Strong Rashba-Edelstein Effect-Induced Spin–Orbit Torques in Monolayer Transition Metal Dichalcogenide/Ferromagnet Bilayers

Qiming Shao,*†‡ Guoqiang Yu,*†‡ Yann-Wen Lan,‡‡ Yumeng Shi,§‖ Ming-Yang Li,§‖ Cheng Zheng,† Xiaodan Zhu,† Lain-Jong Li,‡ Pedram Khalili Amiri,‡ and Kang L. Wang,*†

†Device Research Laboratory, Department of Electrical Engineering, University of California, Los Angeles 90095, United States
‡National Nano Device Laboratories, Hsinchu 30078, Taiwan
§Physical Science and Engineering Division, King Abdullah University of Science and Technology, Thuwal 23955-6900, Kingdom of Saudi Arabia
†‡Research Center for Applied Sciences, Academia Sinica, Taipei 10617, Taiwan
§‖SZU-NUS Collaborative Innovation Center for Optoelectronic Science & Technology and Key Laboratory of Optoelectronic Devices and Systems of Ministry of Education, College of Optoelectronic Engineering, Shenzhen University, Shenzhen 518060, China

ABSTRACT: The electronic and optoelectronic properties of two-dimensional materials have been extensively explored in graphene and layered transition metal dichalcogenides (TMDs). Spintronics in these two-dimensional materials could provide novel opportunities for future electronics, for example, efficient generation of spin current, which should enable the efficient manipulation of magnetic elements. So far, the quantitative determination of charge current-induced spin current and spin–orbit torques (SOTs) on the magnetic layer adjacent to two-dimensional materials is still lacking. Here, we report a large SOT generated by current-induced spin accumulation through the Rashba-Edelstein effect in the composites of monolayer TMD (MoS₂ or WSe₂)/CoFeB bilayer. The effective spin conductivity corresponding to the SOT turns out to be almost temperature-independent. Our results suggest that the charge-spin conversion in the chemical vapor deposition-grown large-scale monolayer TMDs could potentially lead to high energy efficiency for magnetization reversal and convenient device integration for future spintronics based on two-dimensional materials.

KEYWORDS: Spin–orbit torque, transition metal dichalcogenides, Rashba-Edelstein effect, spintronics, charge–spin conversion, two-dimensional materials

The efficient generation of spin current is crucial for improving the energy efficiency of spintronics. The spin current can be used to exert spin–orbit torques (SOTs) on a magnetic layer, enabling the manipulation and even switching of magnetization in an energy efficient way.¹⁻² In the past decade, heavy metals, such as Pt,³⁻⁴ and Ta,⁵⁻⁸ or bulk semiconductors, such as GaAs,⁹,ⁱ⁰ have been extensively studied due to the presence of a strong spin–orbit coupling, allowing the spin Hall effect or the Rashba-Edelstein effect (REE) to generate a spin accumulation. Compared with these three-dimensional materials, the conversion between spin and charge in two-dimensional materials, that is, van der Waals materials, has not been studied until recently.¹¹

Monolayer graphene has been extensively studied as a spin channel due to its weak spin–orbit coupling.²² A modified graphene with an enhanced spin–orbit coupling strength or increased extrinsic spin-dependent scattering rates could give rise to a significant spin Hall effect. However, it requires specific treatments, such as hydrogen bonding or Cu (Au) adatoms,¹⁴ which are hard to control. A giant SOT was demonstrated in heterostructures based on three-dimensional topological insulators, that is, the Bi₂Se₃ family, which are also van der Waals materials.¹⁵⁻¹⁷ The colossal SOT originates from the topological surface states. However, a thickness larger than the hybridization length of two surface states, six quintuple layers (~6 nm), is needed for topological insulators.¹⁸ So far, it remains elusive whether we can have a large spin torque from an ultrathin atomically monolayer film (~1 nm). Monolayer transition metal dichalcogenides (TMDs), such as MX₂ (M = Mo, W; X = S, Se, Te), provide a unique platform for studying the generation of SOTs at the two-dimensional limit because monolayer TMDs have both strong spin–orbit coupling and inversion symmetry breaking.¹⁹⁻⁻²² Very recently, signatures of current-induced SOTs were found in the composite of...
monolayer MoS$_2$/ferromagnet bilayer, but the SOTs have not been quantitatively characterized, and the origin of the SOTs has not been interpreted.

In this Letter, we report the observation of current-induced SOTs in MX$_2$/CoFeB bilayers, where the MX$_2$ is monolayer MoS$_2$ or WSe$_2$. The monolayer MX$_2$ is grown by chemical vapor deposition (CVD) and has a size up to mm scale. Using a second-harmonic method, we succeeded in determining both field-like torque per unit moment (or in-plane spin–orbit effective field) and damping-like torque per unit moment (or out-of-plane spin–orbit effective field). The field-like torque is large in MX$_2$/CoFeB bilayers despite most of the current going through the CoFeB layer. The damping-like torque is negligible within measurement uncertainty, which is consistent with the REE dominated SOT generation in the MX$_2$/CoFeB bilayers. Moreover, the current-induced in-plane spin conductivity due to the REE is almost independent of temperature.

High-quality large-area monolayer MoS$_2$ and WSe$_2$ were grown on sapphire using CVD method, where the transition metal trioxides were vaporized and reacted with the S or Se vapor in a chamber under a controlled temperature and gas environment (see details in Supporting Information Section S1). The insets of Figure 1, panels a and b are the optical images of MoS$_2$ and WSe$_2$, which show the uniformity of the thin film sample. Raman spectra further confirm that the films are monolayers. The Raman spectrum of MoS$_2$ (see Figure 1a) exhibits two characteristic bands: the in-plane phonon mode, E$_{2g}^\text{in}$, centered near 385 cm$^{-1}$, and the out-of-plane phonon mode, A$_{1g}$, centered near 405 cm$^{-1}$, with a peak frequency difference of 20 cm$^{-1}$, which is a clear signature of monolayer MoS$_2$. Similarly, a high-intensity peak (E$_{2g}^\text{in}$) shows near 250 cm$^{-1}$ for WSe$_2$ (see Figure 1b), which indicates that the WSe$_2$ film is a monolayer as well. The sheet resistances of monolayer MoS$_2$ and WSe$_2$ are larger than 10$^6$ $\Omega$/sq as shown in the current–voltage curve (see Figure 1c). To study the current-induced SOTs on the magnetic moment, we deposited 3 nm of CoFeB on top of the monolayer MoS$_2$ and WSe$_2$ using a magnetron sputtering system. The deposition rates were 0.03 nm/s for CoFeB in an argon pressure of 3 mTorr. The CoFeB layer was capped by TaO$_x$ ($\sim$3 nm). For details of the deposition process and Raman characterization of MX$_2$/CoFeB bilayers after the deposition, see Supporting Information Section S2. The MX$_2$/CoFeB bilayers were patterned into Hall bars (channel width is 20 $\mu$m) using standard photolithography as shown in Figure 1, panel d. We used a second-harmonic analysis of both anomalous Hall resistance and planar Hall resistance ($R_{\text{Hall}}^{\text{anom}} = V_{\text{Hall}} / I_{\text{ac,peak}} = R_{\text{HIE}}^{\text{anom}} + R_{\text{HIE}}^{\text{anom}}$) to determine the current-induced spin–orbit effective fields in the MX$_2$/CoFeB bilayers as in refs 26 and 27. The applied a.c. current frequency is $\omega_{\text{ac}} = 35.85$ Hz. Since the magnitudes of

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**Figure 1.** Materials characterization and measurement setup. Raman spectra of monolayer (a) MoS$_2$ and (b) WSe$_2$. The inset is a large scale optical image of monolayer (a) MoS$_2$ and (b) WSe$_2$ on sapphire. The scratches in panel a reveal the color contrast between the monolayer MoS$_2$ and the substrate. (c) Current–voltage characteristics of monolayer MoS$_2$ and WSe$_2$. The inset is an optical image of Hall bar structure used for the measurement. (d) Measurement setup of SOT measurements for the MX$_2$/CoFeB bilayer. The MX$_2$ is a single layer, and the thickness of the CoFeB layer is 3 nm. (e) Illustration of induced spin accumulation by the REE at the interface of MX$_2$/CoFeB under an external electric field. The dashed gray circles are Rashba spin-split Fermi contours in the equilibrium, and the solid red circles are Fermi contours under an applied electric field. The blue arrows represent the spin angular momenta.
harmonic Hall resistance as a function of in-plane azimuthal angle (φ) with an external magnetic field 1000 Oe applied. The black solid curve is fitted curve using $R_\parallel \sin \varphi + R_\perp \cos 2\varphi \sin \varphi$, where the first and second terms are plotted in blue dotted and red solid curves, respectively. (d) The extracted $R_\parallel$ as a function of the external magnetic field along the $\pm y$ direction. The red solid curve is fitted curve using $R_\parallel \frac{H}{H_\parallel}$, where the (field-like) in-plane spin–orbit field $H_\parallel$ is determined to be 0.18 Oe.

second-harmonic signals are proportional to the a.c. current amplitude, here we only present the results using an a.c. current amplitude 1 mA ($r.m.s.$ value).

We examine the magneto-transport properties of the MX$_2$/CoFeB bilayers using a physical properties measurement system at $T = 300$ K unless otherwise stated. In the following, we will first present the results for the MoS$_2$/CoFeB bilayer and subsequently the WSe$_2$/CoFeB bilayer when we discuss the results. The MoS$_2$/CoFeB bilayer shows an in-plane easy plane, and the effective anisotropy field ($H_\parallel$) is $-1$ T (see Figure 2a). Here, we define the perpendicular magnetic anisotropy by a net magnetization $M$ in the direction perpendicular to the film plane, and the effective anisotropy field $H_{\text{an}}$ is defined in the following way:

$$H_{\text{an}} = H_\parallel \cos 2\varphi \sin \varphi + R_\perp \sin \varphi$$

$$= R_\parallel \frac{H_\parallel}{H_{\text{an}}} \cos 2\varphi \sin \varphi + R_\perp \frac{2 R_\parallel}{|H_\parallel|} \sin \varphi$$

where the first term originates from the current-induced in-plane spin–orbit effective field ($H_\parallel$) and the second term comes from the current-induced out-of-plane spin–orbit effective field ($H_\perp$). The magnitudes of both in-plane and out-of-plane spin–orbit fields are proportional to the magnitude of current. When the current is along the $+y$ axis, the $H_\parallel$ is along the $-x$ axis and is independent with the magnetization. Therefore, the $H_\parallel$ gives rise to a field-like torque $\tau_\parallel = m \times H_\parallel$. This oscillating $H_\parallel$ induced by the a.c. current causes the magnetization to oscillate in the film plane and thus gives rise to a $R_\parallel H_\parallel \propto \cos 2\varphi \sin \varphi$ since $\frac{dR_\parallel}{d\varphi} \propto \cos 2\varphi$ and $H_{\text{an}} \propto \sin \varphi$. As shown in Figure 2, panel c, $R_{\parallel\perp}$ reaches minimum $-R_\parallel$ when the $H_{\text{an}}$ is along the $+y$ direction and maximum $R_\parallel$ when the $H_{\text{an}}$ is along the $-y$ direction.

The idea of SOT measurement is described as follows. When the injection charge current passes through the MoS$_2$/CoFeB bilayer, a net spin accumulation could develop in a direction transverse to the current direction in the film plane due to the inversion symmetry breaking and spin–orbit coupling in the monolayer MoS$_2$ or the MoS$_2$/CoFeB bilayer. In other words, the nonequilibrium spin accumulation $\sigma \propto z \times j$, where the mirror symmetry with respect to the $xy$ plane is broken, and $j$ is the current direction (along $\pm y$ direction). This current-induced spin polarization, in general, could give rise to two types of SOTs, the field-like torque ($\tau_\parallel = m \times \sigma$) and the damping-like torque ($\tau_\perp = m \times (m \times \sigma)$). Therefore, the in-plane azimuthal angle dependence of $R_{\parallel\perp}$ can be divided into two major components depending on the symmetry of current-induced SOTs (see Figure 2c and the Supporting Information Section S3):

$$R_{\parallel\perp}^{\text{field-like}} = R_\parallel \cos 2\varphi \sin \varphi + R_\perp \sin \varphi$$

$$R_{\parallel\perp}^{\text{damping-like}} = R_\parallel \sin(2\varphi) + R_\perp \cos 2\varphi \sin \varphi$$

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when the \( H_{\text{ext}} \) is along the –\( y \) direction. The field dependence of \( R_\perp \) follows \( 1/|H_{\text{ext}}| \), which is consistent with the picture of an in-plane spin–orbit field; the larger the external field, the smaller the angle the current-induced field-like torque can induce. By fitting the field dependence of the extracted \( R_\parallel \) with \( R_{\text{HH}} \), where \( H_\parallel \) is the magnetic field along the ±\( y \) direction, we obtain the magnitude of \( H_{1,\text{MoS}_2}/I_{\text{ac, peak}} \approx 0.13 \text{ Oe}/\text{mA} \) (see Figure 2d). To get a more intrinsic property for the SOT generation, we convert the \( H_\parallel \) into the effective spin conductivity \( (\sigma_\perp) \) using \( \sigma_\perp = \frac{1}{e} \frac{H_\parallel}{|H_{\text{ext}}| \epsilon} \), where \( M_{\text{FM}} \) is the saturation magnetization per unit area for the 3 nm CoFeB layer, and \( \epsilon \) is the applied electric field in the MoS\(_2\)/CoFeB bilayer. We independently determine \( M_{\text{FM}} = 2.34 \) mA using superconducting quantum interference device and \( \epsilon = 3.21 \times 10^4 \) \text{V/m} for \( I_{\text{ac, peak}} = 1 \text{ mA} \). Therefore, the corresponding effective in-plane spin conductivity for MoS\(_2\)/CoFeB is \( \sigma_{\perp,\text{MoS}_2} \approx 2.88 \times 10^7 \text{eV/m} \). If we consider that the electronic conductivity of the monolayer MoS\(_2\) is very low, the intrinsic ratio of generated spin current density over charge current density, or the so-called “spin torque ratio”, will be relatively high and comparable with traditional heavy metals such Pt and Ta (will be discussed further).

The current-induced \( H_\parallel \) changes its direction when the direction of magnetization is reversed, and thus it gives rise to a damping-like torque \( \tau_\| = m \times H_\parallel \). The \( H_\parallel \) induced by the a.c. current causes the magnetization to oscillate out of the film plane and thus gives rise to a \( R_{\text{HH},\text{ANE}} \) term \( \propto \sin \phi \) since \( \frac{d\theta_{\text{HH}}}{d\theta} \vert_{\theta=90^\circ} = \frac{R_\perp}{R_\parallel} \frac{d\theta_{\text{ANE}}}{d\theta} \vert_{\theta=90^\circ} = \frac{R_\perp}{R_\parallel} \) and \( H_\perp \propto m \times \sigma \propto \sin \phi \). The \( R_\parallel \) decreases as the external magnetic field increases according to eq 1. As shown in Figure 3, panel a, we do see a distinct field dependence of the extracted \( R_\| \) for the Ta(0.8 nm)/CoFeB bilayer (as the control sample), and the estimated \( H_{1,\text{Ta}}/I_{\text{ac, peak}} \) is around 2.71 Oe per 10\(^{11}\) A/m\(^2\). This value is consistent with the previously reported values on the ultrathin Ta film with the same thickness.\(^{28}\) In addition to the field-dependent \( R_{\text{HH},\text{ANE}} \), there is an additional step function \( R_{\text{HH},\text{ANE}} \) as illustrated in the inset of Figure 3, panel a; the \( R_{\text{HH},\text{ANE}} \) changes sign as the magnetization direction reverses, while the magnitude does not vary with that of the external magnetic field. This step function is due to the anomalous Nernst effect (ANE), a thermoelectric effect\(^{27}\) (see the Supporting Information Section S4). Nevertheless, we can differentiate the thermo-voltage \( (V_{\text{Hall}}) \) and the SOT-induced second-harmonic anomalous Hall resistance \( (R_{\text{HH},\text{ANE}}) \) by their field dependencies. For the MoS\(_2\)/CoFeB bilayer, as shown in Figure 3, panel b, within measurement uncertainty, we do not observe a clear trend that the \( R_\perp \) decreases as field increases (see Supporting Information Section S5 for more information about the measurement uncertainty due to the choice of fitting range of the magnetic field). The observed negative \( V_{\text{Hall}} \) when the magnetization is along the +\( y \) direction is consistent with the ANE picture (see Figure 3b). When the magnetization reverses, the \( V_{\text{Hall}} \) becomes positive as expected from the ANE. Therefore, the ANE dominates in the observed \( V_{\text{Hall}} \) and the damping-like torque or \( H_\parallel \) is not observed within measurement uncertainty.

Here, we interpret that the REE is the mechanism for the observations of a large \( H_\parallel \) and a negligible \( H_{1,\text{Ta}} \) in the sapphire/ MoS\(_2\)/CoFeB/TaO\(_x\) heterostructure (see the Supporting Information Section S6). The REE appears in the presence of a electric potential gradient and strong spin–orbit coupling, that is, typically at the interface between a material with strong spin–orbit coupling and a different material, such as the Bi/Ag interface.\(^{29}\) In our MoS\(_2\)/CoFeB bilayer, the strong spin–orbit coupling and the broken vertical symmetry (together with the intrinsic inversion symmetry breaking in the monolayer MoS\(_2\)) could give rise to a large Rashba-type spin splitting.\(^{30}\) The Rashba Hamiltonian can be expressed by \( H_R = \alpha_R (k \times z) \times \sigma \), where \( \alpha_R \) is the Rashba coefficient, \( k \) is the electron momentum, and \( \sigma \) is the spin Pauli matrices. As shown in Figure 1, panel e, at the equilibrium state, there is no net spin accumulation due to an equal number of electrons moving in two directions. Under an external electric field along the +\( y \) direction, the Rashba spin-split Fermi contours shift, resulting in a net spin accumulation along the –\( x \) direction, which is consistent with the direction of the observed in-plane spin–orbit field. Moreover, theoretical calculation shows that to the first order, the Rashba spin-splitting can only give rise to a field-like torque\(^{31}\) or the Rashba effect gives a much larger field-like torque compared with the damping-like torque\(^{32}\) as we observed in the MoS\(_2\)/CoFeB bilayer. If the spin Hall effect
plays an important role in the MoS2/CoFeB, we should have seen a sizable damping-like torque like the Ta/CoFeB (see Table 1). However, we did not observe any significant damping-like torque in the MoS2/CoFeB. Regarding the charge-spin conversion efficiency, it has been shown that the inverse REE can convert the spin current into the charge current, and the efficiency is quantified as \( \lambda_{\text{REE}} = \alpha \tau / \hbar \), where \( \tau \) is the effective spin relaxation time, and \( \hbar \) is the reduced Planck constant. Since the valley and spin are coupled in monolayer MoS2, the relaxation time of spin polarization could be longer than 1 ns due to the considerable energy required for flipping the valley index. Therefore, the charge-spin conversion efficiency could be very high in the MoS2/CoFeB bilayer.

From the harmonic measurement, we learn that the in-plane effective spin conductivity is \( \sigma_{\parallel, \text{MoS2}} \approx 2.88 \times 10^8 \Omega^{-1} \text{m}^{-1} \), even when most of the current do not flow through the MoS2 layer. The conductivity of MoS2 in the MoS2/CoFeB bilayer is around \( \sigma_{\text{MoS2}} \approx 2.1 \times 10^8 (\Omega^{-1} \text{m}^{-1}) \) (assuming that the thickness of monolayer MoS2 with van der Waals gaps is 0.8 nm), and thus, the spin torque ratio, that is, the ratio of spin current density over charge current density, is given by \( \sigma_{\parallel, \text{MoS2}}/\sigma_{\perp, \text{MoS2}} \approx 0.14 \). If we can find an intrinsic MoS2 with a much higher resistivity (>10⁶ Ω/sq), for example, by putting a monolayer MoS2 on top of a magnetic insulator, and assume that the \( \sigma_{\parallel, \text{MoS2}} \) remains the same, even a larger spin torque ratio (>2.3) could be obtained. A recent experiment on the spin–charge conversion, the Onsager reciprocal process of the charge–spin conversion, in the Co/Al/MoS2 heterostructure shows that the efficiency of the spin–charge conversion is very high, and the estimated \( \lambda_{\text{REE}} \) can be as large as 4.3 nm, which corresponds to a spin torque ratio as large as 12.7. More recently, spin-torque ferromagnetic resonance in the MoS2/Permalloy bilayer reveals a large symmetric Lorentzian peak compared with the antisymmetric Lorentzian peak, which could be ascribed to either a large damping-like torque or a highly efficient spin pumping-driven inverse REE. By combining the results given by the spin-torque ferromagnetic resonance measurement and the present work, we can claim that the large symmetric Lorentzian peak is mainly due to the inverse REE induced by the spin pumping, rather than the damping-like torque generated by the rf current.

To see if other TMDs can produce such a large in-plane spin–orbit field, we carry out the SOT measurement on another TMD material, WSe2. The extracted \( R_{\parallel} \) and \( R_{\perp} \) as functions of an external magnetic field along ±y direction for the WSe2/CoFeB bilayer are plotted in Figure 4, panels a and b, respectively. Similar to the MoS2/CoFeB bilayer, the in-plane spin–orbit field \( H_{\phi, \text{WSe2}} \) is larger than the \( \sigma_{\parallel, \text{MoS2}} \) and is consistent with the stronger spin–orbit coupling in the monolayer WSe2 compared with the MoS2. However, we should notice that although the monolayers MoS2 and WSe2 have very different conductivity (the monolayer MoS2 has much higher resistivity than the monolayer WSe2 in our study), they have similar spin conductivities. This result indicates that spin torques in these bilayers share the same origin, that is, REE.

We also study the temperature dependence of the current-induced in-plane spin conductivity. We do not identify the damping-like torque within the investigated temperature range. We observed that the current-induced in-plane spin conductivity is almost temperature independent (slightly increases as the temperature decreases), as shown in Figure S3, which is similar to the report on the inverse REE in the Ag/Bi interface. A possible explanation for the temperature-independent charge-spin conversion due to REE is the temperature-insensitive strength of Rashba spin-splitting and the Fermi level position. The Rashba spin-splitting developed at the MX2/CoFeB interface relies on the band structure or wave function hybridization between the MX2 and CoFeB. The band structure and the Fermi level position of MX2/CoFeB could be temperature independent as reflected in the temperature independence of resistance (slight increase as the temperature decreases) of MX2/CoFeB bilayers (see the inset of Figure 5). However, detailed theories and more experiments are still required to fully understand the results presented in this paper.

| Table 1. Current-Induced Spin–Orbit Fields in All the Devices |
|------------------|-----------------|-------------------|------------------|
| devices (nm)     | MoS2/CoFeB (3)  | WSe2/CoFeB (3)    | Ta (0.8)/CoFeB (3) |
| in-plane (field-like) spin–orbit field, \( H_{\parallel} (\text{Oe/mA}) \) | 0.13            | 0.19              | 0.15             |
| out-of-plane (damping-like) spin–orbit field, \( H_{\perp} (\text{Oe/mA}) \) | \(-0\)           | \(-0\)            | 0.35             |

Figure 4. Extracted (a) \( R_{\parallel} \) and (b) \( R_{\perp} \) as a function of an external magnetic field along the ±y direction for the WSe2/CoFeB bilayer.
In conclusion, we have shown that a current can generate a large in-plane spin–orbit effective field in a bilayer consisting of a CVD-grown large-scale monolayer TMD and a ferromagnetic layer, and this effective field is temperature-insensitive. Our findings could be beneficial for future design of spintronic devices exclusively based on two-dimensional materials, where monolayer TMDs are coupled with magnetic van der Waals materials to form heterostructures that provide novel functionalities beyond electronics and optoelectronics. For future studies of two-dimensional semiconducting TMDs, on the one hand, systematic measurements on various TMDs need to be carried out to clarify the relation between the spin–orbit coupling strength and the spin torque efficiency. On the other hand, if the ferromagnetic metal we used in this study can be replaced by a magnetic insulator, such as yttrium iron garnet, there will be no shunting problem, and the spin-charge conversion efficiency may be significantly improved. Alternatively, metallic TMDs, such as 1T’ phase WTe$_2$, have also been shown to give rise to a unique out-of-plane damping-like torque due to breaking of the in-plane mirror symmetry, which is preserved in our 1H phase TMDs. At last, we would like to mention that the REE-induced spin polarization at the atomically thin interface is expected to have a broad tunability with an external gate voltage, thus allowing for further improvement of energy efficiency for spintronic devices based on two-dimensional materials.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.6b03300.

Details of CVD of MoS$_2$ and WSe$_2$; deposition details and Raman characterization after deposition; principles of second-harmonic measurement; effect of anomalous Nernst effect on the second-harmonic measurement; discussions on determination of out-of-plane spin–orbit field; discussions on origin of the current-induced in-plane spin–orbit field; properties of investigated films (PDF)

**AUTHOR INFORMATION**

Corresponding Authors

*E-mail: sqm@ucla.edu.
*E-mail: guoqiangyu@ucla.edu.
*E-mail: wang@ee.ucla.edu.

ORCID

Qiming Shao: 0000-0003-2613-3031
Guoqiang Yu: 0000-0002-7439-6920

Author Contributions

Q.S., G.Y., and Y.-W.L. contributed equally to this work.

Notes

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

We thank Haojun Zhang, Dan Wilkinson, and Bruce Dunn for discussions and assistance with experiments. Also, we thank the four anonymous reviewers whose comments and suggestions helped improve and clarify this manuscript. This work is supported as part of the Spins and Heat in Nanoscale Electronic Systems (SHINES), an Energy Frontier Research Center funded by the US Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under Award No. DE-SC0012670. We are also very grateful to the support from the Function Accelerated nanoMaterial Engineering (FAME) Center and Center for Spintronic Materials, Interfaces and Novel Architectures (C-SPIN), two of six centers of Semiconductor Technology Advanced Research network (STAR-net), a Semiconductor Research Corporation (SRC) program sponsored by Microelectronics Advanced Research Corporation (MARCO) and Defense Advanced Research Projects Agency (DARPA). L.-J.L. acknowledges the support from King Abdullah University of Science and Technology (Saudi Arabia), Ministry of Science and Technology (MOST), and Taiwan Consortium of Emergent Crystalline Materials (TCECM).

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7519 DOI: 10.1021/acs.nanolett.6b03300 Nano Lett. 2016, 16, 7514–7520