Temperature- and field-driven spin reorientations in triple-layer ruthenate Sr$_4$Ru$_3$O$_{10}$

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Sr$_4$Ru$_3$O$_{10}$, the $n=3$ member of the Ruddlesden-Popper type ruthenate Sr$_{n+1}$Ru$_n$O$_{3n+1}$, is known to exhibit a peculiar metamagnetic transition in an in-plane magnetic field. However, the nature of both the temperature- and field-dependent phase transitions remains as a topic of debate. Here, we have investigated the magnetic transitions of Sr$_4$Ru$_3$O$_{10}$ via single-crystal neutron diffraction measurements. At zero field, we find that the system undergoes a ferromagnetic transition with both in-plane and out-of-plane magnetic components at $T_c \approx 100$ K. Below $T^* = 50$ K, the magnetic moments incline continuously toward the out-of-plane direction. At $T = 1.5$ K, where the spins are nearly aligned along the $c$ axis, a spin reorientation occurs above a critical field $B_c$ giving rise to a spin component perpendicular to the plane defined by the field direction and the $c$ axis. We suggest that both the temperature- and field-driven spin reorientations are associated with a change in the magnetocrystalline anisotropy, which is strongly coupled to the lattice degrees of freedom. This study elucidates the long-standing puzzles on the zero-field magnetic orders of Sr$_4$Ru$_3$O$_{10}$ and provides new insights into the nature of the field-induced metamagnetic transition.

Ruddlesden-Popper type perovskite ruthenates (Sr,Ca)$_{n+1}$Ru$_n$O$_{3n+1}$ are prototypical $4d$ transition-metal oxides which exhibit a variety of intriguing phenomena. The magnetic and electronic properties of these materials are sensitively dependent on the layer number $n$ and the structural distortions induced by substituting Ca for Sr. For instance, the single-layer Sr$_2$RuO$_4$ ($n=1$) shows an unconventional superconducting state$^1$, whereas the ground state of the double-layer Sr$_3$Ru$_2$O$_7$ ($n=2$) is a Fermi liquid and is close to a ferromagnetic instability$^2$. The three-dimensional SrRuO$_3$ ($n=\infty$) is a ferromagnetic metal with a Curie temperature $T_c = 160$ K$^{3,4}$. On the other hand, Ca$_2$RuO$_4$ ($n=1$) is an antiferromagnetic Mott insulator with a Neel temperature $T_N \approx 110$ K$^5$, while Ca$_3$Ru$_2$O$_7$ ($n=2$) exhibits a quasi-two-dimensional metallic behavior and becomes antiferromagnetic below $T_N \approx 56$ K$^6$, and CaRuO$_3$ is a paramagnetic metal$^7$. To date, compared to the intense efforts invested on the single-, double-layer and three-dimensional ($n=1, 2$ and $\infty$) ruthenates, there have been much fewer studies on the triple-layer ($n=3$) compounds.

Sr$_4$Ru$_3$O$_{10}$ crystallizes in an orthorhombic space group $Pbnm$$^{10}$, as shown in Fig. 1(a), which displays interesting but perplexing magnetic properties. From the magnetic susceptibility measurements, it has been reported that Sr$_4$Ru$_3$O$_{10}$ undergoes a paramagnetic-ferromagnetic transition at $T_p = 105$ K, below which the easy axis is along the $c$ direction$^{10}$. Intriguingly, while an additional transition has been found in the magnetic susceptibility at $T^* = 50$ K$^{10,11}$, no anomaly is revealed in the specific heat measurements$^{42}$. Although several distinct scenarios have been proposed to account for the feature at $T^*$, its intrinsic character remains an open question. On the one hand, below $T^*$ a canted antiferromagnetic structure with a ferromagnetic component along the $c$ axis and an antiferromagnetic component in the $ab$ plane has been proposed$^{14,15}$. Nevertheless, no evidence of antiferromagnetism has been observed via neutron diffraction experiments$^{12,13}$. On the other hand, previous neutron diffraction measurements have yielded contradictory results regarding the direction of the ferromagnetic moments in Sr$_4$Ru$_3$O$_{10}$14,15. It is initially argued that spins are aligned in the $ab$ plane based on the observation of (0 0 $L$)-type magnetic Bragg peaks and no anomaly has been found at $T^*$. As a result, the feature at $T^*$ in magnetic susceptibility measurements is ascribed to a magnetic domain process$^1$. In contrast, a distinct magnetic easy axis

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has been proposed more recently, where the magnetic moments are determined to be along the $c$ axis since the $(0\ 0\ L)$-type magnetic Bragg peaks are found absent. In addition, a kink-like feature has been reported in the temperature dependence of the Bragg reflection $(2\ 0\ 0)$ at $T^*$, which is also claimed to be observed in all other reflections\textsuperscript{15}. Therefore, the easy axis of the magnetic moments and the nature of the anomaly at $T^*$ of Sr$_4$Ru$_3$O$_{10}$ remain elusive.

Another intriguing property of Sr$_4$Ru$_3$O$_{10}$ is the field-induced metamagnetic transition. While a typical magnetic hysteresis loop of ferromagnets is observed as the magnetic field is applied along the $c$ axis, a first-order metamagnetic transition is seen below $T^*$ for the field applied in the $ab$ plane\textsuperscript{10,11}. Experimental signatures of the electronic phase separation during the phase transition have been observed\textsuperscript{16}. Nevertheless, previous neutron diffraction studies reveal controversial features regarding this metamagnetic transition. Across the critical field, while no anomaly has been observed in the field dependence of the magnetic reflection $(0\ 0\ 8)$ in ref.\textsuperscript{14}, a field-induced transition at the same wave vector is reported in ref.\textsuperscript{15}. In addition, it has been reported that the metamagnetic transition is accompanied by a change in the lattice, as evidenced in Raman scattering\textsuperscript{13}, neutron diffraction\textsuperscript{15} and magnetostriction studies\textsuperscript{17}, which suggests the presence of strong spin-lattice coupling in this system. To date, the nature and the origin of this metamagnetic transition are yet to be resolved.

In this paper, we present a single-crystal neutron diffraction study of the magnetic order and the metamagnetic transition in Sr$_4$Ru$_3$O$_{10}$. We find that the material orders ferromagnetically at $T_c \approx 100$ K without any signature of antiferromagnetic components, in agreement with the previous studies\textsuperscript{14,15}. However, the magnetic moments are found to possess both in-plane and out-of-plane components below $T_c$, in contrast to the previous neutron diffraction studies where the spins are proposed to align either in the $ab$ plane\textsuperscript{14} or along the $c$ axis\textsuperscript{15}. In addition, we have observed spin reorientations below $T^* \approx 50$ K, where the magnetic moments incline toward

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**Figure 1.** (a) The crystal structure of Sr$_4$Ru$_3$O$_{10}$. Sr, Ru and O atoms are represented by the green, gray and red balls, respectively. (b) The magnetic structure of Sr$_4$Ru$_3$O$_{10}$. Only the magnetic Ru ions are shown. Note that both moment size and spin direction cannot be uniquely determined due to the lack of enough Bragg peaks with reasonably good magnetic intensity in the measurements.
the out-of-plane direction. Furthermore, we show that while the magnetic moments are nearly along the \( c \) axis at \( T = 1.5 \) K, a magnetic field applied along the in-plane \([1 -1 0]\) direction gives rise to a spin reorientation at a critical field \( B_c \), above which there exists a spin component perpendicular to the plane defined by the field direction and the \( c \) axis, in accordance to the metamagnetic transition. This observation is distinct from the previous studies, where the metamagnetic transition is ascribed to magnetic domain processes\(^{14}\) or a coexistence of zero-field \( c \)-axis component and the field-induced in-plane spin polarization\(^{15}\). Both temperature- and magnetic-field-driven spin reorientations are presumably ascribable to the change in the magnetocrystalline anisotropy that is strongly correlated with the RuO\(_6\) octahedral distortion. Our results reconcile the inconsistency among previous neutron diffraction studies on the magnetic easy axis of Sr\(_4\)Ru\(_3\)O\(_{10}\), resolve the puzzling character of the transition at \( T^* \), and elucidate the nature of the field-induced metamagnetic transition.

Results

We start with the zero-field neutron diffraction data collected at the HB-3A four-circle diffractometer. Figure 2(a) and (b) present the rocking curves of Bragg reflections \((0 0 2)\) and \((1 1 1)\) at \( T = 8, 50 \) and 100 K, respectively. Note that both reflections meet the requirements of nuclear Bragg diffraction. The intensity of \((0 0 2)\) shows a nonmonotonic behavior, which is significantly enhanced at 50 K compared to that measured at 8 K and 100 K; in contrast, the \((1 1 1)\) intensity becomes stronger with decreasing temperature. These observations suggest that there is magnetic intensity superimposed on the nuclear Bragg peaks. To further elucidate the nonmonotonic temperature dependence of the magnetic intensity, Fig. 2(c) and (d) present the peak intensity of \((0 0 2)\) and \((1 1 1)\) reflections as a function of temperature, respectively. Upon cooling, additional reflection intensity in both peaks emerges below 100 K. Similar enhancement has been observed in other nuclear Bragg peaks including \((0 0 6)\) and \((0 0 8)\), indicating the development of a ferromagnetic order below \( T_c = 100 \) K which is in line with the magnetic susceptibility measurements\(^{10}\). Consistent with Fig. 2(a), the intensity of \((0 0 L)\)-type magnetic reflections, \( L = 2, 6 \) and 8, also exhibits nonmonotonic temperature dependence, with a maximum at \( T^* \approx 50 \) K that is reminiscent of the anomaly at \( T^* \) observed in the magnetic susceptibility data\(^{16,11}\). In accordance to Fig. 2(b), the magnetic intensity at \((1 1 1)\) emerges at \( T_c \) and continues to increase below \( T^* \), which suggests a spin reorientation at \( T^* \). On the one hand, the observation of \((0 0 L)\)-type magnetic reflections is in agreement with the previous neutron diffraction experiments reported in ref.\(^{14}\), but is different from that in ref.\(^{15}\) where they are found to be absent. On the other hand, we have revealed distinct temperature-dependent behaviors of the intensity of \((0 0 L)\) and \((1 1 1)\), which is in sharp contrast to the previous studies where either no anomaly was observed\(^{14}\), or a kink feature was claimed to exist in the temperature dependence of all reflections\(^{15}\). Note that the HB-3A diffractometer is equipped with a two-dimensional neutron detector, from which the change in the scattering angle \( 2\theta \) of the...
diffracted neutrons at different temperatures is found too small to be resolved due to the instrumental resolution limitation. This is in agreement with a change in the lattice constants of only ~0.04% upon cooling observed in the previous studies\(^1\). Therefore, the variation in the peak intensity of these Bragg peaks cannot be ascribed to the structural change, but is magnetic in origin.

In order to explore the possible magnetic configurations in Sr\(_4\)Ru\(_3\)O\(_{10}\), we carried out the magnetic representation analysis using the program SARA\(_{\text{H}}\)\(^1\). The space group of the crystal structure is No. 55 Pbam and the propagation vector is \(k = (0 0 0)\). There are four inequivalent Ru atoms in a chemical unit cell which are located at Ru1 (0 0 0), Ru2 (0 0 0.1402), Ru3 (0.5 0 0.3598) and Ru4 (0.5 0 0.5). The analysis shows that an in-plane antiferromagnetic component would give rise to strong magnetic reflections at (1 0 0), (1 0 1) or (0 1 1), which, nevertheless, are all absent in our measurements. Therefore, the feature at \(T^*\) in the magnetic susceptibility cannot be ascribed to the formation of antiferromagnetic components in the \(ab\) plane that was claimed previously\(^1,13\).

As a result, the only symmetry-allowed magnetic configurations in Sr\(_4\)Ru\(_3\)O\(_{10}\) is the ferromagnetic structure with the magnetic moments either along the \(c\) axis or in the \(ab\) plane. Since neutrons couple to the magnetic moment perpendicular to the momentum transfer \(q\), the observation of magnetic intensities at (0 0 \(L\)), \(L = 2, 6\) and 8, at \(T^* < T < T_c\) suggests the development of a magnetic order with spins aligned in the \(ab\) plane. This, combined with the magnetic susceptibility measurements which show an easy axis closer to the \(c\) axis, suggests that the magnetic moments have both in-plane and out-of-plane components in this temperature regime. Note that at \(T^* < T < T_c\), the intensity of (0 0 2) increases more rapidly than that of (1 1 1), which implies that the magnetic moments incline toward the \(ab\) plane, though the easy axis is always closer to the \(c\) axis. Below \(T^*\), the decrease in the intensity of (0 0 \(L\))-type magnetic peaks suggests that the in-plane ferromagnetic moment reduces and the spins incline continuously toward the out-of-plane direction upon cooling. This speculation is supported by the observation of an increase in the reflection intensity of (1 1 1) Bragg peaks below \(T_c\) which was suggested by the \(\alpha\) axis in the field-induced magnetic transition, which is consistent with the previous study that the magnetic easy axis as a function of temperature. This feature is reminiscent of the widely studied spin reorientation

### Discussions

The observation of a spin reorientation at \(T^*\) in Sr\(_4\)Ru\(_3\)O\(_{10}\) at zero field has elucidated the long-standing puzzle on the anomaly in magnetic susceptibility measurements\(^6\). This spin reorientation suggests a change in the magnetic easy axis as a function of temperature. This feature is reminiscent of the widely studied spin reorientation
transitions in rare earth magnets and orthoferrites\textsuperscript{21}, which are ascribed to the change in the magnetic anisotropy constants as a function of external parameters, such as temperature, magnetic field and pressure, etc\textsuperscript{22}. Our finding raises an intriguing question: What is the underlying mechanism responsible for the change in the magnetic easy axis in Sr$_4$Ru$_3$O$_{10}$? It is known that in Ruddlesden-Popper type ruthenates, the magnetic anisotropy is strongly coupled to the structural distortions of the RuO$_6$ octahedra. For instance, magnetocrystalline anisotropy has been extensively investigated in SrRuO$_3$ thin films, where the magnetic anisotropy and the saturated magnetic moments can be readily tuned by controlling the RuO$_6$ octahedral distortion via the epitaxial strain imposed by substrates\textsuperscript{23}. Considering the fact that the ferromagnetic state with $T_c \approx 100$ K in triple-layer ($n = 3$) Sr$_n$Ru$_3$O$_{3n}$ bridges the physics of the double-layer ($n = 2$) Sr$_n$Ru$_3$O$_{3n}$ (Fermi liquid, ferromagnetic instability)\textsuperscript{24} and the three-dimensional ($n = \infty$) SrRuO$_3$ (ferromagnetic, $T_c = 160$ K)\textsuperscript{24}, the change in the magnetocrystalline anisotropy in Sr$_4$Ru$_3$O$_{10}$ across $T^*$ may arise from the corresponding changes in lattice structures. Indeed, anomalies

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**Figure 3.** (a,b) Field dependence of the peak intensity of (0 0 2) and (1 1 1) reflections at $T = 1.5$ K. (c,d) $\theta$-2$\theta$ scans across (0 0 2) and (1 1 1) Bragg reflections at $B = 0$ and 3.5 T, $T = 1.5$ K. Inset shows the $\theta$-2$\theta$ scan over (0 0 16) peak at $B = 0$ and 3.5 T, $T = 1.5$ K.

**Figure 4.** $\theta$-2$\theta$ scans across (0 0 2) and (1 1 1) Bragg reflections at $B = 0$ and 3.5 T, $T = 50$ K.
in the lattice constants with $c$ expanding while $a$ contracting below $T^*$ at zero field have been observed previously\cite{15,17}. These changes in lattice parameters are expected to alter the occupancy of the Ru $t_{2g}$ orbitals in this multiband system, which consequently affects the magnetic anisotropy through spin-orbit coupling\cite{25}.

The nature of the in-plane field-induced metamagnetic transition in Sr$_4$Ru$_3$O$_{10}$ below $T^*$, which has been an unresolved puzzle, can now be readily understood. It is also due to a spin reorientation where the magnetic moments reorient from the nearly $c$ axis toward the $ab$ plane. Intriguingly, we find that there exists a magnetic moment component perpendicular to the plane defined by the field direction and the $c$ axis above the critical field, which is consistent with the observation of a recent study where it has been ascribed to the Dzyaloshinskii-Moriya interaction assuming that the easy axis is along the $c$ axis\cite{26}. On the other hand, it is worth noting that across the field-induced metamagnetic transition below $T^*$ the lattice parameter $a$ expands while $c$ shrinks\cite{27}, a trend which is opposite to that observed upon cooling through $T^*$ at zero field discussed above\cite{15,17}. This suggests that the magnetic anisotropy is altered above the critical field due to the strong spin-lattice coupling, which needs to be taken into account in the future theoretical modeling.

In summary, the magnetic structure and the metamagnetic transition in the triple-layer ruthenate Sr$_4$Ru$_3$O$_{10}$ ($n = 3$) are revisited by neutron diffraction measurements. The magnetic order below $T_1$ is found to be ferromagnetic with both in-plane and out-of-plane spin components, while it evolves toward the $c$ axis below $T^* = 50$ K. This finding elucidates the long-standing puzzle of the nature of the magnetic transition at $T^*$ in Sr$_4$Ru$_3$O$_{10}$. Below $T^*$, upon the metamagnetic transition with a magnetic field applied along the in-plane [1 − 1 0] direction, the magnetic moments undergo a spin reorientation from the $c$ axis toward the $ab$ plane with a spin component perpendicular to the plane defined by the magnetic field direction and the $c$ axis. Both temperature- and field-induced spin reorientations can be attributed to the change of magnetocrystalline anisotropy via spin-orbit coupling due to the change in the Ru$_3$O$_6$ octahedral distortion.

Methods

The single-crystal Sr$_4$Ru$_3$O$_{10}$ was grown by the floating zone method\cite{28}. The lattice constants are $a = 5.5280\ \AA$, $b = 5.5260\ \AA$ and $c = 28.651\ \AA$. The phase and quality of the crystal have been verified by x-ray diffraction measurements. The magnetization as a function of both temperature and magnetic field has been measured using SQUID and the results are in agreement with previous studies\cite{10,11}. A very small amount of intergrowth of SrRuO$_3$ has been revealed in the magnetic susceptibility measurements, which is easy to be separated in the neutron diffraction measurements due to its very different lattice parameter $c$ ($c = 7.8446\ \AA$ in orthorhombic unit cell). Zero-field and field-dependent neutron diffraction measurements were performed using the HB-3A four-circle diffractometer ($\lambda = 1.5426\ \AA$) and the HB-1A triple-axis spectrometer ($\lambda = 2.36\ \AA$) respectively at High Flux Isotope Reactor in Oak Ridge National Laboratory. At HB-3A, the sample was mounted on an aluminum stick and loaded into a closed-cycle Helium displex. At HB-1A, the sample was oriented in the horizontal ($H \perp L$) scattering plane and loaded into a vertical-field cryomagnet such that the magnetic field is applied along the [1 − 1 0] direction. The Bragg reflections ($H \perp L$) were in the reciprocal lattice units $2\pi/a, 2\pi/b$ and $2\pi/c$ of the orthorhombic space group $Pbam$ (No. 55)\cite{29}.

Data availability. The data that support the findings of this study are available from the corresponding author upon reasonable request.

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**Author Contributions**

X.K. conceived the project. P.G.L., Y.W. and Z.Q.M. grew the single-crystal sample. M.Z., H.B.C., W.T., X.K., H.D.Z. and B.D.P. performed neutron diffraction experiments. M.Z., H.B.C., W.T. and X.K. analyzed the data. M.Z. and X.K. wrote the manuscript. All authors commented on the manuscript.

**Additional Information**

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