Phase Bifurcation and Quantum Fluctuations in $Sr_3Ru_2O_7$

A. G. Green,[1] S. A. Grigera,[1] R. A. Borzi,[1] A. P. Mackenzie,[1] R. S. Perry,[1,2] and B. D. Simons[3]

[1]*School of Physics and Astronomy, University of St. Andrews, North Haugh, St. Andrews KY16 9SS, United Kingdom*
[2]*Department of Physics and International Innovation Center, Kyoto University, Kyoto 606-8501, Japan*
[3]*Cavendish Laboratory, Madingley Road, Cambridge CB3 OHE, United Kingdom*

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The bilayer ruthenate $Sr_3Ru_2O_7$ has been cited as a textbook example of itinerant metamagnetic quantum criticality. However, recent studies of the ultrapure 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 tricritial behavior.

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Recently, itinerant electron metamagnetism has seen a resurgence of interest [1–7], much of it connected with quantum criticality [8]. Metamagnetism can be thought of 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. This difference is responsible for interesting and observable new phenomena. First, 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. Second, 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 $Sr_3Ru_2O_7$ [3] where it was shown that the field angle θ (measured with respect to the ab plane) acts as a tuning parameter, allowing the construction of an (H, T, θ) phase diagram for metamagnetism and quantum criticality [4]. The metamagnetic transition tracks a first-order line in the (H, θ) 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 behavior may break down in samples of extremely clean $Sr_3Ru_2O_7$ [5,9]. These new effects are strongly dependent on purity and are seen only in the best crystals with residual resistivity $\rho_0 < 1\ \mu\Omega$ cm. So far, this new behavior has been studied mainly with the applied field oriented along the crystallographic c axis, where the single metamagnetic QCP seen in lower purity samples [3,4] is replaced by two first-order metamagnetic transition lines at approximately 7.8 T and 8.1 T, 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. 1 K [9]. A clear correlation exists between features in magnetic susceptibility and magnetostriction, demonstrating a strong magnetostructural coupling in $Sr_3Ru_2O_7$ [9].

Building on the preliminary angle-dependent resistivity data reported in Ref. [9], the aim of this Letter is twofold: first, we report measurements of resistivity ρ and ac magnetic susceptibility χ 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, θ) plane appears to bifurcate into the two transitions observed for $H \parallel c$. Second, we will show that this complex phase

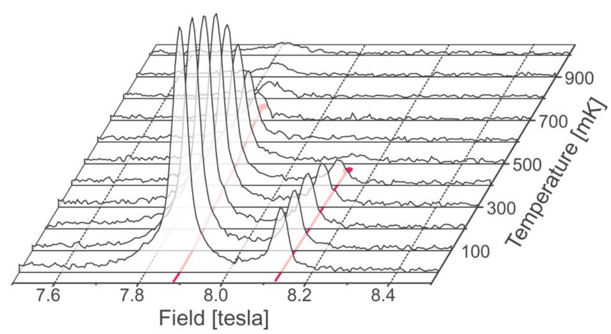

FIG. 1 (color online). Measurements of the imaginary part of the ac susceptibility (χ'') for $H \parallel c$ ($\theta = 90°$) as a function of field for different temperatures. By correlating maxima in χ'' with peaks in χ' (not shown), the magnetic phase diagram can be inferred. The peaks in χ'' are due to dissipation associated with the crossing of a first-order phase boundary [4]. The phase boundaries and critical points inferred from the data at this field orientation are shown in red.

behavior can be accommodated within a Landau phenomenology which ascribes the bifurcation to a "symmetry-broken" tricritical point structure. This phenomenology, combined with the anomalous resistivity behavior in the bifurcated region, places strong constraints on the physical mechanisms active in $Sr_3Ru_2O_7$. Further, we address the potential experimental signatures of quantum tricritical behavior.

The new data reported here are based upon detailed studies of the angular dependence of ac magnetic susceptibility (χ) and resistivity (ρ). 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 [4] (see sample data in Fig. 1). Figure 2 shows the detailed phase diagram inferred from a sequence of measurements taken at different angles θ 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 $\theta \gtrsim 80°$ [4]. Here, in the purer samples, one can see that the dependence is nonmonotonic, with the critical line rising slightly in temperature for large θ [10]. At the same time, a second surface of first-order transitions emerges, with an end point that rises with θ. The complementary study of ρ, shown in Fig. 3, confirms that these first-order transitions enclose a region of anomalously high ρ when H is aligned very close to the c axis [5,9]. Even when $\theta < 85°$, where the anomaly in the ρ is weak, one can identify two distinct ridges bifurcating from a single ridge at an angle of $\theta \approx 60°$, a result consistent with that inferred from a 100 mK section through the magnetic phase diagram [11].

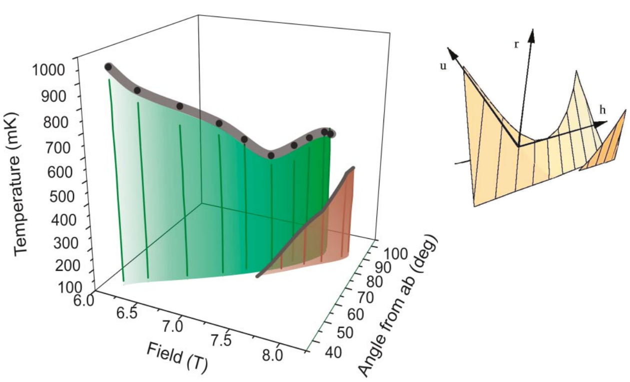

FIG. 2 (color). Experimental phase diagram of ultrapure crystals of $Sr_3Ru_2O_7$ as inferred from ac magnetic susceptibility data measured at 89 Hz using similar techniques to those explained in detail in Ref. [4]. The planes record the loci of peaks in χ'' (cf. Fig. 1), while the thick black lines show absolute maxima in χ'. 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 1.4 K in steps of 100 mK. The inset shows the phase diagram associated with the Landau theory (1) with the parameter s and orientation chosen to match the geometry of experiment (see main text).

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 $\beta F_0 = hm + \frac{r}{2}m^2 + \frac{u}{4}m^4$, where m denotes the deviation of the magnetization density from its value M_* at the critical point $h^* = r^* = 0$. The parameters r and h (themselves functions of T, H, and θ) 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 u is reversed, one must consider the generalization,

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

where the m^3 term is present since, in the metamagnetic system, only the m^5 term may be removed by rescaling.

To understand how the phase behavior is recovered from (1), 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(a), the phase diagram is characterized 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 $h^*(u) = \pm 6u^2(3|u|/10)^{1/2}/25$, $r^*(u) = 9u^2/20$ while the line of degeneracy follows a trajectory $h_{\rm deg} = 0$, $r_{\rm deg}(u) = 3u^2/16$. Restoring the cubic contribution, the point of bifurcation becomes "dislo-

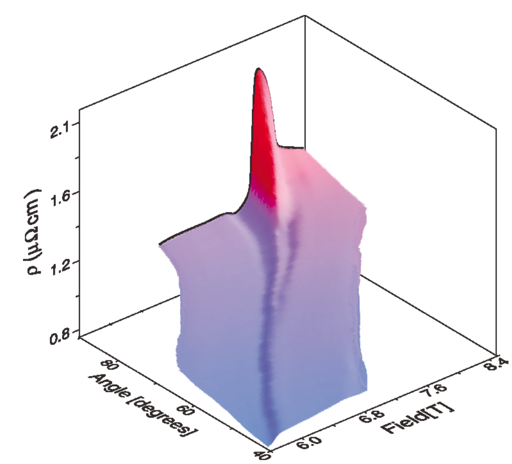

FIG. 3 (color). Resistivity data recorded at $T = 100$ mK taken from the same range of field H and angle θ as that used in Fig. 2.

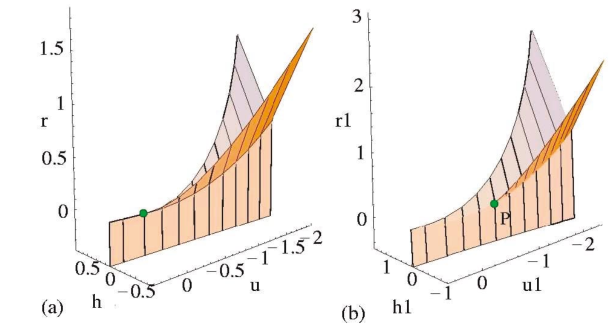

FIG. 4 (color). Phase diagram of the Landau theory (1) with (a) $s = 0$ and (b) $s = 0.2$. The planes of first-order transitions are terminated by lines of critical points. Note that, for $s = 0$, the point of bifurcation occurs at the tricritical point: $h_* = r_* = u_* = 0$ while, for $s \neq 0$, the point of bifurcation becomes dislocated. The zero temperature plane of the physical system forms a plane that bisects the critical lines near the region of bifurcation, with the second critical line emerging at some negative value of temperature (see Fig. 2 inset).

cated'' such that the second line of critical points emerges from the plane of first-order transitions at a point P: $u_p(s) = -(4s)^{2/3}$, $h_p = su_p/4$, $r_{\mathrm{deg}} = 3u_p^2/16$ while, away from the region of bifurcation, an expansion in s shows the critical lines to asymptote to the trajectories $h^*(u,s) \simeq h^*(u) + 2us/5$, $r^*(u,s) \simeq r^*(u) \pm s\sqrt{-6u/5}$ [see Fig. 4(b)]. The corresponding line of degeneracy follows the trajectory $h_{\mathrm{deg}} = su/4$, $r_{\mathrm{deg}} = 3u^2/16$.

When compared with the bifurcation structure of the measured phase diagram, the correspondence with the Landau theory (1) 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.

To go beyond phenomenology, one must identify the physical mechanism 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 observed in the bifurcated region. However, the Landau coefficients may be adjusted indirectly by coupling m to an auxiliary field [14],

$$\beta F[m, \psi] = \beta F_0 + \gamma(M_* + m)^2\psi + \beta F_\psi[\psi]. \qquad (2)$$

Crucially, integrating out ψ imparts a *negative* contribution to the quartic interaction of m; $u \mapsto -4\gamma^2\langle\psi^2\rangle$. What mechanism could give rise to such a coupling?

There are relatively few possibilities whose symmetry allows the simple coupling to m given by (2). Correlations of the magnetostriction data with magnetic susceptibility reported in the $\theta = 90°$ system [9] suggest associating ψ with the lattice strain. (Indeed, this was the coupling originally considered in Ref. [14].) 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 (k-space) DOS [9] (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. Because of the restrictions of symmetry, the Landau theory will take the same form whether one chooses to identify ψ with the lattice strain or with the size of a Fermi surface distortion; 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 [9,15]. If the (diverging) magnetic correlation length exceeds the domain size, the resistivity will become controlled by the scattering from the latter. Further, one may expect that the tilting of the magnetic field away from the c axis would lift the degeneracy, destroying the domain structure and, with it, the peak in resistivity.

To close, we consider further observable consequences of the Landau theory (1). In the symmetric ($s = 0$) theory, 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 [16] (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 behavior: 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 behavior 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 [17],

$$S = \frac{1}{2}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{d\omega}{2\pi}\left(r_0 + \mathbf{q}^2 + \frac{|\omega|}{\Gamma_{\mathbf{q}}}\right)|m(\mathbf{q},\omega)|^2 + \int d\mathbf{r}d\tau[u_0 m(\mathbf{r},\tau)^4 + v_0 m(\mathbf{r},\tau)^6],$$

where $u_0 < 0$, $v_0 > 0$, and $\Gamma_{\mathbf{q}} = v|\mathbf{q}|$. To leading order in the bare parameters r_0, u_0, and v_0, the influence of fluctuations can be incorporated by following a self-consistent renormalization procedure [18] from which one obtains

$$r(T) = r_0 + 12u_0\langle m^2\rangle + 90v_0\langle m^2\rangle^2$$
$$u(T) = u_0 + 15v_0\langle m^2\rangle \qquad v(T) = v_0.$$

Here the averages $\langle\cdots\rangle$ are calculated self-consistently with the renormalized action. To leading order, one need retain only $r(T)$, and perform the calculation with the corresponding quadratic action. Subtracting zero-point fluctuations, one obtains $r(T) = r(0) + 12u(0) \times (\langle m^2\rangle - \langle m^2\rangle_{T=0}) + 90v(0)(\langle m^2\rangle - \langle m^2\rangle_{T=0})^2$, where

$$\langle m^2\rangle - \langle m^2\rangle_{T=0} = \begin{cases} \frac{T^{4/3}\Gamma^{-1/3}}{\pi^2\sqrt{3}} & T \gg r(0) \\ \frac{T^2}{\pi^3\Gamma r(0)} & T \ll r(0) \end{cases},$$

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 $r(T) \propto T^{4/3}$ [17]. 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 $\mathrm{Im}\,\Sigma^R(\mathbf{k},0) \sim T^3/r(T)$ from which one infers a resistivity of $\rho \propto T^{5/3}$ for a conventional quantum critical point and $\rho \propto T^{1/3}$ for the quantum tricritical point.

To conclude, we have shown that the bifurcation structure observed in the magnetic susceptibility of $Sr_3Ru_2O_7$ 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. The bifurcation mechanism described here is quite generic and may provide an opportunity to realize quantum *tricritical* behavior both in $Sr_3Ru_2O_7$ 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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