

Magnetization-dependent and stacking-tunable Edelstein effect in the two-dimensional magnet $2H\text{-VTe}_2$

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The Edelstein effect in magnetic systems enables magnetization switching via the coupling between current-induced spin accumulation and intrinsic magnetic order and is therefore highly promising for next-generation spintronic devices. Realizing and manipulating the Edelstein effect in two-dimensional (2D) magnetic systems is particularly desirable for achieving high-efficiency and multifunctional spintronic applications. In this work, based on first-principles calculations and symmetry analysis, we demonstrate that the Edelstein effect can intrinsically arise in the 2D in-plane ferromagnetic semiconductor $2H\text{-VTe}_2$, with its behavior strongly dependent on the magnetization orientation. For monolayer $2H\text{-VTe}_2$ with D_{3h} crystal symmetry, under an applied current along the $+x$ direction, only the time-reversal-even z component and the time-reversal-odd $y(x)$ component of the spin accumulation are allowed when the magnetization is aligned along $+x$ ($+y$). For ferromagnetic bilayer $2H\text{-VTe}_2$ in AB or BA stacking, where the crystal symmetry is reduced to C_{3v} , additional spin components emerge with the presence of in-plane magnetization. Specifically, for magnetization along $+x$ ($+y$), besides dS_z^{even} and dS_y^{odd} (dS_z^{even} and dS_x^{odd}), extra components such as dS_y^{even} and dS_z^{odd} (dS_y^{even}) become allowed. Notably, these additional components can be reversibly switched by changing the stacking configuration from AB to BA via interlayer sliding, which is equivalent to applying a $M_z T$ operation. The parity dependence of Edelstein coefficients on the in-plane magnetization is also evaluated. Our results not only deepen the understanding of current-induced spin accumulation in 2D magnetic systems from both symmetry and first-principles perspectives but also identify $2H\text{-MX}_2$ materials as a promising platform for realizing intrinsic and tunable Edelstein effects in high-efficiency spin-orbit torque devices.

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I. INTRODUCTION

In noncentrosymmetric systems with strong spin-orbit coupling (SOC), an electric current can induce a nonequilibrium spin accumulation, known as the Edelstein effect [1,2]. In magnetic systems, such current-induced spin accumulation can exert spin-orbit torques on the intrinsic magnetic moments via exchange coupling [3], thereby enabling electrical manipulation of the magnetization orientation [4,5]. This mechanism further underpins various spintronic applications, such as magnetoresistive random-access memory based on magnetization switching and racetrack memory based on domain-wall motion [6–8]. A straightforward strategy to realize the Edelstein effect in magnetic systems is to construct heterostructures composed of ferromagnetic (FM) metals and nonmagnetic materials with strong SOC. In such systems, inversion symmetry is broken at the interface, giving rise to a nontrivial Edelstein effect [9–15]. However, in heterostructures with finite thickness, the interaction between current-induced spin accumulation and localized magnetic moments is typically confined near the interface [9,10,16], rather than extending throughout the entire system, thereby reducing the efficiency of the resulting spin-orbit torque.

Moreover, the finite thickness of such heterostructures not only complicates fabrication but also limits device integration density. Therefore, it is highly desirable to realize the Edelstein effect in intrinsically atomic-thin two-dimensional (2D) magnetic systems with broken inversion symmetry, where the interaction between current-induced spin accumulation and magnetic moment can extend across the entire material. Such systems can not only enhance the efficiency of current-driven magnetic control but also improve the device integration density.

Beyond realizing the intrinsic Edelstein effect in noncentrosymmetric 2D magnets, another key objective is its controllable manipulation, which is essential for tunable spintronic devices. Since the Edelstein effect is fundamentally governed by crystal symmetry, the individual components of current-induced spin accumulation can in principle be selectively activated, suppressed, or even reversed through deliberate symmetry engineering [17–20]. This tunability enables possible diversity in spin-orbit torque behaviors and thus provides a versatile platform for multifunctional spintronic applications. In 2D magnetic systems, crystal symmetry can be effectively engineered via bilayer stacking [21–26], which often reduces the symmetry of the whole system compared to the monolayer case and thereby introduces additional components of the Edelstein effect. Moreover, electrical switching between distinct bilayer stacking configurations can

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be achieved through sliding ferroelectricity [25,27], which is in principle able to switch the sign of Edelstein effect and thus offering a promising route toward nonvolatile spintronic devices [28]. However, such stacking-dependent Edelstein effect in 2D magnets remains largely unexplored, thereby motivating further investigation.

Two-dimensional FM transition-metal compounds MX_2 (M = magnetic elements, X = nonmagnetic elements) in the $2H$ phase (e.g., VSe_2 [29–34], VTe_2 [32,34,35], $FeCl_2$ [36–38], and $FeBr_2$ [37,39]), which have attracted considerable attention due to their rich magnetism-related properties, provide a simple yet powerful platform for addressing the aforementioned two objectives. In particular, their crystal symmetry belongs to the D_{3h} point group, which intrinsically lacks inversion symmetry and further allows the emergence of the current-induced spin accumulation in the presence of magnetism and SOC [40–42]. Moreover, the electronic structure of $2H-MX_2$ is strongly coupled to magnetism, giving rise to a variety of magnetization-dependent physical properties, such as ferrovalley effects [30,37], multiferroicity [32,43], and magnetic topological states [36,38]. Accordingly, the Edelstein effect in these systems is expected to be strongly dependent on the magnetization orientation, likely governed by the corresponding magnetic group symmetry, and thus warrants further systematic investigation. Furthermore, when $2H-MX_2$ layers are stacked into bilayers with specific polar stackings, ferroelectric polarization can emerge [35,44,45], enabling electrical switching between these two distinct stacking configurations. Such switching may lead to a reversal of the Edelstein effect, which calls for a detailed symmetry-based analysis.

In the present work, we focus on $2H-VTe_2$. To date, two distinct phases of VTe_2 have been reported or proposed. One is the experimentally accessible metallic 1T phase, which exhibits, in few-layer samples, charge-density-wave distortions at low temperatures [46–48]. The other is the theoretically predicted [49,50] $2H$ phase, which represents an exciting target for experimental realization owing to the variety of intriguing properties expected for it [32,35,50–52]. In particular, monolayer VTe_2 is predicted to be a magnetic semiconductor which can host valley polarization and higher-order topological phase [50–52]. Magnetism-induced ferroelectricity can emerge upon changing the magnetization direction, making $2H-VTe_2$ a rare example of a type-II multiferroic system [32]. Moreover, sliding ferroelectricity has been predicted in bilayer $2H-VTe_2$, enabling the valley polarization to be controlled by reversing the electric polarization through stacking changes [35]. It is therefore of interest to theoretically explore the magnetic- and stacking-dependent Edelstein effect in $2H-VTe_2$, which may help to reveal further exotic phenomena in this system and provide guidance for future experimental studies.

In this work, using first-principles calculations combined with symmetry analysis, we systematically investigate the Edelstein effect and its relationship with magnetic symmetry in $2H-VTe_2$, which is known to be a ferromagnetic semiconductor in the family of $2H-MX_2$ [32,34]. For monolayer $2H-VTe_2$, we find that in-plane magnetization lowers the symmetry, thereby giving rise to a nontrivial Edelstein effect. Notably, this effect strongly depends on the magne-

tization orientation. For example, when the magnetization is aligned along the $+x$ ($+y$) direction, only the time-reversal-even z component and the time-reversal-odd y component (the time-reversal-even z component and the time-reversal-odd x component) of the spin accumulation are allowed by magnetic symmetry under an electric field applied along the $+x$ direction. For ferromagnetic bilayer $2H-VTe_2$ in AB and BA stacking configurations, additional components of current-induced spin accumulation emerge due to the further reduction of crystal symmetry, some of which can be reversed upon switching between AB and BA stacking. Moreover, rotating the in-plane magnetization leads to symmetry-constrained angular dependence of the spin accumulation in both monolayer and bilayer systems. Our results not only provide symmetry-based insights into charge-spin conversion in 2D magnetic systems but also expand the range of candidate materials for high-efficiency spin-orbit torque devices.

II. METHODS

First-principles calculations are performed using the QUANTUM ESPRESSO package [53]. The Perdew-Burke-Ernzerhof exchange-correlation functional and projector augmented-wave potentials are employed [54,55]. Both structural relaxations and self-consistent calculations are carried out using an $18 \times 18 \times 1k$ -point mesh, with plane-wave energy cutoffs of 60 Ry for the wave functions and 700 Ry for the charge density. Structural relaxations are performed using scalar-relativistic pseudopotentials and the quasi-Newton algorithm until all force components are smaller than 2×10^{-4} Ry/ a_0 , where a_0 denotes the Bohr radius. Grimme D-2 van der Waals corrections is adopted during the relaxation [56,57]. Subsequently, self-consistent calculations are conducted with fully relativistic pseudopotentials to include SOC. A vacuum region of at least 20 Å is introduced to eliminate spurious interactions between periodic images in the slab geometry. To account for the on-site Coulomb interaction of the V-3d orbitals, a Hubbard U parameter of 1 eV is applied, consistent with previous studies [35]. This choice is justified by the fact that $U=1$ eV reproduces the structural and magnetic properties of the closely related $2H-VSe_2$ in good agreement with HSE06 calculations [58], while a recent constrained random-phase approximation study also reported an effective U of approximately 1.5 eV for monolayer $2H-VTe_2$ [32], which is close to 1 eV. Therefore, $U = 1$ eV represents a reasonable and physically motivated choice for the present system. Fully self-consistent calculations for each magnetization angle is individually performed.

After the density functional theory (DFT) calculations, we construct the Wannier Hamiltonian using the Wannier90 package [59]. In the Wannierization procedure, the V- d orbitals and Te- p orbitals are chosen as the projection basis. To ensure that the symmetries of the Wannier functions match those of the system as closely as possible, we do not perform maximal localization. Based on the resulting Wannier Hamiltonian, the WannierTools package is employed to compute the Fermi surface [60]. Furthermore, the Edelstein coefficient is evaluated using the Linres code within the framework of the Kubo formula [61]. Specifically, we consider the linear response of

spin accumulation to an external electric field, expressed as $\delta\mathbf{S} = \chi\mathbf{E}$, where $\delta\mathbf{S}$ denotes the induced spin accumulation, $\mathbf{E}$ is the applied electric field, and χ is the Edelstein response tensor. Within the Kubo formalism, χ can be decomposed into time-reversal-even and time-reversal-odd contributions, respectively [11,62]:

$$\chi_{ij}^{\text{even}} = -\frac{e\hbar}{\pi N} \sum_{\mathbf{k}, m, n} \frac{\Gamma^2 \text{Re}(\langle n\mathbf{k} | \hat{S}_i | m\mathbf{k} \rangle \langle m\mathbf{k} | \hat{v}_j | n\mathbf{k} \rangle)}{[(E_f - \epsilon_{n\mathbf{k}})^2 + \Gamma^2][(E_f - \epsilon_{m\mathbf{k}})^2 + \Gamma^2]}$$

$$\chi_{ij}^{\text{odd}} = \frac{2e\hbar}{N} \sum_{\mathbf{k}, n \neq m}^{n \text{ occ.}, m \text{ unocc.}} \text{Im}[\langle n\mathbf{k} | \hat{S}_i | m\mathbf{k} \rangle \langle m\mathbf{k} | \hat{v}_j | n\mathbf{k} \rangle]$$

$$\times \frac{\Gamma^2 - (\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}})^2}{[(\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}})^2 + \Gamma^2]^2}. \quad (1)$$

Here e is the elementary charge; n, m denote band indices; $\mathbf{k}$ is the Bloch vector; N is the total number of k points used to sample the Brillouin zone; E_f is the Fermi energy; $\mathbf{V}$ is the velocity operator; $\epsilon_{n\mathbf{k}}$ is the eigenvalue; and Γ is the disorder parameter, which is related to the relaxation time τ through: $\tau = \hbar/2\Gamma$. It should be noted that the time-reversal-even component of the Edelstein effect originates solely from electronic states at the Fermi surface and is also termed as “intraband” contribution. This can be understood more clearly in the limit of a small broadening parameter Γ , where the corresponding expression can be approximated as [11]

$$\chi_{ij}^{\text{even}} = -\frac{e\hbar}{2\Gamma N} \sum_{\mathbf{k}, n} \delta(\epsilon_{n\mathbf{k}} - E_f) \langle n\mathbf{k} | \hat{S}_i | n\mathbf{k} \rangle \langle n\mathbf{k} | \hat{v}_j | n\mathbf{k} \rangle, \quad (2)$$

where the δ -function explicitly indicates that only states at the Fermi level contribute to the Edelstein effect. Meanwhile, the time-reversal-odd part of Edelstein coefficient, which is also termed as “interband” contribution, goes to the Γ -independent intrinsic formula when Γ is very small [11]:

$$\chi_{ij}^{\text{odd}} = -\frac{2e\hbar}{N} \sum_{\mathbf{k}, n \neq m}^{n \in \text{occ.}, m \in \text{unocc.}} \frac{\text{Im}[\langle n\mathbf{k} | \hat{S}_i | m\mathbf{k} \rangle \langle m\mathbf{k} | \hat{v}_j | n\mathbf{k} \rangle]}{(\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}})^2}. \quad (3)$$

In this work, for a clear illustration of the current-induced spin accumulation, we restrict the direction of electric field along the $+x$ direction, i.e., $j = x$. In this case, the calculated Edelstein coefficient components χ_{ix} is directly associated with the current-induced spin accumulation components in a unit cell along the i ($i = x, y, z$) direction, δS_i . During the calculation, we set $\Gamma = 10$ meV, which roughly corresponds to a typical sub-100 fs scattering rate (broadening) while remaining much smaller than interband separations (see Appendix B for more details). $600 \times 600 \times 1k$ points are adopted to arrive at the converged Edelstein coefficient.

III. EDELSTEIN EFFECT IN MONOLAYER 2H-VTe₂

The crystal structure of monolayer 2H-VTe₂ is shown in Fig. 1(a), which crystallizes in space group No. 187 (D_{3h} point group). Previous studies have predicted that monolayer 2H-VTe₂ is a stable ferromagnetic (FM) semiconductor [32,34,49]. As illustrated, the V atoms form a triangular lattice, with a nearest-neighbor V-V distance of 3.61 Å after

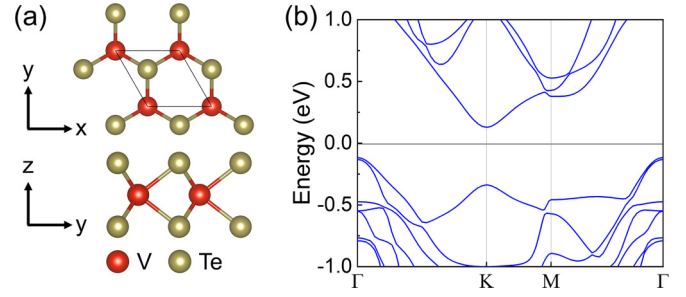


FIG. 1. (a) The atomic structure of monolayer 2H-VTe₂. (b) The band structure of monolayer 2H-VTe₂.

full structural relaxation. Each V atom is coordinated by six Te atoms, forming a local trigonal prismatic environment. This trigonal crystal field splits the V 3d orbitals into three groups: the d_{z^2} orbital (denoted as A_1), the degenerate d_{xy} and $d_{x^2-y^2}$ orbitals (denoted as E_1), and the d_{xz} and d_{yz} orbitals (denoted as E_2). The calculated magnetic moment is $1 \mu_B$ per unit cell, consistent with the $3d^1$ electronic configuration of each V^{4+} ion. Furthermore, the magnetic anisotropy energy (MAE) is evaluated as $E_x - E_z = E_y - E_z = -2.07$ meV per V atom, indicating that the in-plane magnetization is energetically favored over the out-of-plane orientation. This result is consistent with previous reports [32,34]. Therefore, in the following, we focus on the in-plane magnetization states of monolayer 2H-VTe₂. The electronic band structure of monolayer 2H-VTe₂ is presented in Fig. 1(b). The valence band maximum (VBM) is located at the Γ point, while the conduction band minimum (CBM) resides at the K point, resulting in an indirect band gap of approximately 0.23 eV.

Now let us qualitatively explore the Edelstein effect in monolayer 2H-VTe₂ from the viewpoint of symmetry. Considering only the D_{3h} crystal symmetry of monolayer 2H-VTe₂ without the inclusion of magnetism, the Edelstein effect is forbidden. However, the introduction of in-plane magnetization breaks the D_{3h} symmetry, thereby allows a finite current-induced spin accumulation. For instance, when the magnetization is aligned along the $+x$ direction, the magnetic point group reduces to $m'm'2'$, with symmetry generators $\{E, M_x, C_{2y}T, M_zT\}$. Here E denotes identity operation, T denotes time-reversal operation, M_α ($\alpha = x, y, z$) denotes the mirror operation with the normal of mirror plane along the α direction, while $C_{2\alpha}$ ($\alpha = x, y, z$) denotes the twofold rotation operation with respect to α axis. Analysis of these symmetry operations shows that the M_x operator reverses the sign of χ_{xx}^{even} and χ_{xx}^{odd} , while $C_{2y}T$ reverses χ_{yx}^{even} , and M_zT reverses χ_{zx}^{odd} . Consequently, χ_{xx}^{even} , χ_{xx}^{odd} , χ_{yx}^{even} , and χ_{zx}^{odd} are forbidden under the $m'm'2'$ symmetry. In contrast, χ_{zx}^{even} and χ_{yx}^{odd} remain invariant under all symmetry operations and are therefore allowed. As a result, when the magnetization lies along the $+x$ direction, only δS_z^{even} and δS_y^{odd} are induced under an electric field applied along the $+x$ direction. Similarly, when the magnetization is aligned along the $+y$ direction, the magnetic point group becomes $m'm'2$, with symmetry operators $\{E, C_{2y}, M_xT, M_zT\}$. Symmetry analysis yields the following constraints: (i) M_xT forbids χ_{xx}^{even} and χ_{yx}^{odd} , (ii) M_zT forbids χ_{zx}^{odd} , and (iii) C_{2y} forbids χ_{yx}^{even} . Meanwhile, χ_{zx}^{even}

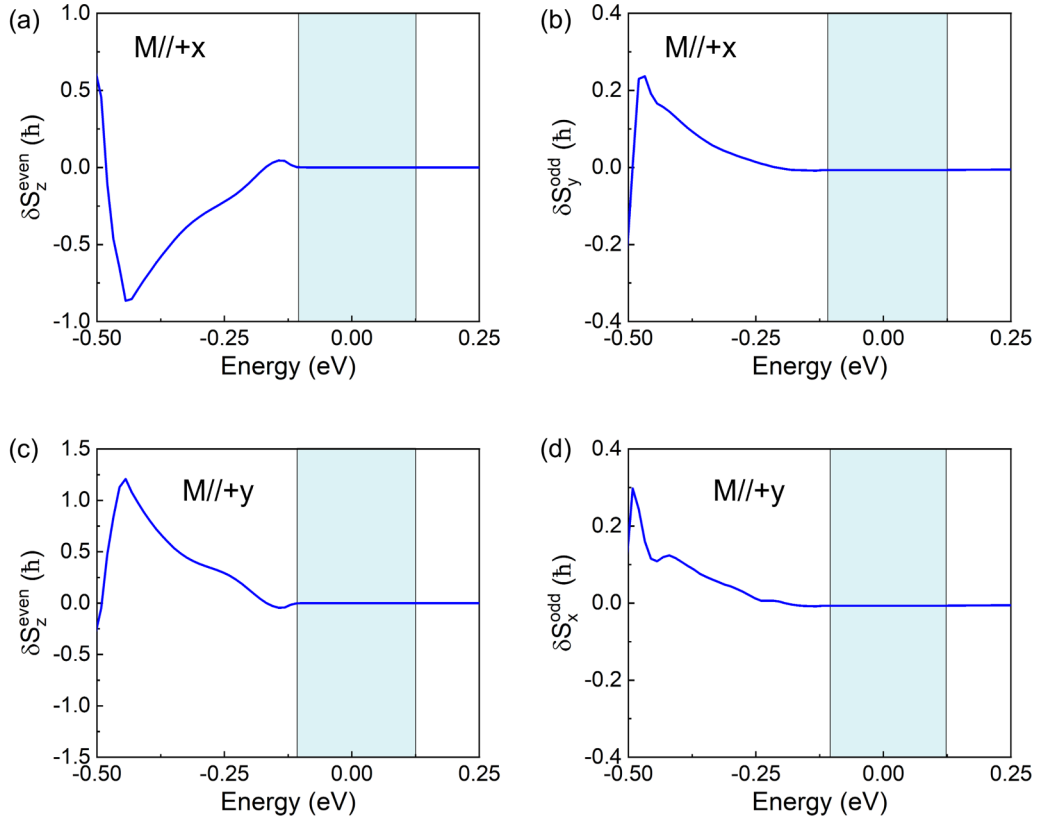


FIG. 2. Calculated symmetry-allowed current-induced spin accumulations in a unit cell as functions of Fermi energy in monolayer $2H$ -VTe $_2$ under a reference electric field $1 \text{ V/\AA}$ along the $+x$ direction. Panels (a) and (b) show the time-reversal-even (δS_z^{even}) and time-reversal-odd (δS_y^{odd}) components of spin accumulation when magnetization is along the $+x$ direction. Panels (c) and (d) show the time-reversal-even (δS_z^{even}) and time-reversal-odd (δS_x^{odd}) components of spin accumulation when magnetization is along the $+y$ direction. The dashed blue region indicates the band gap. Note that the magnitude of the electric field ($1 \text{ V/\AA}$) is difficult to realize experimentally and is used only as a reference value.

and χ_{xx}^{odd} remain invariant under all symmetry operations and are thus allowed. Therefore, for magnetization along the $+y$ direction, only δS_z^{even} and δS_x^{odd} are induced under an electric field applied along the $+x$ direction. Notably, the above symmetry-based analysis is general and can be extended to other $2H$ -MX $_2$ systems with in-plane magnetization.

To gain quantitative insights into the Edelstein effect in monolayer $2H$ -VTe $_2$, we calculate the symmetry-allowed components of current-induced spin accumulation per unit electric field ($\text{V/\AA}$) as a function of the Fermi energy, as shown in Fig. 2. We first consider the case where the magnetization is aligned along the $+x$ direction, for which only δS_z^{even} and δS_y^{odd} are allowed [see Fig. 2(a) and 2(b)]. For δS_z^{even} , its magnitude increases as the Fermi energy shifts downward toward the valence band, reaching a maximum value of about $-0.86 \hbar$ at $E_F = -0.44 \text{ eV}$ (approximately 0.3 eV below the VBM), which is microscopically dominated by the competing contribution from the states near Γ point (see Fig. 9 in Appendix D for more details). Upon further decreasing the Fermi energy, δS_z^{even} decreases in magnitude and changes sign around $E_F = -0.5 \text{ eV}$. The behavior of δS_y^{odd} is qualitatively similar, exhibiting an initial increase followed by a decrease and sign reversal as the Fermi level shifts downward, and the local maxima is dominated by the states near the Γ point

(see Appendix D for more details about microscopic origin). However, two key differences can be identified: (i) δS_z^{even} and δS_y^{odd} have opposite signs within the valence band region; (ii) the maximum magnitude of δS_y^{odd} (about $0.23 \hbar$) is significantly smaller than that of δS_z^{even} (about $0.86 \hbar$). Similarly, when the magnetization is aligned along the $+y$ direction, the symmetry-allowed components δS_z^{even} and δS_x^{odd} are shown in Figs. 2(c) and 2(d). As the Fermi energy shifts downward, both components remain positive and increase in magnitude, reaching their respective maxima with values of approximately $1.2 \hbar$ for δS_z^{even} at $E_F = -0.44 \text{ eV}$ and about $0.3 \hbar$ for δS_x^{odd} at $E_F = -0.49 \text{ eV}$. Notably, for magnetization oriented along either $+x$ or $+y$, both the time-reversal-odd and time-reversal-even spin accumulations are nearly zero in the low-energy conduction-band region. Microscopically, this region is dominated by electronic states near the K points and their symmetry-related partners. The nearly vanishing time-reversal-even spin accumulation arises from an approximate compensation between positive and negative contributions from these states, which is determined by the detailed electronic structure rather than enforced by symmetry (see Fig. 10 for k -resolved time-reversal-even spin accumulation for low-energy conduction bands in Appendix D). Meanwhile, the time-reversal-odd spin accumulation remains negligible

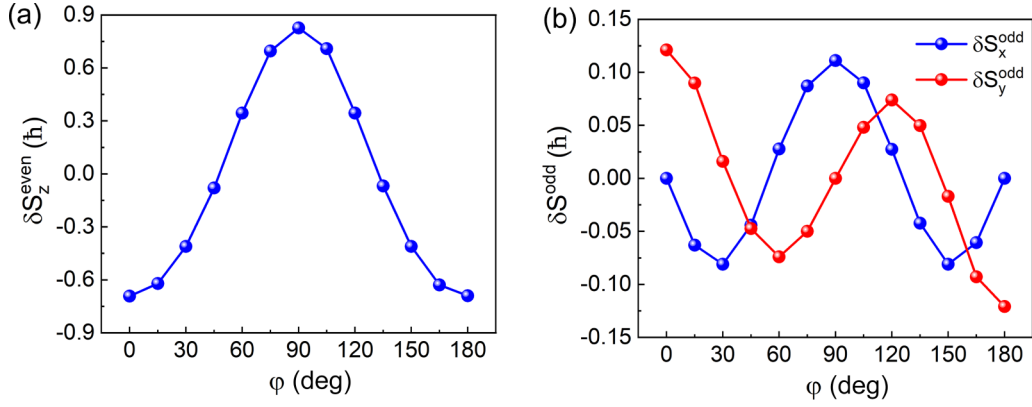


FIG. 3. Angular-dependent spin accumulation in a unit cell of monolayer $2H\text{-VTe}_2$, which is denoted as a function of ϕ (angle between magnetization direction and the $+x$ direction). Panels (a) denotes the symmetry-allowed time-reversal-even components (only δS_z^{even} here) of spin accumulation in a unit cell as function of ϕ , while (b) denotes the symmetry-allowed time-reversal-odd components of spin accumulation in a unit cell as function of ϕ . Note that the magnetization is rotating in plane, which means that $\phi = 0$ and $\phi = 90^\circ$ denotes $+x$ and $+y$ magnetization, respectively. The spin accumulation is calculated at $E_F = -0.4$ eV under a fixed electric field of $1 \text{ V/\AA}$ along the $+x$ direction. Note that this magnitude of electric field is difficult to realize experimentally and is used only as a reference value.

possibly due to the large interband separation at the K point suppresses the corresponding interband contributions to the spin accumulations.

Based on the above analysis, it is evident that the current-induced nonequilibrium spin accumulation in monolayer $2H\text{-VTe}_2$ strongly depends on the magnetization orientation, warranting further investigation. To this end, we evaluate the time-reversal-even and -odd components of the induced spin at $E_F = -0.4$ eV as a function of the in-plane magnetization angle ϕ , as shown in Figs. 3(a) and 3(b). As ϕ varies, only the z component of spin accumulation appears in the time-reversal-even contribution, whereas both x and y components are present in the time-reversal-odd part. When ϕ increases from 0 to 180° , δS_z^{even} first increases, reaching a maximum of approximately $0.9 \hbar$ at $\phi = 90^\circ$, and then decreases symmetrically with respect to $\phi = 90^\circ$. Quantitatively, the δS_z^{even} is roughly proportional to $\cos 2\phi$, which can be understood from the second-order dependence on the magnetization direction of current-induced spin accumulation (see Appendix E). Similarly, δS_x^{odd} initially decreases from zero, then increases to a local maximum of about $0.12\hbar$ at $\phi = 90^\circ$, and subsequently exhibits symmetric behavior about $\phi = 90^\circ$. In contrast, δS_y^{odd} decreases from approximately $0.12\hbar$ to a local minimum of about $-0.08\hbar$ and then increases to zero at $\phi = 90^\circ$, displaying an antisymmetric dependence with respect to $\phi = 90^\circ$. The calculated spin responses, with magnitudes on the order of $0.1 \sim 1\hbar$ per $\text{V/\AA}$, are comparable to those reported in p -wave magnetic systems such as LuFeO_3 [63] and CeNiOAs [64]. By normalizing with the calculated conductivity and assuming an effective thickness of $6 \text{ \AA}$ for the monolayer, we estimate that a current density of 10^7 A/cm^2 induces magnetic moments on the order of 10^{-7} to $10^{-6}\hbar$ per unit cell (see Appendix C). This magnitude is comparable to that of nonmagnetic chiral and ferroelectric systems (e.g., CsGeBr_3 [65], WTe_2 [28], and NbSi_2 [66]) and exceeds that predicted for antiferromagnetic Mn_2Au [42]. To understand the parity dependence of the current-induced spin components on the magnetization angle,

we note that switching the magnetization angle between ϕ to $180^\circ - \phi$ is equivalent to applying the $M_x T$ operation. Under this symmetry, χ_{zx}^{even} and χ_{xx}^{odd} remain invariant, whereas χ_{yx}^{odd} changes sign. Consequently, for an electric field applied along the $+x$ direction, δS_z^{even} and δS_x^{odd} , which are proportional to χ_{zx}^{even} and χ_{xx}^{odd} , respectively, are even functions with respect to $\phi - 90^\circ$, while δS_y^{odd} , proportional to χ_{yx}^{odd} , is an odd function. This analysis establishes a direct connection between the angular dependence of the Edelstein effect and the underlying symmetries and is expected to be generally applicable to other magnetic systems with similar crystal symmetry.

IV. EDELSTEIN EFFECT IN AB- AND BA-STACKED BILAYER $2H\text{-VTe}_2$

The above analysis of monolayer $2H\text{-VTe}_2$ demonstrates that the behavior of the Edelstein effect is strongly governed by the symmetry of the system. By further engineering the crystal symmetry, additional components of the Edelstein effect may emerge, some of which can even be reversibly controlled, offering significant potential for device applications [18,67]. An effective strategy to achieve this is to construct bilayer $2H\text{-VTe}_2$, where the dependence of crystal symmetry—and its constraints on the Edelstein effect—on the stacking configuration warrants systematic investigation. Here we consider two stacking configurations of bilayer $2H\text{-VTe}_2$: AB stacking, in which the V atoms in the top layer align with the upper Te atoms in the bottom layer [Fig. 4(a)], and BA stacking [Fig. 4(b)], where the V atoms in the bottom layer align with the lower Te atoms in the top layer. Previous studies have shown that these two configurations, which are related by an M_z operation, both correspond to local energy minima and can be reversibly switched via sliding ferroelectricity [35]. Based on the previously predicted switching barrier $\Delta E_{\text{barrier}}$ and electric dipole P for bilayer $2H\text{-VTe}_2$ [35], we estimate the critical electric field required for ferroelectric switching, $E_c = \Delta E_{\text{barrier}}/P$, to be on the order of $1\text{--}10 \text{ V/nm}$, which is exper-

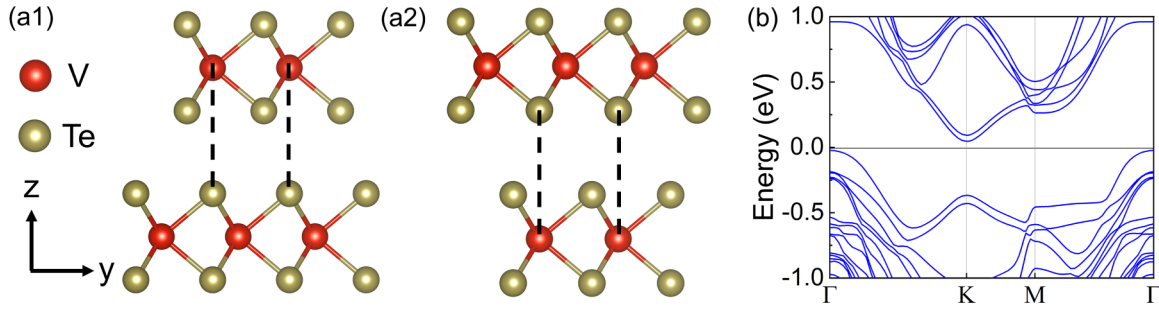


FIG. 4. The atomic structure of bilayer $2H\text{-VTe}_2$ in (a) AB and (b) BA stacking, respectively. (c) The band structure of AB-stacked bilayer $2H\text{-VTe}_2$, which possesses the same qualitative feature (i.e., CBM at K and VBM at Γ) with BA-stacked system.

imentally achievable using piezoresponse force microscopy [68]. It has also been reported that the interlayer magnetic coupling in both AB and BA stackings lies close to the ferromagnetic-antiferromagnetic phase boundary. Moreover, it is predicted that an interlayer compression of approximately 2%, corresponding to a moderate pressure of about 1.2 GPa, can drive AB-stacked bilayer $2H\text{-VTe}_2$ from the interlayer-antiferromagnetic state to the ferromagnetic state [35]. In this work, we focus on the Edelstein effect in the interlayer ferromagnetic configuration of bilayer $2H\text{-VTe}_2$ in order to isolate the role of stacking-dependent crystal symmetry under a fixed magnetic background. The MAE of bilayer $2H\text{-VTe}_2$ is calculated to be $E_x - E_z = E_y - E_z = -1.88$ meV per V atom, indicating an easy-plane magnetization. The electronic band structure of bilayer $2H\text{-VTe}_2$ is shown in Fig. 4(c). Compared with the monolayer case, the band structure becomes more complex due to the coexistence of electronic states from the two VTe_2 layers. Nevertheless, similarly to the monolayer, the VBM is located at the Γ point, while the CBM resides at the K point, resulting in a small indirect band gap of approximately 0.06 eV.

Having understood the basic electronic properties, we now work on qualitatively exploring the Edelstein effect in bilayer $2H\text{-VTe}_2$ from the viewpoint of symmetry. For both AB and BA stackings of bilayer $2H\text{-VTe}_2$, the crystal symmetry belongs to the C_{3v} point group, which allows only the δS_y^{even} component under an applied electric field along the $+x$ direction. Upon introducing in-plane ferromagnetic order, the symmetry is further reduced, thereby enabling additional components of the current-induced spin accumulation. For example, when the magnetization is aligned along the $+x$ direction, the magnetic point group reduces to m , with symmetry generators $\{E, M_x\}$. The M_x symmetry operation forbids both χ_{xx}^{even} and χ_{xx}^{odd} , while allowing other components associated with an electric field along the $+x$ direction, namely χ_{yx}^{even} , χ_{zx}^{even} , χ_{yx}^{odd} , and χ_{zx}^{odd} . Consequently, under an electric field applied along the $+x$ direction, both time-reversal-even and -odd components of δS_y and δS_z can emerge. In contrast, when the magnetization is aligned along the $+y$ direction, the magnetic point group becomes m' , characterized by $\{E, M_x T\}$. Under the $M_x T$ symmetry, χ_{xx}^{even} , χ_{yx}^{odd} , and χ_{zx}^{odd} are forbidden, while the remaining components associated with $+x$ electric field are allowed. As a result, for an electric field along the $+x$ direction, δS_y^{even} , δS_z^{even} , and δS_x^{odd} are permitted. Notably, this symmetry-based analysis is

general and can be extended to other bilayer $2H\text{-MX}_2$ systems with in-plane magnetization.

To further quantify the symmetry-allowed current-induced spin components, we calculate the spin accumulation per unit electric field ($\text{V}/\text{\AA}$) as a function of the Fermi energy for both AB and BA stackings, as shown in Fig. 5. For the case of magnetization along the $+x$ direction, the time-reversal-even component δS_z^{even} exhibits a peak value of approximately $-0.8\hbar$ near $E_F = -0.4$ eV. Meanwhile, a finite δS_y^{even} , which is absent in monolayer $2H\text{-VTe}_2$ but becomes symmetry allowed in the bilayer, also emerges and reaches a local maximum of $0.28\hbar$ near the same energy, which is less than half of δS_z^{even} . Microscopically, these local maxima of spin accumulation primarily originate from electronic states near the Γ point (see Appendix F for more details about microscopic origin), with only minor contributions arising from states near the K point. The time-reversal-odd components, δS_y^{odd} and δS_z^{odd} , increase in magnitude as the Fermi energy shifts downward toward the valence band. For magnetization along the $+y$ direction, both δS_y^{even} and δS_z^{even} exhibit pronounced peaks of approximately $1.0\hbar$ and $1.2\hbar$, respectively, near $E_F = -0.4$ eV, which are again originated from the electronic states near Γ point (see Appendix F for more details). In contrast, δS_x^{odd} increases and gradually saturates at around $-0.5\hbar$ in the same energy region. Notably, switching the stacking configuration from AB to BA—achievable via ferroelectric-controllable interlayer sliding and equivalent to applying an $M_z T$ operation under in-plane magnetization—leads to a sign reversal of δS_y^{even} and δS_z^{odd} , while the other nonzero components remain unchanged. Similarly to the case of monolayer VTe_2 , the spin accumulation in bilayer VTe_2 remains negligible when the Fermi level is shifted upward from the band gap into the low-energy conduction-band region. This behavior arises because the electronic states near the K point, which dominate the low-energy conduction bands, make negligible contributions to the net spin accumulation (see Fig. 15 in Appendix F). Although such sliding-induced sign reversal of current-induced spins has been previously reported for the time-reversal-even contribution in nonmagnetic bilayer WTe_2 [28], its realization in magnetic systems—particularly for both even and odd components—remains largely unexplored and is of great relevance for nonvolatile control of spin-orbit torque. From a symmetry perspective, this selective sign reversal of current-induced spin components originates from the fact that the $M_z T$ operation changes the sign of χ_{yx}^{even} and χ_{zx}^{odd} under

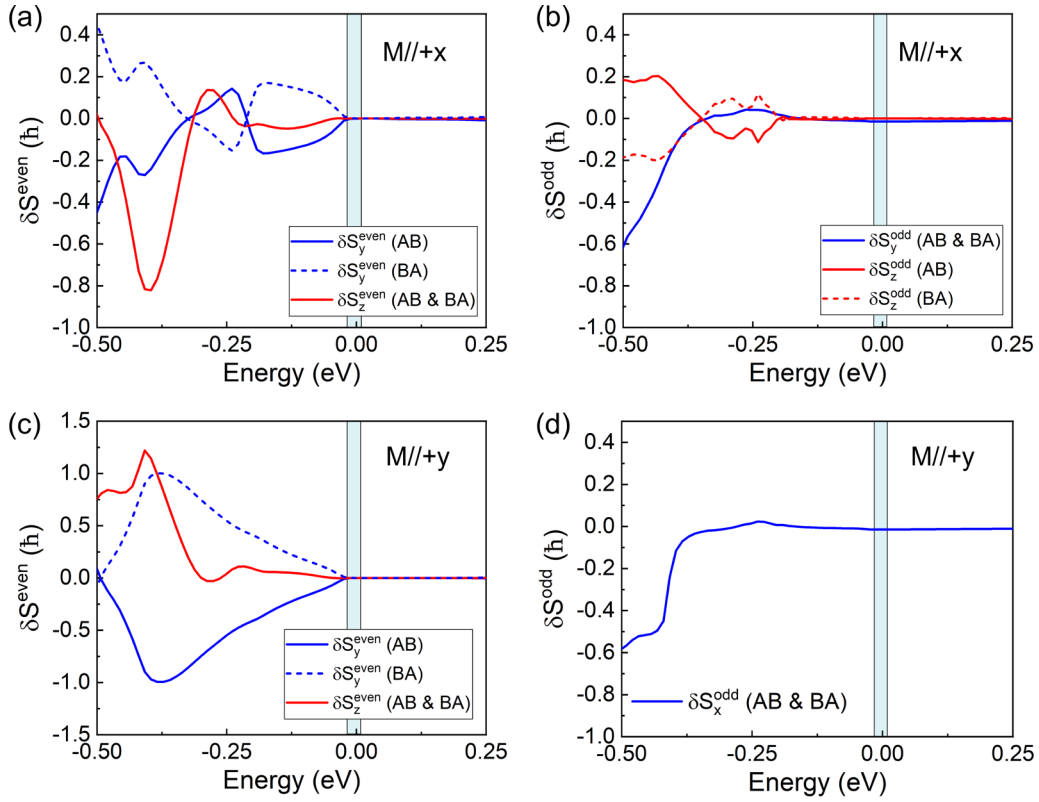


FIG. 5. Calculated symmetry-allowed current-induced spin accumulations in a unit cell as functions of Fermi energy in bilayer $2H\text{-VTe}_2$ with distinct stackings under a reference electric field of $1 \text{ V/\AA}$ along the $+x$ direction. Panels (a) and (b) show the time-reversal-even and time-reversal-odd components of spin accumulation when magnetization is along the $+x$ direction. Panels (c) and (d) show the time-reversal-even and time-reversal-odd components of spin accumulation when magnetization is along the $+y$ direction. AB (BA) denotes the spin accumulation components calculated for AB (BA) stacking in the bilayer system, while AB and BA denote both stackings yield the identical numerical result due to the constraint of symmetry. The dashed blue region indicates the band gap. Note that the magnitude of the electric field ($1 \text{ V/\AA}$) is difficult to realize experimentally and is used only as a reference value.

in-plane magnetization, thereby reversing δS_y^{even} and δS_z^{odd} . These results highlight the potential of manipulating charge-spin conversion in two-dimensional systems via stacking engineering.

The above analysis demonstrates that the current-induced spin accumulation is highly sensitive to the magnetization direction, motivating a detailed investigation of the magnetization-dependent Edelstein effect in bilayer $2H\text{-VTe}_2$. To this end, we calculate the angular dependence of the current-induced spin accumulation for AB-stacked bilayer $2H\text{-VTe}_2$ at $E_F = -0.4 \text{ eV}$ under per applied driven electric field ($\text{V/\AA}$) along the $+x$ direction, as shown in Figs. 6(a) and 6(b). For the time-reversal-even contribution, all three components of δS^{even} emerge during the rotation of the in-plane magnetization. This contrasts with the monolayer case, where only δS_z^{even} is present. Quantitatively, both δS_y^{even} and δS_z^{even} are even functions with respect to $\phi - 90^\circ$, reaching maximum magnitudes of approximately $-1.0 \hbar$ and $1.2 \hbar$ at $\phi = 90^\circ$, respectively. In contrast, δS_x^{even} has a smaller magnitude and exhibits an odd dependence on $\phi - 90^\circ$. For the time-reversal-odd contribution, all three components of δS^{odd} are also present, in contrast to the monolayer case where δS_z^{odd} is absent for in-plane magnetization. Among them, δS_z^{odd} has a relatively small magnitude (always below $0.2 \hbar$), whereas

δS_y^{odd} and δS_x^{odd} can reach values up to approximately $0.4\text{--}0.5 \hbar$. In terms of angular dependence, δS_y^{odd} and δS_z^{odd} are odd functions of $\phi - 90^\circ$, while δS_x^{odd} is an even function (see Appendix E for more details about angular dependence). Overall, the magnitude of the current-induced spin accumulation is on the order of $\sim 0.1\text{--}1 \hbar$ per $\text{V/\AA}$. By normalizing with the calculated conductivity and assuming an effective thickness of $12 \text{ \AA}$ for the bilayer, we estimate that a current density of 10^7 A/cm^2 induces magnetic moments on the order of 10^{-7} to $10^{-6} \hbar$ per unit cell (see Appendix C). To understand the parity dependence of the current-induced spin components on the magnetization angle, we note that changing the magnetization angle from ϕ to $180^\circ - \phi$ is equivalent to applying the $M_x T$ operation. Under this symmetry, χ_{yx}^{even} , χ_{zx}^{even} , and χ_{xx}^{odd} remain invariant, whereas χ_{xx}^{even} , χ_{yx}^{odd} , and χ_{zx}^{odd} change sign. Consequently, for an electric field applied along the $+x$ direction, δS_y^{even} , δS_z^{even} , and δS_x^{odd} (proportional to χ_{yx}^{even} , χ_{zx}^{even} , and χ_{xx}^{odd}) are even functions of $\phi - 90^\circ$, whereas δS_x^{even} , δS_y^{odd} , and δS_z^{odd} (proportional to χ_{xx}^{even} , χ_{yx}^{odd} , and χ_{zx}^{odd}) are odd functions. Notably, when the stacking configuration is switched from AB to BA, δS_x^{even} , δS_y^{even} , and δS_z^{odd} reverse their signs over the entire range of magnetization angles, while the other components remain unchanged (see Appendix G). This

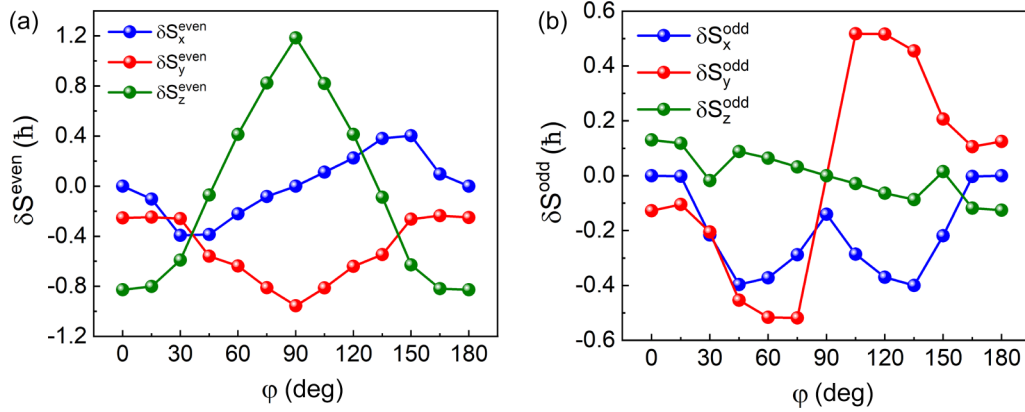


FIG. 6. Angular-dependent spin accumulation in a unit cell of AB-stacked bilayer 2H-VTe₂, which is denoted as a function of ϕ (angle between magnetization direction and the $+x$ direction). (a) The symmetry-allowed time-reversal-even components (only δS_z^{even} here) of spin accumulation in a unit cell as function of ϕ , and (b) the symmetry-allowed time-reversal-odd components of spin accumulation in a unit cell as function of ϕ . Note that the magnetization is rotating in plane, which means that $\phi = 0$ and $\phi = 90^\circ$ denotes $+x$ and $+y$ magnetization, respectively. The spin accumulation is calculated at $E_F = -0.4$ eV under a reference electric field $1 \text{ V/\AA}$ along the $+x$ direction. Note that the magnitude of the electric field ($1 \text{ V/\AA}$) is difficult to realize experimentally and is used only as a reference value.

selective sign reversal originates from the fact that switching from AB to BA under in-plane magnetization is equivalent to applying an $M_z T$ operation. Under this symmetry, χ_{xx}^{even} , χ_{yy}^{even} , and χ_{zx}^{odd} change sign, whereas χ_{xx}^{odd} , χ_{yy}^{odd} , and χ_{zx}^{even} remain invariant. Therefore, for an electric field applied along the $+x$ direction, the corresponding spin components δS_x^{even} , δS_y^{even} , and δS_z^{odd} reverse sign, while the remaining components are unchanged.

V. CONCLUSIONS

In conclusion, based on first-principles calculations and symmetry analysis, we systematically investigate the magnetization-dependent Edelstein effect in monolayer and bilayer ferromagnetic 2H-VTe₂. For monolayer 2H-VTe₂ with D_{3h} crystal symmetry, we find that when the magnetization is aligned along the $+x$ ($+y$) direction, only δS_z^{even} and δS_y^{odd} (δS_z^{even} and δS_x^{odd}) are allowed under an applied electric field along the $+x$ direction. For bilayer 2H-VTe₂ in AB and BA stackings under an applied electric field along the $+x$ direction, where the crystal symmetry is reduced to C_{3v} , additional components emerge. Specifically, for magnetization along $+x$ ($+y$), besides δS_z^{even} and δS_y^{odd} (δS_z^{even} and δS_x^{odd}), extra components such as δS_y^{even} and δS_z^{odd} (δS_y^{even} and δS_x^{odd}) become allowed, some of which can be reversibly controlled by switching between AB and BA stackings. Furthermore, upon rotating the in-plane magnetization, both monolayer and bilayer systems exhibit symmetry-constrained angular dependence of the current-induced spin accumulation. Notably, the symmetry-governed and angle-dependent qualitative behavior of the Edelstein effect identified in 2H-VTe₂ can be generalized to other 2H-MX₂ ferromagnetic systems with the same symmetry, highlighting this material family as a promising platform for efficient charge-spin conversion. Our findings not only deepen the understanding of current-induced spin accumulation in magnetic systems but also pave the way for realizing intrinsic and tunable Edelstein effects for future spin-orbit torque devices.

ACKNOWLEDGMENTS

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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.

APPENDIX A: COMPARISON BETWEEN THE DFT AND WANNIER BAND STRUCTURE

Below we present a comparison between the Wannier-interpolated band structures and the original Quantum ESPRESSO band structures for both monolayer and AB-stacked bilayer 2H-VTe₂ (see Fig. 7). The excellent agreement between the two confirms the accuracy of the Wannier interpolation. For simplicity, we show only the results for the $+x$ magnetization direction, while equally good agreement is obtained for all other magnetization configurations considered in this work, which is not shown here.

APPENDIX B: THE EFFECT OF BROADENING

In the present work, we adopt a phenomenological broadening of $\Gamma = 10 \text{ meV}$. Depending on the convention used to relate the broadening to the carrier lifetime ($\tau = \frac{\hbar}{2\Gamma}$), this value corresponds to a scattering time on the order of 10–100 fs, which is a typical timescale for two-dimensional systems. Therefore, $\Gamma = 10 \text{ meV}$ represents a reasonable choice of broadening [69,70]. We also note that the same broadening value of 10 meV was adopted in a previous first-principles study of the Edelstein effect in monolayer 2H-VSe₂ and 2H-VTe₂ [29].

We nevertheless acknowledge that the choice of broadening can affect the quantitative magnitude of the calculated Edelstein response. To examine this dependence, we

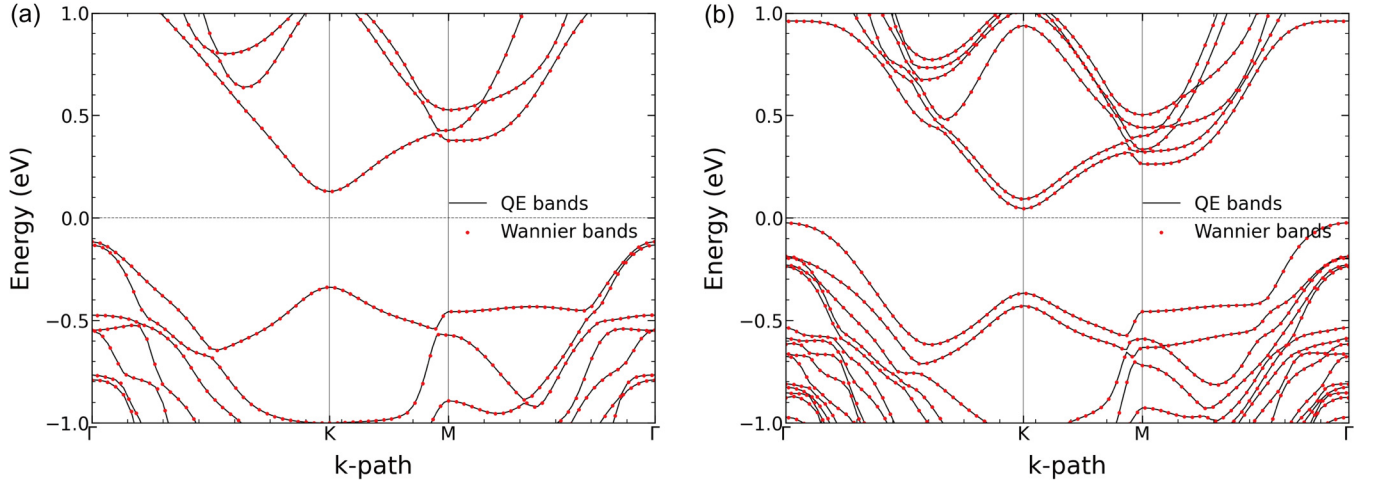


FIG. 7. Comparison between Wannier fitted and DFT-calculated band for (a) monolayer and (b) AB-stacked bilayer VTe_2 with $+x$ magnetization.

calculated the symmetry-allowed current-induced spin accumulations as functions of Γ for monolayer and AB-stacked bilayer $2H-VTe_2$, without losing generality. As shown in Fig. 8, the magnitude of the intraband contribution depends strongly on the broadening and is approximately inversely proportional to Γ . By contrast, the interband contribution is nearly insensitive to Γ . This behavior can be understood from the limiting forms of the response functions. In the small- Γ limit, the time-reversal-even (intraband) contribution reduces to

$$\chi_{ij}^{\text{even}} = -\frac{e\hbar}{2\Gamma N} \sum_{\mathbf{k}, n} \delta(\epsilon_{n\mathbf{k}} - E_F) \langle n\mathbf{k} | \hat{S}_i | n\mathbf{k} \rangle \langle n\mathbf{k} | \hat{v}_j | n\mathbf{k} \rangle, \quad (\text{B1})$$

which explicitly scales as $1/\Gamma$. In contrast, the time-reversal-odd (interband) contribution becomes

$$\chi_{ij}^{\text{odd}} = -\frac{2e\hbar}{N} \sum_{\mathbf{k}} \sum_{\substack{n \in \text{occ.} \\ m \in \text{unocc.}}} \frac{\text{Im}[\langle n\mathbf{k} | \hat{S}_i | m\mathbf{k} \rangle \langle m\mathbf{k} | \hat{v}_j | n\mathbf{k} \rangle]}{(\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}})^2}, \quad (\text{B2})$$

which is independent of Γ in this limit.

Most importantly, although the choice of Γ can substantially modify the magnitude of the Edelstein response, it does not alter the crystal symmetry of the system and therefore does not modify which spin-polarization components are symmetry allowed for a given magnetization direction. Likewise, the symmetry relations between different magnetization directions and stacking configurations remain unchanged. Since the main conclusions of the present work are established based on crystal-symmetry analysis, rather than on the absolute magnitude of the response, they are expected to remain valid for reasonable variations of the Γ .

APPENDIX C: NORMALIZING EDELSTEIN COEFFICIENT WITH CONDUCTIVITY

Besides the original Edelstein effect $\delta S_i = \chi_{ij} E_j$ used in the main text, which denotes the number of spin angular momentum ($\hbar$) in a single unit cell induced by per electric field (1 V/Å) and has the unit of $\hbar \text{ Å/V}$, one can also define a normalized current-induced Edelstein effect, $\delta S_i = \chi'_{ij} J_j$,

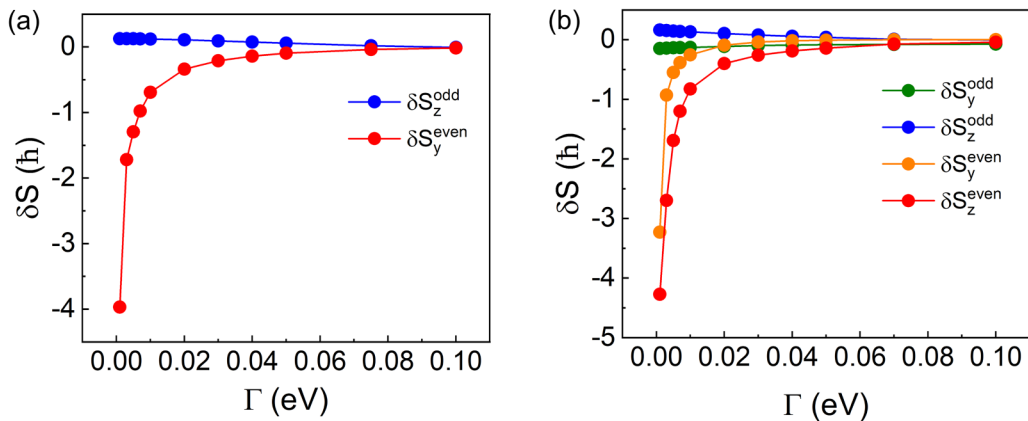


FIG. 8. Γ -dependent current-induced spin accumulation for (a) monolayer and (b) AB-stacked bilayer $2H-VTe_2$ in $+x$ magnetization, under a reference electric field of 1 V/Å applied along the $+x$ direction. The Fermi energy is fixed at $E_F = -0.4$ eV. Only symmetry-allowed components of spin accumulation are displayed.

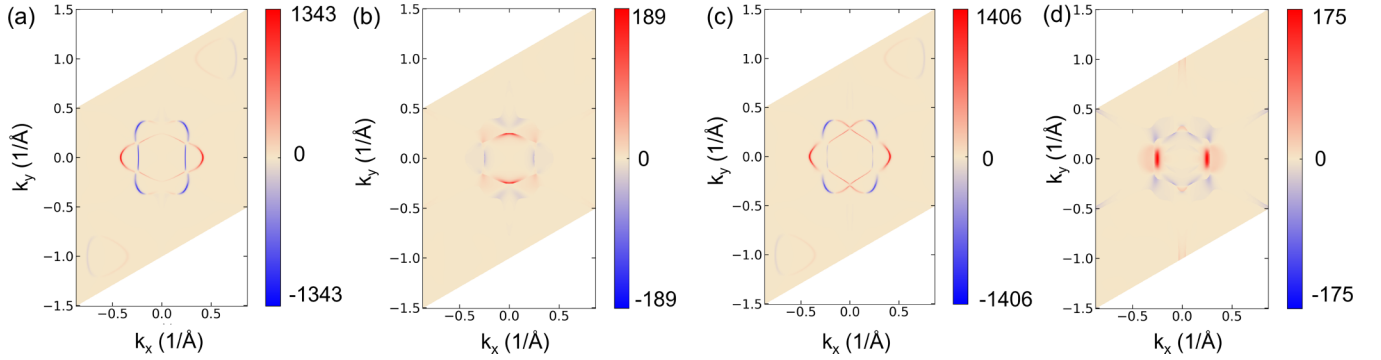


FIG. 9. The k -resolved current-induced spin accumulation under a fixed reference electric field of $1\text{V}/\text{\AA}$ along the $+x$ direction for monolayer $2H\text{-VTe}_2$. (a) δS_z^{even} at $E_F = -0.44$ eV for magnetization along $+x$; (b) δS_y^{odd} at $E_F = -0.47$ eV for magnetization along $+x$; (c) δS_z^{even} at $E_F = -0.44$ eV for magnetization along $+y$; (d) δS_x^{odd} at $E_F = -0.49$ eV for magnetization along $+y$. All selected E_F values are located at local maxima of the calculated spin accumulation. The red and blue denotes the positive and negative values of spin accumulations, respectively. The relationship between k -resolved spin accumulation and total spin accumulation at a specific energy is $\delta S_i = \frac{(2\pi)^3}{VN} \sum_k \delta S_i(E_F, \mathbf{k})$, where V and N is the volume of the unit cell used in DFT calculation and the total number of k points used to sample the Brillouin zone, respectively. The unit of k -resolved spin accumulation is $\hbar \text{\AA}^3$.

which denotes the three-dimensional volume density of spins angular momentum ($\hbar/\text{cm}^3$) induced by per current density (A/cm^2) and possesses the unit of $\hbar/(\text{A cm})$.

Since the electric current component J_i can be related with electric field E_i with $J_i = \sigma_{ii} E_i$, where σ_{ii} is the longitudinal conductivity of the system, the χ_{ij} can be thus converted into χ'_{ij} with the following expression:

$$\chi'_{ij} = \chi_{ij}/(V \cdot \sigma_{ii}), \quad (\text{C1})$$

where V is the effective volume of the system. Here σ_{ii} can be calculated using the Kubo formula implemented in Linres code:

$$\sigma_{ii} = \frac{e^2 \hbar}{\pi V N} \sum_{\mathbf{k}, m, n} \frac{\Gamma^2 \text{Re}(\langle n\mathbf{k} | \hat{v}_i | m\mathbf{k} \rangle \langle m\mathbf{k} | \hat{v}_i | n\mathbf{k} \rangle)}{[(E_f - \epsilon_{n\mathbf{k}})^2 + \Gamma^2][(E_f - \epsilon_{m\mathbf{k}})^2 + \Gamma^2]}. \quad (\text{C2})$$

Here e is the elementary charge, n and m denote band indices, $\mathbf{k}$ is the Bloch wave vector, E_F is the Fermi energy, $\hat{v}$ is the velocity operator, $\hat{S}$ is the spin operator, $\epsilon_{n\mathbf{k}}$ is the eigenvalue, V is the volume of unit cell after considering the effective thickness of two-dimensional monolayer, N is the total number of k points used to sample the Brillouin zone, and $\Gamma = 10$ meV is the disorder parameter. Based on the calculated longitude conductivity and considering the thickness of monolayer and bilayer $2H\text{-VTe}_2$ for about $6 \text{\AA}$ and $12 \text{\AA}$, one can find that a $\chi_{ij} = 1 \text{\AA}^3/\text{V}$ in per unit cell corresponds to $8 \times 10^9 \hbar/(\text{A cm})$ and $4 \times 10^9 \hbar/(\text{A cm})$, respectively. By further considering the strength of driven current and the effective volume of the system, one can arrive at the amount of spin accumulation under certain current. The magnitude of the normalized time-reversal-even Edelstein effect, which dominates the total Edelstein effect in our system, is largely independent of the Γ broadening and can thus serve as a reliable reference value.

APPENDIX D: K -RESOLVED EDELSTEIN EFFECT PATTERN FOR MONOLAYER $2H\text{-VTe}_2$

In Fig. 9 we show the k -resolved current-induced spin accumulation components under a fixed $+x$ electric field ($1 \text{V}/\text{\AA}$) for monolayer $2H\text{-VTe}_2$ at Fermi levels corresponding to the local maxima in the low-energy valance band region. In Fig. 10 we show the k -resolved current-induced spin accumulation components at $E_F = 0.2$ eV, corresponding to the low-energy conduction band region. Note that for $E_F = 0.2$ eV, the time-reversal-odd spin accumulation components vanish throughout the entire k -space and are therefore not plotted.

Below we briefly explain the origin of the maxima spin accumulation components in valance band region. For monolayer $2H\text{-VTe}_2$ with magnetization along either $+x$ or $+y$, the maximum time-reversal-even spin accumulation occurs at approximately $E_F = -0.44$ eV, which can be seen in Figs. 2(a) and 2(c). Since the time-reversal-even spin accumulation originates from the intraband contribution, only the electronic states located at the Fermi surface contribute to the response. Consequently, the k -resolved spin accumulation closely follows the topology of the Fermi surface at the corresponding Fermi energy. To understand its origin, we plot the band structure together with the chosen Fermi level at $E_F = -0.44$ eV. As circled in Fig. 11(a) and 11(c), two bands cross the Fermi level along the Γ - K direction and provide the dominant contributions to the spin accumulation. These two bands possess opposite signs of spin accumulation, giving rise to the two arclike features with opposite signs observed in Figs. 9(a) and 9(c). The total time-reversal-even spin accumulation therefore results from the competition between the contributions of these two Fermi-surface bands.

When the magnetization is along $+x$, the maximum time-reversal-odd spin accumulation occurs at $E_F = -0.47$ eV. In contrast to the intraband response, the time-reversal-odd spin accumulation originates from the interband contribution, which is jointly determined by occupied and unoccupied states. According to the interband Kubo formula [see Eq. (3)

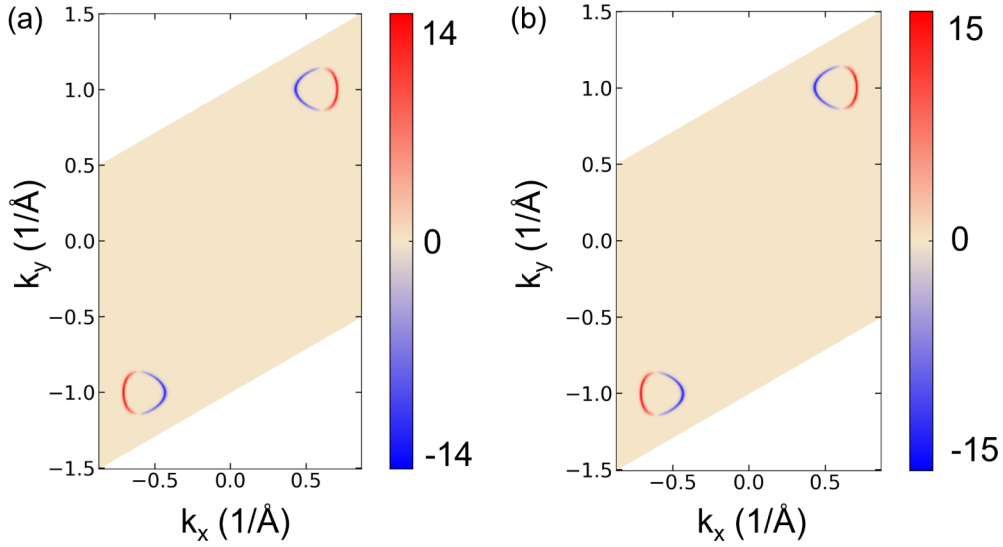


FIG. 10. The k -resolved current-induced spin accumulation under a fixed reference electric field of $1\text{V}/\text{\AA}$ along the $+x$ direction for monolayer $2H\text{-VTe}_2$ under $E_F = 0.2\text{ eV}$. (a) δS_z^{even} for magnetization along $+x$; (b) δS_z^{even} at for magnetization along $+y$. The red and blue denotes the positive and negative values of spin accumulations, respectively. The relationship between k -resolved spin accumulation and total spin accumulation at a specific energy is $\delta S_i = \frac{(2\pi)^3}{VN} \sum_k \delta S_i(E_F, \mathbf{k})$, where V and N is the volume of the unit cell used in DFT calculation and the total number of k points used to sample the Brillouin zone, respectively. The unit of k -resolved spin accumulation is $\hbar \text{\AA}^3$.

in the method part], the response is generally enhanced when two or more bands become nearly degenerate, since the corresponding contribution is inversely proportional to the square of the interband energy separation. As circled in Fig. 11(b), the Fermi energy of $E_F = -0.47\text{ eV}$ passes

through a near-crossing region along the M - Γ direction, where three bands approach each other with very small energy separations. Such a near-degenerate band structure is therefore expected to generate a strong local interband contribution to the time-reversal-odd spin accumulation. This interpretation

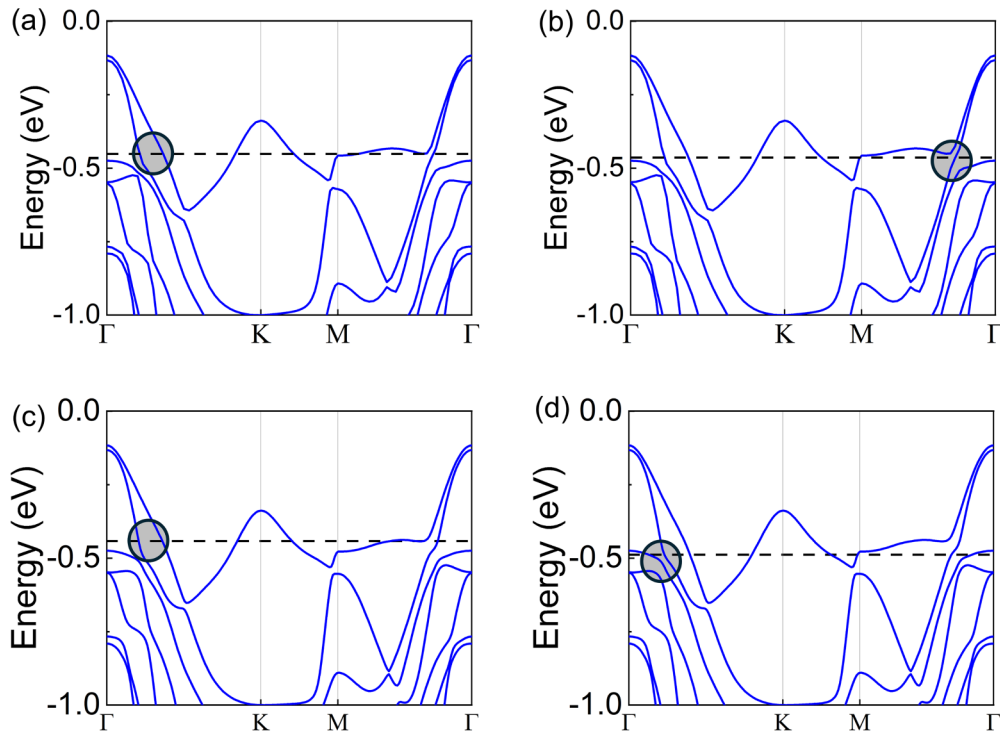


FIG. 11. Band structures for monolayer $2H\text{-VTe}_2$ at specific energies. (a) The band structure at $E_F = -0.44\text{ eV}$ with $+x$ magnetization; (b) the band structure at $E_F = -0.47\text{ eV}$ with $+x$ magnetization; (c) the band structure at $E_F = -0.44\text{ eV}$ with $+y$ magnetization; (d) the band structure at $E_F = -0.49\text{ eV}$ with $+y$ magnetization. The circle denotes the area of interest, while the dashed line denotes the corresponding Fermi energy.

is consistent with the pronounced positive arclike distribution observed in Fig. 9(b), which gives rise to the positive peak in the total time-reversal-odd spin accumulation.

When the magnetization is along $+y$, the maximum time-reversal-odd spin accumulation occurs at $E_F = -0.49$ eV. As circled in Fig. 11(d), the chosen Fermi energy intersects another near-crossing region along the Γ - K direction, where three neighboring bands are separated by only a small energy gap. Similarly to the previous case, such a near-degenerate band structure is expected to enhance the interband contribution locally. This picture agrees well with the two positive arclike features observed in Fig. 9(d), which together produce the large positive peak in the total time-reversal-odd spin accumulation.

APPENDIX E: ANGULAR-DEPENDENCE OF SPIN ACCUMULATION

To qualitatively understand the angular dependence of spin accumulation in monolayer and bilayer $2H$ -VTe₂, one should figure out the symmetry-adapted expression of spin accumulation as a function of in-plane magnetization direction.

For time-reversal-even components, we have $\delta S_i^{\text{even}} = \sum_{j=x,y} \chi_{ij}^{\text{even}} E_j$. Then we express χ_{ij}^{even} as a function of magnetization direction in the even-order dependence up to fourth order:

$$\chi_{ij}^{\text{even}} = \chi^0 + \sum_{ab} \chi^{ab} m_a m_b + \sum_{abcd} \chi^{abcd} m_a m_b m_c m_d. \quad (\text{E1})$$

Here $a, b, c, d = x, y, z$ denotes the coordinate of magnetization.

Similarly, for time-reversal-odd components, $\delta S_i^{\text{odd}} = \sum_{j=x,y} \chi_{ij}^{\text{odd}} E_j$, we express χ_{ij}^{odd} as a function of magnetization direction in the odd-order dependence up to fifth order:

$$\begin{aligned} \chi_{ij}^{\text{odd}} = & \sum_a \chi^a m_a + \sum_{abc} \chi^{abc} m_a m_b m_c \\ & + \sum_{abcde} \chi^{abcde} m_a m_b m_c m_d m_e. \end{aligned} \quad (\text{E2})$$

By applying the symmetry operations of our system (C_{3z} , M_x , and M_z in D_{3h} point group for monolayer $2H$ -VTe₂, as well as C_{3z} and M_x in C_{3v} point group symmetry for bilayer $2H$ -VTe₂) on the above equation, the expression will still be satisfied, which allows us to rule out the symmetry-forbidden terms and thereby lead to a clearer expression. Such approach has been adopted to arrive at the low-order magnetization dependence behavior of Edelstein effect in magnetic systems [29,42].

The above-mentioned symmetry operations will transform the E_x , E_y , m_x , m_y , δS_x , δS_y , and δS_z as

$$\begin{aligned} C_{3z}: \quad E_x &\longrightarrow -\frac{1}{2}E_x - \frac{\sqrt{3}}{2}E_y, \quad E_y \longrightarrow \frac{\sqrt{3}}{2}E_x - \frac{1}{2}E_y, \\ m_x &\longrightarrow -\frac{1}{2}m_x - \frac{\sqrt{3}}{2}m_y, \quad m_y \longrightarrow \frac{\sqrt{3}}{2}m_x - \frac{1}{2}m_y, \\ \delta S_x &\longrightarrow \frac{1}{2}\delta S_x - \frac{\sqrt{3}}{2}\delta S_y, \quad \delta S_y \longrightarrow \frac{\sqrt{3}}{2}\delta S_x - \frac{1}{2}\delta S_y, \\ \delta S_z &\longrightarrow \delta S_z, \end{aligned}$$

$$\begin{aligned} M_x: \quad E_x &\longrightarrow -E_x, \quad E_y \longrightarrow E_y, \\ m_x &\longrightarrow m_x, \quad m_y \longrightarrow -m_y, \\ \delta S_x &\longrightarrow \delta S_x, \quad \delta S_y \longrightarrow -\delta S_y, \\ \delta S_z &\longrightarrow -\delta S_z, \\ M_z: \quad E_x &\longrightarrow E_x, \quad E_y \longrightarrow E_y, \\ m_x &\longrightarrow -m_x, \quad m_y \longrightarrow -m_y, \\ \delta S_x &\longrightarrow -\delta S_x, \quad \delta S_y \longrightarrow -\delta S_y, \\ \delta S_z &\longrightarrow \delta S_z. \end{aligned} \quad (\text{E3})$$

For monolayer $2H$ -VTe₂ with D_{3h} symmetry, by applying the constraint of C_{3z} , M_x , and M_z , and further considering the electric field along $+x$ and magnetization in the xy plane [$\mathbf{m} = (\cos\phi, \sin\phi, 0)$], one can finally arrive at the symmetry-allowed components (δS_z^{even} , δS_x^{odd} , δS_y^{odd}) as a function of ϕ up to three order as

$$\begin{aligned} \delta S_z^{\text{even}} &= C \cos 2\phi \\ \delta S_x^{\text{odd}} &= A \sin 3\phi + B \sin \phi \\ \delta S_y^{\text{odd}} &= -A \cos 3\phi + B \cos \phi. \end{aligned} \quad (\text{E4})$$

The calculated angular dependences in Fig. 3 are well fitted by these expressions, as shown in Fig. 12. As can be seen, δS_z^{even} can be well described by a simple expression of $\cos 2\phi$. On the other hand, the deviation of δS_x^{odd} and δS_y^{odd} from a simple cosine dependence originates from the symmetry-allowed $\cos 3\phi$ and $\sin 3\phi$ harmonic. Therefore, the unconventional angular profile is not anomalous but results from the superposition of the fundamental and third-order angular harmonics.

For AB-stacked bilayer $2H$ -VTe₂ with C_{3v} symmetry, by applying the constraint of C_{3z} and M_x , and further considering the electric field along $+x$ and magnetization in xy plane ($\mathbf{m} = \cos\phi, \sin\phi, 0$), one can finally arrive at the symmetry-allowed components of spin accumulations as a function of ϕ up to fifth order as

$$\begin{aligned} \delta S_x^{\text{even}} &= A_2 \sin 2\phi + A_4 \sin 4\phi, \\ \delta S_y^{\text{even}} &= B_0 + B_2 \cos 2\phi + B_4 \cos 4\phi, \\ \delta S_z^{\text{even}} &= C_2 \cos 2\phi + C_4 \cos 4\phi, \\ \delta S_x^{\text{odd}} &= A_1 \sin \phi + A_3 \sin 3\phi + A_5 \sin 5\phi, \\ \delta S_y^{\text{odd}} &= A_1 \cos \phi + B_3 \cos 3\phi - A_5 \cos 5\phi, \\ \delta S_z^{\text{odd}} &= C_1 \cos \phi + C_5 \cos 5\phi. \end{aligned} \quad (\text{E5})$$

The calculated angular dependences in Fig. 6 are well fitted by these expressions, as shown in Fig. 13. Several components contain appreciable third-, fourth-, or fifth-harmonic contributions, which explains why their angular profiles deviate from the lowest-order $\sin \phi$, $\cos \phi$, or $\cos 2\phi$ forms.

APPENDIX F: k -RESOLVED EDELSTEIN EFFECT PATTERN FOR AB-STACKED BILAYER $2H$ -VTe₂

In Fig. 14 we show the k -resolved current-induced spin accumulation components under a fixed $+x$ electric field (1 V/Å) for AB-stacked bilayer $2H$ -VTe₂ at Fermi levels

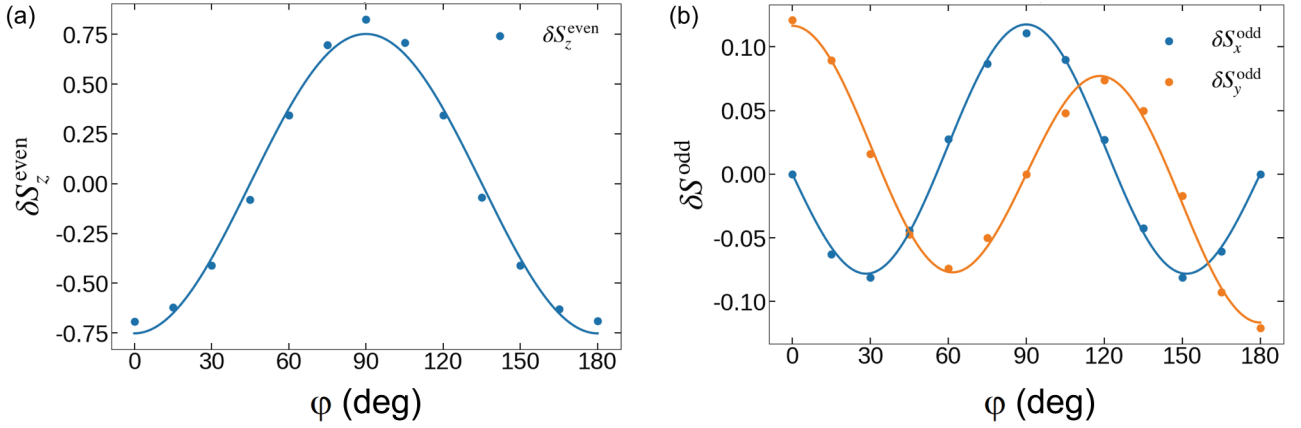


FIG. 12. Fitted angular-dependent spin accumulation in a unit cell of monolayer $2H\text{-VTe}_2$, which is denoted as a function of ϕ (angle between magnetization direction and the $+x$ direction). (a) The symmetry-allowed time-reversal-even components (only δS_z^{even} here) of spin accumulation in a unit cell as function of ϕ , and (b) the symmetry-allowed time-reversal-odd components of spin accumulation in a unit cell as function of ϕ . Note that the magnetization is rotating in plane, which means that $\phi = 0$ and $\phi = 90^\circ$ denotes $+x$ and $+y$ magnetization, respectively. The unit of spin accumulation is $\hbar$. The spin accumulation is calculated at $E_F = -0.4$ eV under a reference electric field $1 \text{ V/\AA}$ along the $+x$ direction. Note that the magnitude of the electric field ($1 \text{ V/\AA}$) is difficult to realize experimentally and is used only as a reference value. The calculated angular dependence is well fitted with equation (E4), which leads to $C = -0.751$, $A = -0.091$, $B = 0.026$.

corresponding to the local maxima in the low-energy valance band region. In Fig. 15 we show the k -resolved current-induced spin accumulation components at $E_F = 0.2$ eV, corresponding to the low-energy conduction band region. Note that for $E_F = 0.2$ eV, the time-reversal-odd spin accumulation components vanish throughout the entire k -space and are therefore not plotted.

Below we briefly explain the origin of the maxima spin accumulation components in valance band region. For AB-stacked bilayer $2H\text{-VTe}_2$, the time-reversal-even spin accumulation reaches its maximum at approximately $E_F = -0.40$ eV for both $+x$ and $+y$ magnetizations, which can be seen in Fig. 5 of the main text. As circled in Fig. 16, the Fermi level intersects five energy bands at this energy. Consequently,

the intraband spin accumulation consists of contributions from multiple Fermi-surface sheets with different magnitudes and signs. The total intraband response is therefore determined by the intricate competition among these individual band contributions, giving rise to the observed maximum around $E_F = -0.40$ eV.

APPENDIX G: ANGULAR-DEPENDENT CURRENT-INDUCED SPIN ACCUMULATION FOR BILAYER $2H\text{-VTe}_2$ WITH BA STACKING

Below we show the angular-dependent current-induced spin accumulation for bilayer $2H\text{-VTe}_2$ with BA stacking, which can be seen in Fig. 17.

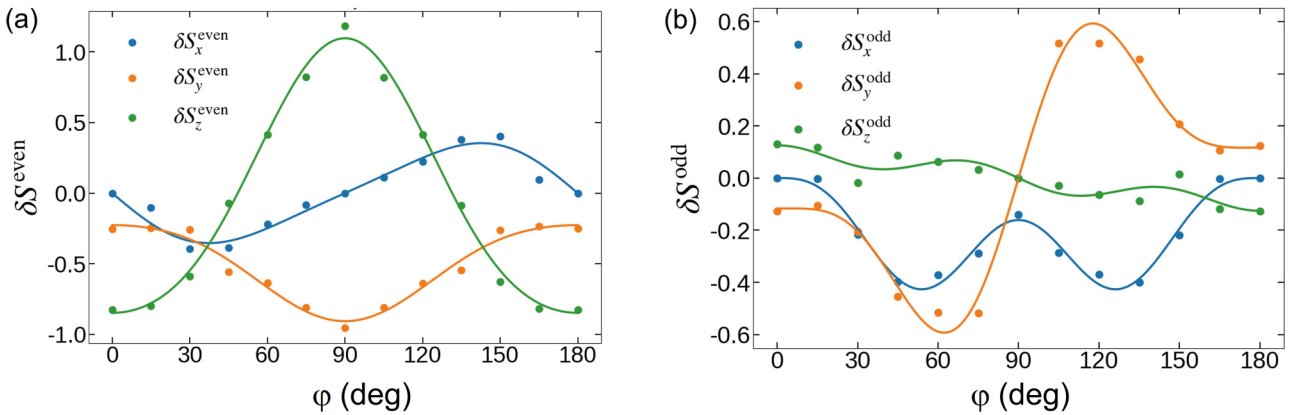


FIG. 13. Fitted angular-dependent spin accumulation in a unit cell of AB-stacked bilayer $2H\text{-VTe}_2$, which is denoted as a function of ϕ (angle between magnetization direction and the $+x$ direction). (a) The symmetry-allowed time-reversal-even components (only δS_z^{even} here) of spin accumulation in a unit cell as function of ϕ , and (b) the symmetry-allowed time-reversal-odd components of spin accumulation in a unit cell as function of ϕ . Note that the magnetization is rotating in plane, which means that $\phi = 0$ and $\phi = 90^\circ$ denotes $+x$ and $+y$ magnetization, respectively. The unit of spin accumulation is $\hbar$. The spin accumulation is calculated at $E_F = -0.4$ eV under a reference electric field $1 \text{ V/\AA}$ along the $+x$ direction. Note that the magnitude of the electric field ($1 \text{ V/\AA}$) is difficult to realize experimentally and is used only as a reference value. The calculated angular dependence is well fitted with Eq. (E5), which leads to $A_1 = -0.354$, $A_2 = -0.340$, $A_3 = -0.076$, $A_4 = -0.052$, $A_5 = 0.117$, $B_0 = -0.521$, $B_2 = 0.340$, $B_3 = 0.354$, $B_4 = -0.046$, $C_1 = 0.089$, $C_2 = -0.973$, $C_4 = 0.124$, and $C_5 = 0.037$.

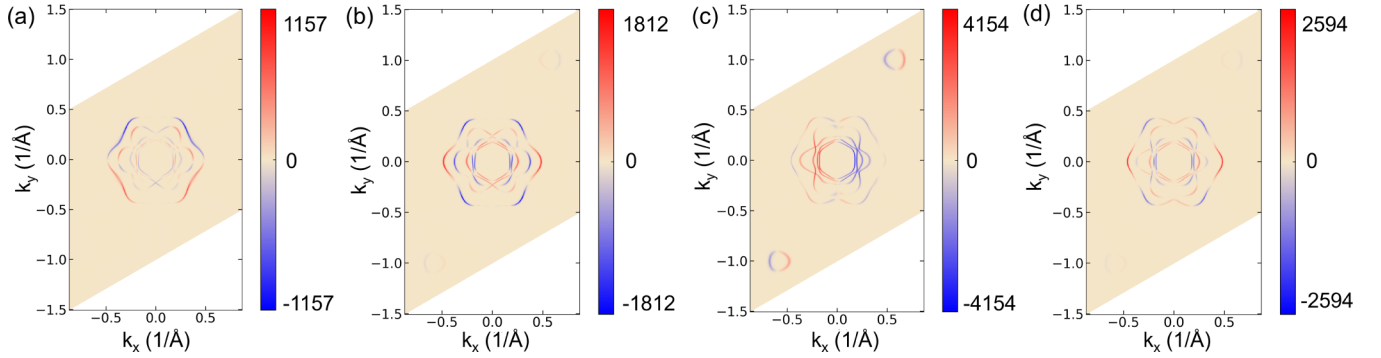


FIG. 14. The k -resolved current-induced spin accumulation under a fixed reference electric field of $1 \text{ V/\AA}$ along the $+x$ direction for bilayer $2H\text{-VTe}_2$. (a) δS_y^{even} at $E_F = -0.4 \text{ eV}$ for magnetization along $+x$; (b) δS_z^{even} at $E_F = -0.4 \text{ eV}$ for magnetization along $+x$; (c) δS_y^{even} at $E_F = -0.4 \text{ eV}$ for magnetization along $+y$; (d) δS_z^{even} at $E_F = -0.4 \text{ eV}$ for magnetization along $+y$. All selected E_F values are located at local maxima of the calculated spin accumulation. The red and blue denotes the positive and negative values of spin accumulations, respectively. The relationship between k -resolved spin accumulation and total spin accumulation at a specific energy is $\delta S_i = \frac{(2\pi)^3}{VN} \sum_k \delta S_i(E_F, \mathbf{k})$, where V and N is the volume of the unit cell used in DFT calculation and the total number of k points used to sample the Brillouin zone, respectively. The unit of k -resolved spin accumulation is $\hbar \text{ \AA}^3$.

APPENDIX H: SPIN ACCUMULATION WITHIN EXTENDED ENERGY RANGE

Below we show the energy-dependent current-induced spin polarization over an extended energy range in the conduction band. For monolayer $2H\text{-VTe}_2$, which can be seen in Fig. 18, when the chemical potential is shifted further into the conduction band, up to 1 eV , all components of the spin accumulation remain very small. This behavior contrasts sharply with the valence-band case, where sizable spin accumulation emerges. As a result, in order to arrive at considerable spin accumulation, one should dope the Fermi level into the valance band.

For bilayer $2H\text{-VTe}_2$, as can be seen in Fig. 19, once the chemical potential is shifted sufficiently far into the conduction band, beyond approximately 0.3 eV , several spin-accumulation components become noticeably enhanced. For example, when the magnetization is oriented along the $+x$ direction, both δS_y^{even} and δS_z^{odd} exhibit pronounced peaks

near 0.5 eV . When the magnetization is oriented along the $+y$ direction, δS_y^{even} displays several peaks of alternating sign over the energy range from approximately 0.4 to 0.7 eV . In contrast to the peaks in the valence band, which originate from electronic states near the Γ point, the aforementioned spin-accumulation peaks in the conduction bands mainly arise from electronic states away from the Γ point, particularly those near the M points and along the paths connecting symmetry-equivalent M points, which can be seen in Fig. 20.

APPENDIX I: SPIN HALL EFFECTS

Besides the Edelstein effect, the spin Hall effect may also exist in present system and even possess magnetization-dependent behavior. To gain more insights, we calculate the spin Hall effect in our system. Note that the spin Hall conductivity, which consists of both the time-reversal-even part

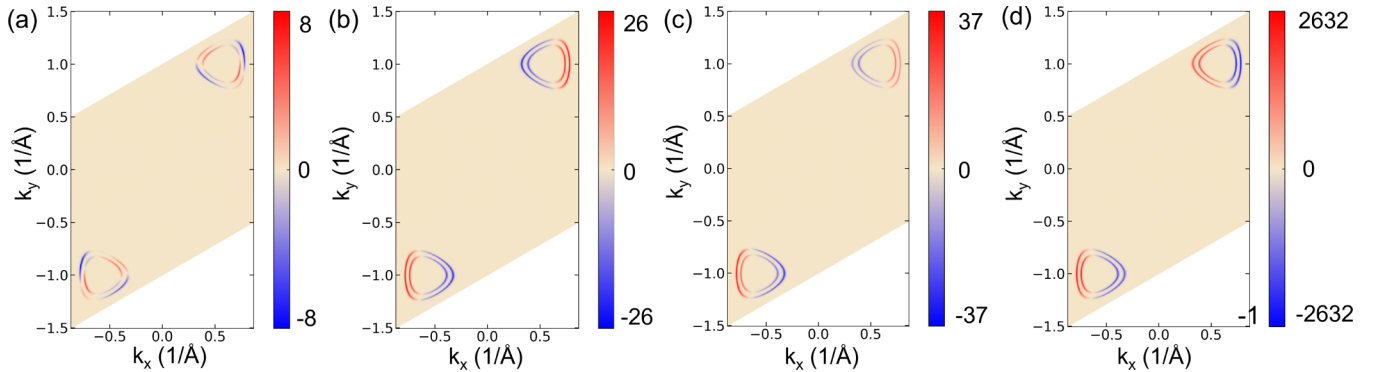


FIG. 15. The k -resolved current-induced spin accumulation under a fixed reference electric field of $1 \text{ V/\AA}$ along the $+x$ direction for AB-stacked bilayer $2H\text{-VTe}_2$ under $E_F = 0.2 \text{ eV}$. (a) δS_y^{even} for magnetization along $+x$; (b) δS_z^{even} for magnetization along $+x$; (c) δS_y^{even} for magnetization along $+y$; (d) δS_z^{even} for magnetization along $+y$. The red and blue denotes the positive and negative values of spin accumulations, respectively. The relationship between k -resolved spin accumulation and total spin accumulation at a specific energy is $\delta S_i = \frac{(2\pi)^3}{VN} \sum_k \delta S_i(E_F, \mathbf{k})$, where V and N is the volume of the unit cell used in DFT calculation and the total number of k points used to sample the Brillouin zone, respectively. The unit of k -resolved spin accumulation is $\hbar \text{ \AA}^3$.

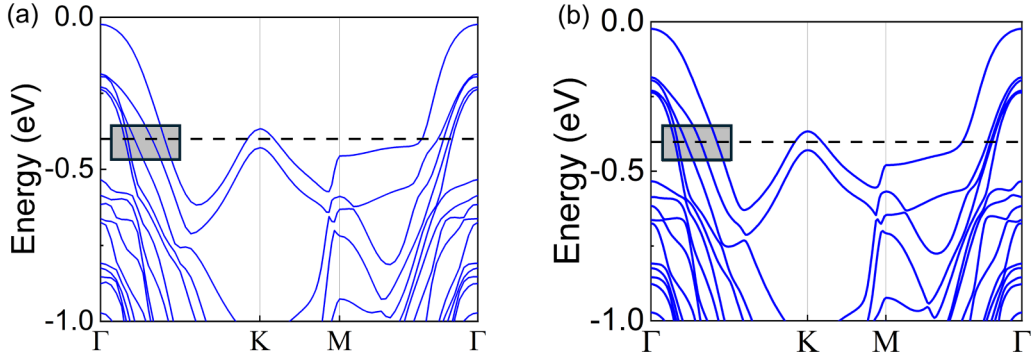


FIG. 16. Band structures for AB-stacked bilayer $2H\text{-VTe}_2$ at specific energies. (a) The band structure at $E_F = -0.4$ eV with $+x$ magnetization; (b) the band structure at $E_F = -0.4$ eV with $+y$ magnetization. The circle denotes the area of interest, while the dashed line denotes the corresponding Fermi energy.

(interband contribution) and time-reversal-odd part (intraband contribution) [11,71,72], is calculated with the Kubo formula:

$$\sigma_{ij}^{k,\text{odd}} = -\frac{e\hbar}{\pi NV} \sum_{\mathbf{k},m,n} \frac{\Gamma^2 \text{Re} [\langle \mathbf{n}\mathbf{k} | \hat{J}_i^k | \mathbf{m}\mathbf{k} \rangle \langle \mathbf{m}\mathbf{k} | \hat{v}_j | \mathbf{n}\mathbf{k} \rangle]}{[(E_F - \epsilon_{n\mathbf{k}})^2 + \Gamma^2][(E_F - \epsilon_{m\mathbf{k}})^2 + \Gamma^2]}, \quad (\text{I1})$$

$$\sigma_{ij}^{k,\text{even}} = \frac{2e\hbar}{NV} \sum_{\mathbf{k}} \sum_{\substack{n \in \text{occ.} \\ m \in \text{unocc.}}} \text{Im} [\langle \mathbf{n}\mathbf{k} | \hat{J}_i^k | \mathbf{m}\mathbf{k} \rangle \langle \mathbf{m}\mathbf{k} | \hat{v}_j | \mathbf{n}\mathbf{k} \rangle] \times \frac{\Gamma^2 - (\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}})^2}{[(\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}})^2 + \Gamma^2]^2}. \quad (\text{I2})$$

Here $\hat{J}_i^k = \frac{1}{2}\{\hat{v}_i, \hat{S}_k\}$ denotes the spin current operator and V denotes the effective volume of the unit cell after considering the effective thickness of two-dimensional monolayer (6 Å for monolayer $2H\text{-VTe}_2$ and 12 Å for bilayer $2H\text{-VTe}_2$). The calculation of spin Hall conductivity is performed with Linres [61] code. $\Gamma = 10$ meV is adopted, being consistent with the calculation of Edelstein effect.

The symmetry-allowed spin Hall conductivity is shown in Figs. 21 and 22. For simplicity, we only consider the case where the electric field is along the $+x$ direction ($j =$

x). For monolayer $2H\text{-VTe}_2$, by changing the magnetization from $+x$ ($m/m'2'$) to $+y$ ($m'/m'2$), the symmetry-allowed time-reversal-even spin Hall component is always σ_{yx}^z , which lies in the order of magnitude of $100 \hbar/e$ (S/cm). However, the symmetry-allowed time-reversal-odd spin Hall components will change from σ_{xx}^x and σ_{yx}^y to σ_{yx}^x and σ_{xx}^y . Moreover, under the current broadening parameter, the time-reversal-odd spin Hall conductivity dominates, being approximately one to two orders of magnitude larger than the time-reversal-even component. The similar situation happens in AB-stacked bilayer VTe_2 . By changing the magnetization from $+x$ (m) to $+y$ (m'), the symmetry-allowed time-reversal-even spin Hall components are always σ_{xx}^x , σ_{yx}^y , and σ_{yx}^z , which all lie in the order of magnitude of $100 \hbar/e$ (S/cm). On the other hand, the symmetry-allowed time-reversal-odd spin Hall components will change from σ_{xx}^x , σ_{yx}^y , and σ_{yx}^z to σ_{yx}^x , σ_{xx}^y , and σ_{yx}^z . Meanwhile, under the current broadening parameter, the time-reversal-odd spin Hall conductivity dominates, being approximately one to two orders of magnitude larger than the time-reversal-even component. The above results demonstrate the magnetic configuration can serve as an effective knob for tuning the spin Hall response in magnetic systems, which is in line with the previous reports [73–75].

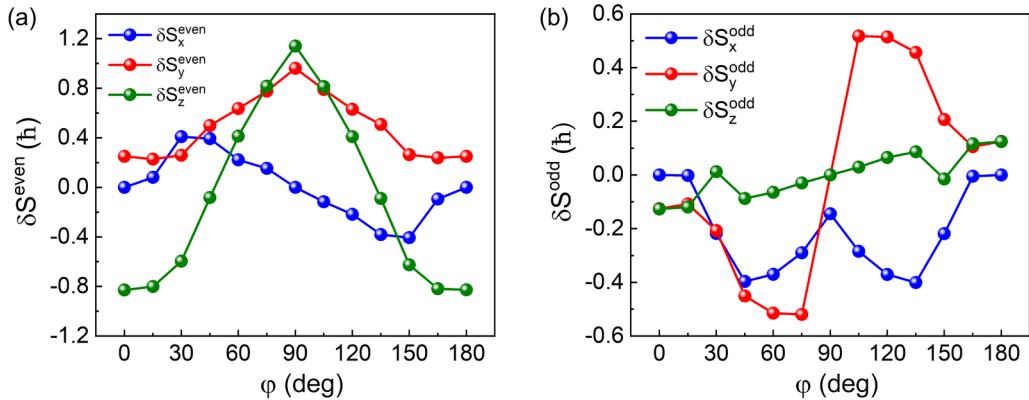


FIG. 17. Angular-dependent spin accumulation of BA-stacked bilayer $2H\text{-VTe}_2$, which is denoted as a function of ψ (angle between magnetization direction and the $+x$ direction). (a) The symmetry-allowed time-reversal-even components (only δS_z^{even} here) of spin accumulation in a unit cell as function of ψ , and (b) the symmetry-allowed time-reversal-odd components of spin accumulation in a unit cell as function of ψ . Note that the magnetization is rotating in plane, which means that $\psi = 0$ and $\psi = 90^\circ$ denotes $+x$ and $+y$ magnetization, respectively. The spin accumulation is calculated under $E_F = -0.4$ eV under a reference electric field of $1 \text{ V/\AA}$ along the $+x$ direction.

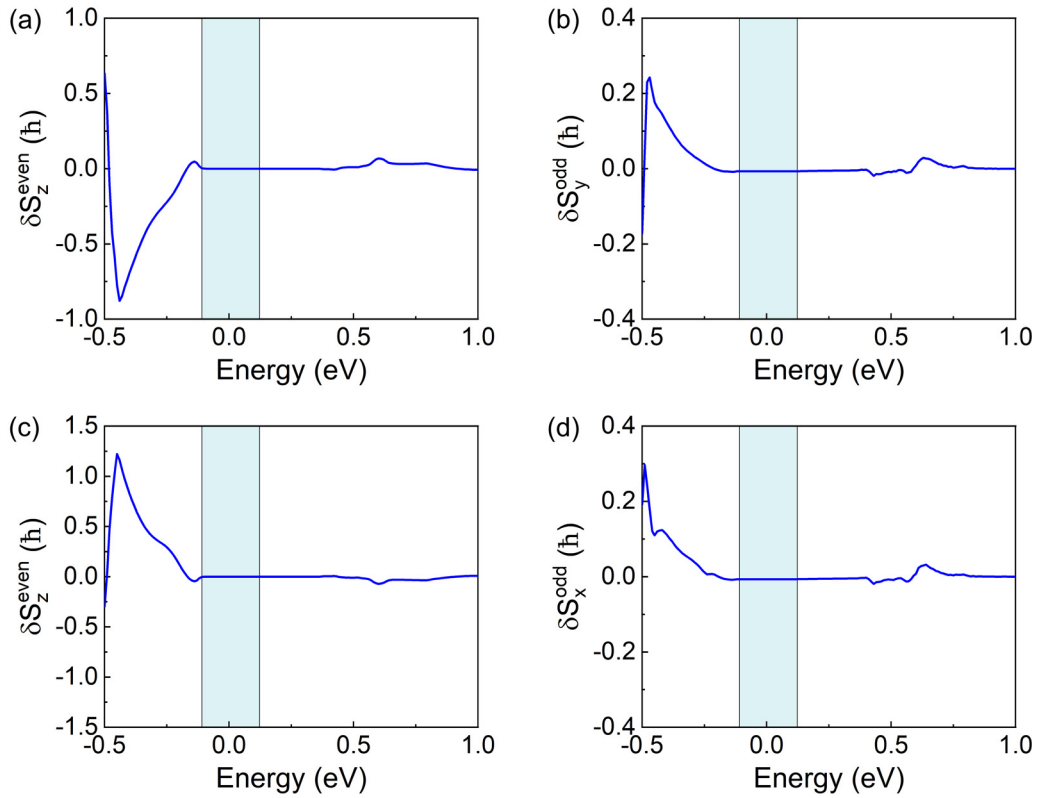


FIG. 18. Calculated symmetry-allowed current-induced spin accumulations in a unit cell in an extended energy range in monolayer $2H$ -VTe₂ under a reference electric field $1 \text{ V/\AA}$ along the $+x$ direction. Panels (a) and (b) show the time-reversal-even (δS_z^{even}) and time-reversal-odd (δS_y^{odd}) components of spin accumulation when magnetization is along the $+x$ direction. Panels (c) and (d) show the time-reversal-even (δS_z^{even}) and time-reversal-odd (δS_x^{odd}) components of spin accumulation when magnetization is along the $+y$ direction. The dashed blue region indicates the band gap. Note that the magnitude of the electric field ($1 \text{ V/\AA}$) is difficult to realize experimentally and is used only as a reference value.

Although a spin Hall response may coexist with the Edelstein effect in the present system, a spin Hall current flowing uniformly in the interior of an ideal material does not by itself generate a homogeneous spin accumulation. A spin-Hall-induced spin accumulation generally appears only when the transverse spin current is scattered, absorbed, or terminated at sample boundaries or interfaces. In a laterally symmetric finite sample, the resulting spin accumulations at opposite boundaries typically have opposite signs and therefore tend to cancel when integrated over the entire system. By contrast, the Edelstein effect directly generates a homogeneous nonequilibrium spin polarization within the bulk of an inversion-asymmetric material under an applied electric field. Therefore, the Edelstein effect is expected to dominate the net current-induced spin accumulation considered here, while the spin Hall effect may only contribute locally near boundaries or interfaces in a finite device, which is beyond the focus of our work.

APPENDIX J: COMPARISON WITH PREVIOUS STUDIES ON SYMMETRY-CONTROLLED SPIN RESPONSES

It is instructive to compare the present work with previous studies on symmetry-controlled spin-orbit torques in low-symmetry transition metal dichalcogenide (TMD)/ferromagnet heterostructures [76–79]. These studies demonstrated that crystal symmetry governs the allowed spin-

orbit-torque components, while changes in the interfacial orientation, layer thickness, or stacking can modify their magnitude and even reverse their sign. They therefore established the fundamental framework of symmetry-controlled unconventional spin-orbit torques for potential field-free switching.

The present work shares two key physical features with these earlier studies. First, lowering the crystal symmetry permits additional current-induced spin-polarization components that are forbidden in higher-symmetry systems, thereby enriching the symmetry-allowed spin response. Second, changing the stacking-related structural polarity reverses selected response components, analogous to the sign reversal of the out-of-plane spin-orbit torque reported for opposite interfacial orientations in WTe₂-based heterostructures [76,77].

Nevertheless, the present system differs fundamentally from these earlier heterostructures in two important aspects. First, unlike WTe₂/FM or MoTe₂/FM heterostructures, where the spin source and ferromagnet are provided by different materials [76–79], bilayer $2H$ -VTe₂ simultaneously hosts intrinsic ferromagnetism and strong SOC within a single van der Waals material. Consequently, no additional metallic ferromagnetic layer is required, and both the current-induced spin accumulation and the magnetic order originate from the same material. Second, in the previously studied TMD/FM systems, the torque symmetry or sign is determined by static structural parameters, such as the crystallographic

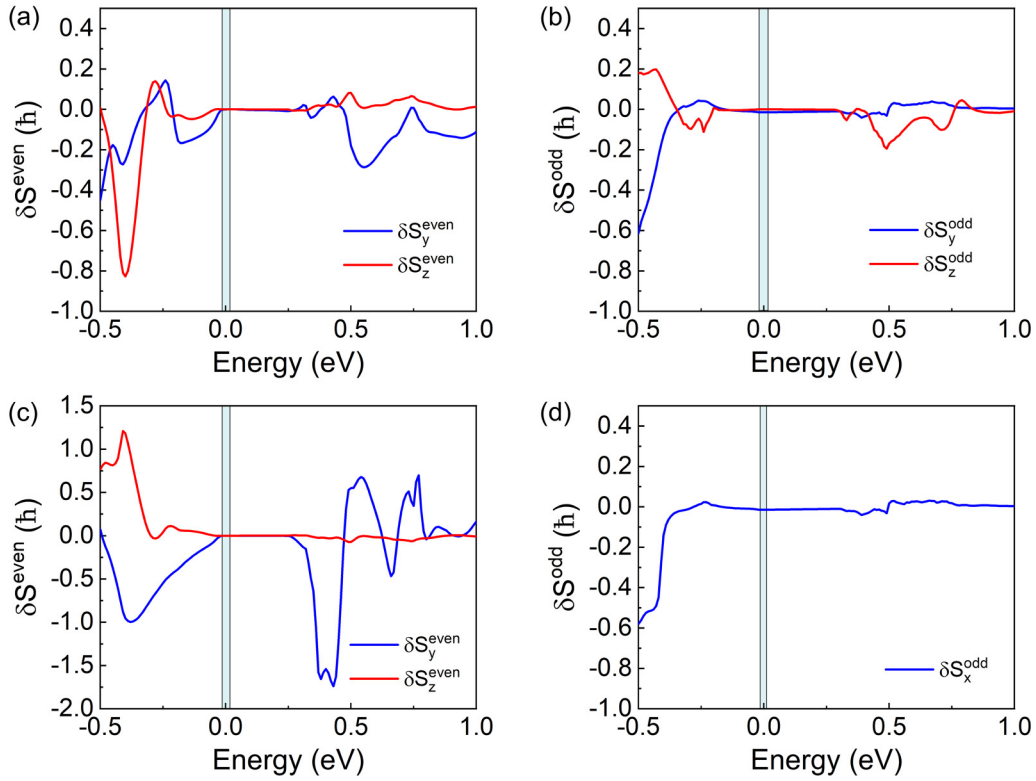


FIG. 19. Calculated symmetry-allowed current-induced spin accumulations in a unit cell in an extended energy range in AB-stacked bilayer $2H\text{-VTe}_2$ under a reference electric field $1 \text{ V/\AA}$ along the $+x$ direction. Panels (a) and (b) show the time-reversal-even and time-reversal-odd components of spin accumulation when magnetization is along the $+x$ direction. Panels (c) and (d) show the time-reversal-even and time-reversal-odd components of spin accumulation when magnetization is along the $+y$ direction. The dashed blue region indicates the band gap. Note that the magnitude of the electric field ($1 \text{ V/\AA}$) is difficult to realize experimentally and is used only as a reference value.

orientation, layer thickness, or interfacial stacking established during device fabrication. Although opposite interfacial orientations were shown to produce opposite torque signs, reversible electrical switching between these configurations has not been demonstrated. In contrast, the AB and BA stackings of bilayer $2H\text{-VTe}_2$ are connected by interlayer sliding and correspond to opposite sliding-ferroelectric polarization

states. As demonstrated in previous work, these two states can, in principle, be reversibly and nonvolatily switched by an external electric field [35], thereby enabling electrical control over both the symmetry and the sign of the current-induced spin response. Such intrinsic and nonvolatile structural control is particularly attractive for programmable spintronic and logic-device applications.

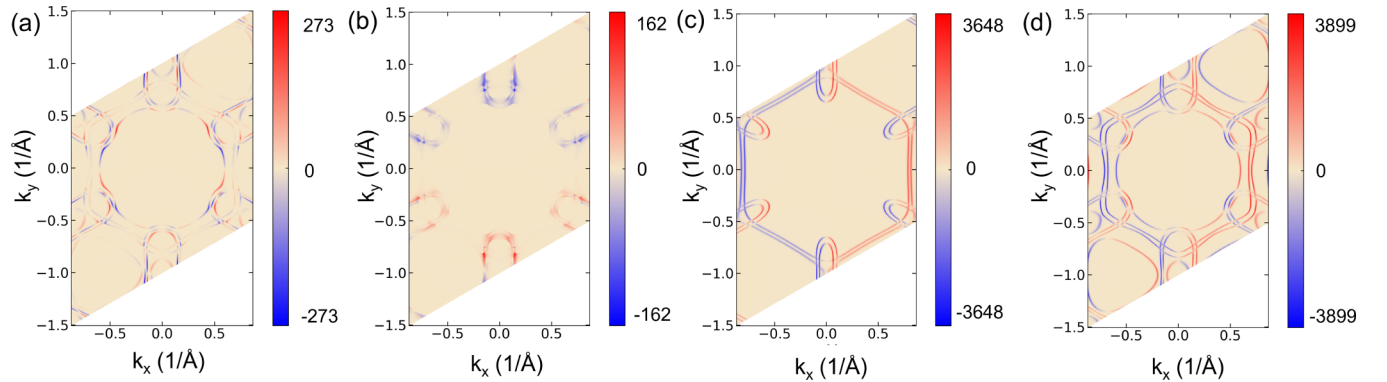


FIG. 20. The k -resolved current-induced spin accumulation under a fixed reference electric field of $1 \text{ V/\AA}$ along the $+x$ direction for bilayer AB-stacked $2H\text{-VTe}_2$. (a) δS_y^{even} for magnetization along $+x$ at $E_F = 0.55 \text{ eV}$; (b) δS_z^{odd} at $E_F = 0.5 \text{ eV}$ for magnetization along $+x$. (c) δS_z^{even} for magnetization along $+y$ at $E_F = 0.4 \text{ eV}$; (d) δS_z^{even} at $E_F = 0.55 \text{ eV}$ for magnetization along $+y$. The red and blue denotes the positive and negative values of spin accumulations, respectively. The relationship between k -resolved spin accumulation and total spin accumulation at a specific energy is $\delta S_i = \frac{(2\pi)^3}{VN} \sum_k \delta S_i(E_F, \mathbf{k})$, where V and N is the volume of the unit cell used in DFT calculation and the total number of k points used to sample the Brillouin zone, respectively. The unit of k -resolved spin accumulation is $\hbar \text{ \AA}^3$.

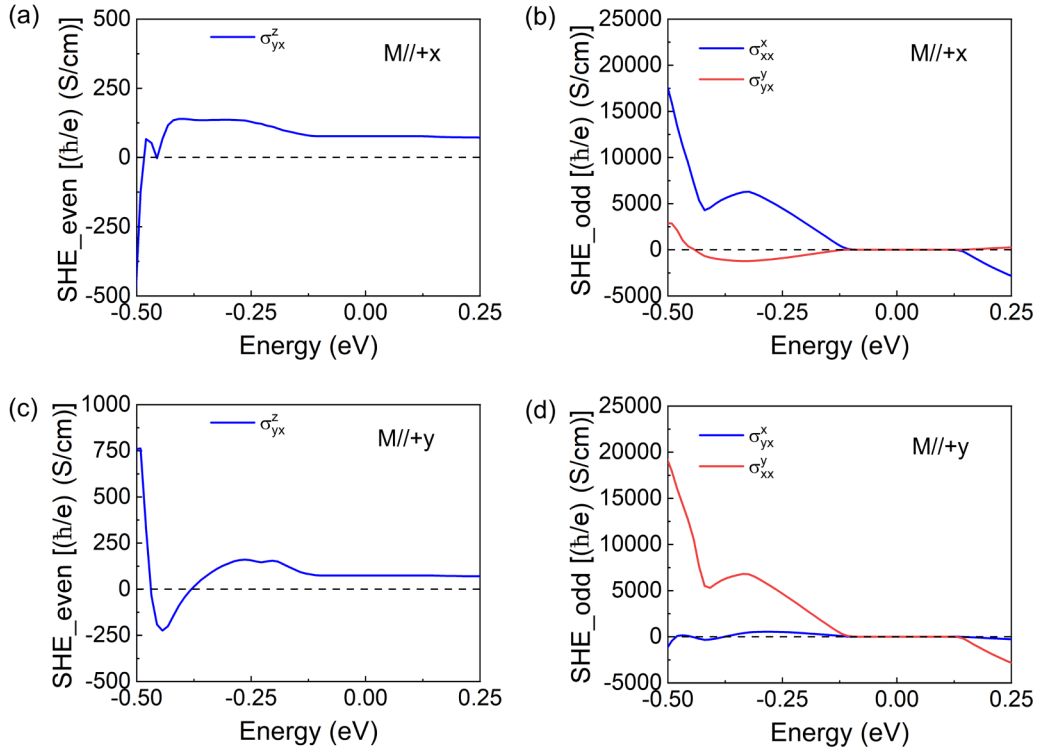


FIG. 21. Symmetry-allowed spin Hall conductivity in monolayer $2H\text{-VTe}_2$. [(a) and (b)] The time-reversal-even and -odd components for $+x$ magnetization, and [(c) and (d)] the time-reversal-even and -odd components for $+y$ magnetization, respectively.

We therefore believe our present work extends the concept of crystal-symmetry-controlled current-induced spin responses from static low-symmetry TMD/ferromagnet

interfaces to electrically switchable, intrinsically magnetic sliding-ferroelectric bilayers, and thus warrants further experimental verification.

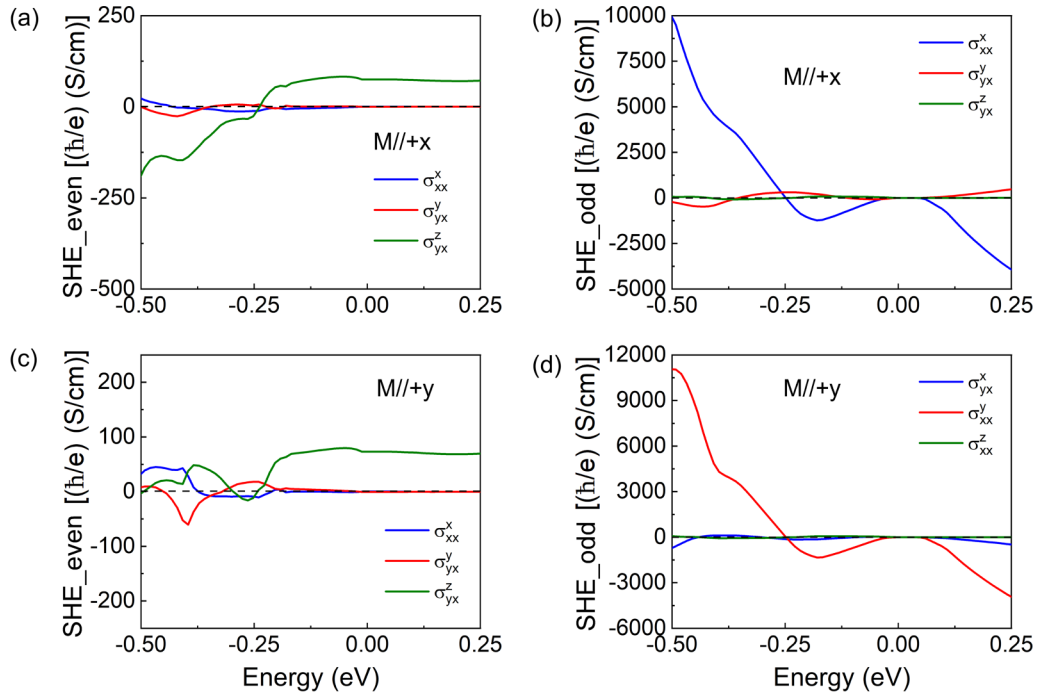


FIG. 22. Symmetry-allowed spin Hall conductivity in AB-stacked bilayer $2H\text{-VTe}_2$. [(a) and (b)] The time-reversal-even and -odd components for $+x$ magnetization, and [(c) and (d)] the time-reversal-even and -odd components for $+y$ magnetization, respectively.

APPENDIX K: DISCUSSION ABOUT MAGNETIC STABILITY

In our work, both monolayer and bilayer $2H$ -VTe₂ possess in-plane magnetic anisotropy. In such easy-plane magnetic anisotropic systems, the long-range in-plane ferromagnetic order might not be established. Instead, the systems are expected to undergo a Berezinskii-Kosterlitz-Thouless transition, below which quasi-long-range magnetic order is established [80]. Nevertheless, this does not forbid the realization of a well-defined ferromagnetic state under experimentally relevant conditions. In realistic samples, several factors can stabilize a preferred in-plane magnetization direction and suppress thermal fluctuations. For example, an external magnetic field can align the magnetization, while magnetic proximity to another material possessing a finite in-plane magnetic anisotropy can effectively pin the magnetic orientation. In addition, applying in-plane uniaxial strain breaks the in-plane rotational symmetry and induces a finite in-plane magnetic anisotropy, thereby selecting a unique easy axis for stabilizing a ferromagnetic state. These experimentally accessible approaches provide practical routes for stabilizing a well-defined ferromagnetic configuration and therefore make the investigation of the current-induced spin response in such a magnetic state physically meaningful.

On the other hand, the negligible intrinsic in-plane magnetic anisotropy also implies that the magnetization direction can be rotated readily by relatively weak external perturbations (e.g., in-plane magnetic field). Consequently, the current-induced spin accumulation, which depends sensitively on the magnetization orientation, becomes highly tunable. We believe that exploring such a tunable spin response is of considerable physical interest, since it not only provides insight into the interplay among magnetic order, spin-orbit coupling, and nonequilibrium spin transport but also offers a potential route toward spintronic devices with flexibly reconfigurable functionalities.

APPENDIX L: ROLE OF SPIN-ORBIT COUPLING

In the present system, SOC plays important roles in two aspects: magnetic anisotropy and the Edelstein effect.

1. Magnetic anisotropy

Magnetic anisotropy is one of the most direct manifestations of SOC in magnetic materials. Microscopically, SOC couples the spin degree of freedom to the anisotropic orbital environment of the crystal and therefore constitutes the

microscopic origin of the MAE. In the absence of SOC, the total energy is invariant under a global rotation of the magnetization. Once SOC is included, different magnetization directions correspond to different SOC matrix elements between occupied and unoccupied electronic states, leading to a finite MAE through second-order perturbation processes [37,81]. Previous first-principles studies systematically compared the magnetic anisotropy of the $2H$ -VX₂ ($X = S, Se, \text{ and } Te$) family and found that all three compounds possess easy-plane magnetic anisotropy [34,82]. More importantly, the magnitude of the MAE, defined as

$$|MAE| = |E_x - E_z|,$$

where E_x and E_z denote the total energies for in-plane and out-of-plane magnetization, respectively, follows the trend

$$|MAE_{VTe_2}| > |MAE_{VSe_2}| > |MAE_{VS_2}|,$$

indicating that the anisotropy becomes progressively stronger from S to Te [34,82]. This trend is consistent with the increasing atomic SOC strength of the chalcogen atoms, suggesting that the stronger SOC associated with Te plays an important role in enhancing the magnetic anisotropy of $2H$ -VTe₂.

2. Edelstein effect

In a system with broken inversion symmetry, SOC generates a momentum-dependent effective spin-orbit field, leading to different spin expectation values for electronic states at different wave vectors, commonly referred to as spin-momentum locking. In equilibrium, the contributions from the occupied states cancel, resulting in no net nonequilibrium spin polarization. When an external electric field is applied, the electronic distribution in momentum space becomes asymmetric. Owing to the SOC-induced spin-momentum locking, this asymmetric occupation also produces an imbalance in the spin distribution, giving rise to a net nonequilibrium spin polarization, namely the Edelstein effect [2]. In general, stronger SOC tends to enhance the spin splitting of the electronic states and can therefore favor a more pronounced Edelstein response. Accordingly, within the $2H$ -MX₂ family, compounds containing heavier M or X elements may exhibit stronger current-induced spin polarization. Indeed, previous calculations reported that the current-induced spin polarization in monolayer $2H$ -VTe₂ is approximately one to two orders of magnitude larger than that in monolayer $2H$ -VSe₂ [29]. This comparison suggests that the stronger SOC associated with Te, together with the resulting modification of the electronic structure, plays an important role in enhancing the Edelstein response of $2H$ -VTe₂.

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