Spin pumping and inverse spin Hall effect in ultrathin SrRuO$_3$ films around the percolation limit

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We demonstrate spin pumping and inverse spin Hall effect (ISHE) in an all oxide heterostructure composed of two oxides with different respective crystal structures. Y$_2$Fe$_2$O$_{12}$ (YIG) is used as a spin source, being a perfect candidate because of its low damping and insulating behavior. SrRuO$_3$ (SRO), a perovskite oxide with metallic conductivity, serves as a spin sink. Spin pumping is shown by the increase of the damping in ferromagnetic resonance for the YIG in the presence of an SRO top layer and by the inverse spin Hall effect appearing in the SRO. Transmission electron microscopy shows that the growth of YIG is fully epitaxial, while the SRO grows in islands of several polycrystalline phases, a fact which strongly influences the measured ISHE. The magnetization of the YIG remains unaffected by the SRO layer while the SRO no longer shows the low temperature ferromagnetism typically observed in monocrystalline layers.

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I. INTRODUCTION

Yttrium-iron-garnet (YIG) plays an important role in spintronics and magnonics due to its extremely low damping constant and the resulting possibility of long-range magnon propagation. In numerous experiments, spin-pumping from YIG into metals and a resulting inverse spin Hall effect (ISHE) have been demonstrated [1–4]. However, only a few results exist for all oxide heterostructures [5,6], especially those including YIG. One reason is that for the investigation of the ISHE a conducting spin sink is mandatory, and the choice of suitable materials is limited. Wahler et al. used La$_0.7$Sr$_0.3$MnO$_3$ (LSMO) as a spin source and SrRuO$_3$ (SRO) as a metallic oxide [5]. These are both perovskites which grow epitaxially on top of each other. For YIG, however, the only materials with matching crystalline structure are obviously garnets, which are insulating and thus cannot exhibit the ISHE. Only Qiu et al. have demonstrated a YIG based heterostructure, employing sputtered indium tin oxide (ITO) [6]; however, no details on epitaxial quality or crystallinity of the ITO were given. Here we present a new crossover system between garnet and perovskite structures, using YIG and SRO. To assess the structural quality of the system, we use transmission electron microscopy (TEM) and reflection high energy electron diffraction (RHEED). The main focus of our characterization, however, lies in the magnetic properties, which are characterized by superconducting quantum interference device (SQUID) magnetometry, ferromagnetic resonance (FMR), and ISHE voltage measurements.

II. SAMPLE FABRICATION

For our experiments a number of YIG/SRO bilayers are fabricated. Both layers are subsequently deposited by pulsed laser deposition (PLD) on Gd$_2$Ga$_5$O$_{12}$ (111) (GGG) (CrysTek GmbH) substrates. The substrates are fixed on the sample holder with silver glue and directly transferred into the evacuated PLD chamber (TSST) at a pressure of approximately 10$^{-9}$ mbar. The sample holder is heated up to 900 °C, which results in a substrate temperature of 816 °C, as measured by a pyrometer. The laser has a wavelength of 248 nm and is operated at a fluency of 2.5 J/cm$^2$ and a frequency of 5 Hz, resulting in a growth rate of 0.5 nm/min. During the deposition an oxygen pressure of 0.025 mbar is used. The nominal thickness of the YIG layers is 25 nm; owing to changes in the deposition rate, the layer thickness as confirmed by transmission electron microscopy varies between 20 and 34 nm (27 ± 7) nm. Subsequently the sample holder is cooled down to 650 °C (substrate temperature 604 °C) and the laser fluency is reduced to 2.4 J/cm$^2$, while all other parameters are kept constant for the deposition of the SRO. A total number of eight samples with different respective nominal SRO-thicknesses $t_{NM}$ are fabricated. In addition a single YIG layer of 20 nm is deposited as a reference, called sample Y. The samples are cut to rectangles approximately 2 mm × 5 mm in size with the substrate [110] direction oriented along the short sides. On these sides copper leads are attached using silver glue to serve as voltage probes for the ISHE measurements. A table of all fabricated samples is shown in Table I. All given thicknesses are nominal thicknesses.

III. STRUCTURAL CHARACTERIZATION

Structural characterization is done by reflection high energy electron diffraction (RHEED) and transmission electron microscopy (TEM).

A. Transmission electron microscopy

For the transmission electron microscopy (TEM) imaging, a JEOL JEM-4010 electron microscope with an acceleration voltage of 400 kV is used. One sample with an SRO thickness of 10 nm, which also shows ISHE, is prepared for cross-sectional view. To protect the surface a thin Pt layer is deposited. The Pt layer is only deposited for TEM

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 TABLE I. List of all fabricated samples with measured nominal SRO thickness $t_{NM}$, FMR linewidth, $R$, $V_{ISHE}$, and calculated $\frac{V_{ISHE}}{R}$. Samples marked with * show more than one absorption line in the FMR spectra.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$t_{NM}$ (nm)</th>
<th>FMR linewidth (Oe)</th>
<th>$R$ (kΩ)</th>
<th>$V_{ISHE}$ (μV)</th>
<th>$\frac{V_{ISHE}}{R}$ (10$^{-2}$ nA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>5</td>
<td>10.6 ± 1</td>
<td>(7.5 ± 2) × 10$^3$</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>B</td>
<td>7.5</td>
<td>13 ± 1 (34.4 ± 7)*</td>
<td>30.6 ± 7.65</td>
<td>1.1 ± 0.3</td>
<td>3.59 ± 1.4</td>
</tr>
<tr>
<td>C</td>
<td>9.2</td>
<td>13 ± 1</td>
<td>38 ± 9.5</td>
<td>0.19 ± 0.05</td>
<td>0.50 ± 0.18</td>
</tr>
<tr>
<td>D</td>
<td>9.5</td>
<td>27.7 ± 2</td>
<td>36 ± 9</td>
<td>0.1 ± 0.03</td>
<td>0.28 ± 0.11</td>
</tr>
<tr>
<td>E</td>
<td>9.8</td>
<td>15.8 ± 1</td>
<td>6.1 ± 1.5</td>
<td>3.0 ± 0.8</td>
<td>49.2 ± 17.9</td>
</tr>
<tr>
<td>F</td>
<td>10</td>
<td>36 ± 7</td>
<td>8.1 ± 2.0</td>
<td>0.31 ± 0.08</td>
<td>3.8 ± 1.4</td>
</tr>
<tr>
<td>G</td>
<td>13.4</td>
<td>27 ± 2</td>
<td>7.5 ± 2</td>
<td>0.85 ± 0.2</td>
<td>11.3 ± 3.9</td>
</tr>
<tr>
<td>H</td>
<td>15</td>
<td>4.8 ± 1</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

characterization. Further characterizations via, e.g., FMR and ISHE were done without Pt on top. After the deposition thin lamellae are cut out using a focused Ga ion beam. The orientation of the samples is chosen for cross-sectional TEM along the cubic crystalline axis. Also scanning electron microscopy images of the sample surface are taken. Figure 1 shows TEM [sample F (a,b), sample B (c), sample H (d)] and SEM [sample B (e)] images. The YIG layer is monocrystalline without any visible inclusions or defects. The SRO layer on top of the YIG, however, shows several polycrystalline and amorphous phases. The insets show the fast Fourier transforms for the marked areas. Because of the inhomogenous growth of the SRO [Fig. 1(b)], it is not possible to define an exact thickness for the SRO. In the following always nominal layer thicknesses are used, which are obtained from calibrations on perovskite substrates under similar growth conditions assuming a sticking coefficient of 1. The top view of sample B in Fig. 1(e) confirms this inhomogeneous growth.

B. Reflection high energy electron diffraction

The atomic order of the layer surface after deposition of YIG followed by SRO is investigated by reflection high energy electron diffraction (RHEED). A high pressure RHEED gun from STAIB INSTRUMENTS is integrated in the PLD chamber. It is operated at a voltage of 30 kV and can be used during the deposition process. Figures 2(a) and 2(b) show detected RHEED patterns of sample F after the 20 nm YIG growth still at a substrate temperature of 817 °C [Fig. 2(a)] and after deposition of 10 nm SRO [Fig. 2(b)] at 604 °C, respectively.

For the YIG surface we observe the typical oriented symmetric RHEED pattern. After the deposition of SRO a new

![FIG. 1. (a) TEM image with fast Fourier transforms (insets) of the marked areas of nominally 10 nm SRO and 20 nm YIG (sample F). YIG shows monocrystalline growth; SRO is deposited in polycrystalline and amorphous phases. In (b) the real thicknesses of sample F are determined. With a variance of 7–13 nm the SRO thickness is not homogenous. SRO grows in islands, visible in (c) (sample B). In sample B the islands are not connected with each other. This can be verified with a top view of the surface via scanning electron microscopy, shown in (e). The TEM image of a 15 nm SRO layer (sample H) is illustrated in (d).](image-url)
FIG. 2. RHEED images between depositions of sample F: (a) RHEED pattern after YIG deposition. (b) Change into an asymmetric pattern after SRO deposition. (c) Repetition of symmetric pattern of SRO every 60°. Apparently asymmetric pattern appears, although the azimuth angle is not changed. However, a detailed investigation, for which the azimuth is rotated, shows that the surface is still highly symmetric. In the azimuth scan the pattern shown in Fig. 2(c) can be observed with a periodicity of 60°. This indicates that the threefold symmetry of the (111) YIG surface is preserved, which for the almost grazing incidence of the beam results in the appearance of identical patterns every 60°.

IV. SQUID MAGNETOMETRY

To determine the saturation magnetization $M_s$, sample Y is characterized by SQUID magnetometry. An in-plane hysteresis loop is taken at 305 K and the linear paramagnetic background caused by the GGG substrate is subtracted. The data are shown in Fig. 3. The observed saturation magnetization is $(108 \pm 3)$ emu/cm$^3$, which is approximately 24 percent below the bulk value [7]. The decreased saturation magnetization may be explained by iron and oxygen deficiencies [8]. The coercive field amounts to $(1.1 \pm 0.1)$ Oe. SQUID magnetometry on sample G shows approximately the same saturation magnetization ($(109 \pm 3)$ emu/cm$^3$). The coercive field, however, is slightly increased to $(4.9 \pm 0.7)$ Oe. To investigate the ferromagnetism of the SRO, which, when grown on strontium titanate substrates, has a Curie temperature of 160 K [9], sample G is cooled down to 50 K and an in-plane hysteresis loop is taken. Nevertheless, besides the magnetization of the YIG, no additional ferromagnetic signal can be detected within the measurement accuracy.

V. FERROMAGNETIC RESONANCE

For FMR the samples are put face down on a coplanar waveguide, which is placed in a homogeneous external magnetic field. A small low frequency modulation field is added to allow for lock-in measurements. The magnetic radio frequency (RF) field resulting from an RF current in the waveguide is used for excitation. The magnetization of YIG is saturated along the external field with both external field and excitation field in plane but perpendicular to each other. The transmitted RF signal is converted to a voltage using a Schottky diode and fed into a lock-in amplifier whose output signal is measured by a nanovoltmeter. As a result the measured absorption curves show the first derivative of the absorption line. Figure 4(a) shows the resonance signal of sample Y at 9.6 GHz. The resonance signal is fitted with the first derivative of a Lorentz function to obtain the resonance field and the peak-to-peak linewidth. After multiplying by a factor of $\sqrt{3}/2$ we determine the linewidth of the absorption to be $(4.8 \pm 0.3)$ Oe [10–12]. This procedure is repeated for every resonance curve of every mentioned sample (see Table I). In Figs. 4(b) and 4(c) the resonance curves for samples E and C are shown, respectively. The respective linewidths after fitting are $(27.7 \pm 0.5)$ Oe (sample E) and $(13 \pm 1)$ Oe (sample C) at 9.6 GHz. The linewidths seem to fluctuate due to of the variation of YIG thicknesses. However, a strong increase of the linewidth compared to the bare YIG is a good indicator of spin pumping from YIG into the SRO in every sample. The permanent flow of spins from YIG to SRO reduces the precession angle and enhances the damping in the YIG layer, resulting in an increased linewidth [4,13–15]. In all of our SRO/YIG samples this effect can be observed. In Table I the linewidths of all measured samples with SRO on top of YIG at 9.6 GHz are listed.

For a detailed analysis the damping constant $\alpha$ is determined for sample Y, sample C, sample F, and sample H, respectively. Resonance curves are measured at different frequencies to obtain the linewidth-frequency dependence. Figure 5(a) displays this dependence for sample Y. The range of the frequency is varied from 2 GHz up to 24 GHz in 2 GHz steps in in-plane geometry and from 2 GHz up to 16 GHz in out-of-plane geometry. Due to inhomogeneities, the fluctuation of the linewidth is about 1 Oe. However, the damping can be determined from the slope, and in the in-plane configuration it amounts to $\alpha = (6.4 \pm 0.5) \times 10^{-4}$. 

FIG. 3. Hysteresis loop as measured by SQUID magnetometry for a 25 nm thick bare YIG (sample Y) after deposition.
FIG. 4. FMR measurements at 9.6 GHz. The linewidths: (a) sample Y with (4.8 ± 1) Oe, (b) sample E with (27.7 ± 1) Oe, and (c) sample C with (13 ± 1) Oe. The lines are the Lorentzian fits for each curve.

Out-of-plane analysis shows a similar value of $\alpha = (7.3 \pm 0.8) \times 10^{-4}$. The linewidth-frequency dependence of sample C is shown in Fig. 5(b). Here the damping is determined using a single fit, and it amounts to $\alpha = (3.7 \pm 0.4) \times 10^{-3}$. A similar value is observed for sample F, Fig. 5(c). In a range of 4 to 9.6 GHz, the calculated $\alpha$ is $(3.88 \pm 0.5) \times 10^{-3}$. Sample H shows a damping of $\alpha = (5.9 \pm 0.4) \times 10^{-3}$ [Fig. 5(d)]. The enhancement of $\alpha$ due to the SRO is at least one order of magnitude.

The variation of the YIG layer thickness of approximately ±25% may influence the damping enhancement by spin pumping as shown by Jungfleisch et al. [16] and thus hinders further quantitative analysis. Nevertheless, a plot of damping over nominal SRO thickness clearly shows a linear dependence (Fig. 6) despite the large error bars. This is at first glance surprising. Wahler et al. determined a spin diffusion length $\lambda = 1.7$ nm in SRO [5], and for layers much thicker than this the damping should no longer depend on the island thickness but only increase more or less linearly with the surface coverage. The dependence of the coverage on nominal layer thickness, however, should only be linear for small degrees of coverage and saturate when the layer starts to close completely. We have added a linear fit as a guide to the eye to Fig. 6. The fact that the deviations between measured data and fit are relatively small and that no saturation appears for thicker layers may be seen as an indicator that all layers are far from being closed, as expected also from our TEM investigation. This assumption is also supported by the sample resistances $R$ (Table I). As we can clearly see, the thicknesses of the respective SRO layers are not inversely proportional to the sample resistances, a fact which will be discussed later in detail together with the ISHE.

According to Tserkovnyak et al. the relation between $g_{\text{eff}}^{1\uparrow \downarrow}$ and the spin-mixing conductance $g^{1\uparrow \downarrow}$ is $1/g_{\text{eff}}^{1\uparrow \downarrow} = 1/g^{1\uparrow \downarrow} + R_{SD}$, where $R_{SD}$ is defined as the resistance per spin of the normal-metal layer of thickness $\lambda$ [13]. As discussed above, the condition of smooth thick layers is not fulfilled. Nevertheless we can estimate a lower limit for $g_{\text{eff}}^{1\uparrow \downarrow}$ using (1). Relating to $\alpha$ of sample H (nominal SRO thickness 15 nm) we find $g_{\text{eff}}^{1\uparrow \downarrow} \geq 9.6 \times 10^{18} \frac{1}{m^{2}}$, which is even larger than for a YIG/Pt interface, $1.3 \times 10^{18} \frac{1}{m^{2}}$ [17].
Using the in-plane Kittel formula we extract a gyromagnetic ratio of \(2.78 \pm 0.01 \text{ MHz/°C}\) and effective magnetization of \(166 \pm 10 \text{ emu/cm}^3\) for sample Y. The same analysis leads to a gyromagnetic ratio of \(2.82 \pm 0.01 \text{ MHz/°C}\) and an effective magnetization of \(166 \pm 10 \text{ emu/cm}^3\) of sample G. Further measurements in out-of-plane geometry show a saturation magnetization of \(127 \pm 14 \text{ emu/cm}^3\) of sample Y and \(133 \pm 15 \text{ emu/cm}^3\) of sample H using the out-of-plane Kittel formula. According to these results the SRO layer has no influence on the magnetocrystalline anisotropy of YIG. A comparison of the saturation magnetization determined by the Kittel formula and by SQUID magnetometry shows the same value within the measurement accuracy. Furthermore the out-of-plane anisotropy field amounts to 490 Oe, which is of the same order as that reported by Hauser et al. [18].

VI. INVERSE SPIN HALL EFFECT

To measure the ISHE the same setup is used. The ISHE is measured by contacting the edges of the sample, which are parallel to the signal line in the waveguide, using copper
wires as voltage probes. To allow for lock-in measurements the amplitude of the RF wave is modulated at a frequency of 197 Hz. In six samples that had previously shown FMR, the ISHE can be detected. The respective ISHE voltages $V_{\text{ISHE}}$ are shown in Table I. Similarly to the analysis of FMR, the data are fitted using a Lorentzian. The listed ISHE voltages are the respective amplitudes of the fitted curves. Figure 7 shows the data with the corresponding fit for sample C. The typical characteristics of the ISHE are clearly visible. The change of the polarity of the applied magnetic field results in a change of the polarity of the measured ISHE voltage. These characteristics can be observed on all samples with ISHE.

As expected from our findings for the layer morphology, the dependence of ISHE and nominal layer thickness is not straightforward. While the damping is simply determined by the surface coverage by the SRO and its thickness, the ISHE depends also on the in-plane resistance. Especially in the case of a layer that consists of islands, we need to distinguish several cases. For very low coverage typically the percolation threshold is not reached, and no conducting path exists between the two ends of the sample [Fig. 8(a)]. In this case no ISHE can be measured, although spin pumping may be present and even the damping can be increased. When more material is deposited, the density of islands and the island size can increase. At some point the percolation limit is reached and one or more conducting paths are created [Fig. 8(b)]. In this case each of the paths can create an ISHE voltage. The magnitude of this voltage, however, is difficult to determine. On the one hand the voltage depends on the inverse spin Hall current and the local resistance of the sample. Due to the island growth this resistance is nonuniform and may even vary between two conducting paths. On the other hand the conducting paths are not straight. They may even at some point run in opposite directions, in which case the locally generated ISHE voltages must be subtracted. For constant conductivity this subtraction should always yield a result similar to a straight path. If the segments that run in different directions, however, have different resistivities, the result is unpredictable. It can be higher or lower than the value for a straight path, depending on the island geometry. In addition all conducting paths, which normally would create different ISHE voltages, are now parallel and influence each other. As a result even the typical assumption of zero current flow is no longer valid. Only for a very large density of islands is an (almost) closed layer created, which can be treated as uniform, although its thickness is not well defined [Fig. 8(c)]. In this case we expect a ISHE that follows approximately the thickness dependence described by Azevedo et al. [15]. A good way to distinguish these three different regimes is to measure the sheet resistance for different samples. We measure the resistance between the two contacts, which are also used for the measurement of the ISHE. As the contacts are made using conducting silver glue, we must assume a certain error for the spacing between the contacts, which is $4.0 \pm 1.0$ mm. As the samples have a width of $2.1 \pm 0.1$ mm the sample area amounts to two squares and the measurements of the resistances yield the values given in Table I. We can clearly see the trend described above. For a sample of approximately 5 nm nominal thickness the resistance is several M$\Omega$, indicating that the percolation limit is barely reached. For 7.5 nm the resistance is already 50 times smaller. For a thickness of 9 nm the resistance is again reduced by a factor of almost 5, showing that at least multiple current paths exist and the layers start to close. For samples only 5 nm thicker the resistance is again smaller by a factor of 5. In this case the layers are almost closed, meaning that we no longer talk about an assembly of connected islands but about a layer with holes. It is interesting to note that one sample with a nominal thickness of 10 nm has a resistance which is closer to a 15 nm layer than to a layer of 9.8 nm nominal thickness. As transmission electron microscopy reveals, this layer for unknown reasons grew in a different morphology. Although the average thickness is less than for the 15 nm layer.
[Fig. 1(d)] we observe that the layer is much smoother. Because the resistance is dominated by the thin parts of the layer, this more uniform thickness results in a relatively low resistance. To check whether the ISHE at least follows the expected trends, we normalize the ISHE voltages to the sample resistance [16,19], resulting in an ISH current. Because all samples have the same size, the area of excitation is identical for all samples, and we do not want to make a quantitative analysis but just a comparison between samples, we can use the total resistance. Furthermore we assume the same susceptibility and excitation and we do not want to make a quantitative analysis but just a comparison between samples, we can use the total resistance. For the group of the three regimes mentioned above. For the high resistance in the different samples (Table I) we can at least differentiate the differences in layer thickness. For the normalized ISHE field for all samples, accepting large error bars because of the mismatches in the area of excitation, the SRO layers are conducting but not ferromagnetic. Spin pumping experiments show enhanced damping in the YIG clearly induced by spin pumping into the SRO. The SRO also shows a clear ISHE. Because the SRO grows in islands, the sample resistance changes along the coalescence of the islands with increasing layer thickness. This fact needs to be taken into account when analyzing ISHE, which may often be of consequence when ultrathin layers are used to detect the ISHE.

VII. CONCLUSION

In our experiments growing SRO on YIG using PLD results in polycrystalline SRO films, however with a clear texture, which relates to the substrate surface orientation. The SRO layers are conducting but not ferromagnetic. Spin pumping experiments show enhanced damping in the YIG clearly induced by spin pumping into the SRO. The SRO also shows a clear ISHE. Because the SRO grows in islands, the sample resistance changes along the coalescence of the islands with increasing layer thickness. This fact needs to be taken into account when analyzing ISHE, which may often be of consequence when ultrathin layers are used to detect the ISHE.

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