

Magnetic, electrical transport, and thermoelectric properties of $\text{Sr}_4\text{Ru}_3\text{O}_{10}$: Evidence for a field-induced electronic phase transition at low temperatures

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We report measurements on resistivity, thermopower, and magnetization as a function of temperature and magnetic field on single crystalline $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ grown by the floating-zone method. The in-plane and c -axis resistivities, magnetization, and the thermopower were found to exhibit a step feature at low temperatures (below roughly 30 K), accompanied by hysteresis behavior when the in-plane field was swept up and down from below 10 kOe to above 20 kOe. In particular, the sharp increase in the thermopower with increasing in-plane magnetic field at low temperatures has not been observed previously in layered transition metal oxides. We propose that these observations can be explained by the existence of a transition between two electronic states in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ in low and high in-plane magnetic fields, respectively, and the alignment of domains is responsible for the emergence of a different electronic state as the in-plane field is ramped up.

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I. INTRODUCTION

Compounds in the Ruddlesden-Popper (RP) homologous series $\text{Sr}_{n+1}\text{Ru}_n\text{O}_{3n+1}$,^{1,2} with $n=1$ to infinity, exhibit various interesting behaviors as the effective dimensionality, determined by the number of perovskite RuO_6 layers in a unit cell, n , is varied. The $n=1$ member of the series, Sr_2RuO_4 , the most two-dimensional member of the series, is superconducting below 1.5 K (Ref. 3) and features an odd-parity, spin-triplet pairing symmetry that was first predicted theoretically^{4,5} and confirmed subsequently by many experimental studies⁶ including recent phase-sensitive measurements.⁷ On the other end of the series, the infinite-layer, three-dimensional member of the series, SrRuO_3 ($n=\infty$), is an itinerant ferromagnet with a T_c of 165 K (Ref. 8) that may exhibit momentum-space magnetic monopoles.⁹ Compounds with intermediate n values, such as $\text{Sr}_3\text{Ru}_2\text{O}_7$ ($n=2$), are also of fundamental interest because they exhibit interesting phenomena stemming from competition between ferromagnetic (FM) and antiferromagnetic (AFM) instabilities¹⁰⁻¹³ and metamagnetic quantum phase transition.^{14,15} The $n=3$ compound, $\text{Sr}_4\text{Ru}_3\text{O}_{10}$, has received recent attention because of several interesting observations. So far, there seems to be consensus that $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ undergoes a FM transition around $T_c \approx 100$ K based on the measurements carried out on single crystalline samples grown by either the flux^{16,17} or the floating-zone¹⁸ method. The magnetization showed strong anisotropy with respect to its response to the in-plane and c -axis fields, reflecting its layered crystalline structure. There is also a strong magnetoelastic coupling, as demonstrated in the Raman spectroscopy measurements,¹⁹ and a possible phase separation within a range of the in-plane field.²⁰

Several basic issues concerning the nature of the FM phase in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ are not settled, however. First, there has been confusion as to whether a second magnetic transition

exists below $T_c \approx 100$ K. Magnetic measurements have shown that with the field applied along the ab plane, the magnetization as a function of temperature shows a pronounced peak as the temperature is lowered to below around 50 K.¹⁶⁻¹⁸ It was suggested previously that this results from the emergence of an interlayer AFM coupling.¹⁹ However, there is fundamental difficulty in this interpretation of the data (see below). Second, as the in-plane field is increased, a steep rise in magnetization was observed in the range between 10 and 25 kOe, which was interpreted as a metamagnetic transition possibly associated with spin reorientation¹⁹ or evolution of the Fermi surface.²¹ However, the initial neutron scattering measurements did not provide support to the conclusion.²² Finally, there seems to be a feature in the magnetization (M) vs temperature (T), $M(T)$, at about 30 K, whose physical origin has not been clarified.

The purpose of this paper is to report our detailed measurements on electrical resistivity, thermopower, and magnetization of $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ as the function of the in-plane magnetic field and show that there exists a field-induced electronic phase transition at low temperature. We will discuss the possible physical origin of the electronic phase transition.

II. EXPERIMENT

Single crystals of $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ were grown by the floating-zone method. The starting materials are SrCO_3 (99.99% purity) and RuO_2 (99.95% purity). The grown crystals were characterized by x-ray diffraction and energy dispersive x-ray (EDX) measurements. The single crystals in this experiment are of a typical size of $2 \times 1 \times 0.4$ mm³. The in-plane residual resistivity ρ_{ab} is about $1.6 \mu\Omega$ cm at $T=2$ K, and the residual resistivity ratio (RRR), the measure of sample quality, is about 160. Temperature and field dependence of magnetization was measured with $H \parallel ab$ and $H \parallel c$ in

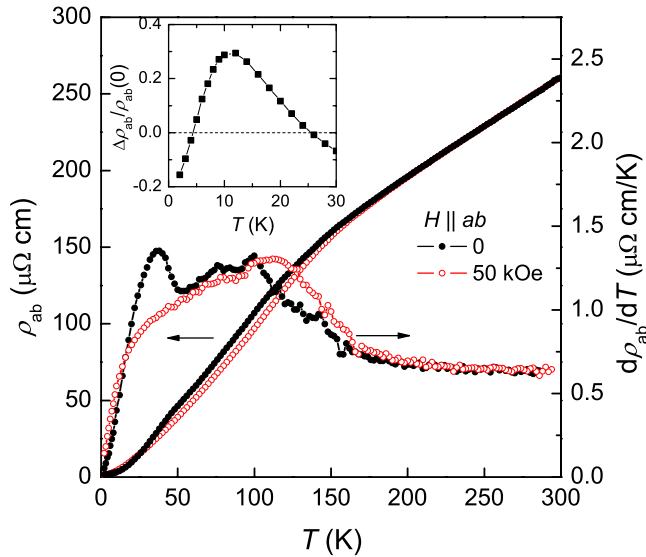


FIG. 1. (Color online) The temperature dependence of the in-plane resistivity and its derivative under 0 and 50 kOe in-plane magnetic field. Inset: magnetoresistance versus temperature for the temperature range from 2 to 30 K.

a Quantum Design MPMS-5 system. The resistance and thermopower were measured with $H \parallel ab$. Resistance was measured by a standard four-terminal method. Thermopower was measured by a steady-state technique. The electric contacts were prepared using silver epoxy with the contact resistance less than 0.5Ω . The temperature gradient applied to the sample was about 0.5 K/mm and was measured by a pair of differential type E thermocouples. The effect of magnetic field on the sensitivity of the thermocouples was carefully calibrated by using calibrated Cernox sensors. All electrical transport measurements were performed in a Quantum Design PPMS-9 system.

III. EXPERIMENTAL RESULTS

A. In-plane electrical resistance and magnetoresistance

Figure 1 displays the temperature dependence of the in-plane resistivity (ρ_{ab}) and its derivative ($d\rho_{ab}/dT$) under 0 and 50 kOe magnetic fields applied along the ab plane. The inset shows the in-plane magnetoresistance (MR), measured at 50 kOe, versus temperature for a range between 2 and 30 K. The MR is seen to grow significantly for temperatures below 150 K, changing its sign twice below 30 K. The derivative of the in-plane resistivity $d\rho_{ab}/dT$ shows two peaks under zero field, at approximately 105 and 35 K. The low-temperature peak vanishes under 50 kOe. In a FM metal, the resistivity as a function of temperature is predicted to feature a change in slope at T_c , which should correspond to a jump in $d\rho_{ab}/dT$ but frequently turns into a peak in $d\rho_{ab}/dT$ experimentally. While the high-temperature peak clearly signals a FM transition, the physical origin of the $d\rho_{ab}/dT$ peak at 35 K in zero field is probably not caused by a magnetic phase transition but rather by FM domain reorganization (see below).

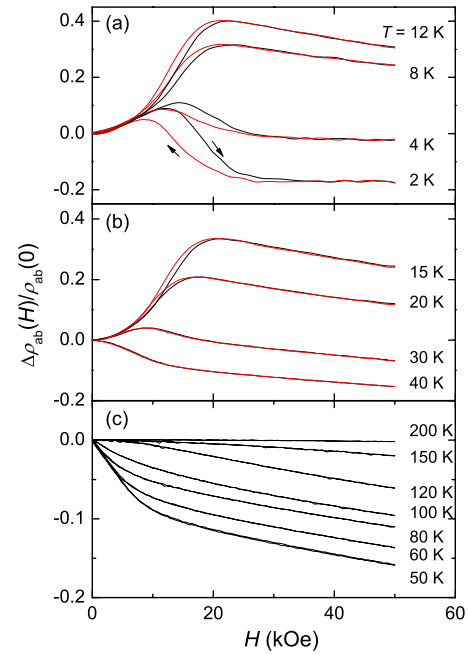


FIG. 2. (Color online) Magnetic field dependence of the in-plane resistivity at several temperatures as indicated. The magnetic field was applied along the ab plane.

Figure 2 shows the in-plane MR vs $H \parallel ab$ at several temperatures. At $T=2$ and 4 K, the in-plane MR increases gradually first when H is lower than 10 kOe, followed by a quick drop when H increases from 13 to 26 kOe and a level-off as the field increases further. As the temperature increases further, the in-plane MR becomes positive even at the highest fields, with its magnitude decreasing as the field increases. However, the two distinct regimes marked by gradual change in MR are clearly discernible. Interestingly, hysteresis behavior was found when the system was brought between the two regimes by the up and down sweep of the in-plane field up to about 30 K, above which the in-plane MR becomes negative in the entire field range. As the temperature increases further, the magnitude of the negative in-plane MR becomes larger. However, the trend is reversed above 50 K. The temperature dependence of in-plane MR and that of $d\rho_{ab}/dT$ described above therefore suggest that fundamental magnetic properties of $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ may still vary even below $T_c=100 \text{ K}$ when the material becomes FM ordered.

B. In-plane thermopower

Figure 3 shows thermopower (S) vs the temperature T . Within the whole temperature range, S is positive, increasing smoothly as the temperature was raised, reaching $34 \mu\text{V/K}$ at room temperature, slightly larger than that of Sr_2RuO_4 ($S=29 \mu\text{V/K}$).²³ For conventional semiconductors, positive thermopower would suggest positive charge carriers (holes). However, this conclusion may not hold here given that the thermopower of Sr_2RuO_4 is also positive in the temperature range measured (4.2–300 K), even though both electrons and holes are known to be present.^{24,25}

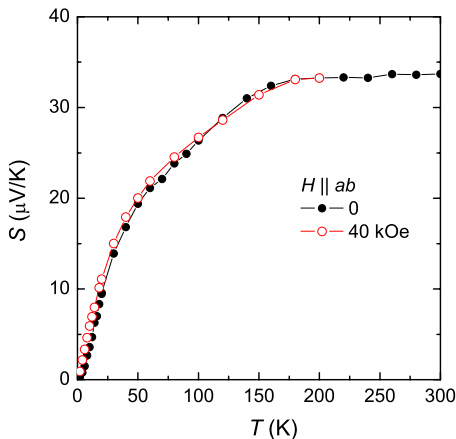


FIG. 3. (Color online) Thermopower (S) vs temperature under 0 and 40 kOe in-plane magnetic field. This piece of the $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ sample is the same one used for resistivity and MR measurements.

The normalized change of thermopower with magnetic field, $\Delta S(H)/S(0)$, exhibits interesting behavior, as shown in Fig. 4, where $\Delta S(H) \equiv S(H) - S(0)$, and H is applied along the ab plane. For $T < 30$ K, $\Delta S(H)/S(0)$ hardly shows any change at low fields, followed by a sharp increase between 8 and 20 kOe, becoming flat as the field increases further. However, the crossover between the two regimes in the low and high in-plane fields is only slightly larger than that of ρ_{ab} . As T is above 50 K, the flat profile in the low-field

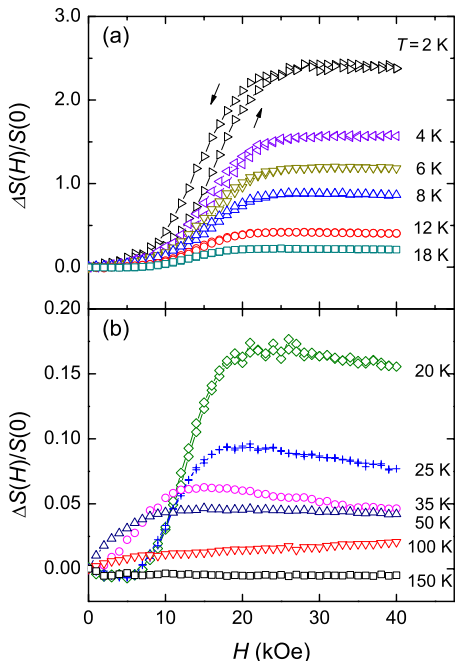


FIG. 4. (Color online) Magnetic field dependence of the normalized thermopower at several temperatures as indicated. The magnetic field is applied along the ab plane. The thermopower values at zero field are 0.283, 0.798, 1.49, 2.66, 4.69, 8.32, 9.57, 11.4, 17.6, 19.4, 26.4, and 31.5 $\mu\text{V}/\text{K}$, for $T=2, 4, 6, 8, 12, 18, 20, 25, 35, 50, 100,$ and 150 K, respectively.

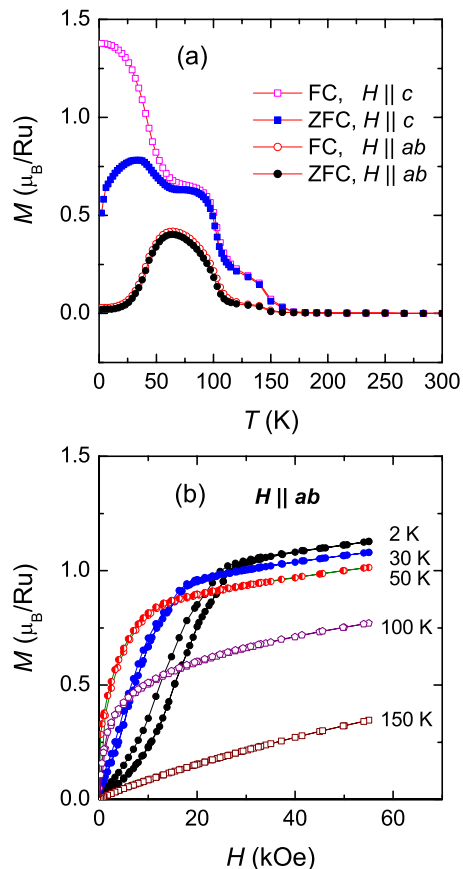


FIG. 5. (Color online) (a) The temperature dependence of magnetization under both in- and out-of-plane magnetic fields and (b) field dependence of magnetization under in-plane magnetic fields.

regime in $\Delta S(H)/S(0)$ is seen to disappear gradually. A large hysteresis with the up and down sweep of the in-plane field was also observed, again up to about 30 K.

C. Magnetization

Figure 5(a) shows the temperature dependence of magnetization (M) under a magnetic field of 1 kOe applied along either the c axis or the ab plane. The measurements were carried out on crystals that were also grown by the floating-zone method, but different pieces from those used in the electrical transport and thermopower measurements. With the magnetic field applied along the c axis, $M(T)$ shows a sharp increase at about 100 K, which is identified as the T_c for the FM transition in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$. Below around 65 K, slightly larger than the temperature at which the in-plane MR trend changes (50 K) as discussed above, $M(T)$ displays a change in slope for $H \parallel c$, increasing more quickly than at higher temperatures as T is lowered. In addition, $M(T)$ curves for the field cooling (FC) and zero field cooling (ZFC) are seen to deviate from one another at this temperature, 65 K. For the magnetic field applied in an in-plane direction (the field was not aligned with any specific in-plane direction), $M(T)$ shows a pronounced peak around 65 K, decreasing sharply with decreasing temperature until it levels

off around 30 K. Moreover, different from the case in which the field was aligned along the c axis, there is little difference between FC and ZFC curves.

Figure 5(b) shows the magnetization as a function of in-plane field, $M(H\parallel ab)$, at several temperatures as indicated. $M(H\parallel ab)$ is seen to show a gradual change in slope around 10–15 kOe below 30 K, before rising more sharply in the range of 15–25 kOe. Above $H\parallel ab \approx 25$ kOe, the magnetization increases much gradually as the field increases further. Again, a hysteresis behavior for the up and down sweep of the in-plane field was observed in the crossover between the two regimes. Above 30 K, the hysteresis disappears and the magnetization displays the behavior of a typical ferromagnet.

Similar behavior of $M(T)$ as well as $M(H)$ was observed in previous reports.^{16–18} Interestingly, the exact shape of the slope change in $M(T)$ and difference between the FC and ZFC behaviors with a c -axis field appear to be highly sample dependent. In addition, the remanent moment shown in Fig. 5(b) is seen to be nearly zero at all temperatures, in contrast to a significant remanent moment seen previously due to the improved sample quality. The RRR is around 160 for the present samples as compared with 20–30 for the previous crystals.¹⁷ The high purity of our samples could account for this difference in the remanent moment. On the other hand, the behavior in $M(T)$ appears to be insensitive to details of the sample. It should also be noted that the bulk magnetization was found to start to increase slightly around $T \approx 150$ K,^{16–18} suggesting possible intergrowth of a tiny amount of ferromagnetic SrRuO₃ that has an intrinsic T_c of 165 K.

D. c -axis resistivity under an in-plane field

The change of the c -axis resistivity as a function of in-plane field, $\Delta\rho_c(H\parallel ab)/\rho_c(0)$, at various temperatures is shown in Fig. 6. Below 30 K, $\Delta\rho_c(H\parallel ab)/\rho_c(0)$ first increases in the small-field regime, decreases surprisingly sharply near the field ranging between 20 and 25 kOe (depending on the temperature), and then varies much more gradually as the in-plane field increases further. Small hysteresis was seen below 30 K as well, as shown in the inset of Fig. 6(a) for $T=2$ K. The small-field regime disappears gradually above 30 K. However, even at 45 K, the drop in c -axis resistivity is still very sharp, especially in comparison with the similar step feature observed in the in-plane resistivity shown in Fig. 2.

IV. DISCUSSION

As shown above, even though $M(T)$ with the field applied along the c axis shows a steep rise around 65 K, $M(T)$ actually starts to decrease below about the same temperature, reaching a value close to zero at the lowest temperatures when the field is applied along the in-plane direction. Similar behavior in $M(T)$ was observed previously. It was proposed that a FM intraplane ordering without interlayer coupling emerges below 100 K and an AFM ordering among the FM layers below the second characteristic temperature, about 50 K (Refs. 17 and 19) (65 K in the present work). The main

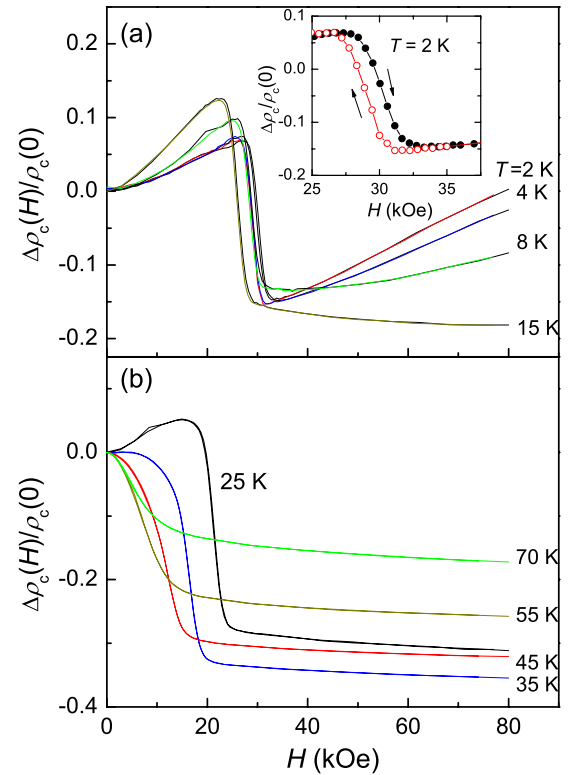


FIG. 6. (Color online) The field dependence of the normalized c -axis resistivity at several temperatures as indicated. The inset in (a) shows the hysteresis at $T=2$ K. The hysteresis disappears as $T > 25$ K. The values of $\rho_c(0)$ are 2.50, 2.51, 2.66, 3.57, 5.75, 7.39, 7.85, 7.76, and 7.59 m Ω cm for $T=2, 4, 8, 15, 25, 35, 45, 55,$ and 70 K, respectively.

problem with the above interpretation is that an interlayer AFM ordering will lead to a vanishing $M(T)$ measured with an in-plane field only if the field is aligned along the easy axis. Since the crystal axes in the magnetic measurements, including those reported here, were not aligned with respect to the field, this interlayer AFM ordering scenario is not likely to be true. Indeed, no AFM ordering of any kind was observed in the initial neutron scattering study.²²

Physical insight may be obtained by considering the strong magnetoelastic coupling found in a Raman study carried out previously in Sr₄Ru₃O₁₀.¹⁹ In that study, the 380 cm⁻¹ B_{1g} phonon frequency which is associated with internal vibrations of the RuO₆ octahedra is highly sensitive to the ferromagnetic order. A distinct change in the slope of the B_{1g} phonon frequency, $d\omega/dT$, is observed below T_c of 105 K. When the magnetic field is applied along the c -axis direction at low temperature (much lower than T_c), the Ru magnetic moments are easily aligned to the c axis by the field, and the B_{1g} phonon frequency exhibits a frequency increase with field. The increase in the B_{1g} phonon frequency implies that there is an increase in the elongation of the RuO₆ octahedra along the c axis and a contraction of the in-plane Ru-O bonds under the c -axis magnetic field. Meanwhile, the B_{1g} phonon frequency exhibits a significant decrease with increasing in-plane field up to about 20 kOe as

$T < 50$ K (actually, this effect became clearly visible only for $T < 30$ K), indicating that there is a distinct increase in the in-plane Ru-O bonds and a decrease in the elongation of the RuO_6 octahedra along the c axis. The decrease in the frequency is more abrupt as the field is around 20 kOe at which the step features in the MR and S were also observed in the present work. This was interpreted as a metamagnetic transition from AFM to FM ordering.¹⁹

We wish to present in this paper a picture based on the domain structure that may explain the results obtained in the present work. In this picture, we envision the existence of two types of domains in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ —the “soft” and the “hard” domains. While soft domains are expected below T_c of 100 K, hard domains start to form only below 50–65 K, growing in size and “hardness” as the temperature is lowered. Below 30 K, these hard domains are pinned and can only be aligned by a large in-plane magnetic field. Because of a strong magnetoelastic coupling, the electronic state of the material can be changed when the entire sample is polarized into a single domain in a strong enough in-plane magnetic field. For fields applied in the c -axis direction, the sample can be polarized into a single domain with a field much smaller than that in the in-plane direction.

The presence of two different types of domain structures could originate from the strong anisotropy. At low temperatures, hard domains are needed to cancel the in-plane magnetization. A small in-plane magnetic field (1 kOe in our case) is not enough to align these hard domains as $T < 30$ K; therefore, a vanishing $M(T)$ is observed due to the cancellation of the magnetic moments of different domains. The magnetization increases nonlinearly with increasing in-plane field and changes dramatically through domain rotation when the field is in the range between 10 and 20 kOe. A well aligned domain structure forms when the field is large enough. The hysteresis observed in M vs $H\parallel ab$ curves below 30 K is a consequence of the domain rotation by the applied magnetic field. The hysteresis in the ab - and c -axis MRs and magnetothermopower could have resulted from the hysteresis in $M(H)$ through a magnetoelastic coupling. It was suggested by a recent paper²⁶ that even a structure transition associated with the reorientation of the magnetic domains is possible, leading naturally to a change in the electronic state. For $H\parallel c$, a small field is enough to align the magnetic moments along the c axis so that the $M(T)$ is larger compared to the case for $H\parallel ab$.

The details of the features found in MR and thermopower support the domain picture. The low-field regime disappears when T is above 30 K, indicating that the domains become soft at high temperatures. The sharp change in the c -axis MR occurs at slightly higher magnetic fields (25–30 kOe) than for the in-plane resistivity. The extremely sharp drop in the c -axis resistivity below 30 K may indicate that a single domain might form at high magnetic field. Actually, the c -axis resistivity drop was observed up to a temperature as high as 70 K, but the low-field shoulder was found to disappear at $T > 30$ K, similar to the case of in-plane resistivity, indicating that the low-field electronic state becomes less well defined above 30 K.

Recently, specific heat measurements²⁷ performed on the $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ single crystals grown by flux method showed that

the specific heat increases gradually in the low-field range, jumps up sharply at $H_c = 29$ kOe, and decreases slowly as $H\parallel ab$ increases further. Such a peak weakens with increasing temperature and disappears above 30 K. This result appears to provide further support to the existence of two distinct electronic states in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ in low and high in-plane magnetic fields. We note, however, that the above cited H_c is larger than that found in the present work. Such a difference in H_c could be due to the difference in samples because the domain structure is also sensitive to possible defects and strains in the crystals.

The increase of thermopower with an increasing magnetic field, as observed in the present work on $\text{Sr}_4\text{Ru}_3\text{O}_{10}$, is quite remarkable. Such an increase in thermopower has never been reported in other RP materials. We note that the thermopower in the cobaltate Na_xCoO_2 is strongly suppressed by the applied magnetic field because the spin entropy which accounts for the large thermopower in cobaltates is efficiently suppressed by the strong magnetic field.²⁸ In view of this result, the observation of the field-induced increase in thermopower in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ can be understood well with the picture that a sufficiently strong in-plane magnetic field leads to the emergence of an electronic phase transition and that the electronic state above the transition field features enhanced entropy. Since the alignment of domain tends to reduce entropy, the increased entropy may have come from orbital rather than spin degrees of freedom, which is very different from the case of cobaltates. However, exactly why the high magnetic field state increases the thermopower in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ is not yet well understood.

V. CONCLUSION

We have measured the temperature and in-plane field dependence of the resistivity, magnetization, and thermopower in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$. It was found that there was a step feature in all these physical properties with increasing in-plane magnetic field at low temperatures, below which significant hysteresis was observed as the in-plane field was swept up and down. We propose here that the step feature marks the transition between the two different electronic states in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$. While metamagnetic transition could account for the existence of the two electronic states, we point out here that the two electronic states could also correspond to the single- or multiple-domain states in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ that feature a strong magnetoelastic coupling. When a sufficiently strong in-plane magnetic field aligns the “hard” domains to form a single domain, a corresponding change in the lattice structure may have taken place because of the magnetoelastic coupling, resulting in a different electronic state. Finally, a sharp increase in the thermopower with increasing in-plane magnetic field at low temperatures, which is highly unusual, was observed, suggesting that the electronic phase transition driven by the in-plane field in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ involves the change of orbital degree of freedom.

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