

Unconventional Unidirectional Spin Hall Magnetoresistance in Epitaxial IrMn/FeNi Bilayer

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Unidirectional spin Hall magnetoresistance (USMR), arising from the interaction between nonequilibrium spin accumulation and magnetization, has been proposed as a simple two-terminal method for electrically detecting magnetic states in heavy-metal/ferromagnet bilayers. However, conventional spin polarization along the y direction restricts USMR to responding only to the transverse component of in-plane magnetization when a charge current is applied along the x direction. As a result, the electrical readout of longitudinal magnetic states via USMR has remained elusive. Here, we overcome this limitation by demonstrating an unconventional USMR induced by an x -polarized spin current in (001)-oriented epitaxial IrMn/FeNi bilayers, which enables electrical detection of the longitudinal magnetic state via USMR. By applying charge currents along different crystal orientations, we show that the x -polarized spin current exhibits a fourfold symmetry, consistent with the crystal mirror symmetry of (001)-oriented IrMn. This unconventional spin current further generates a large spin-orbit torque efficiency, an order of magnitude higher than that reported in antiferromagnetic Mn_3Pt and MnPd_3 systems. Our Letter not only highlights the crucial role of intrinsic crystal symmetry in controlling spin polarization, but also extends the applicability of USMR to the electrical detection of longitudinal magnetization in two-terminal magnetic memory and spin-logic devices.

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Introduction—Symmetry, the invariance of a system or physical law under specific transformations, is a cornerstone of physics. It fundamentally restricts the mathematical form of physical laws by imposing symmetry constraints. Symmetry breaking gives rise to new phenomena and principles; thus, controlling symmetry enables both the realization and interpretation of diverse physical behaviors. This reasoning directly applies to the commonly observed unidirectional spin Hall magnetoresistance

(USMR) in heavy-metal/ferromagnet (HM/FM) bilayers [1–6]. As a nonlinear magnetoresistance, USMR violates the Onsager reciprocity relation, exhibiting odd symmetry under reversal of either magnetization or current. This asymmetric resistance depends on the relative orientation between the spin polarization (σ) and magnetization ($\mathbf{m}$). When σ and $\mathbf{m}$ are parallel (antiparallel), the USMR exhibits a high (low) resistance. Because of its unidirectional nature, USMR enables the electrical detection of magnetic states in HM/FM bilayers using a simple two-terminal configuration [1,7–9].

Although USMR has been widely demonstrated in HM/FM [1,3,10–12], HM/antiferromagnetic insulator [13,14], and topological insulator/FM bilayers [2,7,15–17], systems with polycrystalline or amorphous structures typically

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allow the generation of spin currents with in-plane spin polarization along the y direction (σ_y) when a charge current is applied along the x direction via the spin Hall effect [18–23] and/or the Rashba-Edelstein effect [24,25]. This follows the relation $\mathbf{J}_s = \theta_{\text{SH}} \boldsymbol{\sigma} \times \mathbf{J}_e$, where $\mathbf{J}_s$, θ_{SH} , and $\mathbf{J}_e$ denote the spin current, spin Hall angle, and the charge current, respectively, requiring $\mathbf{J}_s$, $\boldsymbol{\sigma}$, and $\mathbf{J}_e$ to be mutually orthogonal. Under this symmetry constraint, the type-y USMR is defined by the asymmetry in the second-harmonic longitudinal resistance ($R_{xx}^{2\omega}$) as $\Delta R_{xx}^{2\omega} = R_{xx}^{2\omega}(\mathbf{m} \parallel +\mathbf{y}) - R_{xx}^{2\omega}(\mathbf{m} \parallel -\mathbf{y})$ when the magnetization is saturated between opposite orientations along the $\pm y$ direction with an alternating charge current $I_c = I_0 \sin(\omega t)$ along the x direction. Consequently, the type-y USMR is limited to detecting transversal magnetic states in two-terminal geometries. However, the unidirectional nature of USMR does not intrinsically restrict its symmetry. Recently, type-z USMR has been reported in the heterostructures of a topological semimetal and a perpendicular magnetic anisotropy ferromagnetic insulator, enabling electrical readout of perpendicular magnetization [26]. As the final piece of the USMR picture, type-x USMR—which takes the form of $\Delta R_{xx}^{2\omega} = R_{xx}^{2\omega}(\mathbf{m} \parallel +\mathbf{x}) - R_{xx}^{2\omega}(\mathbf{m} \parallel -\mathbf{x})$ when the magnetization is saturated the $\pm x$ direction—remains critically missing. The realization of type-x USMR would enable the electrical detection of longitudinal magnetization states in two-terminal spintronic devices. This capability offers a more direct approach for studying magnetic anisotropy, spin-orbit torque (SOT)-driven magnetization switching, and bistable memory cells, as it allows the full in-plane magnetization vector to be determined using a simple two-terminal geometry.

The unidirectional nature of USMR depends on the relative orientation between $\boldsymbol{\sigma}$ and $\mathbf{m}$; thus, achieving type-x USMR requires the generation of an unconventional spin current with x -polarized spin (σ_x). Unlike conventional charge to spin conversion in polycrystalline or amorphous materials, the lower symmetry of single-crystal systems provides a promising route to control spin polarization [27–33]. For instance, in low-symmetry crystals such as 2D Weyl semimetals WTe_2 [27–29] and TaIrTe_4 [30,31], as well as single-crystal $\text{L1}_1\text{-CuPt/CoPt}$ alloys [32,33], symmetry breaking can produce a spin current with out-of-plane spin polarization σ_z that depends sensitively on the relative orientation between the charge current and the high-symmetry crystal axes. In contrast, an x -polarized spin current is characterized by spin polarization σ_x collinear with the charge current along the x direction. The x -polarized spin current has been reported in noncentrosymmetric ferromagnets like $(\text{Ga,Mn})\text{As}$ via Dresselhaus spin-orbit coupling [34–39], noncollinear antiferromagnets Mn_3Pt via magnetic spin Hall effect [40–44], and ultrathin $[\text{Pt/Co}]_n$ films via the strong orbital magnetic moment of Co [45]. Nevertheless, the critical role of crystal symmetry in controlling spin transport with in-plane unconventional spin polarization σ_x remains

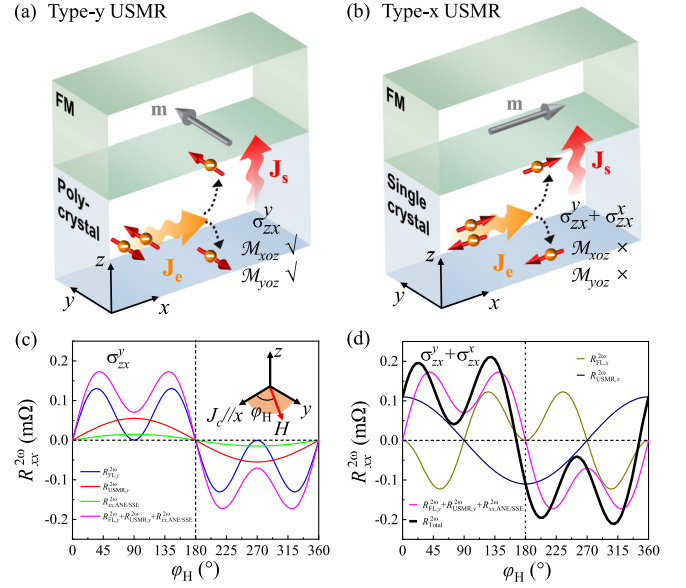


FIG. 1. Schematic illustration of (a) type-y USMR in a polycrystalline HM/FM bilayer and (b) type-x USMR in a single-crystal HM/FM bilayer. The corresponding in-plane angular dependence of the second-harmonic resistance $R_{xx}^{2\omega}$ is shown for (c) the polycrystalline bilayer, which contains the σ_{zx}^y contribution and thermoelectric contribution $R_{xx, \text{ANE/SSE}}^{2\omega}$, and (d) the single-crystal bilayer, which includes both σ_{zx}^y and σ_{zx}^x contributions along with the thermoelectric contribution $R_{xx, \text{ANE/SSE}}^{2\omega}$.

incompletely understood [46–48]; direct experimental evidence for type-x USMR induced by in-plane unconventional spin polarization σ_x , as well as the corresponding electrical detection of longitudinal magnetization in a two-terminal geometry, is still lacking.

In this Letter, to the best of our knowledge, we report the first experimental observation of type-x USMR in epitaxial IrMn/FeNi bilayers, which is driven by unconventional x -polarized spin currents arising from the control of crystal symmetry. Combined with previous reports of type-y and type-z USMR, our work completes the USMR picture (type-x, type-y, and type-z) for electrically reading out all three magnetization orientations—transverse, longitudinal, and perpendicular—within a simple two-terminal geometry. Furthermore, we demonstrate crystal-symmetry-controlled spin transport by determining the dampinglike SOT efficiency for charge currents applied along different crystal orientations. We find that x -polarized spin currents—and, consequently, type-x USMR—appear only when the current direction simultaneously breaks both $\mathcal{M}_{xoz}$ and $\mathcal{M}_{yoz}$ mirror symmetries and vanish when either symmetry is preserved. This on-off switching directly establishes crystal-symmetry-controlled spin transport with in-plane unconventional spin polarization.

Results—Figure 1 illustrates type-y and type-x USMR induced by y -polarized and x -polarized spin currents, along with their respective magnetic-field angular dependences.

In a polycrystalline HM/FM bilayer, both $\mathcal{M}_{xoz}$ and $\mathcal{M}_{yoz}$ mirror symmetries are preserved. This symmetry forbids the generation of an x -polarized spin current, allowing only a y -polarized spin current. The symmetry analysis is derived in Supplemental Material [49]. The resulting type- y USMR, shown in Fig. 1(a), exhibits higher (lower) resistance when the magnetization is parallel (antiparallel) to the $+y$ direction. In contrast, for a single-crystal HM/FM bilayer, the $\mathcal{M}_{xoz}$ and $\mathcal{M}_{yoz}$ mirror symmetries are spontaneously broken along specific crystalline orientations. This broken symmetry permits the generation of both y -polarized and x -polarized spin currents [49]. The type- x USMR arising from the x -polarized spin current, shown in Fig. 1(b), displays higher (lower) resistance when the magnetization is parallel (antiparallel) to the $+x$ direction. Consequently, the unconventional x -polarized spin current is predicted to produce a distinct type- x USMR, quantified as $\Delta R_{xx}^{2\omega} = R_{xx}^{2\omega}(\mathbf{m} \parallel \mathbf{x}) - R_{xx}^{2\omega}(\mathbf{m} \parallel -\mathbf{x})$ when the magnetization is saturated the $\pm x$ direction.

For magnetoresistance measurements, an alternating charge current $I_c = I_0 \sin(\omega t)$ is injected into the HM/FM bilayer. The longitudinal voltage, comprising its first- and second-harmonic components, is recorded as $V_{xx} = V_{xx}^{1\omega} + V_{xx}^{2\omega} = I_0 R_{xx}^{1\omega} + I_0 R_{xx}^{2\omega}$ [1]. The first-harmonic longitudinal resistance $R_{xx}^{1\omega}$ includes contributions from current-independent effects such as the anisotropic magnetoresistance and the spin Hall magnetoresistance [53,54]. The second-harmonic resistance $R_{xx}^{2\omega}$ originates from current-dependent effects, which primarily consist of three contributions: the USMR, the magnetoresistance induced by alternating SOT, and the magnetothermal resistance arising from temperature gradients [1,50,55]. In a polycrystalline HM/FM bilayer, high symmetry permits only the generation of a y -polarized spin current. When an external magnetic field is rotated within the xoy plane at an angle φ_H , the field-angular dependence of $R_{xx}^{2\omega}$ can be expressed by Eq. (1):

$$R_{xx}^{2\omega} = R_{FL,y}^{2\omega} + R_{USMR,y}^{2\omega} + R_{xx,ANE/SSE}^{2\omega}, \quad (1)$$

with

$$\begin{aligned} R_{FL,y}^{2\omega} &= -(R_x - R_y) \frac{H_{FL,y}}{H} \sin(2\varphi_H) \cos \varphi_H, \\ R_{USMR,y}^{2\omega} &= R_{USMR,y} \sin \varphi_H, \\ R_{xx,ANE/SSE}^{2\omega} &= R_{xx,ANE/SSE} \sin \varphi_H, \end{aligned} \quad (2)$$

where $R_{FL,y}^{2\omega}$, $R_{USMR,y}^{2\omega}$, and $R_{xx,ANE/SSE}^{2\omega}$ are the contributions from fieldlike torque, USMR induced by the y -polarized spin current, the anomalous Nernst effect (ANE) and the spin Seebeck effect (SSE), $H_{FL,y}$ is the effective field of the fieldlike torque induced by the y -polarized spin current, and R_x and R_y are the longitudinal resistances when the magnetization is saturated along the x and y directions,

respectively. $R_{xx,ANE/SSE}^{2\omega}$ can be obtained using the relation $R_{xx,ANE/SSE}^{2\omega}/R_{xy,ANE/SSE}^{2\omega} = l/w$, where $R_{xy,ANE/SSE}^{2\omega}$ is determined from the second-harmonic Hall resistance measurements and appears as an intercept in the linear fit of $R_{DL}^{2\omega}$ versus $R_A/(H + H_k)$. The corresponding angular dependence is displayed in Fig. 1(c). In the case of a single-crystal HM/FM bilayer, the spontaneous breaking of the $\mathcal{M}_{xoz}$ and $\mathcal{M}_{yoz}$ mirror symmetries along specific crystalline orientations allows the generation of both y -polarized and x -polarized spin currents. Noting that $m_x \propto \cos \varphi_H$, we point out that the type- x USMR originating from the x -polarized spin current introduces an additional $\cos \varphi_H$ term in the angular dependence of $R_{xx}^{2\omega}$. Including the contributions from both y -polarized and x -polarized spin currents to the SOT and USMR, as well as the SSE and ANE, the full angular dependence of $R_{xx}^{2\omega}$ can be expressed with five terms as in Eq. (3):

$$R_{xx}^{2\omega} = R_{FL,y}^{2\omega} + R_{USMR,y}^{2\omega} + R_{FL,x}^{2\omega} + R_{USMR,x}^{2\omega} + R_{xx,ANE/SSE}^{2\omega}, \quad (3)$$

with

$$R_{FL,x}^{2\omega} = (R_x - R_y) \frac{H_{FL,x}}{H} \sin(2\varphi_H) \sin \varphi_H, \quad (4)$$

$$R_{USMR,x}^{2\omega} = R_{USMR,x} \cos \varphi_H, \quad (5)$$

where $H_{FL,x}$ is the effective field of the fieldlike torque induced by the x -polarized spin current. The corresponding curve is shown in Fig. 1(d). The signals arising from y -polarized and x -polarized spin currents can be distinguished based on their distinct symmetries and field-angular dependences.

To investigate type- x USMR, we fabricate IrMn (20 nm)/FeNi (6 nm)/Ta (2 nm) stacks on MgO (100) single-crystal substrates via magnetron sputtering. The IrMn (20 nm; Ir₂₀Mn₈₀) layer is deposited at 600 °C and subsequently annealed *in situ* at the same temperature for 1 h. After cooling to room temperature, FeNi (6 nm; Fe₂₀Ni₈₀) and a Ta (2 nm) capping layer are deposited. The top Ta layer naturally oxidizes, protecting the underlying FeNi from oxidation. Owing to the high resistivity of the oxidized Ta, its current-shunting effect is assumed to be negligible. The crystal structure of γ -IrMn is face-centered cubic (fcc, point group O_h), with Mn and Ir atoms randomly occupying the fcc lattice sites [51,56] as illustrated in Fig. 2(a). The scanning transmission electron microscopy image in Fig. 2(b) confirms the single-crystalline, (001)-oriented epitaxy of IrMn. Figure 2(c) shows the out-of-plane x-ray diffraction pattern of the stack. The IrMn (002) diffraction peak, with a full width at half maximum (FWHM) of 0.3°, attests to the high-quality [001] epitaxial orientation. The complete absence of superlattice peaks such as (001) or

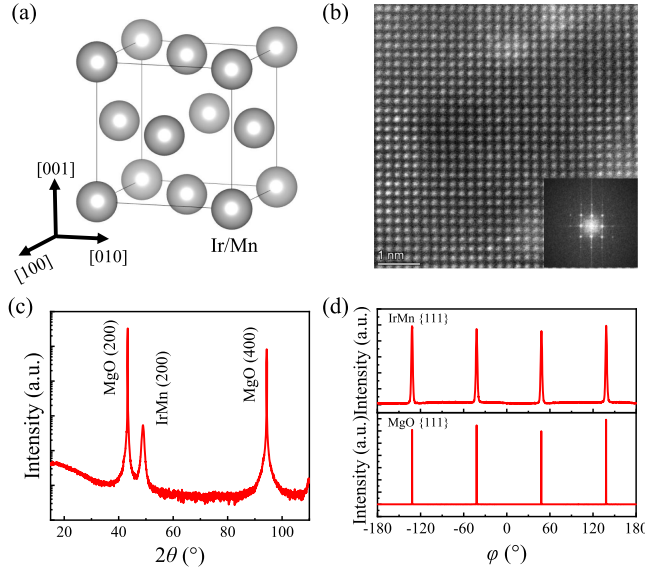


FIG. 2. (a) Crystal structure (unit cell) of the γ -phase IrMn (point group O_h), in which Ir and Mn atoms are chemically disordered. (b) Cross-sectional scanning transmission electron microscopy image. (c) Out-of-plane 2θ - ω x-ray diffraction pattern of the IrMn film grown on a MgO (001) substrate, confirming (001) epitaxy. (d) ϕ scan of the $\{111\}$ family of planes for both the IrMn film and the MgO substrate, illustrating the cube-on-cube epitaxial relationship: $\text{MgO}(001)[110] \parallel \text{IrMn}(001)[110]$.

(003) further confirms that the IrMn film is chemically disordered. Figure 2(d) presents the ϕ scan of the $\{111\}$ lattice planes for both IrMn and the MgO substrate, verifying the epitaxial relationship: $\text{MgO}(001)[110] \parallel \text{IrMn}(001)[110]$. The four equally spaced diffraction peaks demonstrate the in-plane fourfold rotational symmetry of the (001)-oriented IrMn epilayer.

Because of interfacial coupling with the FeNi layer, inversion symmetry is broken at the IrMn/FeNi interface. The point group of the heterostructure thereby reduces to C_{4v} , which contains a fourfold rotational principal axis and four mirror planes intersecting at 45° angles. Figure 3(a) shows the (001)-plane view of the IrMn crystal structure, where four mirror planes perpendicular to the (001) plane are present, namely, (100) and its equivalents and (110) and its equivalents. This defines four high-symmetry in-plane directions: [100] and equivalents and [110] and equivalents. When a charge current is applied along these high-symmetry axes, the $\mathcal{M}_{xoz}$ and $\mathcal{M}_{yoz}$ mirror symmetries are simultaneously preserved, prohibiting the generation of an x -polarized spin current and allowing only a y -polarized spin current. Conversely, when the current is applied between these axes, both $\mathcal{M}_{xoz}$ and $\mathcal{M}_{yoz}$ mirror symmetries are broken, permitting the generation of both x -polarized and y -polarized spin currents. Samples were patterned into Hall bars (length $l = 30 \mu\text{m}$ and width $w = 20 \mu\text{m}$) with various channel directions via optical lithography. The channel direction is defined by the

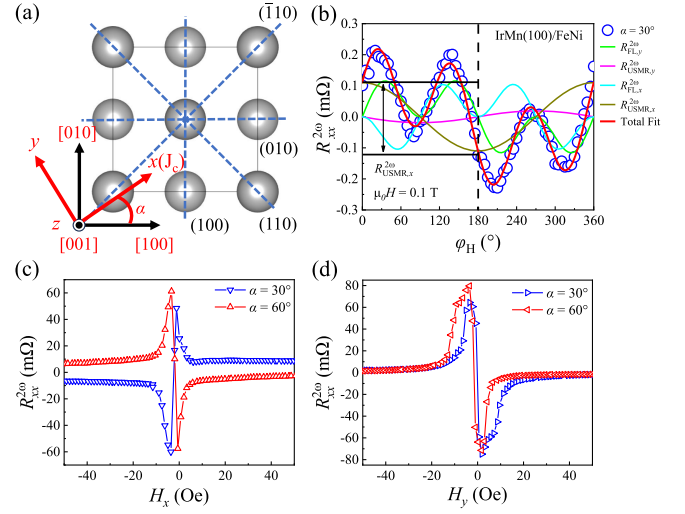


FIG. 3. (a) Definition of the current-flow direction via the azimuthal angle α between the Hall-bar channel and the crystalline [100] axis. The blue dashed line indicates a mirror plane perpendicular to the (001) plane. (b) In-plane angular dependence of the second-harmonic resistance $R_{xx}^{2\omega}$ measured at 300 K, 0.1 T, and a current density of 4.2 MA/cm^2 . (c), (d) $R_{xx}^{2\omega}$ as a function of magnetic field applied along the x (c) and y direction (d) for samples with $\alpha = 30^\circ$ and 60° (300 K, $J_c = 6.8 \text{ MA/cm}^2$).

azimuth angle α relative to the [100] crystal axis. The x and y axes follow the same definitions as in Fig. 3(a). Specifically, the x axis is aligned along the current channel. The longitudinal resistance R_{xx} was measured in a four-terminal configuration (constant current along the Hall bar, voltage across the two longitudinal contacts), eliminating contact and lead resistances. For a Hall bar with $\alpha = 30^\circ$, the magnetic-field angular dependence of $R_{xx}^{2\omega}$ measured at 0.1 T and $J_c = 4.2 \text{ MA/cm}^2$ is shown in Fig. 3(b) and is well fitted by Eq. (3). It is worth noting that, due to the low resistivity ($91 \mu\Omega \cdot \text{cm}$) and large thickness (26 nm) of the epitaxial IrMn/Py bilayer, the determined $R_{xx, \text{ANE/SSE}}^{2\omega}$ is negligible compared with the USMR signal. Further discussion is provided in Supplemental Material [49]. The data reveal an asymmetry between $\varphi_H = 45^\circ, 135^\circ$ and $\varphi_H = 225^\circ, 315^\circ$, attributed to the fieldlike torque from the x -polarized spin current. Furthermore, the asymmetry at $\varphi_H = 0^\circ, 180^\circ$ indicates a contribution from type- x USMR. These asymmetries disappear for Hall bars with $\alpha = 0^\circ, 45^\circ$, and 90° (see Supplemental Material [49]). To highlight the asymmetric signal more clearly, Fig. 3(c) plots $R_{xx}^{2\omega}$ versus the longitudinal magnetic field H_x . For $\alpha = 0^\circ, 45^\circ$, and 90° , no difference in $R_{xx}^{2\omega}$ is observed after magnetization saturation along $\pm x$ (see Supplemental Material [49]). In contrast, for $\alpha = 30^\circ$ and 60° , a notable difference—i.e., type- x USMR—is clearly observed after magnetization saturation along $\pm x$. Moreover, constrained by the fourfold crystal symmetry, a sign reversal of the type- x USMR occurs between $\alpha = 30^\circ$ and 60° , suggesting a

corresponding reversal in the spin-polarization direction. Figure 3(d) shows $R_{xx}^{2\omega}$ versus the transverse field H_y for $\alpha = 30^\circ$ and 60° . The signal after saturation along $\pm y$ exhibits a smaller amplitude and the same sign, consistent with the characteristic of type-y USMR. For comparison with other systems, we have determined the type-x USMR ratio to be 3.5 ppm/(MA/cm²). The electrical detection of longitudinal magnetization switching via type-x USMR has also been demonstrated. Details are provided in Supplemental Material [49].

To characterize the SOT arising from the in-plane unconventional spin polarization in the IrMn/FeNi bilayer, we performed angular-dependent second-harmonic Hall voltage measurements. An ac charge current $I_c = I_0 \sin(\omega t)$ with $\omega/2\pi = 133$ Hz was injected along the longitudinal direction of a Hall bar, and the transverse Hall voltage $V_{xy} = V_{xy}^{1\omega} + V_{xy}^{2\omega} = I_0 R_{xy}^{1\omega} + I_0 R_{xy}^{2\omega}$ was recorded [52,57]. As illustrated in Fig. 4(a), the injected ac current generates spin currents that exert an alternating SOT on the magnetization of the FeNi layer. This torque drives a small oscillatory motion of the magnetization around its equilibrium, thereby inducing a second-harmonic Hall voltage $V_{xy}^{2\omega}$. By rotating an external magnetic field within the xoy plane at an angle ϕ_H , the resulting $R_{xy}^{2\omega}$ reflects the specific geometric symmetry of the underlying spin-current polarization. Considering the three possible spin polarizations (σ_x , σ_y , and σ_z), the in-plane angular dependence of $R_{xy}^{2\omega}$ can be expressed as [45,52]

$$R_{xy}^{2\omega} = -R_{DL,y}^{2\omega} \cos \phi_H + R_{FL}^{2\omega} \cos 2\phi_H \cos \phi_H + R_{DL,z}^{2\omega} \cos 2\phi_H - R_{DL,x}^{2\omega} \sin \phi_H + R_{PNE}^{2\omega} \sin 2\phi_H, \quad (6)$$

where $R_{DL,y}^{2\omega} = \frac{1}{2} R_{AHE} [H_{DL,y} / (H_{ext} + H_k)] + R_{ANE/SSE}$ is the dampinglike (DL) resistance from the y -polarized spin current, $R_{FL}^{2\omega} = R_{PHE} [(H_{FL} + H_{Oe}) / H_{ext}]$ is the fieldlike (FL) resistance, $R_{DL,z}^{2\omega} = R_{PHE} (H_{DL,z} / H_{ext})$ is the DL resistance from the z -polarized spin current, and $R_{DL,x}^{2\omega} = \frac{1}{2} R_{AHE} [H_{DL,x} / (H_{ext} + H_k)]$ is the DL resistance from the x -polarized spin current. Here, R_{AHE} , $R_{ANE/SSE}$, and R_{PHE} denote the anomalous Hall resistance, the resistance from the anomalous Nernst effect (ANE) or spin Seebeck effect (SSE), and the planar Hall resistance, respectively. The terms H_{FL} , $H_{DL,x}$, $H_{DL,y}$, and $H_{DL,z}$ represent the effective fields of the SOT components, $R_{PNE}^{2\omega}$ is the second-harmonic resistance from the planar Nernst effect, H_{ext} is the applied magnetic field, and H_k is the effective anisotropy field. Figure 4(b) shows the angular-dependent $R_{xy}^{2\omega}$ for a Hall bar with $\alpha = 30^\circ$, which exhibits a pronounced asymmetric signal. Fitting the data with Eq. (6) reveals that $H_{DL,x}$ and $H_{DL,y}$ scale linearly with the applied current. The corresponding DL-torque efficiency per unit current density for the σ_x and σ_y components is given by

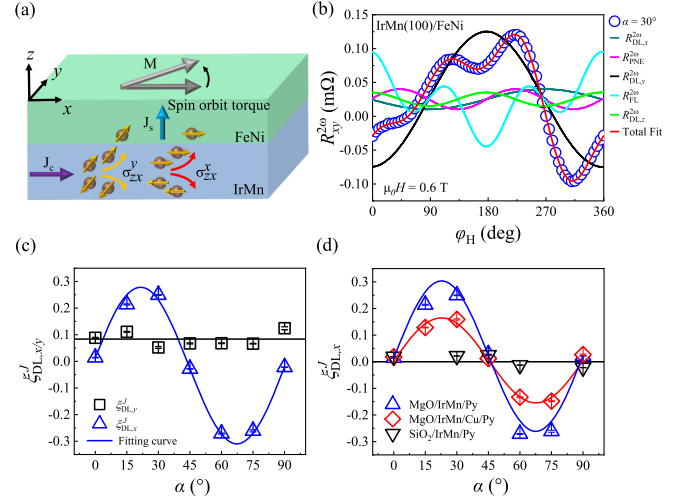


FIG. 4. (a) Schematic of current-induced SOT in the IrMn/FeNi bilayer. (b) In-plane angular dependence of the second-harmonic Hall resistance $R_{xy}^{2\omega}$ measured at 300 K, 0.6 T, and $J_c = 4.2$ MA/cm². (c) Dampinglike SOT efficiency per unit current density from the x -polarized ($\xi_{DL,x}^J$) and y -polarized ($\xi_{DL,y}^J$) spin currents, plotted as a function of the azimuthal angle α . (d) Comparison of $\xi_{DL,x}^J$ versus α for three systems: epitaxial IrMn/FeNi, epitaxial IrMn/Cu/FeNi, and polycrystalline IrMn/FeNi.

$$\xi_{DL,x/y}^J = \frac{2e\mu_0 M_s t_{FM} H_{DL,x/y}}{\hbar J_c}, \quad (7)$$

where e , M_s , t_{FM} , and $\hbar$ are the elementary charge, saturation magnetization, ferromagnetic layer thickness, and reduced Planck constant, respectively. Figure 4(c) plots $\xi_{DL,x}^J$ and $\xi_{DL,y}^J$ as functions of the current-direction angle α . The efficiency $\xi_{DL,x}^J$ shows a strong fourfold crystalline anisotropy, whereas $\xi_{DL,y}^J$ remains nearly constant at approximately 0.08. The maximum value of $\xi_{DL,x}^J$ reaches about 0.28—roughly one order of magnitude larger than reported for antiferromagnetic Mn₃Pt [42] and MnPd₃ [52]. Further discussion and cross-check are provided in Supplemental Material [49]. Previous studies have suggested that strong interfacial coupling in IrMn₃/FeNi can modify the damping factor of FeNi [41]. To examine whether interfacial effects are responsible for the unconventional σ_x spin current in epitaxial IrMn/FeNi bilayer, we inserted a 2-nm Cu spacer to remove the interfacial coupling completely. Figure 4(d) compares $\xi_{DL,x}^J$ versus α for the epitaxial IrMn/FeNi bilayer, a polycrystalline IrMn/FeNi bilayer, and an epitaxial IrMn/Cu/FeNi trilayer. After removing the interfacial coupling by inserting the Cu spacer, $\xi_{DL,x}^J$ remains substantial and retains its fourfold symmetry. The attenuation of $\xi_{DL,x}^J$ with the insertion of the Cu spacer can be attributed to the loss of the spin current. This result rules out interfacial coupling as the origin of the x -polarized spin current. As a further control, we prepared a polycrystalline IrMn/FeNi bilayer on a SiO₂ substrate. In this case, $\xi_{DL,x}^J$ drops nearly to

zero, confirming that the generation of the σ_x spin current depends critically on the crystalline structure of the IrMn layer.

Conclusion—In summary, we have experimentally demonstrated type- x USMR driven by in-plane unconventional spin polarizations in epitaxial IrMn/FeNi bilayers, where the simultaneous breaking of crystal mirror symmetries $\mathcal{M}_{xoz}$ and $\mathcal{M}_{yoz}$ enables the generation of x -polarized spin currents, in addition to the conventional y -polarized ones. The x -polarized spin currents can be controlled by applying charge currents along different crystal orientations. Together with previous reports on type- y and type- z USMR, our demonstration of type- x USMR completes the USMR picture, enabling two-terminal electrical readout of all three magnetization orientations—transverse, longitudinal, and perpendicular. This capability to fully resolve the in-plane magnetization vector in a simple two-terminal geometry simplifies device design, lowers power consumption, and paves the way for higher-density magnetic memory and spin-logic devices.

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Data availability—There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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