Size Effect on the Magnetic Phase in Sr$_4$Ru$_3$O$_{10}$

Yan Liu  
*Chinese Academy of Sciences, China*

Jiyong Yang  
*Chinese Academy of Sciences, China*

Weike Wang  
*Chinese Academy of Sciences, China*

Haifeng Du  
*Chinese Academy of Sciences, China*

Wei Ning  
*Chinese Academy of Sciences, China*

See next page for additional authors

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Authors
Yan Liu, Jiyong Yang, Weike Wang, Haifeng Du, Wei Ning, Langsheng Ling, Wei Tong, Zhe Qu, Zhaorong Yang, Minling Tian, Gang Cao, and Yuheng Zhang

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Size effect on the magnetic phase in \( \text{Sr}_4\text{Ru}_3\text{O}_{10} \)

Yan Liu\(^1\), Jiyoung Yang\(^1\), Weike Wang\(^1\), Haifeng Du\(^1,3\), Wei Ning\(^1\), Langsheng Ling\(^1,3\), Wei Tong\(^1\), Zhe Qu\(^1\), Zhaorong Yang\(^1,4\), Mingliang Tian\(^1,5\), Gang Cao\(^1\) and Yuheng Zhang\(^1,4,5\)

\(^1\) High Magnetic Field Laboratory, Chinese Academy of Sciences, Hefei 230031, Anhui, People’s Republic of China
\(^2\) University of Science and Technology of China, Hefei 230031, Anhui, People’s Republic of China
\(^3\) Hefei Science Center, Chinese Academy of Sciences, Hefei 230031, Anhui, People’s Republic of China
\(^4\) Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, People’s Republic of China
\(^5\) Center for Advanced Materials, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506, USA

E-mail: jyyang@hmfl.ac.cn and tianml@hmfl.ac.cn

Keywords: \( \text{Sr}_4\text{Ru}_3\text{O}_{10} \), metamagnetic transition, size effect, Hall effect

Abstract

High quality \( \text{Sr}_4\text{Ru}_3\text{O}_{10} \) nanoflakes are obtained by the scotch tape-based micro-mechanical exfoliation method. The metamagnetic transition temperature \( T_m \text{ flake} \) is found to decrease in line with the decrease of thickness, while the ferromagnetic (FM) phase, the ordinary, and anomalous Hall effects (OHE and AHE) are independent on the thickness of the flake. Analysis of the data demonstrates that the AHE reflects the FM nature of \( \text{Sr}_4\text{Ru}_3\text{O}_{10} \) and the decrease of thickness favors the Ru moments aligned in the \( ab \)-plane, which induces a decrease of the metamagnetic transition temperature compared with the bulk.

1. Introduction

Ruthenium oxide perovskites \( \text{Sr}_n\text{Ru}_n\text{O}_{3n+1} \) \( (n = 1, 2, 3, \infty) \) are strongly correlated materials involving complex interactions between the charge, spin, orbit and lattice degrees of freedom. Their ground states present a rich of exotic physical properties, such as the unconventional spin-triplet superconductivity in \( \text{Sr}_2\text{RuO}_4 \) \( (n = 1) \) [1], the quantum criticality and nematicity in \( \text{Sr}_2\text{Ru}_2\text{O}_7 \) \( (n = 2) \) [2–4], and the spontaneous itinerant ferromagnetism in the infinite layer of \( \text{Sr}_2\text{RuO}_4 \) \( (n = \infty) \) [5, 6]. \( \text{Sr}_4\text{Ru}_3\text{O}_{10} \) is the \( n = 3 \) member of the \( \text{Sr}_n\text{Ru}_n\text{O}_{3n+1} \) family. It belongs to quasi-two-dimensional metal with an orthorhombic unit cell, which is composed of triple layers of corner-shared RuO\(_6\) octahedra separated by double rock-salt Sr-O layer [7]. This member has attracted considerable attention in recent years due to its unique magnetic properties [7–17]. By applying a magnetic field \( H \) \( (0.01 \text{T}) \) along the \( c \)-axis, the temperature dependent magnetization \( M_T(T) \) exhibits a FM transition at a Curie temperature \( T_C \sim 105 \text{ K} \), followed by another sharp transition at temperature \( T_m \text{ bulk} \) of about \( \sim 50 \text{ K} \) with a large irreversibility. In contrast, by applying a field \( (H = 0.01 \text{T}) \) within the \( ab \)-plane, the \( M_{ab}(T) \) displays a weak cusp at \( T_C \) and a pronounced peak at \( T_m \text{ bulk} \) with a much smaller irreversibility, and below about \( 20 \text{ K} \), the \( M_{ab} \) is almost unmeasurable [7, 8]. Interestingly, if the in-plane field reaches a critical field \( H_c \), a rapid increase of \( M_{ab} \) in a narrow range of magnetic field is observed on the \( M-H \) curves at temperatures below \( T_m \text{ bulk} \), and the \( H_c \) increases with the decrease of temperature \( (H_c \sim 2 \text{ K} \sim 2 \text{ T}) \) [8]. This transition is considered to be the metamagnetic transition.

The metamagnetic transition observed in \( \text{Sr}_4\text{Ru}_3\text{O}_{10} \) below \( T_m \text{ bulk} \) contains rich physics associated with the lattice, spin, orbit, and electronic inhomogeneity, which has not been well understood. Firstly, this transition is accompanied by strong spin-lattice coupling [9, 14, 18]. The crystal structure undergoes a significant change when the applied in-plane field crosses over the critical field \( H_c \) [9, 14]. Secondly, the occurrence of the transition is possibly through a phase separation process with magnetic domain formation, for the in-plane magnetoresistivity near \( H_c \) exhibits large hysteresis or multiple ultra-sharp steps at extreme low temperatures \( (< 1 \text{ K}) \) [10, 11, 12]. Thirdly, the angle-resolved magnetization and magnetoresistivity suggest the metamagnetic transition is orbit-dependent, where the Ru 4d\(_{x^2-y^2}\) orbit is responsible for the metamagnetic transition while the 4d\(_{xy}\) orbit is ferromagnetic in the ground state [12, 13]. The understanding of the metamagnetic transition in
Sr$_4$Ru$_3$O$_{10}$ is challenged by recent neutron study, where the Ru moments at zero field and below $T_c$ are found to be FM-aligned along the $c$-axis only, there is no any signature of either long-range antiferromagnetic (AFM) or FM order in the $ab$-plane \cite{14}. However, the neutron data cannot exclude a possibility that the metamagnetism is a field-induced short-range AFM order to FM transition. To date, the magnetic nature of Sr$_4$Ru$_3$O$_{10}$ below $T_m$ bulk is still unclear and remains elusive.

In this work, we present the electrical transport measurements of two mechanically exfoliated Sr$_4$Ru$_3$O$_{10}$ nanoflakes with thickness of 31 nm and 260 nm. It finds that the metamagnetic transition temperature, $T_m$ flake, in the flake is much smaller than the bulk, $T_m$ bulk $\sim$ 50 K, and decreases with decreasing thickness, but its saturation field $H_s$ along the $c$-axis increases with the decrease of thickness, indicating the Ru moment in the thinner sample is more difficult to be aligned along the $c$-direction. However, the FM transition, the ordinary and anomalous Hall effects are independent on the thickness, where the dominant carriers derived from the ordinary Hall coefficient $R_h$ are always hole-type and the anomalous Hall conductivity $\sigma_{xy}^A$ follows the typical scaling law $\sigma_{xy}^A \propto \sigma_{xx} \varphi$ ($\sigma_{xx}$: longitudinal conductivity) with different scaling exponent, $\varphi$ $\sim$ 1, 0 and 1.6, as the increase of temperature $T$. The decrease of the metamagnetic transition temperature thus cannot be attributed to the changes of the unit cell or the electronic structures, but can be understood by the shape anisotropy induced rearrangement of the Ru moments due to the size effect.

2. Experimental

Sr$_4$Ru$_3$O$_{10}$ single crystal is grown by the flux technique \cite{8}. The temperature dependent magnetization of the bulk crystal along the $c$-axis measured under 0.01 T is shown in figure 1 (a), which is well consistent with those reported previously \cite{7, 8}. The characteristic temperatures $T_m$ bulk $\sim$50 K and $T_c$ $\sim$ 105 K of the single crystal are indicated by the arrows. The Sr$_4$Ru$_3$O$_{10}$ flakes are obtained by the scotch tape-based micro-mechanical exfoliation from the bulk single crystal, and then transferred to a silicon substrate covered with 300 nm-thick silicon dioxide on the top of the surface. The thickness, $d$, of the flake is determined by atomic force microscopy. Conventional six terminal electrical contacts are made using electron-beam lithography (EBL) technique. To

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**Figure 1.** (a) Magnetization as a function of temperature measured along the $c$-axis at 0.01 T of the Sr$_4$Ru$_3$O$_{10}$ single crystal under zero- or field cooling condition. (b) SEM image of a Sr$_4$Ru$_3$O$_{10}$ flake ($d$ = 260 nm) with patterned electrodes. (c) and (d) are, respectively, the temperature dependence of the longitudinal resistivity $\rho_{xx}$ of the Sr$_4$Ru$_3$O$_{10}$ flakes with different thickness $d$; the insets are, respectively, the blow-up of the $\rho_{xx}$-$T$ curves in low-$T$ range measured under in-plane fields.
ensure ohmic contacts, we etch the e-beam patterned contact areas for a few seconds by in situ Ar-plasma to remove the possible residual PMMA resist firstly, and then deposit 5 nm Ti and 150 nm Au working as the electrical leads by thermal evaporation. With this process, the successful rate for ohmic contacts is higher than 90%, and the resistance ratio between the contact resistance and the sample is usually less than 15%. The scanning electron microscope (SEM) image of a device with patterned electrode is shown in figure 1(b). The longitudinal resistivity (ρxx) and the Hall resistivity (ρxy) are measured as functions of temperature (T) and external magnetic field (H) by a physical property measurement system (PPMS, Quantum Design). The ρxx is measured by the standard four-probe configuration, where the contact resistance is believed to be negligible. The Hall resistance ρxy is determined from ρxy = [ρxy(H)−ρxy(−H)]/2 in order to subtract the longitudinal component of ρxx arising from the small misalignment of the transverse contacts.

3. Results and discussion

Figures 1(c) and (d) show ρxx versus T curves of two flakes (d = 31 and 260 nm) measured at zero magnetic field. For each flake, the ρxx decreases monotonically as T decreases. The residual resistance ratio, $\text{RRR} = \rho_{xx}(300 \, \text{K})/\rho_{xx}(2 \, \text{K})$, of each flake reaches about 60 (data not shown), indicating the crystals are high quality, where the residual resistivity of both flakes at 2 K is about 3.77 μΩ cm (d = 31 nm) or 4.15 μΩ cm (d = 260 nm). Two anomalies on the resistivity can be clearly identified from the magnetoresistivity (MR) measured under various in-plane magnetic fields H as shown in figure 2, where MR is defined as $\rho_{xx}(H)/\rho_{xx}(0)$. The data is obtained by the zero field cooling from above 160 K to exclude the influence of the field history. The ‘valley’ observed on the MR-T curves is as expected due to the itinerant FM nature of Sr4Ru3O10 [8], which is caused by the suppression of carrier scattering from spin fluctuations under in-plane magnetic field [19]. The Tc is determined to be ~106 K and 101 K for the 31 nm- and 260 nm-thick flakes, respectively, from the negative maximum of the ‘valley’ [19], which is almost independent on the thickness and consistent with the bulk. With decreasing T below Tc, the Sr4Ru3O10 undergoes another transition called the metamagnetic transition at about $T_m^\text{flake} = 25 K$ and 37 K for the 31 nm- and 260 nm-thick flakes, below $T_m^\text{flake}$ the MR shows a positive behavior, while above which the MR is always negative due to the FM nature of Sr4Ru3O10. This feature is consistent with that reported in the bulk [11], except for the $T_m^\text{bulk}$ is almost 20 K lower than the $T_m^\text{flake} \sim 50 K$. It finds that the metamagnetic transition temperature decreases with decreasing the thickness of the flake, while the $T_c$ remains unchanged. Previously, both the electronic and lattice structures have been suggested to be the driving force for the formation of the metamagnetic phase [7, 12–15]. A fact that our sample preparation process does not make any variations of the electronic or lattice structures provides an indication that the metamagnetic transition in Sr4Ru3O10 should have an origin of internal magnetic orders, which maybe changed as the decrease of thickness. To understand the mechanism in detail, we performed the Hall effect study on the flakes.

Figure 3 shows the H-dependence of ρxy at various temperatures, where H is applied perpendicular to the ab-plane of the flake. As a FM nature of Sr4Ru3O10 below $T_c$, both the OHE and AHE make contributions to the total Hall resistivity [20], characterized by the ‘knee’ profile as shown in figure 3. At temperatures far above $T_c$, the
OHE resistivity $\rho_{xy}$ governs the total $\rho_{xy}$ with a linear dependence of $\rho_{xy} = R_0 H$ (e.g. see the dashed line in figure 3(d)), which is due to the orbital effect of $H$ on the carriers, here $R_0$ is the ordinary Hall coefficient. With decreasing $T$, the AHE gradually dominates the $\rho_{xy}$, and the AHE resistivity $\rho_{xy}^A$ reaches a maximum at about 80 K. Then $\rho_{xy}^A$ decreases rapidly with the further decrease of $T$ and becomes extremely small at 10 K but can still be recognized (e.g. see the dashed line in figure 3(c)). In spite of the FM-induced hysteresis behavior in the low field range, the trace of the observed $\rho_{xy}$ at a fixed temperature can be well described by the equation of $\rho_{xy} = R_0 H + \rho_{xy}^A$, which contains both the OHE and AHE components. The ordinary Hall coefficient $R_0$ can be extracted from the high field slope, i.e. $d\rho_{xy}/dH$ for $H = 5$ T, of the Hall isotherm, and the $\rho_{xy}^A$ below $T_c$ can be simultaneously obtained by extrapolating the linear term to the zero field.

The ordinary Hall coefficient $R_0$, as a function of temperature is shown in figure 4(a). $R_0$ is positive in the whole temperature range, indicating the dominant carriers in Sr$_4$Ru$_3$O$_{10}$ are holes. The $R_0$-$T$ curves for both flakes present almost a same ‘S-shaped’-like behavior. Specifically, the $R_0(T)$ shows a maximum near $T_1 \sim 22$ K and a minimum near $T_2 \sim 60$ K below $T_c$, and these kinks show no direct correlation with the magnetic structures, such as the metamagnetic transition near $T_m$ flake and the FM transition at $T_c$. Generally, the temperature dependent $R_0(T)$ in quasi-two-dimensional metal is related to the changes of the wave vector dependent electron mean free path or the reconstruction of the Fermi surface [21, 22]. The almost identical $R_0(T)$ in both flakes implies that the electronic structures are almost independent on the thickness of the flakes as expected. We noticed that both $R_0(T)$ display a similar $T$-dependent feature as the variations of the lattice

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**Figure 3.** Hall resistivity $\rho_{xy}$ of the Sr$_4$Ru$_3$O$_{10}$ flakes as a function of magnetic field $H$ along the $c$-axis at various temperatures. (a), (b) are for $d = 31$ nm; (c), (d) for $d = 260$ nm.

**Figure 4.** (a) The temperature dependence of the ordinary Hall coefficient $R_0$. (b) The scaling relation between the anomalous Hall conductivity $\sigma_{xy}^A$ and the longitudinal conductivity $\sigma_{xx}$. New J. Phys. 18 (2016) 053019 Y Liu et al.
In summary, we have investigated the transport property of two mechanically exfoliated Sr₄Ru₃O₁₀ nanoflakes. The result shows that the change of thickness has little effect on the FM phase, the OHE and AHE, but can modulate the metamagnetic transition temperature $T_m$ significantly, which is about 25 K for the 31 nm-thick flake and 37 K for the 260 nm-thick flake, respectively. The identical Hall effect in the two flakes suggests the decrease of $T_m$ cannot be attributed to the changes of the unit cell or the electronic structure. However, the reason for such change is possibly attributed to the large variation of the structures in Sr₄Ru₃O₁₀ as well. The result shows that the change of thickness has little effect on the FM phase, the OHE and AHE, but can modulate the metamagnetic transition temperature $T_m$ significantly, which is about 25 K for the 31 nm-thick flake and 37 K for the 260 nm-thick flake, respectively. The identical Hall effect in the two flakes suggests the decrease of $T_m$ cannot be attributed to the changes of the unit cell or the electronic structure. However, the reason for such change is possibly attributed to the large variation of the structures in Sr₄Ru₃O₁₀ as well.
we have found that the saturation field along the c-axis in the two flakes increases as the decrease of thickness, indicating the Ru moment is aligned closer to the ab-plane in the thinner flake due to the size effect, and the metamagnetic transition in Sr₄Ru₃O₁₀ is thus caused by a rearrangement of the Ru moments.

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