Out-of-plane carrier spin in transition-metal dichalcogenides under electric current

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Absence of spatial inversion symmetry allows a nonequilibrium spin polarization to be induced by electric currents, which, in two-dimensional systems, is conventionally analyzed using the Rashba model, leading to in-plane spin polarization. Given that the material realizations of out-of-plane current-induced spin polarization (CISP) are relatively fewer than that of in-plane CISP, but important for perpendicular-magnetization switching and electronic structure evolution, it is highly desirable to search for new prototypical materials and mechanisms to generate the out-of-plane carrier spin and promote the study of CISP. Here, we propose that an out-of-plane CISP can emerge in ferromagnetic transition-metal dichalcogenide monolayers. Taking monolayer VSe₂ and VTe₂ as examples, we calculate the out-of-plane CISP based on linear-response theory and first-principles methods. We deduce a general low-energy model for easy-plane ferromagnetic transition-metal dichalcogenide monolayers and find that the out-of-plane CISP is due to an in-plane magnetization together with intrinsic spin-orbit coupling inducing an anisotropic out-of-plane spin splitting in the momentum space. The CISP paves the way for magnetization rotation and electric control of the valley quantum number.

out-of-plane current-induced spin polarization |
transition-metal dichalcogenides | intrinsic spin-orbit coupling |
valley degree of freedom

In a conducting system with broken inversion symmetry and nonnegligible spin-orbit coupling (SOC), nonequilibrium carrier-spin polarization can be induced by electric currents (1–4). The current-induced spin polarization (CISP) in a magnetic system further applies a torque, i.e., spin-orbit torque, on the magnetic order parameter through the exchange coupling. These inverse spin-galvanic phenomena have seen many spintronic applications in magnetization switching, domain-wall motion, etc., according to Landau–Lifshitz–Gilbert dynamics driven by the spin-orbit torque (5). A bilayer structure composed of a ferromagnetic layer and a heavy nonmagnetic layer is widely used in the study of the CISP (5–7). The inversion symmetry breaking at the interface is minimally captured by two-dimensional (2D) models with Rashba-type SOC (8–10), which gives chiral textures of in-plane spin components in the momentum space. The resulting CISP is always in-plane after the carrier redistribution. The out-of-plane component of CISP arising from further crystalline symmetry breaking has recently been proposed in the bilayer heterostructure (7, 11–14) and quantum well grown in a certain direction (15). However, the examples of the out-of-plane CISP in real materials are still relatively fewer compared with the in-plane CISP that occurs in a large number of Rashba systems. Both new prototypical materials and mechanisms are highly desirable for generating out-of-plane CISP. The out-of-plane CISP is important in device concepts based on perpendicular magnetic anisotropy, where it facilitates perpendicular-magnetization switching by spin-orbit torque. Besides, controlling out-of-plane magnetic ordering enables access to a number of intriguing phenomena, e.g., quantum anomalous Hall effect

(16), semiconductor-to-metal transition (17), and valley splitting (18, 19).

Two-dimensional van der Waals materials provide a plethora of simple and powerful platforms for exploring spin-related physics (20). In particular, monolayer transition-metal dichalcogenides, MX₂ (M = V, Mo, W; X = S, Se, Te, etc.), in the 2H phase have both strong SOC and inversion symmetry breaking (21–24). While special attention has recently been paid to the CISP in MX₂/graphene bilayer (25) and MX₂/ferromagnet bilayer structures (6), the study of possible CISP in 2H-MX₂ monolayers is still lacking. It is worth emphasizing that 2H-MX₂ monolayers are representative 2D systems for the non-Rashba SOC. The intrinsic SOC of monolayer 2H-MX₂ in the low-energy effective theory results from the bulk crystal structure asymmetries, in contrast to Rashba SOC from the interfacial asymmetry. The existence of a mirror plane through the M-atom layer ensures that the potential gradient, ∇V , is in-plane. The SOC-induced effective magnetic field $\propto \nabla V \times \mathbf{p}$, with $\mathbf{p}$ being the 2D momentum, is therefore out-of-plane and results in Zeeman-type spin splitting (21, 22). It is thus an attractive question whether the out-of-plane spin splitting leads to an out-of-plane CISP, which we have fully addressed in this paper. Moreover, there are two valleys in the low-energy bands of 2H-MX₂. Valley, as an electronic degree of freedom, may add a dimension to the CISP, given that valley-contrasting optoelectronic properties offer potential applications in energy-efficient electronics and spintronics (22, 26).

Significance

Although out-of-plane current-induced spin polarization (CISP) is important for perpendicular-magnetization reorientation, and it has been realized by the crystalline symmetry breaking, its material examples are relatively fewer compared with in-plane CISP in Rashba systems. With the help of intrinsic spin-orbit coupling, an intriguing out-of-plane CISP is designed in transition-metal dichalcogenides by the symmetry breaking in the spin space, which provides opportunities for out-of-plane magnetization rotation and electric control of valley splitting. Moreover, the spin polarization is associated with valley-dependent responses to electric current and adds a dimension, valley degree of freedom, to the study of CISP. The symmetry argument and non-Rashba effective model also helps to illuminate these physics and broaden the scope of the CISP.

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In this paper, we use symmetry analysis and first-principles calculations to point out that there is out-of-plane CISP in the prototypical ferromagnet transition-metal dichalcogenides VSe₂ and VTe₂ monolayers (23). Especially for the VSe₂ monolayer, it has been synthesized recently (27, 28). We derive a low-energy model suitable for generic easy-plane ferromagnetic 2H-MX₂ monolayers, and use it to elucidate the physical origin of out-of-plane CISP, which is the anisotropic spin splitting in the momentum space due to both non-Rashba SOC and in-plane magnetization. The symmetry breaking in the spin space enables the out-of-plane CISP in ferromagnet transition-metal dichalcogenides, in contrast to the crystalline symmetry breaking in previous works (7, 11–15). Moreover, two valleys exhibit distinct responses depending on the electric-field direction, which is a convenient experimental knob to tune CISP. This CISP provides opportunities for magnetization reorientation and electric control of valleys. The proposed non-Rashba system and the low-energy model will help to broaden the scope of the study of the CISP and other spin-related physics.

Results

Symmetry Analysis. Symmetry arguments provide a powerful tool for finding possible nonvanishing tensor components of any response function. For the CISP, the corresponding response function is of spin density responding to electric fields. In the good metal limit, we can separate this response function into two contributions: one arises from the nonequilibrium redistribution of carriers in the neighborhood of the Fermi level, under the combined effect of electric field and disorder scattering; the other is from the interband transitions of Fermi sea electrons induced by electric field (29–31). We first study the first contribution that results in out-of-plane CISP and discuss the second contribution at the end of *First-Principles Calculations of CISP*. For the convenience of discussion, we adopt the relaxation time approximation, under which the Fermi surface contribution can be described by

$$\delta\mathbf{s} = \tau\chi\mathbf{E}, \quad [1]$$

where $\delta\mathbf{s}$ is the carrier spin induced by the electric field $\mathbf{E}$, and τ is the relaxation time. χ is the part of the intraband response tensor that does not depend on τ in the good metal limit and is invariant under any unitary symmetry operations of the system in equilibrium. More details can be found in *SI Appendix*.

We now consider symmetries of the 2H-VX₂ (X = Se or Te) monolayer that can help us identify possible nonzero components of χ . Fig. 1 *A* and *B* shows the atomic structure of the VX₂ monolayer with the D_{3h} point group. Two sublattices of a honeycomb lattice are respectively occupied by vanadium and chalcogen atoms. Each vanadium atom lies at the center of a trigonal prismatic cage of six chalcogen atoms. There are therefore three atomic layers stacked as X-V-X. Without considering the magnetization, there are two types of mirror planes. The vanadium layer is a mirror, σ_h , lying in the monolayer (Fig. 1*B*). The other type of mirrors consists of three planes perpendicular to the monolayer and through different V-X bonds. One of them lies in the y - z plane, as denoted by σ_v in Fig. 1*A*. The others are obtained by rotating σ_v through $2\pi/3$ and $4\pi/3$ around the z axis due to the threefold rotational symmetry, C_3 . Since the VX₂ monolayer has an easy-plane magnetic anisotropy (23), the joint transformation of σ_h and time reversal $\mathcal{T}$ becomes a new symmetry, and the C_3 symmetry is broken, when the ferromagnetic order is further taken into account.

Applying the symmetry operation $\sigma_h\mathcal{T}$ on different quantities in Eq. 1, we find the right-hand side is invariant up to a sign change under this operation (*SI Appendix*), if $\mathbf{E}$ is in-plane.

Fig. 1. Atomic and band structures of the VX₂ monolayer. Top view (*A*) and side view (*B*) of the atomic structure. Blue and yellow spheres represent vanadium and chalcogen atoms, respectively. (*C* and *D*) Band structures of VSe₂ with the magnetization along the x and z directions, respectively. Black bands correspond to the pristine monolayer without strain, while blue and red bands correspond to the monolayers under -2 and 2% uniaxial strain along the y direction, respectively. The valence band maximum is set to zero energy. The Brillouin zone is given in *C* (Inset).

Therefore, in order for Eq. 1 to hold, $\delta\mathbf{s}$ must change sign, which means only the out-of-plane component, δs_z , is allowed. The C_3 symmetry breaking by the in-plane magnetization is also crucial for generating δs_z , since both χ and s_z are invariant under the C_3 rotation, but an in-plane $\mathbf{E}$ is not. $\delta\mathbf{s}$ is therefore absent in a perpendicularly magnetized or nonmagnetic monolayer 2H-MX₂ that has the C_3 symmetry. Similarly, a x component of $\mathbf{E}$ is required for nonzero δs_z due to σ_v -related symmetries for a magnetization $\mathbf{M}$ along the high-symmetry x or y directions.

First-Principles Calculations of CISP. Armed with the symmetry analysis, we perform first-principles calculations of the CISP in monolayer VSe₂ and VTe₂ by density functional theory and linear-response theory. The calculated lattice constants are 3.33 and 3.60 Å for VSe₂ and VTe₂, respectively. For both monolayers, the stable ferromagnetic order has a magnetic moment of 1.0 μ_B per unit cell, agreeing with previous results (23).

Fig. 1 *C* and *D* shows band structures of the ferromagnetic VSe₂ monolayer, and similar band structures are also obtained for VTe₂. When the magnetization is in-plane, there are two inequivalent but degenerate valleys at $K_{\pm}$ points of the Brillouin zone. For an out-of-plane magnetization, the valley degeneracy is lifted. There are valley splittings of 78 and 92 meV for VSe₂ and VTe₂, respectively, by comparing valence band maxima at two valleys, similar to magnetized MoX₂ (18, 19). The valley splitting leads to rich valley-spin physics, e.g., valley polarization induced by carrier doping and nonpolarized optical pumping besides the well-known chiral optical field (18, 19, 24). The valley structure and optoelectronic responses are therefore tunable by tilting the magnetization. The out-of-plane CISP, if realized, provides opportunities for electric control of the magnetization and valleys.

We compute the CISP in the VSe₂ and VTe₂ monolayers by the Kubo linear-response formula. For the Fermi surface contri-

bution, the element, $\chi_{\alpha\beta}$, of the response tensor in Eq. 1 is given as (4):

$$\chi_{\alpha\beta} = \frac{e\eta}{\pi S} \text{Re} \sum_{n,\mathbf{k}} \langle s_{\alpha} \rangle_{n\mathbf{k}} \langle v_{\beta} \rangle_{n\mathbf{k}} (G_{n\mathbf{k}}^A G_{n\mathbf{k}}^R - G_{n\mathbf{k}}^R G_{n\mathbf{k}}^A), \quad [2]$$

where e is the electron charge, S is the monolayer's area, and n and $\mathbf{k}$ label bands and wave vectors, respectively. $\langle s_{\alpha} \rangle_{n\mathbf{k}}$ ($\langle v_{\beta} \rangle_{n\mathbf{k}}$) are the diagonal matrix elements of the spin (velocity) operator in the eigenstate basis, and α, β denote Cartesian directions. Note β corresponds to the electric-field direction. $G_{n\mathbf{k}}^{R(A)}$ is the retarded (advanced) Green's function at zero frequency, defined as $G_{n\mathbf{k}}^R = (G_{n\mathbf{k}}^A)^* = 1/(E_F - \varepsilon_{n\mathbf{k}} + i\eta)$, where E_F and $\varepsilon_{n\mathbf{k}}$ are the Fermi energy and the electronic band energy, respectively. $\eta = \hbar/2\tau$ is the band broadening due to the disorder potential. We set $\eta = 0.01$ eV based on previous results of MX₂ monolayers (32, 33). As for the Fermi sea contribution to the CISP, it includes interband matrix elements of the spin and velocity operators (SI Appendix), and it is also calculated. The Fermi surface and Fermi sea contributions can therefore be denoted by intraband and interband CISPs below. They are both shown in Fig. 2.

Fig. 2 A and B shows calculated intraband CISP in the VSe₂ monolayer as a function of the magnetization direction, when $\mathbf{E} \parallel \hat{x}$ and $E_F = -0.2$ eV. The magnetization direction is denoted by the polar angle, θ , and the azimuthal angle, φ . The calculated CISP of VTe₂ and the dependence of CISP on E_F can be found in SI Appendix, Figs. S1 and S2, respectively. While the in-plane CISP is zero, out-of-plane CISP indeed emerges for in-plane magnetization, and it has opposite extreme values for $\mathbf{M}$ along the x and y directions. The out-of-plane CISP still exists as long as there is an in-plane component of the magnetization that breaks the C_3 symmetry, except for some special magnetization directions beyond which the CISP changes its sign. The maximum size of the out-of-plane CISP is about $0.01 \mu_B$ per 1 V/Å for VSe₂. When normalized by a longitudinal conductivity, it is on the order of $10^{-7} \mu_B$ per A/cm (SI Appendix, Fig. S2). With the monolayer thickness of ~ 6 Å considered (34), this corresponds

to an order of $10^{-8} \mu_B$ per 10^7 A/cm², comparable to, e.g., the staggered CISP in Mn₂Au (30, 31). The out-of-plane CISP of the VTe₂ monolayer is much larger than that of the VSe₂ monolayer. In particular, the maximum size of VTe₂ is two orders of magnitude larger than that of VSe₂. Given that the CISP plays a role of effective magnetic field in the magnetization dynamics, the effective magnetic field acting on a magnetic moment of $1 \mu_B$ is also estimated by the relationship $B_{\text{eff}} = \delta s_z J_{\text{ex}} / (\mu_B)^2$, where J_{ex} is the exchange interaction between the carrier spin and the magnetic moment, and estimated to be 0.5 eV according to the spin splitting of ~ 1 eV from the magnetization (SI Appendix, Fig. S3). The effective magnetic fields corresponding to maximum out-of-plane CISP are 89 and 1.5×10^4 T per 1 V/Å for VSe₂ and VTe₂, respectively. Therefore, the electric field needed is much smaller than 1 V/Å, e.g., orders of 10^{-4} to 10^{-3} V/Å, to induce a sizable effective magnetic field, especially for VTe₂. Given that considerable electric fields (35, 36) and considerable doping (37) were achieved in MX₂ monolayers, the CISP and associated torque in VX₂ are expected to be readily observed experimentally by, e.g., the technique of the spin-torque ferromagnetic resonance (7).

When further considering in-plane magnetization rotation ($\theta = \pi/2$) in Fig. 2A, δs_z is proportional to $\cos 2\varphi$. The second-order orientation dependence may be exploited to tune the CISP. When the magnetization is rotated within the x - z plane ($\varphi = 0$) in Fig. 2B, δs_z always points to the same direction, which provides an opportunity for tilting the magnetization perpendicularly, except that it is vanishing for the perpendicular magnetization. Besides, δs_x appears as well. The second-order orientation dependence of the intraband CISP is also found for out-of-plane rotation. For $\mathbf{E} \parallel \hat{y}$, the corresponding δs vanishes when $\mathbf{M}$ is along the highly symmetric x and y directions. These results are consistent with the symmetry analysis.

We also calculated the interband (i.e., Fermi sea) contribution to the CISP (30, 31) and found it to be nearly in-plane, in contrast to out-of-plane CISP from the Fermi surface contribution. δs_x and δs_y are approximately proportional to $\sin\varphi$ or $\cos\varphi$ in Fig. 2C, respectively, demonstrating the first-order orientation dependence. The derivation from the standard sine or cosine function is due to high-order contributions in the magnetization. The similar orientation dependence is also found for the out-of-plane magnetization rotation in Fig. 2D. Besides, it is found that the interband CISP is relatively smaller than the intraband contribution.

Origin of the CISP from a Low-Energy Model. To get a more intuitive understanding of the physical origin of the out-of-plane intraband CISP, we derive an effective Hamiltonian of a generic easy-plane ferromagnetic 2H-MX₂ monolayer in the vicinity of $K_{\pm}$ valleys (38, 39). The derivation details can be found in SI Appendix. By critically considering second-order contributions from the atomic SOC of the X element, and projecting the ferromagnetic exchange interaction term to the low-energy space mainly composed of $d_0^{\uparrow}$, $d_0^{\downarrow}$, $d_{\pm 2}^{\uparrow}$, and $d_{\pm 2}^{\downarrow}$ states of the X atom, we obtain

$$H = H_0 + H_{\text{SOC}} + H_{\text{EI}}, \quad [3a]$$

$$H_0 = v_F (\gamma \sigma_x p_x + \sigma_y p_y) s_0 + \frac{\Delta}{2} \sigma_z s_0, \quad [3b]$$

$$H_{\text{SOC}} = \gamma (\lambda_0 \sigma_+^D + \lambda_2 \sigma_-^D) s_z, \quad [3c]$$

$$H_{\text{EI}} = (M_0 \sigma_+^D + M_2 \sigma_-^D) (s_+^N e^{-i\varphi} + s_-^N e^{i\varphi}) + M_N (\sigma_+^N s_+^N e^{i\varphi} + \sigma_-^N s_-^N e^{-i\varphi}). \quad [3d]$$

Here, the three parts of the Hamiltonian, Eq. 3 b-d, describe massive Dirac states, intrinsic SOC, and exchange interactions (EI) due to an in-plane magnetization with the azimuthal angle φ , respectively. $\sigma_{\alpha}/s_{\alpha}$ ($\alpha = x, y, z$) are Pauli matrices for the

Fig. 2. The evolution of calculated current-induced spin in a unit cell with the magnetization rotation for the VSe₂ monolayer. (A and B) The changes of the Fermi surface contribution with in-plane magnetization rotation and out-of-plane rotation within the x - z plane, respectively. (C and D) The corresponding changes of the Fermi sea contribution. $\varphi = 0$ and $\theta = 0$ correspond to x and z directions, respectively. $\mathbf{E} \parallel x$ and $E_F = -0.2$ eV.

orbital pseudospin/real spin, while σ_0 and s_0 are 2×2 identity matrices. The orbital parts of the basis functions have the relations $d_0 = d_{z2}$ and $d_{\pm 2} = \frac{1}{\sqrt{2}}(d_{x2-y2} \pm id_{xy})$ for $K_{\pm}$ valley, with subscripts 0, ± 2 denoting the magnetic quantum numbers. Up-spin and down-spin are denoted by $\uparrow$ and $\downarrow$, respectively. $\gamma = \pm 1$ labels two valleys. $\sigma_{\pm}^D \equiv \frac{1}{2}(\sigma_0 \pm \sigma_z)$, $s_{\pm}^D \equiv \frac{1}{2}(s_0 \pm s_z)$ and $\sigma_{\pm}^N \equiv \frac{1}{2}(\sigma_x \pm i\gamma\sigma_y)$, $s_{\pm}^N \equiv \frac{1}{2}(s_x \pm is_y)$ are defined to separate the diagonal (D) and nondiagonal (N) elements of the orbital and spin Pauli matrices. v_F and Δ are the Fermi velocity and the crystal-field splitting between d_0 and $d_{\pm 2}$ orbitals, respectively. The two orbitals have different SOC strengths denoted by λ_0 and λ_2 , respectively, which are dependent on the atomic SOC strength of the X atom, λ . We note the SOC terms in H_{SOC} lead to a valley-dependent Zeeman splitting along the z axis (21, 22).

H_{EI} is the major result from this derivation and can be potentially applied to other problems related to MX_2 systems with broken time-reversal symmetry. For our purpose, it turns out that the second term in H_{EI} leads to out-of-plane CISP, when such a low-energy model is applicable. Note the first term in H_{EI} is diagonal in orbital space and leads to a conventional spin splitting, with orbital-dependent exchange-coupling strengths denoted by M_0 and M_2 . The second term is, however, nondiagonal in both spin and orbital basis and arises from a combined effect of the magnetization and atomic SOC. Specifically, M_N is given by

$$\begin{aligned} K_+ : M_N &= \frac{\sqrt{3}\lambda^2 M_1}{\sqrt{2}(\epsilon_{+1} - \epsilon_0)(\epsilon_{+1} - \epsilon_{+2})}, \\ K_- : M_N &= \frac{\sqrt{3}\lambda^2 M_1}{\sqrt{2}(\epsilon'_{-1} - \epsilon'_0)(\epsilon'_{-1} - \epsilon'_{-2})}, \end{aligned} \quad [4]$$

where M_1 stands for the strength of exchange coupling for $d_{\pm 1}$ orbitals, and ϵ_m/ϵ'_m ($m=0, \pm 1, \pm 2$) is the on-site energy of the d_m orbital at K_+/K_- when the SOC and the magnetization are absent, with $\epsilon_m = \epsilon'_{-m}$ owing to time-reversal symmetry. As a result, M_N is valley-independent. The λ^2 factor in M_N suggests this term is due to second-order processes of the atomic SOC. That is, $d_0^{\uparrow}$ and $d_{+2}^{\downarrow}$ ($d_0^{\downarrow}$ and $d_{-2}^{\uparrow}$) states are coupled with $d_{+1}^{\downarrow}$ and $d_{-1}^{\uparrow}$ ($d_{-1}^{\downarrow}$ and $d_{+1}^{\uparrow}$) states at K_+ (K_-) valley, respectively, by the ladder operations of the SOC (38, 39). The d_{+1} (d_{-1}) states with opposite spins are further coupled by the in-plane exchange interaction. The process is summarized as

$$\begin{aligned} K_+ : d_0^{\uparrow} &\xrightarrow{\text{SOC}} d_{+1}^{\downarrow} \xleftarrow{\text{EI}} d_{+1}^{\uparrow} \xrightarrow{\text{SOC}} d_{+2}^{\downarrow}, \\ K_- : d_0^{\downarrow} &\xrightarrow{\text{SOC}} d_{-1}^{\uparrow} \xleftarrow{\text{EI}} d_{-1}^{\downarrow} \xrightarrow{\text{SOC}} d_{-2}^{\uparrow}. \end{aligned} \quad [5]$$

Since the M_N term mixes different orbitals with opposite spins, it converts the anisotropy in the spin space arising from the in-plane magnetization to the orbital space, which is reflected by an anisotropic low-energy band structure with the rotational symmetry breaking at each valley (see Fig. 3D). In contrast, all bands are isotropic in the minimal model with respect to corresponding Brillouin zone corners if $M_N=0$, irrespective of the first term in H_{EI} .

Based on our analysis above, the rotational symmetry breaking in the orbital space is the ultimate reason for the out-of-plane CISP. We nevertheless give a more intuitive explanation by the semiclassical Boltzmann formalism. In a semiclassical Boltzmann formalism, the CISP is obtained as $\delta s = \frac{1}{S} \sum_{n\mathbf{k}} \langle s \rangle_{n\mathbf{k}} \delta f_{n\mathbf{k}}$. $\delta f_{n\mathbf{k}}$ is the nonequilibrium part of the single-particle distribution function and is related to the Kubo formula result through $\delta f_{n\mathbf{k}} = \frac{eE\hbar}{2\pi} \langle \mathbf{v} \rangle_{n\mathbf{k}} \text{Re}(G_{n\mathbf{k}}^A G_{n\mathbf{k}}^R - G_{n\mathbf{k}}^R G_{n\mathbf{k}}^A)$. It can be roughly understood as the change to the equilibrium Fermi-Dirac distribution function under a momentum shift proportional to the electric field,

Fig. 3. Momentum-resolved electronic structures near $K_{\pm}$ valleys. (A and B) Schematics of the carrier redistribution when $\mathbf{E} \parallel x$ and $\mathbf{E} \parallel y$, respectively. (C) $\langle s_z \rangle$ texture. (D) Band-energy contours with respect to the band-edge energy. (E and F) $\delta f_{\mathbf{k}}$ for two electric-field directions. (G and H) $\delta s_{z,\mathbf{k}}$ for two electric-field directions. In C–H, Left and Right correspond to the K_- and K_+ valley, with the gray dot denoting the K_- and K_+ point. The highest energy band is chosen with more eccentric distributions of $\langle s_z \rangle$ and band energy. All model parameters can be found in SI Appendix, and a larger M_N is adopted to amplify its role compared with the fitted value.

which is schematically illustrated in Fig. 3A and B. To understand why δs_z is nonzero, we plot $\langle s_z \rangle_{\mathbf{k}}$, $\epsilon_{\mathbf{k}}$, $\delta f_{\mathbf{k}}$, and $\delta s_{z,\mathbf{k}}$ of the highest band in Fig. 3C–H. Due to the intrinsic $H_{\text{SOC}} \propto s_z$, $\langle s_z \rangle_{\mathbf{k}}$ appears despite the dominant in-plane components induced by the in-plane magnetization, and it is opposite at the two valleys (Fig. 3C). Moreover, due to the M_N term in H_{EI} (Eqs. 3d and 5), $\langle s_z \rangle_{\mathbf{k}}$ becomes eccentric with respect to the K_{γ} point at a given valley but in a way different from that of the band energy (Fig. 3D). It also exhibits the rotational symmetry breaking. As a result, sampling $\langle s_z \rangle_{\mathbf{k}}$ by the $\delta f_{\mathbf{k}}$ in the neighborhood of the Fermi surface determined by $\epsilon_{\mathbf{k}}$ gives a nonzero result at each valley when $\mathbf{E} \parallel x$. Given that two valleys are related by the vertical mirror σ_v , both $\langle s_z \rangle_{\mathbf{k}}$ and $\delta f_{\mathbf{k}}$ are mirror-antisymmetric by comparing their values in two valleys, which ensures that their product $\delta s_{z,\mathbf{k}}$ is mirror-symmetric and two valleys equally contribute to a nonzero total δs_z (Fig. 3C, E, and G). When $\mathbf{E} \parallel y$, the total δs_z is vanishing by combining antisymmetric $\langle s_z \rangle_{\mathbf{k}}$ and symmetric $\delta f_{\mathbf{k}}$ under the σ_v symmetry (Fig. 3C, F, and H). Moreover, the vertical CISP induced by low-symmetric out-of-plane spin splitting applies to other regions of the momentum space, e.g., Γ valley that participates in transport with further hole doping (SI Appendix, Figs. S4 and S5). Besides, the degree

of the deviation from the rotational symmetry in the band structure and spin texture determines the size of out-of-plane CISP. VTe₂ has more considerable derivation in its electronic structure, leading to a larger CISP, compared with VSe₂ (*SI Appendix*, Fig. S6).

The Magnetization Dynamics Induced by the CISP. The magnetization-dynamics calculations are further performed to study the roles of the above magnetization-orientation-dependent CISPs in the dynamics, according to the Landau–Lifshitz–Gilbert equation (5). Given that the monolayer plane is the easy plane, we consider the dynamics with the initial magnetization along the x direction, under the action of $\mathbf{E} \parallel x$. The effective magnetic field and associated spin–orbit torque arises from three contributions, i.e., the intraband CISP, interband CISP, and uniaxial magnetocrystalline anisotropy. By sweeping the strength ratios of the CISP effective fields to the anisotropy field, a phase diagram of the elevation angle of the final magnetization together with representative trajectories of the magnetization are given in Fig. 4, as a function of B_{intra}^0/B_u^0 and B_{inter}^0/B_u^0 . Here, $B_{\text{intra}}^0/\text{inter}$ is an initial effective field of intraband/interband CISP with the magnetization along the x direction, and B_u^0 is the strength of the magnetocrystalline anisotropy field.

The magnetization dynamics can lead to various final resting states and steady-state precessions, which are respectively outside and inside the region surrounded by the green bound-

ary in Fig. 4A. For final resting states, the magnetization can eventually come to rest on the equator, upper hemisphere and lower hemisphere of the unit sphere representing the magnetization direction after the damping motion, as shown in Fig. 4B. For the steady-state precession, the precession axis can be the poles as shown in Fig. 4C and D or the equator. The dynamics is very complicated due to the magnetization-orientation dependency of the CISPs. The out-of-plane resting magnetization and the precession around the two poles have nonvanishing (average) elevation angle, $\xi = \pi/2 - \theta$, measured from the equator plane. It is found that there are considerable out-of-plane magnetization rotations with large ξ in a sizable parameter space. Therefore, the CISPs proposed above are very likely to induce out-of-plane magnetization rotation. Since ξ is zero near the B_{inter}^0/B_u^0 axis, a relatively large ratio of B_{intra}^0 to B_{inter}^0 is preferred for realizing considerable out-of-plane magnetization rotation.

We further discuss the material realization of the magnetization dynamics in ferromagnetic transition-metal dichalcogenides. For VSe₂, the hard axis (out-of-plane) anisotropy field is about 20 T, and B_{intra}^0/B_u^0 is in the order of 0.01 for an electric field of the order of 10^{-3} V/Å. An in-plane rotation occurs in VSe₂ with $\xi = 0$ according to Fig. 4A. However, for VTe₂, the B_{intra}^0/B_u^0 can increase up to ~ 1 for an electric field of the order of 10^{-3} V/Å. The out-of-plane magnetization rotation with ξ of tens of degrees and corresponding electric control of valley splitting are likely to be realized in VTe₂ (*SI Appendix*). Even though the CISPs cannot tilt the magnetization itself out-of-plane, an available effective field of the order of 0.1 T is large enough to be observed experimentally. A heterostructure of VSe₂ and a ferromagnet with a relatively weak anisotropy field of the orders of 10 to 10^2 mT may be constructed to exhibit the role of the CISP by the magnetization rotation of the adjacent ferromagnet and the technique of the spin–torque ferromagnetic resonance (*SI Appendix*). Besides, the CISPs can be increased by, e.g., applied strain, as demonstrated in *Discussion*.

Discussion

We further study the CISP of uniaxially strained VSe₂ monolayer, where the C_3 symmetry is broken by both the magnetization and the crystalline symmetry reduction. For the uniaxial strain, the crystal point group is reduced to C_{2v} , but two valleys are still well preserved (Fig. 1C and D). The symmetry breaking in the orbital space enhances the out-of-plane CISP compared with the unstrained case (*SI Appendix*), which can help the out-of-plane magnetization rotation. Besides, the vertical CISP is nonvanishing for all magnetization directions including perpendicular to the VSe₂ sheet.

It is noted that the symmetry breaking in the spin space that enables the overall symmetry reduction and the creation of out-of-plane CISP in pristine ferromagnetic transition-metal dichalcogenides monolayer, while the uniaxial strain can further enhance the out-of-plane CISP. In contrast, the out-of-plane CISP arises from only the crystalline symmetry breaking in previous studies (7, 11–15). The CISP induced by the time-reversal symmetry breaking has distinct characteristics. The intraband CISP demonstrates a second-order dependence on the direction of the magnetization. The out-of-plane CISP needs the combination role of both a nonvanishing in-plane magnetization component and intrinsic SOC. When the magnetization is parallel to the monolayer plane, the out-of-plane CISP is the only component owing to higher crystalline symmetry, compared with the presence of more nonequilibrium spin components in MX₂ with lower crystalline symmetry (7, 11–14).

Although we have been focusing on the VSe₂ and VTe₂ monolayers that are indeed promising experimental systems, it is worth noting that a large family of 2D ferromagnets with the

Fig. 4. The magnetization dynamics under the action of effective fields arising from CISPs. (A) The elevation angle as a function of the reduced field strengths, B_{intra}^0/B_u^0 and B_{inter}^0/B_u^0 . (B–D) Representative trajectories of the magnetization dynamics on unit sphere representing the magnetization direction. The resting magnetizations are realized in B, with $(B_{\text{intra}}^0/B_u^0, B_{\text{inter}}^0/B_u^0)$ being (1.6, 0), (0, 0.6), and (1.6, 0.6) for blue, black, and red trajectories, respectively. The steady-state precessions around two poles are given in C and D, with reduced strengths of (1.6, −0.6) and (0.6, −0.8), respectively.

symmetry breaking in the spin space can also generate out-of-plane CISP, e.g., VS_2 (23), NbX_2 (40), TaX_2 (41), group-III monochalcogenides (42), transition metal-doped MX_2 (19), etc. On the other hand, the low-energy Hamiltonian proposed here is also expected to become an effective model like the Rashba model when it comes to discussing out-of-plane CISP and other spin-related physics.

Finally, we did not consider higher-order scattering processes such as the vertex correction (4, 25, 43), with the expectation that they will not change the main conclusions of this work qualitatively. As for VSe_2 and VTe_2 , the band splittings induced by the SOC have magnitudes of 0.08 and 0.1 eV in the low-energy bands at $K_{\pm}$ valleys (*SI Appendix, Fig. S3*), respectively, while the band broadening due to the disorder potential, $\eta = 0.01$ eV, is used in our calculation, and it represents the disorder strength. Since the ratio of the disorder strength to the SOC is ~ 0.1 and the SOC induced band splitting is well defined under the action of disorders, our calculation of the CISP in the ferromagnetic transition-metal dichalcogenides are done in the weak disorder regime for which the finite lifetime approximation is appropriate for giving a good qualitative description of nonequilibrium responses. In contrast, the vertex correction is indispensable in the diffusive transport regime with a large strength ratio of the disorder to SOC (44). Nevertheless, quantitative dependence of the out-of-plane CISP on different impurity types, concentrations, and distributions is interesting and worth further study, by, e.g., using Keldysh nonequilibrium Green's function formalism in a two-terminal device (10).

Methods

We study geometric and electronic structures of the VSe_2 and VTe_2 monolayers by density functional theory calculations, which are implemented in

the Vienna Ab initio Simulation Package (45, 46). Here, the projector augmented wave potentials (46) and the Perdew–Burke–Ernzerhof exchange-correlation functional (47) are used. A plane-wave cutoff of 400 eV and a k -mesh of $12 \times 12 \times 1$ are adopted. Vacuum slabs more than 16 Å thick are inserted to minimize the interaction between the monolayer and its periodic images. Structure optimizations are performed with a convergence threshold of 0.01 eV/Å on the interatomic forces. The SOC is included in the calculation of electronic structure, unless otherwise specified.

The ab initio tight-binding model is then constructed based on Wannier functions with first-principles input (48, 49) to calculate the CISP. The Wannier basis set consists of 11 orbitals, i.e., five d -orbitals of one vanadium atom and six p -orbitals of two chalcogen atoms in a unit cell. We perform a real-space truncation of the spinless Hamiltonian up to fifth-nearest neighbors and symmetrize corresponding hopping and on-site energies (38, 39). The symmetrized Hamiltonian reproduces the first-principles bands very well (*SI Appendix, Fig. S3*), and the symmetrization ensures that symmetry-disallowed components of CISP are vanishing. The SOC is further incorporated by operating on-site $\lambda' \hat{\mathbf{L}} \cdot \hat{\mathbf{s}}$ term on atomic orbitals (38, 39), with $\hat{\mathbf{L}}$ ($\hat{\mathbf{s}}$) being the orbital (spin) angular momentum operator. The magnetization is taken into account by the exchange interaction, $m(s_x \sin \theta \cos \phi + s_y \sin \theta \sin \phi + s_z \cos \theta)$. The above strength parameters, λ' and m , are element-dependent and orbital-dependent, respectively. They are obtained by comparing spinless and spinful first-principles calculations.

Data Availability

The data supporting the findings and the parameters for reproducing the calculated results are included in the main text and *SI Appendix*.

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