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Transport and thermodynamic properties of $Sr_3Ru_2O_7$ near the quantum critical point

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The specific heat and electrical resistivity of $Sr_3Ru_2O_7$ single crystals are measured in several magnetic fields applied along the c axis for temperatures below 2 K and at fields up to 17 T. Near the critical metamagnetic field at $B_1^* \sim 7.8$ T, the electronic specific heat divided by temperature increases logarithmically as the temperature decreases, over a large range of T, before saturating below a certain T^* (which is sample dependent), indicating a crossover from a non-Fermi liquid (NFL) region dominated by quantum critical fluctuations to a Fermi liquid (FL) region. This crossover from a NFL to a FL state is also observed in the resistivity data near the critical metamagnetic field for $I \| c$ and $B \| c$. The coefficient of electronic specific heat, γ, plotted as a function of field shows two peaks, consistent with the two metamagnetic transitions observed in magnetization and magnetic torque measurements. At the lowest temperatures, a Schottky-like upturn with decreasing temperature is observed. The coefficient of the Schottky anomaly exhibits a field dependence similar to that of γ, implying an influence by the electrons near the Fermi surface on the Schottky level splitting.

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INTRODUCTION

The $4d$-transition metal layered oxides, especially the layered strontium-ruthenium oxides with perovskite-like structure, have been a focus of intensive research interest since the discovery of superconductivity in single layer Sr_2RuO_4,[1] which appears to possess p-wave spin-triplet pairing,[2] possibly of magnetic origin.[3–5] Studies of the magnetic properties of Sr_2RuO_4 and related compounds may shed light on the mechanism leading to the p-wave pairing in Sr_2RuO_4. Pseudocubic, infinite layer $SrRuO_3$ exhibits a ferromagnetic transition at 165 K.[6] Bilayer $Sr_3Ru_2O_7$ has intermediate dimensionality between the single layer Sr_2RuO_4 and infinite layer $SrRuO_3$. The temperature dependence of the magnetic susceptibility, $\chi(T)$, measured on single crystal $Sr_3Ru_2O_7$ exhibits nearly isotropic behavior and shows no hysteresis between zero-field-cooling (ZFC) and field-cooling (FC) data.[7] A maximum is seen around $T_M = 1$ K in $\chi(T)$ for field applied both parallel and perpendicular to the c axis. A Curie-Weiss fit to the susceptibility in the range 200 K$<T<$320 K, a paramagnetic region well above T_M, returns a negative Curie Weiss temperature, $\Theta_W = -45$ K (-39 K) for $B \| c$ ($B \perp c$), suggesting antiferromagnetic (AFM) correlations.[7] However, no evidence of either ferromagnetic or antiferromagnetic long-range order was observed by neutron scattering for temperatures down to 1.6 K.[8] A metamagnetic transition at $B_1^* = 5.5$ T (7.7 T) was reported in the ab plane (c axis) likely due to a rapid change from a paramagnetic state at low fields to a more highly polarized state at higher fields via a first order phase transition.[9,10] Recently, a second metamagnetic transition was observed by Ohmichi *et al.*[11] at a slightly higher field than B_1^* in magnetization and magnetic torque measurements.

Among the variety of interesting features that $Sr_3Ru_2O_7$ has demonstrated, the magnetic field tuned quantum critical point (QCP) is most intriguing.[9,10,12,13] Tuning the system through a QCP can, in general, be achieved experimentally by applying pressure,[14] varying doping level,[15] or applying a magnetic field. Field-tuning a QCP is a particularly convenient way of approaching the QCP, where a transition, in principle, occurs at absolute zero. Near a QCP, thermal fluctuations are no longer relevant and quantum mechanical fluctuations, determined by Heisenberg's uncertainty principle, become dominant.[16] Critical fluctuations associated with the metamagnetic transition near B_1^* for $B \| c$, have been observed in both transport and thermodynamic properties.[9,12] Magnetotransport data taken between 4.5 and 40 K have shown the quadratic temperature dependence expected in a Fermi liquid (FL) in fields far away from B_1^*.[9,12] However, a nearly linear temperature dependence was reported near the metamagnetic field of $B_1^* \sim 7.8$ T, indicating non-Fermi-liquid (NFL) behavior associated with critical fluctuations.[12] Near $B_1^* \sim 7.7$ T, Perry *et al.*[9] suggest that the Sommerfeld coefficient, $\gamma(T) = C/T$, increases logarithmically with decreasing temperature from approximately 16 to 2 K, giving thermodynamic support for the critical fluctuations suggested by the transport measurements. Since the presence of a symmetry breaking magnetic field prevents a second-order transition which requires a symmetry change, the metamagnetic phase transition is usually first order. The possibility of having a $T\rightarrow 0$ critical end-point of a line of first-order metamagnetic phase transition has been widely discussed. Millis *et al.* presented a renormalization group treatment of metamagnetic quantum criticality in metals.[17] Their detailed results are shown to be in quantitative agreement with the magnetization, transport and specific heat data on $Sr_3Ru_2O_7$ except very close to the critical point itself. Grigera *et al.* reported a study of angular dependence of the differential magnetic susceptibility, demonstrating continuous tuning of a line of end points terminating in a surface of first-order metamagnetic phase transitions.[10] In this article, we present specific heat and c-axis resistivity data taken for temperatures in the range $0.07\,\mathrm{K}<T<2\,\mathrm{K}$ and $0.02\,\mathrm{K}<T<0.8\,\mathrm{K}$, respectively, in various magnetic fields applied along the c axis of $Sr_3Ru_2O_7$. The data was collected at the National High Magnetic Field Laboratory facilities in Tallahassee, FL, and Los Alamos, NM.

EXPERIMENT

Both flux grown and floating zone (FZ) grown single crystals were used in this study. To make the flux grown samples, off-stoichiometric quantities of RuO_2, $SrCO_3$ and $SrCl_2$ (as self-flux) were mixed and heated up to 1500 °C in partially capped Pt crucibles, soaked for 20 h, cooled at 2 °C/h to 1350 °C, and then rapidly cooled to room temperature. The FZ crystals were grown in an image furnace in Princeton, NJ.[7,18] No impurity phases, such as $SrRuO_3$, were detected in powder x-ray-diffraction measurements in either the flux grown or the FZ grown samples. The temperature dependence of the magnetic susceptibility $\chi(T)\equiv M/B$ from 2 K to 350 K was measured on both the flux grown and the FZ grown samples using a Quantum Designs DC SQUID magnetometer. Samples grown with either method show a maximum in $\chi(T)$ for both $B\|ab$ and $B\|c$ at a characteristic temperature $T_M=16$ K. No trace of $SrRuO_3$ was observed in the flux grown samples, while a barely discernable kink was seen in $\chi(T)$ near 150 K in the FZ grown samples corresponding to an impurity phase of a very small amount of $SrRuO_3$ based on the change of magnetization across the kink.[19] Resistance ratios $R(300\,\mathrm{K})/R(4.2\,\mathrm{K})$ of the order 100 and 40 were achieved in the FZ grown samples and the flux grown samples, respectively, for current flow in the ab plane, with residual resistivities of 1.5 $\mu\Omega$ cm and 2 $\mu\Omega$ cm, respectively. For current flow along the c axis, the residual resistivities are approximately three orders of magnitude larger. The dimensions of the FZ crystals employed in the specific heat and resistivity measurements were $2.5\times 2\times 0.4\,\mathrm{mm}^3$ and $1.3\times 1.3\times 0.5\,\mathrm{mm}^3$, respectively, and the corresponding size for the flux grown crystal utilized in the specific heat measurement was $1.5\times 0.8\times 0.2\,\mathrm{mm}^3$.

RESULTS AND DISCUSSIONS

Figure 1(a) shows the specific heat data below 2 K taken on the FZ grown sample for $B\|c$. The data could be smoothly connected with the data reported by Perry *et al.*[9] after the subtraction of the T^3 lattice contribution considering a Debye temperature of 360 K, demonstrating excellent con-

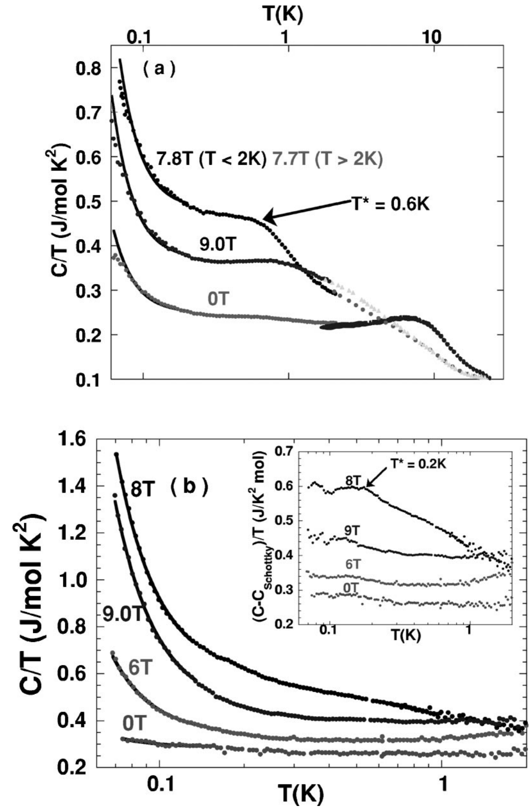

FIG. 1. (a) Specific heat divided by temperature, $C(T)/T$, measured on the FZ grown $Sr_3Ru_2O_7$ single crystal in magnetic fields for $B\|c$. Phonons are subtracted using $\Theta_D=360$ K. Solid lines are fits to the high temperature limit of a Schottky anomaly in the form of $C/T=\gamma+DT^{-3}$ for $0.07\,\mathrm{K}<T<0.20\,\mathrm{K}$. (b) $C(T)/T$, measured on the flux grown $Sr_3Ru_2O_7$ single crystal in magnetic fields for $B\|c$. Solid lines are fits to the high temperature limit of a Schottky anomaly in the form of $C/T=\gamma+DT^{-3}$ for $0.07\,\mathrm{K}<T<0.13\,\mathrm{K}$. Inset: Sommerfeld coefficient after the subtraction of the Schottky term, $\gamma(T)=[C(T)-C(T)_{\mathrm{Schottky}}]/T$.

sistency between the results obtained from different samples. Specific heat data were taken at several fields up to 17 T, but for clarity only three are shown. The data for C/T taken at the critical metamagnetic field as determined by magnetization and magnetotransport measurements, $B_1^*=7.8$ T, follow a logarithmic temperature dependence down to 0.6 K, where there is a crossover to a flat region. This flat region of C/T, where the electronic specific heat is linear in temperature, can be attributed to FL behavior.

A sharp upturn in C/T is observed below 0.2 K, when the temperature is further decreased. The data below 0.2 K can be fit to the high-temperature tail of a Schottky anomaly, $C/T=\gamma+DT^{-3}$, for all fields. The solid lines in Fig. 1(a) are the Schottky anomaly fits. If the Schottky term based on the fit is subtracted, then the data are flat down to the lowest temperature measured. The electronic specific heat coefficient, γ, and the Schottky coefficient, D, were extracted from the fits. Plotted as a function of field in Fig. 2(a), both γ and D show two peaks coinciding with the two metamagnetic transitions observed by Ohmichi *et al.*[11]

Since the large Schottky anomaly tails occur at rather low

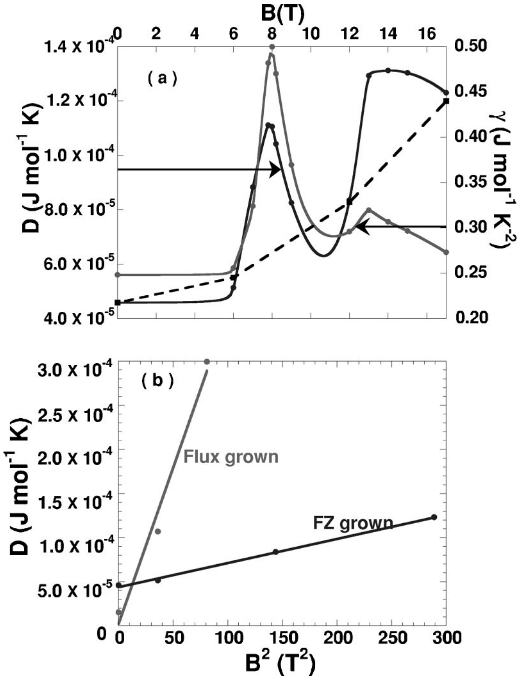

FIG. 2. (a) γ and D extracted from the Schottky anomaly fits. The dashed line shows the calculated hyperfine contribution to the Schottky term due to the applied field. (b) D versus B^2 for the noncritical regions, the fitting line for the FZ grown sample having a larger intercept but smaller slope.

temperatures, they are likely due to the quadrupolar and magnetic spin splitting of Ru and Sr nuclei.[20,21] The hyperfine and quadrupolar contributions of Ru^{99}, Ru^{101}, and Sr^{87} were calculated separately by assuming that the hyperfine splitting in the noncritical regions is mainly caused by the applied field and the quadrupolar contribution is field independent. To obtain the contribution due to the hyperfine splitting we used the effective magnetic moments of the nuclei and their natural abundances. The field dependence of the theoretical value for D is shown in Fig. 2(a) as a dashed line. It agrees well with the experimental data at 6, 12, and 17 T. Field gradient calculations based on a point charge model and the crystal structure given by Huang *et al.*[8] show that the field gradients at the Ru and Sr sites are both along the c axis and have comparable magnitude. From Mössbauer studies of various ruthenates,[22] an upper limit on the field gradient of the Ru sites can be determined to be 130×10^{20} V/m^2. Assuming that the field gradient is the same for Ru and Sr sites, and taking the upper limit value, the calculated quadrupolar contribution (using the quadrupolar moments of the isotopes and their natural abundances) to the Schottky coefficient is 5.65×10^{-6} J mol^{-1} K. This value of D is significantly smaller than the Schottky coefficient, $D=4.58\times10^{-5}$ J mol^{-1} K obtained from the fit to the Schottky anomaly in the form of $C/T=\gamma+DT^{-3}$ for $0.07\ \mathrm{K}<T<0.20\ \mathrm{K}$ and $B=0$ T. This indicates that the quadrupolar contribution itself is probably not large enough to account for the zero-field Schottky anomaly. In Sr_2RuO_4, Langhammer *et al.* also observed a similar low-temperature upturn that could be fit to the high-temperature tail of a Schottky contribution.[23] Again, the reported Schottky term in Sr_2RuO_4 is one to two orders of magnitude larger than the calculated quadrupolar contribution, and it is unlikely to be caused by hyperfine splitting, which would require an unusually large internal magnetic field. Moreover, this term in Sr_2RuO_4 is highly sample dependent.[23]

Specific heat measurements conducted on flux grown samples show quantitatively similar behavior as observed in the FZ grown samples. Figure 1(b) shows C/T of the flux grown $Sr_3Ru_2O_7$ sample versus temperature for several magnetic fields between 0 T and 9 T applied along the c axis. In the inset of Fig. 1(b), the large low temperature Schottky anomaly tails were subtracted. At high magnetic field and low temperature the Schottky term accounts for most of the specific heat measured, and its subtraction results in an increased scatter of the data points. For this reason we did not show C/T with the subtracted Schottky anomaly from the specific heat data on the FZ grown sample displayed in Fig. 1(a). In both samples, near the critical field, $\gamma(T,B)$ shows a logarithmic dependence until a temperature T^*, below which γ saturates to a constant value, indicating a crossover from a NFL region dominated by quantum critical fluctuations to a FL region. This behavior is also seen in other systems that have a QCP, such as $CeCoIn_5$[24] and $YbRh_2Si_2$.[25] γ and D, as functions of field, follow a similar general trend to 9 T, the maximum field applied on the flux grown sample, for both samples. At each field, the two samples have comparable γ values. As shown in Fig. 2(b), for both samples, D versus B^2 can be fit to a straight line if the data points near the critical regions are excluded, however, both the slope and the intercept of the fits on the two samples differ drastically. Differences in the abundances of Ru and Sr isotopes alone are too small to account for such a huge sample dependency.

In order to resolve the discrepancy between the calculated quadrupolar contribution and the zero field Schottky term as well as the two peaks in $D(B)$ which appear to be modulated by γ (at least up to 12 T), we propose that there is another Schottky term, which is electronic in nature, in addition to the quadrupolar and hyperfine contributions. There are several possible sources for such a term. It could arise from the nuclear Knight shift or additional electronic contributions to the electric field gradient at the Ru and Sr nuclei. A more remote possibility is that it is caused by the removal of the degeneracy in the t_{2g} states of Ru. In all cases the term would be roughly modulated by the density of states near the Fermi surface and hence proportional to γ. Factors such as strain in the sample could also influence the nuclear Knight shift and the electric field gradient as a function of magnetic field, causing the discrepancies in the data taken on the two samples.

The crossover from the NFL to the FL region is also exhibited in the resistivity near the critical metamagnetic field. Figure 3 shows the resistivity, ρ, of a FZ grown sample taken from the same growth rod as the specific heat sample for $0.02\ \mathrm{K}<T<0.80\ \mathrm{K}$ and $7.75\ \mathrm{T}<B<8.05\ \mathrm{T}$, with both the current and field along the c axis. The low temperature resistivity for all fields near the critical metamagnetic field can be well fit to $\Delta\rho\equiv\rho(T)-\rho(T=0)=AT^2$, the expected dependence characteristic of a FL. For T above a certain tem-

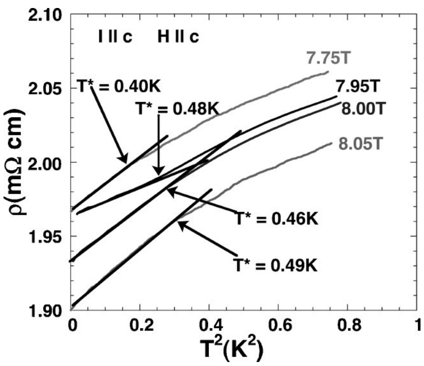

FIG. 3. Resistivity versus T^2 for $I \| c$ and $B \| c$. Solid lines are fits to the T^2 dependence of the resistivity for the low temperature region. T^* denotes the approximate characteristic temperature where the resistivity starts to deviate from the T^2 dependence with increasing temperature.

perature, $T^* \sim 0.5$ K, $\Delta\rho$ starts to deviate from the FL T^2 dependence. The solid lines are guides to the eye and T^* is approximately the point where the resistivity curve starts to digress from the solid line at each field. For $T > T^*$, the resistivity, $\Delta\rho$, follows a power law of T with an exponent less than two, which is common in NFL systems. The values of T^* in the resistivity (see Fig. 3) at fields near B_1^* are very close to each other and consistent with the temperature (~ 0.6 K) below which γ is constant in the specific heat data.

Grigera *et al.*[12] observed similar temperature dependence near the metamagnetic field in their low temperature resistivity data on $Sr_3Ru_2O_7$, with the current *in the ab plane* and field along the c axis. Fine-tuning the magnetic field to the critical point they observe a T^3 dependence in $\rho(T)$ over a small range of T. Note that the resistivity is strongly anisotropic and the residual resistivity is three orders of magnitude larger for current along the c axis as compared to the current in the ab plane.

By its nature a QCP can only be reached if all parameters affecting the transition can be fine-tuned. It involves no energy scale and small perturbations cause deviations from the theoretical QCP. Due to this evasive nature of the QCP, it is not unusual for the system to enter a stable state, such as a FL state or one with long-range order. As the QCP is approached, at a certain point, other parameters than the tuning parameter (magnetic field in this case), e.g., chemical composition and pressure, would also have to be fine-tuned to actually reach the QCP. Numerous disorder driven alloys and systems with field- or (hydrostatic and chemical) pressure-tuned QCP's have been reported in the literature.[26]

CONCLUSIONS

We have presented low temperature specific heat data of $Sr_3Ru_2O_7$ that provide direct thermodynamic evidence of quantum fluctuations as well as the evasive nature of a QCP yielding a crossover to the FL state as the field is tuned in very close to the critical metamagnetic field. Supporting evidence was given by the low temperature resistivity data of $Sr_3Ru_2O_7$ taken near the critical metamagnetic field, which also show a crossover from the NFL state to the FL state at approximately the same temperature as in the specific heat data. We have proposed an additional term to the Schottky anomaly, which is electronic in nature, to account for the extra contribution to the low temperature Schottky term that correlates with the peaks in γ close to the two metamagnetic transitions. The additional term could arise from the electronic structure affecting the nuclear Knight shift or the electric field gradient at the nuclei (quadrupolar contribution), and in either case is roughly proportional to the local density of states.

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