The behavior of matter near zero temperature continuous phase transitions, or “quantum critical points” is a central topic of study in condensed matter physics. In fermionic systems, fundamental questions remain unanswered: the nature of the quantum critical regime is unclear because of the apparent breakdown of the concept of the quasiparticle, a cornerstone of existing theories of strongly interacting metals. Even less is known experimentally about the formation of ordered phases from such a quantum critical “soup.” Here, we report a study of the specific heat across the phase diagram of the model system Sr$_3$Ru$_2$O$_7$, which features an anomalous phase whose transport properties are consistent with those of an electronic nematic. We show that this phase, which exists at low temperatures in a narrow range of magnetic fields, forms directly from a quantum critical state, and contains more entropy than mean-field calculations predict. Our results suggest that this extra entropy is due to remnant degrees of freedom from the highly entropic state above $T_c$. The associated quantum critical point, which is “concealed” by the nematic phase, separates two Fermi liquids, neither of which has an identifiable spontaneously broken symmetry, but which likely differ in the topology of their Fermi surfaces.

One of the most striking empirical facts about quantum criticality is that, in systems with low disorder, the approach to quantum critical points (QCPs) is often cut off by the formation of new broken symmetry phases. Although this phase formation is widely discussed (1–4), thermodynamic data probing how it occurs are surprisingly sparse. The properties of a low temperature ordered phase are usually linked to those of the metal from which it condenses. Many states form from the background of well understood Fermi liquids, so investigations of the metal are used to gain insight into the properties of the ordered phase. For example, the existence of sharply defined quasiparticles in a simple (Fermi liquid) metal implies the well known “Cooper instability” that leads to the formation of a low temperature superconducting state, and the spectrum of phonons and/or magnetic excitations determines the structure of the gap function in that state. The case of phase formation from a quantum critical background is more challenging and possibly richer, because the metal itself is so mysterious; understanding the thermodynamics of phase formation might yield insight into the quantum critical metal as well as the ordered phase.

In-depth studies of the specific heat are difficult in prototypical quantum critical superconductors such as CeIn$_3$ and CePd$_2$Si$_2$ because of the need to work in pressure cells. Both constructing measurement apparatus suited to the high pressure environment and subtracting the huge addenda background due to the pressure cell are challenging experimental problems that have not yet been fully solved. In this paper we give a concrete example in which the material can be tuned through the quantum critical regime using magnetic field rather than pressure as the tuning parameter, enabling comprehensive measurement of the specific heat.

Interest in careful experimental studies of quantum criticality in metals has been further stimulated by a recent theoretical development that cuts across fields of physics, the so called “AdS-CFT” correspondence, based on dualities between conformal field theories (which presumably describe quantum critical systems) and a higher dimensional quantum gravity. In some cases, the quantum gravity is solvable, even though the conformal field theory is strongly interacting and hence impossible to analyze directly (5). As a result, novel strongly interacting “non-Fermi-liquid” critical theories have been characterized, and correspondingly the range of possible critical behaviors that can be imagined has been expanded. It is presently unclear whether any of the new critical theories apply to any realizable condensed matter system. Thermodynamic data from quantum critical materials are particularly desirable in order to investigate this possibility, and to place experimental constraints on the developing theories.

Here, we report the results of a study of the heat capacity of the field-tuned quantum critical system Sr$_3$Ru$_2$O$_7$, the $n = 2$ member of the Sr$_{n+1}$Ru$_n$O$_{3n+1}$ Ruddlesden-Popper series of layered ruthenates. Sr$_3$Ru$_2$O$_7$ consists of strongly coupled Ru-O bilayers, weakly coupled together to form a material with quasi two-dimensional conduction, based on hybridized Ru-4$d$-O-2$p$ bands (6). The purity of our crystals can be characterized in terms of the residual ($T \rightarrow 0$) resistivity at $H = 0$, which we express in terms of an inferred mean-free path, $\ell$. Samples with $\ell \sim 300$ Å display transport properties consistent with the existence of a field-tuned QCP at $\mu H_c = 7.9$ T and ambient pressure (7, 8). In still purer samples with an order of magnitude larger mean-free path, the approach to this QCP is cut off by the formation of a new phase (9, 10) associated with anisotropic magneto-transport suggesting the presence of nematic electronic order (11, 12). A schematic summary of the main features of Sr$_3$Ru$_2$O$_7$ established by previous work is shown in Fig. 1. Although the QCP is hidden by the new phase, its fluctuations still dominate the broader phase diagram. Sr$_3$Ru$_2$O$_7$ is therefore an ideal system in which to study the thermodynamics of phase formation against a background of quantum criticality.
Ru-mol

bution and a Fermi liquid constant term of accuracy with the sum of a Debye model for the phonon contribution. Above a decade of temperature, but the data at higher temperatures are equally striking. For over 10% of the fields studied demonstrates this depression and reemergence of $T^*$. (Fig. 3C).

Further indications that the depression of a single energy scale is at the root of the quantum critical behavior come from plotting the temperature dependence of the entropy at fields across the quantum critical region (Fig. 3D—details are given in the SI Text: SI1). By 15 K, just over 10% of $R ln 2$ entropy per Ru is recovered at each of the applied fields. Interpretations of this behavior will be discussed below, but independent of these, the data in Figs. 2 and 3 provide compelling evidence that the $H_\perp T$ phase diagram of Sr$_2$Ru$_2$O$_7$ is determined by the physics of quantum criticality. As discussed below, features similar to those reported here have been observed in other quantum critical systems, although in those cases they have typically been associated with Kondo lattice physics which is not directly applicable to Sr$_2$Ru$_2$O$_7$. The fact that this quantum critical behavior is observed with such clarity in this d-electron metal is one of our main experimental findings. The value of $T^*(H)$ inferred from the specific heat maximum correlates with the onset of a quadratic scattering rate in the resistivity for $T < T^*(H)$, as shown in the SI Text: SI2, as well as with a peak in the thermal expansion at $T < T^*(H)$ for $H$ close to $H_c$ (14), so it is reasonable to think of $T^*(H)$ as the scale of the renormalized Fermi temperature. By $H = H_c \pm 0.1$ T (the field width of the nematic phase) $T^*$ has fallen to below 1 K, so the transition into the phase at 1.2 K occurs not from a Fermi liquid but directly from a metallic state dominated by quantum critical fluctuations.

In Fig. 4A we show data for $C_{el}/T$ below 1.4 K at eight fields, four outside the ordered phase and four cooling down into it. Outside the phase, $C_{el}/T$ either falls slightly with decreasing $T$ (if the peak of Fig. 3B is at a low enough temperature that its falling edge still affects the data) or is approximately constant, as expected for a fully formed Fermi liquid. At the fields cooling into the phase, qualitatively different behavior is seen. There is a step centered on approximately 1.2 K, followed by a $C_{el}/T$ that rises, approximately linearly, as the temperature drops further. The data are precise; as seen on the expanded scale of Fig. 4B, the midpoint of the step moves systematically down in temperature by approximately 60 mK as the field is increased from 7.9 T to 7.975 T. This shift is in excellent accord with the detailed shape of the phase diagram obtained previously using transport, thermal expansion and susceptibility (10). Here the most important point empirically is that the linear rise in $C_{el}/T$ is observed only above this temperature there is increasing noise due to the range of integration combined with a weak field dependence consistent with the known temperature dependence of the magnetization (see SI Text: SI3).

As the field is increased, the hump sharpens and $T^*$ moves towards $T = 0$ as $H$ approaches $H_c$, before beginning to grow again for fields higher than $H_c$. Plotting the position of the maximum for each of the fields studied demonstrates this depression and reemergence of $T^*$ (Fig. 3C).

Results

In Fig. 2 we show the in-plane resistance ($\rho$) and electronic specific heat ($C_{el}/T$) for Sr$_2$Ru$_2$O$_7$ cooling from 18 K to 250 mK at 7.9 T in a single crystal with $\xi = ~3,000$ Å. Entry into the ordered phase at $T_c = 1.2$ K is marked by a kink in $\rho$ and a step in $C_{el}/T$, but the data at higher temperatures are equally striking. Over a decade of temperature, $\rho$ is nearly perfectly linear in $T$, with $C_{el}/T$ varying as $\ln T$ over the same range. Somewhat similar behavior has been observed in association with quantum criticality in a variety of other materials. In the present case these functional dependences are obeyed with high accuracy all the way down to $T_c$.

Before describing the phase formation in more depth, we turn our attention to the thermodynamic signatures of the hidden quantum critical point. Fig. 3A shows the evolution of the total specific heat below 40 K, in both zero applied field and at 7.9 T. Above 20 K, $C/T$ is field-independent, and can be fitted to high accuracy with the sum of a Debye model for the phonon contribution and a Fermi liquid constant term of $C/T = 70$ mJ/Ru-mol K$^2$. Below 20 K, additional heat capacity is seen at both fields. The logarithmically diverging term highlighted in Fig. 2 dominates the data at 7.9 T, while a broad hump appears in zero applied field. The specific heat data of Fig. 3A suggest that the quantum critical states might be formed from the depression towards $T = 0$ of a low energy scale, $\delta_T$, identified with the hump seen in zero field. As shown in Fig. 3B, this energy scale depression is what occurs.
within the ordered phase and never outside it. Linear terms in $C_d/T$ have been seen in strongly mass-renormalized Fermi liquids at low temperatures (20, 21), and it is known that they can arise due to nonanalytic terms in the Fermi liquid expansion (22). For that physics to be the source of our observations, however, the Fermi liquid parameters of the metals in and outside the ordered phase would have to differ strongly, something for which there appears to be little evidence (16).

Discussion
The data presented in Figs. 2–4 present a number of challenges to our understanding both of quantum criticality and of phase formation from a quantum critical background. The specific heat and entropy data of Fig. 3 B and D are similar to observations in classic heavy fermion compounds (23, 24) and in $f$-electron systems, the local $f$-moments provide the entropy ($R$ln2 per spin) of classical two-state fluctuators at high temperatures. As the temperature is lowered, these spins are incorporated into the Fermi sea via the Kondo effect, after which the entropy drops linearly with $T$ at sufficiently low temperatures as implied by the Pauli principle restrictions of the Fermi liquid and the third law.

The temperature dependence of the specific heat (after subtraction of the phonon background) as the field is tuned through the quantum critical region. The hump seen in zero field sharpens and is depressed to progressively lower temperatures as the critical field is approached before reappearing on the high field side of the transition. The temperature at which the maximum occurs at each measured field is shown in box (C) (blue dots). At 7.9 T no maximum is seen and based on previous data from more disordered samples it is assumed to occur at $T = 0$ (red dot). In box (D) we show the evolution of the entropy at each of the fields for which data are displayed in (B).

As illustrated in Fig. 4, phase formation in Sr$_2$Ru$_3$O$_7$ also has unusual properties. Some insight can be obtained into the observed behavior by considering two different ways in which ordering affects the electronic structure of a system at mean-field level. One is by opening an energy gap on much or all of the Fermi surface. In this case, minimization of $F = U - TS$ is achieved by making a large saving in $U$ that outweighs the costs of reducing the entropy below $T_c$. However, not all order involves gapping, and one would expect that in such situations, the minimization of $F$ would be achieved without as strong a reduction of $S$ below $T_c$.

As an illustration, we have compared the measured temperature dependence of the entropy saving $\Delta S = S_{\text{ord}} - S_{\text{norm}}$ with the same quantity calculated in two simple mean-field models. (Here $S_{\text{ord}}$ is the measured entropy in the ordered phase, and $S_{\text{norm}}$ is the normal state entropy from above $T_c$ extrapolated into the ordered phase.) To represent the behavior of an ordered state which gaps the Fermi surface, we have computed $\Delta S$ in the mean-field description of the transition to an s-wave superconducting state produced by a weak, local attraction between electrons. As a model of a gapless ordered state, we considered the mean-field description of a Pomeranchuk transition to a nematic state with a distorted Fermi surface under circumstances in which the spin-up Fermi surface intersects the van Hove point. Indeed, in both models we have considered fine-tuned conditions in which a portion of the Fermi surface passes through a van Hove singularity so that the normal state $S/T$ is logarithmically divergent as $T \to 0$, as it is in the experiment. (See SI Text, SI4 for the explicit details of the models). From Fig. 4C, one can see that in the fully gapped system (red), $\Delta S(T)$ has a deep minimum at $T = T_c/2$ where $\Delta S/S(T_c) = -0.31$. By contrast, in the nematic model (blue), although the entropy still dips below $T_c$, the dip is much shallower, with a minimum of only $\Delta S/S(T_c) = -0.14$ at $T \approx 0.5 T_c$.

The fact that this entropy is 10% of $RN_\mu$ rather than the full value is not of crucial importance to the arguments given in the text, because not all states in the Brillouin zone need to be contributing to the specific heat peak.
The similarity of the entropy data (black curve in Fig. 4C) to the model calculation for order without gapping strongly suggests that no gap opens at $T_c$ in Sr$_3$Ru$_2$O$_7$. However, the observed $\Delta S$ minimum is even shallower than that in the Pomeranchuk model. The real system has more entropy in the low temperature phase than this calculation or other mean-field calculations predict. Although we cannot definitively rule out other sources for this entropy, the experiments reported here suggest that it is due to remnant degrees of freedom from the highly entropic state above $T_c$. The combination of these extra degrees of freedom and the logarithmically diverging normal state $C/T$ makes the low temperature entropy in the nematic phase higher than that of the adjoining Fermi liquids (19).

Combined with our previous work on the field dependence of the entropy at low temperatures, the results of this paper provide a comprehensive thermodynamic characterization of a model quantum critical material supporting the formation of an unusual low temperature phase. A point worth emphasizing is that our data do not appear to be compatible with a violation of the third law of thermodynamics such as that predicted in some AdS-CFT theories (35). Entropy is balanced within experimental error at $T_c$ if $C/T$ both above and below $T_c$ are extrapolated to $T = 0$ (19). This balance implies that $S \to 0$ as $T \to 0$ within the nematic phase itself (Fig 3C). Moreover, the low temperature entropy of the hidden quantum critical point is associated with a relatively weak logarithmic divergence of $C/T$ that would not lead to a third law violation, even if it were not “hidden” by the nematic phase.

Although this paper is primarily concerned with experiment, we close by outlining a possible theoretical framework for future calculation. Previous proposals (30–32, 36, 37) for the origin of the nematic phase in Sr$_3$Ru$_2$O$_7$ have involved band electrons close to a van Hove singularity (vHs). As the magnetic field is increased, Zeeman splitting brings one spin species closer to the van Hove point, where even weak residual interactions are sufficient to produce broken symmetry phases such as nematic order, enabling the vHs to be effectively avoided. The underlying quantum critical point is thus associated with a change in the Fermi surface topology, rather than with a quantum transition between a broken symmetry phase and a disordered phase. Such models provide a natural source for the energy scale $\kappa_B T(\langle H \rangle)$, which is approximately the distance in energy between the Fermi level and the vHs for this spin species. In the SI Text, we analyze a specific model based on this simple picture and show that it captures some gross features of the behavior of Sr$_3$Ru$_2$O$_7$, while others are not compatible with a simple rigid band Zeeman shift (38, 39).

In summary, we have reported experiments addressing a problem of significance to condensed matter physics and beyond, namely the formation of a quantum critical state in a clean itinerant system and the phenomenology of ordered phase formation directly from such a state. The data point to a mechanism for the formation of heavy Fermi liquids that is more general than the spin-Kondo physics of conventional $f$-electron heavy fermion compounds, and give evidence for unconventional extra degrees of freedom in a phase formed from a quantum critical “normal state.”

Materials and Methods

Transport measurements were performed using standard four-terminal methods at measurement frequencies below 100 Hz in a bespoke adiabatic demagnetization cryostat that enabled continuous temperature sweeps from 100 mK to 20 K. Specific heat was studied above 600 mK in a commercial instrument (Quantum Design PPMS) equipped with a $^3$He cryostat, and between 250 mK and 1.4 K in bespoke apparatus mounted on a dilution refrigerator.

In order to establish the field-independent contribution to the specific heat, we analyzed the data above 20 K. We fitted it with a function of the form

$$ C / T = \gamma + AT^2 \int_0^{T / T_c} \left( \frac{x^4}{e^x - 1} \right) dx $$

with $\gamma$, $A$, and $T_c$ being free fit parameters and $T$ temperature. The first term ($\gamma$) represents a Fermi liquid contribution and the second term is a modified Debye model describing the phonon background. In order to allow for the more complicated phonon density of states of a realistic material we relaxed...
the condition that the prefactor $\alpha$ is fixed by the Debye temperature $T_D$ and allowed them to vary independently.

ACKNOWLEDGMENTS. We acknowledge useful discussions with E. Fradkin, D.-H. Lee, C.A. Hooley, and A.M. Berridge. A.P.M. thanks the Department of Physics at Stanford University for their hospitality during a Sabbatical visit in which this manuscript was written, and the receipt of a Royal Society-Wolfson Research Merit Award. The work was supported by the United Kingdom Engineering and Physical Sciences Research Council and Royal Society, and the Department of Energy Grant # DE-FG02-06ER46287.