

Article

Electrically Activating and Switching the Magneto-Optic Faraday Effect in 2D Antiferromagnets

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Abstract

Two-dimensional (2D) antiferromagnets are highly promising for next-generation spintronics due to their ultrafast dynamics and robustness against stray fields; however, their practical application is severely hindered by the vanishing magneto-optic effects restricted by strict crystal symmetries. In this work, we propose a universal physical mechanism to activate and manipulate the magneto-optic Faraday effect in 2D fully compensated bilayer antiferromagnets using an external vertical electric field. By constructing a comprehensive tight-binding model and performing first-principles calculations on bilayer VSe₂, we demonstrate that the applied electric field explicitly breaks the spatial inversion and combined $\mathcal{PT}$ symmetries. This symmetry breaking lifts the Kramers degeneracy, inducing a pronounced spin splitting that, in conjunction with intrinsic spin–orbit coupling, generates non-vanishing Berry curvature. Consequently, the previously forbidden Faraday rotation angle is activated from zero to a significant non-zero value, and its rotation direction can be deterministically reversed by switching the electric field polarity. Our findings provide profound physical insights into the symmetry-modulated light–matter interactions and pave the way for designing fully electrically controllable, energy-efficient antiferromagnetic opto-spintronic devices.

Keywords: 2D antiferromagnets; magneto-optic Faraday effect; first-principles calculations

1. Introduction

In the exploration of next-generation high-density and ultrafast spintronic devices, antiferromagnets and fully compensated magnets are gradually replacing traditional ferromagnets as a central focus in condensed matter physics and materials science [1–8]. Compared to ferromagnets, antiferromagnetic systems feature zero macroscopic net magnetization, which fundamentally avoids bit crosstalk caused by stray magnetic fields, enabling extremely high-density information storage [9–13]. Meanwhile, they exhibit strong robustness against external magnetic field perturbations, and their spin dynamic responses typically lie in the terahertz (THz) frequency range, which is three orders of magnitude faster than that of ferromagnets [14–16]. In recent years, with the vigorous development of two-dimensional (2D) magnetic systems, researchers have made substantial breakthroughs in the precise synthesis, magnetic order manipulation, and electrical readout of 2D antiferromagnetic materials [17–21]. These advancements have not only profoundly broadened our physical understanding of low-dimensional magnetism but also laid a



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solid foundation for the development of smaller, faster, and more stable antiferromagnetic spintronic devices [22–26].

Despite the tremendous potential of antiferromagnets in electrical transport and dynamics, achieving non-contact, high-resolution detection and optoelectronic applications of their magnetic states remains a major challenge [27–31]. In this context, magneto-optic effects (such as the magneto-optic Faraday effect), serving as a bridge between magnetism and optics, play an indispensable role in opto-spintronic devices and information reading/writing [32–35]. However, for antiferromagnets and fully compensated magnets, their conventional linear magneto-optic effects are often forbidden by severe physical restrictions. This prohibition stems from specific crystal symmetries universally present in their lattice structures, such as the combined spatial inversion and time-reversal symmetry ($\mathcal{PT}$ symmetry), or the combined two-fold rotation and time-reversal symmetry ($\mathcal{C}_2\mathcal{T}$ symmetry) [36–38]. According to the symmetry principles of physics, these symmetry operations force the system's responses to forward and backward propagating light waves to cancel out exactly, resulting in a macroscopic magneto-optic Faraday rotation angle that is strictly zero [39,40]. Therefore, effectively breaking these inherent symmetry restrictions to activate and manipulate the magneto-optic effect in fully compensated antiferromagnetic systems has become a critical obstacle to overcome in the current field of antiferromagnetic optoelectronics.

In this work, we reveal a universal physical mechanism for manipulating the magneto-optic response of 2D antiferromagnetic materials using an external electric field, and conduct an in-depth study using bilayer VSe₂ as a representative material (which exhibits ferromagnetism in the monolayer limit but becomes antiferromagnetic in the bilayer form due to interlayer coupling) [41–43]. Our results demonstrate that without an applied electric field, the two layers of bilayer VSe₂ are strictly constrained by crystal symmetries such as $\mathcal{PT}$ and $\mathcal{C}_2\mathcal{T}$, rendering its magneto-optic Faraday effect completely silent (i.e., the Faraday angle is zero). However, upon introducing an external electric field perpendicular to the 2D plane, the electric field directly and effectively breaks the spatial symmetry connection between the layers. This symmetry breaking disrupts the original optical response balance, successfully realizing the activation of the magneto-optic Faraday effect from zero to a significant non-zero Faraday rotation angle. More importantly, we demonstrate that simply switching the polarity (positive/negative direction) of the perpendicular electric field can directly reverse the direction of the Faraday rotation angle. Our study not only provides profound physical insights into the microscopic mechanism of symmetry and light–matter interactions in 2D antiferromagnetic systems but also offers a practical and universal approach for designing fully electrically controlled magneto-optic switches and next-generation antiferromagnetic optoelectronic information processing devices based on compensated magnets.

2. Results

2.1. Calculations Details

First-principles calculations for VSe₂ were carried out within density functional theory using the Perdew–Burke–Ernzerhof (PBE) functional under the generalized gradient approximation, as implemented in the Vienna ab initio Simulation Package (VASP) [44,45]. A $12 \times 12 \times 1$ k -point mesh was employed to sample the Brillouin zone, with a plane-wave cutoff energy of 600 eV. Structural relaxations incorporated interlayer van der Waals interactions through the DFT-D3 correction [46]. The on-site Hubbard parameter for V was set to $U = 2.7$ eV. Symmetry-adapted Wannier functions were generated using VASP in conjunction with the WANNIER90 package [47].

The magneto-optical Faraday effect was evaluated from the frequency-dependent optical conductivity tensor. For normal incidence, the eigenmodes in the magnetic medium

are left- and right-circularly polarized light. Their corresponding complex refractive indices are given by

$$n_{\pm}(\omega) = \sqrt{\varepsilon_{xx}(\omega) \pm i\varepsilon_{xy}(\omega)}, \quad (1)$$

where n_+ and n_- represent the refractive indices of the two circularly polarized modes, respectively. Because of the nonzero off-diagonal dielectric-tensor component ε_{xy} , the two circularly polarized waves acquire different phases and amplitudes while propagating through the material. Consequently, an incident linearly polarized wave becomes elliptically polarized after transmission.

For a film with thickness d , the exact relation can be expressed as

$$\frac{1 - \tan \eta_F}{1 + \tan \eta_F} \exp(2i\theta_F) = \exp\left[\frac{i\omega d}{c}(n_+ - n_-)\right], \quad (2)$$

where c denotes the speed of light in vacuum. When the Faraday rotation and ellipticity are small, the complex Faraday angle can be approximated as

$$\phi_F(\omega) = \frac{\omega d}{2c}[n_+(\omega) - n_-(\omega)]. \quad (3)$$

Accordingly, the real and imaginary parts of ϕ_F correspond to the Faraday rotation angle and ellipticity, respectively:

$$\theta_F(\omega) = \text{Re}[\phi_F(\omega)]. \quad (4)$$

2.2. Symmetry Analysis and Theoretical Foundation for Electric-Field-Induced Magneto-Optic Effects

In exploring the physical origin of magneto-optic effects in two-dimensional (2D) bilayer antiferromagnetic systems, crystal symmetry plays a decisive role. According to Neumann's principle, any macroscopic physical property of a material must be invariant under all symmetry operations of its point group. For an ideal 2D bilayer fully compensated antiferromagnet (e.g., bilayer VSe₂), in the absence of external perturbations, the upper and lower layers possess magnetic moments of equal magnitude but opposite directions. In this pristine state, although the system as a whole breaks time-reversal symmetry ($\mathcal{T}$), it is often protected by a joint symmetry due to strict geometric symmetry operations connecting the two layers, such as in-plane two-fold rotation ($\mathcal{C}_2$), spatial inversion ($\mathcal{P}$), or interlayer mirror symmetry ($\mathcal{M}$). For instance, the spatial inversion $\mathcal{P}$ maps the spin sites of the upper layer to the lower layer, while time-reversal $\mathcal{T}$ flips the antiparallel spins. Therefore, the entire bilayer system remains invariant under the combined operation $\mathcal{PT}$ (or similarly, $\mathcal{C}_2\mathcal{T}$, $\mathcal{MT}$). It is precisely this strict symmetry that forces the system's responses to forward- and backward-propagating light to cancel out exactly, thereby strictly restricting the macroscopic observables of the anomalous Hall effect (AHE) and the magneto-optic Faraday effect to zero.

To activate these forbidden physical effects in such systems, an external field must be introduced to break the spatial symmetries connecting the upper and lower layers. Applying an external gate electric field (E_z) perpendicular to the 2D plane provides a highly precise and efficient tuning method. A vertical electric field is inherently a polar vector, which changes sign (i.e., reverses direction) under spatial inversion $\mathcal{P}$ and in-plane rotation $\mathcal{C}_2$ operations. Consequently, when an external field $E_z \neq 0$ is applied, it introduces an electrostatic potential difference between the upper and lower layers (i.e., the Stark effect). This interlayer electrostatic asymmetry directly and completely breaks all lattice symmetry operations that can map the upper layer to the lower layer (including $\mathcal{P}$, $\mathcal{C}_2$, and $\mathcal{M}$).

The complete breaking of symmetry triggers a profound physical phase transition. Since the upper and lower layers are no longer physically equivalent under the electric field,

the originally degenerate interlayer electronic band structures undergo relative shifting and reconstruction. As a result, the symmetry relating the sublattices with opposite spin orientations is broken, leading to a transition of the system from a fully compensated antiferromagnetic state to a fully compensated ferrimagnetic state with a non-zero net magnetic moment.

With the destruction of spatial lattice symmetries ($\mathcal{P}$, $\mathcal{C}_2$, $\mathcal{M}$), the original combined symmetry groups (such as $\mathcal{PT}$ or $\mathcal{C}_2\mathcal{T}$) also collapse. The lifting of these symmetry restrictions allows the Berry curvature, which is associated with time-reversal symmetry breaking, to yield a non-zero integral in momentum space. Consequently, macroscopic physical properties that were strictly forbidden in the pristine antiferromagnetic state can now emerge (Figure 1). On the one hand, the system can exhibit an intrinsic anomalous Hall current; on the other hand, the off-diagonal elements of the dielectric tensor become non-zero, thereby directly activating the magneto-optic Faraday effect in the optical frequency range. In summary, by breaking the interlayer symmetry via an electric field, we not only achieve a magnetic phase transition from an antiferromagnet to a fully compensated ferrimagnet but also theoretically establish a solid foundation for the electrically controllable “zero to non-zero” magneto-optic Faraday effect.

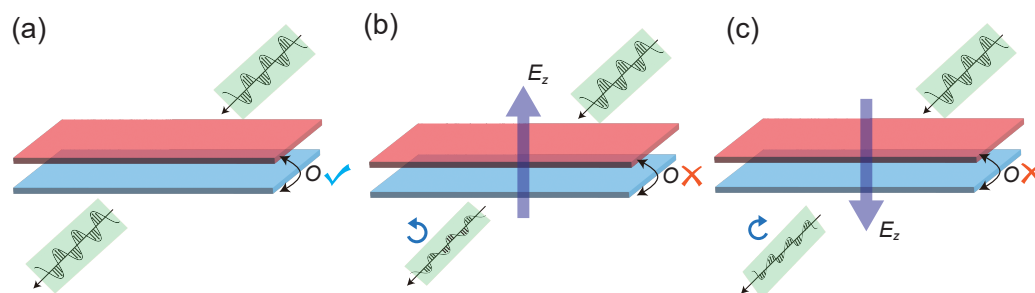


Figure 1. Schematic illustration of the electric-field-tunable magneto-optic Faraday effect in 2D bilayer antiferromagnets. (a) In the absence of an external electric field ($E = 0$), the system exhibits a fully compensated bilayer antiferromagnetic ground state, formed by two ferromagnetic monolayers with opposite magnetizations. The top and bottom layers are geometrically connected by a specific spatial symmetry operation $\mathcal{O}$ (e.g., spatial inversion $\mathcal{P}$). Protected by this symmetry constraint, the macroscopic magneto-optic Faraday rotation angle is strictly zero ($\theta_F = 0$). (b,c) The scenarios under an applied vertical electric field pointing (b) upwards ($+E_z$) and (c) downwards ($-E_z$). The electric-field-induced interlayer potential difference explicitly breaks the symmetry operation $\mathcal{O}$. This symmetry breaking successfully activates the magneto-optic Faraday effect ($\theta_F \neq 0$). Notably, switching the direction of the applied electric field deterministically reverses the rotation direction (sign) of the Faraday angle.

2.3. Tight-Binding Model

To investigate the microscopic mechanism of the electric-field-induced magneto-optic effect, we construct an 8×8 tight-binding (TB) Hamiltonian for the bilayer antiferromagnetic system. The basis is chosen as $\Psi = (\Psi_{\text{top}}, \Psi_{\text{bot}})^T$, where each layer includes two sublattices and two spin degrees of freedom (e.g., $|A \uparrow\rangle, |A \downarrow\rangle, |B \uparrow\rangle, |B \downarrow\rangle$). The total Hamiltonian can be written in a block-matrix form:

$$\mathcal{H}(\mathbf{k}, k_z) = \begin{pmatrix} H_{\text{top}}(\mathbf{k}) & H_{\text{int}}(k_z) \\ H_{\text{int}}^\dagger(k_z) & H_{\text{bot}}(\mathbf{k}) \end{pmatrix} \quad (5)$$

Here, $H_{\text{int}}(k_z) = t_i e^{ik_z d_z} \mathbb{I}_{4 \times 4}$ represents the interlayer coupling, where t_i is the interlayer hopping amplitude, $d_z = 1/10$ is the effective interlayer distance, and $\mathbb{I}_{4 \times 4}$ is the 4×4 identity matrix.

The intralayer Hamiltonians $H_{\text{top}}(\mathbf{k})$ and $H_{\text{bot}}(\mathbf{k})$ are 4×4 matrices given by

$$H_{\text{top}}(\mathbf{k}) = \begin{pmatrix} E_A & T(\mathbf{k}) \\ T^\dagger(\mathbf{k}) & E_B \end{pmatrix}, \quad H_{\text{bot}}(\mathbf{k}) = \begin{pmatrix} E_B & T(\mathbf{k}) \\ T^\dagger(\mathbf{k}) & E_{A'} \end{pmatrix} \quad (6)$$

where the diagonal energy blocks are $E_A = \text{diag}(e_1, e_2)$, $E_B = \text{diag}(e_3, e_3)$, and $E_{A'} = \text{diag}(e_2, e_1)$. The energy splitting in E_A and its exact reversal in $E_{A'}$ ($e_1 \leftrightarrow e_2$) perfectly capture the $\mathcal{PT}$ -symmetric antiferromagnetic order between the two layers.

The intralayer hopping matrix $T(\mathbf{k})$ describes the transitions between the two sublattices and is given by

$$T(\mathbf{k}) = \begin{pmatrix} f(\mathbf{k}) & g_1(\mathbf{k}) \\ g_2(\mathbf{k}) & f(\mathbf{k}) \end{pmatrix} \quad (7)$$

To express the momentum-dependent hopping functions elegantly, we define three nearest-neighbor vectors $\delta_1 = (\frac{1}{3}, \frac{2}{3})$, $\delta_2 = (\frac{1}{3}, -\frac{1}{3})$, and $\delta_3 = (-\frac{2}{3}, -\frac{1}{3})$. The spin-conserving hopping function $f(\mathbf{k})$ is expressed as

$$f(\mathbf{k}) = t \sum_{j=1}^3 e^{i\mathbf{k} \cdot \delta_j} \quad (8)$$

The spin-flipping terms induced by spin-orbit coupling (SOC), denoted as $g_1(\mathbf{k})$ and $g_2(\mathbf{k})$, can be written compactly using complex phase factors:

$$g_1(\mathbf{k}) = t'_2 \left(e^{-i\pi/2} e^{i\mathbf{k} \cdot \delta_1} + e^{i\pi/6} e^{i\mathbf{k} \cdot \delta_2} + e^{i5\pi/6} e^{i\mathbf{k} \cdot \delta_3} \right) \quad (9)$$

$$g_2(\mathbf{k}) = t'_2 \left(e^{-i\pi/2} e^{i\mathbf{k} \cdot \delta_1} + e^{i5\pi/6} e^{i\mathbf{k} \cdot \delta_2} + e^{i\pi/6} e^{i\mathbf{k} \cdot \delta_3} \right) \quad (10)$$

where $t'_2 = \frac{2}{\sqrt{3}} t_2$ is the effective SOC hopping amplitude. This structured decomposition significantly clarifies the underlying physical processes, clearly separating the symmetric hopping (t) from the chiral phase-dependent SOC hoppings (t_2).

The lattice structure and the corresponding interatomic hoppings are schematically illustrated in Figure 2a. The system can be effectively described as a bilayer hexagonal lattice, where each layer contains one magnetic atom per unit cell. To reflect the antiferromagnetic nature, the local magnetic moments of the upper and lower layers are set to be strictly anti-aligned. Geometrically, the two magnetic sublattices—residing in the top and bottom layers with opposite spins—are mapped onto each other by the spatial inversion operation $\mathcal{P}$. Given that the time-reversal operation $\mathcal{T}$ flips the spin orientation, the system preserves the combined $\mathcal{PT}$ symmetry. It is precisely this strict $\mathcal{PT}$ symmetry that guarantees the model represents a perfectly compensated antiferromagnet.

To investigate the fundamental electronic properties of this antiferromagnet, we first analyze the energy spectrum in the absence of spin-orbit coupling (SOC) by setting the spin-flipping hopping parameter $t_2 = 0$. The on-site energies and hopping amplitudes in the tight-binding model are parameterized as $e_1 = 0$, $e_2 = -2$, $e_3 = 1$, $t = 0.8$, and the interlayer coupling $t_i = 0.2$. The calculated electronic band structure is presented in Figure 2b. As expected from Kramers' theorem extended by the spatial inversion, the strict protection of the $\mathcal{PT}$ symmetry enforces a perfect degeneracy between the spin-up and spin-down energy bands throughout the entire Brillouin zone.

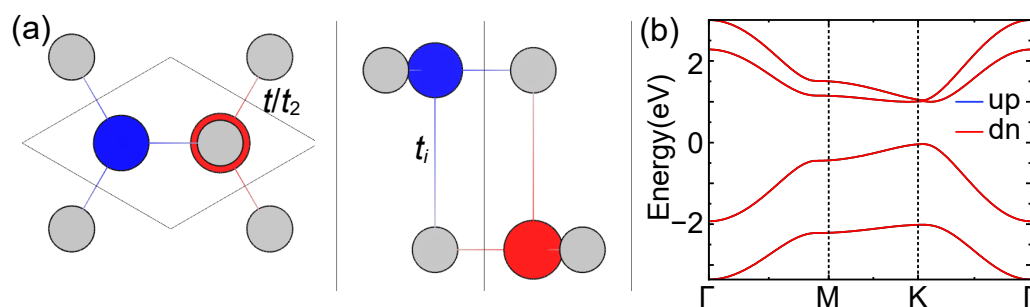


Figure 2. Tight-binding model and its initial electronic band structure. (a) Schematic representation of the lattice sites and interatomic hopping pathways in the bilayer model. The parameters t and t_2 denote the spin-conserving and spin-flipping (induced by spin–orbit coupling) intralayer nearest-neighbor hopping amplitudes, respectively. The parameter t_i represents the vertical interlayer hopping amplitude between the top and bottom layers. (b) The calculated electronic band structure of the model. The band structures of spin up and spin down are completely overlap.

When a vertical gate electric field is applied to the system, it induces an electrostatic potential difference between the upper and lower layers. In our tight-binding model, this electric-field-induced perturbation is rigorously captured by adding a Stark potential term, $\mathcal{H}_E = V_z(\sigma_z \otimes \mathbb{I}_{4 \times 4})$, to the pristine Hamiltonian. Here, V_z represents the electrostatic potential energy modulated by the electric field, σ_z is the Pauli matrix acting on the layer pseudospin space (assigning on-site energies of $+V_z$ and $-V_z$ to the top and bottom layers, respectively), and $\mathbb{I}_{4 \times 4}$ is the 4×4 identity matrix acting on the sublattice and spin degrees of freedom. This interlayer potential difference acts as a symmetry-breaking perturbation that directly and completely destroys all spatial symmetry operations connecting the two layers, such as the spatial inversion $\mathcal{P}$ and the in-plane two-fold rotation $\mathcal{C}_2$. Consequently, the combined $\mathcal{PT}$ symmetry is entirely lifted. At this point, the electronic structure of the system no longer maintains its strictly compensated antiferromagnetic characteristics; instead, it transitions into a fully compensated ferrimagnetic state. Figures 3a and 3b display the calculated band structures under positive ($+V_z$) and negative ($-V_z$) vertical electric fields, respectively. It is clearly visible that the destruction of $\mathcal{PT}$ symmetry lifts the initial Kramers degeneracy, inducing a pronounced spin splitting in the energy bands. Interestingly, when the direction of the applied electric field is reversed, the spin splitting is exactly flipped, indicating that the spin polarization is tightly coupled to the polarity of the external electric field.

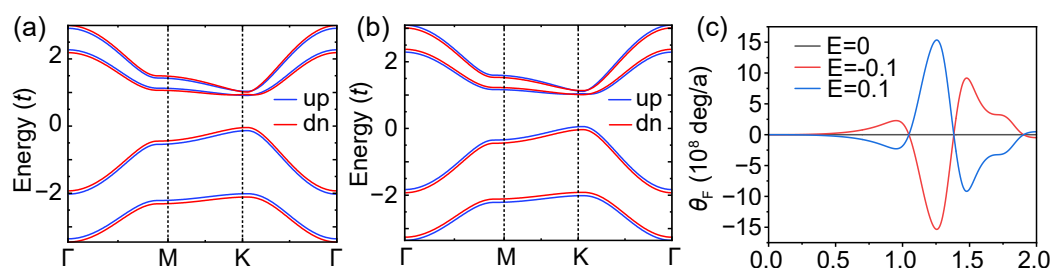


Figure 3. Electric-field-induced spin splitting and the activation of the magneto-optic Faraday effect. (a,b) Calculated electronic band structures of the tight-binding model under an applied vertical electric field pointing (a) upwards ($+E_z$) and (b) downwards ($-E_z$). (c) The calculated magneto-optic Faraday rotation angle (θ_F) of the system with spin–orbit coupling included ($t_2 = 0.2$).

Building upon the symmetry-breaking-induced spin splitting, we further incorporate the spin–orbit coupling (SOC) effect by setting $t_2 \neq 0$ to evaluate the magneto-optic response of the system. In the absence of an applied electric field ($V_z = 0$), the magneto-optic Faraday rotation angle remains strictly zero (Figure 3c), which is dictated by the

preserved $\mathcal{PT}$ and $\mathcal{C}_2\mathcal{T}$ symmetries of the pristine lattice. However, upon the application of a vertical electric field ($V_z \neq 0$), the interplay between the electric-field-induced spin splitting and the intrinsic SOC generates non-vanishing Berry curvature in momentum space. This topological contribution successfully activates the magneto-optic Faraday effect, transitioning the Faraday angle from zero to a significant non-zero value. More importantly, our calculations demonstrate that the rotation direction of the Faraday angle can be deterministically controlled: simply switching the polarity of the external electric field perfectly reverses the direction of the Faraday rotation angle (Figure 3c). This finding provides direct and robust theoretical evidence for the realization of fully electrically controllable, reversible magneto-optic switches in 2D compensated magnets.

2.4. Material Realization in Bilayer VSe₂

To substantiate the physical insights derived from the tight-binding model and verify the universality of our theory in realistic materials, we performed first-principles calculations based on density functional theory (DFT) on bilayer VSe₂. While monolayer VSe₂ exhibits intrinsic ferromagnetism, its bilayer counterpart favors an interlayer fully compensated antiferromagnetic ground state driven by the interlayer antiferromagnetic exchange coupling, as illustrated in Figure 4a. Symmetry analysis reveals that the antiferromagnetic bilayer VSe₂ belongs to the magnetic space group $P\bar{3}'m'1$ (No. 164.88). Crucially, this specific structure strictly preserves the combined spatial inversion and time-reversal symmetry ($\mathcal{PT}$). Under this symmetry operation, the magnetic vanadium (V) atoms located in the top and bottom layers are geometrically mapped onto each other, possessing local magnetic moments of equal magnitude but strictly opposite directions. This rigorous crystallographic and magnetic symmetry leaves a distinct signature on the electronic band structure. As shown in Figure 4b, the DFT-calculated band structure in the absence of an applied electric field exhibits perfect degeneracy between the spin-up and spin-down bands across the entire Brillouin zone, which is a hallmark of $\mathcal{PT}$ -symmetric compensated antiferromagnets.

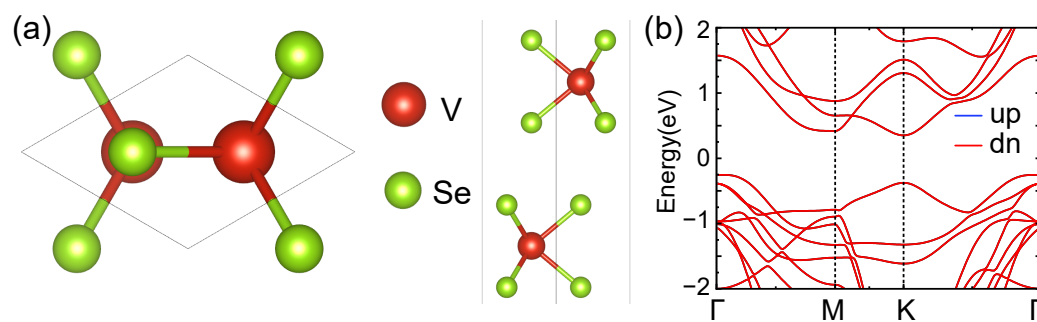


Figure 4. Crystal structure and electronic band structure of bilayer VSe₂ obtained from first-principles calculations. (a) Top and side views of the fully compensated antiferromagnetic bilayer VSe₂ crystal structure. (b) Band structure of bilayer VSe₂. The band structures of spin up and spin down are completely overlap.

Subsequently, we introduced a vertical external electric field in our first-principles calculations. In perfect agreement with the tight-binding model predictions, the applied electric field induces an interlayer electrostatic potential difference that breaks the spatial inversion symmetry $\mathcal{P}$, thereby completely lifting the $\mathcal{PT}$ symmetry constraint. Driven by this symmetry breaking, the originally degenerate energy bands undergo a pronounced spin splitting. Moreover, our calculations confirm that simply reversing the direction of the vertical electric field perfectly switches the polarity of the spin splitting (Figure 5a,b).

By further incorporating the spin–orbit coupling (SOC) effect, we quantitatively evaluated the magneto-optic Faraday responses of bilayer VSe₂ under varying electric field

strengths. At zero electric field, the macroscopic symmetries strictly restrict the Faraday rotation angle to zero (Figure 5c). However, upon the application of a vertical electric field, the Faraday angle immediately transitions from zero to a significant non-zero value (Figure 5c). Notably, as the magnitude of the electric field increases, the absolute value of the Faraday rotation angle exhibits a monotonic increase. Furthermore, the rotation direction (i.e., the sign) of the Faraday angle is deterministically controlled by the direction of the applied electric field. These first-principles results not only provide robust material-level validation for our theoretical mechanism but also highlight the immense potential of bilayer antiferromagnetic VSe₂ for designing highly tunable, fully electrically controlled magneto-optic modulators.

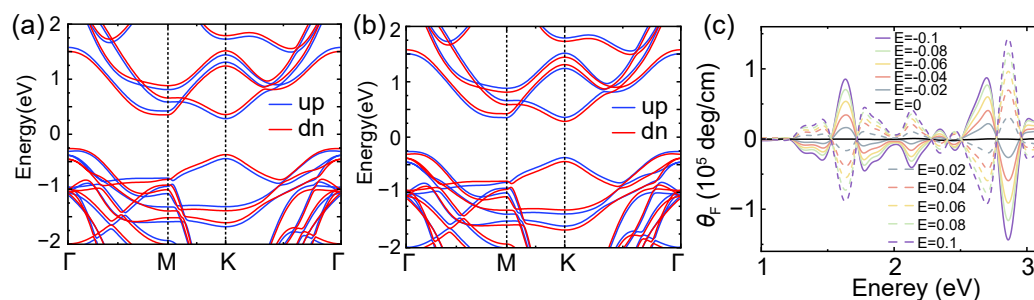


Figure 5. Electric-field-induced spin splitting and the tunable magneto-optic Faraday effect in bilayer VSe₂. (a,b) First-principles calculated spin-projected electronic band structures of bilayer VSe₂ under an applied vertical electric field pointing (a) upwards (+E_z) and (b) downwards (−E_z). (c) The calculated magneto-optic Faraday rotation angle (θ_F) spectra of bilayer VSe₂ under varying electric field strengths, with spin-orbit coupling (SOC) included.

3. Conclusions

In summary, we have theoretically and computationally demonstrated the electric-field-induced activation and reversible tuning of the magneto-optic Faraday effect in 2D fully compensated bilayer antiferromagnets. Using a symmetry-based tight-binding model, we revealed that a vertical gate electric field breaks the inherent spatial inversion ($\mathcal{P}$) and joint $\mathcal{PT}$ symmetries, triggering a phase transition from an antiferromagnetic state to a fully compensated ferrimagnetic state. The interplay between the electric-field-induced spin splitting and intrinsic spin-orbit coupling generates non-zero Berry curvature, which subsequently activates the macroscopic Faraday effect. These theoretical predictions are rigorously corroborated by first-principles calculations on bilayer VSe₂. We show that the magnitude of the Faraday rotation angle increases monotonically with the electric field strength, and its sign can be perfectly switched by reversing the field direction. This work not only broadens our fundamental understanding of symmetry-broken optical responses in low-dimensional magnetism but also establishes a practical paradigm for developing high-density, ultrafast, and fully electrically tunable magneto-optic modulators based on 2D compensated magnets.

Author Contributions: L.Z.: writing—original draft preparation, conceptualization, methodology, validation, visualization; C.L.: writing—review and editing, visualization; C.G.: conceptualization, supervision, writing—review and editing, project administration. All authors have read and agreed to the published version of the manuscript.

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