Electrical transport properties of single-crystal $Sr_3Ru_2O_7$: The possible existence of an antiferromagnetic instability at low temperatures

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We report the results of Hall coefficient R_H and magnetoresistance (MR) measurements on single crystalline samples of $Sr_3Ru_2O_7$ grown by the floating zone method. R_H was found to be positive over the entire temperature range studied (0.3–300 K). Its temperature (T) dependence follows closely that of the magnetic susceptibility including a maximum at a characteristic temperature $T^*=17$ K. We show that R_H can be decomposed into normal and anomalous parts as in the case of skew scattering in heavy-fermion compounds and ferromagnetic metals. This, together with the observation that the longitudinal MR is greater than the transverse MR at the same magnetic field and temperature, suggests that magnetic fluctuations dominate the electrical transport properties in $Sr_3Ru_2O_7$. We found a crossover in the sign of the MR at T^*, from positive to negative as the temperature increased, for both the transverse and the longitudinal configurations. In addition, a nonmonotonic behavior in the field dependence of the MR was found at low temperatures. These observations suggest that the magnetic correlations in $Sr_3Ru_2O_7$ at ambient pressure undergo a qualitative change as the temperature is lowered. Above T^*, they are dominated by ferromagnetic instability. However, below T^*, the system crosses over to a different behavior, controlled possibly by a canted antiferromagnetic instability.

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I. INTRODUCTION

The discovery of superconductivity in the layered perovskite Sr_2RuO_4,[1] which appears to possess a p-wave spin-triplet pairing,[2–4] has renewed interest in the magnetic properties of related compounds such as $SrRuO_3$ and $Sr_3Ru_2O_7$. Understanding the magnetic properties of these compounds may provide clues to the mechanism leading to p-wave pairing in Sr_2RuO_4.

$SrRuO_3$, the three-dimensional cubic perovskite in the Ruddlesden-Popper (RP) series ($Sr_{n+1}Ru_nO_{3n+1}$ with $n=\infty$), is an established ferromagnet with $T_c=160$ K.[5,6] For $Sr_3Ru_2O_7$, the $n=2$ member in the RP series has been the subject of controversy regarding its magnetic properties.[7–11] In the original work of Cava *et al.* on phase pure, polycrystalline $Sr_3Ru_2O_7$,[7] the magnetic susceptibility χ was found to show a peak around 15 K, accompanied by a Curie-Weiss behavior at high temperatures, $\chi=C/(T-\theta_{CW})$, where C is the Curie constant, T is the temperature, and θ_{CW} is the Curie temperature, yielding negative $\theta_{CW}\approx 15$ K and a large moment of $2.5\mu_B$/Ru.

In a subsequent study, using single crystals of $Sr_3Ru_2O_7$ prepared by flux method, Cao *et al.*[8] reported a ferromagnetic (FM) ordering at 104 K under ambient pressure. This result was in sharp contrast with the neutron diffraction results on polycrystalline $Sr_3Ru_2O_7$, which did not reveal any long-range magnetic ordering down to 1.6 K.[9] Recently, Ikeda *et al.*[10] reported the magnetic susceptibility [$\chi(T)$] data of $Sr_3Ru_2O_7$ obtained on crystals prepared by the floating zone (FZ) method, which shows a peak at about 17 K but no ferromagnetic ordering. This result is different from the result of Cao *et al.* but consistent with earlier results obtained in polycrystals. It has been suggested that the FM ordering observed in flux crystals was induced by contamination from impurities of the flux, the crucible, or a combination of both.[10]

Ikeda *et al.* also studied the magnetic properties of $Sr_3Ru_2O_7$ under hydrostatic pressure. Clear evidence for FM ordering was found. The FM transition temperature T_c was found to be 70 K at 1.1 GPa. Therefore the authors argued that $Sr_3Ru_2O_7$ is a nearly FM Fermi liquid. They furthermore suggested that the maximum behavior in $\chi(T)$ might be due to the critical spin fluctuations found close to a quantum critical point, as in the case of $(Ca,Sr)_2RuO_4$ (Ref. 12) and MnSi.[13]

To further clarify the nature of the magnetic fluctuations in $Sr_3Ru_2O_7$, we have carried out Hall coefficient (R_H) and magnetoresistance (MR) measurements using $Sr_3Ru_2O_7$ single crystals grown by the FZ method. In this article, we will present the results of our measurements and discuss their physical implications.

II. EXPERIMENTAL METHODS

Single crystals of $Sr_3Ru_2O_7$ used in this study were grown by the floating-zone method. X-ray diffraction measurements confirmed a crystal structure of the $n=2$ RP compound, showing no impurity phases. For in-plane (MR) and Hall measurements, we used two rectangular-shaped single crystals with dimensions around $0.5\times0.1\times0.1$ and $0.8\times0.3\times0.15$ mm^3, respectively. For each sample, two current contacts covering the opposite ends and four voltage contacts on the two sides of the crystal were prepared. All RuO_2

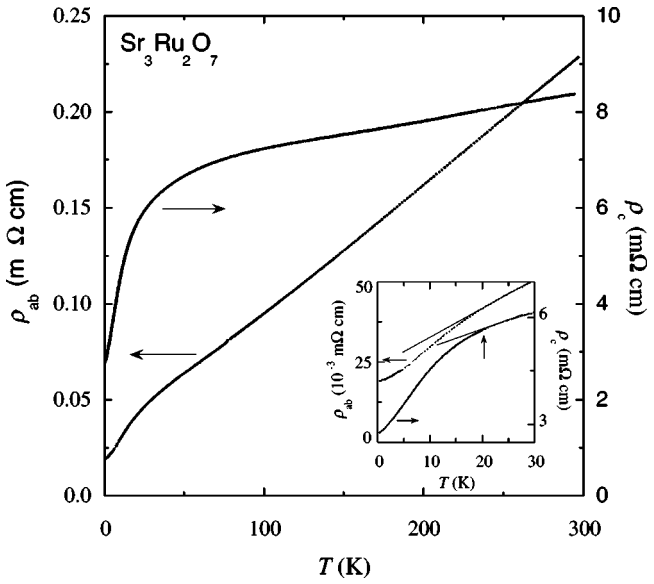

FIG. 1. Temperature dependence of resistivity of $Sr_3Ru_2O_7$ single crystal. The inset shows that both ρ_{ab} and ρ_c have a slope change around 17 K.

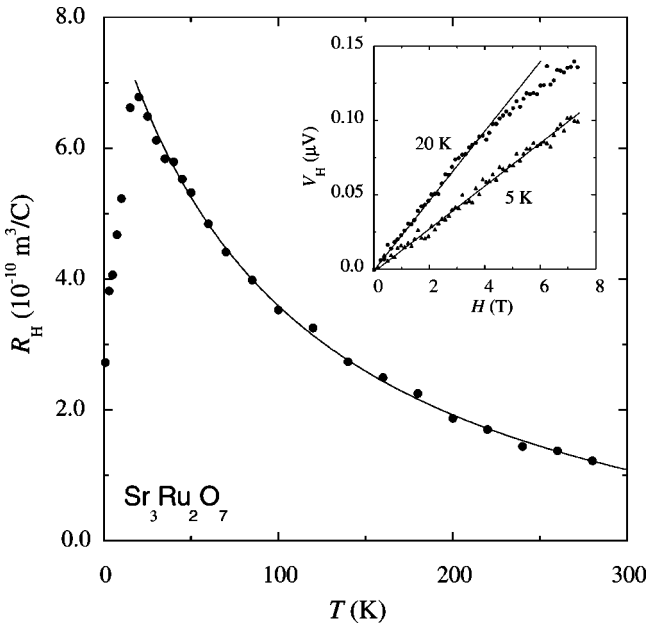

FIG. 2. In-plane Hall coefficient $R_H(T)$ for $Sr_3Ru_2O_7$ with a peak around 17 K. The inset shows the magnetic field dependence of V_H at two different temperatures.

layers were electrically shorted along the c axis to ensure a homogeneous current distribution. The c axis transverse ($H \perp I$) MR measurements were carried out in a single crystal with dimensions around $0.8 \times 0.4 \times 0.2$ mm^3. Two ring-shaped current contacts were prepared on the opposite ab faces. The two voltage contacts were pointlike positioned in the center of the rings.

Electrical measurements were carried out in a ^{3}He and dilution refrigerator. The temperature was measured using a Lakeshore Cernox 1030 thermometer with relative temperature corrections (due to the applied magnetic field, typically 0.15% at 4.2 K and 5.9% at 2 K and 8.0 T). For transverse and longitudinal MR measurements, the magnetic field H was applied perpendicular and parallel to the injected current I, respectively. In order to exclude the Hall contribution to the MR, only the symmetric part of $\Delta\rho_{ab}(H) = \rho_{ab}(H) - \rho_{ab}(0)$ under field reversal was included. For Hall measurements, the magnetic field was applied parallel to the c axis with a current bias applied along the ab plane. The Hall voltage V_H, which contains only the asymmetric contributions under field reversal, was found to vary linearly with H up to 4 T over the whole temperature region. By fitting $V_H(H)$ data using $V_H = R_H HI/d$ (d is the thickness of the sample along the c axis), the Hall coefficient R_H was obtained.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

Figure 1 shows the temperature dependence of electric resistivity for in- and out-of-plane directions of the $Sr_3Ru_2O_7$ crystals (denoted by ρ_{ab} and ρ_c respectively). The overall shape of these data are similar to those reported in Ref. 10. Both $\rho_{ab}(T)$ and $\rho_c(T)$ are metallic in the whole temperature region. The sharp drop in $\rho_c(T)$ below 50 K has been attributed to the suppression of thermal scattering between quasiparticles and phonons.[10] Interestingly, both ρ_{ab} and ρ_c were found to show an apparent slope change around 17 K (see the inset of Fig. 1). This temperature is close to that below which the magnetic susceptibility drops sharply indicating that magnetic fluctuations have strong influence on the electrical transport properties.

The temperature dependence of the Hall coefficient R_H is shown in Fig. 2. R_H is positive over the entire temperature region studied (0.30–300 K) reaching a maximum at the same temperature ($=17$ K) where both $\rho_{ab}(T)$ and $\rho_c(T)$ show a slope change (T^* will be used to denote this characteristic temperature). These observations agree well with a previous report[14] in which R_H was measured on single crystals prepared by the FZ method as well.

The temperature dependence of $R_H(T)$ is strikingly similar to that of $\chi(T)$.[7,10] Such a behavior was observed previously in heavy fermion compounds such as in UPt_3[15] and ferromagnetic metals.[16] It was found that R_H can be fit by

$$R_H = R_0 + R_s \times 4\pi\chi(T). \quad (1)$$

R_0 and R_s are referred to as the normal and anomalous Hall coefficients, respectively. Such behavior of R_S can be understood in the picture of a skew scattering that involves competition between spin-orbit coupling and spin-flip scattering.[17]

For $Sr_3Ru_2O_7$, it has been shown that $\chi(T)$ satisfies the Curie-Wiess law[7,10] at high temperatures. We have attempted to fit our R_H data shown in Fig. 2 using the following equation:

$$R_H = R_0 + R_s'/(R - \theta_{CW}) \quad (2)$$

with three fitting parameters. The best fit (see the solid line in Fig. 2) was obtained at $R_0 = -1.4 \times 10^{-10}$ m^3/C, θ_{CW}

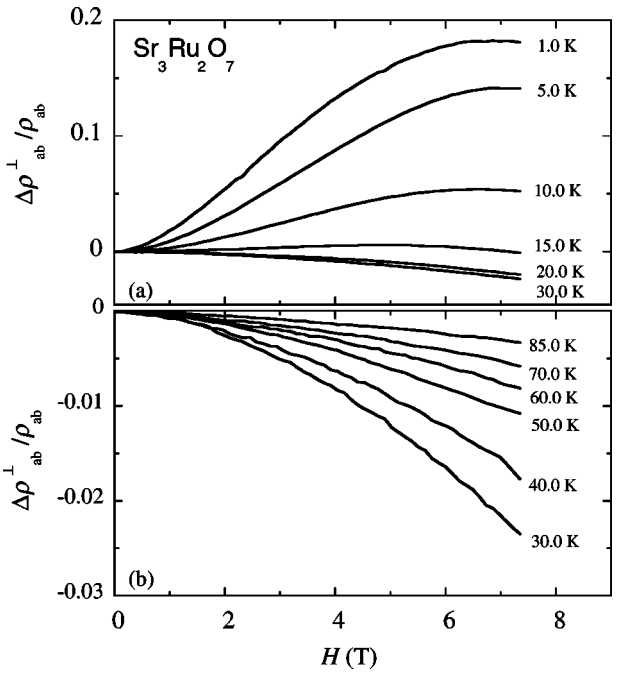

FIG. 3. In-plane transverse MR, $\Delta\rho_{ab}^{\perp}/\rho_{ab}$ $(H\perp ab,\ I\|ab)$, for $Sr_3Ru_2O_7$. A sign change in MR is seen at 17 K.

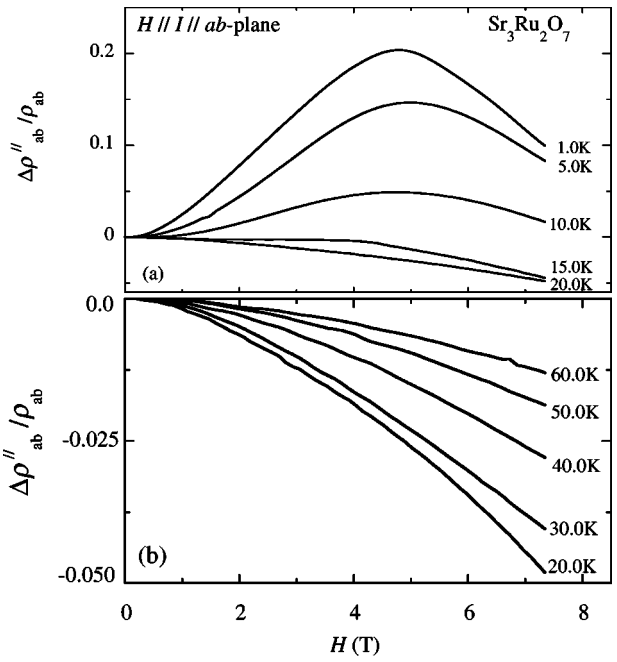

FIG. 4. In-plane longitudinal MR, $\Delta\rho_{ab}^{\parallel}/\rho_{ab}$ $(H\|ab,\ I\|ab)$, for $Sr_3Ru_2O_7$.

$=-102$ K, and $R_S'=1.7\times10^{-7}$ m^3/C. The absolute value of θ_{CW} so obtained is higher than that obtained by fitting $\chi(T)$ curves (around -15 K for polycrystals and -40 K for single crystals), but has the same sign as that found from magnetic susceptibility measurements. R_0 was found to be negative, which means that the normal Hall effect is dominated by electrons. This is consistent with our expectation since band structure calculations reveal that in $Sr_3Ru_2O_7$ there are four electron and two holelike bands crossing the Fermi surface.[18] These observations suggest that skew scattering is a reasonable picture for understanding our $R_H(T)$ result.

As pointed out in Ref. 19, a negative θ_{CW} does not mean that the magnetic fluctuations are necessarily antiferromagnetic (AFM) in nature, as frequently assumed in literature. In fact, we found that V_H as a function of magnetic field deviated from linear behavior above T^* (see inset of Fig. 2) in a manner characteristic of ferromagnetic behavior, consistent with the argument of nearly FM behavior in Ref. 10. However, below T^*, V_H becomes linear with H.

Figure 3 shows the transverse in-plane MR, $\Delta\rho_{ab}^{\perp}/\rho_{ab}$ $(H\perp I)$, between 0 and 7.3 T at various temperatures. At high temperatures, $\Delta\rho_{ab}^{\perp}/\rho_{ab}$ is negative and small, varying monotonically with H. The magnitude of $\Delta\rho_{ab}^{\perp}/\rho_{ab}$ increases with decreasing temperature down to 30 K. As T approaches T^* from high temperatures, the opposite tendency is observed. The value of $\Delta\rho_{ab}^{\perp}/\rho_{ab}$ eventually becomes positive with $T<T^*$ $(=17$ K). Another striking feature is that, below T^*, $\Delta\rho_{ab}^{\perp}/\rho_{ab}$ shows a nonmonotonic field dependence. With increasing H, $\Delta\rho_{ab}^{\perp}/\rho_{ab}$ initially increases, then decreases after reaching a maximum at H_{max}, which appears to increase with decreasing temperature. This nonmonotonic behavior of $\Delta\rho_{ab}^{\perp}/\rho_{ab}(H)$ persists to the lowest temperature measured.

We also measured the longitudinal in-plane MR, $\Delta\rho_{ab}^{\parallel}/\rho_{ab}$, of $Sr_3Ru_2O_7$. As shown in Fig. 4, $\Delta\rho_{ab}^{\parallel}/\rho_{ab}$ exhibits similar features as $\Delta\rho_{ab}^{\perp}/\rho_{ab}$. In particular, it also shows a sign reversal at temperatures slightly lower than T^*. A more pronounced nonmonotonic field dependence was also found for $\Delta\rho_{ab}^{\parallel}/\rho_{ab}$ for $T<15$ K.

We would like to point out another important feature in Figs. 3 and 4: The magnitude of $\Delta\rho_{ab}^{\parallel}/\rho_{ab}$ is greater than that of $\Delta\rho_{ab}^{\perp}/\rho_{ab}$ at the same magnetic field below approximately 10 K. Since the current is not subject to the Lorentz force in the longitudinal configuration, it reflects mostly the contribution of spins. Therefore, this observation, together with that of the skew scattering, suggests that spin scattering has a strong influence on the electrical transport in $Sr_3Ru_2O_7$.

Figure 5 shows the transverse c-axis MR $(\Delta\rho_c^{\perp}/\rho_c)$ of $Sr_3Ru_2O_7$ at different temperatures. It is clear that $\Delta\rho_c^{\perp}/\rho_c$ exhibits all the features observed in the in-plane MR. The only obvious difference is that the negative-to-positive sign reversal in $\Delta\rho_c^{\perp}/\rho_c$ was found at a slightly lower temperature (10 K).

It is remarkable that a maximum in $\chi(T)$ and $R_H(T)$, a slope change in $\rho_{ab}(T)$ and $\rho_c(T)$, a deviation from linear magnetic field dependence in V_H, and a sign change in longitudinal and transverse MR were all found at approximately the same temperature. These observations suggest that in $Sr_3Ru_2O_7$ instead of continuing its trend to move closer to FM ordering as temperature is lowered, as one would naturally expect, the system is side tracked to a different behavior below T^*. This qualitative change in the dynamics of the system has to be magnetic in origin.

Physical insight may be obtained from the $(Ca_{2-x}Sr_x)RuO_4$ solid solution system.[12] For x

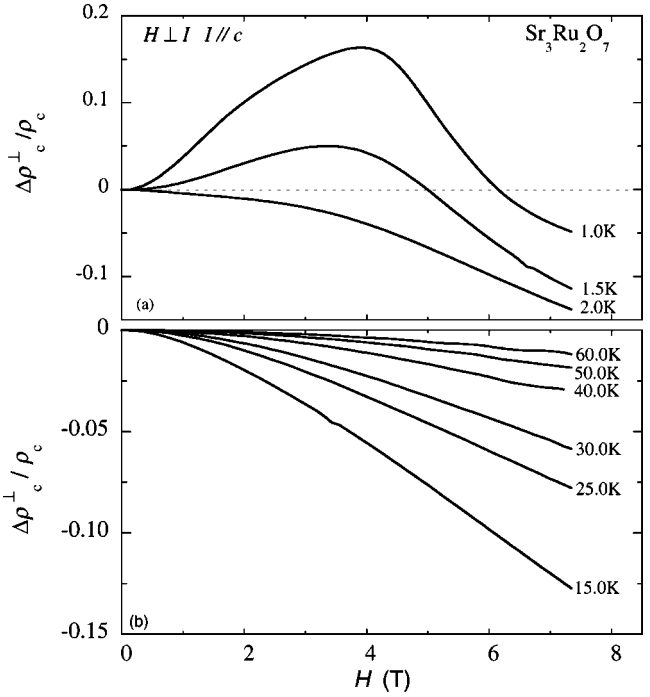

FIG. 5. c axis transverse MR, $\Delta\rho_c^{\perp}/\rho_c$ ($H\perp c$, $I\|c$), for $Sr_3Ru_2O_7$.

<0.2, $(Ca_{2-x}Sr_x)RuO_4$ is antiferromagnetic. For $0.2\leqslant x \leqslant 0.5$, this material system is near a FM instability. In particular, at $x\simeq 0.5$, it is nearly ferromagnetic, evolving from paramagnetic Sr_2RuO_4 through band narrowing. Between $0.2\leqslant x<0.5$, it changes into a state with short-range AFM ordering below a characteristic temperature T_P (T_P, which is about 10 K at $x=0.2$, decreases continuously to zero at $x=0.5$). Below T_P, $\rho_{ab}(T)$ shows a change of slope. MR shows a negative-positive sign reversal and nonmonotonic behavior in field dependence. In addition, both $\chi(T)$ and R_H show a maximum at T_P.

The phenomena seen in $(Ca_{2-x}Sr_x)RuO_4$ are clearly similar to those of $Sr_3Ru_2O_7$. Furthermore, $(Ca_{2-x}Sr_x)RuO_4$ and $Sr_3Ru_2O_7$ have comparable electronic specific heat coefficients and Wilson ratios. All these similarities suggest that the magnetic correlations in $Sr_3Ru_2O_7$ show a qualitative change below T^*. At high temperatures, FM spin fluctuations dominate because of the close proximity to the FM instability while at low temperatures (below T^*) they cross over to AFM fluctuations.

In the magnetization (M) vs field (H) curve obtained on $Sr_3Ru_2O_7$ polycrystals,[7,11] $M(H)$ shows a convex character at low fields below T^*. This is a characteristic feature of AFM correlations. Above T^*, $M(H)$ shows a concave feature typical of FM correlations. This further supports the observation that as the temperature decreases, $Sr_3Ru_2O_7$ at ambient pressure moves away from a FM, but closer to an AFM magnetic instability. This scenario seems to be capable of explaining all features seen in the electrical transport in $Sr_3Ru_2O_7$ described above.

In $(Ca_{2-x}Sr_x)RuO_4$, the change of magnetic correlations is driven by a structural phase transition at temperature above T_P.[19] In $Sr_3Ru_2O_7$, although neutron diffraction measurements did not reveal any structural transition,[9] an unusual change in structural details at low temperature, i.e., a negative thermal expansion along the c axis, was observed. This might be responsible for the change of magnetic coupling at low temperatures.

We note, however, that the anisotropy in $\chi(T)$ between the in- and out-of-plane directions in $Sr_3Ru_2O_7$ single crystals[10] is quite small. In addition, $\chi(T)$ was found to saturate below roughly 6 K. How do we explain these observations in the picture of AFM fluctuations below T^*?

This apparent difficulty may be solved if we assume that the spins aligned antiferromagnetically in $Sr_3Ru_2O_7$ are canted to the c axis. A small net ferromagnetic component in the c axis in the canted AFM can explain the slight hysteresis observed in the $M-H$ curve on polycrystals $Sr_3Ru_2O_7$.[11] The saturation of $\chi(T)$ below 6 K is likely to imply that the crossover from FM to AFM is incomplete probably because $Sr_3Ru_2O_7$ does not undergo a structural phase transition as in $(Ca_{2-x}Sr_x)RuO_4$. This might also be the reason why neutron diffraction did not detect any sizable magnetic ordering.

For $(Ca_{2-x}Sr_x)RuO_4$, the magnetic instability in the $0.2 \leqslant x \leqslant 0.5$ region leads to non-Fermi liquid behavior. Its $\rho_{ab}(T)$ shows a $T^{1.4}$ dependence. For $Sr_3Ru_2O_7$, our resistivity data exhibit a similar behavior. Both $\rho_{ab}(T)$ and $\rho_c(T)$ clearly deviate from T-squared dependence. The best fit is $T^{1.4}$ for $\rho_{ab}(T)$ and $T^{1.2}$ for $\rho_c(T)$, as shown in Fig. 6. However, we note that the residual resistivity ρ_0 estimated from Fig. 1 is about 19.0 (2.8) $\mu\Omega$ cm for ρ_{ab} (ρ_c). It is about five times (for ρ_{ab}) and two times (for ρ_c) greater than that reported by Ikeda in Ref. 10, suggesting that our FZ crystals may contain slightly more impurities and defects than theirs. As a result, it is natural to ask whether the observed non-T^2 behavior is intrinsic to $Sr_3Ru_2O_7$. In Ref. 10, it was argued that $Sr_3Ru_2O_7$ is a Fermi liquid with $\rho_{ab}(T)\propto T^2$. However, their T-squared fitting for resistivities, especially for $\rho_{ab}(T)$, appears to be less than the best fit. A deviation from the T-squared dependence in $\rho_{ab}(T)$ is clearly visible even below 6 K. This deviation suggests an intrinsic deviation from Fermi liquid behavior in $Sr_3Ru_2O_7$ most probably due to the system being close to an AFM instability.

The nonmonotonic field dependence of MR observed in $Sr_3Ru_2O_7$ below T^* can be easily understood in the crossover scenario as discussed above in which the MR should contain both negative and positive terms. The negative term must have originated from the development of FM spin fluctuations as the field increases.

IV. CONCLUSION

In summary, we have studied the transport properties of $Sr_3Ru_2O_7$ under magnetic field using single crystals prepared by the FZ method. The temperature dependence of the in-plane Hall coefficient has been found to resemble that of the magnetic susceptibility showing a maximum at T^*. The field dependence of V_H was found to deviate from linear behavior above T^*. Both $\rho_{ab}(T)$ and $\rho_c(T)$ exhibit a slope change at about T^*. Furthermore, we found that the longitudinal in-

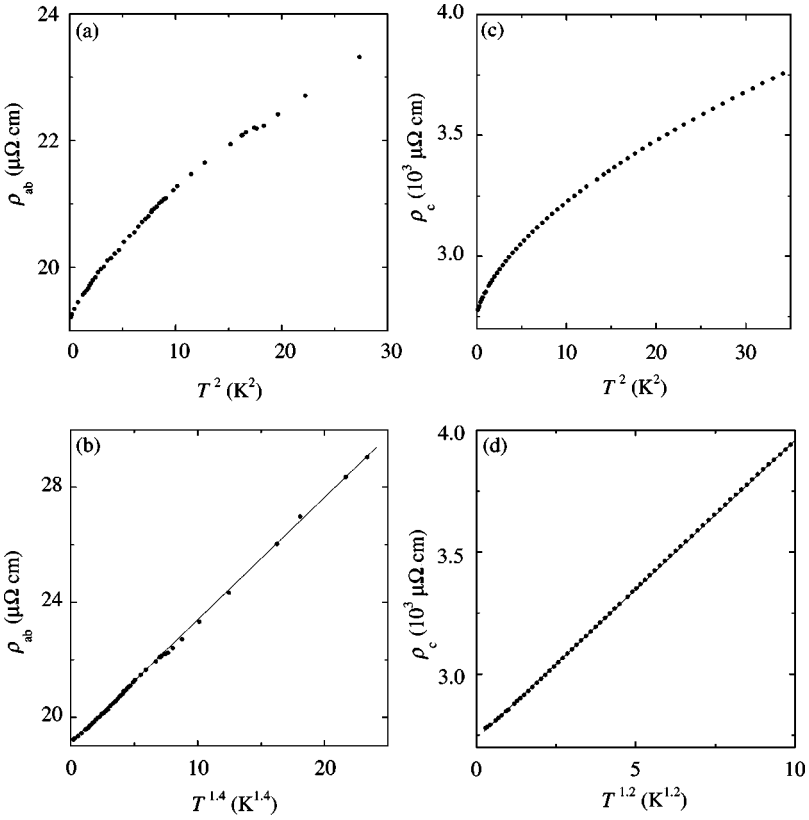

FIG. 6. (a) and (c) show the deviation of ρ_{ab} and ρ_c from T^2 dependence. The best fits obtained are $T^{1.4}$ for ρ_{ab} (b) and $T^{1.2}$ for ρ_c (d).

plane MR is larger than the transverse. Both the in-plane and *c*-axis MR show a negative-to-positive sign reversal below T^*. All these observations support our assessment that the magnetic correlations in $Sr_3Ru_2O_7$ are dominated by FM fluctuations above T^* but cross over to AFM behavior blow T^*. Finally, the ground state may deviate from the conventional Fermi-liquid behavior.

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