Magnetization switching driven by current-induced torque from weakly spin-orbit coupled Zr

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Current-induced magnetization switching (CIMS) via the spin-orbit torque is technologically important for applications of spintronic devices. Existing experimental studies are focused on 5d heavy metals with large spin-orbit coupling but theoretical studies indicate that 3d or 4d light metals with weak spin-orbit coupling may also generate sizable torque through the strong orbital Hall effect. However, CIMS has seldom been experimentally demonstrated using 3d or 4d light metals. Here, we report sizable current-induced torque and a robust CIMS in weakly spin-orbit coupled Zr based perpendicular magnetized multilayers. Current-induced effective torque field changes its sign as the Zr layer thickness varies, indicating the competition of multiple effects. Possible roles of the orbital Hall effect in the competition are discussed. Our findings widen the material scope of the CIMS and imply possible applications of the orbital Hall effect.

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I. INTRODUCTION

The manipulation of the magnetization in a magnetic multilayer can be achieved in an energy-efficient and fast way by current-induced magnetization switching (CIMS) [1–3] in a 5d heavy metal buffered CoFeB/MgO multilayer [4–10], due to the spin angular momentum transfer from the heavy metal to CoFeB by the large spin Hall effect (SHE) in these 5d heavy metals, such as Pt [11], Ta [3], and W [12]. As for 3d or 4d light metals with weak spin-orbit coupling (SOC), few CIMS experiments have been reported [13,14]. However, theories predict that the orbital Hall effect (OHE), a different version of the angular momentum generation effect, can be large in the transition metals despite the weak SOC [15–18]. The calculations show that their orbital Hall conductivities (OHCs) are about an order of magnitude larger than the spin Hall conductivities (SHCs) [15,16]. In particular, a recent calculation reports that intrinsic OHC can be gigantic, $\sigma_{\text{OH}} \sim 10^4 (\hbar/2e)(\Omega \text{cm})^{-1}$, in some materials with negligible SOC [18].

In the presence of SHE, a charge current converts into a spin current due to the spin-orbit interaction, which generates a spin-orbit torque (SOT) in the adjacent ferromagnet (FM) layer. In the presence of OHE, a charge current can be converted to a flow carrying orbital angular momentum, which is defined as orbital current. The orbital current may be either transformed to the spin current within a nonmagnetic metal (thus providing a mechanism to generate SHE of the nonmagnetic metal), or may be injected to an adjacent FM layer. According to a recent theoretical calculation [19], the injection of the orbital current generates an orbital torque (OT) in the FM layer. Since OT has similar properties to SOT, OT should also achieve CIMS in a perpendicularly magnetized multilayer. In this respect, a good test system to experimentally study OT and OHE is a multilayer consisting of a light metal (LM) and a FM with the inversion symmetry broken along the film growth direction. Thus, the search for a LM/FM/MgO multilayer with clear CIMS behavior is valuable not only for widening the material scope of CIMS but also for studying OT and OHE.

Here we demonstrate the CIMS and the measurement of torque in the perpendicularly magnetized Zr/CoFeB/MgO multilayers. Zr is a 4d light metal. Our theoretical calculation shows that the OHC of Zr is more than an order of magnitude larger than its SHC. Similar to the SOT, OT-induced effective fields in the Zr/CoFeB/MgO multilayer also have two components, $H_L$ and $H_T$, namely, longitudinal and transverse.
FIG. 1. Structure of Zr/CoFeB/MgO annealed at 300 °C. (a) High-resolution TEM image of Zr(8)/CoFeB(1.1)/MgO(3) sample annealed at 300 °C. The inset shows the fast Fourier transform pattern from the Zr layer, indicating hcp structure. (b) PNR curves of the Zr/CoFeB/MgO/Ta multilayer measured at the applied magnetic field \( H = 9000 \, \text{Oe} \) at room temperature. Here, the 3-nm-thickness Ta layer is a capping layer to protect the Zr/CoFeB/MgO film. The inset shows the related spin asymmetry (SA) curve. Symbols correspond to the experimental data points, while the solid lines show the fitted curves. (c) The fitted nuclear and magnetic SLD (SLD \(_n\) and SLD \(_m\) ) profiles.

II. EXPERIMENTS

Multilayers films with a structure of Zr(10)/CoFeB(1.1)/MgO(3) (numbers in parentheses are thicknesses in nanometers) were deposited on thermally oxidized Si/SiO\(_2\) substrates at room temperature by a magnetron sputtering system. The thickness of the Zr layer was systematically varied from 2.2 nm to 16.2 nm. The base pressure of the sputtering system was less than 8 \times 10^{-6} \, \text{Pa}. The Zr and Co\(_{40}\)Fe\(_{40}\)B\(_{20}\) (CoFeB) layers were deposited by using a direct current (DC) power source and the MgO layer was deposited by using a radio-frequency power source. The deposition rates for Zr, CoFeB, and MgO layers were 0.54 Å/s, 0.68 Å/s, and 0.1 Å/s, respectively. The films were annealed in a vacuum, which was less than 1 \times 10^{-3} \, \text{Pa}, at 300 °C for 1 h to obtain perpendicular magnetic anisotropy. The Zr/CoFeB/MgO films were patterned into Hall bars of width 10 \, \mu\text{m} by standard photolithography combined with an argon ion etching technique. The anomalous Hall effect (AHE) and CIMS were measured by using a room-temperature multipurpose transport measurement system with a 1 T electromagnet, a Keithley 2182A, and a Keithley 6221, which was employed as the source for the DC and pulse measurements. The transmission electron microscopy (TEM) analysis was performed in a FEI Tecnai G2 F20 microscope, operated at 200 kV. The samples for TEM analysis were prepared by the focus ion beam with Helios Nanolab 600. Figure 1(a) presents a high-resolution TEM image of the cross-sectional Zr(8.1)/CoFeB(1.1)/MgO(3) multilayer after annealing at 300 °C. The CoFeB layer is nearly amorphous, while the MgO layer exhibits a polycrystalline structure. The Zr underlayer is also polycrystalline. The inset is the fast Fourier transform pattern, indicating a clear hexagonal-close-packed (hcp) structure of the Zr layer.

To further understand the layer structure of the annealed Zr/CoFeB/MgO multilayer, the room-temperature polarized neutron reflectometry (PNR) measurement was carried out on the Multipurpose Reflectometer at the China Spallation Neutron Source, which is a unique depth-resolved technique to understand the magnetic properties in a magnetic layer or multilayer due to the high penetration depth of neutrons [20]. The neutron reflectivity curves are recorded at room temperature as the function of momentum transfer \( Q = 4\pi \sin \theta / \lambda \), where \( \lambda \) is the neutron wavelength and \( \theta \) is the incident angle between the neutron beam and the film plane. To access a broad momentum transfer range, reflected neutrons were collected at different incident angles. Figure 1(b) shows the non-spin-flip reflectivity \( R^{++} \) and \( R^{--} \) curves measured under the applied field of 9000 Oe. Non-spin-flip reflectivities indicate that the spin polarizations are the same for the incoming and reflected neutrons. The spin asymmetry (SA) is defined as \( \text{SA} = (R^{++} - R^{--})/(R^{++} + R^{--}) \), which is very sensitive to the magnetization of the film. The obtained \( R^{++} \) and \( R^{--} \) curves were simulated using the SimuReflec program [21]. From the simulation, the depth dependences of nuclear and magnetic scattering length densities (SLD \(_n\) and SLD \(_m\) ) were derived as a function of film thickness as shown in Fig. 1(c).

A reasonable fitting was obtained with a multilayer model for the annealed Zr/CoFeB/MgO sample, containing the interface between the Zr and CoFeB layers and fixing the nuclear scattering length density of the Zr and MgO layers with their bulk values. Due to the noticeable neutron absorption of B nuclei, the imaginary part of SLD \(_n\) is also considered during configurations.
the fitting. As shown in our fitted model, a high region of
the imaginary part of SLDₙ (ImSLDₙ) can be seen in the
interface between Zr and CoFeB, indicating the diffusion of
B into the Zr layer. A similar feature can be seen in Ta/CoFeB
PMA samples by using PNR measurements [22]. The mag-
netic scattering length density is directly proportional to the
saturation magnetization of the sample because CoFeB should
be already saturated by the in-plane magnetic field of 9000
Oe. From the fitting, one obtains SLDₙ = 4.06 × 10⁻⁴ nm⁻².
This gives Mₛ = 1400 emu/cm³, which may be due to the
CoFe formation after some of the B are absorbed by Zr.

III. RESULTS AND DISCUSSION

Figures 2(a) and 2(b) show the setup of the harmonic
Hall voltage measurement, where a low-frequency sinusoidal
current (133.33 Hz) was passed into the patterned Hall bars
and the in-phase first-harmonic and out-of-phase second-
harmonic Hall voltages were measured by two Stanford lock-
in amplifiers. From the harmonic Hall voltage measurements,
the current-induced effective torque fields in two config-
urations, H_L and H_T, namely, longitudinal and transverse
configurations, respectively, can be obtained [23,24]. For H_L
measurement, the magnetic field is swept in the direction
of alternating current, while for H_T measurement, the mag-
netic field is swept perpendicular to the current direction.
In both measurement configurations, the magnetic field is
applied with a small tilting angle (about 2°) to the film plane.

Figures 2(c)–2(f) show the first-harmonic Hall voltages (V₀)
as a function of H_L and H_T for two samples with Zr thick-
nesses of 2.2 nm and 4.9 nm, respectively. It is clear that the V₀
can be fitted using a parabola dependence. The inset of each
figure represents the second-harmonic Hall voltages (V₂₀)
as a function of H_L and H_T, which shows linear dependence. For
the longitudinal field, the slopes of the V₂₀ curves are the same
for magnetizations pointing along +z and −z, whereas their
sign reverses for the transverse field. However, the sign of
the slope changes for both longitudinal and transverse fields
when the Zr thickness is increased from 2.2 nm to 4.9 nm.

The longitudinal effective field (ΔH_L) and the trans-
verse effective field (ΔH_T) can be calculated from the H_L and H_T
field dependencies of harmonic voltages, using the following
equation when the planar Hall effect is negligible compared
to the anomalous Hall effect [25],

\[ \Delta H_{L,T} = -2 \frac{\partial V_{20}/\partial H_{L,T}}{\partial^2 V_{0}/\partial H^2_{L,T}}. \]  

(1)

The longitudinal (transverse) torque efficiencies (βₘ(T)) are
defined as ΔHₘ(T)/Jₑ, to characterize the effective torque
fields. The Zr thickness dependence of the longitudinal effec-
tive field per current density (βₘ = ΔHₘ/Jₑ) and the trans-
verse effective field per current density (β_T = ΔH_T/Jₑ) are
plotted in Figs. 3(a) and 3(b), respectively. Both βₘ and β_T
change their signs when the Zr thickness is above 3 nm.
In principle, current-induced torque can generate the field-like and damping-like terms, when a spin (or orbital) current polarized along the \( \hat{y} \) direction (\( \mathbf{\hat{\sigma}} \parallel \mathbf{\hat{y}} \)) is injected into a FM magnetized along the \( \hat{z} \) direction. The damping-like effective torque field can arise through the SHE (or OHE) and the Rashba effect at the FM/LM interface, which is described as \( \xi_{DL} \propto \mathbf{\hat{m}} \times (\mathbf{\hat{y}} \times \mathbf{\hat{m}}) \). Here \( \mathbf{\hat{m}} \) is the unit vector along the direction of magnetization. Damping-like torque efficiency can be extracted using the following equation:

\[
\xi_{DL} = 2|e|M_{CFCFeB}\Delta H_{Zr}/hJ_s = 2|e|M_{CFCFeB}\beta_s/h,
\]

where \( e \) is the charge of an electron, \( M_r \) is the saturation magnetization of CoFeB, \( t_{CFCFeB} \) is the thickness of CoFeB, and \( h \) is the reduced Planck constant. Figure 3(c) shows the \( t_{Zr} \) dependence of \( \xi_{DL} \) as determined by using Eq. (2). \( \xi_{DL} \) is positive for the thin \( Zr \) layer and negative for the thick \( Zr \) layer.

Current-induced effective torque fields exhibit a significant dependence on the \( Zr \) layer thickness, indicating that there are competing effects, the interfacial Rashba effect and bulk SHE (and possibly OHE). To simplify the description of current-induced torque in a LM with weak SOC, we introduce the effective spin Hall angle \( \theta_{SH} \). If SHE-induced SOT is the dominant source of \( \theta_{SH} \), \( \theta_{SH} \) represents the ratio of the spin current density \( J_s \) absorbed by CoFeB to the charge current density in the \( Zr \) layer, \( \theta_{SH} = J_s/J_c \). If OHE-induced OT is the dominant source of \( \theta_{SH} \), \( \theta_{SH} \) is given instead by \( J_s/J_c \), where \( J_o \) is the effective spin current density in the \( Zr \) layer generated by the orbital current injection into the \( Zr \) layer [19]. When SOT and OT are comparable, \( \theta_{SH} \) should contain both contributions. For simplicity, we assume for now that \( \theta_{SH} \) is dominated by the SHE-induced SOT and consider its competition with the Rashba-based mechanism of the damping-like torque [26–29]. If we follow the diffusion theory [24,30] and assume that the \( Zr/CoFeB \) interface has perfect transparency, the damping-like torque efficiency can be expressed as

\[
\xi_{DL} = \theta_{SH}[1 - \text{sech}(t_{Zr}/\lambda_{Zr})] + C,
\]

where \( \lambda_{Zr} \) represents the characteristic spin diffusion length of \( Zr \) and \( C \) is a constant arising from the interfacial Rashba mechanism. A fit of Eq. (3) to the data [Fig. 3(c)] yields \( \theta_{SH} = -0.03 \pm 0.006, \lambda_{Zr} = 3.1 \pm 1.3 \) nm, \( C = 0.012 \pm 0.01 \). According to the Rashba Hamiltonian, the direction of the Rashba-effect-induced spin accumulation is along \( \mathbf{E} \times \mathbf{p} \), where \( \mathbf{p} \) is the electron momentum. The direction of \( \mathbf{E} \) for a given interface is only determined by the difference of work function (\( \Delta \Phi \)) between two adjacent layers. As for the \( Zr/CoFeB/MgO \) multilayer, the work function difference is expected to be \(-0.75 \) eV between the \( Zr \) (\( \Phi_{Zr} = 4.05 \) eV [31]) and CoFeB (\( \Phi_{CoFeB} = 4.8 \) eV [32]) layers. The spin accumulation generated by the Rashba effect at the \( Zr/CoFeB \) interface is along the \( -z \) direction, which is as same as the directions of \( \mathbf{E} \) at the \( Hf/CoFeB \) interface [24]. Therefore, the sign of the Rashba effective field is opposite to the bulk effective field from SHE.

To further confirm the \( Zr \)-thickness-dependent sign change of the current-induced torques in the \( Zr/CoFeB/MgO \) multilayers, we performed CIMS measurements for different \( Zr \) thicknesses as shown in Fig. 4. We measure the Hall resistance while sweeping the in-plane pulse current. In addition to the current, a small constant in-plane magnetic field is applied along the current direction to break the symmetry, as shown in Fig. 4(a). For the sample with 2.2-nm-thick \( Zr \), by applying an in-plane magnetic field \( H_x = +50 \) Oe, the current switches the magnetization of the CoFeB layer between the up and down directions, and the switching sequence is anticlockwise.
resistance is measured after a 150 μs delay. The switching sequence is also reversed [Fig. 4(b)]. As expected, for reversing the direction of the magnetic field, the switching signs are shown as a function of the Fermi energy $E_F$. The OHC and SHC of Zr were calculated by employing the Kubo formula within the linear response theory. The OHC and the SHC in Zr can be calculated to be $|\sigma_{\text{SH}}| = |\xi_{\text{DL}}/\rho_0| \approx 110 (\hbar/e)/\Omega \text{cm}^{-1}$, where we assume that the observed SOT originates from the SHE of Zr and perfect spin transparency at the Zr/Co-Fe-B interface. Here, $\rho_0 = 135 \mu\Omega \text{cm}$. The estimated lower bound of SHC in Zr is quite close to our calculation. On the other hand, the orbital current may be injected into the adjacent CoFeB layer before OHE completely converts to SHE if the Zr is thinner than its characteristic length.

Finally, we consider the OHE. The electronic band calculation of hcp Zr was performed using a tight-binding model based on the Slater-Koster method [33]. Detailed information on the theoretical calculation is presented in Appendix C. The lattice constants $a = 0.3232 \text{ nm}$ and $c = 0.5147 \text{ nm}$ and the Slater-Koster parameters were taken from Ref. [34]. The spin-orbit coupling was included in the Hamiltonian in the form of $(2\hbar \lambda/h^2) L \cdot S$, where $\lambda$ was set to 0.05 eV [35]. The OHC and SHC of Zr were calculated by employing the Kubo formula within the linear response theory. The integration over the Brillouin zone was performed using a uniform $300 \times 300 \times 300$ k-point mesh. In Fig. 5(a), the theoretically calculated OHC $\sigma_{\text{OH}}$ and the SHC $\sigma_{\text{SH}}$ of hcp Zr are shown as a function of the Fermi energy $E_F$, where $E_F = 0$ corresponds to the actual Fermi energy. The directions of the electric field, orbital (spin) polarization, and orbital (spin) current are set to be along the $x$, $y$, and $z$ axes, respectively [Fig. 5(b)]. We find that $\sigma_{\text{OH}}$ dominates over $\sigma_{\text{SH}}$ by an order of magnitude [5300 $(h/e)/\Omega \text{cm}^{-1}$ and $\sigma_{\text{SH}} \approx -170 (h/e)/\Omega \text{cm}^{-1}$ at $E_F = 0$ eV], and is about 2–3 times larger than the SHC of Pt [16]. Therefore, our experimental observation of the large current-induced torque and magnetization switching from weakly spin-orbit coupled Zr is likely attributed to the OHE and the orbital-to-spin conversion. If the Zr layer is sufficient thick, the conversion may mostly occur in Zr through the SOC of Zr and the resulting spin Hall current will be injected to the adjacent CoFeB layer to generate a SOT. If the Zr layer is not sufficiently thick, the orbital Hall current may not be converted to the spin Hall current within the Zr layer. Then the orbital Hall current itself may be injected into the CoFeB and the conversion may mostly occur in the CoFeB layer instead through the SOC of CoFeB. It was recently proposed that the latter process can generate the OT [19].

Since the spin-orbit correlation is negative in Zr (as evidenced by the opposite signs of $\sigma_{\text{OH}}$ and $\sigma_{\text{SH}}$ of Zr) and positive in CoFeB, the SOT and the OT will have opposite signs and compete. The negative SOT will dominate if the Zr is thicker than its characteristic length (3.2 nm from our experiments), and $\xi_{\text{DL}}$ will be negative as shown in Fig. 3(c) in the thick Zr regime. Furthermore, the lower bound of SHC in Zr can be calculated to be $|\sigma_{\text{SH}}| \approx |\xi_{\text{DL}}|/\rho_0 Zr \approx 110 (h/e)/\Omega \text{cm}^{-1}$, if we assume that the observed SOT originates from the SHE of Zr and perfect spin transparency at the Zr/Co-Fe-B interface. Here, $\rho_0 = 135 \mu\Omega \text{cm}$. The estimated lower bound of SHC in Zr is quite close to our calculation. On the other hand, the orbital current may be injected into the adjacent CoFeB layer before OHE completely converts to SHE if the Zr is thinner than its characteristic length.

In order to assess whether the competition between SOT and OT or the competition between SHE and the Rashba effect is more important for the sign change of torques, we have measured the interfacial Dzyaloshinskii-Moriya interaction (DMI) in our samples using current-induced AHE hysteresis loop shift measurement [13]. However, the DMI-induced effective shifts of AHE loops were within the error bars of the measurement setup, suggesting that the interfacial DMI is small. Considering that both the DMI and the Rashba-induced torque are of interfacial origin, the small DMI implies that the Rashba-induced torque is likely to be small.

The nonmagnetic (NM) thickness dependent sign change of the torque has been reported for NM/CoFeB/MgO for three choices of NM = Zr (this work at ~3 nm), Ta [4] (at ~0.5 nm), and Hf [24] (~2 nm). For NM = Ta and Hf, the sign change has been attributed to the competition between...
SHE and Rashba effect, and it was suggested that the sign-changing thickness increases as the shunting effect becomes severe [24]. In this respect, Zr is abnormal since the sign change occurs at ~3 nm (larger than the sign-changing thickness in Ta and Hf) despite the weaker shunting effect (the resistivity of Zr ~ 135 μΩ cm (see Appendix B) is smaller than those of Ta (~190 μΩ cm) [4] and Hf (~380 μΩ cm) [24]). This raises the possibility that the Rashba effect may not be important for the sign change in Zr. However further studies are necessary to clarify the main origin of the sign change unambiguously.

IV. CONCLUSIONS

To conclude, current-induced torque effective fields exhibit a significant dependence on the Zr layer thickness, indicating that there are several competing effects, the Rashba effect, SHE, and OHE. Our experimental data suggest that the Zr thickness of the sign change in torque effective fields may be influenced by the intrinsic OHE and SHE in Zr. The ξDL may be further enhanced by engineering the interface between the FM and the LM and injecting more orbital current.

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APPENDIX A: CURRENT-INDUCED EFFECTIVE FIELD

Low-frequency harmonic Hall voltage measurement was proposed by Pi et al. [36] and then was developed by [25]. In Ref. [25], the validity of harmonic Hall voltage measurement was tested by numerical calculations based on a macrospin model. In brief, when a current, I, is passed to the Hall device, the related Hall voltage, Vxy = RxyI, typically contains contributions from the anomalous Hall effect (AHE) and the planar Hall effect (PHE). If we define ΔR_{AHE} and ΔR_{PHE} as the change in the Hall resistance due to the AHE and PHE, respectively, the Hall resistance R_{xy} is expressed as [25]

\[
R_{xy} = \frac{1}{2} \Delta R_{AHE} \cos \theta + \frac{1}{2} \Delta R_{PHE} \sin^2 \theta \sin 2\varphi, \quad (A1)
\]

where \( \theta \) and \( \varphi \) are the polar and azimuthal angles, respectively, of the magnetization \( M_z \). When a sinusoidal current is applied, the current-induced effective field oscillates in sync with the sinusoidal current. The second-harmonic Hall voltage \( V_{2\omega} \) contains information on \( \Delta H_{x,y,z} \) through \( \Delta \theta \) and \( \Delta \varphi \). Meanwhile, the current-induced effective field \( \Delta H_{x,y,z} \) can modify the magnetization angle, \( \Delta \theta \) and \( \Delta \varphi \), of the Hall device from its equilibrium value. Thus, we can obtain

\[
B = \frac{1}{2} \left[ (\Delta H_x \mp 2\xi \cos 2\varphi \Delta H_y) \cos \varphi \right] + (\Delta H_x \mp 2\xi \cos 2\varphi \Delta H_y) \sin \varphi H. \quad (A2)
\]

Here, \( \xi \equiv \Delta R_{PHE}/\Delta R_{AHE} \), which is the ratio of the PHE and AHE resistances [25]. Finally, we obtain

\[
\Delta H_x = -2 \frac{(B_x \pm 2\xi B_y)}{1 - 4\xi^2}. \quad (A3)
\]

\[
\Delta H_y = -2 \frac{(B_x \pm 2\xi B_y)}{1 - 4\xi^2}. \quad (A4)
\]

When \( \xi \) is very small—for instance, \( \xi \) is about 0.03 for our perpendicularly magnetized Zr/CoFeB/MgO samples—the above Eqs. (A3) and (A4) can be simplified as Eq. (1) that we used in the text.

As shown in Figs. 6(a)–6(d), we calculated the \( \Delta H_x \) and \( \Delta H_y \), which are plotted as a function of input bias current \( I_0 \) of the sinusoidal signal for the samples with Zr thicknesses of 2.2 nm and 4.9 nm, respectively. It is clear that the effective fields vary linearly with \( I_0 \), indicating that the effects of Joule heating are negligible in the measured \( I_0 \) range [23]. Similarly to the previous reports [4,24], only the sign of the trend for both effective fields is inverse for the samples with Zr thicknesses of 2.2 and 4.9 nm, which indicates the different dominating mechanisms in the current-induced torques.
We introduced 36 orthogonal atomic-orbital basis sets

resistance, binding method based on the Slater-Koster method [33].

sponding eigenstates of hcp Zr, we employed the tight-

we obtain $\rho_L = 119 \mu \Omega \text{cm}$ in the Zr/Ta bilayer, while $\rho_Z = 135 \mu \Omega \text{cm}$ in the Zr/CoFeB/MgO multilayer. The increase of Zr resistivity in the Zr/CoFeB/MgO multilayer may be due to the B diffusion in the Zr, which is consistent with the PNR results in Fig. 1. To further account for the current-shunting effect, the current density in Zr is calculated by $J_z = J_0 [t_{CoFeB}/(t_{CoFeB} + t_Z t_{CoFeB})]$, where $J_0 = I_0 / (W t_Z)$ is the nominal current density in the Zr layer. $t_{CoFeB}$ and $t_Z$ are the thickness of Zr and CoFeB, respectively, and $\rho_Z$ and $\rho_{CoFeB}$ are the resistivities of Zr and CoFeB, respectively. The resistivity of the individual layer was determined by channel resistance of the Hall bar versus thin film thickness measurements, which gives $\rho_Z = 76 \mu \Omega \text{cm}$ and $\rho_{CoFeB} = 135 \mu \Omega \text{cm}$. Both the decrease of resistivity CoFeB and the increase of its magnetization may also be due to the B diffusion.

**APPENDIX C: THEORETICAL CALCULATIONS OF ORBITAL AND SPIN HALL CONDUCTANCE**

In order to obtain the energy eigenvalues and corresponding eigenstates of hcp Zr, we employed the tight-binding method based on the Slater-Koster method [33]. We introduced 36 orthogonal atomic-orbital basis sets $\{|\phi_{n\sigma R}\rangle\}$ for each Bravais lattice vector $R$ with orbital $n = s$, $p_x$, $p_y$, $p_z$, $d_{x^2−y^2}$, $d_{xy}$, $d_{yz}$, $d_{zx}$ for two atomic sites in a unit cell, and two spins $\sigma = \uparrow, \downarrow$. Then the $36 \times 36$ Hamiltonian $H(R)$ matrix for each $R$ is written in terms of the Slater-Koster parameters taken from Ref. [34]. The spin-orbit coupling for the $l = s$, $p$, $d$ orbital was included in the form of $(2\lambda_l/h^2) L^{(l)} \cdot S$, where $\lambda_l$ is the spin-orbit coupling constant, and $S$ and $L^{(l)}$ are the spin and orbital angular momentum operators for the $l$ orbital, respectively. Here $L^{(l)}$ is defined with respect to the center of each atom. We set $\lambda_l$ to be a spin-orbit coupling constant for the outermost electron 0.05 eV [35], which is assumed to be independent of $l$. The $k$-space representation of the Hamiltonian $H(k)$ is obtained from a basis set of Bloch-like states $|\psi_{n\sigma k}\rangle = (1/\sqrt{N}) \sum_R e^{ik \cdot R} |\phi_{n\sigma R}\rangle$. Solving an eigenvalue equation for $H(k)$ gives the electronic band structure of hcp Zr as shown in Fig. 8.

The calculations of OHC and SHC were carried out using the Kubo formula,

$$\sigma_{OH(SH)} = −e^2 \sum_{n,m} \int \frac{d^3k}{(2\pi)^3} \left( f_{nk} − f_{mk} \right) \times \text{Im} \left[ \frac{\langle u_{nk} | j^X_X (k) | u_{mk} \rangle \langle u_{mk} | v_z (k) | u_{nk} \rangle}{(E_{nk} − E_{mk} + i\eta)^2} \right],$$

(C1)

where $e > 0$ is the elementary charge, $\hbar$ is the reduced Planck constant, $f_{nk}$ is the Fermi-Dirac distribution function for room temperature, $|u_{nk}\rangle$ is a periodic part of the Bloch state with energy eigenvalue $E_{nk}$, $v_z (k) = \partial_k H(k)/\hbar$ is the $z$ component of the velocity operator, and $\eta$ was set to 25 meV. Here $j^X_X (k)$ is the $z$ component of the orbital (spin) current operator carrying $y$-directional orbital (spin) angular momentum,

$$j^X_X (k) = \frac{1}{2} [X_X v_z (k) + v_z (k) X_X],$$

(C2)

with $X_X = L_y (S_y)$. The $k$ integration over the Brillouin zone in Eq. (C1) was performed on a uniformly distributed $300 \times 300 \times 300$ $k$-point mesh.

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