Magnetic interlayer coupling between ferromagnetic SrRuO$_3$ layers through a large spin-orbit coupling SrIrO$_3$ spacer

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A key element to tailor the properties of magnetic multilayers is the coupling of the individual magnetic layers. In case of skyrmion hosting multilayers, coupling of skyrmions across the magnetic layers is highly desirable. Here the magnetic interlayer coupling was studied in epitaxial all-oxide heterostructures of ferromagnetic perovskite SrRuO$_3$ layers separated by spacers of the strong spin-orbit coupling oxide SrIrO$_3$. This combination of oxide layers is being discussed as a potential candidate system to host Néel skyrmions. The magnetic coupling between the SrRuO$_3$ layers was assessed using superconducting quantum interference device magnetometry for magnetization loops and for first order reversal curves (FORC). The FORC measurements proved to be useful to distinguish between individual magnetic switching processes and to disentangle the signal of soft magnetic impurities from the samples signal. The type of interlayer coupling can also be assessed with FORC. The interlayer coupling strength estimated from shifts of magnetization major and minor loops was weak for all the heterostructures, with SrIrO$_3$ spacers between 2 monolayers and 12 monolayers thick. No major change occurred with increasing the thickness of the spacer, despite the expectation that the SrIrO$_3$ layer would undergo a transition from insulating to metallic upon thickness increase.

I. INTRODUCTION

Successful integration of magnetic multilayers in applications, such as magnetic memory devices or sensor heads, requires detailed knowledge of the magnetic properties and the characteristics of the magnetization reversal of the system [1]. Of particular interest is the control of the coupling of the magnetic layers to realize for instance sufficient layer-decoupling for applications in magnetic tunnel junctions [2–4], to enable the design of artificial antiferromagnetic systems [5, 6], or to stabilize columnar magnetic skyrmions in specific heterostructures [7], such as Pt/Co/Ir superlattices [8] or Co/Pd multilayers [9].

Recently, the ferromagnetic perovskite oxide SrRuO$_3$ attracted attention due to the proposal of the formation of Néel-type skyrmions when it is interfaced with the large spin-orbit coupling 5$d$ oxide SrIrO$_3$ [10–12]. While the existence of topologically non-trivial textures in SrRuO$_3$-based thin films remains controversial [13–21], epitaxial heterostructures composed of ferromagnetic oxide layers with perpendicular magnetic anisotropy separated by non-magnetic spacers are of general interest due to their versatility in modifying the type and strength of the magnetic interlayer coupling for instance through variation of the spacer thickness or the interactions at the epitaxial interfaces of dissimilar oxides [2, 22].

Strong ferromagnetic coupling of the SrRuO$_3$ layers was realized by introducing a 4 monolayers (MLs) thick metallic LaNiO$_3$ spacer and weak ferromagnetic coupling was observed for the separation of the SrRuO$_3$ layers by 2 MLs of LaNiO$_3$ [23]. Insulating SrTiO$_3$ spacers, 1.6 nm to 2.5 nm thick, were found to result also in weak coupling or in magnetic decoupling of two epitaxial SrRuO$_3$ layers [2, 24]. The interlayer coupling was little investigated for SrRuO$_3$ layers interfaced with SrIrO$_3$, although several studies focused on the discussion of the origin of unconventional features in the magnetotransport of SrRuO$_3$/SrIrO$_3$ heterostructures and multilayers [13–19]. In our previous study [25], the interlayer coupling between SrRuO$_3$ layers separated by an asymmetric spacer of the strong spin-orbit coupling oxide SrIrO$_3$ and the large band gap insulator SrZrO$_3$ was addressed. Weak ferromagnetic coupling was observed with enhanced coupling strength for the reduction of the total spacer thickness from 0.8 nm to 0.4 nm [25]. For symmetric SrRuO$_3$/SrIrO$_3$ multilayers with 2 MLs thick SrIrO$_3$, theoretical calculations by Esser et al. predicted that a ferromagnetic coupling between the SrRuO$_3$ layers is more favorable than an antiferromagnetic type of coupling [26].

Here the magnetic interlayer coupling was investigated for heterostructures in which the SrRuO$_3$ layers were separated by SrIrO$_3$ spacers of various thickness, by means of superconducting quantum interference device (SQUID) magnetometry (full and minor hysteresis loops) and first order reversal curve measurements (FORC). The FORC method has proven to provide valuable information in many different systems that is inaccessible for conventional magnetometry measurements. For example, microstructural information without actual lateral resolution in microstructured and model magnetic sys-

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tems [27–30], information about coercive and interaction field distribution in permanent hard magnetic systems [31–33], as well as interaction strength and interaction type between different magnetic components in systems [27, 33] can be achieved. Performing minor hysteresis loops and FORC measurements enabled us to quantify the type and strength of the magnetic interlayer coupling between the SrRuO₃ layers for various SrIrO₃ spacer thicknesses. For the heterostructure with only 2 MLs SrIrO₃ spacer, the minor loops showed a small positive shift with respect to the major hysteresis loops above 30 K, indicating that the coupling turned weakly antiferromagnetic. However, the estimated coupling strength of about 7 µJ/m² at 40 K led to the conclusion that the two SrRuO₃ layers switch their magnetization almost independently. The spacer material SrIrO₃ is a paramagnetic semimetal in its bulk form [34]. However, metal-insulator transitions can be induced in SrIrO₃ thin films by the reduction of the film thickness to below 4 MLs [35] and by tailoring of epitaxial strain [36, 37]. In our current study, it is therefore expected that the SrIrO₃ undergoes a MIT so that the influence of the SrIrO₃ conductivity on the interlayer coupling could be addressed. We increased the SrIrO₃ spacer thickness to 6 MLs and further to 12 MLs, expecting this to result in improved conductivity of the spacer. It turned out that the coupling strength did not increase upon the increase of the spacer thickness and the two SrRuO₃ layers stayed decoupled.

II. SAMPLE DESIGN AND EXPERIMENTAL METHODS

For investigating the type and strength of the magnetic coupling of the ferromagnetic SrRuO₃ layers, a set of heterostructures with two ferromagnetic SrRuO₃ layers of distinct thicknesses was designed. To make use of the thickness dependence of the coercive field $H_c$ and ferromagnetic transition temperature $T_c$ of SrRuO₃ thin films [38], each multilayer was composed of two separated SrRuO₃ layers with 6 MLs and 18 MLs thickness. The 18 MLs thick SrRuO₃ was deposited directly on the SrTiO₃ (100) substrate, while the top 6 MLs SrRuO₃ layer was grown on top of the spacer layer, as illustrated in the scheme of the heterostructure design in Figure S1a of the supplemental material (Ref. [39]).

The heterostructures were fabricated by pulsed-laser deposition (PLD), using a KrF excimer laser with 248 nm wavelength. The multilayers were grown on SrTiO₃ (100) substrates. The substrates were etched in NH₄F - buffered HF solution and annealed in air at 1000°C for 2 hours to achieve uniform TiO₂- termination of the surface. During the growth, the deposition temperature was 650°C, the oxygen pressure was kept at 0.133 mbar and the laser fluence was set to about 2 J/cm². We used 5 Hz repetition rate for the SrRuO₃ and 1 Hz for SrIrO₃. In order to ensure a smooth epitaxial growth for enhanced thicknesses of the SrIrO₃ spacer, the deposition temperature was increased for the heterostructure RIR12 to 700°C. Employing in situ high-energy electron diffraction (RHEED) enabled the precise control of the SrIrO₃ layer thickness, which grew in a layer-by-layer mode (see Figure S1b in the supplemental information (Ref. [39])). Atomic force microscopy (AFM) investigations confirmed the smooth topography of the heterostructure surface resembling the stepped terrace structure of the SrTiO₃ (100) substrates. Further details on the thin film deposition and structural characterization can be found in the SI (Ref.[39]).

The magnetic interlayer coupling was investigated by a combination of conventional SQUID magnetometry (temperature-dependent and magnetic field-dependent magnetic moment measurements) and FORC investigations. The study was complemented by polar magneto-optical Kerr effect (p-MOKE) and Hall voltage measurements for selected samples. All Hall measurements were performed in the van der Pauw geometry in a custom-built set up.

SQUID magnetometry was performed by a commercially available SQUID magnetometer (MPMS-XL , Quantum Design inc.). In order to extract the magnetic response of the ferromagnetic SrRuO₃ layers, the linear contribution of the diamagnetic SrTiO₃ substrate was subtracted by linear fitting in the high magnetic field range. Furthermore, the nonlinear magnetic moment measured above the Curie temperature of the SrRuO₃ layers was subtracted to correct the additional background response originating from magnetic impurities introduced most likely during the required sample cutting (see section 2 of the supplemental material [39]).

The FORC measurements were performed with a SQUID magnetometer (MPMS 3 , Quantum Design inc.). Processing of raw data was done with LeXtender [40], and the FORC densities were calculated using the gFORC algorithm [41]. For the FORC study, a set of minor loops with various reversal fields was performed. Before each minor loop, the sample was saturated in a positive magnetic field of 5 T. Then the external magnetic field was decreased to the required reversal field $H_r$. The first order reversal curve was determined by measuring the magnetic moment when the magnetic field was increased from $H_r$ to saturation in positive magnetic fields [27, 32, 41]. This procedure was repeated with step-like decreasing of the reversal field until the reversal field reached negative saturation. The FORC-density was calculated by the mixed second derivative of the magnetic moment surface:

$$\rho(H, H_r) = -\frac{1}{2} \frac{\partial^2 m(H, H_r)}{\partial H \partial H_r}$$  \hspace{1cm} (1)

The FORC density was then transformed on the axes of the coercive field $H_c$ and the interaction field $H_u$ via:

$$H_u = \frac{1}{2} (H + H_r); \quad H_c = \frac{1}{2} (H - H_r)$$  \hspace{1cm} (2)
From the FORC-density, plotted as function of the interaction field and the coercive field, the sign of the magnetic interlayer coupling can be assessed.

III. RESULTS

A. Heterostructure with 2 MLs SrIrO$_3$ spacer (RIR2)

Summarized in Figure 1 (a) and (b) are major and minor magnetic hysteresis loops for the heterostructure RIR2 (2 MLs SrIrO$_3$ / 6 MLs SrRuO$_3$ / 2 MLs SrIrO$_3$ / 18 MLs SrRuO$_3$ on SrTiO$_3$, as shown in Figure S1a in the SI (Ref. [39])) at representative temperatures of 10 K and 80 K. The hysteresis loops, acquired by SQUID magnetometry, were corrected by the subtraction of the diamagnetic background of the SrTiO$_3$ substrate and magnetic impurities, following the procedure described in section 2 of the SI (Ref. [39]).

The magnetization of the heterostructure RIR2 reverses its orientation in a two-step reversal process indicating at best weak coupling of the two SrRuO$_3$ layers. Since the 18 MLs thick SrRuO$_3$ layer has a larger magnetic moment than the thinner SrRuO$_3$ layer, it can be concluded that the thicker layer is the magnetically softer layer at 10 K. At elevated temperatures, such as 80 K, the thinner 6 MLs SrRuO$_3$ layer is magnetically softer and switches at smaller magnetic fields than the 18 MLs SrRuO$_3$ layer, as it has been shown already in our previous study on similar SrRuO$_3$-based heterostructures [25].

The temperature dependence of the switching fields of the two ferromagnetic layers of this particular heterostructure RIR2 is shown in Figure S4a in the SI (Ref. [39]). In addition to the sharp two-step magnetization reversal, the magnetic hysteresis loops possess a tail in the high magnetic field range, that can be related most likely to strongly pinned domains in the bottom SrRuO$_3$ layers deposited directly on the SrTiO$_3$ (100) substrate [42].

The minor loop of heterostructure RIR2, drawn in blue in Figure 1(a), did not show a measurable shift with respect to the major hysteresis loop at 10 K, showing that the two SrRuO$_3$ layers of the heterostructure are indeed magnetically decoupled. In contrast, the minor loop of the heterostructure RIR2 is shifted by +30 mT to higher magnetic fields at 80 K (cf. inset in Figure 1(b)). Such a positive shift of the minor loop with respect to the full loop can be an indication for antiferromagnetic coupling of the two SrRuO$_3$ layers (see for instance ref. [1, 43, 44]).

According to the calculation proposed by van der Heijden et al., the magnetic coupling strength is directly proportional to the minor loop shift and the magnetization of the magnetically softer layer [43]. As described in detail in section 4 of the SI (Ref. [39]), we estimated a coupling strength of $35 \mu J/m^2$ for a SrIrO$_3$/SrZrO$_3$ spacers of thickness of about 0.8 nm. As shown in Figure S4 in the SI (Ref. [39]), for heterostructure RIR2, the minor loop shift is almost temperature independent between 40 K and 100 K, when the 6 MLs top SrRuO$_3$ layer is the magnetically softer layer of the heterostructure. Thus, the coupling strength, which is directly proportional to the magnetization of the magnetically softer layer [43], decreases for increasing temperature above 40 K, following the temperature dependence of the magnetization of the thinner SrRuO$_3$ layer.

To confirm the sign and order of magnitude of the minor loop shifts, determined from the magnetometry measurements, Kerr rotation measurements were performed at 10 K and 80 K (see Figure S3 in the SI (Ref. [39])). The Kerr rotation angle, determined in the polar MOKE geometry, scales linearly with the perpendicular component of the magnetization, but is not influenced by magnetic
FIG. 2. First order reversal curve study of the heterostructure RIR2 at 10 K. The measured minor loops, corrected for the diamagnetic contribution originating from the substrate, are presented in (a). The FORC densities plotted as function of the coercive field \( H_c \) and the interaction field \( H_u \) at the corresponding temperatures are shown in (b). Positive FORC density peaks are drawn in red, negative ones in blue. Feature (I) and (II) correspond to the magnetization switching of the 6 MLs (I) and 18 MLs SrRuO\(_3\) (II) layer, respectively. Shown in (c) is the comparison of the major magnetization loop (red), corrected by the subtraction of the diamagnetic substrate and magnetic impurity contribution (see SI [39]), and the reintegrated FORC density (black) after removal of the soft magnetic contribution of the reversible ridge between \(-0.05 \mu_0 H \) to \(0.1 \mu_0 H\) and \(-1.5\) to \(1.5 \mu_0 H\).

Impurities at the backsides or on the edges of the sample for our measurements in reflection geometry and therefore a useful probe of the qualitative interlayer coupling. In agreement with our results from the SQUID investigations, the minorloop at 10 K (Figure S3a in the SI (Ref. [39])) is not shifted within the magnetic field accuracy, while the minorloop at 80 K is also shifted by +38 mT (Figure S3b in the SI (Ref. [39])).

To study further the magnetic interlayer coupling in the heterostructure RIR2, FORC measurements were performed at 10 K (Figure 2) and 80 K (Figure 3). Presented in Figure 2(a) is the set of minor loops of heterostructure RIR2 at 10 K. All minor loops were corrected by the subtraction of the diamagnetic contribution originating from the SrTiO\(_3\) substrate. The soft magnetic contribution visible in the minor loops at small magnetic fields is related to magnetic impurities, often introduced during the sample cutting process, as mentioned earlier. Additionally, the two step-reversal of the magnetization was observed for the minor loops that started close to negative saturation. From these minor loops, the FORC density was calculated according to equation 1 and is shown in Figure 2(b). Three general features are present in the FORC density at 10 K. The positive peaks (I) and (II) correspond to the reversal of the two ferromagnetic SrRuO\(_3\) layers. The intensity of the peaks is proportional to magnetization of the respective layer. Hence, the more intense peak (II) is related to the switching of the 18 MLs bottom SrRuO\(_3\) layer and (I) to the 6 MLs thin SrRuO\(_3\) layer. The positions of the center of the peaks at 620 mT (I) and 450 mT (II) are in good agreement with the switching fields determined from the major magnetization hysteresis loops (see Figure S4a in the SI (Ref. [39])). The FORC investigations of heterostructure RIR2 did not show any hints of the coupling of the two ferromagnetic SrRuO\(_3\) layers at 10 K.
The additional feature located at tiny magnetic field values is the reversible ridge which is dominated by magnetically soft, reversible contributions originating mainly from magnetic impurities. In case of the SQUID hysteresis loop (Figure 1), these contributions were removed by subtraction of the hysteresis loop measured above the transition temperature of the SrRuO$_3$ layers and therefore related to high $T_c$ magnetic impurities (see section 2 of the SI (Ref. 39) for further details). To confirm that the reversible ridge is dominated by the contribution of these magnetic impurities, the FORC density presented in (b) was reintegrated with the exclusion of the contribution between $-0.05 \ T < \mu_0 H_c < 0.1 \ T$ and $-1.5 \ T < \mu_0 H_u < 1.5 \ T$. Such integration of the FORC density was possible, because feature (I) and (II) originating from the magnetization reversal of the layers were sufficiently separated from the reversible ridge. The integration yielded the half of the hysteresis loop from -2 T to 2 T and was mirrored at both x- and y-axis in order to reconstruct the full hysteresis loop. Plotted in Figure 2(c) is the comparison of the reconstructed hysteresis loop of the FORC study (black) and the conventional major magnetization loop (red), which has been corrected for the magnetic impurity contribution. Both hysteresis loops are in good agreement and the switching fields of the two SrRuO$_3$ layers are identical for both techniques within a few mT. The agreement of both hysteresis loops supports our expectation that the reversible ridge is dominated by uncorrelated magnetic impurities that do not influence the switching fields of the magnetic SrRuO$_3$ layers of the heterostructure. This shows that the reintegration of the FORC density without the reversal ridge can be used in this case to obtain a hysteresis loop where the contribution of the soft magnetic impurity is removed, without the need of an additional measurement above the transition temperature of SrRuO$_3$. The FORC study of heterostructure RIR2 at 80 K is summarized in Figure 3. Also the minor loops measured at 80 K, presented in Figure 3(a), show a two-step magnetization reversal. At 80 K, the 6 MLs thin SrRuO$_3$ switches at smaller magnetic fields than the 18 MLs thick bottom SrRuO$_3$ layer. In the FORC density, shown in (b), feature (I) corresponds again to the reversal of the 6 MLs SrRuO$_3$, while feature (II) originates from the magnetization switching of the 18 MLs thick SrRuO$_3$ layer. In contrast to the FORC density map at 10 K, an additional positive-negative-peak pair (structure III) is present at finite interaction field (see Figure 3(b)) at 80 K. According to previous FORC studies on well defined systems of coupled microarrays and on NdFeB samples with components with different coercivites, such additional positive-negative-peak pairs are characteristic for magnetic coupling between two different magnetic sites and denominated as the so called “interaction peak” [27, 32]. The relative position of the positive and negative part of the interaction peak with respect to each other yields information about the nature of the coupling. As shown in [27], the coupling is antiparallel if the negative FORC density part of the interaction peak is at higher coercive and interaction fields than the positive part of the interaction peak and parallel if it is vice-versa. According to this, the interaction peak in Figure 3(b) shows that the SrRuO$_3$ layers in sample RIR2 are coupled antiparallel, which confirms our observation from the conventional SQUID magnetometry. If exchange bias between a ferromagnet and an antiferromagnet was present, this would lead most likely to a positive peak in the FORC density which is elongated along the interaction field [45] rather than a positive-negative peak pair. The FORC density of a SrRuO$_3$-based heterostructure in which the ferromagnetic layers are coupled weakly ferromagnetically is presented in the supplemental material.
for comparison [39]. In this heterostructure RIZR1, the two SrRuO$_3$ layers were weakly ferromagnetically coupled through a spacer of 1 ML SrIrO$_3$ and 1 ML SrZrO$_3$. The FORC density, plotted in Figure S5 in the supplemental material (Ref. [39]), also shows two positive peaks related to the magnetization reversal of the magnetization of the two SrRuO$_3$ layers. The observed interaction peak shows the positive FORC density at higher coercive and smaller interaction field than the negative peak, which indicates the ferromagnetic coupling between the SrRuO$_3$ layers [27].

B. Influence of the metallicity of SrIrO$_3$ spacers on the interlayer coupling

The metal-to-insulator transition reported between 3 and 4 MLs bare SrIrO$_3$ thin films deposited on SrTiO$_3$ [35] motivated the growth of the two heterostructures RIR6 and RIR12 with SrIrO$_3$ spacers that are considerably thicker than 4 MLs. The increased electrical conductivity of the spacer was expected to have major impact on the interlayer exchange coupling, as it was achieved in SrRuO$_3$ based heterostructures separated by LaNiO$_3$ spacers [23]. Presented in Figure 4 (a) and (b) are the full and minor magnetic hysteresis loops of heterostructure RIR6 (6 MLs SrIrO$_3$ spacer) at 50 K and 80 K, acquired by SQUID magnetometry. The major hysteresis loops of the heterostructure RIR6 show a two-step reversal of the magnetization, similar to heterostructure RIR2. At 50 K, the switching at 0.1 T originates from the magnetization reversal of the 6 MLs SrRuO$_3$ layer, while the step at 0.25 T is related to the switching of the 18 MLs SrRuO$_3$ layer, which is the magnetically harder layer at 50 K. Such two-step switching process indicates again the decoupling or weak magnetic interlayer coupling. To determine the interlayer coupling strength, the reversal fields of the minor loop were compared to the switching behavior of the magnetically softer layer during the full loop. As highlighted in the inset of Figure 4(a), the minor loop switching field is equal to the switching field of the major loop. This shows that the minorloop shift and therefore the coupling strength is zero (see section 4 in the SI (Ref. [39])) for further details on the calculation). The two SrRuO$_3$ layers are fully decoupled by 6 MLs SrIrO$_3$ at 50 K. Also at 80 K, where a weak antiferromagnetic coupling was observed for heterostructure RIR2, the minor loop is not shifted in case of heterostructure RIR6 (see inset of Figure 4(b)). As shown in Figure 4(c), such equality of the switching fields of minor loop (drawn as blue triangles) and major loop (full symbols) was observed at all temperatures investigated for heterostructure RIR6. The absence of a measurable minor loop shift shows that 6 MLs SrIrO$_3$ spacer decouple the two ferromagnetic SrRuO$_3$ layers fully at all temperatures.

Even further thickness increase of the SrIrO$_3$ spacer to 12 MLs was still insufficient to couple the two SrRuO$_3$ layers of heterostructure RIR12 considerably. As shown by the loop measurements at 50 K in Figure 4(d), the magnetic hysteresis is consistent with a two step-reversal of the magnetization. The minor loop is not shifted within the magnetic field accuracy indicating the decoupling of the two ferromagnetic layers. The decoupling of the SrRuO$_3$ layers was confirmed by a magnetotransport study of heterostructure RIR12, presented in the SI (Ref. [39] section 6).

Despite the expected metal-insulator transition, no significant change of the interlayer coupling strength was observed for the increase of the SrIrO$_3$ spacer thickness. Most likely, the SrIrO$_3$ spacers did not become metallic and thus could not lead to a major change of the magnetic interlayer coupling. This was adressed by reference thin film investigations, presented in the SI (Ref. [39]), because the details of the SrIrO$_3$ spacer resistivity could not be assessed directly for our heterostructures when the SrIrO$_3$ was sandwiched between the metallic SrRuO$_3$ layers. Semimetallicity has been observed in thin SrIrO$_3$ films, showing multiband-, typically hole-dominated, transport properties with similar carrier densities and mobilities for electrons and holes [35]. Therefore, off-stoichiometry can influence the transport properties of SrIrO$_3$ thin films. Oxygen vacancies for instance, acting as electron dopants, can be responsible for reduced conductivity for hole-dominated transport in SrIrO$_3$ thin films [35]. On the other hand, SrIrO$_3$ thin films are known to be sensitive inhomogeneous Iridium distribution. Biswas et al. have found that SrIrO$_3$ films deposited at high temperatures show a more insulating behavior which was related to increased density of scattering centers due to inhomogeneous cationic elemental distribution [46]. Generally, the coupling of two ferromagnetic layers separated by a nonmagnetic insulator can originate from different mechanisms, such as the direct coupling via pinholes [47], magnetostatic Néel’s coupling due to correlated surface roughness [48, 49], due to shape induced magnetic poles [50], or induced by the coupling of magnetic domain walls [51–54]. Another coupling mechanism is the magnetic exchange coupling by tunneling of spin-polarized electrons through the insulating barrier [44, 55, 56].

The coupling via pinholes, which are often present in heterostructures with ultrathin spacers, would lead to trivial ferromagnetic coupling [47, 57] between the SrRuO$_3$ layers and thus cannot explain the minor loop shift to higher magnetic fields for sample RIR2. On the other hand, we emphasize at this point that the coupling of the two SrRuO$_3$ layers separated by 2 MLs SrIrO$_3$ was found to be very sensitive to the existence of (pin-)holes in the heterostructure. As presented in Figure S8 in the SI (Ref. [39]), a second heterostructure where holes of nanometer depth were observed by atomic force microscopy showed weak ferromagnetic coupling. In contrast, atomic force microscopy did not show the existence of holes in none of the heterostructures RIR2, RIR6, RIR12 so that it can
FIG. 4. Full and minor hysteresis loops of the magnetic moment of heterostructure RIR6 with 6 MLs SrIrO$_3$ spacer at 50 K (a) and 80 K (b). (c) Temperature dependence of the switching fields of the two SrRuO$_3$ layers with 18 MLs (switching field 2) and 6 MLs (switching field 1) thickness, and the switching fields determined from minor loops experiments for the heterostructure RIR6. (d) Major and minor hysteresis loops of the magnetic moment of heterostructure RIR12 with 12 MLs SrIrO$_3$ spacer at 50 K.

be concluded that the density of pinholes connecting the two SrRuO$_3$ layers is most likely small for these samples. The weak antiferromagnetic coupling was observed only in heterostructure RIR2 with 2 MLs SrIrO$_3$ spacer and with a small density of holes seen by AFM. In addition, magnetostatic and interlayer exchange coupling can lead to magnetic coupling of the SrRuO$_3$ layers. Antiferromagnetic coupling between ferromagnetic layers separated by nonmagnetic, insulating spacers has been previously related also to exchange coupling [44] described by the Slonczweski spin-current model [55] or the quantum interference model developed by Bruno [56]. However, the observed decrease of the coupling strength $J_C$ (calculated with equation (1) in the SI (Ref. [39])) with increasing temperature observed for heterostructure RIR2 cannot be explained within the model of quantum interference effects, which predicts an increase for increasing temperatures in case of insulating spacers [56].

Néel’s theory of the magnetostatic coupling due to magnetic surface charges induced by correlated surface roughness was extended by Moritz et al. to magnetic multilayers with perpendicular magnetic anisotropy [49]. Depending on the strength of the magnetic anisotropy constant, the magnetostatic orange-peel coupling has been found to be ferromagnetic for a weak perpendicular anisotropy constant and antiferromagnetic for strong perpendicular magnetic anisotropy [49]. SrRuO$_3$ thin films deposited on SrTiO$_3$(100) typically have a large magnetic anisotropy with the magnetic easy axis close to the [110]$_{orthorh.}$ direction [58] so that the orange-peel coupling would be expected to favor antiferromagnetic coupling between the layers. The heterostructures under study were all deposited on SrTiO$_3$ (100) substrates with a step-and terrace structure of 0.4 nm height and 250-300 nm width that most likely led to unidirectional interface roughness [59]. However, the orange-peel coupling
fields [60] expected for the substrate induced roughness would be too small to explain the observed weak antiferromagnetic coupling. Furthermore due to the similarity of the substrate topography, it would be expected for all heterostructures under study, but the weak antiferromagnetic coupling was observed only for heterostructure RIR2.

A mechanism of domain replication in the hard layer via magnetostatic interactions could explain the very weak antiferromagnetic coupling [61]. When the magnetic field required to reverse the magnetization of the soft layer during the minor loops is close to the nucleation field of the hard layer, inverted domains in the soft layer will generate stray fields that can induce so called replicated domains in the hard layer acting as negative bias field during the second half of the minor loop [61, 62].

IV. CONCLUSION

The magnetic interlayer coupling between ferromagnetic SrRuO$_3$ epitaxial layers separated by the strong spin-orbit coupling SrIrO$_3$ was investigated by the combination of conventional SQUID magnetometry and FORC measurements. The minor loops of the heterostructure with 2 MLs of SrIrO$_3$ spacer showed a small shift to higher magnetic field above 30 K, indicating very weak antiferromagnetic coupling of about 7 $\mu$J/m$^2$. The increase of the SrIrO$_3$ layer thickness above the thickness for which a metal-to-insulator transition was reported for bare SrIrO$_3$ layers did not lead to an increase of the coupling, but to rather fully decoupled layers. This is most likely related to the insulating behavior of the SrIrO$_3$ spacers of our heterostructures. Even the 12 MLs spacer was likely not metallic, as indicated by measurements of a reference sample. Such decoupling or very weak antiferromagnetic coupling of the SrRuO$_3$ by SrIrO$_3$ spacers is undesirable in the context of skyrmion formation. Without ferromagnetic coupling of the magnetic layers, skyrmions would not be coupled through multilayer stacks. Our study further highlights the scientific relevance of first order reversal curve investigations for the study of magnetic interlayer coupling, being capable to detect weak coupling interactions as well as to determine whether the coupling is antiferromagnetic or ferromagnetic. Additionally, FORC measurements have the advantage that a correction for the contribution of magnetic impurities is not necessary, because the peaks representing the various magnetization reversal steps are well separated in the FORC density maps. We could also show that reintegrating the FORC density without the reversible ridge can be an alternative method to correct a samples hysteresis loop for a soft magnetic impurity.

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Supplemental Material for
Magnetic interlayer coupling between ferromagnetic SrRuO$_3$ layers through a large spin-orbit coupling SrIrO$_3$ spacer

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1 Heterostructure deposition and surface topography investigation

Figure S 1: (a) Scheme of the heterostructures under study, exemplarily drawn for heterostructure RIR12, with the thickest spacer layer of this study (12 MLs SrIrO$_3$) (b) integrated RHEED intensity plotted as function of deposition time of the heterostructure RIR12. Atomic force microscopy images (5 µm x 5 µm) of one SrTiO$_3$ (100) substrate after etching and annealing (c), and AFM images of the heterostructures RIR2 (d), RIR6 (e), and RIR12 (f).

The heterostructures under study have the general design depicted in Figure S1 (a). They are composed of two ferromagnetic SrRuO$_3$ layers with deliberately different thicknesses of 18 and 6 monolayers (MLs). The bottom SrRuO$_3$ layer is thicker than the top SrRuO$_3$ layer in each heterostructure and
grown directly on top of the SrTiO$_3$ substrate. A spacer of SrIrO$_3$ separates the two ferromagnetic layers. For the heterostructure RIR2, the top SrRuO$_3$ layer is additionally capped by a 2 MLs SrIrO$_3$ layer.

In case of decoupled or only weakly coupled layers, the magnetic hysteresis loop of such a heterostructure shows a two-step switching behavior due to the different temperature dependence of the coercive fields of the two SrRuO$_3$ layers. This allows to assess the magnetic interlayer coupling.

The deposition was monitored by in-situ reflection high-energy electron diffraction (RHEED). As shown exemplarily for heterostructure RIR12 in Figure S1 (b), the SrIrO$_3$ layers grew in a layer-by-layer growth, which enabled the precise control of the layer thickness. The SrRuO$_3$ layers grew in step-flow mode which has been proven to result in smooth thin films [1]. The surface of the heterostructures possessing SrIrO$_3$ spacers, presented in (d)-(f), are smooth, resembling the structure of the SrTiO$_3$ substrate with uniform terrace width and one unit cell step height (see (c)). During the investigation of the surface topography by AFM, we did not observe any etch pitches on the substrates before deposition, or deep holes in the heterostructures with SrIrO$_3$ spacer (RIR2, RIR6, RIR12).

2 SQUID magnetometry: Background correction procedure exemplarily shown for heterostructure RIR2 at 10 K

![Graph](image)

Figure S2: Magnetic hysteresis loops of heterostructure RIR2 at 10 K (a) and 200 K (b). The loops have been corrected by subtraction of the diamagnetic contribution from the SrTiO$_3$ substrate. (c) Magnetic hysteresis loop at 10 K after subtraction of the 200 K measurement.

The background correction procedure that has been used for the hysteresis loops measured by SQUID magnetometry is presented in Figure S2, exemplarily for heterostructure RIR2 at 10 K. All magnetic hysteresis loops were corrected by the subtraction of the diamagnetic response from the substrate by the perfomance of linear fitting in the high magnetic field range where the sample is in its saturated state. In order to correct for the magnetically soft contribution of magnetic impurities, visible in the hysteresis loop drawn in (a), a reference hysteresis loop was measured at 200 K, above the transition temperatures of the two SrRuO$_3$ layers of the heterostructures. By subtraction of the 200 K measurement shown in (b), this soft magnetic contribution was removed successfully, as shown in (c). However, small peak-like features around 0 T originating from the imperfect background correction remain.
3 Heterostructure RIR2: Magneto-optical Kerr measurements

Figure S 3: Magneto-optical Kerr rotation measurements of heterostructure RIR2 at 10 K (a) and 80 K (b), measured with incoherent light of 540 nm. Drawn with full symbols are the major hysteresis loops; the open symbols are the minor loops.

In order to confirm the sign and magnitude of the observed minor loop shift of heterostructure RIR2 with 2 MLs SrIrO$_3$ spacer and capping layer, polar magneto-optical Kerr effect measurements were performed with our home-built polar-MOKE set up based on the well-established double modulation technique with light from a Xe-lamp. At 10 K, the minor loop is not shifted measurably compared to the full loop, as depicted in Figure S3, which confirms our results from the SQUID magnetometry and FORC study that the SrRuO$_3$ layers are decoupled at low temperatures. The minor loop at 80 K, measured between 2.5 T and -0.1 T is shifted by +38 mT with respect to the full loop. This shift to higher magnetic fields is similar to the minor loop shift that was observed in our SQUID magnetometry and is also in agreement with the (antiferromagnetic) interaction peak seen in our FORC study of this sample.

4 Heterostructure RIR2: Temperature dependence of the switching fields and the magnetic interlayer coupling strength

The full and minor magnetic hysteresis loops of heterostructure RIR2 at 40 K are presented in Figure S4(a). The minor loop (blue) is shifted by +30 mT with respect to the full loop (black). 40 K is the lowest temperature of the current study at which a positive minor loop shift was observed. Shown in Figure S4(b) are the switching fields of the two SrRuO$_3$ layers of heterostructure RIR2 as function of temperature. The two SrRuO$_3$ layers of the heterostructure have the same switching field at 20 K. Below 20 K, the bottom 18 MLs SrRuO$_3$ switches at smaller magnetic fields than the 6 MLs top SrRuO$_3$, while they behave vice-versa above 20 K. Drawn with open triangles are the switching fields of the minor loops at the respective temperatures. The minor loop shift is zero below 20 K, when the bottom, 18 MLs thick SrRuO$_3$ is magnetically softer than the 6 MLs thin SrRuO$_3$ layer. When the thinner layer switches at smaller magnetic fields than the thicker SrRuO$_3$ layer, the minor loop is shifted by 30 mT to elevated fields at all investigated temperatures below the Curie temperature of the thin layer at 110 K (compare the m(T) measurement in (c)). From the difference of the switching fields determined from the minor loops and the full loops of the magnetically softer SrRuO$_3$ layer, the coupling strength $J_C$ can be calculated with the following equation introduced by Heijden et al. [2].

$$J_C = \mu_0 \Delta H_{shift} M_{soft, rev} t_{soft, rev}$$ (1)
Figure S 4: (a) Full and minor magnetic hysteresis loops of heterostructure RIR2 at 40 K. Shown in the inset is the magnetic field range in which the shift of the minor loop to elevated magnetic fields is highlighted. (b) Temperature dependence of the switching fields of the two SrRuO$_3$ layers of heterostructure RIR2 determined from the major hysteresis (circles) and from the minor loops (open triangles). (c) Temperature dependence of the calculated coupling strength (red) and the magnetic moment during warming in 0.1T after field cooling (in 0.1 T).

with the minor loop shift $\Delta H_{\text{shift}}$. $t_{\text{soft,rev}}$ and $M_{\text{soft,rev}}$ are the thickness and the magnetization of the magnetically softer layer, respectively. Due to the direct proportionality to the magnetization of the magnetically softer layer $M_{\text{soft,rev}}$ and the observed almost temperature independent minor loop shift (above 40 K), the total magnitude of the coupling strength is maximum at 40 K and decreases with increasing temperature following the trend of $M(T)$ of the 6 MLs thin SrRuO$_3$ layer. The temperature dependence of the magnetic moment of the heterostructure RIR2 is shown in Figure S4(c) for comparison.

The change of the magnetic interlayer coupling from decoupling below 20 K to very weak antiferromagnetic coupling above 40 K seems to be correlated to the change of the magnetically softer and harder layer of the heterostructure (cp. Figure S4 (b)). When the 18 MLs thick bottom SrRuO$_3$ layer is the magnetically harder layer of the heterostructure, no shift of the minor loop is observed. Only in the temperature range where the 6 MLs thick top SrRuO$_3$ layer has smaller coercivities, the small shift of the minor loop, indicating weak antiferromagnetic coupling, is seen. A similar temperature dependence, namely decoupling at low temperatures and strong coupling at higher temperatures, has been observed in Ni/Co pseudo-spin-valve structures in which the Ni/Co multilayers of different repetition numbers, separated by 4.6 nm Cu, were strongly coupled via magnetic dipolar coupling [3]. Mohnseni et al. attributed this dependence and the decoupling at low temperatures to the increase
of the difference of the coercivities of the magnetically softer and harder layers at low temperatures so that the stray fields of the magnetically harder layer were insufficient to initiate the reversal of the magnetically softer layer [3].

Maybe also in our case the coupling at low temperatures, where the difference between the coercive fields is more than two times larger than above 30 K, is too weak to initiate the switching in the magnetically harder (18 MLs thick) layer.

On the other hand, assuming that the coupling strength was also on the order of a few $\mu J/m^2$ at low temperature, the expected shift of the minor loop would be smaller by a factor of 5-6.5, since the thicker SrRuO$_3$ is the magnetically softer one at low temperature. However, a minor loop shift of 6 mT is close to the detection limit of our experimental set ups and therefore experimentally challenging to observe.

5 First order reversal curve study of heterostructure RIZR1 showing weak ferromagnetic coupling of the SrRuO$_3$ layers

Figure S 5: (a) Major (black) and minor (blue) magnetic hysteresis loops of heterostructure RIZR1 at 10 K, measured with conventional SQUID magnetometry. This heterostructure has 1 ML SrIrO$_3$ and 1 ML SrZrO$_3$ as spacer and capping layers. (b) Minor loops of heterostructure RIZR1 measured at 10 K. (c) FORC density plotted as a function of the coercive field $H_c$ and the interaction field $H_u$ for the heterostructure RIZR1 at 10 K. Positive FORC density peaks are shown in red, negative ones in blue. Feature (I) and (II) correspond to the magnetization switching of the 6 MLs (I) and 18 MLs SrRuO$_3$ (II) layer, respectively. (III) is the interaction peak indicating ferromagnetic coupling.

The major and minor hysteresis loops, determined by SQUID magnetometry of heterostructure RIZR1 are shown exemplarily at 10 K in (a). Similar to heterostructure RIR2, also this heterostructure is composed of two SrRuO$_3$ layers of 18 MLs and 6 MLs thickness. They are separated and capped by 1 ML SrIrO$_3$ /1 ML SrZrO$_3$. As highlighted in the inset of (a), the minor loop is shifted by 30 mT
to smaller magnetic fields indicating a weak ferromagnetic coupling on the order of 35 $\mu$J/m$^2$, as we have found in our previous study [4]. In order to confirm the relation of the interaction peak with the type of magnetic coupling, FORC studies were also performed at 10 K for the heterostructure RIZR1 with heterogeneous spacer of 1 ML SrIrO$_3$/ 1 ML SrZrO$_3$. Depicted in (b) are the individual minor loops of the first order reversal curve study at 10 K. Only the diamagnetic contribution originating from the SrTiO$_3$ substrate has been corrected by linear fitting in the high magnetic field range where the sample is in its saturated state. The soft magnetic contribution is related to magnetic impurities introduced during the sample cutting and handling. The FORC density at 10 K is plotted in Figure S5 (c) as function of the interaction field $H_u$ and the coercive field $H_c$. Similar to heterostructure RIR2 with 2 MLs SrIrO$_3$ spacer, a reversible ridge was observed at small magnetic field values which is related to purely reversible magnetization switching. Due to the relative intensity of the peaks, the more intense peak (II) can be related to the switching field of the 18 MLs thick SrRuO$_3$ layer. Feature (I) corresponds to the irreversible switching of the 6 MLs SrRuO$_3$ layer of the heterostructure. In accordance with the global SQUID magnetometry shown in (a), the 6 MLs SrRuO$_3$ is the magnetically harder layer at 10 K. An additional positive-negative peak pair is present in the FORC density with the orientation opposite to the one observed for heterostructure RIR2. While the orientation of feature (III) in Figure 2 of the paper indicated antiferromagnetic coupling for heterostructure RIR2, the reversed orientation of feature (III) in Figure S5 supports the observation of ferromagnetic coupling at 10 K in heterostructure RIZR1.
6 Magneto-transport investigations of heterostructure RIR12 with 12 MLs SrIrO$_3$ spacer

Figure S 6: Comparison of the magnetic hysteresis loops determined by SQUID magnetometry (black) and the Hall voltage (blue), corrected for the ordinary Hall contribution, as function of applied magnetic field for heterostructure RIR12 with 12 MLs SrIrO$_3$ spacer. To increase the comparability of the magnetic field dependencies of Hall effect and magnetization reversal, the anomalous Hall voltage was plotted from positive to negative values for 10 K, 50 K, 80 K, and 100 K.

As described in detail in the previous section 2, all SQUID magnetometry measurements of this study have been corrected by subtraction of the magnetic hysteresis loop at 200 K, above the Curie temperature of the SrRuO$_3$ layers of the heterostructures. However, this correction leads to artifacts, such like the small peaklike features close to zero T at low temperatures. In order to confirm that this correction does not lead to a misinterpretation of the main physical properties of the heterostructures, Hall measurements were performed for sample RIR12, the heterostructure with the thickest SrIrO$_3$ spacer of this study.

In a single domain ferromagnet, the anomalous Hall constant is directly proportional to the out-of-plane component of the magnetization [5]. As shown also by van Thiel et al., the measured Hall voltage of a magnetic sample that contains several anomalous Hall conduction channels is given by the sum of the different individual contributions [6]. For heterostructure RIR12, the overall anomalous Hall voltage is given by the sum of the contributions of the two SrRuO$_3$ layers with distinct thicknesses and therefore different temperature dependencies of the anomalous Hall constant.

Depicted in Figure S6 is the comparison of the magnetic hysteresis loops (black) and the Hall voltage (after subtraction of the ordinary Hall effect) of heterostructure RIR12 at several temperatures below the ferromagnetic transition temperature of the 18 MLs SrRuO$_3$ layer of the heterostructure. The observation of hump-like features between 50 K and 100 K confirms our expectation of the different
temperature dependencies of the anomalous Hall constant. The anomalous Hall constant of the thin SrRuO$_3$ layer is most likely positive in this temperature range, while the AHE constant of the thicker SrRuO$_3$ layer is still negative up to 100 K. Most relevant for the present study is the magnetic field range in which the two different SrRuO$_3$ layers reverse their magnetization. The comparison of the SQUID and Hall measurements confirms that the hump-like features appear in the same magnetic field range in which the magnetically softer layer reverses its magnetization and disappears when the magnetically harder layer switches its magnetization. This confirms the switching fields determined by SQUID magnetometry. At 100 K, the hump-like feature has an s-shape which is most likely related to the 6 MLs thin layer which is already in its paramagnetic phase with a still measurable contribution to the Hall voltage. At 125 K, the Hall voltage loop is mainly determined by the anomalous Hall voltage of the 18 MLs SrRuO$_3$, which is still in its ferromagnetic phase, again confirming the SQUID magnetometry. Depicted in Figure S7 are the major and minor loop hystereses of the Hall voltage,

Figure S 7: Comparison of the magnetic hysteresis loops determined by SQUID magnetometry (black) and the anomalous Hall voltage (red) as function of applied magnetic field for heterostructure RIR12 with 12 MLs SrIrO$_3$ spacer at 80 K. To increase the visibility in the magnetic field range of the minor loop switching, a zoom- in of (a) is shown in (b).

after subtraction of the ordinary Hall contribution, and compared to the major magnetic hysteresis loop (in black). As highlighted in (b), the minor loop of the anomalous Hall voltage, which is proportional to the magnetization of the magnetically softer layer, is in good agreement with the magnetic field dependence of the major magnetic hysteresis loop for the reversal of the magnetically softer SrRuO$_3$ layer. This confirms that the minor loop is not shifted with respect to the full loop.
Minor loop investigations of a SrRuO$_3$-based heterostructure with 2 MLs SrIrO$_3$ spacer having nanometer-deep holes

The influence of holes in the heterostructures on the magnetic interlayer coupling was investigated by the comparison of heterostructure RIR2 with a second sample with 2 MLs SrIrO$_3$ spacer where holes of minimum 1-2 nanometer depth were observed by atomic force microscopy. As shown in Figure S8 by the time-dependent RHEED intensity plot, this sample also has a SrIrO$_3$ spacer and capping layer of 2 MLs thickness and two SrRuO$_3$ layers of distinct thicknesses. In contrast to the other heterostructures of the current study, this sample was deposited by our new pulsed laser deposition set up at the University of Cologne, manufactured by SURFACE Inc. As depicted in (b), the heterostructure surface of this sample shows the existence of nanometer deep holes. Because there were no holes observed in the AFM investigations of the heterostructures RIR2, RIR6, or RIR12, it was expected that this heterostructure was influenced more strongly by the existence of ferromagnetic bridges by pinholes. Shown in (c) are major hysteresis loops of this sample at various temperatures. Only at 10 K, the switching fields of the two individual SrRuO$_3$ layers are distinguishable. The performed minor loop at 10 K, drawn in (d), shows a small negative shift of 45 mT, indicating weak ferromagnetic coupling. Thus, both samples with 2 MLs SrIrO$_3$ spacer indicate opposite sign of the coupling of the two SrRuO$_3$ layers. The difference could originate from the different densities of nanometer deep (pin-)holes in the heterostructures, which was increased for sample TL06. Such holes are expected to lead to the formation of ferromagnetic bridges by pinholes connecting the two ferromagnetic SrRuO$_3$ layers.
Resistance measurements of heterostructure RIR12, and 6 MLs and 12 MLs bare SrIrO$_3$ thin films deposited on SrTiO$_3$(100)

Figure S 9: (a) Temperature dependence of the voltage measured along the edges of heterostructure RIR12 when 100 $\mu$A were applied. The two configurations that correspond to the current parallel to the two in-plane principal axes of the orthorhombic SrRuO$_3$ layers are sketched in the inset. Temperature dependence of the measured voltage of 6 (grey) and 12 MLs (black) bare SrIrO$_3$ thin films deposited on SrTiO$_3$ (100), for an applied current of 100 $\mu$A. The red axis shows the corresponding temperature dependent resistivity of the reference films. Because it is sandwiched between two SrRuO$_3$ layers with much lower resistivity (about 0.5 $\mu$Ωm at 10 K), the resistivity of the SrIrO$_3$ spacer layers cannot be assessed easily [7,8,9]. The voltage drop measured along the whole heterostructure will therefore be dominated by the SrRuO$_3$ layers. This is confirmed by a resistance measurement of heterostructure RIR12 in comparison to a 12 MLs bare reference SrIrO$_3$ film (see Figure S9). The voltage was measured in van der Pauw geometry as indicated in the inset in Figure S9(a). Although 100 $\mu$A were applied in both cases, the measured voltage across the heterostructure is one order of magnitude smaller than of the 12 MLs bare SrIrO$_3$ layer. Thus, our following estimations of the SrIrO$_3$ spacer resistivity are based on the investigations of 6 and 12 MLs bare SrIrO$_3$ reference thin films deposited on SrTiO$_3$ (100) substrates. Often the charge carrier density can be extracted from the magnetic field dependent Hall resistance. According to Manca et al., the transport in SrIrO$_3$ films can only be described in a multiband- model and the deflection points required to perform proper fits are predicted to be above 33 T, out of our achievable magnetic field range [7]. The temperature dependence of the resistivity, presented in Figure S9(b), indicates a semimetallic behavior with a decrease of the resistivity for doubling the layer thickness from 6 to 12 MLs. The change of slope at 105 K, indicated by the blue arrows, is in accordance with the temperature at which the structural transition of the SrTiO$_3$ substrate from cubic to tetragonal takes place. This imprint of the substrate structural transition in the film resistivity is an indication of the interfacial octahedral connectivity and has been related by van Thiel et al. to SrIrO$_3$ domains that are influenced by the multiple domains in the tetragonal substrate which leads to the reconfiguration of current ways [10]. In contrast to previous experimental studies by Groenendijk et al. and Manca et al. [7,8], our bare 6 and 12 MLs SrIrO$_3$ films show a semimetallic behavior with a comparably large resistivity and a weak upturn for decreasing temperature. The deviation of the electrical properties of the bare SrIrO$_3$ films could be related to off-stoichiometry. Due to the volatility of IrO$_x$, SrIrO$_3$ can show Ir deficiency resulting in an effective hole doping [11]. Also oxygen vacancies, electron dopants, have been found by Manca et al. to effect the electrical conductivity [12]. In case of hole-dominated transport, the
electrical resistivity increased for increasing density of oxygen vacancies. SrIrO$_3$ thin films are also known to be sensitive to inhomogeneous Iridium distribution. Biswas et al. found that SrIrO$_3$ films deposited at high temperatures show a more insulating behavior which was related to increased density of scattering centers due to inhomogeneous cationic elemental distribution [13]. Thus, most likely off-stoichiometry leads to the observed semimetallic behavior of the SrIrO$_3$ reference thin films and most likely of the SrIrO$_3$ spacers of the heterostructures.

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