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

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Abstract

High quality Sr$_4$Ru$_3$O$_{10}$ nanoflakes are obtained by the scotch tape-based micro-mechanical exfoliation method. The metamagnetic transition temperature $T_m$ 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 Sr$_4$Ru$_3$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 Sr$_{n+1}$Ru$_n$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 Sr$_3$Ru$_2$O$_7$ ($n = 1$) [1], the quantum criticality and nematicity in Sr$_3$Ru$_2$O$_7$ ($n = 2$) [2–4], and the spontaneous itinerant ferromagnetism in the infinite layer of Sr$_2$RuO$_4$ ($n = \infty$) [5, 6]. Sr$_4$Ru$_3$O$_{10}$ is the $n = 3$ member of the Sr$_{n+1}$Ru$_n$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 Ru$_6$ 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 T) along the $c$-axis, the temperature dependent magnetization $M(H)$ exhibits a FM transition at a Curie temperature $T_C \sim 105$ K, followed by another sharp transition at temperature $T_m$ of about $50$ K with a large irreversibility. In contrast, by applying a field ($H = 0.01$ T) within the $ab$-plane, the $M_{ab}(T)$ displays a weak cusp at $T_c$ and a pronounced peak at $T_m$ with a much smaller irreversibility, and below about 20 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 $T_m$ and the $H_c$ increases with the decrease of temperature ($H_c \sim 2$ T) [8]. This transition is considered to be the metamagnetic transition.

The metamagnetic transition observed in Sr$_4$Ru$_3$O$_{10}$ below $T_m$ 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 ($< 1$ K) [10, 11, 17]. Thirdly, the angle-resolved magnetization and magnetoresistivity suggest the metamagnetic transition is orbit-dependent, where the Ru 4$d_{x^2-y^2}$ orbit is responsible for the metamagnetic transition while the 4$d_{xy}$ orbit is ferromagnetic in the ground state [12, 13]. The understanding of the metamagnetic transition in
Sr₄Ru₃O₁₀ is challenged by recent neutron study, where the Ru moments at zero field and below Tc 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 [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 Sr₄Ru₃O₁₀ below Tm bulk is still unclear and remains elusive.

In this work, we present the electrical transport measurements of two mechanically exfoliated Sr₄Ru₃O₁₀ flakes with thickness of 31 nm and 260 nm. It finds that the metamagnetic transition temperature, Tm flake, in the flake is much smaller than the bulk, Tm bulk  50 K, and decreases with decreasing thickness, but its saturation field Hs 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 R0 are always hole-type and the anomalous Hall conductivity σxy follows the typical scaling law σxy ∝ σxx (σxx: longitudinal conductivity) with different scaling exponent, φ  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

Sr₄Ru₃O₁₀ single crystal is grown by the flux technique [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 [7, 8]. The characteristic temperatures Tm bulk  50 K and Tc  105 K of the single crystal are indicated by the arrows. The Sr₄Ru₃O₁₀ 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
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 ($\rho_{xx}$) and the Hall resistivity ($\rho_{xy}$) are measured as functions of temperature ($T$) and external magnetic field ($H$) by a physical property measurement system (PPMS, Quantum Design). The $\rho_{xx}$ is measured by the standard four-probe configuration, where the contact resistance is believed to be negligible. The Hall resistance $\rho_{xy}$ is determined from $\rho_{xy} = [\rho_{xy}(H) - \rho_{xy}(-H)]/2$ in order to subtract the longitudinal component of $\rho_{xx}$ arising from the small misalignment of the transverse contacts.

### 3. Results and discussion

Figures 1(c) and (d) show $\rho_{xx}$ versus $T$ curves of two flakes ($d = 31$ nm and $260$ nm) measured at zero magnetic field. For each flake, the $\rho_{xx}$ 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 $\mu\Omega\cdot$cm ($d = 31$ nm) or 4.15 $\mu\Omega\cdot$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 $[\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0)$. 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 Sr$_4$Ru$_3$O$_{10}$ [8], which is caused by the suppression of carrier scattering from spin fluctuations under in-plane magnetic field [19]. The $T_c$ 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 $T_c$, the Sr$_4$Ru$_3$O$_{10}$ undergoes another transition called the metamagnetic transition at about $T_{m,\text{flake}} \approx 25$ 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 Sr$_4$Ru$_3$O$_{10}$. This feature is consistent with that reported in the bulk [11], except for the $T_{m,\text{flake}}$ is almost 20 K lower than the $T_{m,\text{bulk}} \approx 50$ 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 Sr$_4$Ru$_3$O$_{10}$ 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 $\rho_{xy}$ at various temperatures, where $H$ is applied perpendicular to the ab-plane of the flake. As a FM nature of Sr$_4$Ru$_3$O$_{10}$ 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

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 $\sigma_{xx}$. 
parameter $c$ determined by the neutron diffraction study [14], indicating that the variations of $R_{\phi}$ in Sr$_4$Ru$_3$O$_{10}$ might be closely connected to this structural change.

Now we turn to the AHE component and use the universal scaling relation [20], $\sigma_{xy}^A \propto \sigma_{xx}^{\phi}$, to uncover the physical implication below $T_m (\sim 100$ K) in the flakes, where $\sigma_{xy}^A$ and $\sigma_{xx}$ are, respectively, the anomalous Hall and longitudinal conductivity, and $\phi$ is the scaling exponent. Here, $\sigma_{xy}^A$ is estimated by $\sigma_{xy}^A = \rho_{xy}^A / [\rho_{xx}^2 + (\rho_{xy}^A/\rho_{xx})^2]$ since $\rho_{xy}^A$ is at least two orders smaller than $\rho_{xx}$ in our case. As shown in figure 4(b), the $\sigma_{xy}^A$ is also independent on the thickness of the flake, and can be categorized into three regions labeled as I, II and III with an exponent of $\phi \sim 1$.08 in $T < 23$ K ($\sigma_{xx} > 7.5 \times 10^4 \text{ S cm}^{-1}$), $\phi \sim 0$ with $\sigma_{xy}^A = 235 \text{ S cm}^{-1}$ in $23$ K $< T < 55$ K ($\sigma_{xx} < 7.5 \times 10^4 \text{ S cm}^{-1}$) and $\phi \sim 1.61$ or 1.72 in $T > 55$ K ($\sigma_{xx} < 2.2 \times 10^4 \text{ S cm}^{-1}$), respectively. It is well-known that both the extrinsic and intrinsic mechanisms are proposed for the AHE [20], in which the $\sigma_{xy}^A \propto \sigma_{xx} (\phi \sim 1)$ in region I indicates the extrinsic skew-scattering mechanism is dominant based on the asymmetric impurity scattering caused by the spin–orbit interaction (SOI), while $\phi \sim 0$ with a large value of $\sigma_{xy}^A$ in region II suggests the intrinsic Berry-phase contribution governs the AHE [23]. Recently, Onoda et al. [2008] have developed a unified theory of the AHE in ferromagnets and proposed three scaling regions as a function of the carrier lifetime $\tau$ related Born scattering amplitude $h/\tau$ ($h$: Planck constant). For $h/\tau < E_{\text{SOI}} (E_{\text{SOI}}$: spin–orbit interaction energy), $\phi = 1$, the extrinsic skew-scattering arising from the vertex correction yields a dominant contribution; For $h/\tau > E_{\text{SOI}}, \phi = 0$, $\sigma_{xy}^A$ becomes insensitive to the scattering strength because of the intrinsic dissipationless topological Berry-phase contribution. Further increasing $h/\tau$, $\phi = 1.6$ [23, 24]. Usually, it is difficult to observe all the proposed regions in one ferromagnetic sample, possible due to a small variation of $h/\tau$ [20, 25]. The scaling behavior in a Sr$_4$Ru$_3$O$_{10}$ flake can present all the three scaling regions indicates there is a large change of $h/\tau$ in this material. The reason for such change is possibly attributed to the large variation of the structures in Sr$_4$Ru$_3$O$_{10}$ as well [14], for the two crossovers, one at about 23 K and the other at about 55 K, among the three scaling regions match well with the temperatures $T_1$ and $T_2$ shown in figure 4(a). In other words, the AHE reflects the FM nature of the flake below $T_m$, and which is independent on the thickness.

Figure 5 shows the saturation field $H_s$ along the $c$-axis of the two flakes determined from the $\rho_{xy}^A$-$H$ curves (see figure 3), where the inset represents an enlarged $\rho_{xy}^A$-$H$ curve of the 260 nm-thick flake measured at 40 K, and the $H_s$ is 0.65 T indicated by the arrow. The $H_s$ increases as the decrease of thickness at a fixed temperature, indicating that the magnetic moments in the thinner flake are more difficult to be polarized along the $c$-axis. In other words, the shape anisotropy caused by the decrease of thickness favors the magnetic moments aligned in the $ab$-plane (possibly arranged to be a weak FM order). This assignment is consistent with the observation of the metamagnetic transition $T_m^{\text{flake}}$ decreases with the decrease of thickness (see figure 2), where the in-plane magnetic order stabilized by the reduced thickness will suppress the metamagnetic phase.

4. Conclusions

In summary, we have investigated the transport property of two mechanically exfoliated Sr$_4$Ru$_3$O$_{10}$ 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 $T_m^{\text{flake}}$ 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 $T_m^{\text{flake}}$ cannot be attributed to the changes of the unit cell or the electronic structure. However,
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 Sr$_4$Ru$_3$O$_{10}$ is thus caused by a rearrangement of the Ru moments.

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