Topological Signatures in the Hall Effect of SrRuO$_3$/La$_{0.7}$Sr$_{0.3}$MnO$_3$ SLs

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Noncollinear spin textures such as skyrmions lead to a novel Hall effect contribution called the topological Hall effect. In recent years, research has focused on the Weyl metal SrRuO$_3$ with topological signatures reported for SrRuO$_3$ layers adjacent to layers with strong spin–orbit coupling. However, SrRuO$_3$ films are known to be prone to structural modifications when interfaced with different materials. In this work the Hall effect of SrRuO$_3$/La$_{0.7}$Sr$_{0.3}$MnO$_3$ superlattices (SLs) is correlated with structural and magnetic information to study whether a topological Hall effect is induced by interfacing to ferromagnetic layers. High-quality SLs with ultrathin layers are fabricated by pulsed laser deposition on SrTiO$_3$ substrates. The symmetry of the SrRuO$_3$ layers is studied by angular magneto-resistance measurements and the magnetocrystalline anisotropy by magnetization measurements. Two SLs are compared in detail, both with 8 unit cell thick SrRuO$_3$ layers, but with La$_{0.7}$Sr$_{0.3}$MnO$_3$ layers 2 and 4 unit cell thick. Only the sample with 2 unit cell thick manganese layers shows features resembling a topological Hall effect. Depending on the La$_{0.7}$Sr$_{0.3}$MnO$_3$ layer thickness the SrRuO$_3$ crystalline symmetry is orthorhombic or tetragonal. The results indicate that the topological Hall effect features arise from an intricate interplay between magnetocrystalline anisotropy and antiferromagnetic interlayer coupling.

1. Introduction

Topological phenomena in condensed matter systems have attracted intense research activities in recent years.[1,2] Since the Hall effect is directly related to the Berry curvature in momentum space,[3,4] measurements of the Hall resistivity provide a direct means to investigate topologically protected spin structures. It was shown that the presence of skyrmions induces a novel contribution to the Hall resistivity,[5–7] called the topological Hall effect (THE). In the field of oxides, SrRuO$_3$ (SRO) provides an interesting playground to study topological effects, since it was quite early realized that SrRuO$_3$ is a Weyl metal with a monopole-like Berry curvature in reciprocal space.[8,9] There have been attempts to tune the Berry curvature by interfacing SrRuO$_3$ to layers with large spin–orbit interaction,[10,11] to ferroelectric BaTiO$_3$,[12,13] and to ferromagnets.[14–16] In all these cases topological signatures in the Hall resistance were observed, but direct evidence for the formation of topologically protected spin structures such as skyrmions[17] is scarce. Moreover, THE signatures were also observed in single SrRuO$_3$ films[18–21] and were interpreted as arising from the presence of skyrmions[18–19] but could alternatively also be related to sample inhomogeneities.[20,21] Since SrRuO$_3$ is known to be sensitive to strain[22] and structural distortions leading to changes in crystalline symmetry[16,23] and to modified oxygen octahedra rotation patterns,[24] individual heterostructures have to be carefully studied to elucidate the mechanism driving topological-like Hall effect phenomena.

In this work the influence of interfacing SrRuO$_3$ to La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) is investigated. This study goes beyond previous work on bi- and trilayers[14] by using superlattices (SLs) and varying the thickness of the LSMO layers. A dramatic influence of the LSMO layer thickness on the magnetic field dependence of the Hall resistivity was observed, especially the appearance of phenomena indicating a THE. These phenomena are related to the structural symmetry of the SRO layers leading to changes in the magnetocrystalline anisotropy as well as to the antiferromagnetic interlayer coupling and the appearance of magnetic compensation.

In the next section a brief overview of Hall effect measurements on SrRuO$_3$ is given, followed in Sections 3 and 4 by a presentation of results on LSMO/SRO SLs. The article is concluded with a discussion in Section 5.

2. Brief Review

2.1. General Properties

SrRuO$_3$ is a conducting oxide and an itinerant ferromagnet with a Curie temperature of 160 K and a magnetic moment per Ru ion...
of $1.6 \mu_B^{[25,26]}$. In bulk, it crystallizes in the orthorhombic phase (space group Pbnm) with lattice parameters $a = 0.55670$ nm, $b = 0.55304$ nm, and $c = 0.78446$ nm (pseudocubic lattice constant $a_0 = 0.3923$ nm).\textsuperscript{[27,28]} SrRuO$_3$ films grown on cubic SrTiO$_3$ (STO) (001) substrates crystallize in the orthorhombic structure if the films are not too thin, with the [110] direction along the substrate normal and the [110] and [001] directions along the in-plane cubic axes of the STO substrate.\textsuperscript{[29]} The long-range crystallographic orientation of the film is achieved by growth on vicinal substrates with terraces oriented along one cubic substrate direction, since the SRO $c$-axis [001] aligns with the terraces. The magnetocrystalline anisotropy of SRO films leads to a magnetic hard axis along [001] and a magnetic easy axis lying in the (001) plane at an angle of about $25^\circ$ from the [110] direction toward the [100] axis.\textsuperscript{[30]} In comparison, in SRO crystals the [001] axis is magnetically hard and the [100] axis is the magnetic easy axis.\textsuperscript{[31]} On certain substrates, such as DyScO$_3$ (110)\textsuperscript{[32,33]} and GdScO$_3$ (110),\textsuperscript{[34]} and in SrRuO$_3$/manganite SLs,\textsuperscript{[35]} SrRuO$_3$ films grow in a tetragonal phase with the tetragonal [001] axis along the substrate normal. In the tetragonal structure the magnetic hard axis is along the $[\overline{1}00]$ direction.\textsuperscript{[23,36]} As a rule of thumb, a good and easily measurable indicator for the crystalline symmetry of SRO films is a magnetization measurement at low temperatures with a magnetic field applied along the substrate normal. In case of orthorhombic SRO, this shows an easy axis hysteresis loop with sizable coercivity (often of the order of 1 T), whereas tetragonal SRO films are characterized by a nearly reversible magnetization loop characteristic for a magnetization rotation process toward a hard axis. However, there might be exceptions to this rule.\textsuperscript{[37]} There are indications that very thin SRO films grown on STO (001) substrates\textsuperscript{[38]} and very thin SRO layers in SRO/STO SLs grown on STO (001)\textsuperscript{[39]} have tetragonal symmetry. The orthorhombic and tetragonal structures and their orientation with respect to the SrTiO$_3$ substrate are shown in Figure 1.

![Figure 1](image_url)

**Figure 1.** Images of the a) orthorhombic and b) tetragonal SrRuO$_3$ structure viewed along the directions of the cubic axes of the SrTiO$_3$ substrate. The black coordinate axes show the SrTiO$_3$ crystal axes directions, with the colored arrows indicating the directions of the SrRuO$_3$ layers in SRO/STO SLs grown on STO (001).\textsuperscript{[23,36]} The images were drawn using the software VESTA.\textsuperscript{[41]}

### 2.2. Anomalous Hall Effect and Weyl Points

The Hall effect of ferromagnets contains a contribution from the ordinary Hall effect proportional to the magnetic induction $B$ with the Hall constant $R_H$, a contribution from the anomalous Hall effect (AHE) that is proportional to the magnetization component $M_z$ perpendicular to the film, with a Hall constant $R_A$, and—possibly—a contribution arising from the presence of topologically protected spin structures called the THE $\rho_{yx,T}$. It is generally assumed that these contributions add up to the total Hall resistivity

$$\rho_{yx} = R_H B + \mu_B R_A M_z + \rho_{yx,T}. \quad (1)$$

In case of crystalline materials this equation is an approximation in certain instances, since it neglects the dependence of the AHE on the magnetization direction.\textsuperscript{[42]} This article is focused on assessing whether a THE contribution $\rho_{yx,T}$ occurs for these particular LSMO/SRO SLs. Because spin structures such as skyrmions are often not directly visible in the magnetization curves, $\rho_{yx,T}$ is not proportional to the measured magnetization. Therefore, indications for topological Hall contributions are detected by deviations between the field dependence of anomalous Hall resistivity and the field dependence of magnetization. Skyrmions might appear during the remagnetization process after magnetic field reversal. $\rho_{yx,T}$ is thought to be proportional to the density $n_s$ of skyrmions with a flux quantum $\Phi_0$, to the spin polarization $P$ and the ordinary Hall constant $R_H$\textsuperscript{[5,6,43]}

$$\rho_{yx,T} = PR_H n_s \Phi_0. \quad (2)$$

Spin polarization values $P$ for SrRuO$_3$ of $-10\%^{[44]}$ and $55\%^{[45]}$ were reported. If skyrmions appear at all in the remagnetization process, their density might be related to $\delta M/H$. This suggests that the topological Hall contribution has maxima near the coercive fields. In the literature this is sometimes referred to as a "hump structure."\textsuperscript{[10]}

Typical measurements of the Hall resistivity as a function of a magnetic field applied perpendicular to a 40 nm thick SRO film are shown in Figure 2a. The Hall resistivity is proportional to the magnetic field at high fields, which is attributed to the ordinary
Hall effect. At lower fields a hysteresis loop is seen that is rather similar to the magnetization loop (compare the 10 K data for the same film in Figure 2c). Therefore, we attribute the hysteretic contribution to the AHE. The product $\rho_{yx,A} = \mu_0 R_A M_S$ was estimated by extrapolation of the high field line to zero field, as shown in Figure 2a. This yields the anomalous Hall resistivity values for the sample V40 in Figure 3. As the Hall resistivity data in Figure 2a and c do not show any humps, a topological Hall contribution is absent. The magnetoresistance (MR) of the SRO film in the perpendicular field is shown in Figure 2b. It is negative for all fields and, in this temperature range, decreases with increasing temperature.

Data of the anomalous Hall resistivity $\rho_{yx,A}$ of various SRO films are compiled in Figure 3. The data labeled V40, V5, and M were obtained by extrapolation as described in the previous paragraph; the data labeled HK50, HK6, and HK3 were measured in zero field after saturating the magnetization perpendicular to the film. In the case of thick films $\rho_{yx,A}$ approaches zero at low temperatures, since the resistivity is small and the carrier scattering is weak. At a sample-dependent compensation temperature $T_S$ the anomalous Hall resistivity changes sign, from negative values at low to positive values at high temperatures. The product $R_A M_S$ vanishes above the Curie temperature due to the vanishing of $M_S$; the anomalous Hall constant $R_A$, however, increases with increasing temperature. The sign change of the anomalous Hall constant is intriguing, because in general a power-law scaling of the extrinsic anomalous Hall constant $R_A$ with the resistivity $\rho: R_A \propto \rho^n$ with $n = 1$ for skew scattering[50,51] and $n = 2$ for side-jump scattering[52] is expected. The sign change was interpreted as arising from an intrinsic AHE[53] that is related to the Berry-phase mechanism. In modern terminology, SrRuO$_3$ is a Weyl metal[9] having Weyl nodes in its band structure. The Berry phase acquired by an electron moving on a closed trajectory around a Weyl node is equal to the surface integral over the Berry curvature that acts as a virtual magnetic field and has the form of a monopole field close to such a degeneracy.[8,54] A phenomenological scaling relation taking Berry-phase and side-jump contributions into account captures the resistivity dependence of the anomalous Hall constant of SrRuO$_3$ quite well.[47]

The previous results are valid for the orthorhombic SRO phase. In contrast, the Hall effect of tetragonal SRO films is positive.[10,37,55,56] Qualitatively, this might be understood by a shift of the Fermi energy with respect to the location of the nodes in the band structure. It appears, however, that the corresponding theoretical explanations are in the initial stages.[57]

2.3. Possible Topological Hall Effect at the SrRuO$_3$/SrIrO$_3$ Interface

A hump structure in the AHE of SrRuO$_3$ films adjacent to SrIrO$_3$ (SIO) layers was reported in Matsuno et al.[10] This appeared for film thicknesses below 6 unit cells and was attributed to a topological Hall contribution due to the formation of a skyrmion phase. The latter was thought to arise from an interfacial Dzyaloshinskii–Moriya interaction induced by the broken inversion symmetry at the interface in combination with the strong spin–orbit coupling in the SrIrO$_3$ layer. Both the anomalous and topological Hall effects in SrRuO$_3$/SrIrO$_3$ bilayers can be
modulated by electric fields. Similar electrically tunable hump structures of the Hall resistivity were also reported for the SrRuO$_3$/BaTiO$_3$ interface and were related to skyrmion formation.\cite{12}

We have studied these phenomena in asymmetric [SrIrO$_3$/SrRuO$_3$/SrZrO$_3$]$_n$ SLs with six repetitions and layer thickness of 2 unit cells for SrIrO$_3$ and SrZrO$_3$ and 6 unit cells for SrRuO$_3$\cite{59} and present data here to show the demon field dependence of the Hall effect in this system. The Hall effect of these SLs is very similar to that of orthorhombic films, but close to the compensation point at which the anomalous Hall constant changes sign, Hall resistivity loops with hump structure were observed (Figure 4).

Direct evidence for the presence of magnetic skyrmions in SrRuO$_3$ is scarce. High-resolution magnetic force microscopy (MFM) measurements on SrIrO$_3$/SrRuO$_3$ bilayers showed the presence of magnetic bubble domains during the remagnetization processes close to the coercive field, but also clear evidence for the existence of small circular domains with radii of about 10 nm or smaller.\cite{17} The stray-field contrast of the bubble domains indicated that the domain walls were Néel walls with clockwise orientation. Assuming that the wall characteristics of the small bubble domains are the same, the MFM imaging indeed hints at the existence of skyrmions with Néel texture in SrRuO$_3$. In another MFM study of a SrRuO$_3$/SrIrO$_3$/SrZrO$_3$ trilayer with 4 to 5 unit cell thick SrRuO$_3$ layers, the formation of skyrmions was however not observed, although the AHE resistivity loops showed the specific hump structure.\cite{60}

Groenendijk et al.\cite{11} studied the Hall effect of STO/SRO/STO, SIO/SRO/SIO and STO/SRO/SIO heterostructures and found a positive AHE in SIO/SRO/SIO, a negative AHE in STO/SRO/STO, and a compensation point with accompanying hump structures in STO/SRO/SIO. They interpreted the data as tuning of the Berry curvature in SRO by the adjacent SIO layer. In the mixed system STO/SRO/SIO the crossover from negative to positive AHE with different switching fields then generates hump structures without the need of invoking skyrmions.

2.4. Hump Structures in Single SrRuO$_3$ Films

Apart from inducing hump structures in the Hall resistivity by coupling to ferromagnetic and ferroelectric layers and layers with large spin–orbit interaction, such structures already appear in single SRO films, provided these are sufficiently thin. Qin et al.\cite{18} report hump structures for 3–6 nm thick SRO films and Sohn et al.\cite{19} for 4–5 unit cell thick SRO films, in both cases grown on STO (001). The hump structures appear close to the compensation point $T_S$ of the anomalous Hall resistivity. The interpretation was within the skyrmion framework.

Kan et al.\cite{20} observed hump structures in 3 nm thick SRO films grown on GdScO$_3$ and associated those with the formation of Ru vacancies. In a companion paper, Kan and Shimakawa\cite{21} found similar structures in 3.5 nm thick SRO films on NdGaO$_3$. Since in both cases the hump structures appeared close to the compensation point $T_S$, an alternative interpretation based on inhomogeneities was proposed. These inhomogeneities lead to a distribution of compensation temperatures across the film and therefore the measured Hall resistivity might arise from a superposition of contributions with positive and negative AHE. Numerical modeling yielded Hall resistivities similar to the measured curves.\cite{21}

Concluding this brief review, apart from the skyrmion hypothesis, there are three different interpretations invoking heterogeneity: 1) in the compensation temperature driven by defects,\cite{20,21} 2) in the Hall resistivity sign driven by a tuning of the Berry curvature,\cite{21} and 3) in the Hall resistivity sign driven by structural modifications.\cite{16} These interpretations must not be mutually exclusive, but the structural, electronic, and magnetic properties of SrRuO$_3$ are sufficiently complex that different features might dominate in different heterostructures.

3. Experimental Section

LSMO/SRO SLs were made by pulsed-laser deposition at a substrate temperature of 650 °C in an oxygen partial pressure of 0.14 mbar. The growth was on vicinal SrTiO$_3$ (001) substrates with a low miscut angle of about 0.1°. Substrate surfaces were atomically flat with terraces 100 to 500 nm wide and separated by unit-cell-high steps. The substrates had a uniform TiO$_2$ termination. Two samples were chosen for this work; both consisted of 15 bilayers with a SRO layer thickness of 8 unit cells; the LSMO layer thicknesses were 2 and 4 unit cells (Table 1).

Bulk La$_{0.7}$Sr$_{0.3}$MnO$_3$ is a double-exchange ferromagnet with a rhombohedral crystal structure (space group R3c, lattice constant $a_0 = 0.5471$ nm, $a = 60.43^\circ$, pseudocubic lattice constant $a_c = 0.3894$ nm,\cite{28} a Curie temperature of 370 K, and a magnetic moment of $3.7 \mu_B$ Mn$^+$).\cite{61} LSMO films grown on STO (001) substrates are cubic with a tetragonal distortion.

The samples were characterized by X-ray diffractometry and atomic force microscopy measurements and further by high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM), electron energy loss spectroscopy (EELS), and energy dispersive X-ray (EDX) mappings in a TITAN 80–300 FEI microscope (300 keV energy of the primary electrons). From the satellite reflections in the X-ray pattern the bilayer thickness $t_b$ was determined, as shown in Table 1.

![Figure 4. Anomalous Hall resistivity of an asymmetric SrIrO$_3$/SrRuO$_3$/SrZrO$_3$ SL near the compensation temperature of 81.6 K at which the anomalous Hall resistivity at high fields vanishes.](image-url)
agree with the SL periods as determined from HAADF-STEM (shown in the second column of Table 1). Further the mean lattice constant $a$ was calculated \[^{[62]}\] and is also shown in Table 1; this is equal to the SRO value.

Magnetoresistance measurements were performed with the SLs mounted on a rotatable stage with an angular resolution better than 0.1°. The measurements were performed in a He-flow cryostat equipped with an 8 T superconducting solenoid. The measurements were made in van der Pauw configuration. Magnetization measurements were performed in a SQUID magnetometer (Quantum Design) with the magnetic field applied either in-plane or along the SL normal. The diamagnetic contribution from the substrate was subtracted; the magnetization was normalized to the volume of the SL.

### 4. Results

#### 4.1. Structure

Figure 5 shows HAADF-STEM micrographs of both samples imaging the interfaces between the LSMO and SRO layers. The image shows coherent interfaces. As the scattering intensity depends monotonically on the atomic number, the metal elements can be clearly identified; the element assignments are shown in Figure 5b by spheres of variable color and diameter. An intensity scan along the oblique line is displayed in the lower inset. It appears that along the growth direction the SRO layers are predominantly terminated by SrO and the LSMO layers by La$_{0.7}$Sr$_{0.3}$O. However, EDX mapping of the interfaces revealed a slight intermixing of Mn and Ru with the presence of Mn in the first SRO monolayer and the presence of Ru in the first LSMO monolayer. Furthermore, the stoichiometry of the interfacial La$_{0.7}$Sr$_{0.3}$O and SrO layers might vary from that of the bulk. A SrO termination of the SRO layers is consistent with observations on SRO single films \[^{[63]}\]. Overall the HAADF-STEM studies showed excellent quality of the SLs. Further extensive transmission electron microscopy (TEM) characterization was presented elsewhere \[^{[37,64,65]}\].

#### 4.2. Magnetization

For a basic characterization of magnetic parameters, Figure 6 shows the temperature-dependent magnetization of the two samples under field cooling (FC) in an applied field of 0.1 T. In both samples...
cases the manganite layers order ferromagnetically below 300 K and the ruthenate layers below 140 K; the latter ordering leads to a decrease of the global magnetization, since the layers are antiferromagnetically coupled. Because the magnetic moments per unit cell are different, in SL 4/8 the manganite layers and in SL 2/8 the ruthenate layers dominate the global magnetization; in the latter case this even leads to a sign change at low temperatures. In the case of SL 4/8 the in-plane components of both SRO and LSMO magnetization are larger than the perpendicular-to-plane components. This is consistent with a collinear magnetization state with magnetic hard axis along the surface normal. In the case of SL 2/8 the LSMO magnetization component is still small in the perpendicular-to-plane direction, but the SRO magnetization component is larger along the [110] direction than in the other directions. This shows that the magnetization state is not collinear but that spin canting is present, the latter being in agreement with polarized neutron reflectometry data and with magnetization measurements on LSMO/SRO bilayer samples with rather thick layers.

As a new feature, the LSMO layers in SL 2/8 acquired a sizable in-plane uniaxial anisotropy; see the differences in the LSMO magnetization contribution in Figure 6a for the [110] and [001] directions. The uniaxial anisotropy might be due to the antiferromagnetic coupling but is present in the SRO paramagnetic phase. However, whereas the [001] direction is magnetically hard in the SRO layers, it is the magnetic easy axis in the LSMO layers (Figure 6a), so it is not simply the SRO hard axis direction being transferred by antiferromagnetic coupling into the LSMO layers. Furthermore, the anisotropy is unlikely to be due to strain, as the TEM images show heteroepitaxial growth and a fully strained SL adapted to the substrate lattice parameter.

To extend the conclusions on the magnetic anisotropy, hysteresis loops were measured; Figure 7 shows magnetization loops measured at 10 K for magnetic fields applied along various crystallographic directions. In case of SL 2/8 the shapes of the low field loops below 3 T are consistent with orthorhombic symmetry: the largest squareness is obtained in a perpendicular magnetic field along [110], as this is a direction close to the easy axis. In comparison, when the magnetic field is applied in-plane along [001], the loop is stretched out, characteristic of a magnetic hard axis. In large magnetic fields a considerable magnetization rotation is observed due to the strong antiferromagnetic interlayer coupling.

The hysteresis loops of SL 4/8 look distinctly different, with a magnetically hard axis along the SL normal (Figure 7b). In the perpendicular field the magnetization is nearly reversible with the magnetization reversal process being dominated by magnetization rotation from an in-plane to a perpendicular-to-plane orientation. This perpendicular hard axis indicates a structural change of the SRO layers, either with the orthorhombic c-axis along the SL normal or with a change to a tetragonal structure. This will be further discussed in Section 4.4. The in-plane loop shows hysteresis with a low- and a high-field magnetization loop related to the ferrimagnetic layer state at low and the in-plane rotation of SRO and LSMO magnetization at high fields.

4.3. Hall Resistivity

Before discussing the Hall resistivity it is elucidating to have a look at the longitudinal resistivity as shown in Figure 8a. In this figure the resistivities of the SLs are compared to the resistivities of pure SRO and LSMO films. Overall, the temperature dependence of the SL resistivities and the SRO film is quite similar. This indicates that the conductance is dominated by the SRO layers. The single films are rather thick with 5 and 15 nm thickness and in the case of these thick films the resistivity ratio between SRO and LSMO is only between 1.5 and 5 (Figure 8b). However, these values might drastically overestimate the conductivity of the LSMO layers in the SLs, as it is well-known that LSMO films with a thickness of only a few unit cells have much higher resistivities. In contrast, La0.7Ba0.3MnO3 layers embedded in SRO have a rather high conductivity. It is therefore difficult to estimate the resistivity of the LSMO layers in the SLs a priori. In any case, in a first approximation, transport in the SLs can be modelled by parallel conduction in the SRO and LSMO layers. In zero field, when the Hall resistivities vanish, the SL resistivity is calculated from the parallel circuit formula.

Figure 6. Magnetization of SLs a) 2/8 and b) 4/8 as a function of temperature. Measurements were made during FC in a magnetic field of 0.1 T applied along the indicated directions.

Figure 7. Magnetization hysteresis of SLs a) 2/8 and b) 4/8 as a function of applied magnetic field at 10 K.
which yields a SL resistivity smaller than the resistivity of the SRO layers alone. However, as shown in Figure 8a, the measured SL resistivities were higher than the single SRO film resistivity. This clearly shows that there are additional scattering mechanisms—interfacial scattering and increased scattering in near-interface regions with intermixing—active in the superlattices that are not taken into account in the parallel model. From these considerations it is not possible to deduce the current density distribution over the different layers.

In view of topological effects it is most instructive to first search for an uncharacteristic magnetic field dependence of the Hall resistivity, since the THE is often revealed by additional Hall resistivity extrema\cite{10,20,21} that do not appear in the magnetization curves. Figure 9 shows an overview of the measured Hall resistivity of SLs 4/8 and 2/8. The data of sample 4/8 are untypical for SRO epitaxial films grown on STO substrates, since 1) the normal Hall effect is positive and 2) the field dependence is characteristic of a magnetic hard axis along the SL normal. However, there are no indications of any topological signatures. In contrast, the data of sample 2/8 are more characteristic of SRO, with a negative high field slope and a clear central hysteresis loop. Moreover, at 100 K an unconventional low field peak appears in the Hall resistivity that might be related to some unconventional Hall effect mechanism.

As already shown in Figure 2 the saturated anomalous Hall resistivity $\rho_{yx,A}$ was determined by extrapolation and is shown in Figure 10. $\rho_{yx,A}$ of SL 4/8 is negative at all temperatures. This is consistent with the negative $R_A$ of SRO at low temperatures and a superposition of the positive $R_A$ of SRO at higher temperatures with the dominating negative $R_A$ of LSMO at higher temperatures. For comparison the data of a SL with 20 unit cell thick SRO layers are shown; in that case $\rho_{yx,A}$ is dominated by the anomalous Hall constant of the SRO layers with a small negative contribution above 150 K from the LSMO layers. In contrast, the saturated Hall resistivity of SL 2/8 is anomalous, since it has a rather low compensation.
temperature and a positive value in a rather wide temperature range between 35 and 115 K. Especially intriguing is the sharp jump of the saturated Hall resistivity $\rho_{yx,A}$ above 100 K. The comparison of the saturated Hall resistivity with the Hall resistivity measured on FC in $-0.05$ T (see open symbols in Figure 10) shows agreement up to 50 K but large deviations at higher temperatures, even with different signs of the Hall effect. This sign change is certainly related to the different magnetization orientations in the temperature sweep and in the field sweeps. As the global magnetization vanishes between 85 and 95 K, depending on the direction of the applied field (Figure 6a), the appearance of unconventional Hall resistivity loops in this temperature regime is certainly related to this magnetization compensation.

A detailed comparison between anomalous Hall resistivity and magnetization data is shown in Figure 11 for SL 4/8 and in Figure 12 for SL 2/8. In the case of sample 4/8 the field dependence of the magnetization and Hall resistivity nicely agrees below 2 T, with deviations at higher fields due to the high field slope of the magnetization that is due to spin canting close to the interfaces. This agreement is consistent with the collinear magnetization structure. In contrast, the Hall resistivity loops of SL 2/8 significantly deviate from the field dependence of the magnetization. Whereas at 10 K there is a certain correspondence, at the other temperatures the Hall resistivity has hump-like structures just after field reversal, at 50 and 100 K close to the coercive fields as measured by the magnetization loops. The exact field dependence of these humps, however, is different from the topological Hall signatures seen in SRO/SIO bilayers and in our SrIrO$_3$/SrRuO$_3$/SrZrO$_3$ SLs; compare Figure 4.

In Figure 13 the magnetization states in SL 2/8 are probed by anomalous Hall resistivity measurements in a magnetic field applied out-of-plane along [110] and in-plane along $\frac{1}{2}$110 and [001]. For the interpretation of these curves one should keep in mind that the anomalous Hall resistivity at 10 and 75 K is dominated by the SRO layers and at 200 K by the LSMO layers. As at 10 and 75 K the Hall resistivity values at the switching fields are very similar (apart from the [001] direction at 10 K), the Hall resistivity in in-plane fields is dominated by the Hall effect and not the planar Hall effect, since the latter is due to anisotropic MR and has a different magnitude. Therefore, the Hall resistivity can be used to monitor the perpendicular magnetization component, when the magnetic field is changed along certain crystallographic directions. At 10 K, in out-of-plane fields, the standard SRO Hall loop was observed; in an in-plane field along [001] the Hall resistivity is very small, since a magnetic field up to 8 T does not rotate the magnetization vector significantly out of the (001) plane. A magnetic field applied along [110] tilts the magnetization from its near-perpendicular direction in low fields toward [110], thus reducing the anomalous Hall resistivity

![Figure 11. SL 4/8: comparison of anomalous Hall resistivity (left axis) and magnetization (right axis) at a) 10 K, b) 50 K, c) 100 K, and d) 150 K.](image1)

![Figure 12. SL 2/8: comparison of anomalous Hall resistivity (left axis) and magnetization (right axis) at a) 10 K, b) 50 K, c) 100 K, and d) 150 K.](image2)

![Figure 13. SL 2/8: Hall Resistivity measured for magnetic fields applied in the indicated directions at temperatures of a) 10 K, b) 75 K, and c) 200 K. For the [110] direction the ordinary Hall effect was subtracted.](image3)
in high fields. The striking observation is the appearance of a full magnetization loop for magnetic fields applied along [110]. This switching would not occur for a pure magnetization rotation process. It does occur in this case because the SRO easy axis is tilted away from the [110] direction such that it is energetically more favourable for the magnetization to reorient by 180° when the magnetic field is cycled. At 75 K such a switching is also seen for a field applied along [001], albeit at much higher switching fields. This magnetization switching might also be aided by the spin canting due to the antiferromagnetic coupling.

4.4. Anisotropic Magnetoresistance

Measurements of the anisotropic magnetoresistance (AMR) were performed between 10 and 150 K. Here, only 10 K data are presented, as these show MR features induced by the magnetocrystalline anisotropy of the SRO layers most clearly. Generalizing the standard AMR definition, the MR was defined as \( \text{MR} = (R(\theta) - R(0))/R(0) \). Figure 14 shows the MR of sample SL 2/8 measured with the magnetic field rotating in two mutually orthogonal planes. The left panels reveal a pronounced jump at an angle of about –120°. This jump is caused by an abrupt movement of the magnetization vector when the magnetic field vector is swept across the direction of a magnetic hard axis. This feature is characteristic for orthorhombic SRO layers (compare Ziese et al. and Kan et al.[70,71]). In the [001] plane a magnetic easy axis and a magnetic hard axis are located; the easy axis is inclined with respect to the SL normal by about 30° and the hard axis is perpendicular to it.[31,36,70] The symmetry of the AMR proves the symmetry of the SRO layers in SL 2/8 to be orthorhombic.

Figure 15 shows the anisotropic MR of sample SL 4/8 measured at 10 K. The angular dependence of this sample is completely different from that of sample SL 2/8. Large hysteresis appears around \( \theta = 0° \), i.e., around the SL normal. Furthermore, the curve shape and magnitude of the magnetoresistance are identical (within experimental error) for the angular sweeps in Figure 15a,c and Figure 15b,d. This shows that the corresponding rotation planes have equivalent symmetry; i.e., the SRO layers do certainly not have the same orientation with respect to the substrate as SL 2/8. There are three obvious possibilities for the symmetry of the SRO layers in this SL: 1) tetragonal symmetry[71] with the tetragonal c-axis along the normal and the [100] direction rotated by 45° relative to the [100], substrate edge; 2) orthorhombic symmetry with the orthorhombic c-axis along the normal; and 3) twinned orthorhombic SRO layers with [001] axes along the two principal substrate axes and strong anti-ferromagnetic coupling to the LSMO layers forcing the magnetization (which is nearly compensated but with a slightly stronger LSMO component) in-plane. Orthorhombic SRO layers with a perpendicular c-axis would probably show strong a-b-twinning, such that the tetragonal and orthorhombic structure cannot be distinguished by MR measurements. As the c-axis is the magnetic hard axis, these results are consistent with the magnetization results of Section 4.2.

The anisotropy and antiferromagnetic coupling can be further studied by magnetic field–dependent MR measurements, as shown in Figure 16 for SL 2/8 and in Figure 17 for SL 4/8. In both figures the left panels present MR data recorded in perpendicular fields (note again the different orientations of the SRO layers), the right panels in parallel fields. Although in perpendicular orientation the current density is always perpendicular to the magnetic field, SL 2/8 showed a striking MR anisotropy (Figure 16a–c), whereas in this configuration the MR of SL 4/8 is isotropic or only weakly anisotropic (Figure 17a–c). This is consistent with the angle-dependent MR measurements and the assignment of the SRO layer orientation with respect to the substrate.

Since the interpretation of the MR data of SL 4/8 is more evident, the data in Figure 17 are discussed in detail first. In the perpendicular field at 10 and 150 K the curves are reversible...
and are therefore characteristic of a magnetization rotation process in which the magnetization is rotated from the SL plane toward the magnetically hard SL normal. At 100 K irreversibility occurs, especially at the highest fields; this is reminiscent of a spin-flop transition. The MR curves in parallel fields are much more complex (Figure 17d–f), certainly reflecting the antiferromagnetic coupling between LSMO and SRO magnetization. At low fields, very clearly seen by the sharp peaks at 100 K, the coupled magnetic moments are remagnetized, followed at higher fields by a canting of the SRO and LSMO magnetization vectors with respect to one another. In both SRO and LSMO the transverse resistance is positive and the longitudinal resistance negative.\(^{[70,72]}\) The slope change in the MR curves at 5 T and 10 K and 4 T and 100 K in Figure 17d,e might therefore be interpreted as a crossover from longitudinal (transverse) MR characteristic at low fields to transverse (longitudinal) MR characteristic at high fields in the transverse (longitudinal) configuration. Because in this SL the LSMO magnetization is dominant, such a crossover might occur when the canting angle becomes smaller than 45°, under the assumption that the LSMO magnetization is along the magnetic field direction. At 100 K the canting is nearly reversible; at 10 K, however, it is related to magnetic domain formation, since the irreversibility persists up to high fields of at least 7 T. A similar crossover was observed in SL 2/8 at 4 T and 75 K (Figure 16b), but in this case in the out-of-plane MR. This is again consistent with the notion that the easy axis of this SL is close to the normal direction.

Concluding this section, the analysis of the MR has revealed different orientations of the SRO layers in the SLs. The respective differences in the magnetocrystalline anisotropy lead to different magnetization states, with the antiferromagnetically coupled LSMO and SRO magnetic moments in-plane in SL 4/8 and mainly out-of-plane in SL 2/8.

5. Discussion and Conclusions

The Hall effect of two LSMO/SRO SLs with layer thicknesses of 2/8 and 4/8 unit cells was measured and was compared to results from magnetometry and MR. The Hall effect hysteresis curves of the two SLs are strikingly different, although the SRO layer thicknesses are the same and the LSMO layer thicknesses change only from 2 to 4 unit cells: SL 4/8 had a standard AHE, whereas SL 2/8 showed signatures of a THE in the temperature range between 50 and 125 K. At the middle of this temperature range the global SL magnetization is compensated for. AMR measurements clearly showed different magnetocrystalline anisotropies of the SLs, consistent with a SRO layer orientation with normal [110].
in SL 2/8 and with normal [001] in SL 4/8. This might be further related to a tetragonal structure of the SRO layers in SL 4/8 (compare Ziese et al. [16]).

The Hall resistivity of SL 2/8 at 100 K measured in magnetic fields along [110] and [001] is shown in detail in Figure 18. It is striking that the low field Hall resistivity is larger than in attainable magnetic fields up to 8 T. If the Hall resistivity is exclusively due to the AHE, this would mean that the SRO magnetization has a larger perpendicular magnetization component in zero field than in finite fields. At the top of Figure 18 the layer magnetization directions are indicated schematically for the different field regimes. In fields above 4 T (shaded areas) only a magnetization rotation process was observed that rotates the layer magnetization continuously toward the field direction. Near zero field the antiferromagnetically coupled magnetization vectors must be mainly perpendicular to the layers, probably oriented along the easy axis of SRO. The irreversibility in the curves indicates magnetic domain formation. The Hall resistivity minimum between 4 and about 0.5 T might be understood by a rotation of the magnetization vectors toward the layers. It is, however, far from clear why such a rotation should occur.

It is instructive to inspect the corresponding MR curves in more detail (see Figure 19 for the 100 K data). On first glance the curves appear rather standard: field sweeps starting at high fields generate maxima at the coercive fields when the magnetization switches direction. Coercive fields always have opposite polarity to the maximum fields at which the sweeps began. All four MR curves, however, show resistance dips (see blue arrows in Figure 19) at fields with the same polarity as the maximum fields at which the sweeps started. This can occur in ferromagnetic heterostructures with inverted hysteresis loops [64], but the hysteresis curves in Figure 7 prove SL 2/8 has normal magnetization loops. The field range between the MR dip and the MR maximum at the coercive field (between ±0.4 and ±0.5 T for fields along [110] and between ±0.2 and ±0.8 T for fields along [110]) matches the field ranges of maximum Hall resistivity in Figure 17. As this field range is limited by clear magnetoresistance structures, we interpret the Hall resistivity in this regime as arising from the nucleation of very specific domain patterns. It is tempting to relate these to skyrmion-like structures [17], but we do not have evidence from magnetic imaging.

In summary, careful studies of the magnetotransport and magnetic properties of LSMO/SRO heterostructures strongly indicate the formation of magnetic domain patterns that induce the THE. These occur near a compensation temperature of the global magnetization when the magnetocrystalline anisotropy is appropriately adjusted. This could be shown in this work for a LSMO/SRO SL with layer thicknesses of 2 and 8 unit cells, but similar effects were also found for LSMO/SRO bi- and trilayers [14] and for asymmetric LSMO/SRO/STO and LSMO/SRO/BTO SLs [15]. These heterostructures are synthetic ferrimagnets with noncollinear spin structure. The findings are therefore similar to the observation of a THE in the noncollinear phase of the antiferromagnet Mn$_2$Si$_3$ [73,74] and to theoretical results on antiferromagnetic skyrmion crystals [75].

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Conflict of Interest

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Keywords

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