Observation of unconventional spin-polarization induced spin–orbit torque in L12-ordered antiferromagnetic Mn3Pt thin films

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Many recently discovered phenomena associated with the topology of magnetism make antiferromagnets (AFMs) with topologically nontrivial spin structures a rich playground for the investigation of unique topological behaviors, as well as promising candidates for energy-efficient microelectronic applications.1–3 The anomalous transport properties of electrons and magnons that are affected by spin structure-induced non-vanishing Berry curvatures,4,5 or chirality-related Dzyaloshinskii–Moriya interaction,6 known as the anomalous Hall effect (AHE) and magnetic skyrmions, respectively, make it possible to control the charge or magnon transport in AFMs efficiently and stably. AFMs have also attracted interest because it is possible to not only use them as efficient spin current sources7–9 but also to modulate their spin structures electrically,10–12 which leads to great application potential. More recently, the discovery of magnetic spin Hall effect (MSHE)13–15 in non-collinear AFMs with topological triangular spin structures and the observation of Berry curvature–induced spin–orbit torque (SOT)16,17 in collinear AFM with a parallel spin structure have been highlighted, both of which provide the opportunity to manipulate the spin current and generate spin polarization by tuning the spin structures of AFMs.

Although the charge-to-spin conversion phenomenon of MSHE is similar to that of the intrinsic spin Hall effect (SHE), the time-reversal symmetry of MSHE is odd and of extrinsic origin, which makes it a reactive counterpart of SHE and demonstrative of a dissipative nature.15–18 The odd time-reversal symmetry can be regarded as the result of magnetic-order parameter reversal, which indicates that the spin polarization generated by MSHE is dictated by the magnetic order, thereby allowing an in-plane component in the form of a scattering plane created by the charge current and spin current. In other words, in current-induced SOT, MSHE is expected to generate an additional NO-\(y\)-polarization, i.e., \(x\)-polarization and \(z\)-polarization when the charge current is flowing along the in-plane direction (\(x\) direction), whereas only \(y\)-polarization is allowed to exist for SHE due to the restriction of symmetry.13,16,19 Analogous to the \(y\)-polarization generated by SHE, which will give rise to a conventional damping-like (DL) torque in an adjacent ferromagnet (FM) of the form \(m \times (m \times y)\), where \(m\) denotes the magnetization vector, it is expected that the unconventional DL torque of \(x\)-polarization and \(z\)-polarization generated by MSHE will take the forms of \(m \times (m \times x)\) and \(m \times (m \times z)\), respectively. This means that MSHE-induced SOT can be investigated quantitatively under the condition of ferromagnetic resonance. In doing so, we hope to contribute to a deeper understanding of unconventional SOT induced by MSHE.15–18

The non-collinear AFM \(L1_2\) Mn3Pt that we focus on in this paper is an antiferromagnetic material with a topologically nontrivial spin structure.2,20 The crystal structure of Mn3Pt is \(L1_2\) (space group: \(Pm\overline{3}m\)), and the Néel temperature is around 475 K. At room temperature, Mn3Pt has a 120° triangular \(\Gamma_4g\) spin structure lying in a (111) kagome plane where the spin configuration is all-in or all-out, which is similar to Mn3Ir [see Fig. 1(a)].21 Mn3Pt has so far been experimentally demonstrated to exhibit a relatively large AHE22 and spin Hall angle.7 In addition, an intermetallic compound with the same \(\Gamma_4g\) spin structure also shows facet-dependence of the spin Hall conductivity8 and out-of-plane (\(z\) direction) spin accumulation.23 In this work, we use a spin-torque ferromagnetic resonance (ST-FMR) technique to explore the unconventional SOT in an \(L1_2\)-ordered epitaxial Mn3Pt/Ni80Fe20 (Py) heterostructure. We observed the NO-\(y\) spin-polarization-induced SOT in \(L1_2\)-ordered Mn3Pt, which is absent in poly-Mn3Pt whose lattice structure as well as spin structure are regarded as disordered. Such an SOT can be associated with the contribution from the MSHE.

Mn3Pt thin films with the thickness of 15 nm were grown by magnetron sputtering on a MgO (001) and a thermally oxidized Si substrate with the substrate temperature \(T_s\) of 450 °C and RT, respectively. Subsequently, a thin layer of MgAl2O4 (2.5 nm) was deposited at RT to prevent oxidation. The out-of-plane X-ray diffraction patterns for Mn3Pt fabri-

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deposited on MgO forms a well-ordered L12 structure, while the lattice structure of Mn3Pt deposited on Si is disordered. Therefore, we set Mn3Pt deposited on Si substrate as a control sample. In addition, we also note that the aforementioned Γ4g spin structure only exists in the L12 structure. Next, to determine whether L12 Mn3Pt had a triangular spin structure, we characterized the magnetic and magneto-transport properties with magnetic hysteresis curves and performed Hall measurements for the Mn3Pt single film in an out-of-plane applied field. To examine the magnetic properties, we compared the hysteresis curves of L12-ordered Mn3Pt and disordered Mn3Pt at 300 K, as shown in Fig. 1(c). A weak but nonzero net magnetization M (about 10 emu cm−3) was detected along the (001) direction of the L12 Mn3Pt film due to triangle spin canting, which is consistent with previous theoretical and experimental works. The magnitude of M we measured is also similar to that for L12-ordered Mn3Ir. In contrast, such a small magnetization could not be observed for the disordered control sample. For transport properties, we patterned the films by photo-lithography and ion-beam etching into a Hall bar with the channel width of 10 μm and measured transverse resistivity $\rho_{xy}$ by applying a current along the [100] direction. Figure 1(d) shows $\rho_{xy}$ as a function of the external field at 300 K. Negative Hall-resistance loops were observed for L12-ordered Mn3Pt, which means the signature of AHE exists in L12-ordered Mn3Pt and corresponds to the previous observation in the Mn3X (X = Sn, Ge, Ga, Ir, Pt) family of AFM materials.

We next evaluated the symmetry of SOT and examined the SOT components quantitatively using an ST-FMR technique. The stack structures we measured were Si or MgO/Mn3Pt (15 nm)/Py (8 nm)/MgAl2O4 (2.5 nm). Figure 2(a) shows the schematic geometry of the ST-FMR measurement. Coordinate are defined with respect to the lattice of Mn3Pt. The red strip represents Mn3Pt/Py bilayer. (b) Measured $V_{\text{mix}}$ for L12-ordered Mn3Pt/Py and disordered Mn3Pt/Py at 9 GHz with $\phi_H = 40^\circ$. The applied microwave power is 6 dBm. (c) Schematic of the sample geometry and SOTs acting on Py: $\tau_{\parallel}$ is composed of $\tau_{x,DL}$ and $\tau_{y,DL}$, while $\tau_{\perp}$ is composed of $\tau_{x,FL}$ and $\tau_{y,FL}$. (d) Antisymmetric ($V_a$) and symmetric ($V_s$) ST-FMR component of L12-ordered Mn3Pt/Py and disordered Mn3Pt/Py (shown as an inset) with different $\phi_I$ as a function of $\phi_H$. The error bars represent fitting standard deviations. (e, f) Torque ratios $\tau_{x,DL}/\tau_{y,DL}$ and $\tau_{x,FL}/\tau_{y,FL}$ for L12-ordered Mn3Pt/Py and disordered Mn3Pt/Py as a function of $\phi_I$. 

Fig. 1. (Color online) (a) Crystal and magnetic structure of L12-ordered Mn3Pt. Note that the Mn atoms in the unit cell of Mn3Pt construct a kagome lattice in the (111) plane, where the spin structure is triangular and the direction of spins is either all-in or all-out. (b) X-ray diffraction patterns measured for the 15 nm thick Mn3Pt deposited on MgO and Si substrates. (c) Measured magnetic hysteresis curves for the Mn3Pt with and without L12-ordered structure. A weak magnetization can only be detected for L12-ordered Mn3Pt. (d) AHE of samples in (c). Fig. 2. (Color online) (a) Schematic illustration of ST-FMR measurement. Coordinate are defined with respect to the lattice of Mn3Pt. The red strip represents Mn3Pt/Py bilayer. (b) Measured $V_{\text{mix}}$ for L12-ordered Mn3Pt/Py and disordered Mn3Pt/Py at 9 GHz with $\phi_H = 40^\circ$. The applied microwave power is 6 dBm. (c) Schematic of the sample geometry and SOTs acting on Py: $\tau_{\parallel}$ is composed of $\tau_{x,DL}$ and $\tau_{y,DL}$, while $\tau_{\perp}$ is composed of $\tau_{x,FL}$ and $\tau_{y,FL}$. (d) Antisymmetric ($V_a$) and symmetric ($V_s$) ST-FMR component of L12-ordered Mn3Pt/Py and disordered Mn3Pt/Py (shown as an inset) with different $\phi_I$ as a function of $\phi_H$. The error bars represent fitting standard deviations. (e, f) Torque ratios $\tau_{x,DL}/\tau_{y,DL}$ and $\tau_{x,FL}/\tau_{y,FL}$ for L12-ordered Mn3Pt/Py and disordered Mn3Pt/Py as a function of $\phi_I$.
measurement. A microwave current $I$ was applied to Mn$_3$Pt, and the generated torques were therefore exerted to the Py. An in-plane external field was simultaneously swept at an angle $\phi_H$ relative to $I$, after which a mix voltage $V_{\text{mix}}$ modulated by the anisotropic magnetoresistance (AMR) of Py could be detected by a lock-in amplifier. Such a $V_{\text{mix}}$ can be fitted as $V_{\text{mix}} = V_s F_s + V_a F_a$, where $F_s$ and $F_a$ denote the symmetric and antisymmetric Lorentzian functions, and $V_s$ and $V_a$ denote the amplitudes of $F_s$ and $F_a$, respectively. Figure 2(b) shows the ST-FMR spectra of L1$_2$-ordered Mn$_3$Pt/Py and disordered Mn$_3$Pt/Py at 9 GHz with $\phi_H = 45^\circ$ and symmetric and antisymmetric components separated by fitting to $V_{\text{mix}} = V_s F_s + V_a F_a$. The data points were fitted well by the equation above. In order to evaluate the possible DL torque and FL torque generated by NO-polarization due to MSHE, we also performed angle-dependent ST-FMR measurements with respect to $\phi_H$, which is a reliable approach for identifying unconventional torques. According to the symmetric analysis of spin-polarization-induced SOTs that exert to the Py layer, the in-plane torque $\tau_{\parallel}$ and out-of-plane torque $\tau_{\perp}$ components of SOT are proportional to $V_s$ and $V_a$, which can be expressed as

$$V_s(\phi_H) \propto \sin(2\phi_H) \tau_{\parallel} = \sin(2\phi_H)[\tau_{x,\text{DL}} \sin(\phi_H) + \tau_{y,\text{DL}} \cos(\phi_H) + \tau_{z,\text{FL}}],$$

(1)

$$V_a(\phi_H) \propto \sin(2\phi_H) \tau_{\perp} = \sin(2\phi_H)[\tau_{x,\text{FL}} \sin(\phi_H) + \tau_{y,\text{FL}} \cos(\phi_H) + \tau_{z,\text{DL}}],$$

(2)

where $\tau_{x,\text{DL}}$, $\tau_{y,\text{DL}}$, $\tau_{z,\text{DL}}$, $\tau_{x,\text{FL}}$, $\tau_{y,\text{FL}}$, $\tau_{z,\text{FL}}$ are the DL/FL torques generated by spin currents that are polarized along the $x$, $y$, $z$ directions, respectively. Note that effects such as Rashba-like and Dresselhaus-like fields and exchange coupling at the FM and AFM interface may contribute to the above torques, as discussed below. Figure 2(c) depicts the components of SOTs exerting on Py.

We further investigated the unconventional torques ($\tau_x$ and $\tau_y$) by patterning a series of microstrips on the same sample with different $\phi_H$, where $\phi_H$ is defined as the angle between [100] orientation and $I$, thereby measuring the $\phi_H$ dependence of $V_s$ and $V_a$. The resistivity of ordered Mn$_3$Pt, disordered Mn$_3$Pt, and Py is estimated, by standard four-probe method, as 213 $\mu\Omega$ cm, 99 $\mu\Omega$ cm, and 82 $\mu\Omega$ cm, respectively. In Fig. 2(d), we compare the $V_s$ and $V_a$ dependence of $\phi_H$ for L1$_2$-ordered Mn$_3$Pt and disordered Mn$_3$Pt with different $\phi_I$. Next, we describe the experimental phenomenon using $V_a$ as an example. When we focus on these curves in the range of $\phi_H = 0^\circ$ to $\phi_H = 180^\circ$ (plus side), two phenomena could be observed clearly for L1$_2$-ordered Mn$_3$Pt (red and black curves): (1) The $V_a$ values of two peaks bounded by $\phi_H = 90^\circ$ are different regardless of $\phi_I = 30^\circ$ or $\phi_I = 60^\circ$, and (2) the $\phi_H$ position of high and low peaks is exchanged when we switch $\phi_I = 30^\circ$ to $\phi_I = 60^\circ$. These two phenomena could also be observed in the range of $\phi_H = -180^\circ$ to $\phi_H = 0^\circ$ (minus side), while both of them remain absent for the disordered Mn$_3$Pt. The different heights of the peaks in phenomenon (1) are a signal for the appearance of the $\tau_{x,\text{FL}}$-induced $\sin(2\phi_H)$ component, while the position switching of the peaks in phenomenon (2) could be explained by the sign reversal of $\tau_{x,\text{FL}}$. It is also worth noting that although the magnitude of the $V_a$ of L1$_2$-ordered Mn$_3$Pt and disordered Mn$_3$Pt are obviously different due to the distinct impedance mismatch, it does not cause a difference between the two peaks discussed above. $V_a$ also exhibited a similar behavior attribute to the existence of the $\sin(2\phi_H)$ contribution induced by $\tau_{x,\text{DL}}$. Such a result demonstrates a strong correlation between unconventional torques and the triangular spin structure. We, therefore, measured the $V_s$ and $V_a$ as a function of $\phi_H$ from $\phi_H = 0^\circ$ to $\phi_H = 90^\circ$ and fitted them to Eqs. (1) and (2), thereby extracting the associated fitting parameters $\tau_{x,\text{DL}}$, $\tau_{y,\text{DL}}$, $\tau_{z,\text{DL}}$, $\tau_{x,\text{FL}}$, $\tau_{y,\text{FL}}$, and $\tau_{z,\text{FL}}$ and plotting the ratios of $\tau_{x,\text{DL}}/\tau_{y,\text{DL}}$ and $\tau_{x,\text{FL}}/\tau_{y,\text{FL}}$ as a function of $\phi_H$, as shown in Fig. 2(e) and 2(f). Such a torque ratio can help us to quantitatively estimate the $x$ component and $y$ component that contribute to $V_s$ and $V_a$. The FL torque generated by $y$-polarization is negligible so the Oersted field is mainly what contributes to $\tau_{x,\text{FL}}$. Detectable $\tau_{x,\text{DL}}/\tau_{y,\text{DL}}$ of about 0.08 and $\tau_{x,\text{FL}}/\tau_{y,\text{FL}}$ of about 0.07 were observed for L1$_2$-ordered Mn$_3$Pt, and a sign inversion behavior was clearly exhibited for both ratios, which may have originated from the generation and polarization reversal of the spin polarization along the $x$ direction (current direction). However, both the detectable $\tau_x$ and the sign inversion behavior vanish for the disordered one. Notably, compared to the conventional $\tau_x$,

![Fig. 3.](image-url)
which yields a Rashba-like field with the same Rashba-like symmetry, the unconventional FL and DL torques $\tau_{x,FL}$ and $\tau_{x,DL}$ that have been observed above correspond to a Dresselhaus-like symmetry.\textsuperscript{17,30}

The same angular-dependent experiment was carried out in MgO/Mn$_3$Pt (15 nm)/Cu (2 nm)/Py (8 nm) stacks with a Cu insertion layer to rule out possible effects caused by interlayer exchange coupling. We compared torque ratios $\frac{\tau_{x,DL}}{\tau_{x,FL}}$ for samples with and without a Cu layer, as shown in Fig. 3(a). Remarkably, the variation as well as the values of $\frac{\tau_{x,DL}}{\tau_{x,FL}}$ for the sample with a Cu spacer were quite similar to its counterpart without Cu, indicating that the interlayer effects between L1$_2$-ordered Mn$_3$Pt and Py are not likely to account for the appearance of $x$-polarization. The appearance of $x$-polarization in our Mn$_3$Pt sample with a triangular $\Gamma_{4g}$ spin structure can be interpreted phenomenologically by the fact that spin-momentum locking yields MSHE. \textsuperscript{15,18}

Recent reports have demonstrated that the momentum space of non-collinear AFM is related strongly to its spin structure. Figure 3(b) shows the schematics of the spin structure of the triangular $\Gamma_{4g}$ magnetic configuration in momentum space. Upon applying an electric field, the distribution of the electrons at the Fermi level will change, and a spin current with $x$-polarization along the $x$ direction will be generated for both states of the canted kagome plane in our (001)-oriented L1$_2$ Mn$_3$Pt sample will give rise to the detection of magnetic concomitant unique symmetry vanished in disordered Mn$_3$Pt. Invariance of the sample with a Cu spacer excluded the possible effects from interlayer coupling, suggesting that the observation of unconventional SOT can be attributed to the facet-dependent MSHE in L1$_2$-ordered Mn$_3$Pt. Our findings provide insights into how unconventional SOT can be generated by taking advantage of the topological spin structure in non-collinear AFM. In addition, this work offers other perspectives to clarify the underlying physics for charge to spin conversions in non-collinear AFMs.

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