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Phys. Rev. B **99**, 214418 — Published 12 June 2019

DOI: [10.1103/PhysRevB.99.214418](https://doi.org/10.1103/PhysRevB.99.214418)

# Fe-doping induced suppression of the second magnetic transition in $\text{Sr}_4\text{Ru}_3\text{O}_{10}$

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## Abstract

Ruthenium oxide  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  exhibits a second magnetic transition at a temperature  $T_M$  below its ferromagnetic (FM) transition ( $T_C \sim 105$  K), which has attracted considerable attention in the last two decades. Here, we successfully grew  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) crystal and systematically investigated the magnetotransport property of its nanosheets. We found a feature that there is an in-plane FM order aligned along  $\langle 110 \rangle$  directions below  $T_C$  without the occurrence of the second magnetic transition at  $T_M$ , which is in contrast to that expected in pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheets, where the in-plane FM order will rotate to the  $c$  direction below  $T_M$ . Furthermore, a metamagnetic-like transition is also observed below  $\sim 50$  K at about 2 T when the magnetic field is applied along the  $c$  axis. These results unambiguously demonstrate that the second magnetic transition at  $T_M$  is actually a consequence of spin reorientation induced by the inherent coupling between the spin and lattice, which can be effectively tuned by element doping.

## I. INTRODUCTION

Ruddlesden-Popper (RP) series of ruthenium oxides  $\text{Sr}_{n+1}\text{Ru}_n\text{O}_{3n+1}$  ( $n=1, 2, \infty$ ) are interesting materials displaying  $p$ -wave superconductivity ( $\text{Sr}_2\text{RuO}_4$ ,  $n=1$ ) [1], field-tuned quantum criticality ( $\text{Sr}_3\text{Ru}_2\text{O}_7$ ,  $n=2$ ) [2], and itinerant ferromagnetism ( $\text{SrRuO}_3$ ,  $n=\infty$ ) [3]. The  $n=3$  member,  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ , is an exotic ferromagnet with a Curie temperature  $T_C \sim 105$  K, where the easy axis is along the  $c$  direction [4, 5]. Its ferromagnetic (FM) transition is followed by a second magnetic transition at a temperature  $T_M \sim 50$ -70 K, below which a metamagnetic transition, i.e., a steep increase of magnetization near a critical magnetic field, is observed when an in-plane  $H$  is applied [4-7].

The nature of the second transition at  $T_M$  has been a long-lasting controversy. For instance, the magnetization and Raman spectra measurements suggest that this transition might be a FM to antiferromagnetic (AFM) like transition in the  $ab$  plane [5, 8], while no thermal anomaly is observed at  $T_M$  on the specific heat experiments [9, 10]. On the other hand, a neutron diffraction experiment performed by Granata et al. [11] showed that the magnetic moments are only FM coupled along the  $c$ -axis with neither FM nor AFM component in the  $ab$  plane. However, a very recent neutron diffraction study by Zhu et al. [12] found that there are both in-plane and out-of-plane FM components, and below  $T_M$  the in-plane component inclines continuously toward the out-of-plane direction.

To understand the origin of the second transition at  $T_M$  we performed systematic transport studies on single crystalline  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheets recently [13-16]. We

found that  $T_M$  in nanosheets decreases with decreasing thickness and the FM order is within in the  $ab$  plane with magnetic moments along the  $\langle 110 \rangle$  directions for  $T_M < T < T_C$  [14-16], but below  $T_M$ , the in-plane FM order magically transforms to the  $c$  direction spontaneously [16]. We attributed this spin reorientation observed in  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheets to the competition between the shape anisotropy and the inherent magnetocrystalline anisotropy, where the shape anisotropy increases with decreasing thickness and favors spins aligned in the  $ab$  plane, while the magnetocrystalline anisotropy favors spins aligned along the  $c$  direction [14, 16], and hence it is possibly enhanced substantially at low temperatures due to the slight negative thermal expansion of the  $c$  axis near the bulk  $T_M$  [11, 17].

Element doping is an effective way in tuning lattice parameters and thus provides possibility in tuning the magnetocrystalline anisotropy. Here, we successfully grew  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) (nominal concentration) single crystal using the floating-zone technique [18], and investigated the magnetism of its bulk and nanosheet. Since a 0.12 % shrinking of the  $c$  axis by Fe-doping at room temperature is observed [Fig. 1(a)], the intrinsic magnetocrystalline anisotropy is therefore expected to be reduced accordingly, and thus leading to a suppression of the  $c$  axis FM order. Indeed, four distinct features are observed: i) magnetization measurement on the bulk shows that the magnetic easy direction has been changed from the  $c$  axis to the  $ab$  plane due to Fe-doping; ii) only one resistive anomaly at  $\sim 105$  K due to the FM transition in  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheets is found, the second resistive anomaly, which occurs at  $T_M$  in pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  bulk or nanosheets [5, 6, 13], is absent;

iii) the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet shows an in-plane FM order aligned along  $\langle 110 \rangle$  directions in all temperature range below  $T_C$ ; iv) the anomalous Hall effect (AHE) with  $H$  applied along the  $c$  axis in the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet reveals that there is a metamagnetic-like transition at about 2 T below  $\sim 50$  K, which is in contrast to the metamagnetic transition expected in the  $ab$  plane of the pure system. Our results suggest that replacing Ru with a few Fe ions favors magnetic moments aligned in the  $ab$  plane, which highlight the coupling effect between the lattice and spin in the  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  system at temperatures below  $T_C$ .

## II. EXPERIMENTS

Single crystals of  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) were grown by the floating zone method, which is similar to that described in Ref. [18], where the concentration of Fe was analyzed roughly by energy dispersive spectrum (EDS, Oxford instruments). The starting materials were  $\text{SrCO}_3$  (99.99 % purity),  $\text{Fe}_2\text{O}_3$  (99.99 %) and  $\text{RuO}_2$  (99.95 % purity). 25 % excess  $\text{RuO}_2$  was added to compensate the evaporation of  $\text{RuO}_2$  from melting zone. The mixed powder was grinded for  $\sim 1$  h, and then pressed to pellets, after that the pellets were sintered at temperature of  $900^\circ\text{C}$  for 12 h. The sintered pellets were grinded for  $\sim 1$  h again, the grinded powder was pressed to rod, and sintered at temperature of  $1350^\circ\text{C}$  for 4 h. The growth speed was  $\sim 15$  mm/h and the pressure in quartz tube was  $\sim 10$  bar (10 %  $\text{O}_2$ +90 % Ar).

The structural property of the synthetic crystal was studied by a room temperature x-ray diffractometer (XRD) [Fig. 1(a)]. The magnetization of the

$\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) single crystal, which structure was verified firstly by XRD, was measured by a superconducting quantum interference device (SQUID) magnetometer [Fig. 1(b)].

$\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheets were obtained by the scotch tape-based micro-mechanical exfoliation method from the bulk crystal. Nanosheets with different thicknesses,  $d$ , determined by atomic force microscopy were transferred onto a  $\text{SiO}_2$  (300 nm)/Si substrate. Contacts were patterned using an electron-beam lithography technique followed by deposition of Ti/Au (5 nm/100 nm). The scanning electron microscope (SEM) image of a device with patterned electrodes is shown in Fig. 2(a). The transport property measurements of the nanosheets were carried out in a physical property measurement system (PPMS, Quantum Design). The Hall resistance  $R_{xy}$  is determined from  $R_{xy}=[R_{xy}(H)-R_{xy}(-H)]/2$  in order to subtract the component of the longitudinal resistance arising from the small misalignment of transverse contacts.

### III. RESULTS AND DISCUSSION

#### A. Structure and magnetization of bulk crystal

Fig. 1(a) shows the XRD pattern taken on a cleaved (001) surface of the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) crystal. For comparison, the XRD pattern of a pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  crystal grown by the same method is also presented. One can find that the synthetic crystal well matches with the  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  phase without other phases such as  $\text{Sr}_3\text{Ru}_2\text{O}_7$  and  $\text{SrRuO}_3$ , indicating our  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) crystal is high-quality.

Since the ionic size of Fe is smaller than that of Ru, one will expect a shrinkage of the  $c$  axis correspondingly by Fe-doping [19]. Indeed, our XRD measurement shows that the lattice constant  $c$  of the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) single crystal is about 0.12 % shorter than that of the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  single crystal, determined from the most intensity (0014) peak [the inset of Fig. 1(a)].

Fig. 1(b) shows the temperature  $T$  dependence of the magnetization  $M$  of a  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) single crystal measured with a magnetic field of  $H=0.1$  T and different cooling histories [zero field cooling (ZFC) and field cooling (FC)]. The magnetization of a pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  single crystal grown by the same method is also presented in Fig. 1(c).  $M$ - $T$  curve for  $H//ab$  shows a  $T_C$  of about  $\sim 100$  K without distinct irreversibility [Fig. 1(b)]. However,  $M$  for  $H//c$  exhibits only a pronounced peak near  $T_C$  followed by clearly irreversibility below  $\sim 40$  K. Both are completely different compared with the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  [Fig. 1(c)]. The inset of Fig. 1(b) shows the magnetization  $M$  as a function of magnetic field  $H$  measured at  $T=2$  K. For  $H//ab$ ,  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) presents typical FM behavior, as that of pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  with  $H//c$  [inset of Fig. 1(c)]; whereas for  $H//c$ , the moment of  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) shows an abrupt increase at  $\sim 0.8$  T with clearly hysteresis when  $H$  is swept down, resembling the metamagnetic transition observed in pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  with  $H//ab$  [inset of Fig. 1(c)] [5]. Besides, the saturation magnetization for  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) with  $H//ab$  (or  $H//c$ ) is almost the same with that of the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  with  $H//c$  (or  $H//ab$ ). These results show clearly that the magnetic easy direction has been changed from the  $c$  axis to the  $ab$  plane by Fe-doping, though the shrinkage of  $c$  here



is only 0.12%, indicating a close relation between the lattice and spin in the  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  system.

It should be pointed out here that, the structure property of the single crystal used in the magnetization measurement was verified firstly by single-crystal XRD as shown in Fig. 1(a), but a small increase of magnetization at  $\sim 150$  K is still observed, which indicates that there is very likely a small amount of  $\text{SrRuO}_3$  phase in the crystal. The intergrowth is a common problem in synthesizing high-purity large ruthenate single crystals. To make our results more reliable, we use nanosheet rather than bulk to a deep investigation of its magnetic property. For nanosheets with a few tens nanometer in thickness, this intergrowth problem can be avoided effectively. This is because different phases in the  $\text{Sr}_{n+1}\text{Ru}_n\text{O}_{3n+1}$  family have completely different  $R$ - $T$  properties compared with  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  (see, for example: Ref. [20] ( $\text{Sr}_3\text{Ru}_2\text{O}_7$ ); Ref. [6] ( $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ ); Ref. [3] ( $\text{SrRuO}_3$ )). Any other phases in nanosheet samples will give comparable contribution in transport and thus exhibit very different  $R$ - $T$  behavior. We choose nanosheet samples with significant anomaly near  $T_C \sim 100$  K for further study, to ensure that our results are obtained on high purity samples.

## B. Electrical resistance of nanosheet

Figure 2(b) shows the  $T$ -dependent longitudinal resistance  $R$  measured at zero magnetic field of three  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheets with thickness of 12 nm, 21 nm, and 37 nm, respectively. All  $R$ - $T$  curves present very similar metallic behavior, accompanied by a resistive anomaly near  $T_C \sim 105$  K, which can be more clearly

identified from the “ $\lambda$ ”-shaped  $dR/dT$ - $T$  plot shown in Fig. 2(c). In sharp contrast to the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  bulk or nanosheets, where two resistive anomalies at  $T_C$  and  $T_M$  were observed [6, 13] [see also in the inset of Fig. 2(b)], here only one anomaly at  $T_C$  can be identified. These results suggest that the slightly Fe doping has less impact on the FM transition at  $T_C$ , but it suppresses the second magnetic transition at  $T_M$  completely.

### C. Magnetoresistance of nanosheet

Figure 3 shows the  $H$ -dependent  $R$  at various temperatures obtained in a 12 nm-thick  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet with  $H$  applied parallel to the  $ab$  plane [Fig. 3(a)] and the  $c$  axis [Fig. 3(b)]. Other  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheets exhibit very similar  $R$ - $H$  properties as that of the 12 nm-thick sample. As we reported previously in pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheets [14,16], the  $R$ - $H$  curves with  $H//ab$  plane would present a “cusp”-shaped negative magnetoresistance (MR) above  $T_M$ , then transform dramatically into a “valley” behavior with a positive MR below  $T_M$  due to the reorientation of magnetic moments from an in-plane FM order to  $c$  axis [see also in Fig. 4(a)]. Unexpectedly, this phenomenon is not seen in the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet, where only “cusp”-shaped  $R$ - $H$  curves with negative MR effect are observed down to 2 K when  $H//ab$  plane [Fig. 3(a)]. Interestingly, when the direction of the applied magnetic field is changed to the  $c$  direction ( $H//c$ ), the  $R$ - $H$  curves present a “valley” shape in the whole  $T$ -range below  $T_C$  as shown in Fig. 3(b), significantly different to that observed in the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheet [Fig. 4(b)].

We have also measured the temperature dependence of  $R$ - $H$  properties with  $H//ab$  and  $H//c$ , respectively, of the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) bulk single crystal (data not shown), the results are very similar to those observed in the nanosheet. The main difference is that the saturation field  $H_S$  [as indicated in Fig. 3(b)] with  $H//c$  in the bulk is much smaller than that in the nanosheet. For example, the  $H_S$  at 2 K in the bulk is  $\sim 1.2$  T, however, it increases to  $\sim 2.5$  T in the 12-nm thick nanosheet [Fig. 3(b)]. This difference indicates that, similar to the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  [14, 16], the shape anisotropy in  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) still favors spins aligned in the  $ab$  plane.

The above results clearly demonstrate that the easy axis of magnetization is in the  $ab$  plane in the whole temperature range below  $T_C$ , and the second transition at  $T_M$ , which occurs in the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  bulk or nanosheets, has been fully suppressed in the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) due to the lack of spin rotation from the  $ab$  plane to  $c$  direction. We would like to point out here that “disorder effect” induced by Fe-doping might be another reason for the suppression of the second transition. Disorders such as Fe will usually destroy the magnetic order and weak the intrinsic magnetism. However, both the magnetization [Fig. 1(b) and 1(c)] and the  $R$ - $H$  properties show clearly that the Fe-doping here does not weaken the magnetism, but changes the easy magnetic direction from the  $c$  axis to  $ab$  plane, which strongly suggests the “disorder effect” can be excluded. Since the second transition at  $T_M$  is closely related to the competition between shape anisotropy and magnetocrystalline anisotropy in the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheets [14, 16], the Fe-doping caused shrinkage of the  $c$  axis likely results in a suppression of the magnetocrystalline anisotropy so that it is insufficient

to make the in-plane FM order of  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) switching back to the  $c$  direction.

#### D. Planar Hall effect of nanosheet

To assure the conclusion mentioned above, we further probed the in-plane magnetic property by means of planar Hall effect (PHE) [14, 15]. The schematic diagram of the PHE measurement is shown in Fig. 5(a), where the current  $I$  is injected along the [100] crystal direction (i.e.,  $x$  axis), and the magnetic field  $H$  is applied in the  $ab$  plane (i.e.,  $x$ - $y$  plane) with an angle of  $\theta_H$  towards [100]. In this configuration, the planar Hall resistance,  $R_{xy}^p$ , can be written as [21]

$$R_{xy}^p = \frac{k}{d} M^2 \sin 2\theta_M, \quad (1)$$

where  $k$  is a coefficient related to the anisotropic MR,  $M$  is the in-plane magnetization, and  $\theta_M$  is the angle between  $M$  and  $I$ .

Figure 5(b) shows the  $R_{xy}^p$ - $\theta_H$  curves obtained in a 56 nm-thick  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet at different temperatures. When a magnetic field that is higher than the saturation field of the in-plane  $M$ , such as 2 T as representatively shown in Fig. 5(b), is applied, the  $R_{xy}^p$ - $\theta_H$  curve presents a sinusoidal-shaped twofold symmetry as predicted in Eq. (1) with extremes along  $\langle 110 \rangle$  directions, due to the magnetization is fully polarized by the external magnetic field, i.e.,  $\theta_M \sim \theta_H$ . Interestingly, when the field is decreased to 0.2 T, the  $R_{xy}^p$ - $\theta_H$  curves at different temperatures change into a square-wave form with twofold symmetry, accompanied by clearly hysteresis near all  $\langle 100 \rangle$  directions, indicating that the magnetic

polarization no longer linearly follows the external magnetic field, i.e.,  $\theta_M \neq \theta_H$ . The above results suggest that the in-plane FM order in the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet at different temperatures is strongly anisotropic, and the easy axis of the magnetization is along the  $\langle 110 \rangle$  directions. Since this magnetic anisotropy is almost the same as that of the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheet [14], we conclude that the present doping amount of Fe does not change the in-plane magnetic anisotropy of  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$ .

Under this in-plane magnetic anisotropy, “spin-valve”-like switching in  $R_{xy}^p$  with sweeping  $H$  is expected due to the flop of the in-plane magnetization between the easy magnetization directions (i.e.,  $\langle 110 \rangle$ ) [14]. This behavior is demonstrated in Fig. 5(c), which is obtained in a 32 nm-thick sample with an in-plane  $H$  aligned  $\sim 10^\circ$  off the  $[100]$  direction. It is clear seen that  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet presents two significant features: (1) the  $R_{xy}^p$  switching keeps sharp over the entire low temperatures; (2) the turn-off magnetic field (indicated as black arrows) of the switching increases continuously as the decrease of temperatures. These results are in sharp contrast to that observed in the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheet [Fig. 4(c)], where the  $R_{xy}^p$  switching would gradually blur at temperatures below  $T_M$ , accompanied by a decrease of the turn-off magnetic field, due to there is a spontaneous spin-rotation process from the  $ab$  plane to the out-of-plane [14]. These differences indicate that Fe-doping is conducive to retain the magnetic moment in the  $ab$  plane.

### E. Hall effect of nanosheet

As the Fe-doping gives rise to a complete in-plane FM order, an interesting

question is whether the metamagnetic transition survives without the second transition at  $T_M$  when the magnetic field is along the  $c$  direction in the nanosheet. This is because the metamagnetic transition in the pure system was usually considered to be closely related to the second transition previously [8, 11]. To answer this question, we performed Hall effect measurements on  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheets. For a FM sample, both the ordinary Hall effect (OHE) and anomalous Hall effect (AHE) contribute to the Hall effect, the total Hall resistance  $R_{xy}$  can be empirically expressed as [22]

$$R_{xy}=R_0H/d+R_sM_0(H, T)/d, \quad (2)$$

where  $R_0$  and  $R_s$  are, respectively, the ordinary Hall coefficient and anomalous Hall coefficient, and  $M_0$  is the magnetization, which depends on both the temperature  $T$  and the magnetic field  $H$ . Therefore, the anomalous Hall component, i.e., the second term in Eq. (2), can directly reflect the magnetization characteristics of a FM sample with  $H//c$ .

Figure 6 shows the  $H$ -dependent Hall resistance  $R_{xy}$  of a 12 nm-thick  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet measured at various temperatures. To see clearly, the data obtained in the temperature range of 10-80 K are shifted vertically and plotted in Fig. 6(a). All the  $R_{xy}$ - $H$  curves can be described by Eq. (2) as expected. At  $T>140$  K [Fig. 6(b)], only a linear  $H$  dependence of  $R_{xy}$  caused by OHE is observed. With decreasing  $T$  from 140 K, the AHE gradually appears and become dominants the  $R_{xy}$  due to the FM transition, characterized by the ‘knee’ profile of the  $R_{xy}$ - $H$  curve as shown in Fig. 6. In  $50\text{ K}<T<100\text{ K}$ , the  $R_{xy}$ , which is proportional to the magnetization

$M_0$ , increases almost linearly below the saturation field with increasing  $H$ . However, at  $T < 50$  K, a rapid increase of  $R_{xy}$  that reasonably originates from a rapid increase of  $M_0$  in the vicinity of the saturation field is observed [see  $H_c$  indicated in Fig. 6(a)]. This property is very similar to the metamagnetic transition that has been observed for  $H//ab$  below  $T_M$  in the pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  bulk [5] and the  $M$ - $H$  behavior for  $H//c$  of the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) bulk [inset of Fig. 1(b)].

We interpret the metamagnetic transition in the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) for  $H//c$  as follows. At  $50 \text{ K} < T < T_C$ , the magnetic moments in the  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) at the ground state are FM-aligned in the  $ab$  plane but collectively rotate to the  $c$  direction with increasing the perpendicular  $H$ ; at  $T < 50$  K, however, the magnetic moments have been, at least partially, locked into the  $ab$  plane for  $H < H_c$ , and then change their direction to the  $c$  axis for  $H > H_c$ . This picture is supported by the observation of the MR effect shown in Fig. 3(b), where one can see that the “valley” below 50 K gradually changes from a “V”-like shape to a “U”-like shape with clear hysteresis near the saturation field  $H_S$  at low temperatures. Since the MR effect below the critical field  $H_c \sim 2$  T shown in Fig. 3(b) mainly arises from spin scattering, the fact that the MR is almost independent of the perpendicular  $H$  at low temperatures indicates spins are indeed likely locked into the in-plane easy axis, i.e.,  $\langle 110 \rangle$  directions, for  $H < H_c$ . The observation of metamagnetic transition in  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) with  $H//c$  below 50 K clearly suggests that the metamagnetic transition itself in the  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  system is actually less dependent on the second magnetic transition at  $T_M$ , but in fact, closely rely on the spin orientation.

#### IV. SUMMARY

We have investigated the magnetic property of  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ). Results show that the second magnetic transition that observed in pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  bulk or nanosheets is completely suppressed by Fe-doping, and the FM order (along  $\langle 110 \rangle$  directions) remains in the  $ab$  plane in the whole temperature range below  $T_C$ , which is contrasted with that in pure bulk or nanosheet, where the FM order is mostly along the  $c$  direction at low temperatures. This finding clearly demonstrates that the second magnetic transition in  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  is a result of the rearrangement of magnetic moments.

#### ACKNOWLEDGMENTS

The authors would like to thank Professor Lei Zhang in High Magnetic Field Laboratory (Hefei) for help in x-ray diffraction measurements. This work was supported by the National Natural Science Foundation of China (Grants No. 11674323, 11804297, 11774352, 11774305, and U1832214), the support of China Postdoctoral Science Foundation funded project (Grants No.2018M632445), and the International Users of Hefei Science Center of CAS (2016HSC-IU009) and the CAS/SAFEA international partnership program for creative research teams of China. The single crystal growth effort at Tulane was supported by the U.S. Department of Energy under EPSCoR Grant No. DE-SC0012432 with additional support from the Louisiana Board of Regents.





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Figure captions

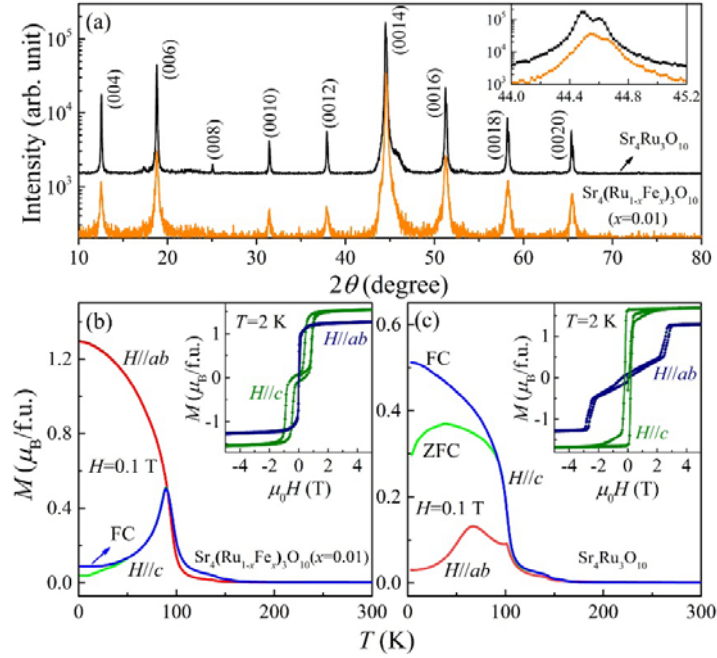


FIG. 1. (a) XRD patterns of a cleaved (001) surface of  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) (orange-line) and  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  (black-line) crystals, grown by floating-zone method. Inset shows the most intensity (0014) peak. Data is shifted vertically for clarity. (b) and (c) are, respectively, the magnetization  $M$  as functions of temperature  $T$  and magnetic field  $H$  (inset, 2K) of a  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) single crystal and a pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  single crystal.

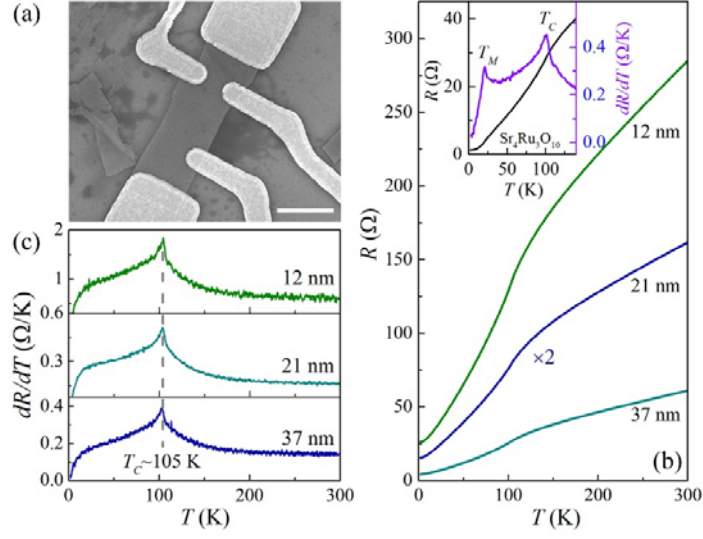


FIG. 2. (a) SEM image of a 12 nm-thick  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet with patterned electrodes. Scale bar: 2  $\mu\text{m}$ . (b) and (c) are, respectively, the temperature  $T$  dependent zero-field resistance  $R$  and  $dR/dT$  of three  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheets with thickness of 12 nm, 21 nm and 37 nm. Inset in (b):  $R$  and  $dR/dT$  versus  $T$  curves of a 34 nm-thick pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheet. Two resistive anomalies at  $T_C \sim 105$  K and  $T_M \sim 25$  K can be identified.

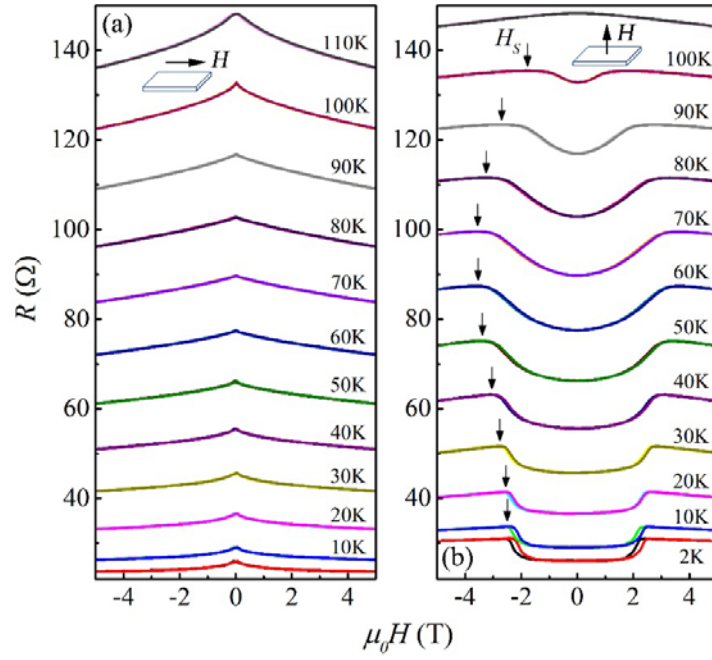


FIG. 3. Temperature dependent  $R$ - $H$  properties of a 12 nm-thick  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet for (a)  $H//ab$  and (b)  $H//c$ , respectively. Arrows indicate the saturation field  $H_s$  of the resistance.

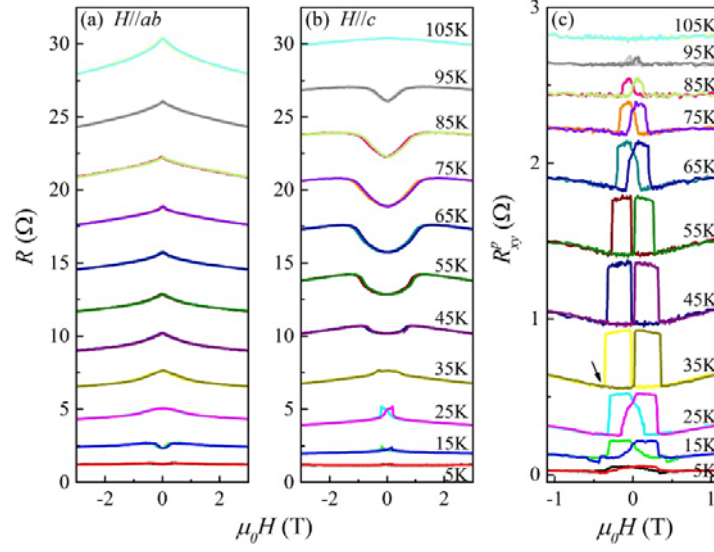


FIG. 4. Temperature dependent  $R$ - $H$  properties of a 34 nm-thick pure  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheet for (a)  $H//ab$  and (b)  $H//c$ , respectively.  $T_M$  of this sample is about 25 K. (c) Planar Hall effect of a 34 nm-thick  $\text{Sr}_4\text{Ru}_3\text{O}_{10}$  nanosheet.  $H$  is aligned near the direction of the applied current, i.e., the  $[100]$  direction. Data is shifted vertically for clarity.

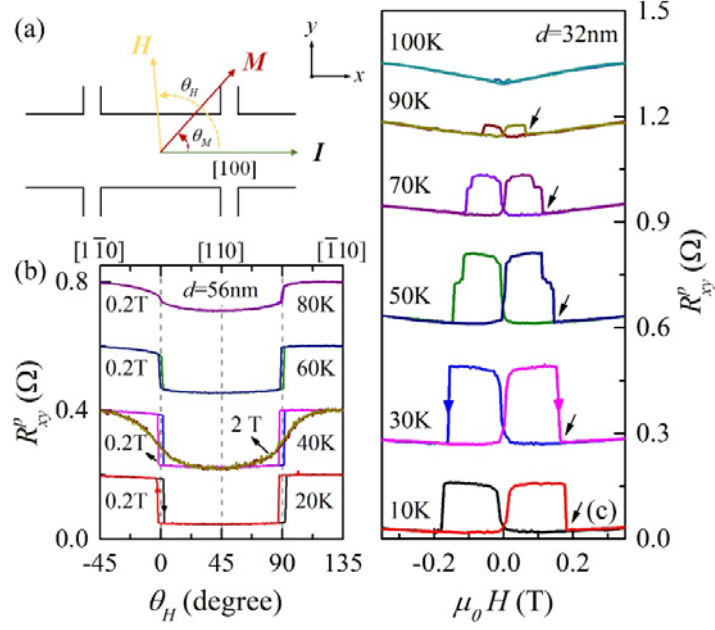


FIG. 5. (a) Sketch of the relative orientations of the applied current  $I$ , the external field  $H$ , and the in-plane magnetization  $M$  of the PHE measurement. (b)  $R_{xy}^p$ - $\theta_H$  curves measured at different temperatures in a 56 nm-thick  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet. (c) Temperature dependent  $R_{xy}^p$ - $H$  curves measured with  $H$  applied about  $10^\circ$  off the  $[100]$  direction in a 32 nm-thick  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet. Curves in (b) and (c) have been shifted vertically for clarity.



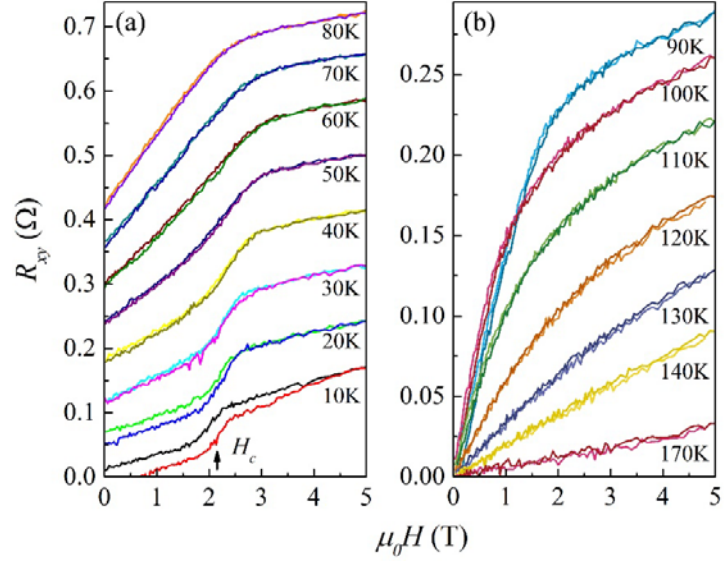


FIG. 6. Hall resistance  $R_{xy}$  of a 12 nm-thick  $\text{Sr}_4(\text{Ru}_{1-x}\text{Fe}_x)_3\text{O}_{10}$  ( $x=0.01$ ) nanosheet as a function of perpendicular magnetic field  $H$  at various temperatures. Data in (a) has been shifted vertically for clarity.