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Multidirectional Spin–Orbit Torque Magnetization Dynamics in beyond Room Temperature Van der Waals Magnet Devices

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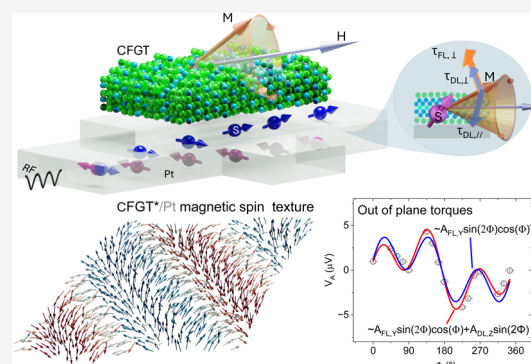
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ABSTRACT: van der Waals (vdW) magnets with room-temperature ferromagnetism offer exciting opportunities for energy-efficient spintronic devices, yet their magnetization dynamics remain largely unexplored despite their importance for high-speed memory technologies. Here, we investigate spin–orbit torque phenomena in the room-temperature vdW magnet (Co_{0.15}Fe_{0.85})₅GeTe₂ (CFGF)/Pt heterostructure using spin-torque ferromagnetic resonance and second-harmonic Hall measurements. Alongside a conventional in-plane spin Hall conductivity of 3.68×10^5 ($\hbar/2e$) ($\Omega \text{ m}$)^{−1}, we identify a sizable out-of-plane component of -0.33×10^5 ($\hbar/2e$) ($\Omega \text{ m}$)^{−1} that generates unconventional damping-like torques. Density functional theory and Monte Carlo simulations suggest that this torque can originate from interface-induced spin reorientation arising from a modified magnetic anisotropy landscape and strongly enhanced Dzyaloshinskii–Moriya interaction at the CFGF/Pt interface. The combination of low effective magnetization (0.321 T), moderate Gilbert damping (0.027), and efficient multidirectional torques highlights vdW magnets as promising platforms for next-generation spintronic devices.

KEYWORDS: spin–orbit torque (SOT), vdW magnets, 2D materials, room temperature, ST-FMR, second harmonic Hall, magnetization dynamics



Continued technological advancements drive the need for smaller, faster, nonvolatile, and energy-efficient spintronic devices for information and communication technologies.^{1,2} Spintronic memories based on spin-transfer torque (STT) phenomena have already been integrated into microcontrollers for autonomous systems, automobiles, space, gaming, artificial intelligence, and machine learning platforms.^{3–5} In recent years, the chip industry has become interested in spin–orbit torque (SOT) nonvolatile memory for high-performance computing, due to its energy efficiency, faster speeds, and higher endurance and retention compared to its counterparts.^{6–11} However, key challenges remain in achieving lower energy consumption, magnetic field-free operation, and tunability, necessary for widespread adoption in higher-level memory, memory-in-logic, and applications in artificial intelligence and neuromorphic computing.^{4–6,12,13}

The emergence of two-dimensional (2D) vdW magnets has opened a new paradigm for spintronic device engineering because of ultralow dimensionality, tunable magnetic properties, and the ability to enable energy-efficient and field-free magnetization switching mechanisms.^{14–18} Recent demonstrations of room-temperature vdW ferromagnets offer robust magnetization and a high degree of anisotropy control,^{17,19} spanning both ferromagnetic and antiferromagnetic regimes,^{17,19–23} as well as exotic spin textures^{24,25} and coexisting

magnetic orders.²⁶ These properties have enabled proof-of-concept magnetic tunnel junctions,^{27–29} lateral spin-valves^{22,23,25} and spin–orbit torque (SOT) memory devices.^{26,30–35} However, systematic experimental studies of magnetization dynamics, specifically their spin–orbit torque efficiencies, magnetization damping, and charge-to-spin conversion, remain sparse but are essential for realizing fast and energy-efficient spintronic technologies.

Here, we report multidirectional SOT magnetization dynamics using the above room temperature vdW magnet (Co_{0.15}Fe_{0.85})₅GeTe₂ (CFGF) in heterostructure with Pt. Using both spin-torque ferromagnetic resonance (ST-FMR) in the frequency range of 2–14 GHz and second (2nd) harmonic Hall measurement techniques, we observe a large in-plane spin Hall conductivity and a significant unconventional out-of-plane spin Hall conductivity. Further DFT calculations combined with Monte Carlo simulation show that proximity with Pt can introduce complex noncollinear spin textures in the

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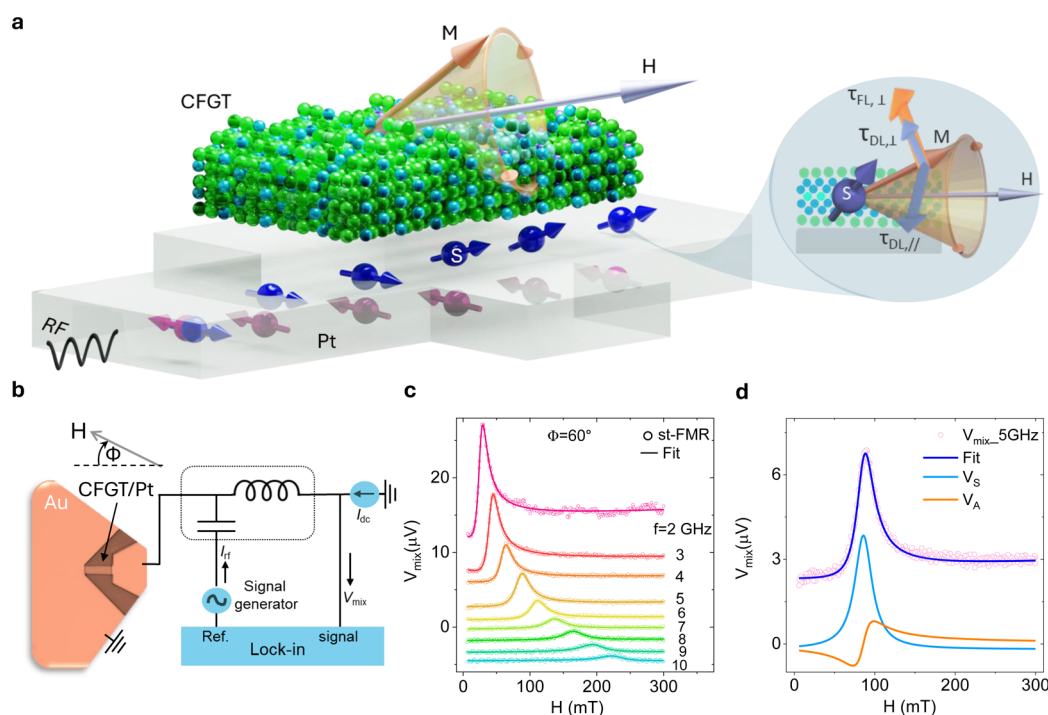


Figure 1. Magnetization dynamics measurements using CFGT/Pt heterostructure at room temperature. (a) Schematics of the CFGT/Pt heterostructure with magnetization M oscillation driven by SOTs and external B field. The inset shows the multidirectional spin current induced by conventional and unconventional SOT in the CFGT/Pt heterostructure. (b) Schematics of the spin-torque ferromagnetic resonance (ST-FMR) measurement setup with CFGT/Pt heterostructure device. (c) Representative ST-FMR data, showing V_{mix} as a function of the magnetic field at different radio frequencies in the range of $f = 2$ – 10 GHz at $\Phi = 60^\circ$. (d) ST-FMR signal at 5 GHz with the fitted symmetric (V_S) and antisymmetric (V_A) components. Measurements are performed on the CFGT(25 nm)/Pt(10 nm) ST-FMR device.

CFGT, which can be attributed to the strongly enhanced Dzyaloshinsky–Moriya interaction and weakened magnetic anisotropy energy with tilted magnetic easy axis. These results reveal crucial parameters governing efficient spin dynamics and unconventional SOT in vdW magnets for next-generation spintronic devices.

We were motivated to study magnetization dynamics in vdW magnet CFGT because of their above-room-temperature ferromagnetism with strong in-plane magnetic anisotropy.²² Here, we employed the spin–orbit torque (SOT) induced ST-FMR and 2nd harmonic Hall measurements to investigate the magnetization dynamics of the vdW magnet CFGT/Pt heterostructure (Figure 1a). For the ST-FMR device fabrication, the CFGT crystals were first exfoliated onto a Si/SiO₂ substrate inside a glovebox, followed by Pt deposition to create the CFGT/Pt heterostructure (see the Methods section). Subsequently, the CFGT/Pt heterostructure was patterned into rectangular microbar devices with an integrated microwave guide with Ti/Au contacts for the ST-FMR measurements (Figure 1b). The acquired measurements unveiled information about effective magnetization $\mu_0 M_{\text{eff}}$, effective Gilbert damping constant α , magnetic anisotropy H_k , and SOT efficiency at room temperature.

In ST-FMR measurements, an in-plane radio-frequency (RF) current I_{RF} is applied along the CFGT/Pt stripe, while an in-plane magnetic field H is applied at an angle Φ with respect to the I_{RF} . The spin–orbit coupling in Pt generates a spin current perpendicular to the direction of I_{RF} , which is injected into the adjacent CFGT layer and exerts a torque on the magnetization of CFGT, causing it to precess. The I_{RF} causes the anisotropic magnetoresistance (AMR) to oscillate and produces a DC mixing voltage V_{mix} which is then measured

using a lock-in amplifier. The representative ST-FMR signals V_{mix} for the CFGT/Pt device at room temperature are shown in Figure 1c. The obtained V_{mix} is fitted by eq 1,^{36–38}

$$V_{\text{mix}} = V_S F_S(H) + V_A F_A(H) \quad (1)$$

where $F_S(H) = \frac{\Delta H^2}{\Delta H^2 + (H - H_R)^2}$ and $F_A(H) = F_S(H) \left[\frac{(H - H_R)}{\Delta H} \right]$ are symmetric and antisymmetric Lorentzian functions, respectively. H , ΔH , and H_R are the applied external magnetic field, ferromagnetic resonance line width, and ferromagnetic resonance field, respectively. V_S and V_A are the amplitudes of symmetric and antisymmetric signals and are proportional to the current-induced in-plane torque and out-of-plane torque, respectively.³⁹ The obtained V_{mix} at 5 GHz is shown in Figure 1d, where symmetric and antisymmetric components are extracted and shown for reference. The extracted values of ΔH and H_R are used to obtain the effective magnetization $\mu_0 M_{\text{eff}}$ by fitting the frequency f versus H_R data to the Kittel equation^{38,40,41} $f = \left(\frac{\gamma}{2\pi} \right) \mu_0 \sqrt{(H_R - H_k)(H_R - H_k + M_{\text{eff}})}$ as given in Figure 2a. The Gilbert damping constant α is evaluated by fitting the ΔH versus f using $\Delta H = \Delta H_0 + \frac{2\pi\alpha f}{\gamma}$, as shown in Figure 2b. The $\mu_0 M_{\text{eff}} = 0.321 \pm 0.014$ T and $\alpha = 0.027 \pm 0.001$ are obtained for the CFGT/Pt device, as given in parts a and b of Figure 2, respectively (additional results at $\Phi = 240^\circ$ are presented in Supplementary Figure S1).

To estimate the effective damping-like SOT efficiency, the DC bias-dependent ST-FMR measurements are done on the ST-FMR device as well, and the representative data at different values of DC current (I_{dc}) are shown in Figure 2c,d. The resonance line width, ΔH at different I_{dc} is extracted, and

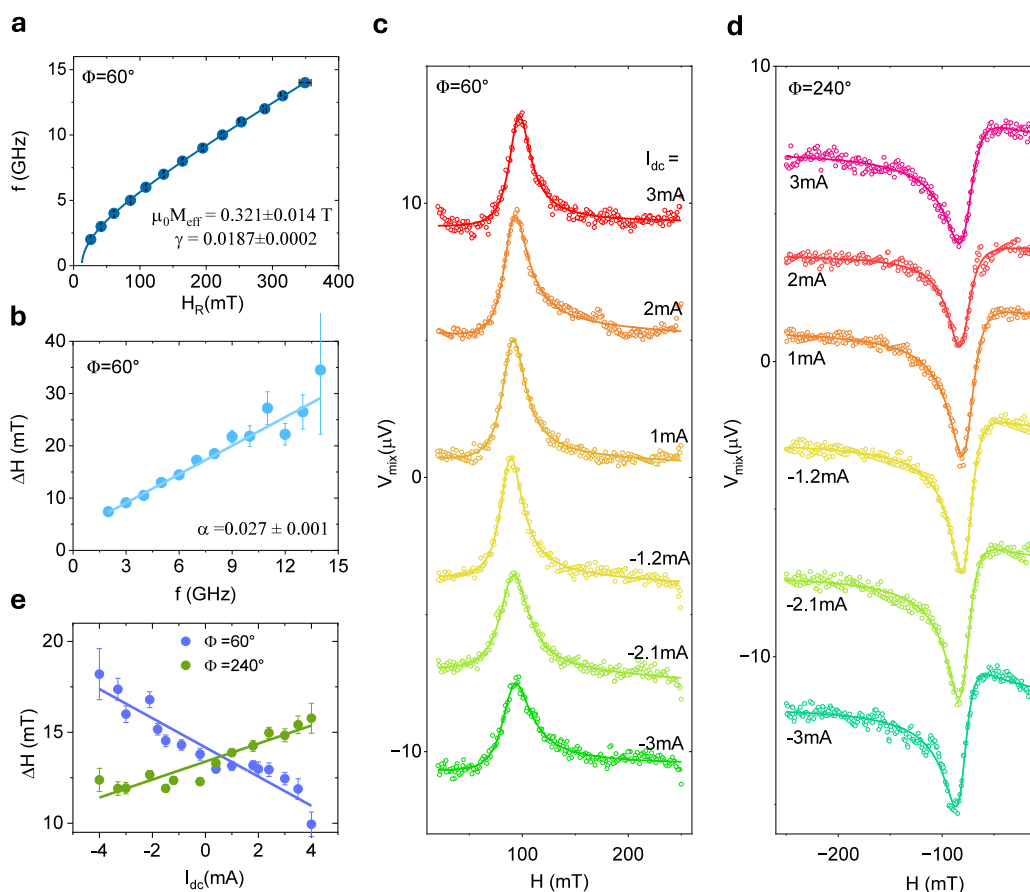


Figure 2. Evaluation of magnetization dynamic parameters and effective spin–orbit torque efficiency from DC bias-dependent ST-FMR measurements. (a) Fitting of resonant frequency f versus the extracted resonant field H_R . (b) Fitting of resonant extracted line width ΔH versus resonant frequency f (GHz). The error bars are calculated from the standard deviation of the signal background. (c and d) DC current tuned ST-FMR plots for positive/negative fields at a frequency of 5 GHz using different values of I_{dc} . Hollow dots denote experimental data while solid lines correspond to fitting performed using eq 1. (e) Line width versus DC current plot at 5 GHz for positive (blue line) and negative (dark green line) fields. The fitted slope of these lines is used to calculate the SOT efficiency of damping-like torque using eq 2. All the solid curves are the fitting results.

shown in Figure 2e. The strength of damping-like SOT efficiency is estimated using the slope of linearly fitted ΔH versus I_{dc} data,^{36–38}

$$\xi_{DL}^{eff} = \frac{2e}{\hbar} \frac{(H_R + 0.5M_{eff})\mu_0 M_S t_{CFGT}}{\sin \phi} \frac{\gamma}{2\pi f} \frac{\delta \Delta H}{\delta(I_{dc,Pt})} A_C \quad (2)$$

where Φ is the azimuthal angle between I_{dc} and H . M_{eff} , $\mu_0 M_S$, t_{CFGT} , and $\frac{\gamma}{2\pi}$ are the effective magnetization, saturation magnetization, thickness, and effective gyromagnetic ratio of the CFGT layer, respectively. e and $\hbar$ are the elementary charge and the reduced Planck's constant. $I_{dc,Pt}$ is the current in the Pt layer, and A_C is the cross-sectional area of the ST-FMR microbars. The effective spin Hall conductivity ($\sigma_{SHC} = \sigma_{cSDL}^{eff}$) values are evaluated to be 1.13×10^6 ($\hbar/2e$) ($\Omega \text{ m}$)^{−1} for CFGT/Pt heterostructure. Noticeably, the effective SOT efficiency value obtained from DC bias-dependent measurements might have unwanted contributions due to thermal effects, which could result in deviations in SOT efficiency compared to its intrinsic values.⁴²

To gain more insight into different SOT components, we also performed the in-plane angle dependence of the ST-FMR measurements, as shown in Figure 3a,b. The symmetric (V_S) and antisymmetric (V_A) components are extracted by fitting

the ST-FMR signals at different in-plane angles Φ with eq 1, as illustrated in Figure 3c,d. Traditionally, the relation of the symmetric and antisymmetric components as a function of the in-plane angle Φ can be expressed as follows:^{39,43}

$$V_S = S_{DL,Y} \sin(2\Phi) \cos(\Phi) \quad (3)$$

$$V_A = A_{FL,Y} \sin(2\Phi) \cos(\Phi) \quad (4)$$

$S_{DL,Y}$ and $A_{FL,Y}$ are the coefficients for the damping and field-like torques with spin polarizations along the y direction. However, we found that the antisymmetric component is not fitted properly with eq 4 (blue curve in Figure 3d). Then another term $A_{DL,Z} \sin(2\Phi)$ was added in eq 4, and we have

$$V_{AI} = A_{FL,Y} \sin(2\Phi) \cos(\Phi) + A_{DL,Z} \sin(2\Phi) \quad (5)$$

$A_{DL,Z}$ is the coefficient for the damping-like torque with spin polarizations along the z direction. The new formula (eq 5) shows a better fitting result (red curve in Figure 3d), which suggests the presence of an additional out-of-plane SOT component (see a more detailed analysis in Supplementary Note 1, Supplementary Figure S2, and Table S1). The in-plane ($\xi_{DL,j}^Y$) and out-of-plane ($\xi_{DL,j}^Z$) damping-like torque efficiencies per unit current density could be extracted using

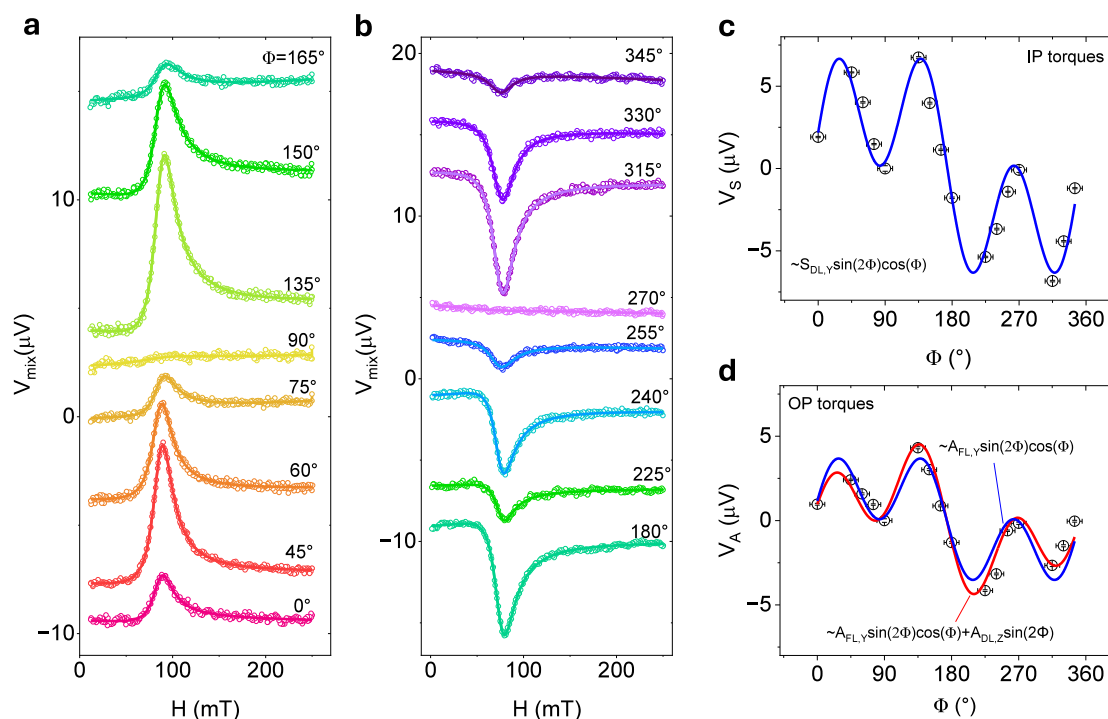


Figure 3. Unconventional spin-orbit torque in CFGT/Pt heterostructures from angular ST-FMR measurements. (a and b) Angular dependence of the ST-FMR signals at a frequency of 5 GHz. The open circles denote the output V_{mix} signal, and the solid lines represent the fitting by eq 1. (c and d) Angular dependence of the extracted symmetric component V_S and antisymmetric component V_A , respectively. Solid curves denote the fitting to the data using equations indicated in the figures.

$$\xi_{\text{DL},j}^Y = \frac{S_{\text{DL},Y}}{A_{\text{FL},Y}} \frac{e\mu_0 M_S t_{\text{Pt}} t_{\text{FM}}}{\hbar} \sqrt{1 + \frac{M_{\text{eff}}}{H_R}} \quad (6)$$

$$\xi_{\text{DL},j}^Z = \frac{A_{\text{DL},Z}}{A_{\text{FL},Y}} \frac{e\mu_0 M_S t_{\text{Pt}} t_{\text{FM}}}{\hbar} \quad (7)$$

The in-plane and out-of-plane damping-like torque efficiencies of $\theta_{\text{SH},Y} = 0.147 \pm 0.015$ and $\theta_{\text{SH},Z} = -0.013 \pm 0.005$ are obtained for spin polarization S_y and S_z , respectively. Correspondingly, the effective spin Hall conductivity components are evaluated to be $\sigma_{\text{SH},Y} = 3.68 \times 10^5 (\hbar/2e) (\Omega \text{ m})^{-1}$ and $\sigma_{\text{SH},Z} = -0.33 \times 10^5 (\hbar/2e) (\Omega \text{ m})^{-1}$, respectively, which are comparable with TaIrTe₄^{44,45} and PtTe₂/WTe₂⁴⁶ and larger than WTe₂.³⁹ Interestingly, such an unconventional out-of-plane damping-like torque component is observed in the CFGT/Pt heterostructure using ST-FMR measurements.

To further verify this unconventional torque, we performed second harmonic Hall measurements in microfabricated Hall bar devices (see Figure 4a and the Methods section). Specifically, the in-plane angle dependence harmonic Hall measurement is used to extract the effective spin-orbit field $H_{\text{DL(FL)}}$ along x -, y -, and z -axis directions and thermal contributions V_{th} with the 2nd harmonic Hall voltage

$$V_{xy}^{2\omega} = D_{\text{DL},Y} \cos(\Phi) + D_{\text{DL},Z} \cos(2\Phi) + F_{\text{FL},Y} \cos(2\Phi) \cos(\Phi) + F_{\text{FL},Z} \quad (8)$$

where

$$D_{\text{DL},Y} = -\frac{H_{\text{DL},Y} V_{\text{AHE}}}{2(H + H_k)} + V_{\text{th}} \quad (9)$$

$$D_{\text{DL},Z} = -\frac{H_{\text{DL},Z} V_{\text{PHE}}}{H} \quad (10)$$

$$F_{\text{FL},Y} = -\frac{H_{\text{FL},Y} V_{\text{PHE}}}{H} \quad (11)$$

Φ is the angle between the applied current I_{ac} and the magnetic field H . V_{AHE} and V_{PHE} are the AHE voltage and planar Hall (PHE) voltage, respectively. V_{th} is the thermal-related contribution, like anomalous Nernst and spin Seebeck effects, and H_k is the magnetic anisotropy field. By measuring the in-plane angle dependence of the first harmonic signal $V_{xy}^{1\omega}$ at a fixed large in-plane field, we can extract the V_{PHE} with $R_{xy}^{1\omega} = R_{\text{PHE}} \sin(2\Phi) + R_{\text{AHE}} \cos(\Phi) + c$ (as shown in Figure 4b). The 2nd harmonic signal $V_{xy}^{2\omega}$ in Figure 4c can be fitted with eq 8. The extracted components $F_{\text{FL},Y}$ and $D_{\text{DL},Y(Z)}$ as a function of the external field are plotted in Figure 4d,e. To further evaluate the effective fields induced by the SOT, eqs 9–11 are adopted, respectively. The bias dependence of the effective fields and thermal contribution are presented in (Figure 4f), which exhibit strong linear and parabolic relations with the bias current, respectively.

Because the damping-like torque is attributed to the longitudinal spin-orbit effective field H_{DL} , the spin-orbit efficiency can be calculated.^{47,48}

$$\xi_{\text{DL},Y(Z)} = T_{\text{ini}} \theta_{\text{SH}} = \frac{2e}{\hbar} \mu_0 M_S t_{\text{CFT}}^{\text{eff}} H_{\text{DL},Y(Z)} / J_{\text{ac}} \quad (12)$$

where J_{ac} is the current density of Pt, e is the electron charge, $\hbar$ is the reduced Planck constant, and $\mu_0 M_S$ and $t_{\text{CFT}}^{\text{eff}}$ are the saturation magnetization and the effective thickness of CFGT, respectively. By assuming the fully transparent interface, i.e., $T_{\text{ini}} = 1$, and effective saturation magnetization $\mu_0 M_S = \mu_0 M_{\text{eff}}$

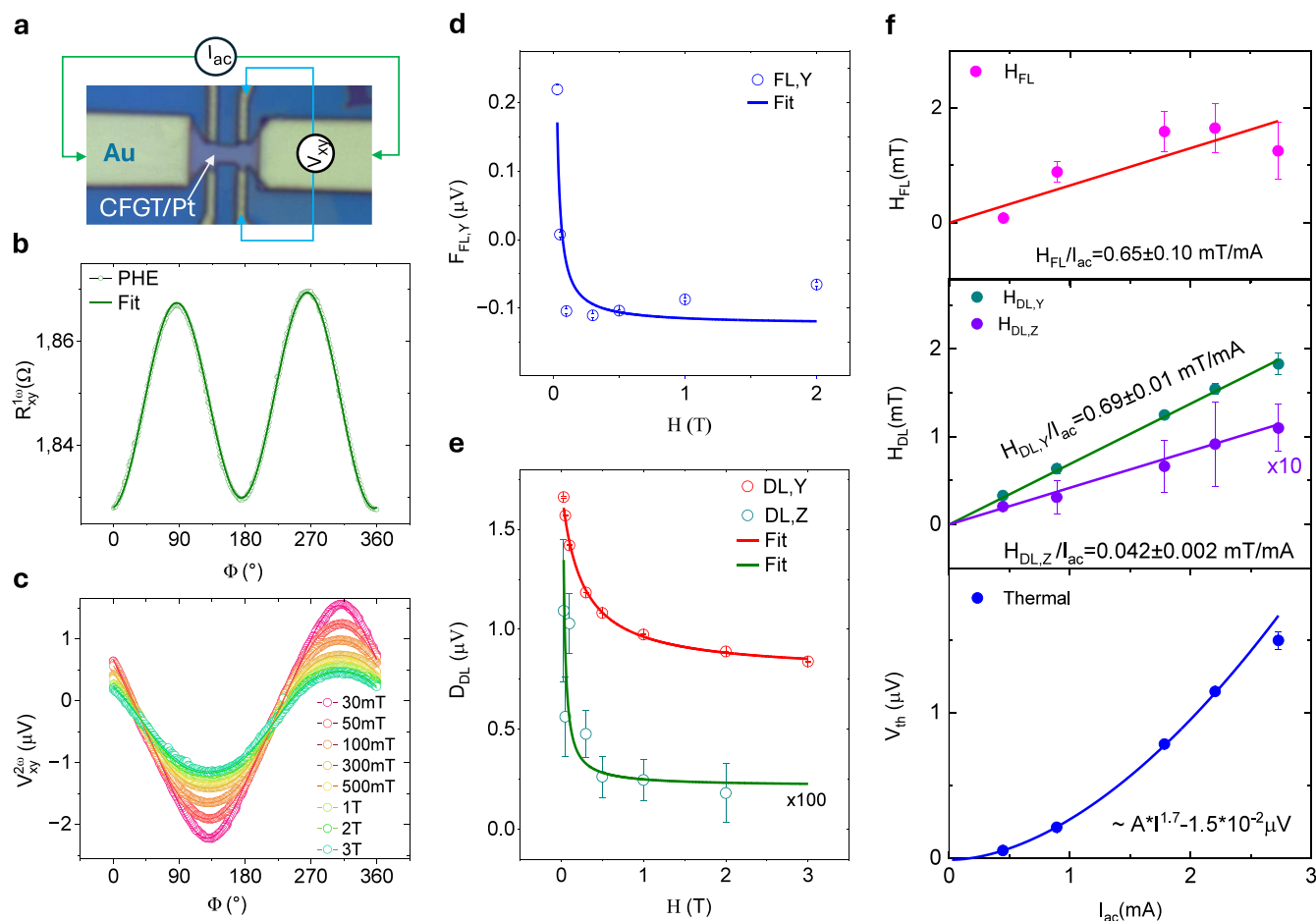


Figure 4. Harmonic measurements of the CFGT/Pt heterostructure at room temperature. (a) Schematics of the harmonic measurement setup with CFGT(15 nm)/Pt(10 nm) heterostructure Hall bar device. (b) First harmonic signal as a function of the in-plane angle with $H_{ip} = 1$ T and fitting result (solid curve). (c) In-plane angle dependence of the 2nd harmonic signals with different fields and corresponding fitting (solid) curves at $I_{ac} = 1.78$ mA. (d and e) Extracted effective H_{FL} and D_{DL} components as a function of the external field with fitting results. (f) Extracted effective field H_{FL} , H_{DL} , and thermal contribution as a function of the bias currents. The solid curves are the linear and nonlinear fitting results.

we obtain spin Hall angle $\theta_{SH,Y} = 0.157 \pm 0.002$ and $\sigma_{SH,Y} = (3.93 \pm 0.05) \times 10^5 (\hbar/2e) (\Omega \text{ m})^{-1}$ and $\theta_{SH,Z} = 0.010 \pm 0.001$ and $\sigma_{SH,Z} = (0.23 \pm 0.03) \times 10^5 (\hbar/2e) (\Omega \text{ m})^{-1}$, which are comparable to the results obtained from ST-FMR experiments.

Our study of mechanisms governing magnetization dynamics and SOT in vdW magnet CFGT with in-plane magnetic anisotropy at room temperature uncovers their unique and unconventional spin–orbit torque behavior. Both ST-FMR and second harmonic Hall measurements show that the SOT efficiency of CFGT/Pt along S_y polarization is comparable to that of the other vdW magnets in a heterostructure with Pt.^{26,31–34,49} However, in addition, a large unconventional SOT component S_z is also observed. Our control experiments of a single CFGT nanolayer with second harmonic Hall measurements prove that the self-torque from the single CFGT nanolayers can be ruled out (see detailed results in Supplementary Note 2 and Supplementary Figure S3). Furthermore, Pt is a well-studied SOT material with a dominant S_y component, so any bulk material-related unconventional SOT effects can be ruled out. Furthermore, our previous study on CFGT/graphene²² suggests that the in-plane magnetic anisotropy of CFGT is independent of thickness variation. Therefore, the unconventional spin component should not originate from bulk CFGT, but rather

from an interface-driven phenomenon. To confirm this, our DFT calculations combined with Monte Carlo simulation show that proximity with Pt introduces complex noncollinear spin textures in the CFGT (Figure S5a,b), which can be attributed to the strongly enhanced Dzyaloshinsky–Moriya interaction (Supplementary Figures S5 and S6), and weakened magnetic anisotropy energy with tilted magnetic easy axis ($\sim 16^\circ$; Figure S5c and Supplementary Figure S4). This is in contrast to the pristine CFGT, which exhibits in-plane magnetic anisotropy, consistent with our previous experimental results (see a more detailed discussion in Supplementary Note 3). These observations suggest that the reported unconventional torque is not specific to a particular thickness or a single surface but is instead a robust feature of the CFGT/Pt interface.

Although such interface-related spin reorientation phenomena in conventional ferromagnetic materials heterostructures have been reported before, the origin remains elusive.^{50–52} Here, we show that the modified magnetic anisotropy and interfacial DMI at the CFGT/Pt interface can induce the spin reorientation toward an unconventional spin polarization, in addition to the traditional spin Hall effect-induced counterpart (Figure S5c, bottom panel). This largely expands the library for multiple spin polarization mechanisms (Figure S5d), including (1) lower crystal symmetry related SHE (such as WTe_2 ,^{39,53,54}

for ST-FMR measurements and Hall bars for harmonic measurements using the Ar ion milling with e-beam resist as the etching mask.

Anisotropic Magnetoresistance Measurements

In-plane angular dependence, anisotropic magnetoresistance (AMR) measurements are performed on microbars using a rotatable projected vector field magnet with a fixed applied magnetic field of 1 T and applied DC current of 0.5 mA. The resistance of the devices was measured while rotating the magnetic field in the plane.

Spin Torque Ferromagnetic Resonance (ST-FMR) Measurements

ST-FMR measurements are performed at room temperature on the microbar devices to estimate the spin-orbit torque (SOT) efficiency and other magnetodynamical parameters. The measurements for effective SOT analysis are performed with a fixed in-plane angle $\phi = 60^\circ$, the radio-frequency (RF) current modulated at 98.76 Hz is applied to the device through a high-frequency bias-T at a fixed frequency (ranging from 3 to 14 GHz) with an input RF power $P = 4$ dBm. The in-plane angle ϕ dependence ST-FMR measurements are performed using a rotatable projected field magnet with $\phi = 0-360^\circ$ (step of 15°) at a fixed frequency.

2nd Harmonic Hall measurements

The 2nd harmonic measurements in the high magnetic field range were carried out in the Quantum Design DynaCool PPMS system with an external electronic connection to Lockin SR830 to measure the first and second harmonic voltages with fixed frequency $\omega = 213.34$ Hz.

■ ASSOCIATED CONTENT

Data Availability Statement

The data supporting the findings of this study are available from the corresponding authors upon reasonable request.

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.6c02494>.

Analysis of ST-FMR results (Supplementary Note 1), control experiments for self-torque in CFGT (Supplementary Note 2), magnetic dynamic parameters of this work at room temperature (Supplementary Table S1), and DFT supplemental results (Supplementary Note 3) (PDF)

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Author Contributions

^{††}B.Z. and L.B. contributed equally to this work. B.Z. and L.B. fabricated and measured the devices with the help of P.N.S. and R.N.. S.E., J.R., and B.S. performed theoretical calculations and analysis. B.Z., L.B., J.Å., and S.P.D. analyzed and interpreted the experimental data and wrote the manuscript with comments from all of the authors. S.P.D. coordinated and supervised the project.

Notes

The authors declare no competing financial interest.

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