

Interface-relevant out-of-plane spin polarization in IrMn₃/permalloy bilayers

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Out-of-plane spin polarization (σ_z) has potential applications for magnetic memory with high storage density and low energy consumption. Several noncollinear antiferromagnets have been confirmed to generate σ_z due to their triangular spin configuration on kagome planes, including IrMn₃. Apart from the spin configuration of the (110)-oriented IrMn₃, we demonstrate that the interface of IrMn₃/permalloy also contributes to the generation of σ_z . With Cu insertion between IrMn₃ and permalloy, interfacial σ_z vanishes, which further supports the interfacial origin of σ_z . We are not only convinced that the interface-relevant σ_z is independent of exchange coupling between IrMn₃ and permalloy but also propose several possible origins of interface-relevant σ_z . Our findings enrich the understanding of generating σ_z in antiferromagnets/ferromagnets bilayers. The interface-relevant σ_z broadens the scope of material design and proposes a potential path to optimize magnetic memories with low power consumption.

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I. INTRODUCTION

According to the classic scenario of spin-orbit torque (SOT) induced by the spin Hall effect (SHE), a charge current flowing in the in-plane direction (x axis) generates a spin current along the out-of-plane direction (z axis) with spin polarization along the y axis (σ_y) [1,2]. In this case, ferromagnets (FM) with perpendicular magnetic anisotropy, which benefit high-density storage and device miniaturization, usually need an external magnetic field to break the symmetry in order to achieve deterministic switching [3–7]. Recently, out-of-plane spin polarization (σ_z) has received a great deal of interest due to its ability to realize field-free switching of perpendicular magnetization with high efficiency [8–10]. In general, σ_z can be generated not only from nonmagnets (NM) with low crystalline symmetry [8,11–13], but also from several antiferromagnets (AFM) owing to the reduced symmetry considering the magnetic structure [14–22]. The noncollinear antiferromagnet IrMn₃ has compensated moments with 120° triangular spin texture in the {111} kagome plane as shown in Fig. 1(a) [17], giving rise to nontrivial magnetoelectrical transport phenomena [23,24]. Previous studies have reported that IrMn₃(100) can generate σ_z and corresponding unconventional torques, making IrMn₃ a promising spin source [17,18].

Interfaces play a pivotal role in SOTs. In addition to the σ_y from the Rashba-Edelstein effect [3,25–27], σ_z has also been investigated at the FM/NM interface. The origin of σ_z includes the spin-orbit precession due to the interfacial spin-orbit field [28], anomalous SHE [29], magnetization-dependent SHE [30,31], and nonequilibrium spin swapping [32]. Compared with FM, AFM has vanishing magnetization with fast dynamics and negligible stray field [33,34]. Moreover, the

magnetic structure of AFM may bring about a new dimension for the interface-relevant σ_z . However, the generation of σ_z at AFM/FM interfaces still remains elusive.

In this work, we demonstrate (110)-oriented IrMn₃ can generate out-of-plane fieldlike torques relevant to the IrMn₃/Permalloy (Ni₈₀Fe₂₀, Py) interface as well as the bulk of IrMn₃ via spin-torque ferromagnetic resonance (ST-FMR) measurements. Figure 1(b) shows the bulk origin of σ_z that relates to the spin configuration of IrMn₃. When the applied current (J_c) is parallel to the cluster magnetic octupole moment (T), spins will precess along the spin-orbit field (H_{SO}) produced by the current, bringing about out-of-plane components. Besides the bulk of IrMn₃, the interface of IrMn₃/Py can generate σ_z even though $J_c \perp T$, as displayed in Fig. 1(c). In the IrMn₃/Cu/Py control sample, the vanishment of σ_z further reveals the relevance between the additional σ_z and the IrMn₃/Py interface. After performing the field annealing, the interface-relevant σ_z degenerates while the bulk-induced σ_z keeps still no matter which direction the annealing field is along.

II. EXPERIMENTS

We grew 23-nm-thick IrMn₃ (110) films on MgO (110) substrates at 773 K by DC magnetron sputtering followed by 1-h annealing at 773 K *in situ*. X-ray-diffraction spectrum of IrMn₃ indicates obvious peaks of IrMn₃ (110) and (220), apart from the peaks of MgO (220) as shown in Fig. 1(d). Secondary phases do not emerge in the present IrMn₃ (110). Figure 1(e) shows the Φ -scan measurement of IrMn₃ {100} and MgO {100} at $\chi = 45^\circ$, confirming the quasiepitaxial growth mode of IrMn₃. The crystallographic orientation of IrMn₃ relative to the substrate is epitaxial IrMn₃ {110}{100} || MgO{110}{100}, demonstrated by the same peak position of IrMn₃ and MgO substrate. Magnetic hysteresis loop measured by the superconducting quantum interference device (SQUID)

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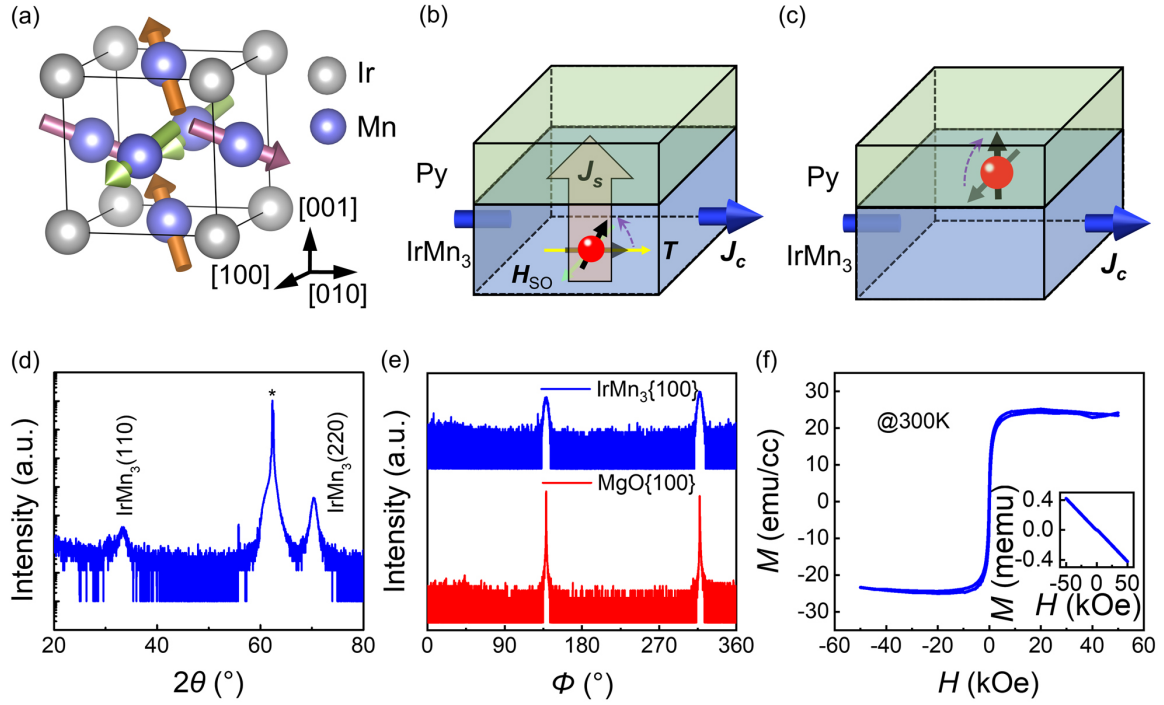


FIG. 1. Schematic of dual generation of out-of-plane spin polarization and basic properties of IrMn₃ film. (a) Magnetic structure of IrMn₃. (b) Schematic diagram of σ_z related to the relative orientation between the applied current J_c and the cluster magnetic octupole moment T . (c) Schematic diagram of σ_z related to the interface of IrMn₃/Py. (d) θ - 2θ measurement of IrMn₃, indicating the strong (110) orientation of IrMn₃ film. (e) Φ -scan measurement of IrMn₃ (110), demonstrating the quasiepitaxial growth mode of IrMn₃. (f) Magnetic hysteresis loop of IrMn₃ measured by SQUID. The inset is the raw data, appearing as a diamagnetic behavior with a kink around the zero field.

at 300 K with an in-plane magnetic field is shown in the inset of Fig. 1(f), appearing as a diamagnetic behavior with a tiny kink around zero field, which indicates the antiferromagnetic characteristic of IrMn₃. For better understanding, the diamagnetic base is deduced in Fig. 1(f). Saturated magnetization is around 24 emu/cc, which is reasonable in noncollinear antiferromagnets [19,35]. The prepared IrMn₃ films with strong (110) orientation provide a precondition for further study of charge-to-spin conversion.

We deposited 15-nm-thick Py and 2-nm-thick Al on the (110)-oriented 12-nm-thick IrMn₃ after cooling to the room temperature *in situ*. IrMn₃/Py/Al samples were patterned into microstrips with a size of 30 μm \times 20 μm by standard photolithography and Ar-ion milling techniques. Top electrodes of Ti/Au were then deposited by e-beam evaporation. Devices with different orientations were fabricated to investigate the symmetry of the generation of σ_z . The current direction in conjunction with the cluster magnetic octupole moment can be controlled by choosing different ST-FMR devices.

ST-FMR was used to measure the spin torques in IrMn₃/Py bilayers. As shown in Fig. 2(a), microwave current was applied along the stripe, which arouses an alternating torque on the upper Py with the same period as the rf current. Successively, the precession of magnetic moments in Py excited by the rf current drives the resistance of the stripe to change periodically due to the anisotropy magnetoresistance. Mixing with the alternating current through Py, alternating resistance generates a double-frequency voltage signal as well as zero-frequency signal, namely, the rectified voltage V_{mix} which is measured by a nanovoltmeter.

III. RESULTS AND DISCUSSION

The discussion of ST-FMR spectrum is based on the V_{mix} as a function of the external magnetic field H shown in Fig 2. At a fixed frequency, V_{mix} around the resonance magnetic field H_{res} can be fitted as a function of magnetic field H by [36]

$$V_{\text{mix}} = V_S \frac{\Delta H^2}{\Delta H^2 + (H - H_{\text{res}})^2} + V_A \frac{\Delta H(H - H_{\text{res}})}{\Delta H^2 + (H - H_{\text{res}})^2}, \quad (1)$$

where H_{res} is the resonance magnetic field, and ΔH is the linewidth. The amplitude of the Lorentzian symmetric line shape V_S and the antisymmetric V_A is proportional to the amplitude of the in-plane torque τ_{\parallel} and out-of-plane torque τ_{\perp} , respectively, as illustrated in Fig 2(a). Note that the angle between the applied current J_c and the magnetic field H is denoted by ϕ_H . Figure 2(b) displays the measured V_{mix} at $\phi_H = 40^\circ$, 10 GHz, and 15 dBm in IrMn₃/Py, where the applied current is along the $[1\bar{1}0]$ crystal axis of IrMn₃. Note that the materials with high symmetry only allow conventional torques corresponding to SHE or the Rashba-Edelstein effect, where $V_{\text{mix}}(H)$ should be equal to $-V_{\text{mix}}(-H)$. Strikingly, the amplitude of V_S in the positive field is much larger than that in the negative field in IrMn₃/Py as depicted in Fig. 2(b), revealing the unconventional torque triggered by σ_z . Then, $4\pi M_{\text{eff}}$ is extracted from the Kittel equation $f = \frac{\gamma}{2\pi} \sqrt{H_{\text{res}}(H_{\text{res}} + 4\pi M_{\text{eff}})}$ in Fig. 2(c), whose value is 9320 Oe, indicating the resonance peak originates from the ferromagnetic resonance signal of Py.

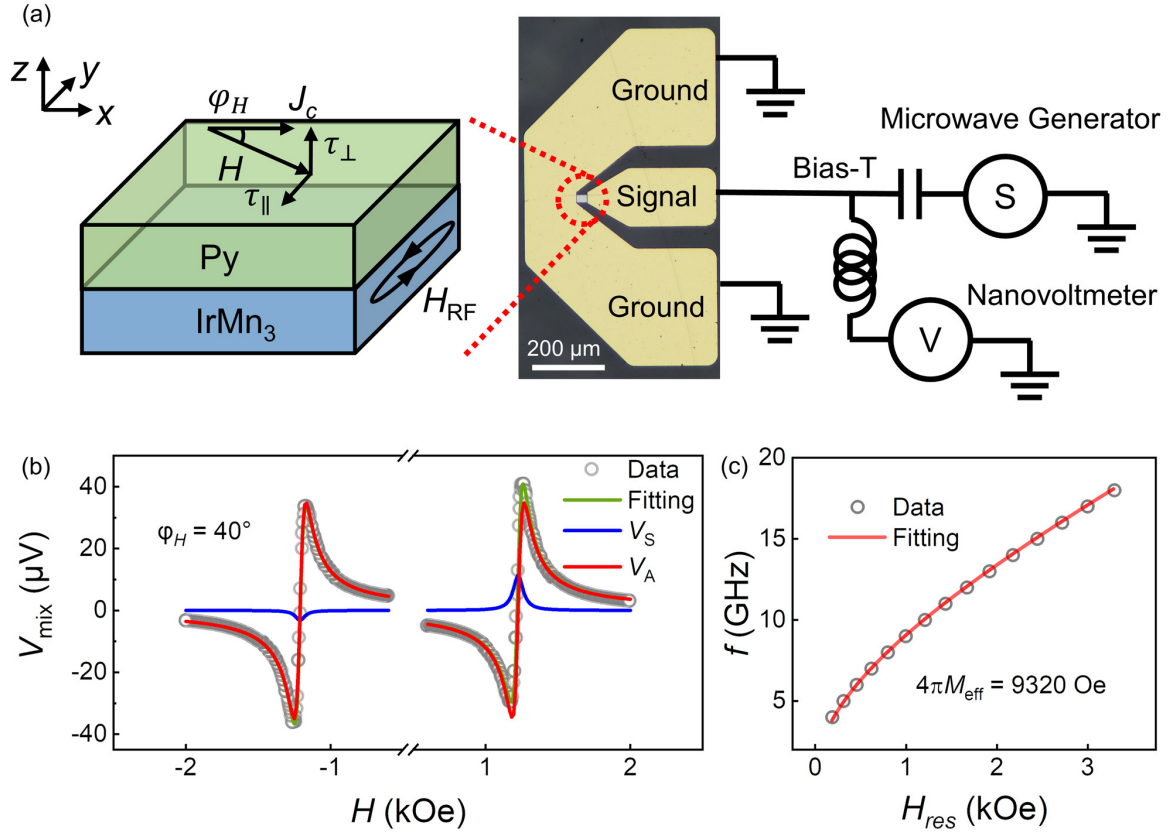


FIG. 2. ST-FMR measurement of IrMn₃/Py. (a) Schematic of ST-FMR geometry of IrMn₃/Py bilayer, corresponding optical microscope graph of the device, and the setup. φ_H is the angle between rf current J_c and magnetic field H . rf current is triggered in microwave generator and the rectified voltage is measured by nanovoltmeter. (b) ST-FMR spectrum of IrMn₃/Py at 10 GHz and $\varphi_H = 40^\circ$. Raw data V_{mix} are shown as gray circles, while the fitting line is depicted in green. V_S and V_A extracted from fitting lines are illustrated in blue and red, respectively. (c) Frequency fitting of Kittel equation from 4 to 18 GHz at $\varphi_H = 45^\circ$ and the corresponding M_{eff} . Resonance field H_{res} is extracted from the fitting of V_{mix} likewise.

f , γ , and M_{eff} are the microwave frequency, the gyromagnetic ratio, and the effective magnetization, respectively.

To investigate the components of the torque generated by IrMn₃, angular-dependent ST-FMR measurements are performed. Antidamping torques exhibit a common form of $\tau_{\text{AD}} \propto \mathbf{m} \times \boldsymbol{\sigma} \times \mathbf{m}$, in which $\boldsymbol{\sigma}$ is the spin polarization and \mathbf{m} stands for the unit vector of magnetization. The common form of fieldlike torques is $\tau_{\text{FL}} \propto \mathbf{m} \times \boldsymbol{\sigma}$. The direction of $\boldsymbol{\sigma}$ determines the variation of corresponding torques with φ_H . The spin polarization along the x axis (σ_x), σ_y , and σ_z contribute to $\sin \varphi_H \sin 2\varphi_H$, $\cos \varphi_H \sin 2\varphi_H$, and $\sin 2\varphi_H$ components, respectively. By fitting V_S and V_A as a function of φ_H , components of torques triggered by IrMn₃ can be differentiated individually. V_S and V_A can be expressed, respectively, as

$$V_S = \tau_{\text{AD},x} \sin \varphi_H \sin 2\varphi_H + \tau_{\text{AD},y} \cos \varphi_H \sin 2\varphi_H + \tau_{\text{FL},z} \sin 2\varphi_H, \quad (2)$$

$$V_A = \tau_{\text{FL},x} \sin \varphi_H \sin 2\varphi_H + \tau_{\text{FL},y} \cos \varphi_H \sin 2\varphi_H + \tau_{\text{AD},z} \sin 2\varphi_H, \quad (3)$$

where $\tau_{\text{AD},x}$, $\tau_{\text{AD},y}$, and $\tau_{\text{AD},z}$ are the coefficients related to antidamping torques. Likewise, $\tau_{\text{FL},x}$, $\tau_{\text{FL},y}$, and $\tau_{\text{FL},z}$ are the coefficients corresponding to fieldlike torques. Given that

fieldlike torques triggered by σ_y have the same symmetry, $\cos \varphi_H \sin 2\varphi_H$, but a negligible amplitude compared with torques generated by the Oersted field, $\tau_{\text{FL},y}$ is considered as the contribution from Oersted field [36,37]. V_A can be well fitted by $\cos \varphi_H \sin 2\varphi_H$ in each device, indicating that V_A is mainly contributed by the Oersted field (Fig. S1 in Supplemental Material [38]). Hence, only V_S is shown and discussed in this study.

When the applied current (J_c) is parallel to [001] ($J_c \parallel \mathbf{T}$), the emergence of σ_z can be well interpreted by the cluster magnetic octupole (\mathbf{T}). Each cluster is defined as six Mn atoms related to each other by crystal-symmetry operators without space translation in a magnetic unit cell [39]. \mathbf{T} can be considered as a ferroic ordering, pointing to the same direction as the tiny canted moment, illustrated by yellow arrows in Figs. 3(a) and 3(d) [14,40,41]. When the charge current is applied to IrMn₃, a spin-orbit field \mathbf{H}_{SO} perpendicular to the current arises [42], making spins precess around it. The generation of σ_z depends on the conjunction between \mathbf{T} and J_c , namely, $\sigma_z \propto \mathbf{H}_{\text{SO}} \times \mathbf{T}$ [20]. The principle of generating σ_z mentioned above is equal to the magnetic symmetry analysis regarding IrMn₃ [17]. The magnetic mirror symmetry M' contains a crystal mirror symmetry M and a time-reversal symmetry. Although IrMn₃ has high crystal symmetry, even twofold

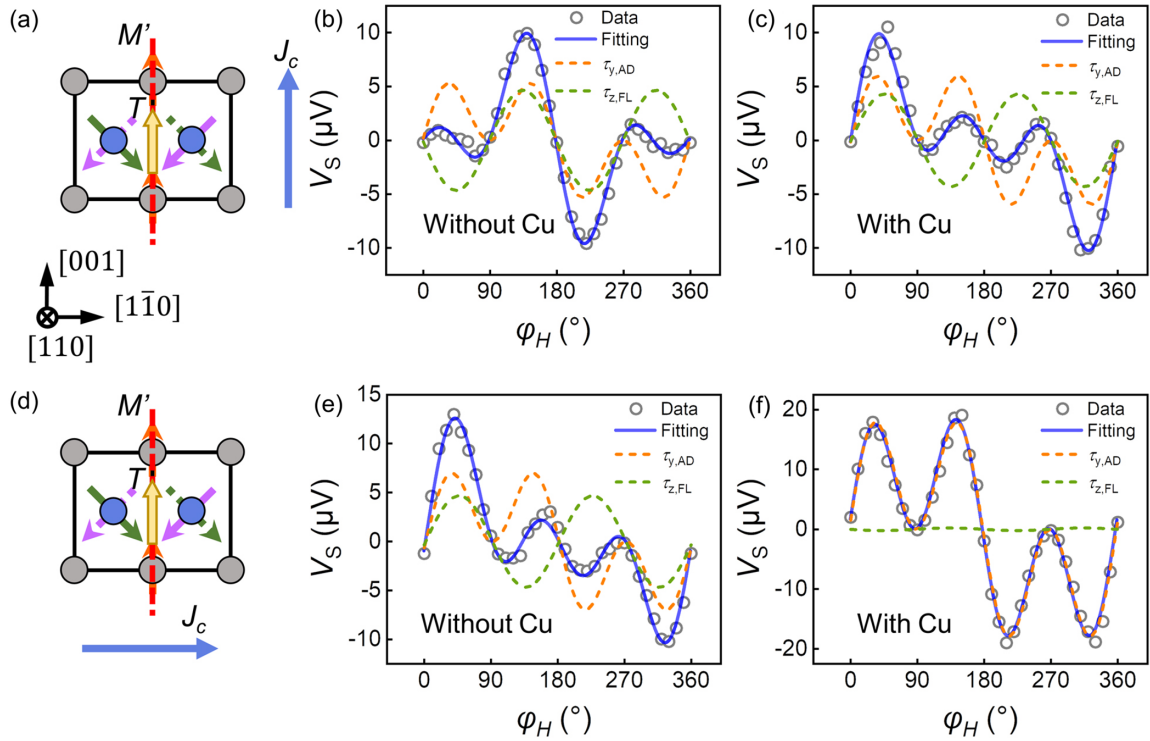


FIG. 3. V_S extracted from ST-FMR spectrum at varied φ_H in IrMn_3/Py and $\text{IrMn}_3/\text{Cu}/\text{Py}$. (a), (d) Projection of crystal structure and spin texture on the (110) plane. Applied current (J_c) is parallel to the cluster magnetic octupole moment T (a) and perpendicular to T (d). Red dashed line indicates the magnetic mirror M' . Yellow arrow represents the direction of T . Gray and blue balls denote Ir and Mn atoms, respectively. (b), (c) Angular-dependent symmetric signal and its components when $J_c \parallel T$ ($J_c \parallel M'$) without (b) or with Cu insertion (c). (e), (f) Angular-dependent V_S and its components when $J_c \perp T$ ($J_c \perp M'$) without (e) or with (f) Cu insertion.

rotational variation is lost if taking the magnetic structure of IrMn_3 into consideration. The magnetic mirror symmetry is broken on condition that $J_c \parallel M'$, which triggers σ_z . The magnetic mirror M' in IrMn_3 is parallel to the $(1\bar{1}0)$ plane, whose projection is indicated as red dashed lines in Figs. 3(a) and 3(d). Note that the arrows through the blue balls denote the magnetization of each Mn atom. Arrows with the same color indicate the same direction, and atoms with solid arrows are above those with dashed arrows. (See 3D structure of M' and T in Fig. S3 in the Supplemental Material [38].) In Fig. 3, the angle-dependent V_S can be well fitted by the sum of $\cos \varphi_H \sin 2\varphi_H$ and $\sin 2\varphi_H$, which are corresponding to the antidamping torque generated by σ_y ($\tau_{\text{AD},y}$) and the fieldlike torque generated by σ_z ($\tau_{\text{FL},z}$), respectively. If $J_c \parallel T$ ($J_c \parallel M'$) as shown in Fig. 3(b), $\tau_{\text{FL},z}$ has a comparable amplitude with $\tau_{\text{AD},y}$, showing the robust emergence of σ_z in IrMn_3/Py , which is consistent with the principle above. Surprisingly, considerable σ_z still arises when $J_c \perp T$ ($J_c \perp M'$), as depicted in Fig. 3(e). To probe the derivation of the unexpected σ_z , control samples with 2-nm-thick Cu insertion between IrMn_3 and Py are prepared, as the long diffusion length of Cu. In $\text{IrMn}_3/\text{Cu}/\text{Py}$ samples, currents applied along $T(M')$ can trigger σ_z as shown in Fig. 3(c) while currents perpendicular to $T(M')$ cannot [Fig. 3(f)]. The crystallographic orientation-dependent σ_z is consistent with previous studies in noncollinear antiferromagnets [17,19]. The Cu insertion allows the transmission of spin currents and eliminates the possible interfacial effect in the IrMn_3/Py bilayer. Therefore, the disappearance of σ_z with the applied current J_c

perpendicular to $T(M')$ suggests the additional contribution to σ_z is probably related to IrMn_3/Py interface.

Among the interfacial effects of AFM/FM bilayers, exchange coupling between AFM and FM should be considered first, whose intensity is usually reflected by the exchange bias. We control the exchange coupling of IrMn_3/Py via annealing and cooling at 473 K in an in-plane magnetic field of 8 kOe. The obtained exchange bias in IrMn_3/Py is confirmed by the magnetization measurements (Fig. S2 in Supplemental Material [38]). Notably, $\tau_{\text{AD},y}$ in IrMn_3/Py bilayer is attributed to the SHE due to the large spin-orbit coupling of IrMn_3 , which has been proven to be independent of exchange coupling [43]. Considering that $\tau_{\text{AD},y}$ in IrMn_3/Py bilayer cannot be influenced by the field annealing, hence we use $\tau_{\text{FL},z}/\tau_{\text{AD},y}$ to evaluate the amplitude of fieldlike torque generated by σ_z .

As for the device where $J_c \perp T$, $\tau_{\text{FL},z}$ and $\tau_{\text{AD},y}$ are acquired from the fitting of V_S as a function of φ_H on condition that the annealing field H_{FA} is applied along different directions, as depicted in Fig. 4. After carrying out the field annealing, $\tau_{\text{FL},z}/\tau_{\text{AD},y}$ declines significantly. Compared with the considerable amplitudes of $\tau_{\text{FL},z}/\tau_{\text{AD},y}$ in Fig. 3(e), the amplitude of $\tau_{\text{FL},z}$ (shown as green dashed lines) is much lower than that of $\tau_{\text{AD},y}$ (shown as orange dashed lines) in Figs. 4(a)–4(c), indicating the negligible σ_z in IrMn_3/Py when $J_c \perp T$ after field annealing. Note that the annealing fields H_{FA} are parallel to the applied current J_c with opposite directions in Figs. 4(a) and 4(b). Despite the sign of $\tau_{\text{FL},z}/\tau_{\text{AD},y}$ in Fig. 4(b) being opposite after H_{FA} switches, the amplitudes of $\tau_{\text{FL},z}$ are both negligible compared with those of $\tau_{\text{AD},y}$, indicating the

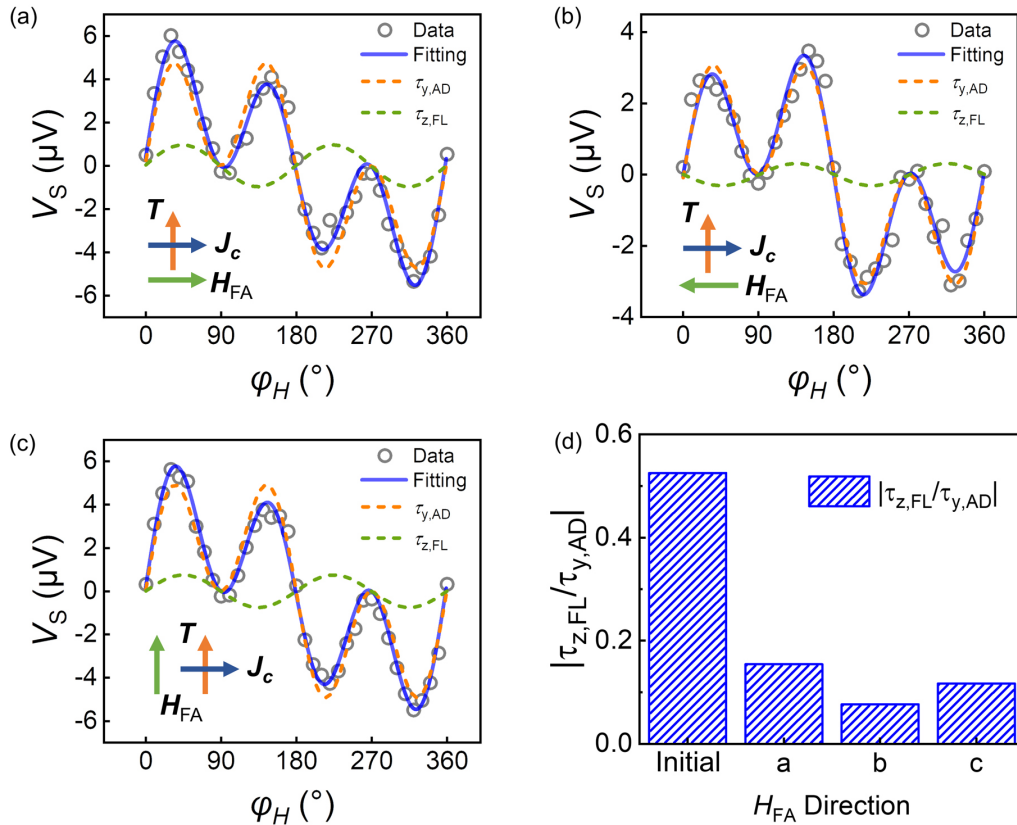


FIG. 4. Angular-dependent V_S and its components with the current \mathbf{J}_c applied perpendicular to the cluster magnetic octupole moment \mathbf{T} after field annealing in IrMn₃/Py. (a), (b) Annealing field \mathbf{H}_{FA} perpendicular to \mathbf{T} while parallel or antiparallel to \mathbf{J}_c , respectively. (c) Annealing field \mathbf{H}_{FA} parallel to \mathbf{T} while perpendicular to \mathbf{J}_c . (d) Summary of $\tau_{FL,z}$ calibrated by $\tau_{AD,y}$ in different annealing conditions.

irrelevant between generating σ_z and the sign of exchange-bias field. Similarly, the exchange-bias field perpendicular to the applied current \mathbf{J}_c cannot make σ_z recurrent as shown in Fig. 4(c). The absolute value of $\tau_{FL,z}/\tau_{AD,y}$ is summarized in Fig. 4(d). The considerable decline of σ_z probably results from the disappearance of perpendicular moments at the interface during the field annealing, which will be discussed below. The degeneration of σ_z suggests that the in-plane exchange bias has no significant influence on the generation of σ_z relevant to the interface.

Field annealing was carried out on devices where $\mathbf{J}_c \parallel \mathbf{T}$ to probe the influence of in-plane exchange bias on bulk σ_z induced by the magnetic structure of IrMn₃, as shown in Fig. 5. The angle-dependent symmetry signal V_S can be decomposed into $\cos \phi_H \sin 2\phi_H$ (indicating the antidamping torques contributed by σ_y) and $\sin 2\phi_H$ (indicating the fieldlike torques contributed by σ_z), shown as orange dashed lines and green dashed lines, respectively, in Fig. 5. All of the figures show a common feature that the amplitudes of $\tau_{FL,z}$ are comparable to those of $\tau_{AD,y}$, indicating the distinguished generation of σ_z in IrMn₃/Py after field annealing. Nevertheless, the sign of $\tau_{FL,z}/\tau_{FL,y}$ remains negative in all of the configurations. Considering the significant decline of interfacial σ_z after field annealing (Fig. 4), σ_z in cases of $\mathbf{J}_c \parallel \mathbf{T}$ (Fig. 5) is mainly from the spin configuration of IrMn₃, namely, the cluster magnetic octupole moments \mathbf{T} of IrMn₃.

As depicted in Figs. 5(a) and 5(b), both the amplitude and the sign of σ_z have no significant change regardless of

whether the annealing field is parallel or antiparallel to \mathbf{T} in parallel conjunction with \mathbf{J}_c . Assuming that the current direction reverses, the sign of σ_z will reverse, during which case the relative orientation between \mathbf{T} and \mathbf{J}_c also reverses. Thus, the inverse of \mathbf{T} will result in the sign inverse of σ_z when the direction of \mathbf{J}_c remains unchanged. The unchanging of σ_z in Figs. 5(a) and 5(b) indicates that the direction of the cluster magnetic octupole moment \mathbf{T} would not be modified by field annealing at 473 K, which is relatively lower than the growing and annealing temperature *in situ*. Furthermore, as shown in Figs. 5(c) and 5(d), field annealing perpendicular to \mathbf{J}_c as well as \mathbf{T} has no influence on the generation of σ_z contributed by the magnetic structure of IrMn₃. Therefore, not only the field annealing but also the exchange coupling between IrMn₃ and Py would not influence the σ_z arising from the bulk of IrMn₃.

The interfacial σ_z can be explained by the interfacial perpendicular moment of Py due to Dzyaloshinskii-Moriya (DM) interaction [38]. The inversion symmetry is broken at the interface of IrMn₃/Py, bringing about the DM interaction between Mn atoms and Ni (Fe) atoms, which favors the perpendicular alignment of Mn moments and Ni (Fe) moments. Ni (Fe) moments cant to the direction normal to the kagome plane of IrMn₃ slightly after summing up the effective DM fields on each Ni (Fe) atom per unit cell [44]. In our case of IrMn₃(110)/Py, the interfacial moments of Ni (Fe) perpendicular to the kagome plane {111} give the components perpendicular to {110} planes, namely, the interfacial moments normal to the interface of IrMn₃/Py. Out-of-plane spin

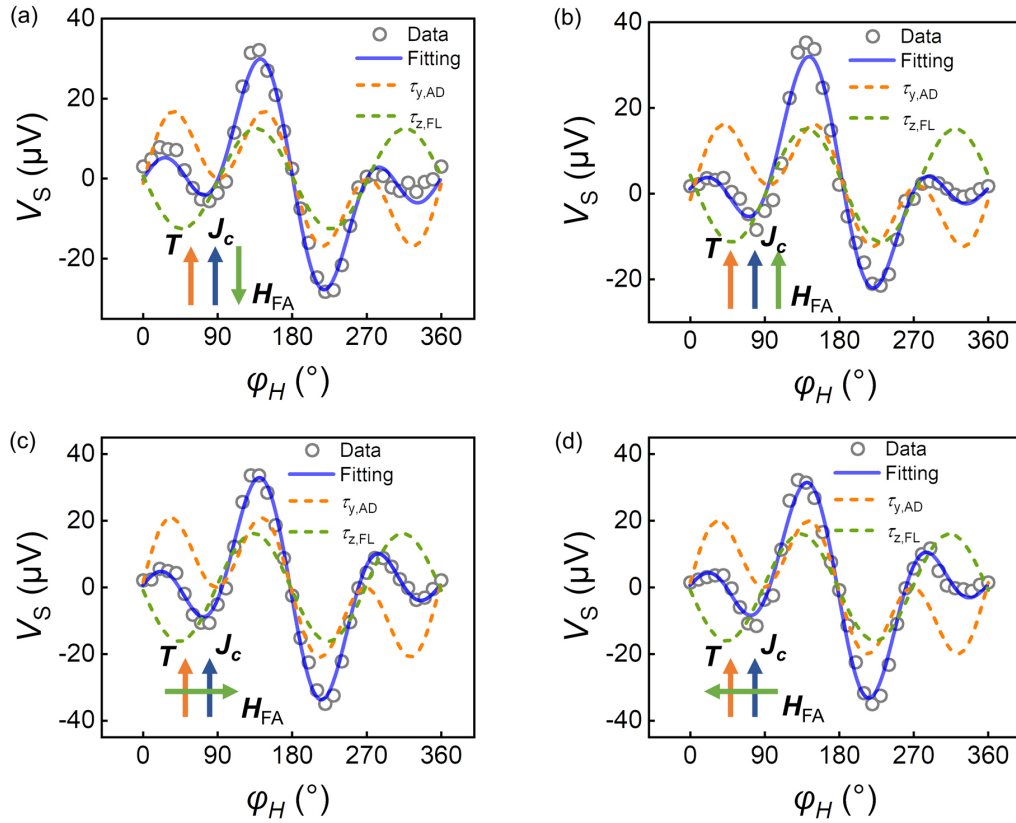


FIG. 5. Invariance of σ_z that is related to spin texture of IrMn₃ with the current J_c applied parallel to the cluster magnetic octupole T after field annealing in IrMn₃/Py. (a), (b) Annealing field H_{FA} parallel to T as well as J_c . (c), (d) Annealing field H_{FA} perpendicular to T as well as J_c .

polarization is generated by the exchange of the spin angular momentum between σ_y produced by the SHE of IrMn₃ and the interfacial moments, similar to the spin transfer in FMs [45].

Besides, the anomalous spin-orbit torque has the same form as the fieldlike torque generated by σ_z , namely, $m \times z$ [46]. Given the strong spin-orbit coupling of IrMn₃, strong spin-orbit scattering arises at the IrMn₃/Py interface while weak scattering at Py/Al, resulting in the asymmetric anomalous spin-orbit torques at the two interfaces of Py. Hence, a net anomalous spin-orbit torque arises and may also contribute to the interface-relevant σ_z . Another possible source of interface-relevant σ_z is the nonequilibrium spin-swapping effect due to scattering related to local moments at IrMn₃/Py interface [32].

IV. CONCLUSION

In summary, we have investigated the dual sources of out-of-plane spin polarization in the IrMn₃/Py bilayer via ST-FMR. The fieldlike torque of σ_z arises in two current directions that are orthogonal to each other. The control sample with Cu insertion confirms the bulk source of σ_z related to

the noncollinear spin texture of IrMn₃. Specifically, $J_c \parallel T$ triggers strong σ_z while $J_c \perp T$ does not in IrMn₃/Cu/Py. The bulk σ_z preserves its amplitude after annealing in the magnetic field; σ_z relevant to interface degenerates after the field annealing. Regardless of the direction that the in-plane annealing field is along, the amplitude of fieldlike torque related to σ_z is relatively low compared with antidamping torque related to σ_y , indicating that the interfacial σ_z is independent of the exchange coupling between IrMn₃ and Py. We suppose several possible origins may give rise to the interfacial σ_z at IrMn₃/Py interface. Our findings deepen the understanding of σ_z in noncollinear AFM/FM systems and will trigger the interests and further exploration of the derivation of interface-relevant σ_z in antiferromagnets. The interface-relevant σ_z provides potential candidates for next-generation magnetic memory devices.

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