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Disorder-Sensitive Phase Formation Linked to Metamagnetic Quantum Criticality

S. A. Grigera,[1*] P. Gegenwart,[1,2] R. A. Borzi,[1] F. Weickert,[2] A. J. Schofield,[3] R. S. Perry,[1,4,5] T. Tayama,[6] T. Sakakibara,[6] Y. Maeno,[4,5] A. G. Green,[1] A. P. Mackenzie[1*]

Condensed systems of strongly interacting electrons are ideal for the study of quantum complexity. It has become possible to promote the formation of new quantum phases by explicitly tuning systems toward special low-temperature quantum critical points. So far, the clearest examples have been appearances of superconductivity near pressure-tuned antiferromagnetic quantum critical points. We present experimental evidence for the formation of a non-superconducting phase in the vicinity of a magnetic field–tuned quantum critical point in ultrapure crystals of the ruthenate metal $Sr_3Ru_2O_7$, and we discuss the possibility that the observed phase is due to a spin-dependent symmetry-breaking Fermi surface distortion.

The field of quantum criticality continues to attract widespread theoretical and experimental attention because of its importance to the global effort to understand the behavior of correlated electron systems (*1–4*). It gives the rare opportunity for controlled study of the collective behavior that arises in many-body quantum physics in the presence of strong interactions. Much of the interest has focused on the effects that quantum critical fluctuations have on itinerant systems, notably the link between their strength and the breakdown of Landau's Fermi liquid theory (*5*). Quantum criticality is now understood not to be a trivial extension of the classical case; quantum critical fluctuations can have surprising strength and subtlety, including important mode-mode interaction terms (*3*, *4*).

Recently several examples have been discovered of a phenomenon that is potentially even more exciting, namely the use of quantum critical points to create regions of phase space in which systems are highly susceptible to the emergence of new ordered phases. Qualitatively, the high susceptibility to novel phase formation is thought to arise because tuning a thermal phase transition toward zero temperature flattens the free energy landscape, causing competing low-temperature phases to become nearly degenerate. The characteristics of the quantum critical fluctuations associated with particular quantum critical points (QCPs) can then tip the balance in favor of a particular new phase. A notable example is the formation of unconventional superconductivity in the vicinity of antiferromagnetic QCPs (*6*). In this case, the diverging spin fluctuations are thought to provide the effective bosons for the Cooper pair binding.

Superconductivity is not the only form of low-temperature order that can be adopted by correlated electron systems; the fractional quantum Hall state is a spectacular example of the subtle quantum self-organization that is possible (*7*). A particularly interesting issue is whether forms of magnetically tuned QCPs can be used to explicitly promote the formation of other new types of quantum order (*8*) in the special situation in which the magnetic field disfavors superconducting ground states. We demonstrate the formation of a previously unknown, non-superconducting phase in an applied magnetic field of 8 T, near a metamagnetic QCP in $Sr_3Ru_2O_7$. The experiments place strong constraints on the order parameter of this phase, which appear to be satisfied by invoking a spin-dependent Fermi-surface instability, the possibility of which was first discussed theoretically nearly half a century ago by Pomeranchuk in phenomenological calculations using Landau's Fermi liquid theory (*9*).

[1]School of Physics and Astronomy, University of St. Andrews, North Haugh, St. Andrews, Fife KY16 9SS, Scotland. [2]Max-Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany. [3]School of Physics and Astronomy, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK. [4]International Innovation Center, Kyoto University, Kyoto 606-8501, Japan. [5]Department of Physics, Kyoto University, Kyoto 606-8502, Japan. [6]Institute of Solid-State Physics, University of Tokyo, Kashiwa, Chiba 2778581, Japan.

*To whom correspondence should be addressed. E-mail: sag2@st-and.ac.uk; apm9@st-and.ac.uk

$Sr_3Ru_2O_7$ is the bilayer member of the Ruddlesden-Popper series of layered perovskite ruthenates whose single-layer member, Sr_2RuO_4, is a well-studied unconventional superconductor (*10*). The growth of high-quality single crystals in an image furnace revealed it to be a strongly enhanced paramagnet in zero field, displaying an itinerant metamagnetic transition in applied fields in the range from 4.9 T (field parallel to *ab* plane) to 7.9 T [field parallel to *c* (*11*, *12*)]. Good evidence has been acquired that, for fields parallel to *ab*, the metamagnetism is due to a line of first-order phase transitions terminating in a finite temperature critical point. As the field is rotated away from the *ab* planes, this critical end point is tuned downward in temperature, becoming quantum critical at an angle less than 10° from *c* (*12–14*). The crystals on which the work of (*12–14*) was performed were of sufficiently high purity (residual resistivity $\rho_{res} \sim 3\ \mu\Omega$ cm) that disorder might not have been expected to play an important role in determining their properties. However, a further improvement of nearly an order of magnitude to $\rho_{res} \sim 0.4\ \mu\Omega$ cm (*15*) revealed substantial changes in the observed behavior. Away from the QCP, the properties are indeed essentially independent of purity, but in its immediate vicinity a large and striking peak was observed in the resistivity, in a region bounded at 100 mK by two first-order magnetic phase transitions (*16*). Those observations opened the possibility of the formation of a non-superconducting phase in the vicinity of the metamagnetic QCP in $Sr_3Ru_2O_7$.

We show the magnetic field (H) dependence (for H // c) of the resistivity of a high-purity single crystal of $Sr_3Ru_2O_7$ as temperature increased from 0.1 to 1.3 K (Fig. 1). At 0.1 K, the large, steep-sided feature is similar to that reported and discussed in (*16*). By 1.3 K, the observed resistivity behavior is unsurprising, with a pronounced thermal broadening. The surprising results come at intermediate temperatures. At all temperatures between 0.1 and 1.1 K, the anticipated broadening is seen outside the field range from 7.8 to 8.1 T; but once that field range is entered, ρ appears to "lock in" to the field and temperature dependence seen at the lowest temperature. The value of ρ at which this occurs is temperature-dependent, but there is almost no temperature dependence to the field at which it occurs or to its field dependence once it has locked in to the anomalous resistivity.

The data of Fig. 1 suggest the presence of a low-temperature phase in which the strong temperature-dependent (inelastic) scattering associated with quantum critical fluctuations is cut off at well-defined boundaries and replaced by a large and essentially temperature-independent (elastic) scattering mechanism. It is not, however, compelling thermodynamic evidence of the kind necessary to support the postulate of a phase change. Such thermodynamic signatures are provided by the susceptibility, magnetostriction, thermal expansion, and magnetization. A sample comparison of alternating current (ac) susceptibility (χ) and linear magnetostriction (λ) is shown (Fig. 2). There is a clear correlation between the real part of χ and λ, indicating a strong magnetostructural coupling. A low-field peak at about 7.5 T, which is probably a crossover, is followed by two sharp peaks at fields corresponding precisely to the steep walls in the resistivity of Fig. 1. As discussed in (*13*, *16*), the appearance of a dissipative peak in χ″ indicates that each of these two peaks corresponds to a first-order phase boundary at 100 mK (*17*). All peaks in both χ′ and λ are positive, showing that both sample moment and length increase at each transition.

Temperature scans of thermodynamic quantities also show well-defined features at the boundaries of the anomalous region seen in the resistivity, for example, the temperature dependence of the magnetization in a fixed field of 7.9 T (Fig. 2, inset). At temperatures above 1.1 K, the data show upward curvature consistent with divergent fluctuations on the approach to a QCP, but this is then cut off at a well-defined kink, followed by a much weaker temperature dependence. The kink is seen only on entry to the anomalous region; scans at fields outside this range remain smooth to the lowest temperature reached (50 mK).

The data shown in Fig. 2 represent only a small subset of the experiments performed. A collation of all the relevant information is presented (Fig. 3), which also includes points

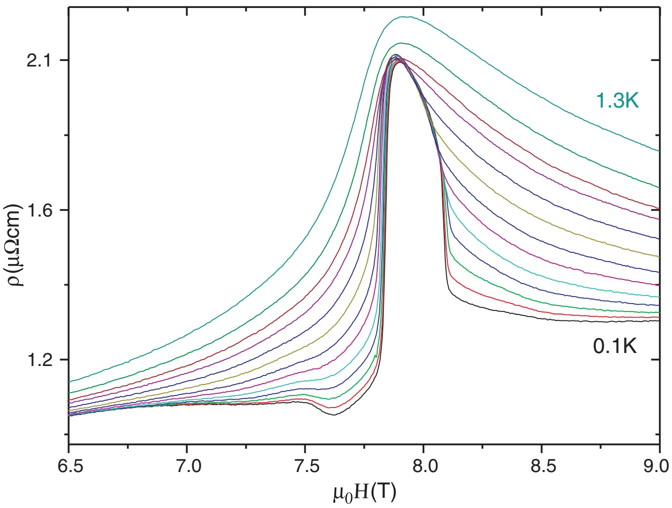

Fig. 1. The resistivity ρ of very pure single-crystal $Sr_3Ru_2O_7$ as a function of magnetic field (applied parallel to *c*) at a series of temperatures between 0.1 and 1.3 K in steps of 100 mK. Increasing the temperature broadens and increases the resistivity outside the field range of the pronounced central feature, but within that field range and below 1.1 K, it locks in to the peak. The value of ρ at which this occurs is strongly temperature-dependent, but both the lock-in field and the variation of ρ through the peak are strikingly independent of temperature.

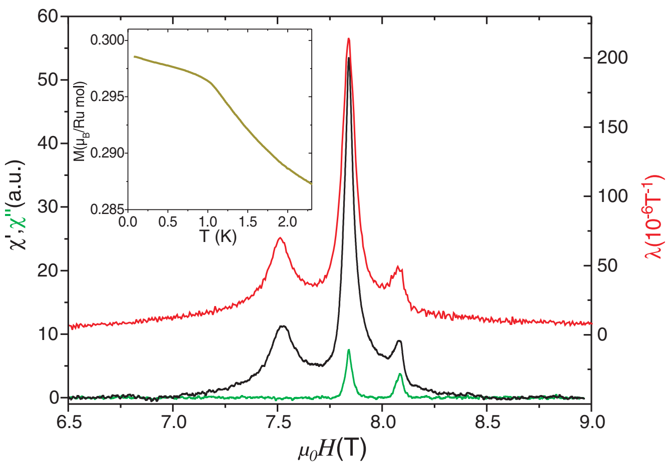

Fig. 2. Sample ac magnetic susceptibility (χ) and linear magnetostriction (λ) data at 100 mK. The susceptibility was measured at a low frequency of 17 Hz to avoid excessive finite-frequency effects (*13*). Both the main field and the small ac field of 3×10^{-5} T root mean square were applied parallel to *c*, and $\lambda = d(\Delta c/c)/dB$. The close similarity between λ and χ′ demonstrates a strong magnetostructural coupling, whereas the peaks in χ″ accompanying the two higher-field peaks in λ and χ′ identify two first-order phase transitions [also (*17*)]. a.u., arbitrary units. (**Inset**) Sample dc magnetization (*M*) data. As the temperature is lowered at a fixed field (in this case 7.9 T), the incipient divergence of fluctuations seen in *M* is cut off at a clearly identifiable kink at the same temperature as that of the top of the peak seen in Fig. 1.

obtained from plotting the positions of peaks in the thermal expansion α. Data obtained from five different crystals of similar purity studied independently in four separate laboratories yield consistent information. Surrounding the temperature and field at which a QCP had been identified in more disordered samples is a region in the (*H*,*T*) plane enclosed by well-defined phase boundaries. At sufficiently low temperatures these are first-order, but they then change to second-order as indicated (Fig. 3, red arrows) (*18*).

The existence of this new phase is the central experimental result of our paper. The anomalous transport properties that accompany it are strongly purity-dependent (*14*, *16*); it can be defined with the clarity shown (Fig. 3) only in the very best samples with $\rho_{res} < 1$ μΩ cm. A strong purity dependence like this is characteristic of some of the best-known ordered phases in itinerant interacting electron systems such as the fractional quantum Hall state (*7*) and unconventional superconductivity (*10*). Several key pieces of experimental evidence further indicate that the previously unknown phase is intrinsically linked with the existence of the previously identified metamagnetic QCP. First, as stated above, it encloses the characteristic field of that QCP as estimated either by direct measurement on more disordered samples or by extrapolation of transport and magnetic measurements taken at temperature $T > 1$ K on the highest purity crystals. Second, we draw attention to the relative orientation of the first-order phase boundaries (Fig. 3). These are seen to have opposite slopes and curvatures with respect to field variation, similar to the quantum critical contours that can be calculated by using a Hertz-Millis model for the underlying QCP (*19*). In contrast, the naïve expectation for two successive first-order metamagnetic transitions unrelated to the QCP would be lines of qualitatively similar slope and curvature, whereas the boundary of a new phase might have been expected to be dome-shaped.

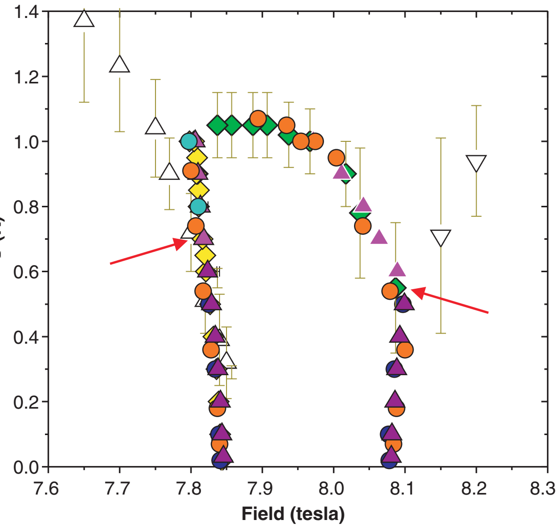

Fig. 3. An empirical phase diagram from collating data from the four measurements contributing to Figs. 1 and 2, combined with the results of thermal expansion measurements. The colored triangles are the loci of peaks in ac susceptibility; the white triangles, the loci of maxima (up) and minima (down); the orange circles are derived from dc magnetization; the yellow diamonds, the loci of maxima in $d\rho/dH$; the green diamonds, the loci of maxima in $d^2\rho/dT^2$; and the blue circles, the loci of maxima in the magnetostriction. The red arrows mark the temperatures of critical points deduced from the temperatures above which no peak in χ'' is observable. We believe that all the data are consistent with the presence of a phase enclosed with first-order phase boundaries for temperatures below these critical points and a second-order line linking them at higher *T*.

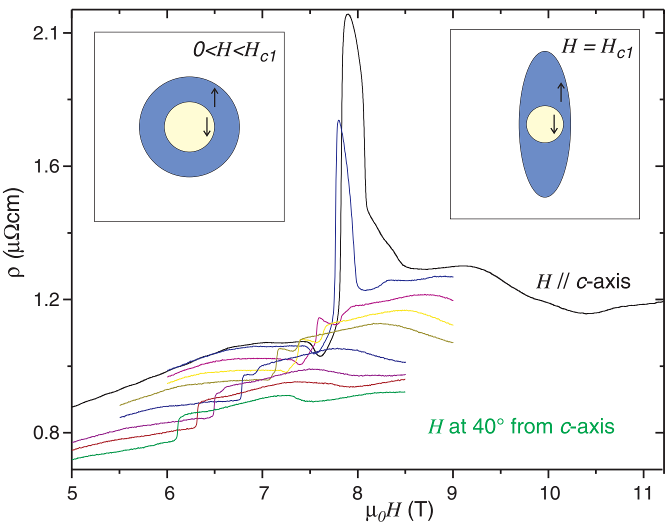

Fig. 4. The resistivity, ρ, at 100 mK as the magnetic field is rotated away from the *c* axis toward the *ab* plane at intervals of 5°. The striking peak that we associate with formation of the phase for *H* // *c* (black trace) exists only over a narrow range of angle. For angles < 80°, features are still seen, but they are much weaker. **(Insets)** Sketches of the kind of spin-dependent Fermi surface distortion that we discuss in the text. They are not meant to be representative of the true Fermi surfaces but are designed to illustrate the kind of symmetry breaking that we discuss. H_{c1} denotes the field (about 7.8 T) at which the symmetry-broken phase is entered. Below this field and above H_{c2} (the field of about 8.1 T at which the phase is left), both the spin-up and spin-down Fermi surfaces have the same symmetry (left inset). Between H_{c1} and H_{c2}, the spin-up Fermi surface distorts to a lower symmetry. This symmetry-breaking distortion could equally well be rotated by 90°, so domain formation is likely.

The third piece of experimental support for a connection with the quantum critical physics of the underlying metamagnetism comes from an initial study of the angular dependence of ρ. Previous work on more disordered samples has shown that rotating the field from the *c* axis toward the *ab* plane raises the characteristic temperature of the underlying critical point, "detuning" the quantum criticality (*13*). It is, therefore, natural to investigate the effect of changing field angle on the formation of the anomalous phase. To do this for all the thermodynamic quantities contributing to Fig. 3 will be a formidable experimental task, but the resistivity data (Fig. 4) show that the anomalous behavior is well bounded in field angle as well as in field and temperature. Each method of tuning is therefore seen to affect the underlying critical point and the appearance of new phase in a qualitatively similar way.

Although itinerant metamagnets have been studied fairly extensively in the past [for example, (*5*, *20*, *21*)], novel phase formation in the vicinity of the metamagnetism has been reported only in URu_2Si_2 (*22*, *23*). The phenomena seen there may have a similar origin to those reported here, but there are important differences in the physics of the two materials. In URu_2Si_2, *f* electrons play an important role, and both superconductivity and a much-studied "hidden order" phase are observed in zero applied field. Also, the phase formation seems relatively insensitive to the presence of disorder, in contrast to the present case. Although we certainly do not rule out relevance to the fascinating physics that has been discovered in URu_2Si_2, the following discussion will have its basis purely in the observations reported here on $Sr_3Ru_2O_7$.

The key facts that any model for the new phase needs to address are (i) the existence of a large enhancement in the absolute value of ρ accompanied by a suppression of its temperature dependence, (ii) an itinerant rather than a localized picture, as evidenced by the observation of dHvA oscillations at both high and low fields (*24*) and the absence of a large change in the Hall coefficient as the anomalous phase is entered (*25*), (iii) a magnetization that increases both on entry to and exit from the new phase as a function of field, (iv) a magnetization whose divergence is cut off as the phase is entered from high temperatures at constant field and whose absolute value shows a kink but no sharp decrease as the phase boundary is crossed, and (v) existence of the phase over only a narrow range of applied field angle.

These facts place some fairly tight constaints on theory. For example, they appear to rule out a partially gapped state such as that recently discussed in relation to the hidden order of URu_2Si_2 (*26*). One scenario, however, seems to offer a plausible explanation for all the observations. In a standard picture of itinerant metamagnetism, the system develops the additional moment by a sudden extra exchange polarization of the Fermi surface, changing the relative volumes of the spin-up and spin-down parts (Fig. 4, left inset). The extra exchange splitting at the metamagnetic transition is driven by the Zeeman splitting of the Fermi surface in an applied field, allowing one or both spin species to access a higher density of states than is available in zero field so that the system satisfies the Stoner criterion. Many-body enhancements also play the role of reducing the energy scale of the important features in the density of states [to ~10 K in $Sr_3Ru_2O_7$ (*11*, *12*)].

A natural assumption is that in **k** space, the metamagnetic exchange splitting will respect the fourfold symmetry of the RuO_2 planes. We postulate, however, that something more unusual can happen in $Sr_3Ru_2O_7$. Its quasi-two-dimensional electronic structure will incorporate fourfold density of states maxima because of van Hove singularities in the electronic structure. Indeed, the importance of the Fermi level lying close to these has been discussed in the context of the standard metamagnetism (*27*). We believe that the main features of our observations would be explained by the onset of a spontaneous, twofold, symmetric Fermi-surface distortion in which the Fermi surface of one spin species elongates along the direction of two of the four available density of states peaks (Fig. 4, right inset). This Fermi surface, which breaks the original fourfold symmetry, persists throughout the anomalous new phase until a second phase transition restores the original symmetry by polarizing equally along the orthogonal direction. This scenario has a number of attractive features. It would naturally account for the facts that the magnetization of the system rises at each transition and that it is not diminished on entering the new phase as a function of temperature at constant field. It involves no partial gapping of the Fermi surface but still gives a way to understand the large resistive peak in terms of domain formation between regions in which the symmetry-breaking distortion is rotated by 90°. Alignment of these domains by the inplane field component would also likely be a factor in the rapid disappearance of the peak with field angle.

Symmetry-breaking Fermi-surface distortions have been discussed theoretically in various contexts since the advent of Landau's Fermi liquid theory and the subsequent work of Pomeranchuk (*9*, *28–33*). However, there have been, to our knowledge, no conclusive observations in real metals. This raises the question of what is so special about $Sr_3Ru_2O_7$. We believe that part of the answer lies in the underlying metamagnetic QCP. As stressed in (*12–14*), the fluctuations associated with an itinerant metamagnetic QCP are rather unusual. They are fluctuations of the Fermi surface itself and so would act to soften the original fourfold Fermi surface, making it easier for it to lower its total energy by adopting a symmetry-breaking distortion. The other important feature of $Sr_3Ru_2O_7$ is the level of purity to which it can be grown. Just like anisotropic superconducting gaps, anisotropic Fermi-surface distortions would be averaged away by sufficiently strong disorder scattering (*34*). The fact that we observe a strong dependence of the new phase on elastic scattering is consistent with this behavior being driven by a Fermi-surface distortion rather than, for example, a structural transition.

We suggest the above scenario as an intuitively appealing possible explanation for the observations that are the core of this paper but do not claim it to be the only way to account for the data. Real-space phase separation or valence transitions are two other possibilities worthy of investigation (*35*). More theoretical and experimental work will be necessary to tell for certain (*36*).

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17. The dissipative peak in χ'' is accompanied both by a peak in the imaginary part in field-modulated resistivity measurements and by hysteresis between up and down sweeps in χ' (*25*).
18. Our identification of the second-order phase boundary in Fig. 3 has its basis in the locus of the kinks in the magnetization coinciding with that of maxima in $d^2\rho/dT^2$.
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28. It is, perhaps, debatable whether the Fermi-surface distortion discussed here should be classed as a Pomeranchuk instability. In Pomeranchuk's original work, the presence of a lattice was not necessary for the many-body distortion to take place. Here, it is likely that the lattice does play a role by modulating the **k** dependence of the density of states and possibly also through the strong magnetostructural coupling demonstrated in Fig. 2. This does not, however, mean that many-body effects are unimportant to the observed behavior. In fact, they are probably crucial to what takes place. First, they renormalize the energy scale of the density of states down to a few kelvin, and secondly they dominate the quantum critical fluctuations that we presume to be vital in softening the undistorted Fermi surfaces. In this sense, we believe that it is appropriate to describe the model that we propose with use of the term "spin-dependent Pomeranchuk instability."
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35. A real-space phase separation giving a coexistence region between 7.8 and 8.1 T cannot be completely ruled out on the basis of our data. However, we believe that it is unlikely. Both hysteretic dissipation (measured by χ'') and hysteresis of the dc magnetization are confined to narrow regions about the first-order phase boundaries identified in Fig. 3. A phase coexistence might be expected to yield maximum dynamic dissipation where the mixture is closest to 50/50, namely in between these two lines.
36. Some experiments that would provide strong supporting evidence of "Pomeranchuck domains" are study of the resistivity under uniaxial inplane pressure anisotropy and study of the inplane resistivity tensor as a function of the polar angle in tilted magnetic fields. Further insight might be gained by using spatially resolved probes such as scanning tunnelling spectroscopy. All these experiments are planned.

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