

Direct and Inverse Spin Splitting Effects in Altermagnetic RuO₂

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Recently, the altermagnetic materials with spin splitting effect (SSE), have drawn significant attention due to their potential to the flexible control of the spin polarization by the Néel vector. Here, the direct and inverse altermagnetic SSE (ASSE) in the (101)-oriented RuO₂ film with the tilted Néel vector are reported. First, the spin torque along the *x*-, *y*-, and *z*-axis is detected from the spin torque-induced ferromagnetic resonance (ST-FMR), and the *z*-spin torque emerges when the electric current is along the [010] direction, showing the anisotropic spin splitting of RuO₂. Further, the current-induced modulation of damping is used to quantify the damping-like torque efficiency (ξ_{DL}) in RuO₂/Py, and an anisotropic ξ_{DL} is obtained and maximized for the current along the [010] direction, which increases with the reduction of the temperature, indicating the present of ASSE. Next, by way of spin pumping measurement, the inverse altermagnetic spin splitting effect (IASSE) is studied, which also shows a crystal direction-dependent anisotropic behavior and temperature-dependent behavior. This work gives a comprehensive study of the direct and inverse ASSE effects in the altermagnetic RuO₂, inspiring future altermagnetic materials and devices with flexible control of spin polarization.

transfer torque (STT), i.e., the electrical current can be polarized by the local magnetization in the magnetic layer ($\sigma // m$) and thus the spin-polarized current can be used to exert a torque for another magnetic layer for magnetization switching or oscillation, which has been adopted in current STT-MRAM technology.^[3,4] Spin-orbit torque (SOT) is another phenomenon that typically generates the transversal spin current (J_s) by spin-orbit coupling with the spin-polarized direction $\sigma // J_e \times J_s$, which has been demonstrated in abundant material systems such as heavy metals,^[5–7] topological insulators,^[8–13] oxide materials,^[14] and so on.^[15–19] SOT-MRAM can separate the writing and reading paths and thus has an improved device endurance.^[20,21]

Generally, antiferromagnets are considered to possess negligible spin polarization due to the fully-cancelled spin sub-bands. Recently, the theoretical work predicts a new class of altermagnets by decoupling the spin and crystal spaces, with the zero net magnetization and an alternating

spin-momentum locking (spin splitting effect, SSE) in energy bands originating from the inequivalent crystal environments for two spin sublattices, which can provide a spin current with the polarization aligned along the Néel vector N and an induced spin splitting torque (SST) to the adjacent magnetization.^[22–28] Recently, researchers experimentally observed the spin splitting band structures of altermagnets, MnTe₂^[29] and MnTe,^[30]

1. Introduction

Spintronic materials and devices have potential applications in the next-generation low-power and high-speed memory and logic technology, such as magnetic random-access memory (MRAM).^[1,2] The crucial question of spintronics is how to efficiently generate the spin current by electrical methods. Spin

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by using the spin-resolved and angle-resolved photoemission spectroscopy (SARPES) and angle-resolved photoemission spectroscopy (ARPES) measurements, respectively. These observations are consistent with the band structures theoretically predicted. Altermagnetism candidates are diverse, including quasi-2D oxide insulator V_2Se_2O , 3D rutile fluoride or oxide insulators FeF_2 ,^[31] MnO_2 ,^[32] and metal RuO_2 .^[26] Additionally, materials like pnictide with metal–insulator transition $FeSb_2$ ^[33] and metal $CrSb$ ^[22] among others,^[26,34,35] are also noteworthy candidates for altermagnetism. SSE in the altermagnets possesses the controllable spin polarization direction (σ) and controllable spin current flow direction (J_s) simultaneously by tuning the Néel vector N and the electric current J_e directions, respectively, and therefore providing a more flexible way for the electrical control of magnetic order, such as an out-of-plane spin component for field-free switching of the perpendicular magnet.

Ruthenium dioxide (RuO_2) is a conductive rutile oxide with a collinear antiferromagnetic structure, exhibiting a high Néel temperature ($T_N > 300$ K).^[36] A recent study^[37] has introduced a dynamic approach to characterize the antiferromagnetic ordering in RuO_2 , unveiling a spatially periodic spin structure. Additionally, an anomalous Hall effect in RuO_2 was observed,^[38] with an anomalous Hall conductivity exceeding $1000 \Omega^{-1} \text{ cm}^{-1}$ in thin films along different crystal orientations. Further, the research^[39] delves into the crystal thermal transport phenomena in RuO_2 , which result from the breaking of crystal time-reversal symmetry. Based on the RuO_2/Py heterostructures, several experimental works have observed the altermagnetic spin splitting effect (ASSE) in the altermagnetic RuO_2 -based structures by the spin–torque ferromagnetic resonance (ST-FMR)^[40,41] and the SST-driven field-free magnetization switching measurements,^[42] where the Néel vector-dependent and crystal direction-dependent properties indicate the presence of SST. However, the inverse ASSE is still limited to the spin Seebeck measurement,^[43] where the thermal effect-free spin pumping measurement is still waiting to be discovered. Moreover, the comprehensive study of direct and inverse ASSE in the same material systems is important for the consistent understanding of the microscopic mechanism of ASSE.

In this work, we study the direct and inverse ASSE in (101)-oriented RuO_2 film with tilted Néel vector N . First, the spin torque along the x -, y -, and z -axis is obtained from the ST-FMR measurement, and z -axis spin torque is only obvious when the electric current is along the [010] direction. By employing the current-induced modulation of damping (MOD) method to evaluate ξ_{DL} in RuO_2/Py more directly, the anisotropic ASSE contribution is further confirmed which dominates at lower temperatures. Then, we perform the spin pumping measurement for the study of the inverse ASSE (IASSE) for the (101)- RuO_2 , where the crystal direction-dependent spin pumping signals (maximized along the [010] direction) and the enhancement of anisotropic spin pumping signals at lower temperature indicates the IASSE in RuO_2 . This work gives a comprehensive and consistent understanding of the spin splitting effect in altermagnetic RuO_2 .

2. Results and Discussion

RuO_2 is a conductive rutile oxide, with the Ru atoms at the center and corners of the unit cell having opposite spins, forming

a collinear antiferromagnetic ordering with the Néel vector N oriented along the [001] axis,^[28,44] as shown in **Figure 1A**. The center and corners of the Ru atoms experience different oxygen environments, which leads to the anisotropic spin sub-bands splitting.^[25,27] As a result of this spin splitting, it can generate a spin current J_s with the polarization σ along the N occurring under an electric field (E) (**Figure 1B**) via ASSE,^[40–42] even without the presence of spin–orbit interaction (SOI), and it is odd under time reversal ($\mathcal{T}$ -odd). Moreover, when introducing a pure spin current polarized parallel to N into RuO_2 , this spin current induces a non-equilibrium shift of the Fermi surface, leading to the emergence of a charge current J_c through its inverse effect called IASSE, as depicted in **Figure 1C**. This distinctive spin splitting mechanism, along with its inverse effect, contributes significantly to both the generation and detection of spin current.

To investigate the ASSE, we fabricate the epitaxially grown RuO_2 thin films (15 nm) on the $TiO_2(101)$ single crystalline substrates by magnetron sputtering. And then an in-plane magnetized $Ni_{81}Fe_{19}(Py)$ layer (8 nm) with the capping layers of $MgO(2 \text{ nm})/Ta(3 \text{ nm})$ are deposited, as shown in **Figure 1D**. High-resolution cross-sectional scanning transmission electron microscopy (HRTEM) is conducted in $TiO_2(101)/RuO_2(101)$, as shown in **Figure 1E**, showing the high-quality epitaxy growth of the $RuO_2(101)$ film. The high-resolution high-angle annular dark field (HADDF) image reveals highly ordered alternatively stacked Ru and O atoms. Next, the devices for ST-FMR and spin pumping measurements are fabricated by the standard photolithography combined with the ion etching method. First, we use the technique of ST-FMR^[45,46] to quantify the SST by ASSE at room temperature. The measurement setup is shown in **Figure 2A**, including a bias-tee, a microwave signal generator, and a lock-in amplifier. An RF current (I_{RF}) is applied through the sample, which excites the magnetization precession by the ASSE-induced spin torque. The magnetization resonance contributes to RF resistance due to the anisotropic magnetoresistance (AMR), and the mixing between the RF resistance and the RF current results in a DC voltage signal V_{mix} , i.e., the spin rectification effect. The geometry of the ST-FMR device with dashed blue rectangles is illustrated in **Figure 2B**, which has a dimension of $20 \mu\text{m}$ in width and $100 \mu\text{m}$ in length.

The detected ST-FMR signal V_{mix} as a function of the applied in-plane magnetic field H for the (101)-oriented $RuO_2(15)/Py(8)/MgO(2)/Ta(3)$ (thicknesses in nanometers) sample is obtained at $\theta_H = 40^\circ$ and $\varphi_C = 0^\circ$ (**Figure S4**, Supporting Information), with a microwave of 18 dBm and 10 GHz. Py film has an ultralow saturation field and isotropic in-plane magnetic anisotropy, and thus the magnetization is approximately aligned with the external magnetic field $\theta_M = \theta_H$. The V_{mix} can be fitted by combining the symmetric ($V_{S,ST}$) and antisymmetric ($V_{A,ST}$) Lorentzian peak shapes^[45–47]:

$$V_{mix} = V_{offset} + V_{S,ST} \frac{\Delta H^2}{(H - H_t)^2 + \Delta H^2} + V_{A,ST} \frac{\Delta H(H - H_t)}{(H - H_t)^2 + \Delta H^2} \quad (1)$$

where V_{offset} is the offset, $V_{S,ST}$ and $V_{A,ST}$ are the coefficients of symmetric and antisymmetric terms, respectively. H_t is the

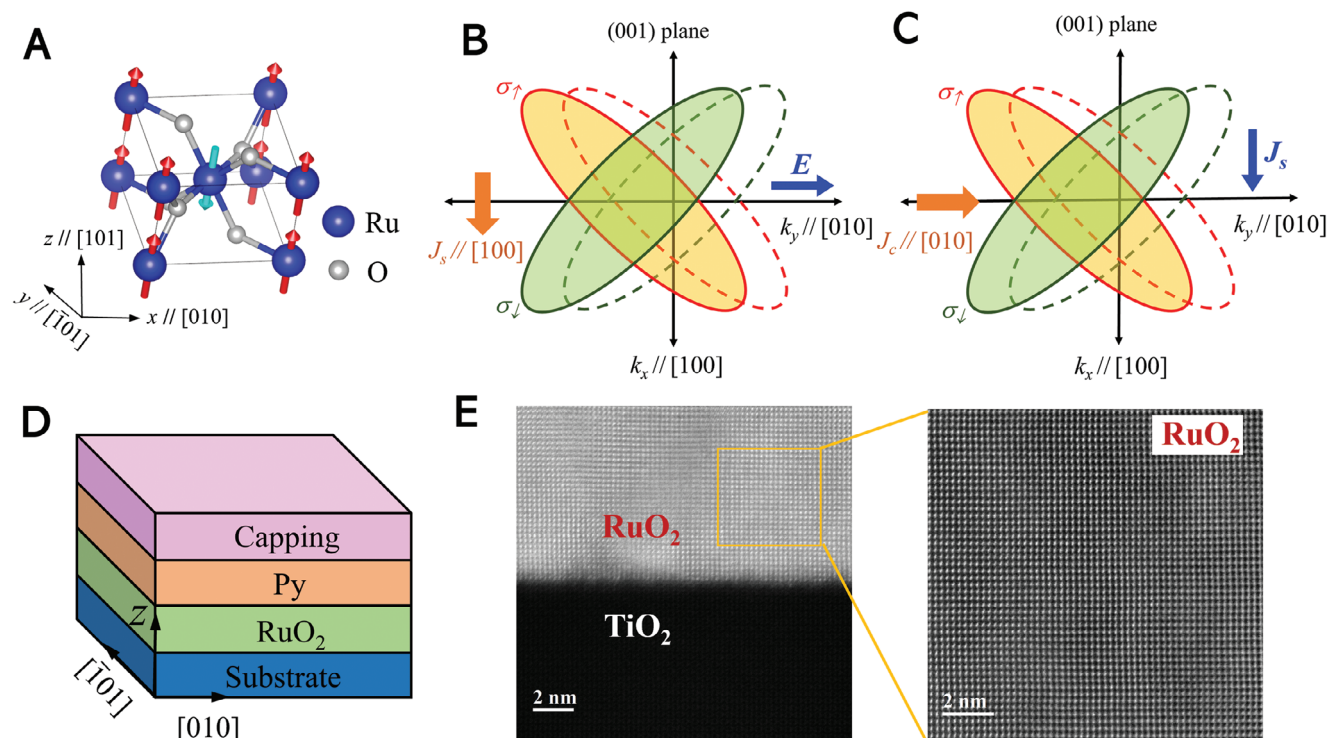


Figure 1. Direct and inverse spin splitting effect in RuO₂. A) The crystal structure of (101)-oriented RuO₂ with the Néel vector **N** (arrows) tilted from the out-of-plane direction. Blue and grey balls represent Ru and O atoms, respectively. B) Schematic diagrams of charge-to-spin and C) spin-to-charge conversion via ASSE and IASSE in RuO₂ from the view of momentum space. D) Schematic structure for the ASSE and IASSE in (101)-RuO₂/Py system. E) High-resolution HAADF image of the cross-section of TiO₂//RuO₂ (left) and RuO₂ (right) thin film.

resonant field and ΔH is the linewidth. The angle θ_H dependence of the V_{mix} as an external magnetic field is obtained by rotating the sample plane relative to the magnetic field direction. The amplitude of the damping-like (DL) and field-like (FL) torques of x -, y - and z -polarized spin current can be separated from the φ_H dependence from the fitting by Equations. (2) and (3):

$$V_S = S_y^{\text{DL}} \cos \theta_H \sin 2\theta_H + S_x^{\text{DL}} \sin \theta_H \sin 2\theta_H + S_z^{\text{FL}} \sin 2\theta_H \quad (2)$$

$$V_A = A_y^{\text{FL}} \cos \theta_H \sin 2\theta_H + A_x^{\text{FL}} \sin \theta_H \sin 2\theta_H + A_z^{\text{DL}} \sin 2\theta_H \quad (3)$$

where S_i^{DL} and A_i^{DL} (S_i^{FL} and A_i^{FL}) represent the amplitude of the DL (FL) torque generated by the x -, y - and z -directional spin polarization components, respectively, corresponding to the subscript i . Through the analysis by Equations. (2) and (3), the amplitudes of the DL and FL torques are effectively fitted and separated. Figure 2C,D depict the results of $V_{S,\text{ST}}$ and $V_{A,\text{ST}}$ measurements at $\varphi_C = 0^\circ$, respectively. It is evident that the observed data cannot be fitted by only considering the conventional spin Hall effect (SHE) ($S_y^{\text{DL}} \cos \varphi \sin 2\varphi$) term due to the strict orthogonal relationship among the applied charge current, generated spin current and the spin polarization. The fitting amplitude is derived from the contributions of τ_z^{FL} and τ_z^{DL} , indicating the emergence of z -spin polarizations. Furthermore, the corresponding ST efficiency, $\xi_y^{\text{ST}} = \frac{S_y^{\text{DL}}}{A_y^{\text{FL}}} \frac{e\mu_0 M_s t_{\text{Py}} t_{\text{RuO}_2}}{h} \sqrt{1 + \frac{M_{\text{eff}}}{H_0}}$ and $\xi_z^{\text{ST}} = \frac{A_z^{\text{DL}}}{A_y^{\text{FL}}} \frac{e\mu_0 M_s t_{\text{Py}} t_{\text{RuO}_2}}{h}$, can be estimated through the extracted amplitudes V_S and V_A .^[40] The first term A_y^{FL} in Equation (3) is predominant from the Oer-

sted field.^[45,48,49] Here, e , t_{Py} , t_{RuO_2} , $\hbar$, and H_0 are the elementary charge, thickness of Py, thickness of RuO₂, the reduced Planck constant, and Py resonance field at 10 GHz, respectively. $4\pi M_{\text{eff}}$ (9479 Oe) is the effective demagnetization field by fitting the resonance fields by Kittel equation, which is consistent with the saturation magnetization $4\pi M_s$ of 10053 Oe measured by the vibrating sample magnetometry (VSM), see the Supporting Information. The calculated results show that the z -spin torque efficiency for $\varphi_C = 0^\circ$ ($\xi_z^{\text{ST},[010]} = 0.0155$) is stronger than that of $\varphi_C = 90^\circ$ ($\xi_z^{\text{ST},[101]} = 0.0037$), indicating the crystal direction-dependent anisotropic ASSE contribution of RuO₂. The y -spin torque efficiency is 0.137 ($\xi_y^{\text{ST},[010]}$) for $\varphi_C = 0^\circ$, and the one for $\varphi_C = 90^\circ$ is 0.0166 ($\xi_y^{\text{ST},[101]}$), as listed in Table 1. For the (101)-oriented RuO₂ film with tilted **N**, besides the y -polarization, the z -spin polarizations emerge due to the anisotropic spin band splitting of RuO₂, which agrees with previous experiments^[40–42] and theoretical predictions.^[22,23,26]

Through the ST-FMR measurements taken along two orthogonal directions, the obtained ST efficiencies encompass both DL and FL torques. Furthermore, a modulation of damping (MOD) method is performed to further characterize the DL torque efficiency ξ_{DL} ,^[45] where the ΔH of resonance can be tuned by the DC current-induced SST in the RuO₂ layer. By varying the DC, the modulation of ΔH can be expressed as^[45,49]:

$$\Delta H = \frac{2\pi f}{\gamma} \left[+ \frac{J_{\text{DCDL}} \sin \theta_H}{(H + 2\pi M_{\text{eff}}) \mu_0 M_0 t_{\text{FM}} 2e} \hbar \right] \quad (4)$$

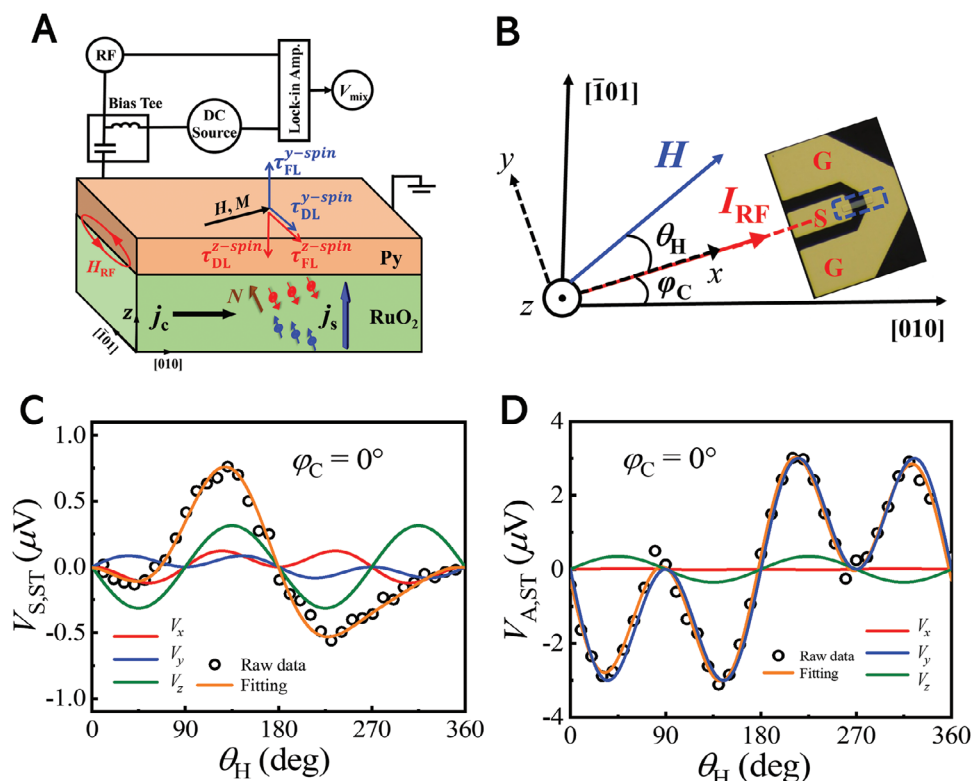


Figure 2. Spin-torque ferromagnetic resonance measurements. A) Schematic of ST-FMR measurement setup. The I_{RF} -induced charge-to-spin conversion in RuO_2 injects a spin current into the Py and excites the FMR. $\tau_{\text{DL}}^{\text{y-spin}}$ are in-plane (out-of-plane) for γ -spin with the blue arrows. $\tau_{\text{DL}}^{\text{z-spin}}$ are out-of-plane (in-plane) for z -spin with the red arrows. B) Schematic illustration of the ST-FMR measurement in $(101)\text{-RuO}_2/\text{Py}$. The heterostructures are patterned into long stripes within dashed blue rectangles. θ_{H} is the angle orientation of the magnetic field H relative to the direction of the applied microwave current I_{RF} in the sample, and φ_{C} is the angle between the applied current and the crystal direction $[010]$ for $\text{RuO}_2(101)$. C) $V_{\text{S,ST}}$ and D) $V_{\text{A,ST}}$ voltage amplitudes as a function of θ_{H} at $\varphi_{\text{C}} = 0^\circ$. Orange lines represent angle-dependent voltage signals contributed by the V_x (red), V_y (blue), and V_z (green), based on the analysis by using Equations (2) and (3).

As shown in **Figure 3A**, the approximately linear relation between ΔH and j_{c} is extracted. Here, the current is applied along the $\varphi_{\text{C}} = 0^\circ$ and the external magnetic field is oriented at the angle $\theta_{\text{H}} = 40^\circ$ with respect to the current direction. The applied frequency and power of microwave are 7 GHz and 18 dBm, respectively. According to Equation (4), we examine the current-induced change in the effective resonance linewidth $\Delta H/j_{\text{c}}$ (slope of the linear part fit in **Figure 3A**) as a function of in-plane magnetic field θ_{H} , which fits to $\sin\theta_{\text{H}}$. It can be clearly seen that the amplitude of $\Delta H/j_{\text{c}}$ at $\varphi_{\text{C}} = 0^\circ$ ($\xi_{\text{DL},[010]} = 0.2581$) is much stronger than that of $\varphi_{\text{C}} = 90^\circ$ ($\xi_{\text{DL},[101]} = 0.0155$) at room temperature. The MOD measurement has the advantage of eliminating the spin pumping contribution in ST-FMR and therefore gives a more

accurate value of ξ_{DL} . Furthermore, the extracted ξ_{DL} as a function of the temperature for $(101)\text{-RuO}_2(15)/\text{Py}(8)/\text{MgO}(2)/\text{Ta}(3)$ sample at $\varphi_{\text{C}} = 0^\circ$ and $\varphi_{\text{C}} = 90^\circ$ is shown in **Figure 3C**, from which we observe a strong enhancement of ξ_{DL} with decreasing temperature at $\varphi_{\text{C}} = 0^\circ$, indicating a much-dominated spin splitting contributions at lower temperatures. In contrast, conventional $\mathcal{T}$ -even mechanism resulting from intrinsic SHE generally exhibits negligible temperature dependence when $\varphi_{\text{C}} = 90^\circ$. Hereby, we have demonstrated the generation of the γ - and z -polarized spin current via ASSE in $\text{RuO}_2(101)$. Besides, the DL torque is a strong enhancement with decreasing temperature related to the crystal axes of RuO_2 , provides further evidence to the nontrivial charge-to-spin conversion due to the existence of the

Table 1. The ST efficiencies and pure spin pumping signal from the $[010]$ and $[101]$ orientations in $(101)\text{-RuO}_2(15)/\text{Py}(8)$ sample.

Structure	Method	$\varphi_{\text{C}} = 0^\circ$ [010] [μV]	$\varphi_{\text{C}} = 90^\circ$ [101] [μV]	Ratio
$\text{RuO}_2(15)/\text{Py}(8)$	ST-FMR	$\xi_{\text{y}}^{\text{ST},[010]}$	$\xi_{\text{y}}^{\text{ST},[101]}$	$\xi_{\text{y}}^{\text{ST},[010]}/\xi_{\text{y}}^{\text{ST},[101]}$
		0.137	0.0166	8.250
	SP	$V_{\text{S,O,SP}}^{[010]}$	$V_{\text{S,O,SP}}^{[101]}$	$ V_{\text{S,O,SP}}^{[010]}/V_{\text{S,O,SP}}^{[101]} $
		1.85	0.21	8.81

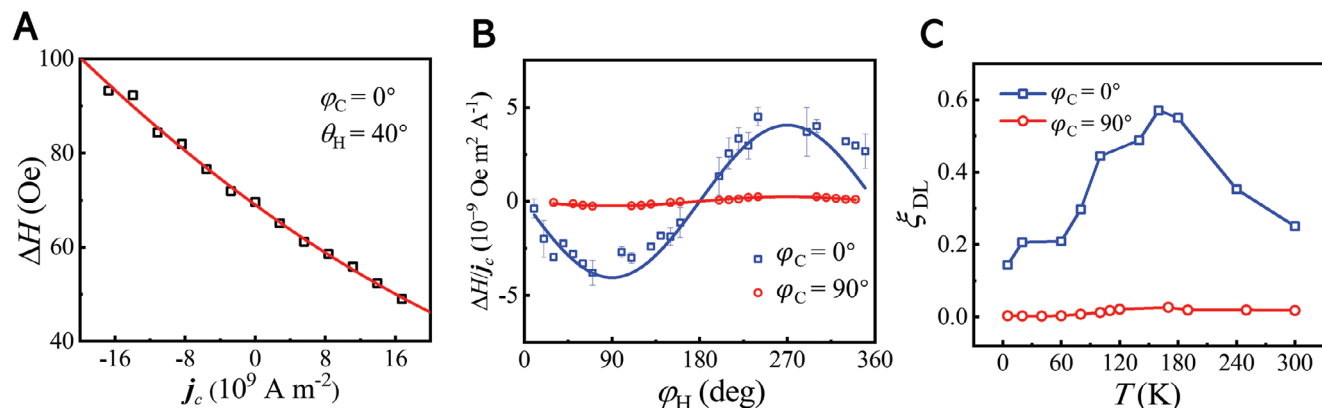


Figure 3. Experimental results for the modulation of damping. A) ΔH as a function of current density j_c in (101)-RuO₂(15)/Py(8)/MgO(2)/Ta(3) sample at $\theta_H = 40^\circ$ and the applied current along the $\varphi_C = 0^\circ$. B) The current-induced modulation of the effective resonance linewidth $\Delta H/j_c$ as a function of external magnetic field angle θ_H at $\varphi_C = 0^\circ$ (blue square) and $\varphi_C = 90^\circ$ (red square), respectively. The solid lines represent the fit to the $\sin\varphi_H$. C) The extract ξ_{DL} as a function of the temperature for (101)-RuO₂(15)/Py(8)/MgO(2)/Ta(3) sample at $\varphi_C = 0^\circ$ and $\varphi_C = 90^\circ$.

ASSE in RuO₂(101), where the decreasing trend below ≈ 160 K originates from the structural distortion of the TiO₂ substrate (see the Supporting Information). The observation of temperature-dependent characteristics of the ASST prompts us to perform its inverse effect in RuO₂(101)/Py heterostructure.

Next, the IASSE is studied by the spin pumping measurement in the stack of (101)-RuO₂(15)/Py(8)/MgO(3)/Ta(3), as shown in **Figure 4A**. The spin-pumping device configuration has a dimension of 8 μm in width and 360 μm in length. The stripe located in the gap of the coplanar waveguide (CPW) experiences an

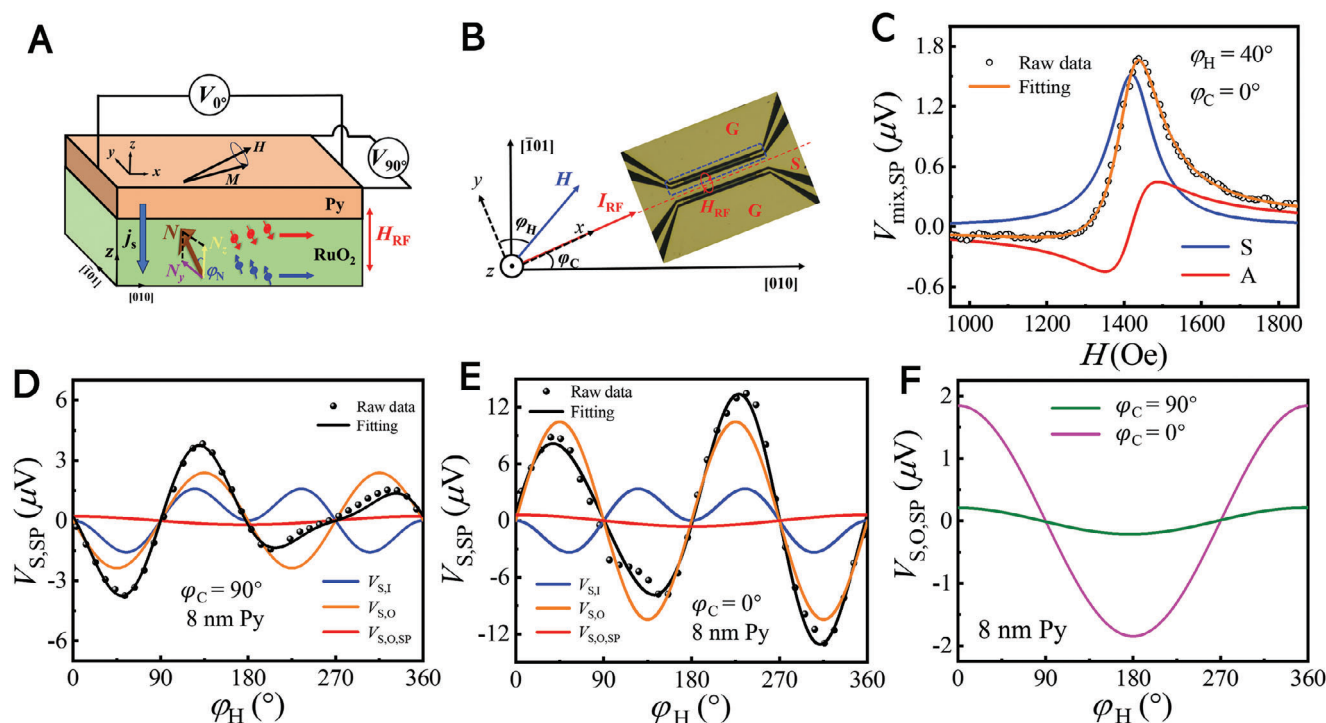


Figure 4. Angular dependence of DC voltages for the spin pumping measurement. A) The schematic diagram of the spin pumping measurement for device configuration with an out-of-plane microwave excitation field H_{RF} in (101)-RuO₂/Py. φ_N is the angle between the $\mathbf{N}$ and the z -axis. B) The actual depiction of the spin pumping device configuration with an out-of-plane microwave excitation field H_{RF} in (101)-RuO₂/Py. φ_H is the angle of the applied magnetic field H relative to the y -axis and the layout includes two orthogonal directional components V_{0° and V_{90° . C) The DC voltage signals ($V_{mix,SP}$) obtained for (101)-RuO₂(15)/Py(8)/MgO(3)/Ta(3) sample at $\varphi_H = 40^\circ$ and the spin pumping device layout along the $\varphi_C = 0^\circ$, where the fitting results include symmetric and antisymmetric parts. Symmetric voltage amplitudes ($V_{S,SP}$) as a function of angle φ_H for D) $\varphi_C = 90^\circ$ and E) $\varphi_C = 0^\circ$, respectively. Black plots and curves show the raw data and fitting parameters, contributed by the $V_{S,I}$ (blue), $V_{S,O}$ (yellow), and $V_{S,O,SP}$ (red), fitting by the Equations (5) and (6). F) The extracted pure spin pumping voltage signals ($V_{S,O,SP}$) as a function of φ_H .

out-of-plane microwave excitation field via the ground-source-ground (GSG) pads, which has the advantage of the separation of spin pumping and parasitic anisotropic magnetoresistance,^[50] as shown in Figure 4B. Here, the spin-pumping device utilizes the same thin film as the ST-FMR device in Figure 2, to enable a comparison between the direct and inverse spin-splitting effects in RuO₂. Figure 4C shows a typical spin pumping signal of $V_{\text{mix,SP}}-H$ curve (offset voltage V_{offset} is subtracted) under the microwave power $P = 23$ dBm, an excitation frequency of 10 GHz, $\phi_H = 40^\circ$ and $\phi_C = 0^\circ$ at room temperature. The symmetric (blue line) and antisymmetric Lorentzian (red line) curves are employed to describe the line-shapes of the spectra, and the $V_{\text{mix,SP}}$ is fitted well by the sum of $V_{\text{S,SP}}L_{\text{S,SP}}$ and $V_{\text{A,SP}}L_{\text{A,SP}}$, where $V_{\text{S,SP}}$ and $V_{\text{A,SP}}$ are the magnitudes of symmetric and antisymmetric components, respectively. After considering the finite inductive currents induced in the conductive RuO₂ layer which can lead to an additional in-plane magnetic excitation field, the DC voltages as a function of ϕ_H are fitted by the following equations:

$$V_{\text{A,SP}} = V_{\text{A,I}} \sin^2 \phi_H \cos \phi_H + V_{\text{A,O}} \sin 2\phi_H \quad (5)$$

$$V_{\text{S,SP}} = V_{\text{S,I}} \sin^2 \phi_H \cos \phi_H + V_{\text{S,O}} \sin 2\phi_H + V_{\text{S,O,SP}} \cos \phi_H \quad (6)$$

Here, $V_{\text{A,I}}$ ($V_{\text{A,O}}$) and $V_{\text{S,I}}$ ($V_{\text{S,O}}$) are the coefficients of $V_{\text{A,SP}}$ and $V_{\text{S,SP}}$ induced by the in-plane (out-of-plane) excitation,^[51] and $V_{\text{S,O,SP}}$ is the magnitude of the spin pumping signal (SP) induced by the out-of-plane excitation (see the Supporting Information). The ϕ_H represents the angle of the applied magnetic field H relative to the y -axis. The antisymmetric part in Figure 4C solely arises for the AMR-spin rectification as described in Equation (5). However, the amplitude of the symmetric part contains pure SP ($V_{\text{S,O,SP}}$) and AMR-spin rectification parts. The $V_{\text{S,SP}}$ signal as a function of ϕ_H is extracted from the Lorentzian line-shape to acquire the $V_{\text{S,O,SP}}$ by Equation (6), as shown in Figure 4D,E, corresponding to $\phi_C = 90^\circ$ and 0° , respectively. For the pure SP contribution, the obtained value of $V_{\text{S,O,SP}}^{[\bar{1}01]}$ is 0.21 μV in Figure 4D with $\phi_C = 90^\circ$ owing to the conventional spin-to-charge ($\sigma_{y(x)}$) conversion with the x -axis spin polarization, without the spin polarization component from N in yz -plane (the schematic diagram of Figure 4A). Note that the J_s is along the z -axis and the detected DC voltage V_{90° is along the y -axis. In contrast, the $V_{\text{S,O,SP}}$ in the case with $\phi_C = 0^\circ$ is increased to 1.85 μV ($V_{\text{S,O,SP}}^{[010]}$) in Figure 4E when V_{90° is detected along the x -axis. Here, the spin polarization in the y -axis (σ_y) includes the conventional $\sigma_{y(y)}$ and N -dependent $\sigma_{Ny(y)}$, indicating that the enhanced charge current arises from the IASSE-induced spin-to-charge conversion in the RuO₂ layer. Subsequently, Figure 4F shows the comparison of the pure SP contributions with the voltage signal along the orthogonality directions ($\phi_C = 90^\circ$ and $\phi_C = 0^\circ$) obtained from the fitting of Figure 4D,E, and a substantial enhancement of $V_{\text{S,O,SP}}$ is observed at $\phi_C = 0^\circ$ compared to $\phi_C = 90^\circ$, where this difference exhibits the features of IASSE with the Néel vector-dependent spin-to-charge conversion, which is also consistent with theoretical prediction^[25] and experimental results.^[43] As presented in Table 1, a comparison of both spin pumping and ST-FMR for the (101)-RuO₂(15)/Py(8)/MgO(3)/Ta(3) film at $\phi_C = 90^\circ$ [$\bar{1}01$] and $\phi_C = 0^\circ$ [010], it reveals that the anisotropy in the spin-pumping signal ($|V_{\text{S,O,SP}}^{[010]}/V_{\text{S,O,SP}}^{[\bar{1}01]}| = 8.25$) is consistent with the anisotropy

observed in the ST-FMR signals ($\xi_y^{\text{ST},[010]}/\xi_y^{\text{ST},[\bar{1}01]} = 8.81$). The direct and inverse spin splitting effects, as well as the crystal direction-dependent anisotropic behavior for the samples, confirming the spin splitting effects in the (101)-RuO₂.

Table 1 summarizes the ST efficiencies and pure spin pumping signal in (101)-RuO₂ for the two orthogonal directions $\phi_C = 0^\circ$ ([010]) and $\phi_C = 90^\circ$ ($[\bar{1}01]$).

To obtain the temperature dependence of IASSE, we perform the in-plane spin-pumping measurement in the (101)-RuO₂(15)/Py(20)/MgO(3)/Ta(3) film where a 20 nm Py film is used to ensure a sufficiently strong signal in FMR measurement, by attaching the film to a CPW holder which is integrated inside the cryogenic chamber of the physical property measurement system (PPMS). Figure 5A illustrates the actual illustration (top) and schematic diagram (bottom) of the spin pumping measurement. The (101)-RuO₂(15)/Py(20)/MgO(3)/Ta(3) film is positioned on top of the CPW and has tight winding around it, causing the microwave field to align in-plane. The detected voltage, orthogonal to I_{RF} , is measured by attaching two electrodes at the ends of a 4.5 mm distance to quantify the induced voltages. Initially at room temperature, the (101)-RuO₂(15)/Py(20)/MgO(3)/Ta(3) film is measured with $\phi_H = 0^\circ$ and $\phi_C = 0^\circ$, at 6 GHz and varied P ranging from 13 to 23 dBm, as shown in Figure 5B. The measured symmetric component is represented by $V_{\text{sym}}(\phi_H) = V_{\text{SP}}^{\text{sym}} \cos \phi_H + V_{\text{AMR}}^{\text{sym}} \sin 2\phi_H \sin \phi_H$, where $V_{\text{SP}}^{\text{sym}}$ and $V_{\text{AMR}}^{\text{sym}}$ denote the amplitudes of the pure SP and AMR-spin rectification contributions to the symmetric component, respectively. Notably, the AMR-spin rectification contribution becomes null at $\phi_H = 0^\circ$ for a pure Py layer, resulting in a symmetric signal for the RuO₂/Py sample solely attributed to SP. Hence, we concentrate on the $\phi_H = 0^\circ$ configuration with the magnetic field being parallel to the I_{RF} . In Figure 5B, we extract the symmetric component ($V_{\text{SP}}^{\text{sym}}$) for SP analysis, where the slight asymmetric signal might be from the induced RF driving current through the sample.^[50,52] The inset of Figure 5B shows the $V_{\text{SP}}^{\text{sym}}$ as a function of P , with a linear relation.

Then, we carry out the spin pumping measurement with $\phi_H = 0^\circ$ at different temperatures. The typical temperatures of 20 K (red) and 300 K (black) are displayed in Figure 5C, showing the H -dependent voltage signals at $P = 23$ dBm, where the reversed polarity of $V_{\text{mix,SP}}$ at positive and negative magnetic fields indicating the evidence of spin pumping effect. The extracted $V_{\text{SP}}^{\text{sym}}$ the component at varied T is summarized in Figure 5F (yellow circles). Notably, as the T decreases from 300 to 100 K, even the FMR intensity reduces at lower temperatures as shown in Figure 5D,^[53] the $V_{\text{SP}}^{\text{sym}}$ gradually increases and reaches a double value compared to that at 300 K, as shown in Figure 5F, where the transition below 100 K comes from the crystal phase transition of the TiO₂ substrate.^[54] Further to clarify the detailed relationship between the crystal phase transition of the TiO₂ substrate and the altermagnetic spin splitting effect, the Raman spectroscopy measurements and first-principles calculations of the spin splitting energy in the band of RuO₂ are performed (see the Supporting Information). Generally, the spin splitting of the band structure is much dominated at lower temperatures due to the suppression of the thermal fluctuations, which is consistent with the enhanced $V_{\text{SP}}^{\text{sym}}$ by IASSE in RuO₂ with reducing the temperature to 100 K.

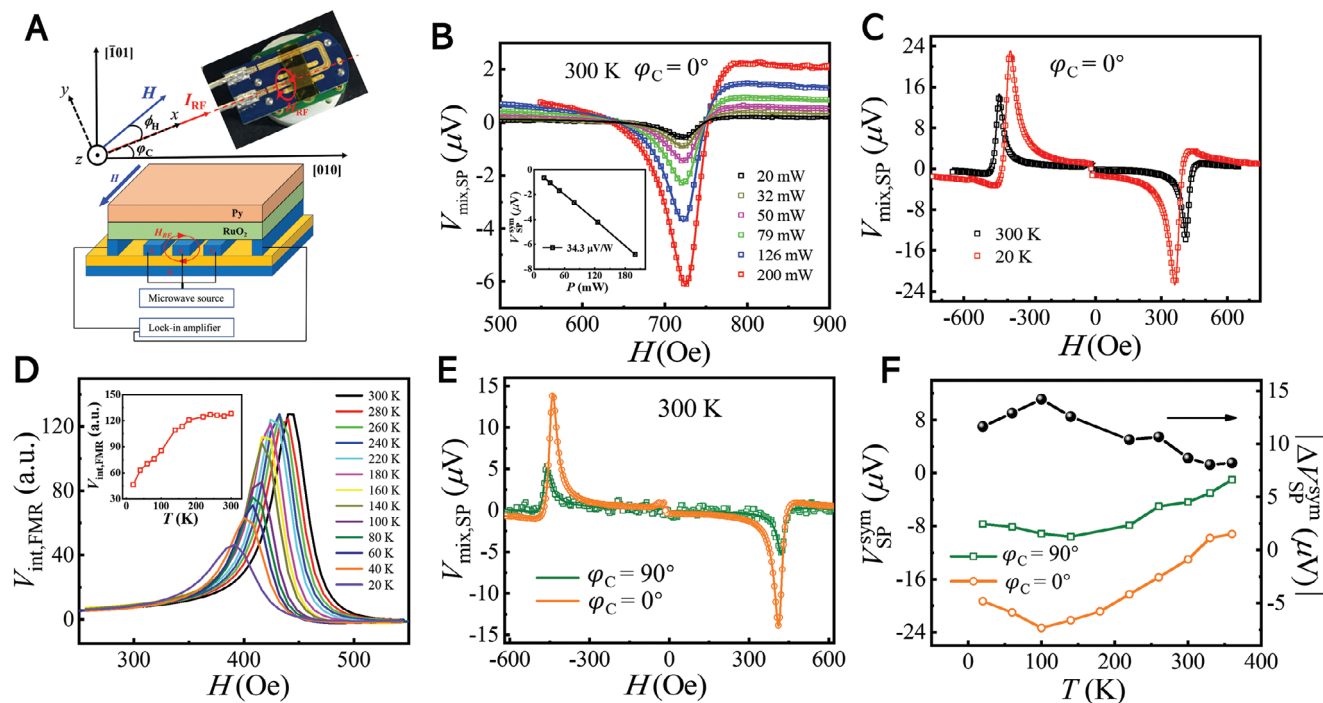


Figure 5. Temperature dependence of spin pumping signal. A) The actual illustration (top) and schematic diagram (bottom) of the spin pumping measurement with an in-plane H_{RF} in a $5 \times 5 \text{ mm}^2$ dimension of (101)- RuO_2 (15)/Py(20)/MgO(3)/Ta(3) film, attached at the top of the CPW. ϕ_H is the orientation of the applied magnetic field H relative to I_{RF} . B) The resulting DC voltages acquired with microwave power P ranging from 13 to 23 dBm at $\phi_C = 0^\circ$. The inset illustrates the P -dependence of magnitudes for the symmetric (V_{SP}^{sym} , black squares) components of the DC voltages. C) The dependence of voltage signal on magnetic field at typical temperatures T of 20 K (red) and 300 K (black). D) Integral FMR spectra for different T , the inset shows the dependence of the FMR amplitude on T . E) Comparison of magnetic field-dependent voltage signals for $\phi_C = 0^\circ$ (yellow) and $\phi_C = 90^\circ$ (green) orientations, at 300 K. F) T -dependent behavior of the V_{SP}^{sym} signal for $\phi_C = 0^\circ$ (yellow) and $\phi_C = 90^\circ$ (green) orientations, on the left axis. The right axis shows the temperature dependence of the V_{SP}^{sym} difference between these two orientations, i.e., $|\Delta V_{SP}^{sym}| - T$ (black). The above measurements are taken with $\phi_H = 0^\circ$ where the magnetic field is parallel to the I_{RF} .

The same method is performed for the detection voltage along another direction $\phi_C = 90^\circ$. In Figure 5E, the $V_{mix,SP}$ signal for $\phi_C = 0^\circ$ is 2.9 times larger than that for $\phi_C = 90^\circ$, confirming the crystal orientation-dependent IASSE of RuO_2 . Furthermore, both $\phi_C = 0^\circ$ and $\phi_C = 90^\circ$ exhibits a consistent trend as varying T ranging from 10 to 300 K, and V_{SP}^{mix} signal for $\phi_C = 0$ is significantly stronger than that of $\phi_C = 90^\circ$ at all range of temperature. The difference ($|\Delta V_{SP}^{sym}|$) between these two orientations is much more significant at lower temperatures (8.7 μV at 300 K and 14.2 μV at 100 K), indicating the much-dominated anisotropic spin splitting effect at a lower temperature. Furthermore, as the temperature increases, the spin-pumping signal gradually decreases. At 390 K, the signal-noise ratio is very low, and fitting with the Lorentzian curve is not feasible (see Supporting Information).

In the altermagnetic RuO_2 , the spin polarization direction is dependent on the Néel vector of RuO_2 due to the spin splitting effect. The crystal structure of (101)-oriented RuO_2 with the Néel vector tilted from the out-of-plane direction in y - z plane determines the spin polarization direction (y - and z -spin). We performed a comprehensive analysis of anisotropy in both ST-FMR and SP signals using a $\text{RuO}_2/\text{Py}(8 \text{ nm})$ sample. Simultaneous measurements are conducted for both ST-FMR and spin pumping devices on this film. The observed anisotropy in the spin-pumping signal aligns consistently with the anisotropy ob-

served in the ST-FMR signals. The presence of direct and inverse spin-splitting effects, along with the crystal direction-dependent anisotropic behavior in the samples, provides confirmation of the spin-splitting effects in the (101)- RuO_2 orientation.

The analysis of the differences between the spin torque efficiency (ξ_y^{ST} by the angle-dependent ST-FMR) and the DL torque efficiency (ξ_{DL} by current-modulated damping) has been discussed and refined. For $\phi_C = 90^\circ$ ($[101]$ orientation), the ASSE can be ignored, thus one can analyze the ST-FMR data by the method of general heavy metals. The spin torque efficiency quantified by angle-dependent ST-FMR contains the contribution of FL torque and DL torque. The relationship of ξ_y^{ST} , ξ_{DL} , and ξ_{FL} are written as $\frac{1}{\xi_y^{ST}} = \frac{1}{\xi_{DL}} (1 + \frac{\hbar}{e \mu_0 M_s t_{Py/RuO_2}} \xi_{FL})$. Obviously, ξ_y^{ST} is not usually equal to ξ_{DL} if ξ_{FL} cannot be ignored. For the Py/ RuO_2 bilayer, the difference between ξ_y^{ST} and ξ_{DL} is quite significant, compared with traditional heavy metals, which may be caused by the conductivity evaluation for the RuO_2 layer. We estimate the current distribution in Py/ RuO_2 by using the conductivity values that are measured in separated Py and RuO_2 films. As we know, ξ_{DL} calculated by current-modulated damping needs an accurate current density inside the RuO_2 layer in the Py/ RuO_2 heterostructure. However, the diffusion of oxygen and the interfacial electric field at the interface between the Py and RuO_2 layer changes the distribution of charge carriers near the interface and thus affect

the conductivity inside the RuO₂ layer, which leads to the difficulty of the accurate current density calculations inside the RuO₂ layer in the Py/RuO₂ heterostructure. We put most of our efforts into the crystal dependence for one specific measurement to analyze the properties of ASSE in the RuO₂ layer. Nevertheless, the differences in measured data between different methods are also very interesting and need to be further studied in the future.

3. Conclusion

In summary, we have studied the direct and inverse spin splitting effect in (101)-oriented altermagnetic RuO₂, by the ST-FMR, current-induced modulation of damping and spin pumping measurements. The direct ASSE is detected by the ST-FMR and MOD methods: the spin currents along the z-direction arising from the ASSE show an anisotropic behavior and are maximized when an electric current is along the [010] direction; MOD gives consistent anisotropic ASSE result, where the ASSE is enhanced at lower temperatures due to much-dominated spin splitting of the band structure. The spin pumping method is employed to detect the spin current by the IASSE in (101)-orientation RuO₂, and crystal direction-dependent spin pumping signals demonstrate the IASSE in RuO₂ which is along the [010] direction. Furthermore, the pronounced temperature-dependent spin pumping signals and the V_{SP}^{sym} difference between two crystal directions ([010] and $\bar{1}10$) further confirm a dominated spin splitting in RuO₂ at lower temperatures. These results give a consistent understanding of the direct and inverse spin splitting effect in altermagnetic RuO₂, with great potential for future altermagnetic materials and devices with flexibly controlled spin polarization.

4. Experimental Section

Sample Growth and Characterizations: The RuO₂/Ni₈₁Fe₁₉(Py)/MgO/Ta film was grown on the TiO₂ (101) substrate by magnetron sputtering in a system maintained at a base pressure of 3×10^{-8} Torr. During the deposition, the Ar atmosphere was maintained with a pressure of 2.5 mTorr. The RuO₂ layer was deposited by radio frequency (RF) sputtering. The Py and Ta layers were deposited by direct current (DC) sputtering and the sputtering power was 30 W. The MgO layer was deposited by RF sputtering with 80 W.

The thickness and crystal structure were characterized by X-ray reflection (XRR) and X-ray diffraction (XRD) techniques with a Bruker D8 Discover diffractometer using Cu K α radiation ($\lambda = 0.15419$ nm). By using the technique of photolithography and ion etching, the multilayer films were patterned into standard rectangular-shaped strips with $20 \times 100 \mu\text{m}^2$ for ST-FMR, $8 \times 360 \mu\text{m}^2$ for spin pumping measurements, and $20 \times 100 \mu\text{m}^2$ for longitudinal resistivity, respectively. Cr(10 nm)/Au(100 nm) metal stacks were deposited as coplanar waveguide (CPW) transmission lines or contacts for electrical measurements.

The magnetic properties of the samples were measured by the vibrating sample magnetometer (VSM) module of a superconducting quantum interface device (SQUID).

Characteristics and Methods of Magnetic Dynamics: For ST-FMR measurement, a microwave produced by a microwave signal generator was applied to the device. The RF power ranged from 13 to 23 dBm, and the frequency was ranging from 3 to 10 GHz. A mixing DC voltage would be measured by a lock-in amplifier. A bias-tee was used to divide RF and DC signals.

For spin-pumping measurements, the devices were placed in a vacuum chamber with a base pressure of 2×10^{-4} mbar and at varied temperatures

from 10 to 300 K. The microwave current with a frequency ranging from 3 to 10 GHz was used and the input microwave power ranging from 13 to 23 dBm. A microwave probe was attached to the G–S–G electrode to produce an RF magnetic field (H_{RF}) around the signal line. The heterostructure devices were placed at the gap between the signal and ground line. A DC voltage was measured by a lock-in amplifier.

For the spin pumping measurement of films, the film was tightly wound on the CPW by Kapton tape. The FMR measurement system, designed by NanoSC company, was used to obtain voltage signals. In this setup, two electrons were attached to the ends of the distance of 4.5 mm to acquire the induced voltages. The spin-pumping measurement was performed in a physical property measurement system (PPMS) at various temperatures from 10 to 300 K.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Keywords

altermagnetic materials, inverse spin splitting effect, spin pumping, spin splitting effect, spin–torque ferromagnetic resonance

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