Observation of Strong Bulk Damping-Like Spin-Orbit Torque in Chemically Disordered Ferromagnetic Single Layers

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Strong damping-like spin-orbit torque (\( \tau_{DL} \)) has great potential for enabling ultrafast energy-efficient magnetic memories, oscillators, and logic. So far, the reported \( \tau_{DL} \) exerted on a thin-film magnet must result from an externally generated spin current or from an internal non-equilibrium spin polarization in non-centrosymmetric GaMnAs single crystals. Here, for the first time a very strong, unexpected \( \tau_{DL} \) is demonstrated from current flow within ferromagnetic single layers of chemically disordered, face-centered-cubic CoPt. It is established here that the novel \( \tau_{DL} \) is a bulk effect, with the strength per unit current density increasing monotonically with the CoPt thickness, and is insensitive to the presence or absence of spin sinks at the CoPt surfaces. This \( \tau_{DL} \) most likely arises from a net transverse spin polarization associated with a strong spin Hall effect, while there is no detectable long-range asymmetry in the material. These results broaden the scope of spin-orbitronics and provide a novel avenue for developing single-layer-based spin-torque memory, oscillator, and logic technologies.

1. Introduction

Currently, exerting a nonzero damping-like spin-orbit torque (\( \tau_{DL} \)) on a ferromagnet (FM) layer requires an external transversely polarized spin current from the bulk spin Hall effect (SHH)\(^{[1-9]}\) of an adjacent spin-orbit layer, an interfacial spin-orbit effect at magnetic interfaces\(^{[8,10-11]}\) or the anomalous Hall effect (AHE)\(^{[13]}\) of an adjacent FM layer. Very recently, the generation of spin-orbit torques (SOTs) via current flow inside FMs has become an emerging focus of spin-orbitronics. The FM layer inside a magnetic interfaces, or the anomalous Hall effect (AHE)\(^{[13]}\) of an adjacent spin-orbit layer, an interfacial spin-orbit effect at magnetic interfaces, or the anomalous Hall effect (AHE)\(^{[13]}\) of an adjacent FM layer. Very recently, the generation of spin-orbit torques (SOTs) via current flow inside FMs has become an emerging focus of spin-orbitronics. The FM layer inside a

\[ H_{DL(FL)} = \frac{2eH_{DL(FL)} M_s}{\mu_0} \]

(1)

where \( H_{DL(FL)} \) are damping-like (field-like) SOT effective field (see Section S2, Supporting Information), \( h \) is the reduced Planck’s constant, \( \mu_0 \) the permeability, \( M_s \) the saturation magnetization of the spin current detector, and \( j \) the charge current density of the spin current generator.

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As plotted in Figure 1b, when $t_{\text{CoPt}}$ exceeds 4 nm a nonzero $\xi_{\text{DL}}$ emerges and increases monotonically with $t_{\text{CoPt}}$. This $\xi_{\text{DL}}$ behavior is not consistent with it arising from an interfacial SOT since then $\xi_{\text{DL}}$ would be approximately independent of $t_{\text{CoPt}}$. In Figure 1c, we plot the efficiency of $\tau_{\text{DL}}$ per thickness, $\xi_{\text{DL}}/t_{\text{CoPt}}$. This indicates that $\tau_{\text{DL}}$ has the characteristic of a bulk effect that has an onset at $\approx$4 nm and then increases in strength until it reaches its full bulk intensity at $\approx$16 nm. We have reaffirmed this unique thickness dependence of $\tau_{\text{DL}}$ with separate spin-torque ferromagnetic resonance (ST-FMR, Figure 1d) measurements. Figure 1e shows a representative ST-FMR spectrum for the 24 nm CoPt film, which shows a strong symmetric ($S$) component due to $\tau_{\text{DL}}$. As discussed in detail in Sections S3 and S4 of the Supporting Information, for a given rf current sourced into a magnetic microstrip, we have

$$\frac{\xi_{\text{DL}}}{t_{\text{CoPt}}} \approx \frac{S\sqrt{J_{\text{CoPt}}/C_{\text{MR}}}}{\chi}$$

(2)

where $C_{\text{MR}}$ is the longitudinal magnetoresistance ratio and $\chi$ is a parameter that accounts for the reflection of rf power due to the impedance mismatch of the device and the rf circuits. The measured values of $S\sqrt{J_{\text{CoPt}}/C_{\text{MR}}}$ (10 GHz and 15 dBm) for the CoPt films (Figure 1f) exhibit a $t_{\text{CoPt}}$ dependence that is well consistent with the $\xi_{\text{DL}}/t_{\text{CoPt}}$ results obtained from the harmonic response measurements (Figure 1c).

We find that $\tau_{\text{DL}}$ in these CoPt layers is insensitive to the details of the CoPt interfaces, whether those interfaces are formed with neighboring insulating materials (SiO$_2$ and MgO), or with metallic layers that are either spin reflectors (Hf) or spin sinks (Tb and Ru). As summarized in Table 1, $\xi_{\text{DL}}$ is $-0.12$ for all the symmetric stacks MgO/CoPt 16/MgO, Hf/CoPt 16/Hf, Tb/CoPt 16/Tb, and Ru/CoPt 16/Ru, and also for the asymmetric SiO$_2$/CoPt 16/MgO. Both the unique thickness dependence and the robust insensitivity to the nature of CoPt interface make the observed $\tau_{\text{DL}}$ distinctly different from any interfacial SOTs. [12, 15]
that we observe requires the generation of an $\xi_j$ as being due to the reflection becomes detectable for $\xi_j$ as determined from in-plane harmonic response measurement. CoPt denotes Co$_{0.2}$Pt$_{0.8}$. The Co concentration $x$ of the Co$_{0.2}$Pt$_{1-x}$ ($x = 0.75 \rightarrow 0.25$) layer is varied continuously from 0.75 at the bottom to 0.25 at the top, while for Co$_{0.25}$Pt$_{1-x}$ ($x = 0.25 \rightarrow 0.75$) $x$ is varied continuously from 0.25 at the bottom to 0.75 at the top. This was achieved by subsequently varying the composition of each 0.2 nm subatomic layer during the growth of the Co$_{0.2}$Pt$_{1-x}$ layer.

The harmonic measurements also determine a field-like torque which, in contrast, is sensitive to the nature of the CoPt interfaces. As plotted in Figure 1b, for a series of SiO$_2$/CoPt/MgO samples, $\xi_{DL}$ becomes detectable for $t_{CoPt} = 4$ nm, and then is essentially constant for all $t_{CoPt}$ Values beyond that point. As shown in Table 1, when spin sinks (Tb and Ru) are placed at the two surfaces of the CoPt, $\xi_{FL}$ is found to be negligible; while it has close to the same value when the interfaces are formed by different spin reflectors (MgO, SiO$_2$, Hf). We interpret this $\xi_{FL}$ as being due to the reflection, with some spin rotation, of the spin current that is generated in the bulk of the CoPt due to the bias current.

### 2.2. Exceptionally Strong Spin Hall Effect

The bulk $\xi_{DL}$ that we observe requires the generation of an internal spin current (spin accumulation) within the CoPt, and one that has a net spin polarization when averaged over the thickness of the material. Spin currents can be generated by either the SHE, the AHE, or the PHE. Here the AHE and the PHE can be ruled out because the in-plane angle-dependent harmonic response technique we employed is sensitive only to transversely polarized spins, as in the SHE, and cannot capture torques due to spin accumulations whose polarization rotates with the external field as in the AHE and PHE (Section S5, Supporting Information).

A strong SHE in the CoPt should of course exert a substantial torque on an adjacent FM layer, as we found to be the case. For multilayer structures that include two FM layers which are both freely rotatable by external fields, the harmonic response analysis is quite challenging due to the entanglement of the Hall voltage responses from the two FMs. Therefore, we used the ST-FMR technique to examine the spin current emission and resultant SOT in multilayers of CoPt 24/Ti 0.8/FeCoB (FeCoB = Fe$_{0.8}$Co$_{0.2}$B$_{0.2}$). The FeCoB, whose thickness varied from 1.4 to 4 nm, was chosen as the detector of the spin current emitted from the top surface of the CoPt layer because the FeCoB dominates the FMR spectrum of the trilayer stack (Figure S3a, Supporting Information), while the Ti layer was used to suppress the exchange coupling between the CoPt and the FeCoB layers. If we define the apparent FMR spin-torque efficiency ($\xi_{FL}^{FM}$) from the ratio of the symmetric (S) and antisymmetric (A) components of the magnetoresistance response of the ST-FMR (Section S3, Supporting Information), the efficiency of the damping-like torque acting on the FeCoB by the external spin current from the CoPt ($\xi_{DL,ext}^{S}$) in the CoPt/Ti/FeCoB sample is determined as the inverse intercept in the linear fit of $1/\xi_{FL}^{FM}$ versus $t_{FeCoB}$. As shown in Figure 2a, we obtained $\xi_{DL,ext}^{S} = -0.12 \pm 0.01$ for a CoPt 24/Ti 0.8/FeCoB multilayer, which is twice the amplitude that we measured for a baseline Pt 4/FeCoB bilayer series ($\xi_{DL,ext}^{S} = 0.06$). We confirmed that the interfaces and thin layers of Ti 0.8/FeCoB bilayer have a negligible torque contribution ($\xi_{DL,ext}^{S} = 0.001$, Figure 2a) to the overall SOT through ST-FMR on the FeCoB. We also find no sin2$\varphi$ term in the A component of the ST-FMR spectrum.

**Table 1.** Effects of interfaces and vertical composition gradient. Damping-like (field-like) spin-orbit torque efficiency per unit current density $\xi_{DL,FL}^{j}$ as determined from in-plane harmonic response measurements. CoPt denotes Co$_{0.2}$Pt$_{0.8}$. The Co concentration $x$ of the Co$_{0.2}$Pt$_{1-x}$ ($x = 0.75 \rightarrow 0.25$) layer is varied continuously from 0.75 at the bottom to 0.25 at the top, while for Co$_{0.25}$Pt$_{1-x}$ ($x = 0.25 \rightarrow 0.75$) $x$ is varied continuously from 0.25 at the bottom to 0.75 at the top. This was achieved by subsequently varying the composition of each 0.2 nm subatomic layer during the growth of the Co$_{0.2}$Pt$_{1-x}$ layer.

| Structure          | $\xi_{DL}^{j}$ | $\xi_{FL}^{j}$ |
|--------------------|---------------|---------------|
| SiO$_2$/CoPt 16/MgO 2 | $-0.125 \pm 0.008$ | $0.018 \pm 0.002$ |
| MgO 2/CoPt 16/MgO 2  | $-0.130 \pm 0.004$ | $0.014 \pm 0.002$ |
| Hf 2.5/CoPt 16/Hf 2.5 | $-0.115 \pm 0.004$ | $0.019 \pm 0.002$ |
| Tb 2.5/CoPt 16/Tb 2.5 | $-0.119 \pm 0.013$ | $-0.002 \pm 0.002$ |
| Ru 1/CoPt 16/Ru 1    | $-0.124 \pm 0.002$ | $0.003 \pm 0.002$ |
| SiO$_2$/CoPt$_{0.75}$  | $-0.031 \pm 0.002$ | $0.057 \pm 0.001$ |
| SiO$_2$/CoPt$_{0.25}$  | $-0.023 \pm 0.004$ | $-0.043 \pm 0.001$ |

![Figure 2.](image-url)
of the CoPt/Ti/FeCoB (Figure S3b, Supporting Information), revealing minimal perpendicularly polarized spin emission\cite{24} and spin-orbit precession effect at the CoPt/Ti interface.\cite{32} We note that $\xi_{DL,\text{ext}} = -0.12$ represents only a lower bound for the internal value of $\theta_{SH}$ for the CoPt because of the cumulative effects of spin attenuation in Ti, interfacial spin backflow,\cite{25,26} and spin memory loss\cite{21} on the ST-FMR measurement. The spin memory loss is likely to be substantial as indicated by the significant interfacial magnetic energy density of $1.4$ erg cm$^{-2}$ for the Ti/FeCoB interface\cite{21} ($\approx 0.4$ erg cm$^{-2}$ for the Pt/FeCoB interface, Section S6, Supporting Information).

As shown in Figure 2b, the spin Hall spin current emitted from a 4 nm CoPt layer can switch a perpendicularly FeCoB layer (with a layer thickness of 1 nm, a perpendicular coercivity of 230 Oe, and an effective perpendicular anisotropic field from a 4 nm CoPt layer) when a control sample of 16 nm Co$_{1-x}$Pt$_x$ layer with $x$ continuously varying from 0.75 to 0.25 ($\xi_{DL} = -0.031$), compared with that of the uniform 16 nm CoPt layer ($x = 0.5$), and similarly for a 16 nm Co$_{1-x}$Pt$_x$ layer with the reversed composition gradient direction ($\xi_{DL} = 0.025$). This indicates that neither the magnitude nor the sign of the observed $\tau_{DL}$ in the CoPt is caused by a composition gradient. The resistivity ($\rho_{xx}$) of the CoPt layer remains constant at $\approx 70$ $\mu$Ohm cm as a function of $t_{CoPt}$ until $t_{CoPt}$ is reduced to 2 nm where the interfacial scattering begins to contribute significantly to $\rho_{xx}$.

We do find a significant vertical compressive strain in the CoPt layer as revealed by the fcc (111) lattice plane spacing as measured by X-ray diffraction which is smaller than the bulk value\cite{27} of 2.214 Å (Figure 3g). However, the strain is essentially invariant as a function of $t_{CoPt}$. Averaged electron diffraction patterns from the top and bottom regions of a sufficiently thick CoPt indicate no difference in the strain within the experimental resolution (Section S8, Supporting Information).

Finally, there is no evidence of any significant role of thermal gradient in our samples. $H_{DL}$ for each CoPt layer scales linearly with the input voltage ($V_{in}$) and thus electric field ($E$) even when $V_{in}$ is increased from 1 to 5 V, which results in anomalous Nernst voltage ($V_{ANE}$) and thus the thermal gradient ($VT$) being enhanced by a factor of 23 (Figure 3h,i). This linear behavior excludes any role of the vertical thermal gradient in the generation of the non-equilibrium spin accumulation. In fact, the thermal gradient during the harmonic measurement ($E = 1.67$ kV cm$^{-1}$, $V_{in} = 1$ V) remains too small ($\leq 0.4$ mK nm$^{-1}$, Figure 3j and Section S9, Supporting Information) to yield a significant variation of the magnetic or structural properties across the film thickness.

Although there is no theory predicting the novel bulk $\tau_{DL}$ reported here, our experimental observations imply that, in magnetic materials with a strong SOC effect (SHE), a strong bulk damping-like torque can be generated without the need of an obvious long-range symmetry breaking. We speculate that the non-equilibrium spin polarization in CoPt could be associated with a “hidden” local inversion asymmetry (e.g., short-range order or local SOC effect) that can cause asymmetric spin accumulation, spin relaxation, or exchange interaction. The bulk $\tau_{DL}$ of Co$_{1-x}$Pt$_x$ (x = 0.75 → 0.25) and Co$_{1-x}$Pt$_x$ (x = 0.25 → 0.75) is much smaller than that of CoPt (Table 1), suggesting that such bulk $\tau_{DL}$ of a uniform Co$_{1-x}$Pt$_x$ layer may be tuned by its composition, for example, quite strong at $x = 0.5$ and much weaker at Pt-rich and/or Co-rich region. Our findings of the strong SHE (with the spin current independent of its magnetization orientation and transversely polarized) and bulk $\tau_{DL}$ in the metallic ferromagnetic single layers should fundamentally broaden the scope of spin-orbitronics and stimulate future experimental and theoretical efforts on intriguing SOC effects (generation, relaxation,\cite{28} and

2.3. Absence of Detectable Long-Range Asymmetry

We have found no evidence for a long-range symmetry breaking in our CoPt films. X-ray diffraction (Figure 3a) and electron diffraction patterns (Figure 3b and Figure S7, Supporting Information) reveal that these CoPt layers consist of chemically disordered fcc (A1) polycrystalline grains, which are globally centrosymmetric. There is also no indication of a vertical gradient in grain size, composition, electron scattering, strain, or temperature in the CoPt samples. As revealed by the cross-sectional scanning transmission electron microscopy (STEM) images in Figure 3b,c, the CoPt layers have good homogeneity across the thickness, without any statistically significant vertical gradient in the dimension of the columnar crystalline grains. There is also no significant vertical composition gradient as indicated by the fairly constant electron energy loss spectrum (EELS) intensities of Co and Pt in the depth profile (Figure 3d) and by the independence of $M_s$ ($\approx 790$ emu cm$^{-3}$) on $t_{CoPt}$ (Figure 3e). Indeed, we find that a strong artificial vertical composition gradient degrades rather than enhances $\tau_{DL}$ in this system. As summarized in Table 1, $\xi_{DL}$ is four times smaller for a control sample of 16 nm Co$_{1-x}$Pt$_x$ layer with $x$ continuously varying from 0.75 to 0.25 ($\xi_{DL} = -0.031$), compared with that of the uniform 16 nm CoPt layer ($x = 0.5$), and similarly for a 16 nm Co$_{1-x}$Pt$_x$ layer with the reversed composition gradient direction ($\xi_{DL} = 0.025$). This indicates that neither the magnitude nor the sign of the observed $\tau_{DL}$ in the CoPt is caused by a composition gradient. The resistivity ($\rho_{xx}$) of the CoPt layer remains constant at $\approx 70$ $\mu$Ohm cm as a function of $t_{CoPt}$ until $t_{CoPt}$ is reduced to 2 nm where the interfacial scattering begins to contribute significantly to $\rho_{xx}$.

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transport of spin current and local inversion asymmetry) in FMs. The novel bulk $\tau_{DL}$ we demonstrate here may also provide a possible interpretation for the absence of $\tau_{DL}$ in thin $L_1_0$ FePt single crystal,[28] the presence of self-induced switching in very thick $L_1_0$-FePt single crystal,[29] and the presence of a nonzero $\tau_{DL}$ in Co/Pd multilayer nanowires[30] and in a 15 nm-thick nearly compensated ferrimagnetic Tb-Co film.[31] However, the existing models involving a long-range composition gradient[29,32,33] are clearly irrelevant to the novel bulk $\tau_{DL}$ we establish in this work.

3. Conclusion

We have demonstrated the first experimental observation of an unexpected, strong $\tau_{DL}$ from current flow within uniform ferromagnetic single layers of chemically disordered, face-centered-cubic CoPt. We identify that the novel $\tau_{DL}$ is a bulk effect, with the strength per unit current density increasing monotonically with the FM thickness, and is insensitive to the presence or absence of spin sinks at the CoPt surfaces. This $\tau_{DL}$ most likely arises from a net transverse spin polarization associated with a strong SHE.
while there is no detectable long-range asymmetry in the material. These results provide a novel avenue for generating strong damping-like spin-orbit torque ($\tau_{SO}$) in disordered magnetic single layers. This work also establishes the technological possibility of self-driven single-layer spintronics devices (e.g., memory, oscillator, and logic) with simplified architecture and improved scalability.

4. Experimental Section

Sample Preparation: The samples were deposited at room temperature by sputtering onto oxidized Si substrates with an argon pressure of 2 mTorr and a base pressure of $\approx 10^{-9}$ Torr. Each sample was capped by a MgO 2 nm/Ta 1.5 nm bilayer that was fully oxidized upon exposure to the atmosphere. The samples were patterned by photolithography and ion milling into $5 \times 60 \mu m^2$ Hall bars and $10 \times 20 \mu m^2$ microstrips, followed by deposition of 5 nm Ti and 150 nm Pt as electrical contacts for harmonic response measurements and for ST-FMR measurements.

Structural Characterizations: A Rigaku Smartlab diffractometer was used for X-ray diffraction measurements. A FEI Strata 400 STEM focused ion beam system was used to prepare the sample for STEM. An electron microscope pixel array detector in FEI F20 STEM (200 kV) was used to collect the 4D STEM diffraction data. A FEI/Thermo Fisher Titan Themis STEM system at 300 kV was used for the STEM imaging and EELS measurements.

Magnetic and Electrical Measurements: The saturation magnetization of each sample was measured at 300 K with a vibrating sample magnetometer (sensitivity $\approx 10^{-7}$ emu) in a Quantum Design physical property measurement system. Anomalous Hall voltage and effective anisotropic field were measured electrically using PPMS. During the harmonic response measurements, a Signal Recovery DSP lock-in amplifier (Model 7625) was used to source a sinusoidal voltage onto the Hall bars and to detect the first and second harmonic Hall voltage responses. For the ST-FMR measurements, an rf signal generator and a Signal Recovery DSP lock-in amplifier (Model 7625) were used and an in-plane magnetic field was swept at 45° with respect to the magnetic microstrip. An electromagnet with a maximum in-plane field of 3.5 kOe was used during the spin-torque measurements. For the switching measurements, a Signal Recovery DSP lock-in amplifier (Model 7625) was used to source a sinusoidal voltage of 0.1 V onto the Hall bars and to detect the first harmonic response Hall voltage, and a Yokogawa 7651 was used to source direct current to electrically drive the magnetization switching. All the measurements were performed at room temperature.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

ferromagnetic resonances, inversion symmetry breaking, spin currents, spin-orbit torques

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