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Electric-field control of spin-orbit torques in WS₂/permalloy bilayers

Weiming Lv,[†] Zhiyan Jia,[‡] Xin Luo,[†] Yuan Lu, Bochong Wang,[#] Zhongming Zeng,^{*,†}

Zhongyuan Liu^{*,‡}

[†] Key Laboratory of Nanodevices and Applications, Suzhou Institute of Nano-tech and Nanobionics, Chinese Academy of Sciences, Suzhou 215123, China

[‡] State Key Laboratory of Materials Science and Technology, Yanshan University, Qinhuangdao 066004, China

[‡] Institut Jean Lamour, UMR 7198, CNRS-Université de Lorraine, BP 239, 54506 Vandœuvre, France

[#] Key Laboratory for Microstructural Material Physics of Hebei Province, School of Science, Yanshan University, Qinhuangdao 066004, China

（作者顺序请曾老师安排）

ABSTRACT: Transition metal dichalcogenides (TMDs) have drawn great attention owing to their potential for electronic, optoelectronic and spintronics applications. In TMDs/ferromagnetic bilayers, efficient spin current can be generated by the TMDs for the manipulation of magnetic moment in the ferromagnetic layer. In this work, we report on the electric-field modulated spin-orbit torques (SOT) in WS₂/NiFe (Py) bilayers by spin-torque ferromagnetic resonance technique (ST-FMR). It is found that the RF current can induce a spin accumulation at WS₂/Py interface due to the interfacial Rashba-Edelstein effect. More importantly, the SOT ratio between field-like torque and anti-damping-like torque can be effectively controlled by applying back-gate voltage in WS₂/ NiFe bilayers.

These results provide a strategy for controlling spin-orbit torque with field-effect by using semiconducting TMDs.

KEYWORDS : spintronics, spin-orbit torques, transition metal dichalcogenides, Rashba-Edelstein effect, electric-field

With the development of modern magnetic memory and logic devices, the exploration of efficient and low energy consumption mechanisms to change the magnetization states has become a key issue.^{1, 2} An effective way is utilizing the strong spin-orbit interactions (SOIs)^{3, 4} to induce spin current which can be used to exert spin-orbit torques (SOT) on adjacent magnetic layer for realizing the manipulation and switching of magnetization.^{5, 6} Previous studies have confirmed the influence of SOIs in layered heavy metal/ferromagnet bilayers through the spin Hall effect in heavy metal, such as Pt,^{5, 7} Ta,^{8, 9} and W¹⁰, and other interface spin-orbit effect¹¹ in ferromagnet, such as Rashba-Edelstein effect,¹²⁻¹⁷ which can produce strong current-driven torques on the magnetic layer. Recent experiments in the material search for providing more efficient spin-orbit-induced torques have demonstrated that a large SOT can exist in topological insulators^{18, 19} and two-dimensional (2D) materials.^{20, 21} Transition metal dichalcogenides (TMDs) as one kind of 2D materials,^{12, 20, 22-24} such as MX₂ (M = Mo, W; X = S, Se, Te), have been extensively studied in the electronic and optoelectronic field due to their layered structure and unique band structure depending on thickness.^{25, 26} Moreover, the strong SOC effect and the breaking of inversion symmetry for monolayer TMDs also promote the research works on their spintronic applications, especially for the generation of SOT.²⁷⁻²⁹ Current induced field-like and damping-like torques were observed in Py/MoS₂ bilayers and the strong SOT exhibited purely interfacial nature.³⁰ A further study has experimentally shown that one can change the allowed

symmetries of SOT in spin-source/ferromagnet bilayer devices with low crystalline symmetry. An out-of-plane anti-damping torque was generated when the current was applied along a low-symmetry axis of WTe₂/Py bilayers which provided a new strategy to control SOT.³¹

Electric-field control of magnetization dynamics has been widely studied for realizing high-performance, low-power spintronic memory and logic devices. For example, the electric-field control of ferromagnetism in magnetic semiconductors through modulation of the carrier concentration,³² electric-field manipulation of magnetization reversal by using multiferroics,³³ and voltage-controlled magnetic anisotropy in ultrathin ferromagnet/oxide junctions.³⁴ The electric-field control of SOT has been reported in a Cr-doped topological insulator thin film using a top-gate field-effect transistor structure, in which the SOT strength can be modulated by a factor of four within the accessible gate voltage range, and it shows strong correlation with the spin-polarized surface current in the film.³⁵ However, the related research of electric-field control of SOT in 2D materials is still inadequate. Monolayer WS₂ is a n-type semiconductor of 2D material which shows excellent electronic and optoelectronic properties with relatively high mobility up to $\sim 100 \text{ cm}^2 \text{Vs}^{-1}$ and on/off ratio up to $\sim 10^8$.^{36, 37} It is possible to change the carrier distribution in monolayer WS₂ by applying a back-gate voltage through the dielectric layer. And, the WS₂ monolayer has extremely strong SOIs: several hundreds of millivolts in the valence bands and several tens of millivolts in the conduction band.²⁰ Therefore, it is much interesting to investigate the current induced spin transfer torque resonance phenomenon and the electric-field control of SOT effect in WS₂/Permalloy bilayer device.

In this work, high quality WS₂ monolayers were grown using CVD method and the WS₂/permalloy bilayer device was fabricated by electron beam evaporation and photolithography technique. The effect of SOT in WS₂/ NiFe (Py) bilayer was investigated by

the spin-torque ferromagnetic resonance (ST-FMR). Current induced field-like torque ($\tau_{\perp}$) and anti-damping-like torque ($\tau_{\parallel}$) were observed. Furthermore, the ratio of $\tau_{\perp}/\tau_{\parallel}$ in WS₂/Py bilayer could be modulated by applying back-gate voltage which can adjust the semiconducting nature of WS₂. Our work provided a strategy to electrically control SOT by utilizing the semiconducting TMDs.

RESULTS AND DISCUSSION

High-quality large-area monolayer WS₂ was grown on Si(p++)/SiO₂ substrate using CVD method. The structure and morphology were characterized by optical microscopy, atomic force microscopy (AFM), Raman and photoluminescence (PL) spectroscopy. As shown in Figure 1a, the distinct contrast of the optical image indicates a triangular shape of as-grown WS₂ monolayers with size up to several tens and hundreds of μm .^{38, 39} By AFM measurement (see Figure 1b) with a selected triangular WS₂ sample, a typical height of WS₂ is determined to be ~ 0.83 nm which is consistent with the theoretically and experimentally reported values of monolayer WS₂.⁴⁰ Figure 1c shows the Raman spectrum of WS₂ with characteristic modes of E_{12g} and A_{1g} at 351 cm⁻¹ and 418 cm⁻¹, respectively. The peak frequency difference of 67 cm⁻¹ indicates a clear signature of monolayer WS₂.⁴¹ Figure 1d gives a PL spectrum of the as-grown WS₂, showing a single characteristic peak at 630 nm (1.97 eV), which also indicates the WS₂ is a monolayer.⁴²

For the fabrication of WS₂/Py bilayer device, the Py (Ni₈₁Fe₁₉) was deposited directly on the surface of WS₂ with ~ 10 nm thickness to form pure interfacial contact. Photolithography and etching process were used to obtain the WS₂/Py bilayer stripe with a size of 45 μm in length and 8 μm in width. The ground-signal-ground (GSG) electrical contacts were patterned by

photolithography and Ni (10 nm)/Au (100 nm) metal electrode was deposited using electron beam evaporation. Moreover, pure Py device was fabricated on the same Si(p++)/SiO₂ substrate using the same technological process for comparison. This technological process can avoid the contact of WS₂ with photoresist and is different from the process used in the work of MoS₂/Py bilayer.³⁰ Figure 2a shows the optical image of one representative WS₂/Py bilayer device including contact GSG pads and circuit of the ST-FMR measurement. The ST-FMR technique^{5, 43} was used to measure the SOT produced by WS₂/Py bilayers at room temperature. Figure 2b illustrates the schematic device geometry and circuit of electrical-field control of the ST-FMR measurement. A RF current with fixed microwave frequency and in-plane magnetic field were applied through the bilayer at the ferromagnetic resonance condition. The oscillating current-induced torque causes the Py magnetization to precess, yielding resistance oscillations due to the anisotropic magnetoresistance (AMR) of Py layer. This change in resistance mixes with the alternating current to create a DC voltage, V_{dc} , across the stripe.

As shown in Figure 2b, two vector components of the current-induced torque, in the $\hat{\mathbf{m}} \times (\hat{\mathbf{y}} \times \hat{\mathbf{m}})$ ($\parallel$, in-plane) and $(\hat{\mathbf{y}} \times \hat{\mathbf{m}})$ ($\perp$, perpendicular) directions, are respectively obtained from the amplitudes of the symmetric and anti-symmetric components of the resonance lineshape.^{18, 30, 31} We interpret the ST-FMR signals within a macrospin approximation for the magnetization direction using the Landau-Lifschitz-Gilbert-Slonczewski (LLGs) equation of motion.⁵ The magnetization equation of motion can be written as follows:

$$\frac{d\hat{\mathbf{m}}}{dt} = -\gamma \hat{\mathbf{m}} \times H + \alpha \hat{\mathbf{m}} \times \frac{d\hat{\mathbf{m}}}{dt} + \gamma \tau_{\perp} \hat{\mathbf{m}} \times \hat{\mathbf{y}} + \gamma \tau_{\parallel} \hat{\mathbf{m}} \times (\hat{\mathbf{y}} \times \hat{\mathbf{m}}) \quad (1)$$

where γ is the gyromagnetic ratio, α is the Gilbert damping parameter and H is the applied magnetic field.³⁰ The out-of-plane $\tau_{\perp}$ is the magnitude of the field applying field-like torque and

the in-plane $\tau_{//}$ is the magnitude of the field applying anti-damping-like torque. The ST-FMR mixing voltage has the following form:

$$V_{dc} = V_S \frac{\Delta H^2}{4(H-H_0)^2 + \Delta H^2} + V_A \frac{4\Delta H(H-H_0)}{4(H-H_0)^2 + \Delta H^2} \quad (2)$$

where H_0 is the applied magnetic field at ferromagnetic resonance, and ΔH is the linewidth.^{31, 44}

The symmetric and anti-symmetric amplitudes, V_S and V_A , can be obtained by fitting eq 2 to measured DC voltage as a function of applied magnetic field. The symmetric and anti-symmetric amplitudes, V_S and V_A , are related to the two components of torque as follows:³¹

$$V_S = - \frac{I_{RF}}{2} \left(\frac{dR}{d\phi} \right) \frac{1}{\alpha\gamma(2H_0 + \mu_0 M_{eff})} \tau_{//} \quad (3)$$

$$V_A = - \frac{I_{RF}}{2} \left(\frac{dR}{d\phi} \right) \frac{\sqrt{1 + \mu_0 M_{eff}/H_0}}{\alpha\gamma(2H_0 + \mu_0 M_{eff})} \tau_{\perp} \quad (4)$$

The torque ratio $\tau_{\perp}/\tau_{//}$ can be obtained from eq 3 and eq 4 as follows:

$$\frac{\tau_{\perp}}{\tau_{//}} = \frac{V_A}{V_S} \frac{1}{\sqrt{1 + \mu_0 M_{eff}/H_0}} \quad (5)$$

where μ_0 is the permeability in vacuum, M_{eff} is the effective saturation magnetization of Py and can be obtained from the frequency dependent of H_0 using the Kittel formula.^{32, 43}

$$f = \frac{\gamma}{2\pi} \sqrt{H_0(H_0 + \mu_0 M_{eff})} \quad (6)$$

Figure 3a shows the V_{dc} comparison for WS₂/Py and pure Py device and the fitted symmetric and antisymmetric amplitude components of the line shape. The in-plane magnetic field was oriented at 45° relative to the length direction of the Py stripe and the applied microwave power and frequency is 10 dBm and 5 GHz, respectively. In comparison with the pure Py device, the amplitudes of the symmetric and antisymmetric components of WS₂/Py both increase obviously, which is similar to that of MoS₂/Py layer.³⁰ For the pure Py device, normally we should observe no resonance signal at all. However, a very small and symmetric signal was

observed in the Py (10 nm) sample. The symmetric signal may arise from the relative phase of microwaves⁴⁵ or an Oersted field due to nonuniform current flow at the electrode contacts which lead to a self-induced precession of the magnetization.⁴⁶ Figure 3b shows the typical ST-FMR spectra for a WS₂/Py bilayer excited at different rf frequency from 4 to 10 GHz and fixed power of 15 dBm. The obtained f vs. resonant field curve is well fitted by Kittel formula eq 6 as shown in Figure 3c. From the fitting, we obtained $M_{\text{eff}} = 949.4 \pm 18.2 \text{ KAm}^{-1}$ and $4\pi M_{\text{eff}} = 1.19 \text{ T}$, which is comparable to commonly used Py ($\sim 1 \text{ T}$).⁴⁷ The microwave frequency dependence of the torque ratio $\tau_{\perp}/\tau_{\parallel}$ was calculated using eq 5 as shown in Figure 3e. The values of torque ratio $\tau_{\perp}/\tau_{\parallel}$ vary obviously from ~ 0.1 to ~ 0.5 with increasing frequency because the ratio $\tau_{\perp}/\tau_{\parallel}$ is directly proportional to the resonant field H_0 . The strong spin-orbit coupling and broken vertical symmetry together with the intrinsic inversion symmetry breaking in the monolayer WS₂ could give rise to a large Rashba-type spin splitting in our WS₂/Py bilayer.^{48, 49} Such type of spin-splitting can give rise to a field-like torque base on the theoretical calculation^{50, 51}. In the measurement, large anti-symmetric voltages V_A indicate the existence of significant field-like torques. And the potent SOT is obtained which supports the interfacial nature of the torque in WS₂/Py bilayer device. The ST-FMR measurements under different microwave power were also performed with a fixed frequency at 5 GHz. The amplitude of V_{dc} increase with applied microwave power, but the resonant fields have little change. The values of torque ratio $\tau_{\perp}/\tau_{\parallel}$ are around 0.19 with tiny fluctuation, seen in Figure S1.

Then, we performed comprehensive full angular (φ) dependence of the ST-FMR signal V_{dc} . The ST-FMR measurement was conducted at a series of angle φ from 0° to 360° at fixed frequency of 5 GHz and fixed power of 15 dBm. By fitting the ST-FMR spectra using eq 2, the symmetric ST-FMR resonance components V_S and antisymmetric components V_A as a function

of in-plane magnetic-field angle φ are obtained, shown in Figure 4a and b, respectively. Accordingly, in the heavy-metal/ferromagnet bilayer, the current induced torque amplitudes follow a $\cos\varphi$ behavior due to the spin Hall effect, the Rashba-Edelstein effect, or the Oersted field.^{5, 18} The anisotropic magnetoresistance (AMR) in Permalloy follows an $\cos 2\varphi$ angular dependence, which enters V_{dc} as $dR/d\varphi \sim \sin 2\varphi$. Two contributions yield a same angular dependence for the symmetric and anti-symmetric ST-FMR components. Therefore, V_S and V_A can be well fitted by the $\cos\varphi \sin 2\varphi$ behavior. The $\cos\varphi \sin 2\varphi$ angular dependence behavior confirms the FMR rectification is due to the interaction of the AMR mixed on the resonance with microwave.

Figure 5a shows the ST-FMR resonances signals V_{dc} at a series of back-gate voltage from -80 V to 80 V at fixed frequency 5 GHz and fixed power 15 dBm. When the external field is over 260 Oe, there is no obvious difference in the lineshape under different back-gate voltage; but below that, the curves shift downwards at back-gate from -80 V to 40 V and shift upwards from 40 V to 80 V. We also applied back-gate voltage on the Py/SiO₂/Si reference sample for the same measurement as shown in Figure S2. The lineshape remains unchanged which means influence of back-gate electric field on FMR measurement can be excluded. Taking the same analysis process, the ST-FMR measurement signals are fitted to obtain the symmetric and anti-symmetric ST-FMR components of V_S and V_A under different back-gate voltages, and the torque ratio $\tau_{\perp}/\tau_{\parallel}$ can be calculated sequentially. As shown in Figure 5b, the symmetric and antisymmetric components of V_S and V_A of WS₂/Py bilayers shows inverse variation tendency with applied back-gate voltage. The variation of V_A with back-gate voltage demonstrates similar variation tendency with the field-effect transistor transfer curves of monolayer WS₂ (as shown in Figure 5d). The torque ratio $\tau_{\perp}/\tau_{\parallel}$ was also calculated using eq 5. Figure 5c intuitively shows that

the torque ratio $\tau_{\perp}/\tau_{\parallel}$ increases with back-gate voltage from -80 V to 40 V and keep nearly unchanged from 40 V to 80 V. The variation of torque ratio implies that the SOT in WS₂/Py bilayer heterostructure can be effectively regulated and controlled by the electrical field. Interestingly, the variation of torque ratio with the back-gate voltage demonstrates similar tendency with the field-effect transistor transfer curves of the monolayer (ML) WS₂, as shown in Figure 5d. This phenomenon can be explained by the regulation of the carrier density in WS₂ and the interfacial characteristics between WS₂ and Py (including the contact properties and spin-mixing conductance) by electrical field. At positive electric-field, the carrier concentration in WS₂ increases resulting in an enhancement of spin current. The WS₂/Py bilayer shows a non-Ohmic character contact and the back-gate voltage may produce a strong Schottky barrier leading to a strong electric field. Relevant effect has been studied on spin valve structure using TMDs as spacer, such as NiFe/MoS₂/NiFe⁵² and NiFe/WSe₂/NiFe⁵³ spin valves. This effect may further induce Rashba-type spin splitting. Most importantly, the electric-field can regulate the interface transparency between WS₂ and Py. The back-gating allows the electrical field control of spin-relaxation rate of MoS₂-metallic stack (SiO₂//MoS₂/Al/Co/Al/Cu) on the spin to charge conversion in MoS₂ monolayer with spin pumping.⁵⁴ By applying back-gate voltage, the interface transparency to spin current increase and the effective spin-mixing conductance gives rise to larger total spin-relaxation rate. All mention mechanism above indicates the electrical field can effectively regulate the interface properties between WS₂ and Py and confirm the interfacial origin of SOT in WS₂/Py bilayer.

CONCLUSION

In conclusion, we experimentally investigated SOT in WS₂/Py bilayers. Field-like and anti-damping-like torques were observed in WS₂/Py bilayer due to spin accumulation of WS₂ arising

from the interfacial Rashaba-Edelstein effect. More importantly, we demonstrated the effective electric-field control of SOT ratio $\tau_{\perp}/\tau_{\parallel}$ in WS₂/Py bilayer taking advantage of the semiconductor property of WS₂. The electric-field control of torque ratio effect demonstrated similar tendency with the field-effect transistor transfer curves of the monolayer WS₂. We ascribed this phenomenon to the regulation of the carrier density in WS₂ and the interfacial characteristics between WS₂ and Py. Our results provided a strategy for manipulating spin-orbit torque compatible with field-effect semiconductor devices based on two-dimensional materials TMDs and could be beneficial for the improvement of energy efficiency for spintronic devices in the future.

METHODS

Growth of WS₂ Monolayer. High quality WS₂ monolayers were grown on a Si/SiO₂ (300 nm) substrate (Silicon Quest International Inc., USA) using a CVD method in a quartz tube. During the process of growth, sulfur powder (1 g, 99.99% purity, Alfa Aesar) was put on the upstream of quartz tube, WO₃ powder (500 mg, 99.8% purity, Alfa Aesar) was put on the downstream with an argon flow at 100 sccm at ~900°C for 50 min. The WS₂ monolayers in a triangular shape and size up to hundreds of μm were finally obtained.

Fabrication of WS₂/Py bilayer device. The permalloy (Py) was deposited directly on the surface of WS₂ at a thickness of ~10 nm using electron beam evaporation (EBE-09, China) with deposition rate of 1.5 Å/s. The Py stripe at a size of 45 μm in length and 8 μm in width was patterned using photolithography (MA6) and the outer part of the stripe was etched away (IBE-A-150). The ground-signal-ground (GSG) electrical contacts was patterned using

photolithography and Ni (10 nm)/Au (100 nm) metal electrode was deposited using electron beam evaporation.

Measurements. The monolayer WS₂ was characterized by Raman and PL spectroscopy and AFM. Both Raman and PL spectroscopy were carried out using a Horiba Jobin Yvon LabRAM HR-Evolution Raman microscope with a laser radiation of 532 nm and power of 10 μ W. The surface morphology images of sample were obtained with AFM (MultiMode8, Veeco Instruments Inc., USA). We determine the strength of current-induced torque by using a spin torque ferromagnetic resonance (ST-FMR) technique. Microwave signal produced by a generator was applied to the devices through a bias tee using an rf probe. The oscillating current-induced torques cause the Py magnetization to precess, yielding resistance oscillations. The rf current and oscillated resistance generated direct voltage V_{dc} was recorded by a nano-voltmeter. A voltage source meter (Keithley 2400) from -80 V to 80 V was applied on the gate to provide an electric field.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI:

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ST-FMR measurement results at a series of power, the power dependence of torque ratio $\tau_{\perp}/\tau_{\parallel}$, The back-gate dependence of ST-FMR signals V_{dc} for Py.

AUTHOR INFORMATION

Corresponding Author

* zmzeng2012@sinano.ac.cn.

* lzy0319@ysu.edu.cn.

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

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Graphic abstract

Figure 1. Characterization of WS₂ sample. (a) Optical image of CVD-grown WS₂ samples. The scale bar is 100 μm . (b) Height profile measured along the dash line in the insert figure. The insert is the AFM image of a selected triangular WS₂ sample region. (c) Raman spectrum and (d) Photoluminescence spectrum of WS₂ sample in (b).

Figure 2. WS₂/Py device geometry and ST-FMR measurement circuit. (a) Optical images of WS₂/Py (10 nm) bilayers device including contact pads and schematic illustration of the spin-torque FMR measurement circuit. The applied external magnetic field oriented at 45° relative to the current direction (the device long axis). The stripe size is 45 μm×8 μm. The scale bar is 50 μm. (b) Schematic of the WS₂/Py bilayer device geometry. The back-gate voltage was applied through the SiO₂ dielectric layer.

Figure 3. ST-FMR measurement results of Py and WS₂/Py bilayers. (a) Comparison of ST-FMR resonances signals V_{dc} of WS₂/Py bilayers and pure Py at fixed frequency of 5 GHz and fixed power of 10 dBm. (b) ST-FMR spectra at a series of frequencies from 4 to 10 GHz with fixed power of 15 dBm. (c) Kittel fitting of frequency vs. resonance field. (d) The frequency dependence of torque ratio $\tau_{\perp}/\tau_{\parallel}$.

Figure 4. Angular dependence of ST-FMR measurement for WS₂/Py bilayer at a series of angle φ from 0° to 180° at fixed frequency 5 GHz and fixed power 15 dBm. (a) Symmetric ST-FMR resonance components V_S and (b) antisymmetric components V_A as a function of in-plane magnetic-field angle φ . The line represent the corresponding fitting by theoretical function of $\cos(\varphi)\sin(2\varphi)$.

Figure 5. Electrical-field control of ST-FMR measurement for WS₂/Py bilayer. (a) The back-gate dependence of ST-FMR signals V_{dc} at a series of back-gate voltage from -80 to 80 V at fixed frequency 5GHz and fixed power 15 dBm. (b) The fitted symmetric ST-FMR resonance components V_s and antisymmetric components V_A at back-gate voltage from -80V to 80V. (c) The torque ratio $\tau_{\perp}/\tau_{\parallel}$ dependence on back gate voltage V_g and electric field E for Py and WS₂/Py bilayer. (d) The transfer curve of a typical monolayer (ML) WS₂ field effect transistors device.