Spin current and magnetic measurements in heterostructure SrIrO$_3$/La$_{0.7}$Sr$_{0.3}$MnO$_3$

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Abstract. The dc voltage on the film SrIrO$_3$ (SIO) generated under ferromagnetic resonance condition has been studied in the heterostructure based on manganite thin epitaxial films La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO). The SIO film is a paramagnetic metal with strong spin-orbit interaction in the temperature range 20-300K. The effect is shown to be caused by two different phenomena: (1) the resonance dc electromotive force related to anisotropic magnetoresistance (AMR) in the manganite film and (2) pure spin current (spin pumping) registered by means of the inverse spin Hall effect in upper layer. We also give the results of the magnetic parameters measuring for the heterostructure SIO/LSMO using ferromagnetic resonance in a wide temperature range.

1. Introduction

The 3d transition metal oxides (TMOs) have various functionalities caused by the presence of strong electron-electron correlation. The pronounced spin-orbit coupling has attracted attention in recent years due to the emergence of new topological states [1] and applications in spintronics [2]. The contact between 3d and 5d TMOs provides unique interface. At the 5d TMOs interface with a ferromagnet may occur a violation of topological symmetry in the region of the interface and the occurrence of a gap in the excitation spectrum which in turn can lead to fairly strong magnetoelectric effects [3].

The pure spin current is one of the most interesting and prospective effects. This phenomenon occurs in bilayer structures in which the ferromagnetic layer (FM) contacts with the normal metal layer (NM). The FM layer is in the conditions of ferromagnetic resonance (FMR) [4, 5]. The pure spin current is not related to transfer of electric charge and corresponds to the transfer of the spin momentum from the excited FM to adjacent NM. Spin pumping has been demonstrated in various combinations of materials, even the magnetic layer may be insulator (e.g., yttrium iron garnet). The nonmagnetic conductor may be a transition metal, semiconductor, conductive polymer, or topological insulator [6-12].

In this paper, we present the experimental results of the studying spin current at the interface and magnetic characteristics of heterostructures SrIrO$_3$/La$_{0.7}$Sr$_{0.3}$MnO$_3$. SrIrO$_3$ is interesting due to the presence of strong electron-electron and spin-orbit interaction at the same time. As a result, unusual resistive, ferromagnetic and magnetoelectric phenomena appear. Effect of a strong spin-orbit
interaction in SrIrO$_3$ makes it possible to obtain an increase in the magnitude of the spin current flowing through the interface in heterostructure.

2. Samples and experimental technique

The epitaxial films La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) were deposited by magnetron sputtering on the (110) plane of a single-crystal substrate of NdGaO$_3$ (NGO) with the sizes 5x5x0.5 mm$^3$. 10 nm of SrIrO$_3$ (SIO) were sputtered in situ just after LSMO film deposition. The SIO films was found to have the orthorhombic Pnma structure with lattice parameters a = 0.559 nm, b = 0.788 nm and c = 0.556 nm, which can be approximately considered as a "pseudo cub" with the parameter a = 0.398 nm. The structure of the LSMO film can be considered "pseudocubic" with the parameter a = 0.389 nm [13].

The magnetic characteristics were measured by magnetic resonance technique using ER-200 EPR spectrometer from Bruker (frequency 9.76 GHz). A synchronous detection was used at the modulation of an external magnetic field with the frequency 100 kHz. The samples under study were located in the microwave cavity of the spectrometer in such a way that the plane of the sample was always parallel to the direction of the dc external magnetic field and the magnetic component of the microwave field (parallel orientation). The parallelism of the plane of the samples to a constant magnetic field was controlled by the minimum of the resonance value of the field of the ferromagnetic resonance lines. This arrangement of the samples eliminated the change in the magnetic resonance spectra due to shape anisotropy. Rotation of the samples was carried out around an axis perpendicular to the plane of the substrate.

3. Magnetic parameters of the films

In the absence of any external action the magnetization vector $\vec{M}$ in a ferromagnet is directed along the effective magnetic field $\vec{H}_{\text{eff}}$. There are several contributions to $\vec{H}_{\text{eff}}$ such as the external magnetic field and the magnetic anisotropy fields of the sample. If the magnetization is deflected from the direction $\vec{H}_{\text{eff}}$ then the motion of the vector $\vec{M}$ in the ferromagnet is described by the Landau–Lifshitz (LL) equation. As a result, the magnetization precesses around the $\vec{H}_{\text{eff}}$ vector with a frequency determined both by the magnitude of the external magnetic field and by the values of the magnetic anisotropy parameters. In this case, the precession is damped due to various relaxation processes. Most often one uses the relaxation process proposed by Gilbert, who phenomenologically added a term to the LL equation that causes the precession decay [14]. As a result, a rule, the motion of the vector $\vec{M}$ in ferromagnets, taking into account the damping, is described by the Landau-Lifshitz-Hilbert equation (LLG) in the form:

$$\frac{d\vec{M}}{dt} = -\gamma \vec{M} \times \vec{H}_{\text{eff}} + \alpha \vec{M} \times \frac{d\vec{M}}{dt}.$$  

(1)

Here $\alpha$ is the Gilbert damping coefficient, $\gamma$ is the gyromagnetic ratio, $H_{\text{eff}}$ is the local effective magnetic field including the external, demagnetization and magneto anisotropy fields. Significant relaxation processes in thin ferromagnetic films [15] are spin-orbit relaxation damping, two-magnon scattering and eddy current damping.

The method for determining the parameters of the magnetic anisotropy consists in processing the angular dependences of the resonant fields of the FMR spectra. The solution of the Landau-Lifshitz equation for the evolution of the magnetization $M$ in an external constant magnetic field $H$ under the action of the magnetic component of the radio-frequency field is used, which gives an analytic relation for the resonance field $H_0$ and the frequency $\omega$ [16]

$$\left( \frac{\omega}{\gamma} \right)^2 = \left( 4\pi M_0 + H_0 + \frac{2K_1}{M_0} \cos^2 \varphi_a + \frac{2K_2}{M_0} \frac{1 + \cos^2 2\varphi_a}{2} \right) \left( H_0 + \frac{2K_1}{M_0} \cos 2\varphi_a + \frac{2K_2}{M_0} \cos 4\varphi_a \right).$$

(2)
Here $M_0$ is equilibrium magnetization, $K_u$ and $K_c$ are the uniaxial strain and the biaxial cubic anisotropy constants correspondingly. $\varphi_u$ and $\varphi_c$ are the angles between the easy axis of the uniaxial and cubic anisotropies and the external magnetic field correspondingly. $K_u$, $K_c$, $M_0$ as well as angles $\varphi_u$ and $\varphi_c$ were obtained by the fitting of the angular dependence (see figure 1).

Figure 1 shows the angular dependence of the resonance value of the magnetic field $H_0$ for the SIO/LSMO heterostructure at room temperature and external magnetic filed is rotated around the normal to the film plane by an angle $\varphi$. The angle was measured from one of the facets of the substrate. The external magnetic field and the magnetic component of the microwave field were in the plane of the film. The change of resonant field when the angle changes is due to the planar magnetic anisotropy of the SIO/LSMO heterostructure. The angular dependence was described by a resonance relation (2) taking into account magnetic uniaxial and cubic plane anisotropies [16].

![Figure 1](image-url)

**Figure 1.** Angular dependence of resonance field for SIO/LSMO heterostructure at $T=300$ K. Dots are measured data. The solid line demonstrates a fit of experimental data by Equation (1) that gives the following magnetic parameters of LSMO film: $M_0=300$ Oe, $H_u=69$ Oe, $H_c=11$ Oe.

4. The linewidth of FMR

The initial and final points of which $\Delta H$ measured constitute the position of the maximum $H_p$, and minimum $H_p$ of the first derivative $dP/dH$ of the high-frequency field absorption signal (see figure 2). The resonance field ($H_0$), defined as the point where $dP/dH$ changes sign. The deposition of a SIO film on top of LSMO film increases $\Delta H$ (figure 2).

We deposited SIO layer on LSMO to obtain and the efficiency of generation of spin current at the interface of a ferromagnetic 3d TMO and 5d TMO. Using the following expressions, we calculated the spin mixing conductance in heterostructure SIO/LSMO.
Figure 2. FMR line (dP/dH) for single LSMO film (filled squares) and SIO/LSMO heterostructure (open circles). The increasing of linewidth is clearly observed. The spectra were shifted relative to the ordinate axis.

\[ g_{\text{eff}}^{\uparrow\downarrow} = \frac{4\pi g_f M_s t_{\text{LSMO}}}{g\mu_B \omega_f} \left( \Delta H_{\text{SIO/LSMO}} - \Delta H_{\text{LSMO}} \right). \] (3)

For the following parameters: \( M_s = 300 \text{ Oe} \) is the magnetization LSMO film, \( t_{\text{LSMO}} = 12 \text{ nm} \) is the thickness for LSMO film, \( \mu_B = 9.274 \times 10^{-21} \text{ erg/G} \) is the Bohr magneton, \( g = 2 \) is the Lande factor, \( \gamma = 17,605 \times 10^{16} \text{ s}^{-1} \text{ G}^{-1} \) is the gyromagnetic ratio for electron, \( \omega_f = 2\pi \times 9.51 \times 10^{9} \text{ s}^{-1} \) is the microwave angular frequency in our case. At the room temperature we got \( \Delta H_{\text{SIO/LSMO}} - \Delta H_{\text{LSMO}} = 20 \text{ Oe, } g_{\text{eff}}^{\uparrow\downarrow} = 0.95 \times 10^{18} \text{ m}^{-2} \) for SIO/LSMO heterostructures. For comparison for interfaces SrRuO\(_3\)/La\(_{2/3}\)Sr\(_{1/3}\)MnO\(_3\), Pt/Ni\(_{80}\)Fe\(_{20}\) and Pt/YIG (YIG is yttrium iron garnet) were obtained \( g_{\text{eff}}^{\uparrow\downarrow} = 5 \times 10^{18} \text{ m}^{-2} \) [17], \( 2.1 \times 10^{19} \text{ m}^{-2} \) [18] and \( 4.8 \times 10^{20} \text{ m}^{-2} \) [19] correspondingly.

5. The temperature dependence

Temperature dependence of FMR spectrum and in particular the width of the resonance lines as well as other parameters in the SIO/LSMO heterostructure was measured. Possible sources of additional damping could be the sputtered of SIO film on LSMO. The spin pumping mechanism creates a spin current from a ferromagnet to a normal metal. As a result, an additional channel of torque outflow from the ferromagnet is formed, which leads to an increase in the damping parameter [18-21].

The figure 3 shows the temperature dependence of the resonant field \( H_0 \) with the external magnetic field \( \mathbf{H} \) directed along the hard axis of the in-plane uniaxial magnetic anisotropy of LSMO. Such a direction of the external magnetic field was chosen from the condition of the minimal contribution of magnetic anisotropy to the resonance relation for FMR (see Equation 2). In this approximation, it can be seen that a decrease in the value of the resonant field is due to an increase of \( M_0 \) parameter in the Equation 2.

From an analysis of the temperature dependences of resonance field \( H_0 \) (figure 3), it follows that at a temperature about 50 K the resonant field decreases sharply that means increase \( M_0 \) parameter. So we revealed the influence of SIO layer on the FMR spectrum of the LSMO film. A similar increase in the \( M_0 \) parameter of the LSMO layer at temperatures below 150 K in the LSMO/SRO heterostructure (here SRO is SrRuO\(_3\)) was observed in [22] and interpreted as the appearance of interlayer exchange interaction [23] after the SRO layer passed into ferromagnetic state. For clarity, we demonstrate the
temperature dependence of the resonant field for a single film, on which it is seen how, starting from 110K, the resonant field, due to saturation of magnetization, stops changing.

![Figure 3](image.jpg)

**Figure 3.** Temperature dependence of FMR resonance field for LSMO (filled squares) film (40 nm) and SIO/LSMO (filled pentagons) bilayer with a layer thickness of iridium 10 nm and manganite 12 nm.

### 6. Spin current

For the detection of spin current we used the method based on inverse spin Hall effect (ISHE) [24]. In the material with strong spin-orbit interaction a spin current gives rise to an electric current. The relation between spin and electric currents is determined by the dimensionless spin Hall angle $\theta_{\text{SH}}$:

$$J_{\text{ISHE}} = \theta_{\text{SH}} \frac{e}{h} [\vec{H} \times \vec{J}_s].$$

where $\vec{H}$ is a unit vector in the direction of the spin momentum flow.

The sample LSMO/SIO is a strip with electric silver contacts at the edges for the voltage measurements. It was placed in the central plane of the rectangular $TE_{102}$ microwave cavity. The microwave pumping was produced by Gunn diode with output up to 130 mW and at the frequency 9 GHz. The signal $V(H)$ was accumulated by sweeping the external field $H$ across the resonance. By rotating the external field $H$ in the film plane we measured the voltage for angles in the range 180 degrees with 10 degrees step.

The typical signal $V(H)$ detected at the SIO film is shown at figure 4. Spin current is converted into electric current due to ISHE. In turn, this current creates the dc voltage. In addition, one should take into account dc voltage arising due to the presence of anisotropic magnetoresistance (AMR) in FM layer. As a result, the full dc voltage should be written as

$$V(H, \phi_0) = V_{\text{SP}} \cdot L(H) \cdot \cos \phi_0 + [V_{\text{AMR}}^{\text{Sym}} \cdot L(H) + V_{\text{AMR}}^{\text{Antisym}} \cdot L'(H)] \cdot \sin 2\phi_0 \cdot \sin \phi_0.$$  

Here FMR absorption line $L(H) = (\Delta H)^2/[(H - H_0)^2 + (\Delta H)^2]$ is the symmetric Lorentzian function with the resonance field $H_0$ and half-width $\Delta H$. $L'(H) = \Delta H(H - H_0)/[(H - H_0)^2 + (\Delta H)^2]$ is an antisymmetric function [17,21].

The voltage $V_{\text{SP}}(H, \phi_0)$ and $V_{\text{AMR}}(H, \phi_0)$ changes sign upon inversion of the H direction. Since parasitic contribution is constant for the opposite orientations of the magnetic field we use the
difference for the signals with opposite orientations of the magnetic field in order to exclude this parasitic contribution.

In order to divide the symmetric signal into the effects of spin pumping and anisotropic magnetoresistance we measured the voltage dependence upon the angle $\phi_0$ between directions of charge current and the magnetic field $H$ (the magnetization is parallel of $H$ in our case). By fitting the angular dependence with formula (5) we obtain the following ratio between amplitude of the symmetric signal of anisotropic magnetoresistance and signal of spin pumping:

$$\frac{V_{SP}}{V_{Sym}} = 0.23 \pm 0.07$$

(6)

Figure 4. The voltage arising in the SIO/LSMO heterostructure during the magnetic field swiping at room temperature (blue points). The dashed green and red lines represent symmetric and antisymmetric part of the AMR signal and magenta line represents signal from the spin pumping. Solid blue line is the sum of three contributions (see Equation 5).

Figure 4 demonstrates how the experimental response is described by the sum of two contributions due to the spin current through the interface and the anisotropic magnetoresistance in LSMO.

7. Conclusion
We have measured the dc voltage caused by spin pumping and the anisotropic magnetoresistance in NM/FM sample, with FM made from 3d ferromagnets (LSMO), and the top layer (NM) from 5d paramagnetic (SIO). The temperature dependence of FMR linewidth and magnetic parameters of SIO/LSMO bilayer was investigated. From FMR linewidth obtained for the LSMO film and iridate heterostructure with SIO on top of epitaxial LSMO film the spin mixing conductance was determined. In the temperature range from 60 to 20 K, a sharp decrease in the value of the resonant field for the heterostructure under study was observed. One of the possible causes may be the appearance of ferromagnetism in the upper paramagnetic SIO film.

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