and perform the calculation with the corresponding quadratic action. Subtracting zero-point fluctuations, one obtains \( \rho(T) = \rho(0) + 12\rho(0)(m^2) - \langle m^2 \rangle_{T=0} + 9\rho(0)(m^2) - \langle m^2 \rangle_{T=0} )^2 \), where

\[
\langle m^2 \rangle_{T=0} = \frac{T^{4/3} - T^{1/3}}{T^{2/3} - T^{1/3}},
\]

\( T \gg r(0) \)

\[
T \ll r(0),
\]

denotes the thermal contribution to the critical fluctuations. At a conventional quantum critical point, \( r(0) = 0 \) and \( u(0) > 0 \), leading to the characteristic temperature dependence \( r(T) \propto T^{4/3} \) \([20]\). By contrast, at the quantum tricritical point, \( r(0) = u(0) = 0 \), leading to an enhanced temperature dependence \( r(T) \propto T^{8/3} \) reflecting the shallow potential for fluctuations. Translated to the electron self-energy, the magnon fluctuations contribute a factor \( \text{Im} \Sigma^R(\mathbf{k},0) \sim T^3/r(T) \) from which one infers a resistivity of \( \rho \propto T^{8/3} \) for a conventional quantum critical point and \( \rho \propto T^{4/3} \) for the quantum tricritical point.

To conclude, we have shown that the bifurcation structure observed in the magnetic susceptibility of Sr\(_3\)Ru\(_2\)O\(_7\) is consistent with a Landau phenomenology reflecting a ‘dislocated’ tricritical point structure. Further, we have argued that, by coupling lattice fluctuations to a Fermi surface instability, the Landau phenomenology provides a natural explanation of the resistance anomaly. It is interesting to note that the bifurcation mechanism described here is quite generic and may provide an opportunity to realize quantum tricritical behavior both in Sr\(_3\)Ru\(_2\)O\(_7\) and potentially more widely. Indeed, even within the ruthenate system, there is growing evidence that the neighboring metamagnetic transitions revealed in the pure system are also accompanied by bifurcation structures. The distortion of the Fermi surface through lattice instabilities or strong interactions may provide a general mechanism for clean materials to mask a magnetic quantum critical point.

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[18] Formally, a two-fold degeneracy of the lattice instability can be accommodated by introducing an Ising field into the Landau functional, viz. \[ \beta F[m, \psi, \sigma] = \beta F_0[m] + \gamma(M_+ + m)^2 \psi + \beta \psi \sigma \], where \( \sigma = \pm 1 \) indexes the orientation of the distortion. Such a term does not compromise the effect of the strain field fluctuations on the magnetisation while, by separately coupling \( \sigma \) to \( \theta \), the sensitivity of the domain structure to the orientation of the magnetic field can be addressed. Note that the domain formation does not necessitate a symmetry breaking of the Ising field in the bifurcated region.

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