We report the results of Hall coefficient $R_H$ and magnetoresistance (MR) measurements on single crystalline samples of Sr$_3$Ru$_2$O$_7$ grown by the floating zone method. $R_H$ was found to be positive over the entire temperature range studied (0.3 - 300K). Its temperature ($T$) dependence follows closely that of the magnetic susceptibility, including a maximum at a characteristic temperature $T^* \approx 17$ 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$_3$Ru$_2$O$_7$. 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 non-monotonic behavior in the field dependence of the MR was found at low temperatures. These observations suggest that the magnetic correlations in Sr$_3$Ru$_2$O$_7$ 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.

I. INTRODUCTION

The discovery of superconductivity in the layered perovskite Sr$_2$RuO$_4$, which appears to possess a $p$-wave spin-triplet pairing, has renewed interest in the magnetic properties of related compounds, such as SrRuO$_3$ and Sr$_3$Ru$_2$O$_7$. Understanding the magnetic properties of these compounds may provide clues to the mechanism leading to $p$-wave pairing in Sr$_2$RuO$_4$.

SrRuO$_3$, the three-dimensional (3D), cubic perovskite in the Ruddlesden-Popper (R-P) series (Sr$_{n+1}$Ru$_n$O$_{3n+1}$, with $n = \infty$), is an established ferromagnet with $T_c = 160$ K. For Sr$_3$Ru$_2$O$_7$, the $n=2$ member in the R-P series, has been the subject of controversy regarding its magnetic properties. In the original work of Cava et al. on phase-pure, polycrystalline Sr$_3$Ru$_2$O$_7$, the magnetic susceptibility $\chi$ 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 $\theta_{CW}$ 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$_3$Ru$_2$O$_7$ prepared by flux method, Cao et al. 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$_3$Ru$_2$O$_7$, which did not reveal any long-range magnetic ordering down to 1.6 K. Recently, Ikeda et al. reported the magnetic susceptibility ($\chi(T)$) data of Sr$_3$Ru$_2$O$_7$ obtained on crystals prepared by the FZ method, which shows a peak at about 17 K but no ferromagnetic ordering. This result is different from Cao’s result, 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.

Ikeda et al. also studied the magnetic properties of Sr$_3$Ru$_2$O$_7$ 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$_3$Ru$_2$O$_7$ is a nearly FM Fermi liquid. They further showed 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)$_2$RuO$_4$ and Mn$\text{S}_2$.

To further clarify the nature of the magnetic fluctuations in Sr$_3$Ru$_2$O$_7$, we have carried out Hall coefficient ($R_H$) and magnetoresistance (MR) measurements using Sr$_3$Ru$_2$O$_7$ 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$_3$Ru$_2$O$_7$ used in this study were grown by the floating-zone method. X-ray diffraction measurements confirmed a crystal structure of...
For in-plane MR and Hall measurements, we used the crystal were prepared. All RuO$_2$ 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 $0.8 \times 0.4 \times 0.2 \text{mm}^3$. Two ring-shaped current contacts were prepared on the opposite $ab$ faces. Two voltage contacts were point-like 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 2K and 8.0T). 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 $V_H = R_H \cdot H \cdot I/d$ ($d$ is the thickness of the sample along the $c$-axis), the Hall coefficient $R_H$ was obtained.

![FIG. 1. Temperature dependence of resistivity of Sr$_3$Ru$_2$O$_7$ single crystal. The inset shows that both $\rho_{ab}$ and $\rho_c$ have a slope change around 17 K.](image1)

![FIG. 2. In-plane Hall coefficient $R_H(T)$ for Sr$_3$Ru$_2$O$_7$ with a peak around 17K. The inset shows the magnetic field dependence of $V_H$ at two different temperatures.](image2)

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

Figure 1 shows the temperature dependence of electric resistivity for in- and out-of-plane directions of the Sr$_3$Ru$_2$O$_7$ crystals (denoted by $\rho_{ab}$ and $\rho_c$ 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 phonon scattering between quasi-particles and phonons. Interestingly, both $\rho_{ab}$ and $\rho_c$ 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 (0.30-300K), 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 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 $\chi(T)$. Such a behavior was observed previously in heavy fermion compounds such as in UPt$_3$ and ferromagnetic metals. It was found that $R_H$ can be fit by

$$R_H = R_0 + R_\alpha \times 4\pi\chi(T).$$  

(1)
Following equation, line in Fig. 2) was obtained at 

\[ R \] attempted to fit our with three fitting parameters. The best fit (see the solid R\(_5\)) can be understood in the picture of a skew scattering that involves competition between spin-orbit coupling and spin-flip scattering. 

For Sr\(_2\)Ru\(_2\)O\(_7\), it has been shown that \( \chi(T) \) satisfies the Curie-Wiess law at high temperatures. We have attempted to fit our \( R_\text{H} \) data shown in Fig 2 using the following equation,

\[ R_\text{H} = R_0 + R'_s/(T - \theta_{CW}), \]  

with three fitting parameters. The best fit (see the solid line in Fig. 2) was obtained at \( R_0 = -1.4 \times 10^{-10} \text{ m}^3/\text{C}, \theta_{CW} = -102 \text{ K}, \) and \( R'_s = 1.7 \times 10^{-7} \text{ m}^3/\text{C}. \) The absolute value of \( \theta_{CW} \) 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\(_2\)Ru\(_2\)O\(_7\) there are four electron- and two hole-like bands crossing the Fermi surface. These observations suggest that skew scattering is a reasonable picture for understanding our \( R_\text{H}(T) \) result.

As pointed out in Ref. 19, a negative \( \theta_{CW} \) does not mean that the magnetic fluctuations are necessarily antiferromagnetic (AFM) in nature, as frequently assumed in literature. In fact, we found that \( V_\text{H} \) 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 \( T^* \), \( V_\text{H} \) becomes linear with \( H \).

Figure 3 shows the transverse in-plane MR, \( \Delta\rho^\perp_{ab}/\rho_{ab} \) (\( H \perp ab, I \parallel ab \)) for Sr\(_2\)Ru\(_2\)O\(_7\). A sign change in MR is seen at 17K.

We also measured the longitudinal in-plane MR, \( \Delta\rho^\parallel_{ab}/\rho_{ab} \), of Sr\(_2\)Ru\(_2\)O\(_7\). As shown in Fig. 4, \( \Delta\rho^\parallel_{ab}/\rho_{ab} \) exhibits similar features as \( \Delta\rho^\perp_{ab}/\rho_{ab} \). In particular, it also shows a sign reversal at temperatures slightly lower than \( T^* \). A more pronounced non-monotonic field-dependence was also found for \( \Delta\rho^\parallel_{ab}/\rho_{ab} \) for \( T < 15 \text{ K} \).
We would like to point out another important feature in Fig. 3 and 4. The magnitude of \( \Delta \rho_{ab} / \rho_{ab} \) is greater than that of \( \Delta \rho_{ab} / \rho_{ab} \) 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\(_3\)Ru\(_2\)O\(_7\). Figure 5 shows the transverse c-axis MR (\( \Delta \rho_{c}^⊥ / \rho_{c} \)) of Sr\(_3\)Ru\(_2\)O\(_7\) at different temperatures. It is clear that \( \Delta \rho_{c}^⊥ / \rho_{c} \) exhibits all the features observed in the in-plane MR. The only obvious difference is that the negative-to-positive sign reversal in \( \Delta \rho_{c}^⊥ / \rho_{c} \) 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\(_3\)Ru\(_2\)O\(_7\), 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. For \( x < 0.2 \), (Ca\(_{2-x}\)Sr\(_x\))RuO\(_4\) is antiferromagnetic. For \( 0.2 \leq x \leq 0.5 \), this material system is near a FM instability. In particular, at \( x \approx 0.5 \), it is nearly ferromagnetic, evolving from paramagnetic Sr\(_2\)RuO\(_4\) through band narrowing. Between 0.2 \( \leq x \leq 0.5 \), 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 non-monotonic 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\(_3\)Ru\(_2\)O\(_7\). Furthermore, (Ca\(_{2-x}\)Sr\(_x\))RuO\(_4\) and Sr\(_3\)Ru\(_2\)O\(_7\) have comparable electronic specific heat coefficients and Wilson ratios. All these similarities suggest that the magnetic correlations in Sr\(_3\)Ru\(_2\)O\(_7\) 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.
tions is driven by a structural phase transition at temperature above $T_\text{f}$. In Sr$_3$Ru$_2$O$_7$, although neutron diffraction measurements did not reveal any structural transition, 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$_3$Ru$_2$O$_7$ single crystals 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 anti-ferromagnetically in Sr$_3$Ru$_2$O$_7$ are cantled to the $c$-axis. A small net ferromagnetic component in the $c$-axis in the canted antiferromagnet can explain the slight hysteresis observed in the M – H curve on poly-crystals Sr$_3$Ru$_2$O$_7$. The saturation of $\chi(T)$ below 6 K is likely to imply that the crossover from FM to AFM is incomplete, probably because Sr$_3$Ru$_2$O$_7$ does not undergo a structural phase transition as in (Ca$_{1-x}$Sr$_x$)RuO$_4$. That might also be the reason why neutron diffraction did not detect any sizable magnetic ordering.

For (Ca$_{1-x}$Sr$_x$)RuO$_4$, the magnetic instability in the 0.2 $\leq x \leq 0.5$ region leads to non-Fermi liquid behavior. Its $\rho_{ab}(T)$ shows a $T^{1.4}$ dependence. For Sr$_3$Ru$_2$O$_7$, 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. 4. However, we note that the residual resistivity $\rho_0$ estimated from Fig. 4 is about 19.0 (2.8) $\mu\Omega\cdot$cm for $\rho_{ab}$ ($\rho_c$). It is about five times (for $\rho_{ab}$) and two times (for $\rho_c$) 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$_3$Ru$_2$O$_7$. In Ref.10, it was argued that Sr$_3$Ru$_2$O$_7$ 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$_3$Ru$_2$O$_7$, most probably due to the system being close to an AFM instability.

The non-monotonic field-dependence of MR observed in Sr$_3$Ru$_2$O$_7$ 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$_3$Ru$_2$O$_7$ 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-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$_3$Ru$_2$O$_7$ are dominated by FM fluctuations above $T^*$, but cross over to AFM behavior below $T^*$. Finally, the ground state may deviate from the conventional Fermi-liquid behavior.

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