

Interfacial Rashba magnetoresistance of the two-dimensional electron gas at the LaAlO₃/SrTiO₃ interface

Kulothungasagaran Narayanapillai,¹ Gyungchoon Go,² Rajagopalan Ramaswamy,¹ Kalon Gopinadhan,¹ Dongwook Go,³ Hyun-Woo Lee,³ Thirumalai Venkatesan,^{1,4} Kyung-Jin Lee,^{2,5,*} and Hyunsoo Yang^{1,†}

¹*Department of Electrical and Computer Engineering, NUSNNI, National University of Singapore, Singapore 117576, Singapore*

²*Department of Materials Science and Engineering, Korea University, Seoul 02841, Korea*

³*PCTP and Department of Physics, Pohang University of Science and Technology, Pohang 37673, Korea*

⁴*Department of Physics, National University of Singapore, Singapore 117542, Singapore*

⁵*KU-KIST Graduate School of Converging Science and Technology, Korea University, Seoul 02841, Korea*

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We report the angular dependence of magnetoresistance in the two-dimensional electron gas at the LaAlO₃/SrTiO₃ interface. We find that this interfacial magnetoresistance exhibits a similar angular dependence to the spin Hall magnetoresistance observed in ferromagnet/heavy metal bilayers, which has been so far discussed in the framework of the bulk spin Hall effect of the heavy metal layer. The observed magnetoresistance is in qualitative agreement with a theoretical model calculation including both Rashba spin-orbit coupling and an exchange interaction. Our result suggests that magnetic interfaces subject to spin-orbit coupling can generate a non-negligible contribution to the spin Hall magnetoresistance, and the interfacial spin-orbit coupling effect is therefore key to the understanding of various spin-orbit-coupling-related phenomena in magnetic/nonmagnetic bilayers.

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I. INTRODUCTION

Recent advances in deposition techniques have enabled film growth control at the molecular level with atomic precision. These advances allow us to study exotic oxide materials with interesting properties. Since the discovery of two-dimensional electron gas (2DEG) formation at the interface of two insulating materials, LaAlO₃ (LAO) and SrTiO₃ (STO) [1], the LAO/STO system has emerged as one of the central material systems in the oxide community as it exhibits intriguing properties including superconductivity [2,3] and ferromagnetism [4–7]. The ferromagnetism, which occurs on the Ti site at the interface, has been evidenced by various measurement techniques such as scanning superconducting quantum interference device (SQUID) [6,8], x-ray magnetic circular dichroism [7], torque magnetometry [9], and magnetotransport [10–16] driven by the Rashba spin-orbit coupling (SOC). Furthermore, broken inversion symmetry at the interface results in the Rashba SOC which can be tuned by a gate voltage [15,16]. Therefore, the LAO/STO interface is subject to both interfacial Rashba SOC and exchange coupling.

Magnetoresistance (MR) is a fundamental means to investigate charge and spin transport in condensed matter systems. In magnetic systems in the presence of SOC, the longitudinal resistance depends on the magnetization direction, e.g., anisotropic MR [17]. Recently, another type of angle-dependent MR, called spin Hall MR, was observed in ferromagnet (FM)/heavy metal (HM) bilayers [18–20]. Including both anisotropic MR and spin Hall MR, the longitudinal resistivity is given as

$$\rho = \rho_0 + \Delta\rho_1 m_x^2 - \Delta\rho_2 m_y^2, \quad (1)$$

where ρ_0 is the magnetization-direction-independent resistivity, $\Delta\rho_1$ ($\Delta\rho_2$) is the longitudinal resistivity change due to the anisotropic MR (spin Hall MR), and m_x (m_y) is the normalized magnetization component longitudinal (transverse) to the current-flow direction in the film plane. The original theory [21] described the spin Hall MR as a consequence of the bulk spin Hall effect (SHE) in HM by assuming no interfacial SOC effect. Recent theories, however, have predicted an important role of the interfacial SOC at the FM/HM interface in the spin Hall MR [22,23].

Furthermore, several experiments on FM/HM bilayers suggested that the role of interfacial SOC should be carefully examined. For example, it was reported for spin pumping and inverse SHE experiments on Co/Pt bilayers [24] that the total dissipated transverse spin current from the Co layer (measured through the effective damping) is substantially different from the spin current absorbed in the bulk part of the Pt layer (measured through inverse SHE). This difference was ascribed to spin memory loss [25] describing the spin flipping due to SOC at the Co/Pt interface [26]. There were also spin-orbit torque experiments that cannot be explained by the bulk SHE mechanism alone but require an essential role of the interfacial SOC effect [27–30]. A recent experiment [31] also found a close correlation, predicted for interfacial SOC [32], between the Dzyaloshinskii-Moriya interaction and a fieldlike spin-orbit torque [32].

Recent theories also suggested that the interfacial SOC effect is important for various SOC-related phenomena in FM/HM bilayers. A first-principles approach [33,34] showed that both SHE and inverse SHE are largely enhanced at the FM/HM interface, i.e., a manifestation of the interfacial SOC effect. Boltzmann transport calculations also suggested the importance of the interfacial SOC effect for various spin transport phenomena [35–37].

Consequently, it is natural to raise a question about the role of interfacial SOC in the spin Hall MR. Answering this

*kj_lee@korea.ac.kr

†eleyang@nus.edu.sg

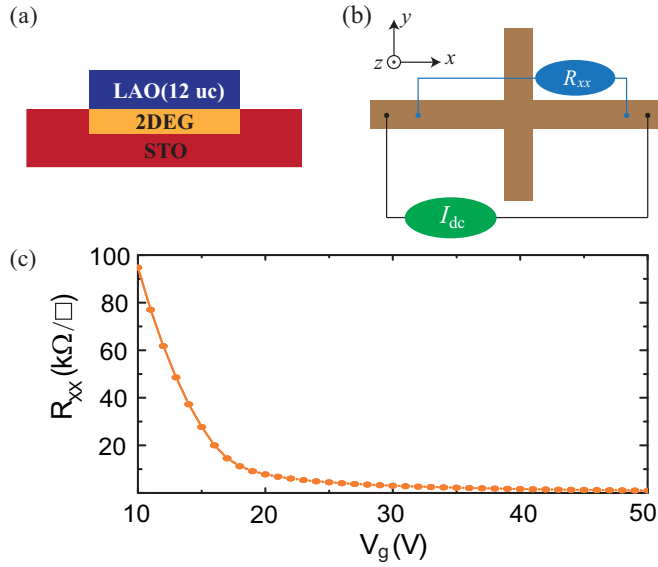


FIG. 1. (a) Schematics of LAO/STO stack layers, (b) Hall bar device with a measurement schematic for R_{xx} , and (c) the sheet resistance R_{xx} as a function of gate voltage V_g . At low gate voltages, the sample is insulating and the resistance monotonically decreases with increasing V_g .

question is of critical importance not only for understanding the underlying physics of SOC-related phenomena in FM/HM bilayers but also for enhancing the SOC effects for applications. For this purpose, we investigate the angular dependence of MR in 2DEG formed at the LAO/STO interface which has interfacial Rashba SOC and ferromagnetism. Unlike FM/HM bilayer structures where both bulk and interfacial SOC contributions coexist and it is therefore hard to differentiate one from the other, the LAO/STO 2DEG is an ideal system to investigate a pure interface effect because it is a single interface just as the FM/HM interface but does not have the bulk SHE.

II. EXPERIMENTS

The devices were prepared as follows. As shown in Fig. 1(a), LAO (12 unit cells; 1 u.c. = 0.379 nm) was grown on a TiO_2 terminated atomically smooth STO (001) single-crystalline substrate, which was pretreated with a buffered oxide etch and air annealed at 950 °C for 1.5 h, by pulsed laser deposition (PLD) at 750 °C in an oxygen partial pressure of 1 mTorr. The growth was monitored by *in situ* reflective high-energy electron diffraction. The sample was postannealed at 750 °C in the presence of oxygen in order to remove oxygen vacancies. Electron beam lithography was utilized to define the Hall bar structure using a negative tone resist. A blanket AlN_x was deposited by PLD at room temperature, followed by a lift-off process. Thereby, a 2DEG LAO/STO interface was formed only at the defined device structure. The Hall bar channels were designed parallel to the sample edge to align the channels along the crystallographic axis (001).

A measurement scheme for longitudinal resistance (R_{xx}) is shown in Fig. 1(b). The electrical transport characterizations

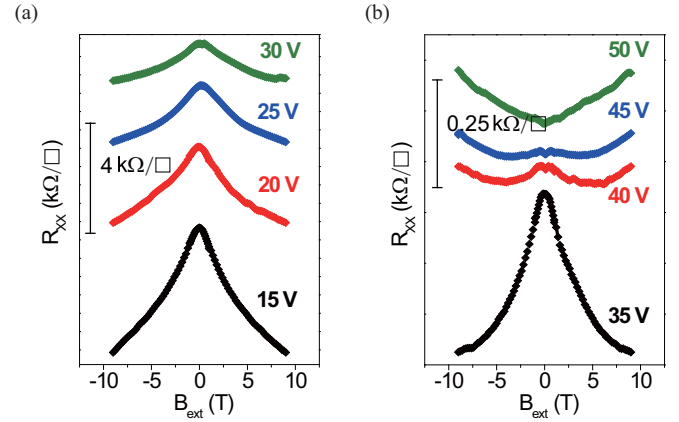


FIG. 2. Gate voltage dependence of R_{xx} . Perpendicular magnetic field dependence of R_{xx} for (a) $V_g = 15, 20, 25,$ and 30 V, and (b) $V_g = 35, 40, 45,$ and 50 V.

were performed in a physical property measurement system with a rotator used for angle-dependent MR measurements. We performed MR measurements as a function of various parameters including magnetic field B_{ext} , gate voltage V_g , and the rotation plane of the magnetic field. For the gating, a back-gate voltage (V_g) was applied through STO as the dielectric and silver as the back-gate contacts.

In Fig. 1(c), we depict a typical R_{xx} vs V_g curve at a temperature T of 2 K. We observe a monotonic decrease in R_{xx} with increasing V_g , showing that the device characteristic changes from insulating to conducting as V_g increases. Figure 2 shows the perpendicular magnetic field (B_{ext}) dependence of R_{xx} at various gate voltages. At low gate voltages ($V_g < 40$ V), R_{xx} decreases with increasing $|B_{ext}|$, which results from weak localization (WL). At high gate voltages ($V_g > 45$ V), on the other hand, R_{xx} increases with increasing $|B_{ext}|$, which implies that the WL correction is subdominant in conducting regimes.

Figure 3 shows a representative result of the angle-dependent R_{xx} for three rotation angles of B_{ext} , i.e., α rotation in the xy plane, β rotation in the yz plane, and γ rotation in the zx plane (see Fig. 3 for the definition of the angles). Here, the current is always applied in the x direction. We observe that

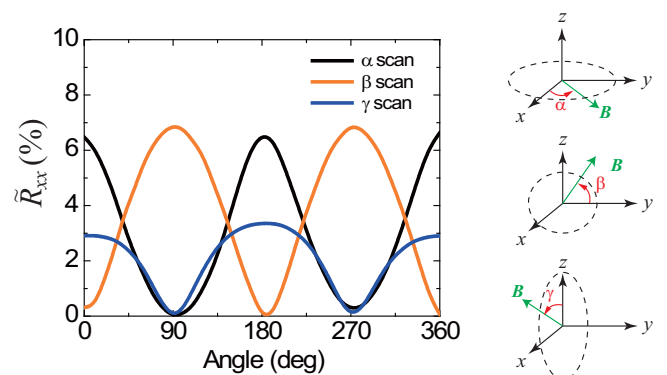


FIG. 3. Measured angular dependence of the normalized MR (\tilde{R}_{xx}) as a function of the rotating angle (α , β , and γ) with an applied field of $B_{ext} = 9$ T and gate voltage $V_g = 45$ V.

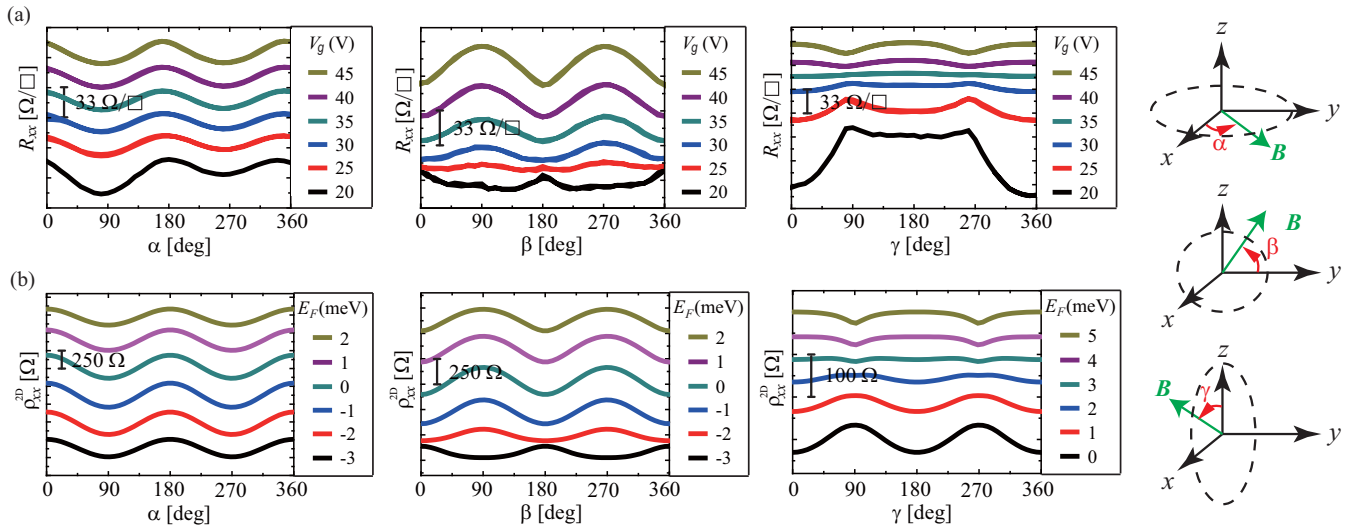


FIG. 4. (a) Experimental results of angular dependence of R_{xx} as a function of α , β , and γ rotations with various V_g , and applied field $B_{\text{ext}} = 9$ T. (b) Theoretical calculations for angular dependence of the resistivity ρ_{xx}^{2D} as a function of α , β , and γ rotations with various Fermi energies. The curves are offset along the y axis for clarity.

the normalized MR [$\equiv \tilde{R}_{xx} = (R_{xx}^{\text{max}} - R_{xx}^{\text{min}})/R_{xx}^{\text{min}}$] is about 7% in the α and β rotations, while it is about 3% in the γ rotation [38]. An important observation is that the angle-dependent change in \tilde{R}_{xx} is nonzero for the β rotation. As the LAO/STO interface has no contribution from the bulk SHE, this noticeable change in \tilde{R}_{xx} for the β rotation proves that the $\Delta\rho_2$ term of Eq. (1) is nonzero for the LAO/STO interface even without the bulk SHE. This angle-dependent MR at the LAO/STO interface can be called the interfacial Rashba MR as it originates from the Rashba SOC at the interface. Furthermore, it is noteworthy that the interfacial Rashba MR is much larger than the reported spin Hall MR of FM/HM structures (0.01%–1%) [18–20].

We further measured the angular dependences of interfacial Rashba MR for the three rotations at various back-gate voltages [Fig. 4(a)]. In the α (β) rotation, the interfacial Rashba MR in general follows $\cos^2 \alpha$ ($-\cos^2 \beta$), consistent with Eq. (1). An exception appears for $V_g = 20$ V. At this gate voltage, R_{xx} in the α rotation is asymmetric between 90° and 270° . This asymmetry is attributed to a non-negligible contribution from current-induced spin polarization, as reported previously [12]. For a detailed description of the current-induced spin polarization to the magnetoresistive effect, see Ref. [39]. In the β rotation, on the other hand, the sign of interfacial Rashba MR changes at $V_g = 20$ V. This sign change is attributed to the WL correction as the β rotation involves the contribution of the out-of-plane component of the magnetic field B_{ext} and the WL correction to R_{xx} becomes significant at low gate voltages (see Fig. 2).

III. THEORETICAL ANALYSIS

In order to understand the interfacial Rashba MR, we compute charge transport in the 2DEG at the LAO/STO interface. As an effective model which captures the qualitative features of the experimental results, we choose a simple model

Hamiltonian \mathcal{H} as

$$\mathcal{H}(\mathbf{k}) = \frac{\hbar^2 \mathbf{k}^2}{2m} + \alpha_R \boldsymbol{\sigma} \cdot (\hat{\mathbf{z}} \times \mathbf{k}) + J \boldsymbol{\sigma} \cdot \hat{\mathbf{m}}, \quad (2)$$

where $\mathbf{k} [= (k_x, k_y)]$ is a two-dimensional wave vector, m is the effective mass of the d_{xy} band, \hbar is the reduced Planck constant, α_R is the Rashba constant, and J is the exchange coupling between the conduction electron spin $\boldsymbol{\sigma}$ (Pauli matrices) and unit localized magnetic moment $\hat{\mathbf{m}}$. Here, we assume that the magnetization orientation ($\hat{\mathbf{m}}$) is aligned along the external magnetic field direction. In general, the Rashba constant depends on the magnetization orientation [40,41]. In our model calculation, we ignore this angular dependence of the Rashba constant and show that qualitative features of the angle-dependent MR observed in the experiment are described by a simple free-electron model without considering an additional angular dependence of the physical properties. The choice of this Hamiltonian demands an explanation. Since the STO is a cubic perovskite, three t_{2g} orbitals are degenerate at the Γ point in the bulk STO. For the STO-based interface, the d_{xy} band lies lower than the degenerate d_{yz} and d_{xz} bands due to the interface confinement [42–44]. The 2DEG geometry leads to a circular Fermi surface for the d_{xy} band and degenerate elliptical Fermi surfaces for the d_{yz} and d_{xz} bands. In our experiment, the sample is conducting at high gate voltages and becomes insulating as the gate voltage decreases. Moreover, the α -rotation results of R_{xx} [Fig. 4(a)] show a $\cos^2 \alpha$ -like dependence without the Lifshitz transition which was observed in Ref. [10]. These results indicate that the Fermi level lies in the lowest d_{xy} band and lowers as the gate voltage decreases. Therefore, in our theoretical analysis, we use a free-electron Hamiltonian with Rashba SOC and s - d exchange interaction by focusing on the single d_{xy} band. For simplicity, we treat the vector potential contribution to MR separately as the WL correction.

In our model calculation, we focus on the qualitative description of the longitudinal conductivity. In the high gate voltage regime where the WL correction is subdominant, the

qualitative feature of the longitudinal conductivity is captured by the Kubo formula with the relaxation time approximation, given as

$$\sigma_{xx}^0 = 2e^2\tau \sum_{n=\pm} \int \frac{d^2k}{(2\pi)^2} (v_x^n)^2 \delta(E_n - E_F), \quad (3)$$

$$E_{\pm} = \frac{\hbar^2 \mathbf{k}^2}{2m} \pm |\alpha_R(\mathbf{k} \times \hat{\mathbf{z}}) + J\hat{\mathbf{m}}|, \quad (4)$$

$$v_x^{\pm} = \frac{\hbar k_x}{m} \pm \frac{\alpha_R}{\hbar} \left(\frac{Jm_y + \alpha_R k_x}{|\alpha_R(\mathbf{k} \times \hat{\mathbf{z}}) + J\hat{\mathbf{m}}|} \right), \quad (5)$$

where $+/-$ represents the spin up/down band, e is the electron charge, τ is the relaxation time, and E_F is the Fermi energy. In the experimental result depicted in Fig. 4(a), there are sign changes between low gate and high gate voltage regimes. In order to describe the electron transport in the low conductance (i.e., low gate) regime, we adopt the theoretical results of Refs. [45,46], which compute the WL correction as

$$\Delta\sigma_{xx}(B) = -\frac{e^2}{4\pi^2\hbar} \left\{ \frac{1}{a_0} + \frac{2a_0 + 1 + H_{SO}/B}{a_1[a_0 + H_{SO}/B] - 2H_{SO}/B} + 2 \ln \frac{H_{tr}}{B} + \psi \left(\frac{1}{2} + \frac{H_{\phi}}{B} \right) + 3C \right. \\ \left. - \sum_{n=1}^{\infty} \left[\frac{3}{n} - \frac{3a_n^2 + 2a_n H_{SO}/B - 1 - 2(2n+1)H_{SO}/B}{[a_n + H_{SO}/B]a_{n-1}a_{n+1} - 2(H_{SO}/B)[(2n+1)a_n - 1]} \right] \right\}, \quad (6)$$

where $a_n = n + \frac{1}{2} + \frac{H_{\phi}}{B} + \frac{H_{SO}}{B}$, $\psi(1+z) = -C + \sum_{n=1}^{\infty} \frac{z}{n(n+z)}$, and C is the Euler constant. We note that Eq. (6) is a quantum correction to the conductivity due to a perpendicular magnetic field [47]. We use the following parameters for the model calculations: $m = 0.7m_e$ (m_e is the free-electron mass) [16], $J = 2.5$ meV, $\tau = 0.33$ ps, $H_{\phi} = 3.0$ T, and $H_{SO} = 1.2$ T. For the gate voltage effect on the Rashba coefficient, we assume $\alpha_R = (60 + \lambda E_F)$ meV Å, where $\lambda = 4/\text{meV}$.

In Fig. 4(b), we show the theoretical results for the angular dependence of the resistivity as functions of the rotating angles in α , β , and γ . The change in the gate voltage is considered as the change in the Fermi energy. The Fermi-energy shift for the γ rotation is taken into account in order to reflect the resistance hysteresis from the voltage cycle [38]. In all three rotations, the theoretical calculations of MR qualitatively match well with the experimental results [Fig. 4(a)]. Even though our model Hamiltonian and calculation scheme is simplified, this good agreement shows that the simple Hamiltonian [Eq. (2)] considering the coexistence of Rashba SOC and exchange coupling describes the experimental result reasonably well and the interfacial Rashba SOC is key to the interfacial Rashba MR whose angular dependence is similar to that of the spin Hall MR.

In order to get insight into the similarity between the interfacial Rashba MR and the spin Hall MR, we focus on the second term in Eq. (5), which is an additional velocity originating from the Rashba interaction. This additional velocity includes the magnetization orientation that gives the angular dependence of the longitudinal conductivity. In other words, a strong m_y dependence of ΔR_{xx} is natural because of the symmetry of Rashba SOC. For instance, one obtains $v_x^{\pm} \approx \hbar k_x/m \pm Jm_y\alpha_R/\hbar$ for $J \gg \alpha k_F$. There is also an additional source of the angular dependence of the interfacial Rashba MR. When Rashba SOC and exchange coupling coexist, the Fermi surface distorts, depending on the magnetization direction [50]. The Fermi-surface distortion is maximized for an in-plane magnetization whereas it is absent for an out-of-plane magnetization. Because of these two

contributions, additional velocity and Fermi-surface distortion, the interface subject to both Rashba and exchange interactions exhibits an interfacial Rashba MR similar to the spin Hall MR.

IV. SUMMARY AND DISCUSSION

In summary, we report interfacial Rashba MR for LAO/STO 2DEG and find that its angular dependence is similar to that of the spin Hall MR observed in FM/HM structures. Our model calculations describe the experimental results reasonably well and show that the spin-Hall-MR-like behavior originates from the combined effects of interfacial Rashba SOC and exchange coupling. As the bulk spin Hall effect is absent in the LAO/STO 2DEG system, our finding evidences that interfacial Rashba SOC gives rise to spin-Hall-like MR. Therefore, our result suggests that the interfacial SOC effect is key to understanding various SOC-related phenomena in magnetic/nonmagnetic bilayers.

In the theory part of this work, we note that there are potential complexities left aside. In this paper, we use the free-electron model of a single orbital (d_{xy}) with the linear Rashba interaction. However, the LAO/STO 2DEG is composed of t_{2g} orbitals with nonquadratic energy dispersion and there is a report suggesting the existence of a cubic Rashba interaction [51]. Moreover, in our model, many parameters are difficult to be determined in experiments and a quantitative description including a quantum correction is not reliable. Thus, we restrict the purpose of the model calculation to a qualitative description of experimental observations.

We end this paper by noting that a recent experiment for a Bi/Ag/CoFeB metallic trilayer found the Rashba-Edelstein MR [52], whose angular dependence is similar to that of the spin Hall MR. This Rashba-Edelstein MR originates from the combined action of two separate interfaces, a Bi/Ag interface with Rashba SOC [53] and a Ag/CoFeB interface with exchange splitting, through a spin diffusion process. In this trilayer including a conducting bulk, therefore, the

charge-to-spin and spin-to-charge conversions at the Bi/Ag interface and the spin-dependent reflection at the Ag/CoFeB interface are separated. Because of this separation in the trilayer, it is not straightforward to get insight into the bulk versus interface contributions to the spin Hall MR of the FM/HM bilayers. In contrast, our result gives a clear indication about the pure interface contribution to the spin Hall MR as the LAO/STO 2DEG, just as the FM/HM interface, is a single interface subject to both Rashba SOC and exchange coupling with no conducting bulk.

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K.N. and G.G. contributed equally to this work.

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