Large Damping-Like Spin–Orbit Torque in a 2D Conductive 1T-TaS₂ Monolayer

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ABSTRACT: A damping-like spin-orbit torque (SOT) is a prerequisite for ultralow-power spin logic devices. Here, we report on the damping-like SOT in just one monolayer of the conducting transition-metal dichalcogenide (TMD) TaS₂ interfaced with a NiFe (Py) ferromagnetic layer. The charge-spin conversion efficiency is found to be 0.25 ± 0.03 in TaS₂(0.88)/Py(7), and the spin Hall conductivity $\sigma_{xy} = (14.9 \times 10^{10} \text{Ω}^{-1} \text{m}^{-1})$ is found to be superior to values reported for other TMDs. We also observed sizable field-like torque in this heterostructure. The origin of this large damping-like SOT can be found in the interfacial properties of the TaS₂/Py heterostructure, and the experimental findings are complemented by the results from density functional theory calculations. It is envisioned that the interplay between interfacial spin–orbit coupling and crystal symmetry yielding large damping-like SOT. The dominance of damping-like torque demonstrated in our study provides a promising path for designing the next-generation conducting TMD-based low-powered quantum memory devices.

KEYWORDS: Transition-metal dichalcogenide, Damping-like torque, Spin-torque ferromagnetic resonance, Planar Hall effect

Spin–orbit torques (SOTs) induced by spin currents are prerequisite to controlling the magnetization ($m$) in next-generation nonvolatile three-terminal memory devices, spin-torque nano-oscillators for microwave-assisted switching and neuromorphic computing. SOT-based magnetic memories are considered to be more reliable by utilizing low energy induced switching of the magnetization in contrast to the low endurance and low speed of two-terminal spin-transfer torque (STT)-based random access memories. A spin current with spin polarization vector $\sigma$ generated by the spin Hall effect (SHE) and/or the Rashba–Edelstein effect (REE) in the presence of high spin–orbit coupling (SOC) in a material may give rise to two types of SOTs, damping-like ($\tau_{DL} = m \times (m \times \sigma)$) and field-like ($\tau_{FL} = m \times \sigma$) torques, and have been reported for a number of heavy metals (HMs). In contrast to STT devices where the spin polarization of the charge current passing through the free layer enforce the switching of the magnetization, the physical origin of SOTs is the transfer of spin and orbital angular momenta through exchange interaction process, the latter via contributions mainly from different d orbitals. The SOT in HMs is reported to be a bulk-like phenomenon, which requires the thickness of the HM layer to be larger than its spin diffusion length in order to produce appreciable torque. However, it is difficult to control the crystallinity of the HM in the low thickness regime. Recently, very large SOT has been reported in topological insulators (TIs), but the topological surface states are quenched if the TI is deposited next to a metallic ferromagnet.
and hence a ferromagnetic insulator is required to render high SOTs,13 which implies industrial compatibility issues.14

To overcome the bulk-like effect in HMs, with perceived industrial compatibility, two-dimensional transition-metal dichalcogenides (2D-TMDs) were proposed a few years ago for spintronic applications.15−18 By replacing the HMs with TMDs, one can anticipate two positive outcomes for spin devices. First, a pure spin current can be produced by just a monolayer thick TMD without any bulk-like effect. Second, being a layered material, it is possible to realize smooth surfaces with atomic-scale flatness, i.e., in the Ångström scale. Although there are few reports on the observation of SOTs in TMDs,18,19 they are, however, encountered with the problem of a dominating field-like torque due to their semiconducting nature.16,12 The SOC in TMDs/FM at the interface can also give Dzyaloshinkii−Moriya interaction (DMI),20 which can generate chiral structures such as skyrmions. The surface quality of TMD exfoliated films grown by chemical vapor deposition is also compromised due to high roughness and strain.21,22 TMD films produced in this way exhibit inhomogeneity and are thus not suitable for spintronic device applications. Thus, we are forced to face two challenges to realize the requirement of dominating damping-like torques in TMDs. One is to grow large-area TMD films directly on SiO2 substrates and concomitantly to provide large damping-like torques by using conducting TMD/ferromagnet bilayers. Keeping in mind the growth problem of conducting TMDs, the 1T-tantalum-disulfide (TaS2) system is yet to be explored, which can be easily fabricated by the sputtering technique along with the distinctive plasma sulphurization process. The prefix 1T in 1T-TaS2 refers to 1 layer per trigonal (T) unit cell. Being conducting with high SOC,23 the 1T-TaS2 system also possesses exotic temperature-dependent properties, owing to several charge density wave (CDW) transitions,24 thus contributing with the rich physics of CDWs to the field of SOTs. Previously, researchers have reported the growth of TaS2 by various methods,24−26 which is yet to be explored for spin−orbitronics.

Here, we report a dominating damping-like torque in conducting TaS2/Pt bilayer heterostructures. The high-quality large-area TaS2 monolayers have been prepared by ion-beam sputtering combined with plasma-assisted sulfurization (see methods and materials for details in Supporting Information S1). The SOTs were measured by using spin-torque ferromagnetic resonance (ST-FMR) and angle-dependent planar Hall effect (PHE) measurements. First-principles calculations based on density functional theory envisage the possible source of the damping-like torque in the TaS2/Pt heterostructures. Micromagnetic simulations corroborate the experimental results.

Results. Monolayer Characteristics. Figure 1 shows the transmission electron microscopy (TEM) cross-sectional image of the TaS2(1 ML)/Py(7)/Al(3) heterostructure. (b) Raman spectra recorded using two lasers (532 and 785 nm) and mapping (10 × 10 μm²) in an inset on single-layer TaS2 using a 785 nm laser. (c, d) XPS spectra of Ta and S recorded on a single-layer TaS2 sample.

Figure 1. (a) High-resolution cross-sectional TEM image of the TaS2(0.88)/Py(7)/Al(3) heterostructure. Insets: (left) low-magnification cross-sectional image; (right) elemental EDX mapping of the trilayer sample. (b) Raman spectra recorded using two lasers (532 and 785 nm) and mapping (10 × 10 μm²) in an inset on single-layer TaS2 using a 785 nm laser. (c, d) XPS spectra of Ta and S recorded on a single-layer TaS2 sample.
corresponding to the 1T-TaS$_2$ phase. The uniformity of the film can be seen in the Raman mapping as recorded around the most intense Raman peak (shown in the inset). Elemental analysis has been performed by X-ray photoelectron spectroscopy (XPS) as presented in Figure 1c,d for Ta and S, respectively. Observed peaks are deconvoluted into the two spin–orbit split peaks, which confirms the TaS$_2$ formation without residual phases.

We also observed a finite shift around 1.5 ± 0.9 eV for the Ta(4f$_{7/2}$) orbital peak position in TaS$_2$ in comparison to the metal Ta, which indicates a large spin-orbit splitting in TaS$_2$. Further, surface topography and step height scans were also recorded using atomic force microscopy and confirm the monolayer thickness and the smooth interface with Ångström scale flatness of the TaS$_2$ layer (see Supporting Information, S4).

**ST-FMR Measurements.** The magnitude of the SOT efficiency governed by the spin-torque efficiency ($\theta_s$) was measured using ST-FMR. The applied field makes an angle of 45° with respect to the current as shown in Figure 2a (scanning electron microscopic (SEM) image of the device). An SEM image with the measurement circuit is shown in Figure 2b. A schematic of the torques acting on the magnetization due to the microwave current is shown in Figure 2c. The ST-FMR measurements were performed in a field-sweep mode in the frequency range of 5–16 GHz. We have used a lock-in detection technique with an electronic lock-in amplifier. The ST-FMR measurements were performed in a field-sweep mode in the frequency range of 5–16 GHz. We have used a lock-in detection technique with an electronic lock-in amplifier.

Figure 2d shows the ST-FMR spectra together with fits using the equation for $V_{\text{mix}}$ which gives the line-shape parameters. The ST-FMR spectra for positive and negative frequency scans are shown in the inset (at 12 GHz). It is to be noted that the peak changes its sign on changing the direction of the external magnetic field, indicating an damping-like torque $\tau_{DL}$ and ruling out the possibility of a dominating Oersted field generated torque ($\tau_{FL}$). The symmetric and antisymmetric amplitudes have been separately fitted to the spectra; an example for the spectrum recorded at 15 GHz is shown in Figure 2e. The effective damping ($\alpha_{\text{eff}}$) of the TaS$_2$(0.88)/Py(7) bilayer is evaluated by fitting the $\mu_0\Delta H$ versus $f$ data (as shown in Figure 2f) using the equation, $\mu_0\Delta H = \mu_0\Delta H_0 + 2\alpha_{\text{eff}}/\gamma$, where $\mu_0\Delta H_0$ is the line width contribution from inhomogeneity in the magnetic film, $\gamma$ (≈2πf) is the microwave frequency, and $\gamma/2\pi$ (28.8 GHz/T) is the magentogenic ratio. From the fitting, the values of effective damping are found to be 0.0067 ± 0.0007. The inhomogeneous line width is found to be 0.20 ± 0.02 mT, which is quite small and indicative of a smooth and clean interface of the Py/TaS$_2$ heterostructure. Further, $\mu_0M_{S\text{eff}}$ and the anisotropy field ($\mu_0H_K$) values have been calculated by fitting the $f$ versus $\mu_0H$ data to the Kittel equation, $f = \mu_0\gamma/2\pi(H + H_K)(H + H_K + H_L)$.
$M_{\text{eff}} \sim 0.02$ T (see Supporting Information, S5), which is consistent with the $\mu_0 M_{\text{eff}}$ value extracted from the ST-FMR results considering the out-of-plane anisotropy field contribution to the effective magnetization.

From the line-shape parameters, the value of the spin-torque efficiency $\theta$ is evaluated using the standard line-shape analysis method and found to be 0.023 ± 0.01. However, in this method, it is assumed that the symmetric component is purely from a damping-like torque, disregarding a possible contribution from spin pumping due to the inverse spin Hall effect and can, therefore, yield erroneous values for the SOT efficiency. Concomitantly, the antisymmetric component is considered as an Oersted field generated torque component, but it is again a naive approximation. Moreover, the line-shape analysis also shows a frequency dependency, which may lead to the wrong estimation of the effective spin-torque efficiency. Hence, to determine a reliable value of the effective spin-torque efficiency and evidence of damping-like torque, we use the so-called damping modulation scheme by applying a dc during the ST-FMR measurement, where spin pumping due to the ISHE and field-like contributions are insignificant.

**Current Induced Modulation/Changes of Effective Damping in the TaS$_2$(0.88)/Py(7) Device.** In this method, the dc-induced nonequilibrium spin accumulation at the interface, resulting due to the SHE in TaS$_2$, acts as a torque on the Py magnetization, resulting in a change of the effective damping as described by

$$\delta \alpha_{\text{dc}} = \alpha_{\text{dc}}(I_{\text{dc}} \neq 0) - \alpha_{\text{dc}}(I_{\text{dc}} = 0) = -\frac{\sin \phi}{H_0 + 0.5 M_{\text{eff}}} \frac{h_1}{2 \pi \sigma_{\text{dc}}}$$

The effective spin-torque efficiency using the dc-induced change of the damping is defined as the ratio of spin to charge current density: $\theta_{\text{eff}}^\alpha = \frac{I_{\text{dc}}}{J_{\text{dc}}}$. Here, $J_{\text{C,dc}} = \frac{\nu}{\kappa} + \frac{R_{\text{Py}}}{R_{\text{TAS}}} + \frac{R_{\text{Py}}}{R_{\text{TAS}}} R_{\text{TAS,py}}$, and $A_4$ are the resistances of permalloy and TaS$_2$, respectively, and the cross-sectional area of the device. $\sigma_{\text{dc}}$ is the angle between the magnetization and the applied field, which is 45° in our case. Figure 3a shows the change/modulation of effective damping as a function of the dc ($I_{\text{dc}}$). For comparison, the $\delta \alpha_{\text{dc}}$ values are plotted for the two directions of the magnetic field scan. The corresponding changes of $\mu_0 \Delta H$ with dc, i.e., $\delta \mu_0 \Delta H$, are shown in Figure 3b. The slopes of the $\delta \alpha_{\text{dc}}$ versus $I_{\text{dc}}$ curves for the two field directions are almost equal, which confirm that the damping-like torque acting on the magnetization in our TaS$_2$/Py bilayer is due to the SHE generated spin current. The damping values are reversed on current polarity reversal. This reversal is also observed when reversing the magnetic field direction. Therefore, the spin Seebeck and anomalous Nernst effect induced modulation of the damping can be ruled out in our measurements. The slope of the change in $\alpha_{\text{dc}}$ with dc ($\frac{\delta \alpha_{\text{dc}}}{\delta I_{\text{dc}}}$) is 2.17 ± 0.21 $\times 10^{-4}$/mA and 1.95 ± 0.17 $\times 10^{-4}$/mA for positive and negative applied fields, respectively. Using the measured resistances of Py (225 $\Omega$) TaS$_2$ (952 $\Omega$) (see Supporting Information, S8) in the equation for $J_{\text{C,dc}}$, the $\theta_{\text{eff}}^\alpha$ value is found to be 0.25 ± 0.03. Within the experimental uncertainty, the values of the spin-torque efficiency are the same for both positive and negative field scans. The obtained value is better than the values reported for other TMDs. The intercept with the current axis is known as the critical current density for auto-oscillations and estimated to be $5.13 \times 10^{10}$ A/m$^2$. ST-FMR measurements were also performed on a controlled Ta(0.9)/Py(7) device, which shows a weak signal-to-noise ratio (see Figure S11). We further measured the efficiency of various Py thicknesses with a fixed TaS$_2$ layer (see Supporting Information, S10b). The Hall conductivity (SHC) $\sigma_{\text{dc}} = \sigma_{\text{dc}} \times \theta_{\text{eff}}^\alpha$ is found to be $14.9 \times 10^4 \frac{h}{2 \pi} (\Omega \cdot m)^{-1}$, using $\sigma_{\text{dc}} = 5.9 \times 10^6 (\Omega \cdot m)^{-1}$ of TaS$_2$ and $\theta_{\text{eff}}^\alpha = 0.25$. The SHC value is $\sim$10 times smaller than the value reported for the TI Bi$_2$Se$_3$/TiO$_2$/.
The dc-induced changes of the effective damping can also be seen in the $\mu_d H$ versus $j$ results shown in Figure 3c for positive and negative direct currents. The current distribution in the heterostructure was evaluated, and it was found that 19% of the current is flowing through the TaS$_2$(0.88) layer (see Supporting Information, S8). Consequently, the Oersted field $\mu_d H_{\text{Oer}}$ in the TaS$_2$(0.88) layer is found to be $\sim 0.012$ mT/µA, which is very small, and it is, therefore, concluded that the field-like torque contribution generated by dc passing through the TaS$_2$ layer can be neglected. The ST-FMR spectra and resonance field plots shown for two currents in Figure 3d,e, respectively, show no change during a current polarity reversal, which is indicative of negligible field-like torque contributions. However, a small field-like torque contribution can arise from the unavoidable interface symmetry breaking, which is discussed in Supporting Information, S10a.

We validate the dc-induced damping-like torque in the TaS$_2$(0.88)/Py(7) device by means of magnetization in the in-plane field scan in two opposite current directions, which yield the opposite hysteresis formation, discussed in Supporting Information, S12. The magnetization switching indicates the presence of dominating damping-like torque in TaS$_2$(0.88)/Py(7). The Hall voltage hysteresis was also recorded using a Hall bar structure as is shown in Supporting Information, S11d. Further, using micromagnetic simulations, the switching of the in-plane magnetization for different current amplitudes has also been studied (see Supporting Information, Figure S13).

**Figure 4.** (a) SEM image of the TaS$_2$(0.88)/Py(7) Hall bar (100 × 20 µm$^2$) device together with a schematic of the circuit used for planar Hall effect measurements. (b) Vector components of the applied field $H$, magnetization, and the fields generated by SOTs. (c) Planar Hall effect signal recorded for two directions of the dc (magnitude = $5 \times 10^{10}$ A/m$^2$) in the presence of an in-plane magnetic field of 0.4 T. (d) Plot of the Hall resistance difference observed for two current directions versus angle $\phi$.

**Angle-Dependent Planar Hall Effect (PHE) for SOTs.** Planar Hall effect (PHE) measurements have received much attention for characterizing the SOTs in the in-plane magnetized systems. $44-46$ Figure 4a shows a scanning electron microscope (SEM) image of the Hall device used for PHE measurements. A schematic of the measurement circuit has been added to the image. In the planar Hall measurement, the sample is rotated 360° at fixed dc. The vector representation of the PHE is shown in Figure 4b, where $\phi$ is the angle between the current direction and the applied field, and $\theta$ is the angle between the current direction and the magnetization vector. The theoretical background of the PHE is discussed in Supporting Information, S11a. Figure 4c shows the PHE signal ($R_{\text{PHE}}$) versus $\phi$ recorded for two direct currents of the same magnitude but of opposite polarity. A magnetic field of 0.4 T was used for the measurements, which was enough for suppressing the field-like torque contribution. The PHE measured at different magnetic fields and currents are discussed in the Supporting Information, S11a. The difference between the curves ($R_{\text{DH}}$) is plotted as a function of $\phi$ in Figure 4d, which embraces the dominance of the damping-like torque. The $R_{\text{DH}}$ versus $\phi$ curve was fitted using eq S12 with $H_{\text{IL}}$ and $H_{\text{SO}}$ as fitting parameters. The $H_{\text{SO}}$ field determines the amount of damping-like torque acting on the ferromagnetic layer. The value is found to be 1 Oe per $10^{10}$ A/m$^2$ from which the spin-torque efficiency ($\zeta_S$) has been obtained by using the expression, $\zeta_S = \frac{2\pi \mu M_{\text{SO}} H_{\text{SO}}}{\hbar}$. The field-like contribution $H_{\text{FL}}$ has been found to be negligibly small. The dc density $J_c$ can be calculated by considering the dimensions of the device and the conductivity values at a dc value of 4.5 mA, yielding a dc density of $5 \times 10^{10}$ A/m$^2$.
and saturation magnetization, $\zeta$ is found to be 0.19 ± 0.01, which is comparable to the value obtained using the ST-FMR analysis. We have also measured the Py thickness-dependent efficiency (see Supporting Information, S11b). For comparison, we have measured the angle-dependent PHE on the Ta(0.9)/Py(7) reference sample, which shows a very weak signal-to-noise ratio (see Supporting Information, S11c). Therefore, it is confirmed without a doubt that a monolayer of 1T-TaS$_2$ produces a damping-like torque acting on the Py magnetization.

**Discussion.** It is to be pointed that the possibility of a finite field-like torque contribution is not ruled out (see Figure S10a) in this work, which is reasonable and cannot be disentangled in SOT-based systems.$^{42,43,47}$ There has been no clear evaluation of the critical current density and spin-torque efficiency in previously reported results for TMD/FM heterostructures.$^{17-19}$ The quantitative estimation of the SHC and auto-oscillations current density in our TaS$_2$/Py hold valuable information for several spintronic applications. Evidently, the interface of our TaS$_2$/Py bilayer, as confirmed by cross-sectional TEM and supported by parameters extracted from X-ray reflectivity measurements, is clean in contrast to other works using exfoliated sheets and nonuniform growth where extrinsic contributions from strain and defect-related issues$^{17,48,38}$ reduce the charge-spin conversion efficiency. First-principles calculations based on the density functional theory (DFT) reveal the role of SOC for lifting the degeneracy in the band structure of TaS$_2$/Py. Figure S2. For pristine TaS$_2$, the effects of SOC are clearly observed at $\Gamma$ and K points (see the expanded views). Specifically, at the $\Gamma$ point, the degenerate $d_{xz}$ and $d_{yz}$ bands are split due to SOC. It should be noted that this degeneracy is already lifted by the lower symmetry present at the interface between TaS$_2$ and Py due to the distorted atomic structures. On top of that, further splitting occurs due to the presence of SOC. Therefore, one can conclude that a sizable redistribution of band structure and hence the splitting of states due to the interface occurs, which becomes responsible for prominent damping-like torque. Moreover, a low-symmetry-induced out-of-plane unconventional torque$^{49}$ which mimics the out-of-plane Rashba-like or field-like torque, can also be present in this heterostructure (see Supporting Information, S10a). To highlight the contribution from different $d$ orbitals, we show in Figure 5d the orbital projected DOSs of Ta in pristine TaS$_2$ and also for the TaS$_2$/Py bilayer. Moreover, the projected DOSs of $d$ orbitals of Fe and Ni in Py are shown to reveal features of hybridization. As our energy range of interest is in the vicinity of the Fermi level, we will consider the electronic states within that energy range. It is observed that in the pristine material, $d_{xy}$, $d_{z^2}$, and $d_{xz}$ are the orbitals of interest. However, for the bilayer, the $d_{z^2}$ orbital for both spin channels is quite prominent at the Fermi level and its vicinity. Moreover, for the spin-down channel, hybridization between the $d_{z^2}$ orbitals of Ta and Ni is seen for the spin-down channel.

In conclusion, the damping-like spin-torque efficiency has been carefully investigated in the one monolayer of TaS$_2$ using ST-FMR and PHE measurements. Employing effective damping modulation or changes with dc, the effective spin-torque...
efficiency is found to be $0.25 \pm 0.03$ in TaS$_2$ (1 ML)/Py(7). Angle-dependent PHE measurements verify the spin-torque efficiency and clearly reveal the dominance of damping-like torque in our 1 ML TaS$_2$/Py bilayers. Further, the microscopic origin of the observed dominance of damping-like torque has been substantiated by DFT calculations. The observation of a dominating damping-like torque in just one monolayer provides a path for how to use TaS$_2$ in future spin–orbitronic devices.

**ASSOCIATED CONTENT**

*Supporting Information*

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.0c01955.

Full growth and characterization details, density functional theory, crystal structure and thickness analysis, atomic force microscopy, magnetization measurements, angular dependent FMR for in-plane anisotropy measurement, power calibration for ST-FMR measurement, resistivity measurement and current distribution, RF-current measurement in TaS$_2$(0.88)/Py(7) device, field-like contributions in TaS$_2$(1 ML)/Py($t_{Py}$), Py thickness-dependent damping modulation using ST-FMR, current modulation in Ta(0.9)/Py(7), planar Hall effect (PHE) for spin-orbit torques, Py thickness-dependent SOT using PHE, comparison between Py, Ta/Pt, and TaS$_2$/Py, magnetization switching in the presence of in-plane field at constant dc, micromagnetic modeling, and elemental mapping using STEM (PDF).

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**Notes**

The authors declare no competing financial interest.

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