1. Introduction

Spintronics involves active control and manipulation of spin degrees of freedom in solid-state systems. The generation, manipulation, and detection of spin current play an important role in spintronic devices such as spin-orbit torque (SOT) magnetic random access memory. Among the several ways of creating and controlling spin current, the spin Hall effect, which converts an unpolarized charge current into a pure spin current through spin-orbit interaction, has attracted much attention since it was first observed 18 years ago. The spin Hall angle is an intrinsic property of the material which represents the efficiency of converting charge current into spin current and vice-versa, and is defined as \( \theta_{\text{SH}} = \rho_{xy}/\rho_{xx} \). Materials with low resistivity and large spin Hall angles are indispensable for achieving spintronic devices with high efficiency and low energy dissipation.

Much research has been done on spin Hall materials to meet the requirement of spintronic devices. Such materials include heavy metals with strong spin–orbit coupling such as Pt, Ta, and W. Although the resistivities of these heavy metal materials are low, their spin Hall angles are not large enough, and are far short of the requirements of commercial spin devices. In recent years, large spin Hall angles greater than 1 have been observed on topological insulator materials, such as Bi$_2$Se$_3$, Bi$_x$Se$_{1-x}$, and Bi$_x$Sb$_{1-x}$. However, the application of topological materials in spintronic devices is limited by their ultra-high resistivity and process incompatibility with semiconductors. Thus, it is still a challenge to explore spin materials with large spin Hall angles and low resistivities.

The spin Hall effect originates from three distinct microscopic mechanisms: the intrinsic mechanisms, skew scattering and side-jump. The spin Hall angle can be enhanced by introducing impurities into the metal materials, such as Au or Pd doping into Pt. Zhu et al. improved the spin Hall angle by about 60% by doping MgO into Pt, and proved that the enhancement of spin Hall effect comes from intrinsic mechanism. Besides, the skew scattering mechanism has been observed in CuIr$_2$, CuBi, and PtBi alloys, and the enhancement of the spin Hall effect in AuTa and CuPt is due to the side-jump mechanism. On the basis of magnetron sputtering growth and inverse spin Hall measurement, we report the great enhancement of spin Hall effect in Pt$_{1-x}$(TiO$_2$)$_x$ nanocomposites. A giant spin Hall angle of 1.607 ± 0.04 was obtained in Pt$_{0.94}$(TiO$_2$)$_{0.06}$, which still has a relatively low resistivity of \( \approx 65 \ \mu\Omega \, \text{cm} \), making it a strong and particularity advantageous spin Hall material from the viewpoint of energy efficiency of a spintronic device. The SOT in Pt$_{1-x}$(TiO$_2$)$_x$/Co/Pt was evaluated by harmonic Hall voltage analysis and current-induced...
magnetization switching measurement. The critical switching current density of Pt0.94(TiO2)0.06/Co/Pt is reduced to $2.5 \times 10^6$ A cm$^{-2}$. We also prove that the enhancement of spin Hall angle is due to the side-jump induced by TiO$_2$ impurities in Pt. The discovery of a giant spin Hall effect in Pt$_{1-x}$(TiO$_2$)$_x$ provides new route for constructing highly efficiency spin Hall materials for SOT magnetic random access memory.

2. Results and Discussion

2.1. Sample Details

We grew yttrium-iron-garnet (YIG) 200/Pt$_{1-x}$(TiO$_2$)$_x$ d bilayers (200 and d are thicknesses in nm) on gadolinium gallium garnet (GGG) substrates via liquid phase epitaxy and magnetron sputtering. The crystal structure of YIG was characterized by transmission electron microscope (TEM) (Section A, Supporting Information). The X-ray diffraction (XRD) $\theta$–2$\theta$ patterns of Pt$_{1-x}$(TiO$_2$)$_x$ is shown in Figure 1a. There are mainly two diffraction peaks of Pt, namely fcc (111) diffraction peak near $2\theta = 39.7^\circ$ and fcc (200) diffraction peak near $2\theta = 45.9^\circ$. With TiO$_2$ doping, the fcc (111) peak does not shift and is located at the Bragg angle of Pt, indicating that the diffraction is from the periodic Pt lattice and that the TiO$_2$ are primarily dispersed in the Pt as interstitial impurity rather than being substituted into the Pt lattice. Figure 1b shows the x-ray photoelectron spectroscopy (XPS) spectrum of the Pt$_{0.94}$(TiO$_2$)$_{0.06}$ (50 nm) for the survey range 0–1100 eV, indicating that the sample composed of Pt, Ti, and O. As shown in Figure 1c, the Pt 4f$_{7/2}$ peak is located at 71.6 eV, which is close to $71.6$ eV for Pt$^{0}$.**[1] In contrast, the binding energy of Ti 2p$_{3/2}$ and O 1s peaks in the sample are located at 458.25 eV, 463.23 eV, and 530 eV, respectively, which are close to 458.46, 464.23, and 530 eV for TiO$_2$.**[1,2] Furthermore, the elemental content ratio of Ti and O obtained by calculating
the peak area is about 1:2. Therefore, we can judge that Ti atoms are oxidized while Pt atoms are not oxidized, and Ti and O exist in Pt$_{1-x}$(TiO$_2$)$_x$ film in the form of TiO$_2$. The O 1s peak located at 532.2 eV originates from water vapor attached to the surface of the film during testing. Figure 1d shows cross-sectional super energy-dispersive x-ray spectroscopy (EDS) mapping Pt, Ti, and O in the composite Pt material under the high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM), which indicates that TiO$_2$ molecules were evenly distributed in Pt without obvious aggregation within the resolution. The high-resolution transmission electron microscopy (HR-TEM) image of YIG/Pt$_{1-x}$(TiO$_2$)$_x$ in Figure 1e indicates that the Pt$_{1-x}$(TiO$_2$)$_x$ layer has a polycrystalline structure. Figure 1d,f shows HAADF-STEM image and aberration correction transmission electron microscope (AC-TEM) image of Pt$_{1-x}$(TiO$_2$)$_x$ layer, which indicates that TiO$_2$ molecules in Pt did not aggregate to form clusters and Pt essentially maintained long-range fcc order.

2.2. Spin Pumping and Inverse Spin Hall Effect

Among the several approaches,[4,23,24] spin pumping[25,26] is an effective and widely used method to generate spin current. Spin pumping and inverse spin Hall effect (ISHE) experiments are common methods to measure the spin Hall angle of heavy metals.[27] In our study, a spin current was injected from YIG into Pt$_{1-x}$(TiO$_2$)$_x$ through spin pumping excited by ferromagnetic resonance (FMR) in YIG/Pt$_{1-x}$(TiO$_2$)$_x$ bilayer structure. Because of inverse spin Hall effect, the spin current generated is converted into charge current, which can be probed when charge accumulates at the edges of the sample, as showed in Figure 3a. The injection efficiency of the spin current is expressed by the spin mixing conductance $g_{\text{eff}}^{\uparrow \downarrow}$, obtained from the damping enhancement in FMR using[28–31]

$$g_{\text{eff}}^{\uparrow \downarrow} = \frac{4 \pi M_s t_{\text{YIG}}}{g_B}(\alpha_{\text{YIG/NM}} - \alpha_{\text{YIG}})$$  \hspace{1cm} (1)

Here, $4 \pi M_s$ and $t_{\text{YIG}}$ are the saturation magnetization and thickness of the YIG film; $g_B$ and $\mu_B$ are Lande factor and Bohr magneton; $\alpha_{\text{YIG/NM}}$ and $\alpha_{\text{YIG}}$ are effective damping constants of the YIG/nonmagnetic material (NM) bilayer and the bare YIG bilayer, which are obtained from the frequency dependence of the FMR linewidth measured with a microstrip transmission line. Notably, $g_{\text{eff}}^{\uparrow \downarrow}$ is the real part of the complex spin mixing conductance $G^{\uparrow \downarrow}(G^{\uparrow \downarrow} = g_{\text{eff}}^{\uparrow \downarrow} + ig_{\text{eff}}^{\uparrow \downarrow})$.[32] The $g_{\text{eff}}^{\uparrow \downarrow}$ describes an exchange magnetic field that causes a precession of the accumulated spin. Because $g_{\text{eff}}^{\uparrow \downarrow}$ is difficult to determine and much lower than $g_{\text{eff}}^{\uparrow \downarrow}$,[32] the contribution of $g_{\text{eff}}^{\uparrow \downarrow}$ is negligible in our calculation. For the FMR measurements samples were capped on a microstrip line during tests, and the external magnetic field $H$ was parallel to the microstrip line. The variation in the S21-parameter was tested using a vector network analyzer with a magnetic field at different microwave frequencies. This enabled determination of the FMR position and linewidth. Figure 2a shows the FMR results of YIG (200 nm) and YIG (200 nm)/Pt$_{1-x}$(TiO$_2$)$_x$ (10 nm) at a microwave frequency of 7 GHz. The FMR linewidth ($\Delta H_{\text{FMR}} = \sqrt{3} \Delta H_{p-p}$) of YIG clearly increased after capping by the Pt$_{1-x}$(TiO$_2$)$_x$ composite film, which indicated the injection of spin current. Figure 2b shows the relationship between FMR microwave frequency $f$ and the peak area is about 1:2. Therefore, we can judge that Ti atoms are oxidized while Pt atoms are not oxidized, and Ti and O exist in Pt$_{1-x}$(TiO$_2$)$_x$ film in the form of TiO$_2$. The O 1s peak located at 532.2 eV originates from water vapor attached to the surface of the film during testing. Figure 1d shows cross-sectional super energy-dispersive x-ray spectroscopy (EDS) mapping Pt, Ti, and O in the composite Pt material under the high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM), which indicates that TiO$_2$ molecules were evenly distributed in Pt without obvious aggregation within the resolution. The high-resolution transmission electron microscopy (HR-TEM) image of YIG/Pt$_{1-x}$(TiO$_2$)$_x$ in Figure 1e indicates that the Pt$_{1-x}$(TiO$_2$)$_x$ layer has a polycrystalline structure. Figure 1d,f shows HAADF-STEM image and aberration correction transmission electron microscope (AC-TEM) image of Pt$_{1-x}$(TiO$_2$)$_x$ layer, which indicates that TiO$_2$ molecules in Pt did not aggregate to form clusters and Pt essentially maintained long-range fcc order.

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FMR field $H_{\text{FMR}}$. We can obtain $4\pi M_s = 1750 \pm 20 \text{ Oe}$ and the gyromagnetic ratio $\gamma = 2.81 \pm 0.002 \text{ MHz Oe}^{-1}$ using [33]

$$f = \gamma \mu_0 \sqrt{H_{\text{FMR}} + 4\pi M_s}$$

(2)

where $\mu_0$ is the magnetic permeability. As shown in Figure 2c, to obtain the damping value of YIG (200 nm) and YIG (200 nm)/Pt$_{1-x}$(TiO$_2$)$_x$ (10 nm), we extracted the FMR linewidths of the samples at different frequencies and conducted fitting through [13]

$$\Delta H_{\text{FMR}} = \frac{2\alpha_{\text{eff}}}{f} + \Delta H_0$$

(3)

Here, $\Delta H_0$ is the inhomogeneous broaden. Study have shown that the value of $\alpha_{\text{eff}}$ changes little and tends to be saturated with the thickness of NM layer covering YIG.[19] Therefore, we used the damping value of the Pt$_{1-x}$(TiO$_2$)$_x$ layer with a thickness of 10 nm to calculate $g_{x}^{\text{eff}}$ in YIG/Pt$_{1-x}$(TiO$_2$)$_x$ through Equation (1).

Figure 2d shows the variation in $\alpha_{\text{eff}}$ and $g_{x}^{\text{eff}}$ with the content of TiO$_2$ in Pt, indicating that $\alpha_{\text{eff}}$ and $g_{x}^{\text{eff}}$ decrease with increasing $x$. This is because TiO$_2$ is insulated at room temperature. When TiO$_2$ doped into Pt, the contact area between YIG and Pt will be reduced, which affects the injection efficiency of spin current, thus reducing the spin mixing conductivity of Pt/YIG. These results imply that the spin-current injection efficiency in the YIG/Pt$_{1-x}$(TiO$_2$)$_x$ heterostructure is lower than that of YIG/Pt. The specific values of the $\alpha_{\text{eff}}$ and $g_{x}^{\text{eff}}$ in YIG/Pt$_{1-x}$(TiO$_2$)$_x$ are listed in Table 1.

Spin pumping, excited by the precession of the magnetic moment caused by the microwave magnetic field ($h_{rf}$) in YIG, produces a spin current in Pt$_{1-x}$(TiO$_2$)$_x$. Owing to the ISHE, upward-flowing spin-up electrons and downward-flowing spin-down electrons deflect in the same direction. turning the spin current $J_s$ into a charge current $I_c$ given by [34]

$$J_s = \theta_{\text{SH}} \left( \frac{2e}{\hbar} \right) I_c \times \sigma$$

(4)

where $\hbar$ and $e$ is the reduced Planck constant and the electron charge, and $\sigma$ is the spin polarization vector.

In Figure 3b, we show the detected voltage as a function of external magnetic field $H$ for the YIG/Pt$_{1-x}$(TiO$_2$)$_x$ (10 nm) bilayers for an incident microwave power of 10 mW at a frequency of 5 GHz. A clear signal peak appears around the resonance field $H_{\text{FMR}}$. This must be caused by the ISHE because the detected voltage amplitude is constant and the sign is reversed when the external magnetic field is reversed, and YIG is a remarkable magnetic insulator. The voltage contribution by the anisotropic magnetoresistance or the anomalous Hall effect, assumed to produce an asymmetric Lorentzian shape in a Py/NM bilayer, are negligible.[35–37] The inverse spin Hall voltage of Pt$_{1-x}$(TiO$_2$)$_x$ is obviously higher than that of pure Pt, while the spin Hall current injection efficiency in YIG/Pt$_{1-x}$(TiO$_2$)$_x$ is lower than that in YIG/Pt, which implies that the spin Hall angle of Pt$_{1-x}$(TiO$_2$)$_x$ is greater than that of pure Pt. Although the value of $V_{\text{ISHE}}$ linewidth is different from that of FMR, their trends are consistent, and both of them gradually decrease with the increase of TiO$_2$ content (see Section B, Supporting Information for details).

The relationship between the spin Hall angle and inverse spin Hall voltage can be expressed as [10,31,35]

$$I_c = \frac{V_{\text{ISHE}}}{R} = \theta_{\text{SH}} \frac{\omega}{2\lambda_{SD}} \tanh \left( \frac{d}{2\lambda_{SD}} \right) J_{\text{eff}}^{\text{f}}$$

(5)

where $R$ and $d$ are the resistance and thickness of the NM layer, $\omega$ is the NM-layer width, $\lambda_{SD}$ is the spin–diffusion length of the NM film, and $J_{\text{eff}}^{\text{f}}$ is the effective spin–current density with $J_{\text{eff}}^{\text{f}} = \begin{pmatrix} \frac{2e}{\hbar} \gamma f \end{pmatrix} J_{\text{f}}^{\text{f}}$, in which $J_{\text{f}}^{\text{f}}$ is the spin current density from spin pumping at the YIG/NM interface expressed as [35]

$$J_{\text{f}}^{\text{f}} = \frac{\alpha_{\text{eff}}}{4\pi} \frac{\gamma f}{g_{x}^{\text{eff}}} P_c \sin^2 \theta$$

(6)

where $\alpha = 2\omega f$ is the angular frequency of microwave excitation; $\theta$ is the magnetization precession cone angle given by $\theta = \frac{h_{rf}}{2\omega f}$, in which $\alpha$ is the damping constant of YIG; and $h_{rf}$ is calibrated to be 0.05 Oe at maximum rf power $P_{rf} = 10 \text{ mW}$ (see Section C, Supporting Information for details). $P_c$ is a correction factor for the elliptical precession of the ferromagnetic magnetization. $P_c = 1.075$ is calculated from [35,38]

$$P_c = \frac{2\alpha \pi \gamma \hbar s + \sqrt{(\gamma \pi \hbar s)^2 + 4\alpha^2}}{(\gamma \pi \hbar s)^2 + 4\alpha^2}$$

(7)

Figure 3c shows the inverse spin Hall voltage $V_{\text{ISHE}}$ and charge current $I_c$ as functions of TiO$_2$ content. $V_{\text{ISHE}}$ and $I_c$ increases gradually with the increase of TiO$_2$ content, which is opposite to the behavior of $g_{x}^{\text{eff}}$. Furthermore, $I_c$ reaches the maximum when the TiO$_2$ content is 0.06. The charge current produced by ISHE in Pt$_{0.94}$(TiO$_2$)$_{0.06}$ is greater than that in pure Pt, while the spin

| Sample                  | $\rho$ [\Omega cm] | $\alpha_{\text{eff}}$ | $g_{x}^{\text{eff}}$ [m$^{-2}$] | $\lambda_{\text{SD}}$ [nm] | $\theta_{\text{SH}}$ |
|------------------------|---------------------|------------------------|-------------------------------|--------------------------|----------------------|
| YIG/Pt                 | 36                  | 7.86 x 10$^{-4}$       | 1.09 x 10$^{18}$              | 4.43 ± 0.2               | 0.051 ± 0.008       |
| YIG/Pt$_{0.94}$(TiO$_2$)$_{0.06}$ | 10               | 0.02 1.33 ± 0.005       | 0.813 ± 0.005               |
| YIG/Pt$_{0.96}$(TiO$_2$)$_{0.04}$ | 54             | 0.04 1.71 ± 0.02       | 0.517 ± 0.02               |
| YIG/Pt$_{0.95}$(TiO$_2$)$_{0.05}$ | 61             | 0.06 1.43 ± 0.04       | 0.831 ± 0.005               |
| YIG/Pt$_{0.93}$(TiO$_2$)$_{0.07}$ | 69             | 0.08 1.41 ± 0.06       | 1.607 ± 0.04               |

Table 1. Resistivity, effective Gilbert coefficient, the calculated interfacial spin mixing conductance, spin diffusion length, and spin Hall angle for each sample.
current produced by spin pumping in Pt$_{0.94}$(TiO$_2$)$_{0.06}$ is less than that in pure Pt. This indicates that Pt$_{0.94}$(TiO$_2$)$_{0.06}$ has a giant spin Hall angle relative to that of pure Pt. Figure 3d shows the thickness dependent $I_c$ for pure Pt and Pt$_{1-x}$(TiO$_2$)$_x$. The microwave frequency was 5 GHz and the incident microwave power was 10 mW. Combining these with the results from Figure 3d and Equation (5), we extracted the spin Hall angle $\theta_{SH}$ and spin diffusion length $\lambda_{SD}$ of pure Pt and Pt$_{1-x}$(TiO$_2$)$_x$; these values are shown in Table 1.

The spin Hall angle of Pt$_{1-x}$(TiO$_2$)$_x$ reaches the maximum when the TiO$_2$ content is 0.06. The maximum spin Hall angle 1.607 $\pm$ 0.04 for Pt$_{0.94}$(TiO$_2$)$_{0.06}$, which is an order of magnitude higher than 0.051 $\pm$ 0.008 for pure Pt. It is worth noting that the resistivity of Pt$_{0.94}$(TiO$_2$)$_{0.06}$ is as low as $\approx$65 $\mu\Omega$ cm. Figure 4 summarizes the resistivities $\rho$ and spin Hall angles $\theta_{SH}$ of several heavy metals, alloys, and topological insulators at room temperature. In terms of spin Hall angle, which is considered as the figure of merit for spin Hall materials, Pt$_{0.94}$(TiO$_2$)$_{0.06}$ outperforms some alloy by a factor of 2 and heavy metals (Ta, Pt) by a factor of 10, and is comparable to some topological materials (Bi$_2$Se$_3$). In addition, the very low resistivity and semiconductor compatible process of Pt$_{0.94}$(TiO$_2$)$_{0.06}$ make it more advantageous than topological materials in SOT magnetic random access memory devices.

2.3. Spin–Orbit Torque in Pt$_{1-x}$(TiO$_2$)$_x$/Co

In order to further study the advantages of the Pt$_{1-x}$(TiO$_2$)$_x$ nanocomposite films in SOT devices, Pt$_{1-x}$(TiO$_2$)$_x$(5 nm)/Co(0.8 nm)/Pt(1 nm) ($x=0, 0.04, 0.06$) multilayer structures were fabricated by magnetron sputtering and fabricated into Hall bar devices by photolithography and argon ion etching. The efficiency of SOT is tested by the second harmonic Hall voltage method (see Section D, Supporting Information for details). Figure 5a shows the first and the second harmonic Hall voltage test structure. The anomalous Hall resistance of Pt$_{1-x}$(TiO$_2$)$_x$/Co/Pt multilayer structure was tested as shown in Figure 5b. The per-
pendicular magnetic anisotropy of multilayer structures was still observed after TiO₂ doped into Pt. We measured the first and the second harmonic Hall voltage of Pt₀.₉₄(TiO₂)₀.₀₆/Co/Pt under large magnetic field scanning, which is shown in Section E, Supporting Information. The first harmonic Hall resistance does not change with the current density, while the second harmonic Hall resistance increases with the current density. This is because the second harmonic Hall resistance is related to the equivalent magnetic field generated by SOT. With the increase of TiO₂ content, the second harmonic Hall resistance signal generated by unit current density gradually increases, which indicates that with the increase of TiO₂ content, the SOT of multilayer structure increases gradually.

Figure 5c shows the first and the second harmonic Hall voltage under a small magnetic field scan when an ac current with the current density of.J = 3 × 10⁹ A m⁻². We evaluated the effective field of SOT through Equation (S5) (Supporting Information). The SOT effective field generated by different current densities for Pt₀.₉₄(TiO₂)₀.₀₆/Co/Pt is shown in Figure 5d. Through fitting calculation, we can get that when x = 0.06, the effective SOT field generated by unit current density is 629 Oe/(10¹¹ A m⁻²), which is about 35 times higher than that of pure Pt. This result is consistent with the spin Hall angles of Pt₀.₉₄(TiO₂)₀.₀₆ and Pt measured previously. Figure 5e,f shows current-induced magnetization switching of Pt₀.₉₄(TiO₂)₀.₀₄/Co/Pt and Pt₀.₉₄(TiO₂)₀.₀₆/Co/Pt. The critical switching current density of Pt₀.₉₄(TiO₂)₀.₀₄ and Pt₀.₉₄(TiO₂)₀.₀₆ is about 5 × 10⁶ and 2.5 × 10⁶ A cm⁻², which are one order of magnitude lower than that of pure Pt (Jc = 4.3–5.75 × 10⁷ A cm⁻²).

2.4. The Origin of Giant Spin Hall Effect in Platinum-Titanium Oxide Nanocomposite Films

The spin Hall effect relies on spin–orbit coupling in materials and is derived from intrinsic or extrinsic mechanisms. In intrinsic mechanisms, the spin Hall effect is typically proportional to the resistivity of the heavy metal. For extrinsic mechanisms, there
are two particular scattering mechanisms: the skew scattering, which provides a spin Hall resistivity proportional to the longitudinal resistivity caused by impurities, and side-jump, for which the spin Hall resistivity is proportional to the square of the resistivity caused by impurities. Figure 6a shows the dependence of resistivity $ρ_{Pt_{1-x}(TiO_2)_x}$, spin diffusion length $λ_{SD}$ and spin Hall angle $θ_{SH}$ on TiO2 content $x$ for Pt1−x(TiO2)x. The $ρ_{Pt_{1-x}(TiO_2)_x}$ is linearly related to the TiO2 content. It is interesting that the resistivity of Pt1−x(TiO2)x is almost twice as large as that of pure Pt when the TiO2 content is only 0.06. A small amount of TiO2 leads to a rapid increase of the resistivity of Pt1−x(TiO2)x because of the high dielectric constant of TiO2. [41] TiO2 impurities in Pt will concentrate electrons to create a local internal electric field, which causes an increase in the resistivity of Pt1−x(TiO2)x. The spin Hall angle of Pt1−x(TiO2)x increases with the TiO2 content. To identify the origin of the giant spin Hall effect in Pt1−x(TiO2)x, we plot $θ_{SH}$ as a function of $ρ_{Pt_{1-x}(TiO_2)_x}$, as shown in Figure 6b, from which we can clearly observe that there is a linear relationship between $θ_{SH}$ and $ρ_{Pt_{1-x}(TiO_2)_x}$. Unfortunately, both intrinsic and side jump lead to a linear contribution from $ρ_{Pt_{1-x}(TiO_2)_x}$ to $θ_{SH}$. Therefore, we separate the intrinsic and extrinsic contributions to the spin Hall resistivity using the following equation:

$$ρ_{SH} = σ_{SH}^int ρ_{Pt_{1-x}(TiO_2)_x} - ρ_{SH}^imp$$  \hspace{1cm} (8)$$

where $ρ_{SH}$ is the total spin Hall resistivity of Pt1−x(TiO2)x, given by $θ_{SH} = ρ_{SH}/ρ_{Pt_{1-x}(TiO_2)_x}$, $σ_{SH}^int$ is the intrinsic spin Hall conductivity of Pt, and $ρ_{SH}^imp$ is the extrinsic spin Hall resistivity induced by the TiO2. Here, we ignore the phonons contribution to the spin Hall resistivity.[42–45] For Equation (8), we consider the case where the TiO2 content $x = 0$, for which $ρ_{SH}^imp = 0$, $ρ_{Pt_{1-x}(TiO_2)_x} = ρ_{Pt}$ and $σ_{SH}^int = θ_{SH,Pt}/ρ_{Pt}$, where $θ_{SH,Pt}$ is the spin Hall angle of pure Pt. Thus, we can calculate the value of $ρ_{SH}^imp$ via\[20\]

$$ρ_{SH}^imp = ρ_{Pt_{1-x}(TiO_2)_x}θ_{SH} - (θ_{SH,Pt}/ρ_{Pt})ρ_{Pt}^2$$  \hspace{1cm} (9)$$

The plot of $ρ_{SH}^imp$ as a function of $ρ_{imp}^2$ is shown in Figure 6c. Here, $ρ_{imp}$ is the resistivity caused by TiO2 impurities, given by $ρ_{imp} = ρ_{Pt_{1-x}(TiO_2)_x} - ρ_{Pt}$. There is an obvious linear dependence of the $ρ_{SH}^imp$ on $ρ_{imp}^2$, which is consistent with the previous reported properties of the side jump.[20,46] Thus, the enhancement of the spin Hall angle originates from the giant extrinsic spin Hall effect manifested in the side-jump induced by TiO2 impurities in Pt.

For materials with strong spin–orbit coupling, such as Pt and Ta, there are always two sources of side-jump scattering:[13] extrinsic side jump arising from the non-spin–orbit-coupled part of the wave-packet scattering off the spin–orbit-coupled disorder, and intrinsic side jump arising from the non-spin–orbit-coupled part of the wave-packet scattering off the spin–orbit-coupled disorder. To identify the physical origin that the side-jump dominates rather than skew scattering in Pt1−x(TiO2)x, we determine the relationship between the two scattering mechanisms using the following equation:[47]

$$θ_{SH}^{SS} = \frac{2π}{3} \cdot \frac{k_F v_F}{(2π/h) n_{imp} v_{imp}} \cdot θ_{SH}^{SS}$$  \hspace{1cm} (10)$$
where $\theta_{\text{SH}}^{\text{imp}}$ and $\theta_{\text{SS}}^{\text{imp}}$ are the contributions of skew scattering and side-jump to spin Hall angle, respectively. $k_F$ and $n_F$ are the Fermi momentum and the Fermi velocity, $n_{\text{imp}}$ and $V_{\text{imp}}$ are the impurity concentration and the impurity potential. It can be concluded from Equation (10) that the contribution of side-jump to spin Hall angle will be far greater than that of skew scattering when the impurity potential is large. Fortunately, we have concluded above that TiO$_2$ impurities in Pt generates a large potential. Thus, giant side-jump in Pt$_{1-x}$(TiO$_2$)$_x$ originates from the scattering of strong orbital coupled electrons by the scalar potential generated by TiO$_2$ impurities, which is consistent with the contribution of intrinsic side-jump.

3. Conclusion

In summary, we fabricated Pt$_{1-x}$(TiO$_2$)$_x$ nanocomposite films with giant spin Hall angles using magnetron sputtering. The spin Hall angle of Pt$_{1-x}$(TiO$_2$)$_x$ at different TiO$_2$ contents was investigated via a combination of ferromagnetic resonance, spin pumping, and inverse spin Hall measurements. The SOT in Pt$_{1-x}$(TiO$_2$)$_x$/Co/Pt was evaluated by films with harmonic Hall voltage analysis and current induced magnetization switching measurement. A giant spin Hall angle of 1.607 $\pm$ 0.04 is obtained in Pt$_{0.94}$(TiO$_2$)$_{0.06}$ nanocomposite films with a relatively low resistivity of $\approx$65 $\mu$Ω cm, which is comparable to that of topological insulators. The critical switching current density of Pt$_{0.94}$(TiO$_2$)$_{0.06}$/Co/Pt is reduced to 2.5 $\times$ 10$^6$ A cm$^{-2}$. Particularly, this giant spin Hall effect and low resistivity make Pt$_{0.94}$(TiO$_2$)$_{0.06}$ more advantages for manipulating spin current than other heavy metals and topological insulators. This enhancement of spin Hall angle is due to side-jump induced by the impurity TiO$_2$ in Pt. Our findings provide a new route for the constructing highly efficient spin Hall nanocomposite films. This combines the advantages of giant spin Hall angle, low resistivity, and excellent process compatibility with semiconductors, for developing low power dissipation SOT magnetic random access memory or other spintronic devices.

4. Experimental Section

**Sample Fabrication:** After photolithography and Ar ion etching, the nonmagnetic layer was patterned into a Hallbar shape. Two 100 nm thick SiO$_2$ was fabricated into a Hallbar with a size of 200 $\times$ 1400 $\mu$m.

**FMR and ISHE Experiments:** Two 100 nm thick Cu was fabricated into a Hallbar with a size of 200 $\times$ 1400 $\mu$m. A Keithley 2400 source meter was used to provide a dc current. Keithley 2182A nanovoltmeter was used to detect spin Hall voltage. A Keithley 6221 source meter was used to provide a sinusoidal current and pulse current. A sinusoidal current with a frequency of 13 Hz was applied to the SOT device, and the first and second harmonic Hall voltages were detected by two phase-locked amplifiers. For current-induced magnetization switching measurement, A pulse current is first applied to the device, followed by a current of 0.1 mA to detect Hall resistance.

**Image Analysis:** The Digital Micrograph software was used to process TEM images. XPS Peak Fit software was used to perform peak splitting processing on XPS and calculate the proportion of each element in the sample. The FMR results were differentiated by Origin software. ISHE voltage was fitted by Lorentz function. ISHE currents were the results of ten averages and spin Hall angle values were represented with standard deviation error bars.

**Supporting Information**

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

**Acknowledgements**

Funding for the project was supported by the National Natural Science Foundation of China under Grant Nos. 62171096 and 61734002, National Key Scientific Instrument and Equipment Development Project No. 51827802, Sichuan Science and Technology Support Project under Grant Nos. 2021YFG0091 and 2021YFG0347.

**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

**Keywords**

ferromagnetic resonance, memory devices, nanocomposite films, spin Hall effect, spin pumping

Received: December 9, 2021
Revised: March 5, 2022
Published online: April 7, 2022
