Magnetic Field–Tuned Quantum Criticality in the Metallic Ruthenate $Sr_3Ru_2O_7$

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The concept of quantum criticality is proving to be central to attempts to understand the physics of strongly correlated electrons. Here, we argue that observations on the itinerant metamagnet $Sr_3Ru_2O_7$ represent good evidence for a new class of quantum critical point, arising when the critical end point terminating a line of first-order transitions is depressed toward zero temperature. This is of interest both in its own right and because of the convenience of having a quantum critical point for which the tuning parameter is the magnetic field. The relationship between the resultant critical fluctuations and novel behavior very near the critical field is discussed.

Understanding the behavior of a dense assembly of interacting, or "correlated," electrons in metallic systems remains one of the outstanding challenges of modern physics. It is a many-body quantum-mechanical problem of such complexity that it cannot be solved by direct, "first principles" calculation (*1*), so it has been tackled through a series of ingenious alternative approaches that have been developed over the past 50 years (*2*). A concept that has attracted considerable attention recently is quantum criticality, which can be explicitly produced by depressing the characteristic temperature of a second-order phase transition toward absolute zero by using some externally applied control parameter such as chemical composition (*3*) or hydrostatic pressure (*4*). The physical properties of the system are then dominated by the critical fluctuations associated with this quantum critical point (QCP) or quantum phase transition (QPT). This concept has generated considerable excitement for several reasons. First, it is now generally acknowledged that these fluctuations can influence the behavior of correlated electron systems even at very high temperatures, up to room temperature and beyond (*5*). Second, because a whole variety of tuning parameters can produce a QCP, many materials of scientific and technological interest may lie close to one in some general parameter space. Third, proximity to a QCP may stabilize novel ground states in itinerant systems. Examples of these include unconventional forms of superconductivity (*4, 6*) and magnetism (*7*).

The concept of the QCP has now been extensively investigated theoretically (*5, 8, 9*), but many open questions remain, particularly in itinerant systems. One of the problems is the restricted range of experiments that have been possible on systems where QCP's are explicitly introduced. If pressure is used as the tuning parameter, a variety of desirable thermodynamic measurements are currently difficult to obtain, whereas the chemical composition is not continuously tunable and also has the significant disadvantage of introducing disorder. Perhaps the most useful tuning parameter at present is a magnetic field. The above restrictions would not apply, and the complete characterization of a QCP and its consequences for an itinerant system would become a realizable goal.

Here, we show that the bilayer ruthenate metal $Sr_3Ru_2O_7$ exhibits behavior consistent with proximity to a metamagnetic (i.e., magnetic field–tuned) quantum critical point. This constitutes both the observation of a new class of QCP (the quantum critical end point) and an opportunity to exploit the experimental precision of field-tuned studies.

Metamagnetism is empirically defined as a very rapid increase of magnetization, M, over a narrow region of applied field, H. In localized systems, where it was first discovered, metamagnetism is the result of "spin-flop" or "spin-flip" spin reorientation processes (*10*); however, it may also occur in itinerant systems, even those which are nonmagnetic at $H = 0$. A natural question is how such behavior could lead to a QCP, because the application of a symmetry-breaking field forbids a second-order magnetic phase transition. A QCP can arise in the following way: In general, metamagnetism is expected to be a first-order phase transition occurring along some line $H = H_c(T)$ in the field and temperature (T) plane. A line of first-order transitions must terminate in a critical end point (H^*,T^*) characterized by diverging susceptibilities. A quantum critical end point would occur if $T^* \rightarrow 0$. One may also imagine a situation in which $T^* < 0$; this would correspond to a rapid (but analytic) crossover in the low-T $M(H)$ curve. This spectrum of behaviors has been observed. There is good evidence for a first-order transition [discontinuity in $M(H)$] in UPd_2Al_3 at low T (*11*) and for a crossover in $CeFe_2Ge_2$ (*12*). The possibility of a metamagnetic criticality has been suggested in relation to observations on materials such as $CeRu_2Si_2$ (*13–15*) and UPt_3 (*15, 16*), but little discussion was given of exactly how it could occur. We believe that the work reported here clearly identifies the existence and consequences of a metamagnetic quantum critical end point. The fundamental difference between this and a QCP produced by depressing a second-order phase transition to absolute zero is the absence of spontaneous symmetry breaking.

The ruthenate $Sr_3Ru_2O_7$ is the $n = 2$ member of the Ruddlesden-Popper series of layered perovskites $Sr_{n+1}Ru_nO_{3n+1}$. The end members of the series, with $n = \infty$ $n = 1$, respectively, are pseudo-cubic $SrRuO_3$ [an itinerant ferromagnet (*17*)] and the strongly two-dimensional Sr_2RuO_4 [a well-known unconventional superconductor (*18*)]. All compounds in the series contain Ru^{4+}, and have Fermi surfaces with strong character of the $4d_{xy}$ and $4d_{xz,yz}$ orbitals of Ru, hybridized with O $2p$ states. $Sr_3Ru_2O_7$ displays metamagnetism for magnetic fields oriented both in the ab plane and along the c axis (*19*). For $B \parallel ab$, the transition occurs between 5 and 6 tesla (T), and clearly has some structure. For $B \parallel c$, it is a single, well-defined transition at approximately 7.8 T. In our previous work, there was evidence that critical fluctuations exist in $Sr_3Ru_2O_7$ for temperatures >1.8 K (*19*). Although interesting, this does not establish the existence of a QCP. The results presented here form a careful study of low-temperature transport for the apparently most definitive field orientation of $B \parallel c$. We show that a "coarse-grained" view of the data reveals all the hallmarks of a QCP at the metamagnetic field H_c. Additionally, detailed investigation gives evidence for a marked change in transport properties in an extremely narrow window of temperature and field around H_c, an effect new to these systems.

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Experiment. The high-quality single crystals on which this work depends were grown in Kyoto and characterized in Birmingham using methods described previously (*19–21*). Resistance ratios $R(300K)/R(4.2K)$ in excess of 100 were achieved. These are approximately a factor of 10 higher than those achieved elsewhere (*22*), and our highest quality crystals show no sign of the ferromagnetic inclusions that are sometimes seen in $Sr_3Ru_2O_7$. The transport experiments were carried out in ^{4}He cryostats and dilution refrigerators in Birmingham and Cambridge using standard four-terminal techniques.

High-temperature resistivity. Although the evidence for low-temperature criticality comes from a number of thermodynamic and transport measurements (*19*), the most precise data have been obtained from magnetotransport. The data are analyzed using the general expression $\rho(T) = \rho_{res} + AT^{\alpha}$, where ρ_{res} is the resistivity due to elastic scattering at $T = 0$, A is a temperature-independent coefficient related to a quasi-particle effective mass, and the exponent α contains valuable information about the nature of the metallic state. One of the key predictions of the Fermi liquid theory of correlated electron metals is that $\alpha = 2$ as $T \rightarrow 0$. A characteristic signature of non-Fermi liquid metals has been the observation of $\alpha < 2$ in this limit (*23*). An indication of the existence of a metamagnetic QCP in $Sr_3Ru_2O_7$ comes from analysis of "high-temperature" data taken between 4.5 and 40 K in a ^{4}He cryostat. In the field/temperature contour plot of α (Fig. 1), $\alpha = 2$ at low fields, falls to a power close to 1 near the metamagnetic field, and then grows again as the field is increased further (*24*).

Low-temperature resistivity. Although these results are suggestive of a metamagnetic QCP, they are not conclusive, as neither a first-order transition/low-temperature critical point nor a crossover can be ruled out. It is not even clear whether $\alpha = 2$ will be recovered on the high-field side of the transition. To address these issues, we have extended the study to the low-temperature region using a dilution refrigerator (Fig. 2). As the field is changed through the metamagnetic transition field, large changes are observed in both ρ_{res} and the temperature-dependent part of ρ. Analysis of the data shows that $\alpha = 2$ at low temperatures. The clearest way to demonstrate this is to fit the data below 350 mK to the form $\rho(T) = \rho_{res} + AT^2$, subtract the value of ρ_{res}, and plot $(\rho - \rho_{res})/T^2$ versus T (Fig. 3). The results contain two of the key signatures expected of a QCP (*9*). First, A (which can be read off as the zero temperature intercept of the flat part of the data at each field) changes by at least a factor of 7 from its low-field value and shows behavior consistent with a divergence at a single field, as can be seen more clearly when plotted explicitly as a function of field (Fig. 4). The second feature is that the rise in A is accompanied by a fall in the characteristic temperature below which the T^2 behavior is observed. This is more difficult to quantify because its definition requires the use of a slightly arbitrary criterion, but the trend can clearly be seen in the data.

These results (Figs. 2 to 4) are very good evidence that the properties of $Sr_3Ru_2O_7$ are controlled by a QCP associated with the metamagnetic transition. They are entirely consistent with the trend suggested by the high-temperature transport (Fig. 1), because $\alpha = 2$ is seen to be recovered at both low and high fields, and $\alpha < 2$ persists to progressively lower temperatures as the QCP is approached. This is perhaps our key experimental result, because it shows that $Sr_3Ru_2O_7$ will give an ideal opportunity to study the effect of a

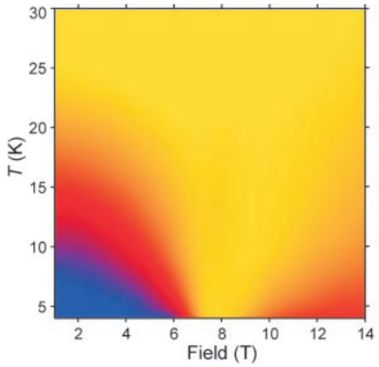

Fig. 1. Summary of the high-temperature resistivity, ρ, near the metamagnetic transition in $Sr_3Ru_2O_7$, for magnetic field applied parallel to the c axis. The plot shows the temperature and field evolution of the exponent, α, derived from the expression $\rho = \rho_{res} + AT^{\alpha}$. At low field, the quadratic temperature dependence expected in a Fermi liquid is seen below 10 K. Near the metamagnetic field of ~7.8 T, a power close to 1 persists down to 4.5 K, before rising again as the field is increased.

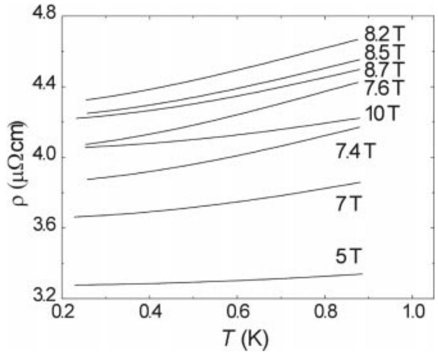

Fig. 2. Sample raw resistivity (ρ) data for $Sr_3Ru_2O_7$ for $B \parallel c$ below 1 K. Passing through the metamagnetic field of ~7.85 T leads to a maximum in the strength of both the elastic and inelastic scattering.

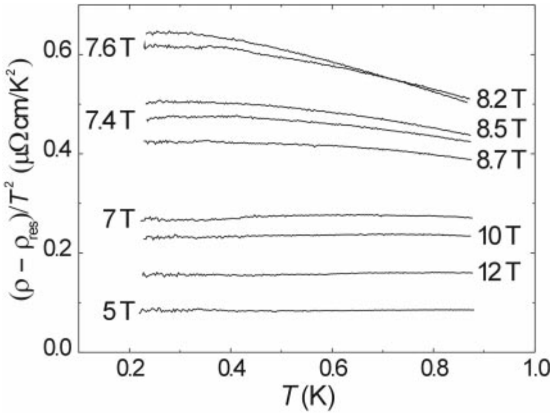

Fig. 3. The inelastic resistivity as a function of temperature in $Sr_3Ru_2O_7$, for a number of applied fields. Passing through the metamagnetic field depresses the crossover to the quadratic behavior expected of a Fermi liquid and leads to a sharp maximum in the strength of the quasi-particle–quasi-particle scattering.

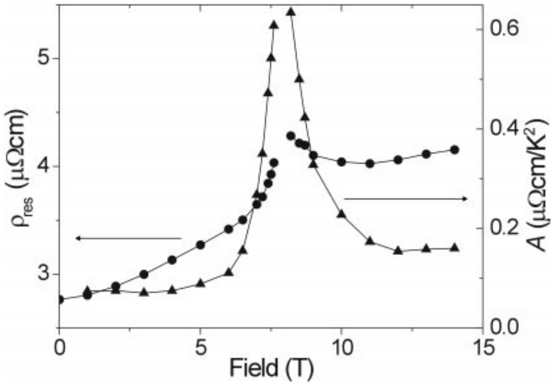

Fig. 4. A summary of the data of Figs. 2 and 3, expressed in terms of the expression $\rho = \rho_{res} + AT^{\alpha}$. The value given for A is from the lowest temperature portion, when $\alpha = 2$. The sharp peaking of A is very strong evidence that when the field is sampled at this resolution, the low-temperature data are governed by the existence of a metamagnetic quantum critical point. Extrapolation of the low- and high-field data gives a critical field of 7.85 ± 0.05 T.

new class of QCP in a clean itinerant electron system. The convenience of magnetic field tuning will open the way to a host of thermodynamic and spectroscopic measurements that cannot easily be performed on other known metallic QCP systems, in which the tuning parameter is pressure or chemical composition (*25*).

Detailed behavior very near the metamagnetic transition. Careful analysis of the transport properties for very closely spaced fields through the metamagnetic transition reveals further interesting behavior (Fig. 5). The data (plotted after the subtraction of ρ_{res}) show three quite distinct characteristic shapes. Those traced in black (taken at fields in the ranges 7.5 to 7.75 T and 7.95 to 8.5 T) have the same basic shape as those in Figs. 2 and 3, ($\alpha = 2$ at low temperatures). Those shown in red, which come from the extremely narrow field region between 7.82 and 7.86 T, have a completely different characteristic shape, which is much flatter at low temperature. The traces shown in blue are from 7.8 and 7.9 T, and seem to delineate the boundary between the "conventional" (black) behavior and the unconventional (red) behavior.

An obvious question to ask is whether the data from the unconventional region display any fixed unusual power law. As shown in Fig. 6, they vary approximately as T^3 for temperatures below about 500 mK. The insets to Fig. 6 show (i) how poor the agreement with T^2 is and (ii) that the crossover to the higher power is very sharp in temperature. To distinguish the notation from that used when the conventional quadratic temperature dependence is observed, we use A_3 to denote the weight of the T^3 term. Extrapolation of this T^3 term was used to extract ρ_{res} for these values of the field. For the intermediate fields of 7.8 and 7.9 T, noninteger α values of approximately 2.7 were observed down to the lowest temperatures. The values of ρ_{res} are plotted against magnetic field in the inset to Fig. 5, color coded in the same way as the inset. They show that the fields at which the unusual temperature dependence is seen in ρ are those at which ρ_{res} peaks, and are hence those closest in field to the metamagnetic transition. The combination of ρ_{res} data shown in Figs. 4 and 5 can be subjected to an independent check. A field sweep at a constant temperature of ~40 mK closely reproduces the field dependence of the extracted values of ρ_{res}, giving us confidence in the quality of our analysis procedure [data not shown here; similar data shown in (*19*)].

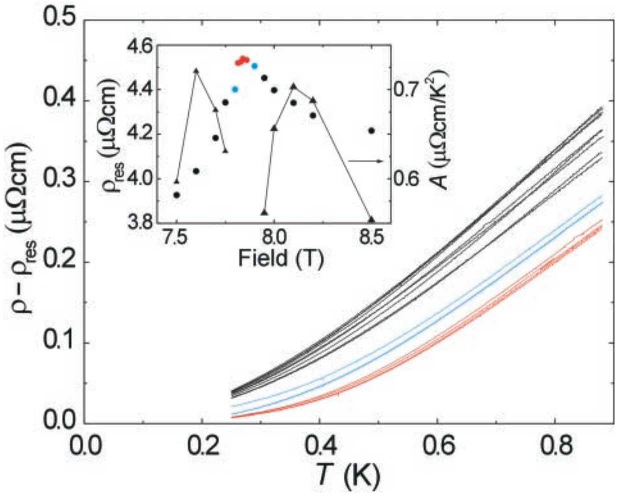

Fig. 5. The inelastic resistivity as a function of temperature in $Sr_3Ru_2O_7$, for applied fields very close to the metamagnetic field. The data shown in black, from 7.5 to 7.75 T and 7.95 to 8.5 T, have the same characteristic shape as those of Figs. 2 and 3, with the expected quadratic temperature dependence at sufficiently low temperature. The data shown in red, from the extremely narrow region between 7.82 and 7.86 T, have a completely different temperature dependence, with much weaker variation at low temperature. Those in blue, for 7.8 and 7.9 T, appear to mark the boundary between the two distinct classes of behavior. (Inset) The field dependence of ρ_{res} extracted from the same field sweeps (circles, color coded as in the main figure) and A (black triangles and guide to the eye) for the fields at which a quadratic temperature dependence is observed.

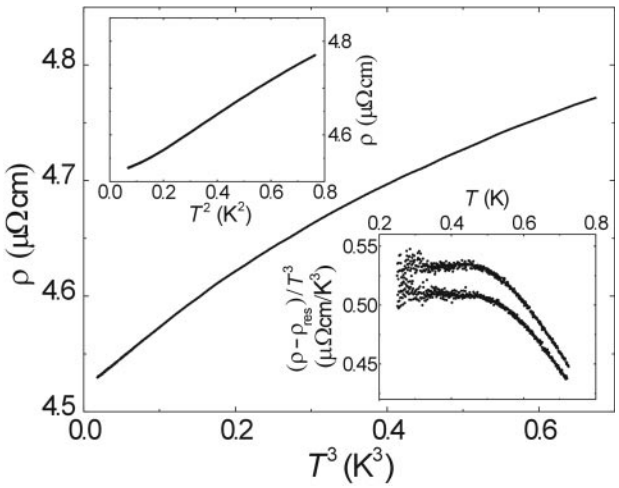

Fig. 6. The resistivity for one of the fields (7.82 T) at which anomalous behavior is shown in Fig. 5. The data show an accurate T^3 variation below ~0.5 K. The degree of deviation from a T^2 dependence is demonstrated by the top inset. The cubic axis on the main figure strongly emphasizes the data above 0.5 K for which almost linear behavior is seen (Fig. 5). For this reason, we include the bottom inset, which shows data from both 7.82 and 7.86 T. It emphasizes the quality of the T^3 variation, that it is essentially the same for the entire range of fields shown in red in Fig. 5, and that the onset of this highly anomalous behavior is very sharp in temperature as well as in field.

Discussion and relationship to other data. The overall picture that emerges from our data can be summarized as follows. If a "coarse-grained" approach is taken, very strong evidence is obtained for the existence of a single metamagnetic QCP with a characteristic field of 7.85 ± 0.05 T (Fig. 4). Although metamagnetic features have been studied at ambient pressure in a number of other materials [e.g., (*11–16, 26, 27*)], the steep, sevenfold increase in the A coefficient is by far the most pronounced such feature seen near a metamagnetic transition. In $CeRu_2Si_2$, for example, an equivalent study revealed at most a factor of 2 change in A (*13*). We believe, therefore, that these observations open the way to the careful study of a new class of QCP, the quantum critical end point.

We now make some remarks about the appropriate theoretical approach in such a situation. This critical end point has Ising symmetry, so the order parameter ϕ has one component, which is the deviation of the long-wavelength magnetization from the value M_c found at the metamagnetic critical end point $H = H_c$, $T = 0$. This order parameter is expected to be governed by an overdamped quantum Ginzburg-Landau model of the type discussed in (*8, 9*), in 2*d* [in light of recent neutron-scattering results (*28*)]. We have analyzed such a model (*29*), and a more complete treatment of the theory will be presented elsewhere. However, the data contain a further remarkable feature which is not predicted by this or any other conventional quantum critical theory, as we now discuss.

The "fine-grained" study (Fig. 5) shows that the apparent divergence of A is cut off in a rather remarkable way. In the sharply defined anomalous region of the (B,T) plane (7.82 T < B < 7.86 T), very unusual transport properties are seen, with $\rho = \rho_{res} + A_3,T^\alpha$, $\alpha \approx 3$. Although we do not have sufficient data to show $\alpha \approx 3$ over a complete decade of temperature, the accuracy with which it is obeyed below 500 mK seems fairly persuasive. A more complete picture comes from further analysis of the "conventional" data of Fig. 5, for which $\alpha = 2$ at low temperature. Extracting the A coefficient using the procedure described above yields the data shown by the black triangles in the inset to Fig. 5. As a precursor to the collapse to the new power, the apparently diverging A (Fig. 4) turns over and begins to fall as the field comes very close to the metamagnetic field.

The quantum critical theory discussed above does not predict this behavior. Indeed, we argue that any continuous transition consistent with our data and scaling yields $\Delta\rho \sim T^x$ with $x < 2$, if the field is tuned to the critical value. We write $\Delta\rho = \rho - \rho_{\mathrm{res}} = T^x R(h/T^y)$, where $R(u)$ is a well-behaved function, $h = (H - H_c)/H_c$ is the reduced field, and $y > 0$ because both h and T are relevant fields which scale the system away from the critical point. In the limit $u \rightarrow 0$, near criticality, a leading order expansion yields $R(u) = R_0 + \mathcal{O}(u)$, and in the opposite limit, the asymptotic behavior of R is characterized by an exponent w, $R(u \rightarrow \infty) \sim u^{-w}$. For $h = 0$, $\Delta\rho \sim R_0 T^x$, so a nondivergent resistivity at this field requires $R_0 < \infty$; $w > 0$ because $\Delta\rho$ decreases away from the QCP. The observed $\Delta\rho \sim T^2$ as $T \rightarrow 0$ and $h \neq 0$ implies $x + yw = 2$; the positivity of y and w then implies $x < 2$, provided $R_0 \neq 0$.

The question of why ρ_{res} peaks is also an interesting one. This feature, common in metamagnetic systems, seems to suggest that the change in the quasi-particle mass is accompanied by a rather pronounced change in the scattering cross-section. An implicit assumption of our analysis throughout this paper is that the elastic and inelastic contributions to the scattering rate are separable. This is at least partially justified by the facts that (i) conventional QCP behavior of A is seen even when the peak in ρ_{res} is fairly well developed (Fig. 4) and (ii) the "double peak" structure in A very near the transition is not seen in ρ_{res} (Fig. 5, inset).

To our knowledge, observation of $\alpha > 2$ at low temperatures near a QCP is unprecedented. The scaling analysis given above confirms the intuitive expectation that such behavior cannot be a simple consequence of critical fluctuations, so we are forced to speculate about other causes. The first that merits careful investigation is that the tuning of the critical point is not perfect at ambient pressure. We would then be observing the consequences of a weakly first-order transition below approximately 0.5 K, perhaps accompanied by a narrow region of mesoscopic electronic phase separation producing the anomalous transport properties. The precise mechanism for these properties is not, however, understood within this picture.

A more intriguing possibility is that we are seeing the signs of a novel form of quantum order (*1*). Clean itinerant systems very near QCP's often adopt ordered states such as unconventional superconductivity at low temperatures (*4, 6, 23*). The unusual aspect of the current situation is that the QCP is dependent on the existence of a large symmetry-breaking magnetic field, whose presence is likely to rule out most of the known "escape routes," opening the way to genuinely novel behavior. We cannot, on the basis of the data presented here, distinguish between the above possibilities or rule out others. A key challenge is likely to be a convincing explanation for $\alpha \approx 3$ in a situation where scattering is clearly dominated by pure electron-electron interactions. Whatever its eventual explanation, this is, we believe, a very interesting observation. The well-known result of $\alpha = 2$ is a consequence of phase space restriction for quasi-particles near a Fermi surface; the current observation clearly indicates that one or more of the central planks of this picture is breaking down.

Conclusion. In summary, we have presented a detailed study and discussion of metamagnetism in the bilayer ruthenate $Sr_3Ru_2O_7$, and have argued that it has the potential to yield new insight into one of the most important current areas of research in correlated electron physics, namely quantum criticality. We have shown how metamagnetism can, in principle, be the source of a quantum critical point, and have presented data that strongly suggest the existence of such a critical point in $Sr_3Ru_2O_7$ at ambient pressure. However, as the field is tuned in very close to the critical metamagnetic field at the lowest temperatures, there is the sudden onset of behavior that cannot be explained as a simple consequence of strong critical fluctuations. One possibility is that because standard ordered states such as unconventional superconductivity are disfavored by the presence of a large magnetic field, the correlated electron system is adopting a novel form of low-temperature order. The convenience of magnetic field as a tuning parameter should lead to future work on $Sr_3Ru_2O_7$ that will improve our understanding of quantum criticality and its consequences.

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23. For a succinct, modern review, see P. Coleman, C. Pépin, Q. Si, R. Ramazashvili, preprint at http://xxx.lanl.gov/abs/cond-mat/0105006.
24. For $B \parallel ab$, $\alpha = 2$ is recovered at high field at temperatures >2.5 K (*19*).
25. A magnetic field has an important effect on pressure- or composition-tuned criticality in several metallic systems {e.g., $CeCu_{5.8}Au_{0.2}$ [H. von Löhneysen, C. Pfleiderer, T. Pietrus. O. Stockert, B. Will, *Phys. Rev. B* **63**, 134411 (2001)]}, but in these cases, the data seem to suggest that the applied field takes the system away from a zero-field critical point. The power of field-tuning was demonstrated in studies of $LiHoF_4$ [D. Bitko, T. F. Rosenbaum, G. Aeppli, *Phys. Rev. Lett.* **77**, 940 (1996)]. This system is an insulator and, in ambient field, a dipolar-coupled Ising ferromagnet; its QPT is of the conventional "Ising model in a transverse field" variety.
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29. Our approach is as follows: The crystal symmetry suggests that the *c*-axis magnetization is conserved even in the presence of spin-orbit scattering, so the dynamics are conservative and overdamped. Recent neutron-scattering experiments (*28*) suggest that interplane spin correlations are negligible. In the conventional formalism (*9*), critical phenomena are expected to be described by the action
$$S = \sum_{\omega} \int \frac{d^2q}{(2\pi)^2} \left[\frac{i|\omega|}{q} + q^2 + r\right] |\Phi_{q,\omega}|^2 + \ldots$$
where the ellipsis denotes nonlinearities irrelevant near the critical point and $r \sim (H - H_c)^{1/3}$ parametrizes the deviation from the critical point at $T = 0$ (the 1/3 is the usual mean-field exponent for field tuning). Standard calculations yield $A \sim (H - H_c)^{-2/3}$ and, at criticality, $\rho \sim T^{4/3}$, not inconsistent with data outside the anomalous region 7.82 T $< H <$ 7.86 T. We also note that Belitz and Kirkpatrick (*30*) have shown that in many 2*d* situations, the term we have written as $q^2\Phi^2$ should be replaced by $|q|\Phi^2$, which could lead to rather different behavior. An important question to be addressed in the future is the relevance of these considerations to $Sr_3Ru_2O_7$.
30. See, for example, T. R. Kirkpatrick, D. Belitz, *J. Stat. Phys.* **87**, 1307 (1997).
31. We acknowledge useful discussions with P. Coleman, R. A. Cowley, E. M. Forgan, S. M. Hayden, C. Pfleiderer, and P. Raychaudhuri. We also thank the Leverhulme Trust, the UK Engineering and Physical Sciences Research Council, and the Royal Society for valuable financial support.

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