Applied Surface Science 589 (2022) 152854

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Full length article

Giant magnetoresistance response in Sr_2FeMoO_6 based organic spin valves

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ARTICLE INFO

Keywords:
Sr_2FeMoO_6
Alq_3
Spin valve
Magnetoresistance

ABSTRACT

We report the fabrication of the first Sr_2FeMoO_6 based organic spin valve device using Tris(8-hydroxyquinolinato) aluminum (Alq_3) as a spin transport layer. The characterization of the device confirms hysteretic magnetoresistance with approximately 20%–30% switching between high and low resistance states at low temperatures. The results demonstrate that organic semiconductors can form a suitable interface with double perovskite, half metallic Sr_2FeMoO_6, for efficient low temperature operation and have a potential to improve the room temperature performance significantly in tunneling devices where decay in spin diffusion length of organic layer does not affect the transport.

1. Introduction

Sr_2FeMoO_6 (SFMO) is a potential material regarding spintronics due to theoretically suggested 100% spin polarization, according to band structure calculations, and high Curie temperature (T_C) around 420 K [1]. SFMO based magnetic tunnel junctions (MTJ) have been reported in literature [2–4] with perovskite oxides as the tunnel barrier layer. For all these devices, a very complicated junction fabrication was crucial due to nanometer scale defects at the layer SFMO/oxide interfaces. Organic semiconductors (OS) offer great promise for incorporation with transition metals and ferromagnetic epitaxial perovskite metal oxides [5,6]. This is due to their structural flexibility and a highly spin-polarized interface with the ferromagnets, known commonly as the spinterface [5]. Additionally, due to their considerable spin diffusion time, a longer spin lifetime is expected of these devices [7]. Indeed, promising results have already been demonstrated in giant magnetoresistance (GMR) devices with manganite oxides, used as one of the ferromagnetic layers [8–11]. Although during the last decade, unquestionable improvements were reported in the field of OS spintronics, open questions and challenges remain. For example, despite the notable magnetoresistance at room temperature [5,12–16], magnetoresistive response is greatly diminished at room temperature and even at lower temperatures when compared with magnetoresistance at cryogenic temperatures [8,11–14,16–20]. Understanding the magnetoresistance phenomenon in these complex heterostructures and across hybrid interfaces remained an active research topic in the field of OS spintronics [5,21–23].

In OS spintronic devices $La_{1-x}Sr_xMnO_3$ (LSMO) is a commonly used halfmetallic material [5]. Other candidates used as the ferromagnetic electrode include different materials, with varying spin polarization, such as Fe, Co, Fe_3O_4, Co_2MnSi and NiFe [5]. In the current study, we provide the first report on GMR response from a SFMO based spin valve with an OS spin transport layer. The OS transport layer used here is the most widely reported small molecule semiconductor Tris(8-hydroxyquinolinato) aluminum (Alq_3)-layer. In addition, the spintronic community has demonstrated magnetoresistive results with prototypes combining ferromagnetic layers with organic semiconductors such as T_6, rubrene, fullerene, RRP3HT and CuPc [5]. However, the earlier work does not include prototypes with SFMO electrodes.

2. Experimental details

The SFMO layer was fabricated with pulsed laser deposition on a single crystal $SrTiO_3$ substrate. The deposition was conducted in a 9 Pa Ar-atmosphere with 2000 laser pulses, resulting in ≈150 nm thickness, at 900 °C. After the deposition, the film was patterned with UV-lithography technique. The patterned film consisted of 100 μm wide SFMO stripe, which was 4 mm in length.

The rest of the spin valve multilayer structure was deposited by thermal evaporation on top of the patterned SFMO electrode. The pressure in the evaporator chamber during the deposition was kept between 500 Pa and 1000 Pa. The thickness of the layers was monitored with a quartz microbalance resonator during the deposition. The Alq_3 layer thickness was later determined using S neox confocal microscope provided by Sensofar. First, the Alq_3 layer was deposited on top of the SFMO layer resulting with approximately 45 nm thickness, followed by a ferromagnetic 16 nm Co layer and a protective 2 nm Al layer. The

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https://doi.org/10.1016/j.apsusc.2022.152854
Received 1 June 2021; Received in revised form 9 February 2022; Accepted 14 February 2022
Available online 25 February 2022

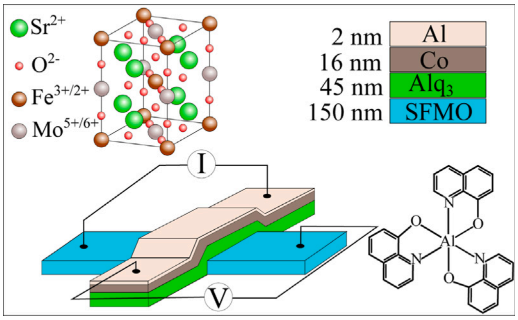

Fig. 1. A schematic illustration of a single spin valve junction along with Sr_2FeMoO_6 and Alq_3 structures.

deposition was performed with a slit mask resulting with the total of nine SFMO/Alq_3/Co/Al junctions. A schematic illustration of a single junction together with chemical structures of Alq_3 and SFMO crystal structure is presented in Fig. 1.

Resistive measurements were done using Quantum Design Physical Property Measurement System (PPMS) with resistivity option. Current/voltage characteristics were recorded with a separate Keithley 2614B SourceMeter. Resistive measurements with PPMS were performed at 10 K, 100 K, 200 K and 300 K temperatures between −500 mT and +500 mT magnetic field range. Magnetic field was applied along the film in-plane. Current transport measurements with Keithley were performed in zero magnetic field at 10 K, 100 K, 200 K and 300 K temperatures.

Magnetic measurements were done using Quantum Design MPMS SQUID magnetometer. The measurements included zero field cooled (ZFC) and field cooled (FC) magnetization measurements in 100 mT magnetic field between 10 K and 400 K temperatures. Magnetization hysteresis measurements with magnetic field were performed between −500 mT and +500 mT at 10 K, 100 K, 200 K, 300 K and 400 K temperatures. SQUID measurements were done with another SFMO thin film, patterned as the one used in multilayer fabrication. This sample lacks the following Alq_3, Co and Al layers. However, the actual multilayer device was also measured. An additional Co thin film was deposited on a glass plate and measured accordingly with SQUID magnetometer. Magnetic field was applied along the film in-plane. Coercive field (B_c) for SFMO was evaluated as an average of absolute magnetic field with zero magnetization from the hysteresis measurements. Remanence magnetization (M_{rem}) for SFMO was determined as an average of absolute magnetization value in zero magnetic field from the hysteresis measurements.

3. Results and discussion

3.1. Sr_2FeMoO_6 thin film and Co film on glass

In order to assess the quality of the SFMO and Co electrodes used in the spin valves, zero-field-cooled, field-cooled (ZFC and FC) and magnetic field hysteresis measurements for magnetization were conducted on identical SFMO and Co films. Fig. 2(a) shows the 100 mT ZFC and FC magnetizations of SFMO as a function of temperature. The results have been normalized according to FC magnetization at 10 K temperature and the diamagnetic background arising from the substrate and the sample holder has been subtracted from the signal. The results demonstrate a regular decay of magnetization with increasing temperature, along with ferri-paramagnetic transition at T_C. The inset in Fig. 2(a) shows the first order derivative of FC-magnetization as a function of temperature. The minimum of the first order derivative was used to define the T_C and the minimum falls close to 325 K, which is well within the range of our previously reported values [24,25]. The signal instability close to T_C (seen in the inset of Fig. 2(a)), is due to a distortion in the raw scan signal when the sum of the background and sample moments approaches zero. This causes a distortion in the original data. Fig. 2(b) shows the normalized magnetization hysteresis loops for the patterned SFMO thin film (blue) and the Co film (red) evaporated on a glass plate. The results for the SFMO stripe show the coercive field ($\mu_0 H_c$) of approximately 46 mT and 68% remanence magnetization (M_{rem}) from the saturation. The normalized magnetization hysteresis for Co at 10 K shows a large exchange bias, with $\mu_0 H_c$ values of −149 mT and 19 mT. This would set the bias field ($\mu_0 H_b$) to −65 mT. Determined from $\mu_0 H_b$, the value for M_{rem} is approximately 94% from the saturation. The large exchange field seen in Co magnetization hysteresis could be due to Co oxidation. This could lead to an exchange bias between the antiferromagnetic Co oxide and the metallic cobalt [26]. Since the magnetization hysteresis measurements were preceded by the ZFC/FC measurement, the hysteresis measurement corresponds to an FC hysteresis and therefore the exchange bias can emerge. Based on the field and temperature dependent magnetization measurement results obtained for the SFMO film, it can be argued that the film used in the spin valve prototype is similar in quality when compared with our other work regarding SFMO films [24,25].

Magnetoresistive measurements were done for a 100 μm wide SFMO stripe. The measured stripe was included in the SFMO/Alq_3/Co spin valve, but this measurement concerns only the SFMO stripe. The low field magnetoresistance (LFMR) results for the SFMO stripe used in the spin valve prototype are presented in Fig. 2(c). The relative magnetoresistance was calculated according to $MR = (R_H - R_{500\ mT})/R_{500\ mT}$, where R_H is the resistance in the applied field H and $R_{500\ mT}$ is the resistance in 500 mT magnetic field. The inset in Fig. 2(c) shows the LFMR responses between −80 mT and 80 mT with scalar offsets. The results at 10 K temperature show a clear hysteresis in the magnetoresistance, with maxima around –50 mT and 50 mT. From the zero field to ±500 mT, the sample shows a decrease in resistance, with LFMR of 5.3%. The LFMR signal diminishes with increasing temperature. The results demonstrate that 5.3% magnetoresistance at 10 K goes down to 1.9% at 100 K and to 0.8% at 200 K and to 0.7% at 300 K. While the LFMR is still visible at 100 K, the signal becomes negligible at 200 K and above. Thus, the spin dependent scattering and tunneling through grain boundaries are dominating the magnetoresistance signal at 10 K and 100 K in the SFMO stripe. Despite the proclaimed 100% spin polarization and demonstrated high Curie temperature above room temperature [1], the decay of magnetoresistance in SFMO and in similar materials is a well documented result. The explanations for this phenomenon could for example be due to the possible deterioration of grain boundary surface and film surface spin polarization [27,28]. Magnetoresistance results for SFMO thin films have been demonstrated in previous research publications by us [29–31] and by others [4,32,33]. Comparison between our previous data and the data presented here shows that previously magnetoresistance has been measured with a much higher magnetic field, up to ±8 T. Considering the applied field range here, the magnetoresistive response at 10 K is one of the highest compared to our previous results. The differences in research methods is simply due to different research interests. While the previous research has focused on magnetoresistance phenomenon including high field magnetoresistance, the interests here remain within the field range where GMR response is expected.

The magnetoresistance results presented for the SFMO stripe are typical responses in systems where the spin-dependent transport of charge carriers combined with magnetic hysteresis between grains induces scattering of charge carriers [1]. Our previous work confirms the fabrication of texturized and *c* axis oriented SFMO thin films on $SrTiO_3$ substrates with identical deposition conditions [25,34]. Therefore, the scattering of charge carriers probably takes place due to defects like low angle grain boundaries, or antiphase boundaries [33]. These results confirm that we do not have damages induced by the lithography and wet etching process of SFMO, which could have caused additional trapping sites causing increased LFMR.

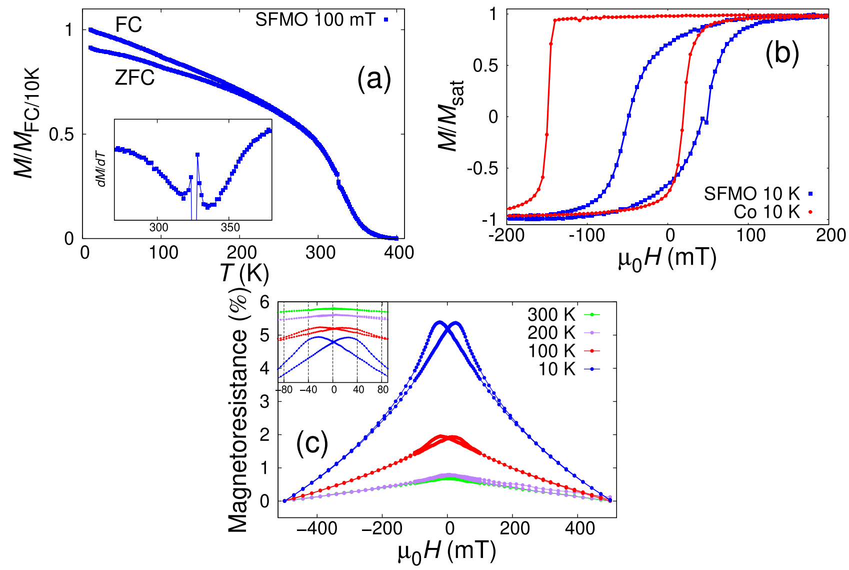

Fig. 2. (a) ZFC/FC measurement results for the patterned SFMO film. (b) Magnetization hysteresis measured at 10 K for the patterned SFMO film (blue) and the Co thin film (red) deposited on a glass plate. The considerable exchange bias in Co hysteresis is likely due to the oxidation of Co since the sample lacks the protective Al layer. (c) Magnetoresistive response from a 100 μm wide SFMO stripe, used in the spin valve structure, measured at 10 K, 100 K, 200 K and 300 K. The inset shows the magnetoresistance results with scalar offsets.

3.2. Sr_2FeMoO_6 based spin valves

In this work, we report the results for a typical multilayer junction with 100 μm SFMO stripe, named hereafter as S100. Fig. 3(a) and (b) present the magnetoresistance measurement results for S100 at 10 K and 300 K temperatures, respectively. The results of 10 K measurements show approximately 30% magnetoresistance. The magnetoresistance response showed decay with increasing temperature from 30% at 10 K, to 25% at 100 K, to 10% at 200 K and to 2% at 300 K, as shown in Fig. 3(c) as normalized magnetoresistance according to 30% magnetoresistance response at 10 K. Although the signal near the zero and 100 mT fields at 300 K, seen in Fig. 3(b), suggests higher magnetoresistance response, this signal takes place likely due to contact modification. This demonstrates a slight instability in the contacts connecting the spin valve prototype to the PPMS. The hysteresis in magnetoresistance loops is still visible at 100 K, but practically absent at 200 K. Although the change in resistance is still significant at this temperature around the zero field. However, no clear switching close to the zero field takes place at 300 K. The zero field resistance at 10 K was ≈1.2 MΩ.

Fig. 3(d) shows the data from Fig. 3(a) with field-cooled magnetic moment magnetic field hysteresis results (M-H) for the device. These results demonstrate the magnetic switching originating from SFMO and Co layers. Results for the hysteresis measurements performed at various temperatures are presented for SFMO film and for S100 in Fig. 3(e) and (f), respectively. The results for SFMO film show regular hysteretic magnetic field response in magnetic moment expected from SFMO thin film. The device shows one gradual and one sharp switching. Comparison with individual electrode's magnetic switching pattern (Fig. 2(b)) suggests the sharper switching originates from magnetization reversal of Co layer while the gradual transition might have its partial origin from SFMO magnetization reversal. The effect of exchange bias at 10 K temperature is apparent, but is clearly diminished at higher temperatures.

The presented magnetoresistive results are in a good agreement with the few existing previous publications for SFMO based tunneling magnetoresistance (TMR) junctions measured at low temperatures, where approximately 10%–50% magnetoresistance responses have been reported [2,3]. A slight asymmetry in the switching field in the results for S100 seen in Fig. 3 could be related to a partial Co oxidation resulting in an exchange bias between ferro- and antiferromagnetic layers. Since the vacuum condition between Alq_3 and Co deposition was never broken, our hypothesis suggests that there is insufficient protective Al layer, as the observed exchange bias in magnetic hysteresis measurements for the device suggests.

Despite the fact that multiple research groups have demonstrated possible spin valve operation at cryogenic temperatures in organic semiconductor devices, deposited of electrode materials with high T_C, much of the reported work show rather poor, or at least clearly diminished, magnetoresistance performance at elevated temperatures [5, see citations within]. Although research articles at the same time report relatively high room temperature magnetoresistance allegedly due to high T_C electrode [35], the loss of magnetoresistance performance has also been demonstrated in devices deposited of materials, which ideally should better preserve spin polarization at higher temperatures [17]. One of the reasons causing the magnetoresistance decay could be the loss of spin polarization at the ferromagnetic electrode and semiconductor transport layer interface [36,37]. This in our case would result with an insufficient spin injection in spite of high T_C, close to 325 K in our SFMO film. Indeed, research reports argue the loss of surface spin polarization in LSMO electrode as a contributing factor in the demise of device magnetoresistance response at high temperatures [36,37]. The decrease of SFMO magnetoresistance at high temperatures seen in Figs. 2(c) and 3(c) support this claim [11]. Although room temperature magnetoresistance has been detected in similar systems compared to our SFMO based system [38], device performance has remained far from ideal low temperature scenario [37–39]. This might also suggest that the surface spin polarization in our case is lost already at lower temperatures, since we do not detect considerable magnetoresistance at room temperature. In addition, other interfacial properties, for example metal molecule hybridization, are reported influencing the device magnetoresistance in various systems [40–42], which may also contribute to our device performance.

The decrease in spin diffusion length was directly demonstrated by Drew et al. with direct evidences of spin injection into organic transport layer [43]. These results also demonstrated that the spin diffusion length in Alq_3 decreases from 35 nm to nearly 10 nm when the temperature is increased from deep cryogenic to 50 K. In the event of spin injection in our device at high temperatures, from the high T_C electrode, the spin information would therefore be lost along with the

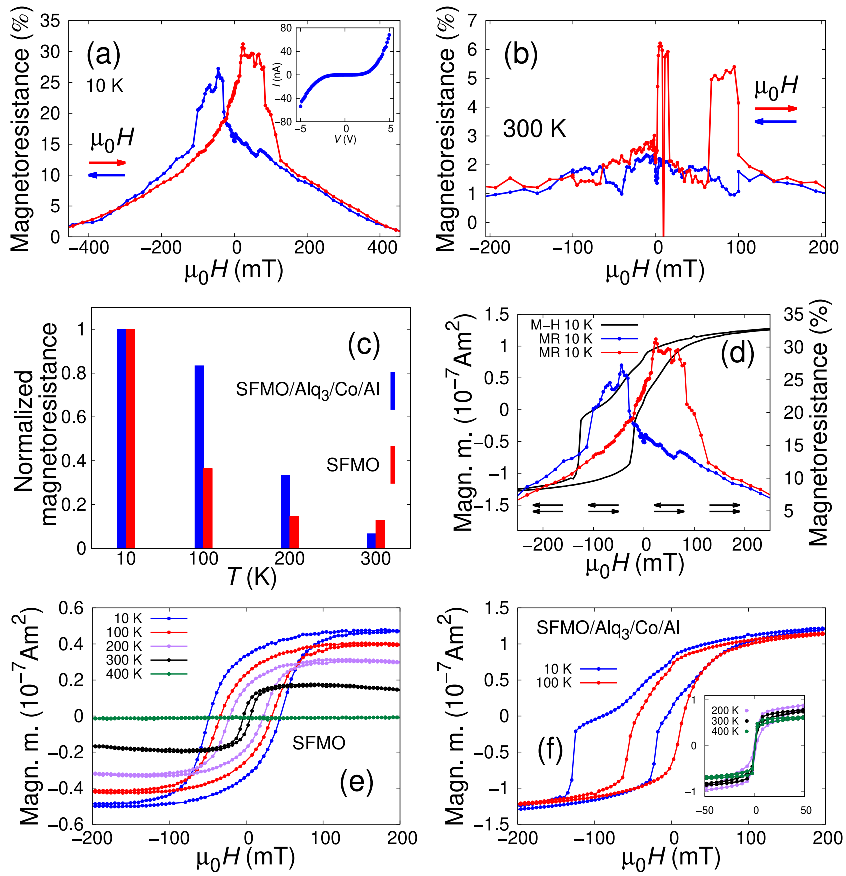

Fig. 3. (a) The magnetoresistive response of the multilayer junction with a 100 μm SFMO stripe at 10 K. The colors (blue/red) demonstrate the scanning direction of the magnetic field. The inset shows the zero field current/voltage characteristics. (b) The hysteretic magnetoresistance obtained at 300 K. Temperature dependence of normalized magnetoresistance signals are presented in (c) for SFMO film and multilayer junction. (d) Field-cooled magnetic moment field dependence measurement results (M-H) at 10 K are presented with the data from (a) (MR). Arrows are presented to illustrate magnetic switching of two ferromagnetic layers. (e) and (f) present magnetic field hysteresis measurements at various temperatures for SFMO film and S100, respectively.

hysteretic magnetoresistance. Spin relaxation in Alq_3 has been reported as a dominating mechanism behind magnetoresistance decrease in similar systems [44]. The probable contributors for spin relaxation include the spin orbit coupling and the hyperfine interaction [7,45,46]. It is worth noting that while in Alq_3 based GMR devices, the magnetoresistance decays consistently, in Alq_3 based magnetic tunneling junctions magnetoresistance shows more robust magnetoresistance retention at higher temperature [47]. However, due to the fundamental differences between conduction mechanisms and between device structures, the differences are as could be expected when considering spin relaxation in device transport layer [48]. In organic semiconductor based GMR devices the spin transport happens by drift or diffusion through the transport layer, while in TMR devices the transport takes place due to quantum mechanical tunneling of spin polarized carriers across the barrier layer. Therefore, the loss of spin diffusion length in the barrier layer does not effect the device response. To sum up the discussion around the poor magnetoresistance performance at elevated temperatures, we can say that possible explanations for the phenomenon include at least the demise of surface spin polarization at SFMO/Alq_3 interface despite the high T_C of the electrode materials and the decrease in spin relaxation length in organic transport layers. Same, although sometimes rather speculative, conclusions are widely addressed in the relevant literature [5].

Fig. 4 shows a schematic image about the discussion topics regarding the magnetoresistive results. While multiple research publications report a spin dependent transport of charge carriers between two ferromagnetic layers separated by a semiconductive organic transport layer, it is also reported that in similar systems a similar signal depicting spin transport could be achieved with only one ferromagnetic layer and applying a nonmagnetic counter electrode [49]. These findings have been argued through tunneling anisotropic magnetoresistance (TAMR) [49]. TAMR is a phenomenon requiring no spin-transport through an interface layer between electrodes and is directly related to magnetocrystalline anisotropy. In addition, some reports showed concern on the possibility of spin injection into organic semiconductors [50]. However, considerable evidence for spin injection has been provided [43]. The questions regarding the magnetoresistance and reported spin valve performance are far from obvious and remain practically unsolved and somewhat controversial. Indeed, more research is required in order to fully understand these topics. The polar coordinate image in Fig. 4 illustrates a common $\sin^2\alpha$ response in systems where TAMR gives rise to a spin valve-like operation in multilayer device when the angle α between magnetic field and crystalline axis is altered in magnetoresistance measurements. It remains inconclusive whether the measured magnetoresistance signal presented here originates from TAMR, GMR phenomenon or whether the signal is the sum of both mechanisms. This topic will be covered in more detail in future research projects.

Another interesting aspect is the sign of the magnetoresistance. On a first sight, our results would seem to contradict the previously reported negative magnetoresistance response of the Alq_3 spin valves. There are reports for both positive [10,11,39,51] and negative [8,9,37,39,51,52] magnetoresistance responses in OS spin valves with manganite oxides, the positive response being the increase in resistance with an antiparallel orientation of magnetic moments between two ferromagnetic layers. The early reports suggest the negative magnetoresistance response in the LSMO/Alq_3/Co spin valve can originate from the negative

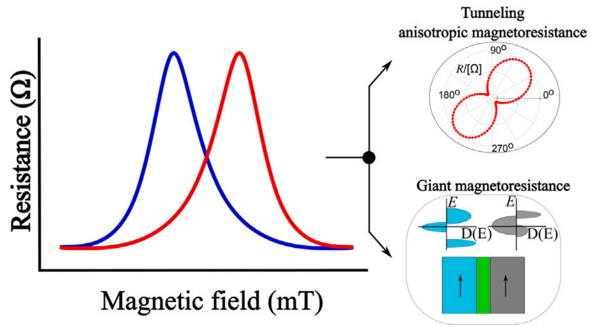

Fig. 4. Schematic image illustrating the discussion regarding the spin-valve-like magnetoresistance response. The results can be explained with giant magnetoresistance phenomenon but similar results can be understood as tunneling anisotropic magnetoresistance.

spin polarization of the Co layer while the LSMO spin polarization is positive [8].

In our experiment, we have demonstrated a resistance increase with an antiparallel magnetization alignment between the ferromagnetic (ferri) SFMO and Co layers separated by Alq_3. Earlier, Bibes et al. [3] reported a 50% positive TMR response from SFMO/$SrTiO_3$/Co MTJ whereas a negative 50% TMR response was recorded in LSMO/$SrTiO_3$/Co MTJ [53]. From this result, it was concluded that LSMO and SFMO have similar spin polarization value of nearly 80%, however, with an inverse sign. Since LSMO spin polarization is positive, SFMO spin polarization is negative. Therefore, our experimental results are in line with the previously observed sign of magnetoresistance in SFMO based junctions.

4. Conclusions

To summarize, we report the fabrication and magnetoresistive response of Sr_2FeMoO_6 based organic semiconductor spin valves. The results confirm hysteretic magnetoresistance with approximately 20%–30% switching between high and low resistance states at low temperatures. Large magnetoresistive switching between the parallel and antiparallel states decreases significantly with increasing temperature. The results presented here demonstrate that the spin valve devices with SFMO as the spin injection and organic semiconductor as the spin transport layer are a promising combination for high-efficiency spintronic components. With optimization, further improvement in room temperature magnetoresistance response is expected making SFMO an important choice as spin polarized electrode for room temperature operating spintronic devices.

CRediT authorship contribution statement

I. Angervo: Conceptualization, Investigation, Writing – original draft. **M. Saloaro:** Writing – review & editing. **H. Palonen:** Writing – review & editing. **H. Huhtinen:** Supervision, Writing – review & editing. **P. Paturi:** Resources, Supervision, Writing – review & editing. **T. Mäkelä:** Investigation, Resources. **S. Majumdar:** Conceptualization, Resources, Supervision , Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Jenny & Antti Wihuri foundation and Turku University foundation are acknowledged for financial support. We thank M. Sc Asko Sneck for his assistance in Alq_3 layer thickness measurements. S. Majumdar acknowledges the Academy of Finland (Project 13293916) for financial support.

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