

Magnetic Routes to Valley Polarization in Monolayer Transition Metal Dichalcogenides

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(Dated: October 2026)

Monolayer transition metal dichalcogenides (TMDs) host two inequivalent valleys at the K and K' points of the Brillouin zone, offering a binary degree of freedom that can encode information. Lifting the degeneracy between these valleys is the central challenge of valleytronics. In this review, we present the theoretical foundations and experimental progress of magnetic routes to valley polarization in monolayer TMDs. We first develop the relevant theory, including the valley Zeeman effect, spin-valley locking, and the valley Hall effect, before surveying four magnetic mechanisms: external magnetic fields, magnetic proximity effects, magnetic doping, and intrinsic ferrovalley materials. For each mechanism, we compare reported valley splitting energies and operating conditions across a range of material systems. External fields applied to monolayer MX_2 yield modest splittings, while magnetic proximity coupling and doping can enhance the splitting by one to three orders of magnitude. Intrinsic ferrovalley materials such as H-VSe_2 bypass external magnetization entirely, exhibiting spontaneous valley polarization with splittings reaching hundreds of meV and Curie temperatures well above room temperature. We conclude with an outlook on the prospects and remaining challenges for integrating valley polarization into practical information-encoding devices.

Keywords: valleytronics, valley polarization, transition metal dichalcogenides, valley Zeeman effect, magnetic proximity effect, magnetic doping, ferrovalley materials, spin-valley locking

I. INTRODUCTION

Finding new quantum degrees of freedom of electrons has been a widespread goal in condensed matter physics research. Historically, applications of every new quantum degree of freedom often lead to significant development in the science and technology industries [1]. The development of modern electronics and semiconductor devices (such as the transistor) relied on the manipulation of the charge degree of freedom of electrons. Later, the field of spintronics focused on manipulating the spin degree of freedom to encode information in electronic devices such as magnetic random-access memory (MRAM) and magnetic tunnel junctions (MTJs). The manipulation of the charge and spin degrees of freedom kickstarted a drive to miniaturize devices and improve processing speeds by orders of magnitude, but in reaching the thermodynamic and quantum limitations of these approaches, researchers have begun investigating a third degree of freedom for the electron: the “valley” degree of freedom [1]. In this paper, we discuss this third degree of freedom, including reviewing the relevant theory and discussing possible applications of it in valleytronic devices that encode information.

In a crystalline solid, electrons do not have arbitrary energies; instead, their allowed energies form bands that depend on the electron’s crystal momentum $\mathbf{k}$, giving rise to the energy dispersion $\varepsilon_n(\mathbf{k})$ in the Brillouin zone.

In general, valleys refer to local minima and maxima in the crystal’s conduction and valence band structure. Typically, the band extrema (valleys) occur at the inequivalent K and K' points of the Brillouin zone in two-dimensional hexagonal transition metal dichalcogenides (TMDs), shown in Fig. 1. Electrons occupying the K and K' valleys form a two-state system known as the valley degree of freedom, often called valley pseudospin. It is

termed a *pseudospin* because the valley index behaves mathematically like a spin- $\frac{1}{2}$ two-level system—it can be described by the same Pauli-matrix formalism—even though it originates from the two inequivalent valleys rather than from intrinsic angular momentum. As one might note, the K and K' points alternate around the hexagonal lattice.

This review primarily discusses magnetic routes to valley polarization in nonmagnetic monolayer TMDs (MX_2 , where $M = \text{Mo}, \text{W}$ and $X = \text{S}, \text{Se}, \text{Te}$), though we also examine how intrinsically magnetic 2D TMDs exploit their own exchange interactions to spontaneously lift valley degeneracy.

In monolayer TMDs, the two inequivalent valleys are not only separated in momentum space, but also possess distinct geometric properties associated with the electron’s wavefunction. The K and K' valleys hold opposite Berry curvatures $\Omega_n(\mathbf{k})$, and opposite electrical, magnetic, and optical properties [1, 3].

More importantly, such monolayer materials exhibit time-reversal symmetry ($\mathcal{T}$), enforcing

$$\Omega_n(-\mathbf{k}) = -\Omega_n(\mathbf{k}).$$

In comparison, spatial inversion symmetry ($\mathcal{P}$) enforces

$$\Omega_n(\mathbf{k}) = \Omega_n(-\mathbf{k}),$$

which is broken in MX_2 .

Note that when a material possesses both time-reversal and spatial inversion symmetry, $\Omega_n(\mathbf{k}) = 0$. Monolayer graphene is an example of a material possessing both symmetries.

As a consequence of these valley-dependent properties, electrons in the K and K' valleys interact differently with light. Theory predicted, and experiments later confirmed, that σ^+ and σ^- circularly polarized light excite carriers

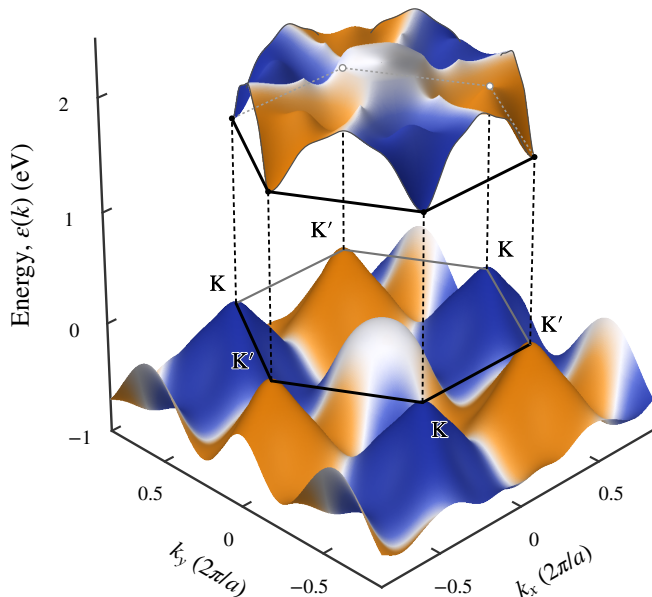


Figure 1. Electronic band structure of a 2D transition metal dichalcogenide: the highest valence band and lowest conduction band of monolayer MoS₂ over the $\mathbf{k}$ -plane, computed from the three-band tight-binding model of Ref. [2] (GGA parameters, without spin-orbit coupling). Direct gaps open at the K and K' corners of the hexagonal Brillouin zone. Blue and orange mark the K and K' valleys, and the conduction band is cut off at 2.3 eV near Γ .

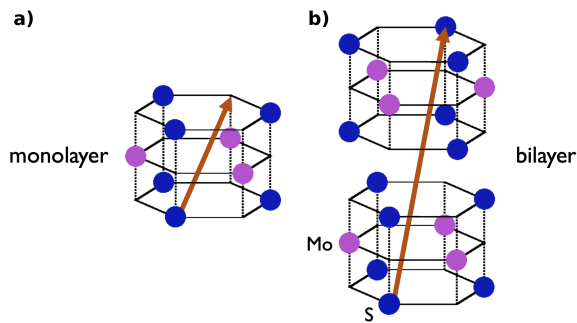


Figure 2. (a) Breaking of spatial inversion symmetry in monolayer MoS₂. (b) Bilayer of MoS₂ with preserved spatial inversion symmetry. Reproduced from Ref. [4] with permission of the author.

in the K and K' valleys, respectively [5, 6]. This valley-selective excitation enables the optical generation and control of valley polarization in monolayer TMDs.

These valley-dependent optical properties are only one manifestation of the unique electronic structure of monolayer TMDs. The interplay between Berry curvature, orbital magnetic moments, and spin-orbit coupling gives rise to several phenomena including the valley Zeeman effect, spin-valley locking, and the valley Hall effect. Understanding and controlling these effects is essential for realizing stable valley polarization and practical valleytronics devices [6].

This review specifically focuses on magnetic mecha-

nisms that lift valley degeneracy and enable controllable valley polarization in monolayer TMDs.

II. THEORETICAL FRAMEWORK FOR MAGNETIC VALLEY POLARIZATION

The treatment developed throughout this section is based on the reviews and foundational works of Refs. [1, 6, 7], and further detail may be found by consulting them.

II.A. Terminology

For a monolayer TMD, the two inequivalent valleys are labeled by their valley index:

$$\tau = +1 (K), \quad \tau = -1 (K').$$

Spin-orbit coupling (SOC) is the interaction between an electron's spin and its orbital motion within the crystal potential. In monolayer TMDs, SOC splits the valence bands by hundreds of meV and is responsible for spin-valley locking. In monolayer MX₂, strong spin-orbit coupling pins the electron spin orientation to the valley index: in the topmost valence band, spin-up states reside predominantly at K and spin-down states at K' , while in the conduction band this ordering is reversed. This coupling, known as *spin-valley locking*, is a direct consequence of broken $\mathcal{P}$ -symmetry together with time-reversal symmetry.

The *valley splitting energy*, $\Delta E_{\text{valley}} = E_K - E_{K'}$, quantifies the energy difference between the K and K' valley band edges. When $\Delta E_{\text{valley}} \neq 0$, valley degeneracy is lifted. The primary mechanism for achieving this in MX₂ is the breaking of $\mathcal{T}$ -symmetry, which enables $E_K \neq E_{K'}$.

Valley polarization measures how unevenly carriers occupy the two valleys. If n_K and $n_{K'}$ denote the carrier populations in the K and K' valleys, respectively, then the valley polarization P_v is defined as

$$P_v = \frac{n_K - n_{K'}}{n_K + n_{K'}}. \quad (1)$$

When $P_v = 0$, both valleys are equally populated; when $P_v = \pm 1$, all carriers reside in the K or K' valley, respectively. Prolonged valley polarization is of great value because it enables the potential development of valleytronic devices. However, Luo *et al.* report that the lifetime of this unbalanced distribution is short, posing an obstacle to utilizing the valley degree of freedom for stable information processing [1].

An *exciton* is a bound electron-hole pair created by optical excitation. In magneto-photoluminescence experiments, shifts in exciton emission energies under σ^+ and σ^- circular polarization are used to measure valley splitting.

Many magnetic substrates used in valleytronic heterostructures are ferromagnetic materials. In ferromagnets, the magnetic moments of electrons align parallel

to each other due to exchange interactions, producing a net magnetization even in the absence of an external magnetic field. The *Curie temperature*, T_C , is the critical temperature above which a ferromagnetic material loses its spontaneous magnetization and becomes paramagnetic. For valleytronic applications, a high T_C is desirable because it ensures that spontaneous valley polarization persists at or near room temperature—an important practical consideration, since electronic components generate substantial heat during operation.

II.B. Theory of the Valley Hall Effect

We begin with the concept of the Berry curvature, $\Omega_n(\mathbf{k})$, which plays a key role in understanding the valley Hall effect.

To understand the Berry curvature, we must start with the Berry connection. In a crystal, electron wavefunctions $\psi_{n\mathbf{k}}$ are Bloch states that can be written in the form

$$\psi_{n\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r})$$

where $u_{n\mathbf{k}}(\mathbf{r})$ is the periodic part of the Bloch wavefunction.

The Berry connection $\mathbf{A}_n(\mathbf{k})$ is defined as

$$\mathbf{A}_n(\mathbf{k}) = i \langle u_{n\mathbf{k}} | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle.$$

The inner product $\langle u_{n\mathbf{k}} | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle$ measures how the Bloch state changes as the crystal momentum $\mathbf{k}$ varies, while the factor i corresponds to the phase change of the wavefunction as $\mathbf{k}$ changes. Therefore, the Berry connection describes the geometric phase acquired by the wavefunction under an infinitesimal displacement in crystal momentum.

The total accumulated Berry phase γ is then given by

$$\gamma = \int \mathbf{A}_n(\mathbf{k}) \cdot d\mathbf{k}.$$

The Berry curvature is defined as the curl of the Berry connection. Therefore

$$\Omega_n(\mathbf{k}) = \nabla_{\mathbf{k}} \times \mathbf{A}_n(\mathbf{k}).$$

For a two-dimensional system, $\mathbf{k}$ lies in the (k_x, k_y) plane, so the curl points entirely in the z -direction. The only persistent component is therefore

$$\Omega_{n,z}(\mathbf{k}) = i \left(\left\langle \frac{\partial u_{n\mathbf{k}}}{\partial k_x} \left| \frac{\partial u_{n\mathbf{k}}}{\partial k_y} \right\rangle - \left\langle \frac{\partial u_{n\mathbf{k}}}{\partial k_y} \left| \frac{\partial u_{n\mathbf{k}}}{\partial k_x} \right\rangle \right).$$

Because it arises as the curl of a vector potential, the Berry curvature can be interpreted as an effective magnetic field in momentum space.

In the semiclassical description of electron dynamics in crystals, the velocity of the Bloch electron in band n is determined by the band dispersion $\varepsilon_n(\mathbf{k})$ as [3, 8]

$$\mathbf{v}_n(\mathbf{k}) = \frac{1}{\hbar} \nabla_{\mathbf{k}} \varepsilon_n(\mathbf{k}).$$

However, once the Berry curvature is introduced, we obtain a new term regarded as the “anomalous velocity”:

$$\mathbf{v}_n(\mathbf{k}) = \frac{1}{\hbar} \nabla_{\mathbf{k}} \varepsilon_n(\mathbf{k}) - \frac{e}{\hbar} \mathbf{E} \times \Omega_n(\mathbf{k}).$$

This additional term gives rise to the *valley Hall effect*.

In monolayer MX_2 , K and K' valleys are correlated by time reversal symmetry. Therefore, under an applied electric field $\mathbf{E}$, carriers at the K and K' points acquire transverse velocities of equal magnitude but opposite direction [8].

The sideways deflection of the carriers is consistent with our interpretation of the Berry curvature, as seen from the cross product $\mathbf{E} \times \Omega_n(\mathbf{k})$. If an electric field is applied in the x -direction and the Berry curvature points out of the plane, then the cross product points in the y -direction, resulting in the sideways deflection. Refer to Fig. 3(b).

The valley Hall effect generates a transverse voltage, enabling electrical detection of valley polarization [3, 10], which is critical for the development of valleytronic devices.

Because valley polarization is imperative to valleytronic applications, a major focus of current research is the development of methods to achieve it. These methods include optical pumping, electric fields, mechanical strain, electrical injection, geometric engineering, and magnetic approaches [5, 8, 11–14].

In the following sections, we discuss magnetic mechanisms that can induce band splitting and lift valley degeneracy, thereby enabling valley polarization.

II.C. Orbital Magnetic Moment

Along with Berry curvature, K and K' valley states can be distinguished by the orbital magnetic moment [6].

As mentioned above, electrons in a crystal are described by Bloch states $\psi_{n\mathbf{k}}$. However, a single Bloch state may extend over the entire crystal, with a precise momentum $\mathbf{k}$. From the uncertainty principle $\Delta r \Delta k > 1$, we cannot localize an electron in both momentum and position [6]. Therefore, electron dynamics in crystals are described by the motion of *wave packets* [6, 15]:

$$\Psi(\mathbf{r}) = \sum_{\mathbf{k}} a(\mathbf{k}) \psi_{n\mathbf{k}}.$$

In other words, we have a superposition of nearby $\mathbf{k}$ -states.

The motion of a wave packet is characterized through the motion of its center of mass $\mathbf{r}_c$. Since the packet is not perfectly localized but instead has a finite spread, it also may exhibit a self-rotational motion around its center $\mathbf{r}_c$, shown in Fig. 4 [6, 7].

This self-rotation induces an orbital magnetic moment of the wave packet, $\mathbf{m}_{\text{orbital}}$ (along with the spin moment), exactly the same way a current loop in electromagnetism produces a magnetic dipole.

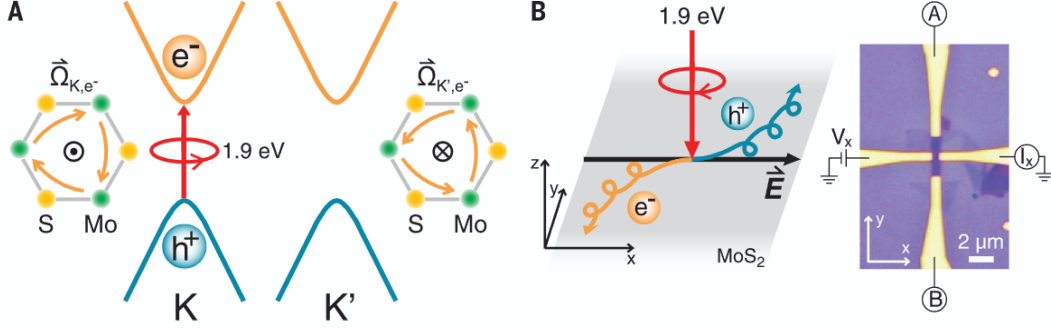


Figure 3. (a) Valley-dependent optical selection rules at K and K' valleys possessing opposite Berry curvatures. The orange arrows indicate clockwise and counterclockwise motion of the K and K' electrons. (b) An applied electric field $\mathbf{E}$ induces a transverse anomalous velocity, leading to opposite deflection of carriers from K and K' valleys. Reproduced from Ref. [9] with permission of K. F. Mak; peer-reviewed version in Ref. [3].

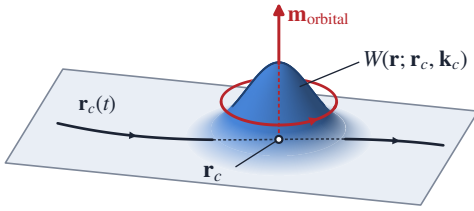


Figure 4. Depiction of a wave packet $W(\mathbf{r}; \mathbf{r}_c, \mathbf{k}_c)$ moving along the trajectory of its center of mass $\mathbf{r}_c$ while rotating about $\mathbf{r}_c$. The red loop shows the self-rotation, with its arrow giving the sense of the circulating current, so the orbital magnetic moment $\mathbf{m}_{\text{orbital}}$ points out of the monolayer plane. Redrawn after Ref. [7].

The total magnetic moment of the electron is then $\mathbf{m}_{\text{total}} = \mathbf{m}_{\text{spin}} + \mathbf{m}_{\text{orbital}}$. It should be noted that the orbital component arises entirely from the band structure, a novel property in monolayer MX_2 .

As mentioned briefly in the introduction, the orbital magnetic moment shows exactly the same symmetry transformation as the Berry curvature—that is to say, it exists only in $\mathcal{T}$ -broken or $\mathcal{P}$ -broken symmetry systems [6, 16].

Focusing henceforth on the orbital contribution, we find that in TMDs $\mathbf{m}_K = -\mathbf{m}_{K'}$, providing a natural means of distinguishing the K and K' valleys.

II.D. Valley Zeeman Effect

In general, the magnetic-field-induced energy difference between the K and K' valleys is known as *valley Zeeman splitting* [17]. We first consider the case of an external magnetic field.

II.D.1. External Magnetic Fields

For a particle with a magnetic dipole moment $\mathbf{m}$ in an external magnetic field $\mathbf{B}$, the potential energy is given by $E = -\mathbf{m} \cdot \mathbf{B}$.

In monolayer TMDs, the orbital motion of our electrons is confined in-plane. As a result, their orbital magnetic moment points out-of-plane and couples to the out-of-plane magnetic field, B_z [6].

Therefore, for an out-of-plane field, the energy shift

$$\Delta E = -m_z B_z.$$

In a valley system, the magnetic moment depends on the valley index:

$$m_z = \tau m_v$$

where m_v is the magnitude of the valley magnetic moment.

So the energy shift of a band state is

$$\Delta E_\tau = -\tau m_v B_z.$$

Using $\tau = \pm 1$ for the K or K' valleys, the corresponding energies become

$$\begin{aligned} E_K &= E_0 - m_v B_z \\ E_{K'} &= E_0 + m_v B_z. \end{aligned}$$

Because we are ultimately interested in the magnitude of the valley splitting, we find that the valley splitting energy

$$\begin{aligned} |\Delta E_{\text{valley}}| &= |E_K - E_{K'}| \\ &= |(E_0 - m_v B_z) - (E_0 + m_v B_z)| \\ &= 2m_v B_z. \end{aligned}$$

Defining an effective Landé g_{eff} -factor by

$$m_v = \frac{g_{\text{eff}} \mu_B}{2}$$

gives

$$|\Delta E_{\text{valley}}| = |g_{\text{eff}}| \mu_B B_z$$

where μ_B is the Bohr magneton. In monolayer MX_2 it has been shown theoretically and confirmed experimentally that $g \approx -4$ [17, 18].

Because the orbital magnetic moments in the K and K' valleys are opposite in sign, our external magnetic field causes opposite energy shifts in the bands of inequivalent valleys, and hence, lifts valley degeneracy [19].

This splitting enables selective population of valleys under magnetic field, offering a mechanism for valley polarization.

Note that $\mathcal{T}$ -symmetry requires $E_K = E_{K'}$. Because the applied magnetic field lifts valley degeneracy, we obtain $E_K \neq E_{K'}$, and therefore $\mathcal{T}$ -symmetry is broken.

Although this derivation was presented for an external magnetic field, the same Zeeman-like splitting framework also applies for the following magnetic mechanisms.

II.D.2. Magnetic Proximity Effects

In very recent years, the use of magnetic substrates has attracted significant interest from researchers aiming to lift valley degeneracy.

A magnetic substrate is simply a magnetic material placed beneath a TMD. In Sec. II.D.1, we discussed the use of an *external* magnetic field; conversely, a magnetic substrate makes use of proximity effects, effectively inducing an *exchange* magnetic field inside the monolayer. Similar to an applied external magnetic field, exchange fields constitute effective Zeeman-like splitting, lifting valley degeneracy in the TMD.

Magnetic proximity effects are commonly realized in van der Waals heterostructures, where a TMD layer is stacked on a magnetic material and held together by weak van der Waals forces [20]. An important characteristic of van der Waals materials is their ability to form vertical heterostructures with an ideal, atomically sharp interface [6].

The origin of this effect can be understood from the exchange interaction at the interface. In a magnetic material, the electrons have aligned spins due to exchange interactions, generating a net magnetization $\mathbf{M}$. When a monolayer TMD is placed directly on top, the electrons' wavefunctions marginally overlap across the interface. These interactions shift the energies of the valley states in a manner analogous to Zeeman splitting produced by an external magnetic field.

More precisely, the low-energy physics at the K and K' points of a TMD/magnetic substrate heterostructure is captured by the effective Hamiltonian [21]

$$H = H_0 + H_{\text{SOC}} + H_{\text{ex}} + H_R.$$

The first term

$$H_0 = at(\tau k_x \sigma_x + k_y \sigma_y) + \frac{\Delta}{2} \sigma_z$$

is the massive Dirac fermion Hamiltonian that produces the gapped band structure at each valley, where a is the lattice constant, t is the nearest-neighbor hopping energy, $\tau = \pm 1$ is the valley index, σ_i are Pauli matrices acting on the orbital pseudospin, and Δ is the band gap.

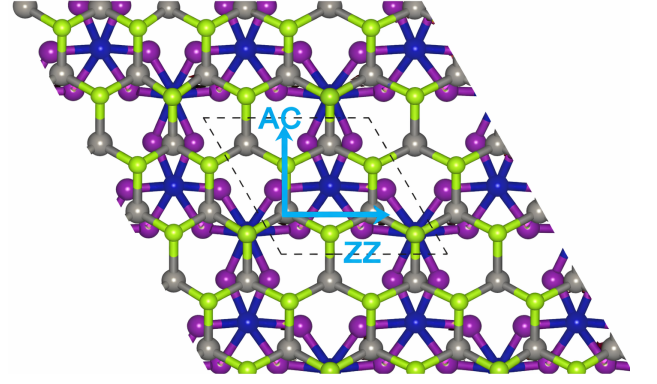


Figure 5. Structure model of twisted WSe₂/CrI₃ heterostructure [24]. Reproduced from Ref. [24] under a CC BY 4.0 license.

The second term,

$$H_{\text{SOC}} = \frac{\tau \lambda_{\text{SOC}}}{2} s_z (\sigma_z - 1)$$

describes the spin-orbit coupling, which produces opposite spin splitting in opposite valleys and establishes spin-valley locking; however, because H_{SOC} preserves $\mathcal{T}$ -symmetry, it cannot by itself lift valley degeneracy.

The third term, H_{ex} , is the proximity-induced exchange interaction between the localized spins of the magnetic substrate and the carriers in the monolayer. Developed from the Kondo model [22], this interaction takes the form

$$H_{\text{ex}} = -\mathbf{J} \mathbf{M} \cdot \mathbf{s}$$

where $\mathbf{M}$ is the magnetization vector of the substrate, $\mathbf{s}$ is the electron spin, and J is the exchange coupling constant. This term breaks $\mathcal{T}$ -symmetry by shifting spin-up and spin-down states uniformly regardless of the valley index. Because spin-valley locking pins opposite spins to opposite valleys, this uniform spin shift translates into a valley-dependent energy shift, thereby lifting valley degeneracy.

The final term, H_R , is the Rashba contribution arising from the broken inversion symmetry at the heterostructure interface; it is typically small and acts as a correction.

Crucially, valley polarization requires both terms acting together: H_{SOC} locks spin to valley, while H_{ex} breaks $\mathcal{T}$ -symmetry, so the resulting valley splitting grows with the strength of the exchange field.

The use of magnetic substrates carries an important advantage, in that the magnitude of valley splitting can be tuned by controlling the magnetization of the substrate [23]. One of the more experimentally popular heterostructures is monolayer WSe₂ coupled with CrI₃, shown in Fig. 5.

II.D.3. Magnetic Doping

Magnetic doping can be broken into two subcategories: substitutional doping and adsorption. Substitutional dop-

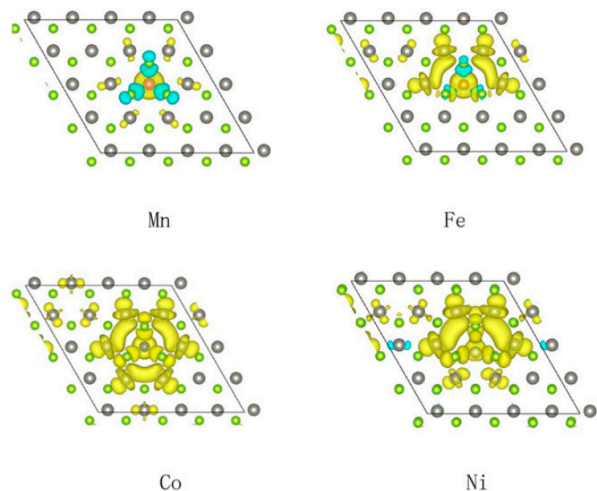


Figure 6. Spin density distribution for a single transition metal dopant atom (Mn, Fe, Co, Ni) substituted in monolayer WSe₂, depicting localized magnetic moments introduced by dopants [25]. Reproduced with permission of the authors.

ing inserts magnetic impurities directly into the crystal lattice. For example,



Generally, the dopant atoms contain unpaired *d*-electrons, giving them a localized magnetic moment. At sufficient concentration, they can produce a net magnetization $\mathbf{M}$ inside the material, breaking $\mathcal{T}$ -symmetry.

Adsorption, by contrast, places the dopant atom *on top of* the surface rather than inside the lattice. The atom sits at a preferred surface site (often above a hollow site or directly above a host atom) and bonds to the TMD through weaker surface interactions.

In both cases, the exchange interaction discussed in the previous section applies, where the localized spin of the magnetic dopant interacts with the carriers in the monolayer. However, because a dopant does not introduce the global vertical asymmetry of a heterostructure interface, the Rashba term is absent and the effective Hamiltonian reduces to

$$H = H_0 + H_{\text{SOC}} + H_{\text{ex}}.$$

As before, valley polarization arises from the exchange term acting together with spin-valley locking. Because the magnetic dopant creates a preferred spin direction, H_{ex} shifts the valley energies differently through spin-valley locking, thereby lifting valley degeneracy. An example of substitutional doping is shown in Fig. 6.

II.D.4. Intrinsic Ferrovalley Materials

Intrinsic ferrovalley materials are two-dimensional ferromagnetic semiconductors whose spontaneous magnetization lifts valley degeneracy without the need for external fields or substrates. In the presence of magnetization, an

exchange interaction shifts the spin-resolved bands. Because the spin and valley degrees of freedom are strongly coupled in monolayer TMDs, this exchange field produces opposite energy shifts at the K and K' valleys. Therefore, this ultimately boils down to Zeeman-like splitting, $\Delta E = g_{\text{eff}} \mu_B B_{\text{eff}}$ (B_{eff} is the induced effective magnetic field), lifting valley degeneracy.

Ferrovalley behavior requires a specific crystal symmetry. Monolayer TMDs can crystallize in two principal phases: the H-phase (trigonal prismatic coordination), in which the metal atom sits at the center of a trigonal prism formed by the chalcogen atoms, and the T-phase (octahedral coordination), in which the chalcogen atoms form an octahedron around the metal [1]. The H-phase possesses the same broken inversion symmetry and hexagonal structure as the conventional MX₂ compounds discussed throughout this work, and therefore supports inequivalent K and K' valleys with spin-valley locking. The T-phase, by contrast, has higher point-group symmetry that generally does not produce the same valley contrast. For this reason, ferrovalley materials are predominantly found in the H-phase. Common examples of intrinsic H-phase ferrovalley TMDs include the vanadium dichalcogenides VSe₂, VS₂, and VTe₂, which we examine in detail in Sec. III.

As in the previous magnetic mechanisms, the effective Hamiltonian at the valley points is $H = H_0 + H_{\text{SOC}} + H_{\text{ex}}$, with the Rashba term again absent since there is no heterostructure interface. The crucial distinction is that H_{ex} now arises from the material's own spontaneous magnetization rather than from an external field, a magnetic substrate, or a dopant. Because the exchange interaction is intrinsic, valley degeneracy is lifted as an equilibrium property of the ground state, requiring no external intervention.

III. COMPUTATIONAL STUDIES AND EXPERIMENTAL RESULTS IN MAGNETIC MECHANISMS

While the previous section discusses theoretical mechanisms of lifting valley degeneracy, experimental investigations are necessary to confirm these predictions and quantify resulting valley polarization in a number of monolayer TMDs. Recently, a variety of experiments have explored magnetic routes to valley polarization including the application of an external magnetic field, the use of magnetic proximity effects, the realization of magnetic doping, and the utilization of intrinsic ferrovalley materials. In this section, we review key studies that demonstrate valley splitting and polarization under the techniques introduced in Sec. II. However, working with monolayer TMDs presents several experimental challenges—among them the difficulty of preparing high-quality samples, achieving atomically clean interfaces, and operating at cryogenic temperatures. When direct experimental investigation is prohibitive, computational studies, most often based on

density functional theory (DFT), become invaluable for advancing our understanding of the magnetic mechanisms described above. As shown below, computational results largely agree with experimental findings when both are available, demonstrating the reliability of DFT in predicting valley polarization.

In a majority of studies, valley polarization is measured using magneto-photoluminescence spectroscopy, where a monolayer TMD or heterostructure is optically excited by a laser. The emitted photoluminescence is then analyzed in a circular polarization basis where σ^+ (σ^-) selectively excites electron-hole pairs in the K (K') valleys. The spectra are then recorded to measure the degree of valley polarization.

III.A. External Magnetic Field

The most straightforward route to lifting valley degeneracy is the application of an out-of-plane magnetic field, which directly breaks time-reversal symmetry and induces Zeeman-like splitting between the K and K' valleys. We first examine results from monolayer TMDs before turning to van der Waals heterostructures, which exhibit substantially larger splittings.

MacNeill *et al.* directly experimented with a monolayer TMD, MoSe₂ [17]. They report polarization-resolved luminescence spectra for back-gated MoSe₂ at 4.2 K in magnetic fields up to 6.7 T.

Fig. 7(a) illustrates the opposite optical and magnetic responses of the K and K' valleys. At $B = 0$ T, the two circular polarizations (σ^+ and σ^-) exhibit nearly identical energy and intensity. At $B = \pm 6.7$ T, however, the degeneracy is lifted, and the relative positions of the σ^+ and σ^- spectra reverse with the sign of the magnetic field.

Fig. 7(b) then shows a linear dependence of the valley splitting energy on the applied magnetic field, confirming the valley Zeeman relationship covered in Sec. II.D:

$$\Delta E = g\mu_B B_z.$$

The fitted slope of -0.22 ± 0.01 meV/T [17] yields the effective Landé g -factor upon division by the Bohr magneton. For monolayer MoSe₂, the effective Landé g -factor was found to be $g_{\text{eff}} = -3.80 \pm 0.17 \approx -4$, as discussed in Sec. II.D.1.

Additional findings on related TMDs report similar relationships between the applied magnetic field and the lifting of valley degeneracy. Srivastava *et al.* studied monolayer WSe₂ at 4.2 K under magnetic fields up to ± 8.4 T using polarization-resolved photoluminescence. After the addition of a magnetic field to WSe₂, an exciton splitting of 0.21 meV/T was exhibited, corresponding to an effective g -factor of $g_{\text{eff}} \approx -3.63$ —a similar result to the experiment conducted by MacNeill *et al.* [18]. Using DFT, Liu *et al.* computed a comparable valley splitting of 0.21 meV/T for fields up to ± 10 T, in agreement with

these measurements [13]. This is shown by their first-principles DFT calculations in Fig. 8.

Numerous experiments have demonstrated the ability of monolayer TMDs to lift valley degeneracy under the application of an external magnetic field. However, one limitation of the applied-field scheme is the resulting relatively low valley splitting, ranging from 0.11–0.25 meV/T in MX₂ systems such as MoSe₂ and WSe₂ [1, 13, 17–19]. Additionally, the magnitude of the valley splitting reaches only 1–5 meV as a whole. As such, it has become evident that the application of an external magnetic field to bare monolayer TMDs is neither practical nor broadly applicable for the development of valleytronic devices.

However, the utilization of van der Waals heterostructures composed of monolayer TMDs under an applied magnetic field has been much more effective in lifting valley degeneracy. Nagler *et al.* investigated a nearly 60° stacked WSe₂/MoSe₂ heterostructure, exfoliated onto a SiO₂/Si substrate, at 4 K [27]. An out-of-plane magnetic field ranging from 0 to 30 T was applied while polarization-resolved spectra were recorded. The valley splitting exhibited an approximately linear dependence on the magnetic field, establishing Zeeman-like splitting in the heterostructure. More specifically, the splitting reached 26 meV (much higher than the 1–5 meV splitting exhibited by a monolayer MX₂) at $B = 30$ T, indicating a strong magnetic response of about 0.87 meV/T. The graph of Fig. 9(b) suggests an effective g -factor of -15.1 ± 0.1 , much higher than reported for monolayer MoSe₂ or WSe₂.

Thus, the WSe₂/MoSe₂ heterostructure exhibits a substantially larger valley splitting, and hence a much greater effective Landé g -factor, than those typically reported for monolayer TMDs. Heterostructures provide a stronger mechanism than monolayer TMDs for lifting valley degeneracy, enabling greater promise for valleytronic device applications.

In this sense, while external magnetic fields provide a direct and highly tunable route to valley polarization, the magnitude of the resulting splitting depends strongly on the material system under study. Bare monolayer TMDs proved the least promising, yielding only 1–5 meV of splitting; the TMD-based heterostructures discussed above performed markedly better, yet the large fields and cryogenic conditions they still require motivate the alternative magnetic mechanisms explored below.

III.B. Magnetic Proximity Effects

Experimentally, magnetic proximity effects are commonly realized by vertically stacking a monolayer TMD on a 2D magnetic substrate to form a van der Waals heterostructure. Several theoretical studies using first-principles DFT have predicted that magnetic substrates can induce large valley splitting in monolayer TMDs through interfacial exchange coupling [19, 24, 27, 29]. The limited feasibility of such experiments largely stems

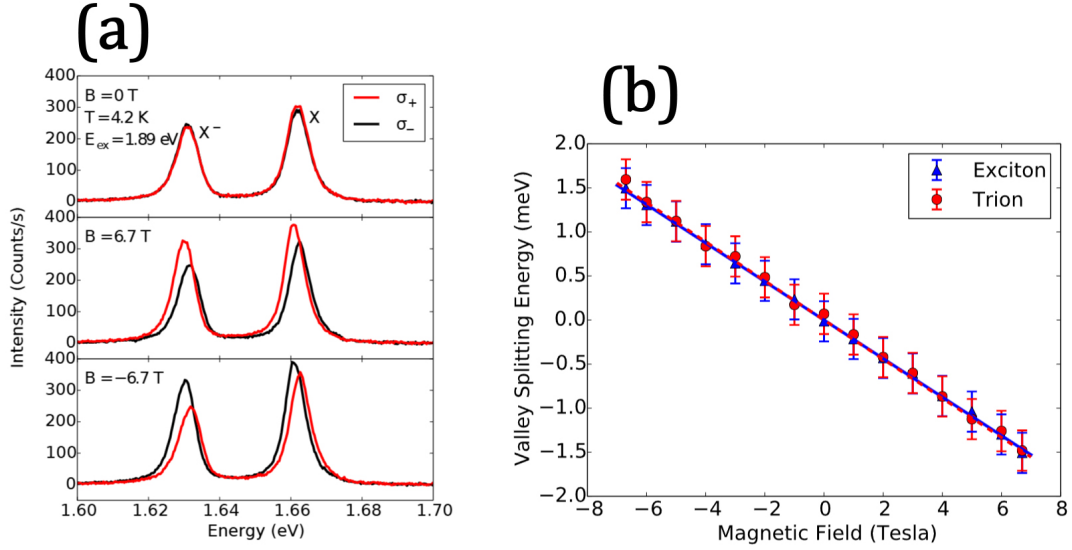


Figure 7. (a) Polarization-resolved luminescence spectra from monolayer MoSe₂ at 4.2 K for σ^+ and σ^- detection. Spectra taken (top-to-bottom) from out-of-plane magnetic fields of 0 T, 6.7 T, and -6.7 T. Polarization and splitting change with the reversed sign of the magnetic field. (b) Linear splitting of -0.22 ± 0.01 meV/T in both excitons (blue triangles) and trion (red circles). Reproduced from Ref. [26] with permission of the authors; peer-reviewed version in Ref. [17].

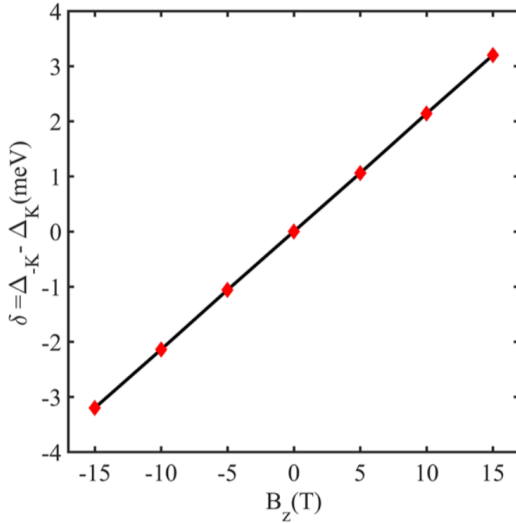


Figure 8. DFT calculations, demonstrating a valley splitting of 0.21 meV/T in monolayer WSe₂ [13]. Reproduced from Ref. [13] under a CC BY 4.0 license.

from the difficulty of achieving atomically flat interfaces, integrating two-dimensional magnetic semiconductor insulators, maintaining extremely low disorder, and operating at cryogenic temperatures. Many experiments involving the use of magnetic proximity effects are often accompanied by an external magnetic field to bolster the magnitude of valley splitting.

Qi *et al.* investigated magnetic proximity effects in a computationally modeled MoTe₂/EuO heterostructure using first-principles DFT and a low-energy effective Hamiltonian [21]. Fig. 10 depicts the schematic.

The EuO substrate was chosen because it is a ferromag-

netic semiconductor with a band gap of 1 eV, an exchange moment of approximately $7\mu_B$ per Eu ion, and a lattice mismatch of only 2.7% with monolayer MoTe₂.

The group utilized the low-energy Hamiltonian

$$H = H_0 + H_{\text{SOC}} + H_{\text{ex}} + H_R$$

where the proximity-induced exchange term H_{ex} ,

$$H_{\text{ex}} = -s_z(B_c\sigma_+ + B_v\sigma_-),$$

lifts valley degeneracy together with the Rashba term, producing valley-dependent band shifts. Here, s_z is a Pauli matrix eigenvalue (± 1), $\sigma_{\pm} = \frac{1}{2}(\sigma_0 \pm \sigma_z)$ project onto the conduction and valence orbitals, and B_c and B_v are “effective Zeeman fields” in the conduction and valence bands, respectively.

In their model, the effective Zeeman fields were gargantuan: using $B_c = 206$ meV and $B_v = 170$ meV, they find a translated magnetic field of over 2937 T. This is far larger than the bare dipolar field of only ~ 0.007 T from the Eu moments at the MoTe₂ layer, demonstrating that the valley splitting arises from exchange fields.

Their calculations predict enormous valley splitting ranging from 321 to 419 meV, far greater than the 1–5 meV splitting typically achieved with external fields.

Zhao *et al.* observed enhanced valley splitting in WSe₂ in van der Waals WSe₂/EuS heterostructures [30]. Experimentally, it was found that the valley splitting energy increased by nearly an order of magnitude, from 0.21 meV/T to 2.5 meV/T, with the addition of the magnetic substrate EuS.

In 2017, Zhong *et al.* similarly investigated valley splitting in WSe₂ [23]. However, they employed a CrI₃ magnetic substrate, which enabled unprecedented control of

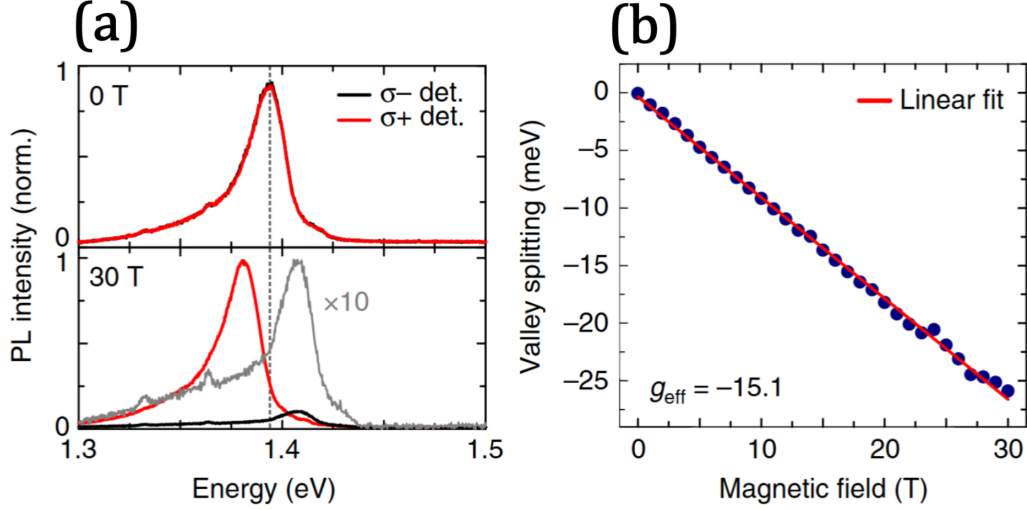


Figure 9. (a) Comparison of photoluminescence spectra of the interlayer for 0 T and 30 T. At $B = 0$ T, both circular polarizations show the same energy and intensity. At $B = 30$ T, valley degeneracy is lifted at the σ^+ transition. (b) Corresponding valley-selective splitting of the interlayer exciton. Red line represents a linear fit in the strength of the magnetic field and the valley splitting energy, with $g_{\text{eff}} = -15.1 \pm 0.1$. Reproduced from Ref. [28] with permission of the authors; peer-reviewed version in Ref. [27].

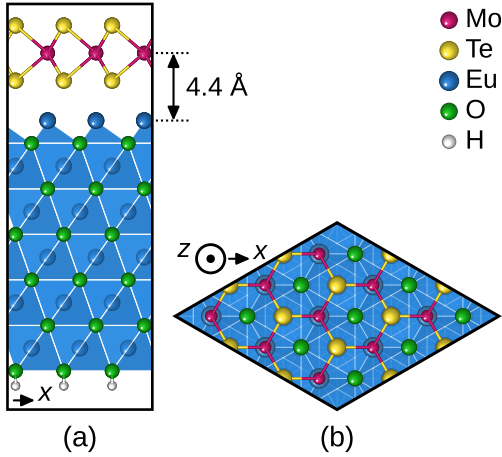


Figure 10. (a) Side view and (b) top view of a MoTe₂ monolayer on an EuO(111) substrate, drawn from a structural model based on Ref. [21]. The Mo plane sits 4.4 Å above the top Eu plane.

spin and valley pseudospin in monolayer WSe₂. Their study is particularly compelling because it reports a direct experimental demonstration rather than a first-principles DFT calculation.

Ultrathin CrI₃ with a thickness of 9–12 nm is an especially appealing substrate, as it is a newly discovered 2D ferromagnetic semiconductor [32]. It also exhibits an out-of-plane easy axis—the energetically preferred direction of its magnetization—which is imperative to its ability to induce a valley-splitting exchange field, since the valley Zeeman coupling responds to the out-of-plane component of the magnetization. Additionally, an h-BN encapsulation was implemented to prevent degradation. The stack was assembled as h-BN / CrI₃ / monolayer

Valley splitting (meV)	$\Delta_{\text{val}}^{v,+}$	$\Delta_{\text{val}}^{c,+}$	$\Delta_{\text{val}}^{v,-}$	$\Delta_{\text{val}}^{c,-}$
MoTe ₂ [†] (DFT & LEH)	0	0	0	0
MoTe ₂ /EuO (DFT)	-342	-386	-321	-419
MoTe ₂ /EuO (LEH)	-319	-412	-340	-391
Spin splitting (meV)	$\Delta_{\text{spin}}^{v,+}$	$\Delta_{\text{spin}}^{c,+}$	$\Delta_{\text{spin}}^{v,-}$	$\Delta_{\text{spin}}^{c,-}$
MoTe ₂ [†] (DFT & LEH)	214	-27	-214	27
MoTe ₂ /EuO (DFT)	-168	-449	-496	-356
MoTe ₂ /EuO (LEH)	-142	-455	-517	-348

[†] For free-standing MoTe₂, DFT and LEH give the same results.

Table I. Valley splitting and spin splitting for free-standing monolayer MoTe₂ and the MoTe₂/EuO heterostructure, comparing density functional theory (DFT) and low-energy effective Hamiltonian (LEH) results. Magnetic proximity to EuO induces large valley and spin splittings in monolayer MoTe₂. Data from Ref. [21].

WSe₂ / h-BN / SiO₂. A schematic of the experiment is shown in Fig. 11.

At 5 K, the valley splitting energy was measured to be 3.5 meV, which the authors converted to an effective magnetic field of approximately 13 T. The authors further noted that the magnitude is independent of the excitation power, helping rule out carrier-density effects as the main cause of the lifted valley degeneracy.

Near an applied external magnetic field of -0.85 T, the valley splitting changes from -1.3 meV to 3.4 meV over only about 30 mT, corresponding to a valley splitting rate of more than 150 meV/T. The authors note that this is nearly three orders of magnitude larger than can be achieved by the Zeeman effect in bare WSe₂, thereby highlighting the practicality and enhanced tunability of

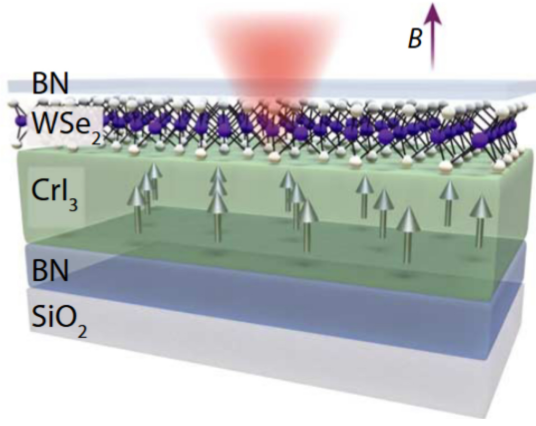


Figure 11. Schematic of the van der Waals heterostructure formed by monolayer WSe₂ and ultrathin ferromagnetic semiconductor CrI₃, encapsulated by h-BN. Reproduced from Ref. [31] with permission of the authors; peer-reviewed version in Ref. [23].

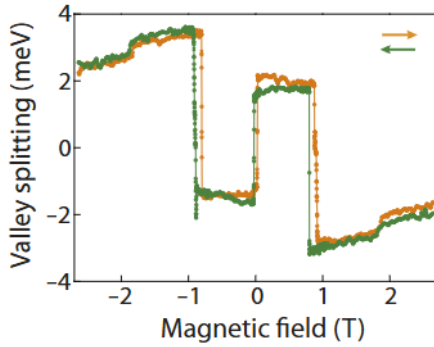


Figure 12. Valley splitting in WSe₂ versus applied magnetic field, showing hysteretic step-like switching due to reversal of the CrI₃ substrate magnetization; the large splitting is dominated by the proximity-induced exchange field rather than the external field alone. Reproduced from Ref. [31] with permission of the authors; peer-reviewed version in Ref. [23].

magnetic proximity effects, even under a comparatively small applied magnetic field.

Fig. 12 shows the valley splitting in WSe₂ as a function of the out-of-plane magnetic field. Rather than varying linearly with field, the splitting exhibits abrupt, hysteretic jumps, indicating that it is controlled primarily by the magnetization state of the CrI₃ substrate. The external field mainly acts to switch the CrI₃ magnetization, whereas the large measured splitting arises from the proximity-induced exchange field in WSe₂. The step-like behavior therefore provides strong evidence that the lifted valley degeneracy is dominated by magnetic proximity coupling rather than by the direct Zeeman effect of the applied field alone.

To further verify that the valley splitting arises from the exchange field, the authors measured the photoluminescence spectra of the valleys above the Curie temperature of CrI₃, at 65 K. They found an equal distribution of right-circularly polarized and left-circularly polarized

light, further demonstrating the central role of the CrI₃ substrate in lifting the valley degeneracy of WSe₂. This comparison is shown in Fig. 13(a) and (b).

The experiment of Zhong *et al.* ultimately demonstrates the practicality of magnetic heterostructures for lifting valley degeneracy in prospective valleytronic devices.

Finally, we turn to a recent theoretical study of magnetic proximity effects. In 2025, Zhang *et al.* investigated CrGeTe₃/MTe₂ (M = Mo, W) van der Waals heterostructures using first-principles DFT and a low-energy $k \cdot p$ model, showing that magnetic proximity to monolayer CrGeTe₃ lifts the K/K' valley degeneracy in the TMD layer [29].

The respective H1, H2, and H3 heterostructures are shown in Fig. 14(a), (b), and (c).

At equilibrium, they found that the strongest valley splitting arises from the H1 stacking, with a splitting of 5.90 meV in the Mo-based heterostructure and 19.74 meV in the W-based heterostructure. Naturally, the W-based system exhibits the larger valley splitting because of its stronger SOC. Their findings demonstrate that the stacking registry matters in great detail; it is not a minor structural feature, but instead directly determines the scale of the valley splitting.

The tunability results are arguably the most valuable part of the paper. Rotating the substrate magnetization causes the splitting to follow an approximate cosine law:

$$\Delta E_s(\theta) = \Delta E_s(0) \cos(\theta).$$

This relationship is consistent with our current understanding, as the valley splitting goes to zero for an in-plane magnetic field at 90°.

Additionally, they introduced a normal strain that reduces the interlayer spacing by 0.5 Å, which increases the splitting to 21.97 meV in the Mo-based heterostructure and 38.09 meV in the W-based heterostructure.

While these results exceed the ~ 1 –5 meV values shown in monolayer WSe₂, they do not surpass the valley splitting of 321 to 419 meV shown by Qi *et al.* [21] in the MoTe₂/EuO heterostructure discussed above.

From the collection of studies utilizing magnetic proximity effects, we observe a variety of methods and valley splitting energies. However, as both theoretical calculations and experiments continue to become more prevalent, it is apparent that researchers are focused not only on the magnitude of the valley splitting achieved, but also on the precise stacking structures through which the same substrate and monolayer may be van der Waals coupled. Furthermore, tunability is of even greater value in the context of valleytronic applications, and the 2025 study of Zhang *et al.* demonstrates the importance and practical promise of such tunability. This therefore represents one further step toward the improvement of valleytronic applications through magnetic proximity effects.

Overall, magnetic substrates have proven to be one of the strongest options for valleytronic devices. The combination of practicality, as no external field is required,

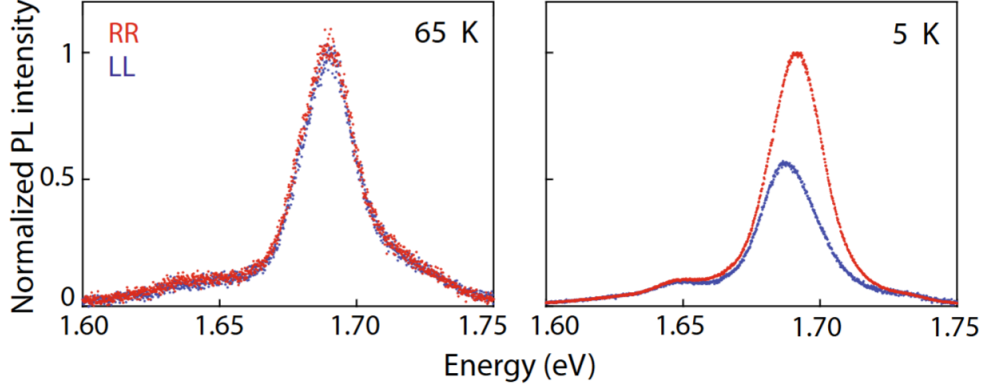


Figure 13. Circularly polarized photoluminescence spectra above T_c of CrI_3 (65 K) and below T_c (5 K) in the absence of an external magnetic field. Reproduced from Ref. [31] with permission of the authors; peer-reviewed version in Ref. [23].

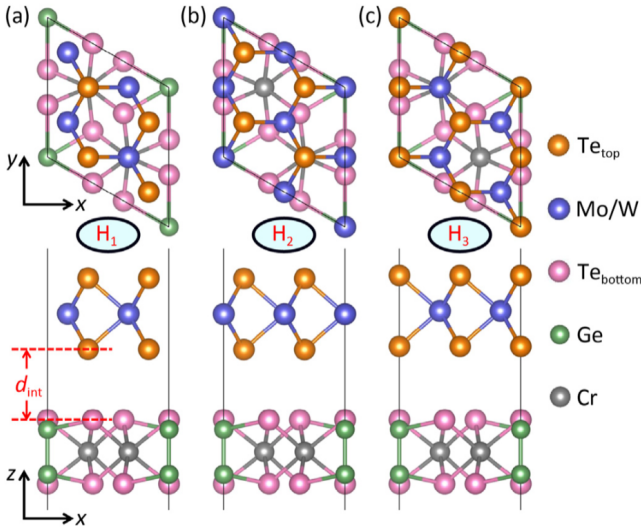


Figure 14. Top and side views of 2D $\text{CrGeTe}_3/\text{MTe}_2$ ($M = \text{Mo}, \text{W}$) with three different stacking patterns: (a) H1 model, (b) H2 model, and (c) H3 model [29]. Reproduced with permission of the authors.

and usability, as such systems can be realistically implemented, makes them especially promising in comparison with external-field approaches.

III.C. Magnetic Doping

Magnetic dopants can be incorporated into TMD monolayers through substitutional replacement of host atoms (substitutional doping) or surface adsorption during growth. Substitutional doping typically produces stronger exchange interactions but can introduce lattice distortion and defect states, whereas adsorbed magnetic atoms preserve the lattice at the expense of weaker coupling. The resulting magnetic response depends sensitively on dopant concentration: isolated dopants act as localized moments, while higher densities may lead to collective magnetic ordering. In addition to inducing magnetism, doping modifies the electronic structure by introducing impurity

states, shifting the Fermi level, and altering carrier mobility and optical properties. However, the presence of dopants inevitably introduces disorder and carrier scattering, which can broaden spectral features and degrade device performance.

Theoretical studies have shown that the adsorption of transition metals such as Sc, Fe, Mn, and Cu on MoS_2 produces a valley splitting of 16–41 meV [1, 33].

Recently, Sahoo *et al.* investigated enhanced room-temperature spin-valley coupling in V-doped (vanadium-doped) MoS_2 . In their study, they combined experimental measurements with DFT calculations in order to compare the observed valley response with its theoretical origin [34].

To probe the valley-selective optical response, the authors used chirality-resolved photoluminescence under opposite circular polarizations of light (σ^+ and σ^-). They also carried out weak-value-amplification-based spin Hall effect of light (SHEL) measurements as an additional optical probe, while first-principles DFT calculations were used for theoretical comparison.

A computer-generated schematic of the V-doped MoS_2 system is shown in Fig. 15.

Sahoo *et al.* calculate the polarization degree, also known as the valley polarization, as

$$\rho = \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)}, \quad (2)$$

where $I(\sigma^\pm)$ is the emitted photoluminescence intensity in each circular polarization. This is the optical analogue of the valley polarization defined in Eq. (1): whereas Eq. (1) counts the carrier populations directly, this expression captures the same valley imbalance through the intensities of the emitted light. Using this relation, they obtain about 12% for pristine MoS_2 and 42% for V-doped MoS_2 , demonstrating the strong role of the dopant in lifting valley degeneracy. More importantly, for the V-doped MoS_2 , the photoluminescence spectra they report show a splitting of about 35 meV between opposite chiralities.

The SHEL measurements further support this result. Because SHEL detects very small polarization-dependent

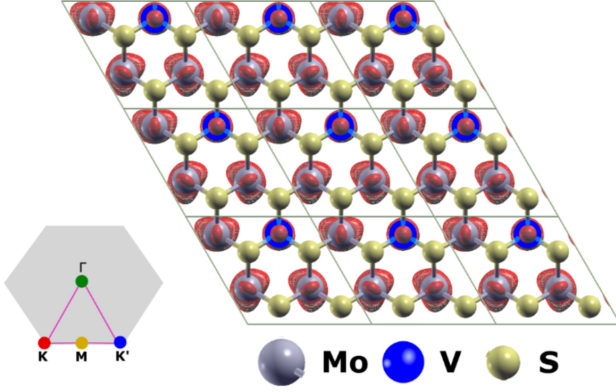


Figure 15. Atomic model of the V-doped MoS₂ system with local density of states [34]. Reprinted figure with permission from K. R. Sahoo, J. J. Panda, S. Bawari, R. Sharma, D. Maity, A. Lal, R. Arenal, G. Rajalaksmi, and T. N. Narayanan, Phys. Rev. Materials **6**, 085202 (2022), doi:10.1103/PhysRevMaterials.6.085202. Copyright (2022) by the American Physical Society. Also reproduced with permission of the authors.

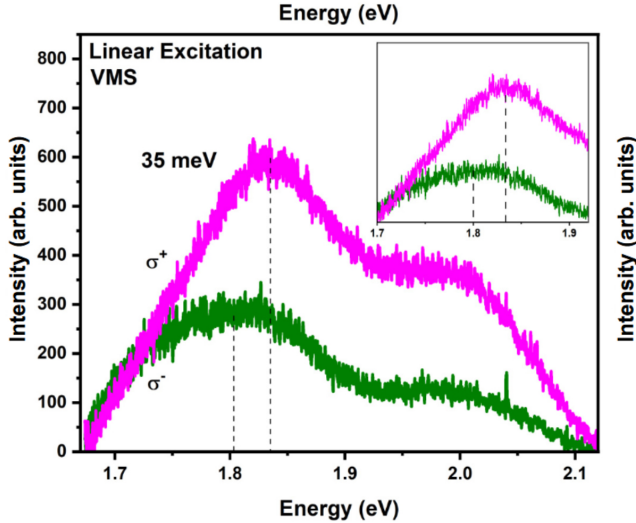


Figure 16. Photoluminescence spectra under excitation, extracting the two circular polarizations of light (right σ^+ and left σ^- circular) for V-doped MoS₂ at room temperature, illustrating a valley splitting energy of ~ 35 meV [34]. Reprinted figure with permission from K. R. Sahoo, J. J. Panda, S. Bawari, R. Sharma, D. Maity, A. Lal, R. Arenal, G. Rajalaksmi, and T. N. Narayanan, Phys. Rev. Materials **6**, 085202 (2022), doi:10.1103/PhysRevMaterials.6.085202. Copyright (2022) by the American Physical Society. Also reproduced with permission of the authors.

shifts of reflected light, it allows the authors to show that V-doped MoS₂ absorbs σ^+ and σ^- differently, providing additional evidence for room-temperature valley polarization.

The DFT calculations are consistent with this overall picture. For V-doped MoS₂, they recover a valley splitting energy of about 120 meV between the K and K' points at the valence band. This value is noticeably larger than the

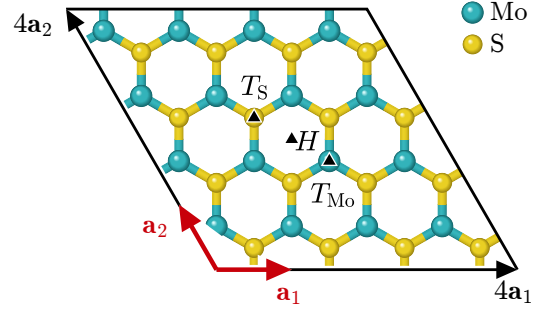


Figure 17. A 4×4 supercell of monolayer MoS₂ with the adsorption sites T_S , H , and T_{Mo} , drawn from a structural model based on Ref. [33].

experimentally measured splitting of 35 meV, which the authors attribute to the fact that temperature and lattice quality were not incorporated into the calculations.

Because the experiment was conducted at room temperature, these results open the possibility of developing atomically ultrathin optoelectronic spintronic as well as valleytronic devices [1, 34].

While Sahoo *et al.* provide a promising route to valley polarization through substitutional doping, we now turn to adsorption-based approaches, which may offer greater tunability and control.

In 2016, Chen *et al.* theoretically explored the variability of valley splitting in MoS₂ across many different transition metals such as Sc, Ti, V, Cr, Mn, Fe, Co, Ni, and Cu [33].

In their paper, adsorption means that a metal atom binds on the TMD surface, specifically at a pre-existing chalcogen vacancy, rather than replacing a lattice atom; this is surface-bound defect formation, not substitutional doping. This preference arises because the vacancy creates an under-coordinated local environment with altered charge density, allowing strong hybridization between the adatom orbitals and the surrounding TMD atoms.

Strong hybridization, that is, the mixing of the electronic orbitals of the adatom and MoS₂, can lead to indistinguishable valleys. Adsorption energy tells us how energetically favorable it is for the atom to stick to the surface at a given site; in this case, it tells us how strongly the adatom binds to MoS₂. The evidence suggests that a larger adsorption energy corresponds to stronger hybridization, which can prevent the lifting of valley degeneracy.

From the adsorption-site analysis, the authors find that the most stable site for all $3d$ adatoms they study is the top of the Mo atom, labeled T_{Mo} . This is shown in Fig. 17 [33]. Additionally, it is determined that for Sc, Mn, Fe, and Cu, the valley character remains identifiable and valley degeneracy is lifted. By contrast, for Ti, V, Cr, and Co, the impurity bands hybridize too strongly with the MoS₂ highest valence bands, so the valleys become indistinguishable.

Computational results for the adatoms whose structures

Table II. Adsorption energy, magnetic moment, and valley splitting for selected dopants. Data from Ref. [33].

Dopant	E_{ads} (eV)	M_{μ} (μ_B)	$\Delta_{KK'}$ (meV)
Sc	0.90	3.0	41
Mn	0.41	5.0	-16
Fe	0.91	4.0	-28
Cu	1.12	1.0	21

lift valley degeneracy are shown in Table II. Here the valley splitting $\Delta_{KK'}$ is the energy difference between the highest valence band at the K and K' points.

Surprisingly, the findings demonstrate that the adsorption energy does not track valley splitting directly. Stronger binding does not simply mean larger valley splitting. The binding stabilizes the adatom, but the splitting depends on the resulting exchange coupling and hybridization.

Table II also reports the total magnetic moments of the selected adatoms. Similarly, we find that a large moment alone is not enough to significantly lift valley degeneracy. Therefore, magnetism is needed without excessive band mixing [33]. Adsorption-based valley polarization requires not only local magnetism, but also sufficiently weak band hybridization so that the intrinsic valley structure of MoS₂ remains intact.

The authors also compute the tunability of valley splitting $\Delta_{KK'}$ for Mn adsorbed on MoS₂. They do this by applying an external magnetic field that rotates the magnetic moment of Mn, thereby changing the valley splitting. As the direction of magnetization rotates from the $+x$ direction (in-plane) to the $+z$ direction (perpendicular to the monolayer plane), they find an approximate sine relation between the angle and the magnitude of valley splitting, as demonstrated by Fig. 18 [33]. From the graph, the valley splitting energy varies roughly from about -16 meV to $+15$ meV, crossing through zero as the magnetization direction changes. As they describe, the valley splitting can thus be controlled by an external magnetic field, a readily accessible experimental knob.

Magnetic doping can be realized through two approaches: substitutional doping and adsorption. As shown, substitutional doping generally provides larger valley-splitting energies, whereas adsorption offers greater tunability. For now, however, substitutional doping may remain the more promising route because of its demonstrated experimental results, including operation at room temperature, which suggests stronger practicality for valleytronic devices.

III.D. Intrinsic Ferrovalley Materials

Typically studied monolayer TMDs, MX₂ ($M = \text{Mo, W}$; $X = \text{S, Se, Te}$), are intrinsically nonmagnetic and therefore cannot exhibit spontaneous valley polarization without external magnetization. In contrast, intrinsic ferrovalley materials possess valley polarization as an equilibrium

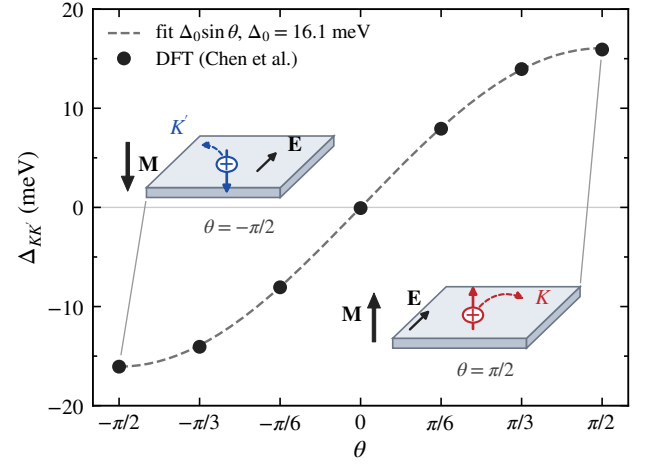


Figure 18. Valley splitting $\Delta_{KK'}$ for Mn adsorbed on a 4×4 MoS₂ supercell as a function of the magnetization direction θ , the angle between the magnetic quantization axis and the monolayer plane. Points are DFT values digitized from Ref. [33]; the dashed line is a fit $\Delta_0 \sin \theta$ with $\Delta_0 \approx 16$ meV. Insets sketch the anomalous valley Hall effect at $\theta = \pm\pi/2$: the circled + denotes a hole, $\mathbf{M}$ is the magnetization, and $\mathbf{E}$ is the applied in-plane electric field. For $\theta = \pi/2$, spin-up holes from the K valley dominate; for $\theta = -\pi/2$, spin-down holes from the K' valley dominate, and they deflect in opposite directions.

property of the crystal itself. Since they do not rely on an additional mechanism to induce valley polarization, they are regarded as especially promising for valleytronic applications because of their greater practicality in device implementation. We now look beyond the most commonly studied TMDs to a broader family of compounds that gives rise to many ferrovalley materials.

In Sec. II.B, we introduced the valley Hall effect and discussed how the transverse voltage it generates can be useful for valleytronic applications. In ferrovalley materials, this response acquires an additional charge Hall contribution arising from spontaneous valley polarization, which is referred to as the anomalous valley Hall effect.

Among the proposed intrinsic ferrovalley TMDs, monolayer VSe₂ has received particular attention in recent years. Monolayer VSe₂ exists in two main phases, the T- and H-phases [1]. In 2016, Tong *et al.* identified monolayer 2H-VSe₂ as a promising ferromagnetic valleytronic material using a two-band $k \cdot p$ model together with first-principles calculations [10]. They predicted an intrinsic magnetic moment of about $1.01 \mu_B$ on the V-3d orbitals and an estimated Curie temperature of about 590 K, suggesting that 2H-VSe₂ could serve as a room-temperature ferrovalley material.

To understand the origin of the ferrovalley state, Tong *et al.* individually examined the roles of spin-orbit coupling (SOC) and ferromagnetism. When SOC is included without ferromagnetism, the system exhibits spin splitting of about 0.01 eV near one band edge and 0.08 eV near the other, as shown in Fig. 19 [10]. This indicates

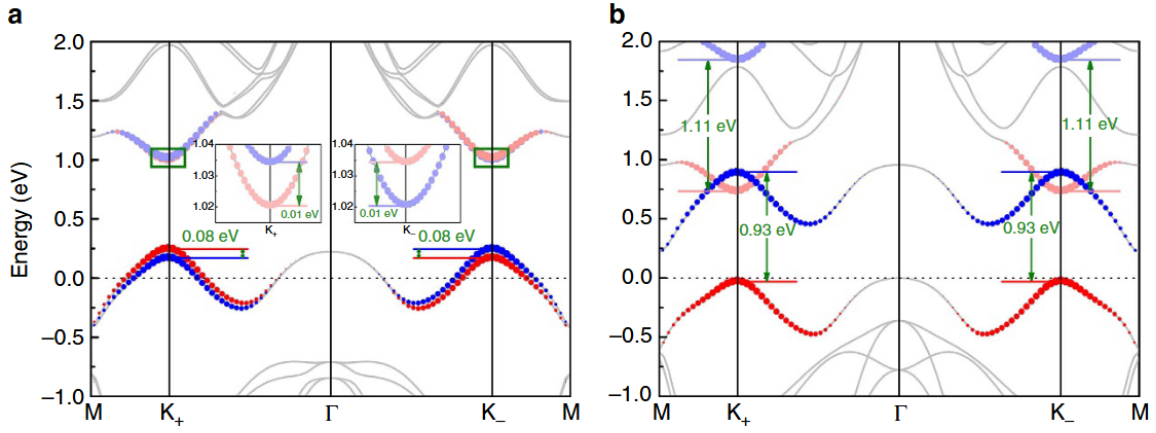


Figure 19. Band structure of monolayer 2H-VSe₂. (a) With SOC but without ferromagnetism. (b) With ferromagnetism but without SOC. Reproduced from Ref. [35] with permission of the authors; peer-reviewed version in Ref. [10].

that SOC alone modifies the spin-resolved band structure, but is insufficient to produce the full ferrovalley state.

By contrast, when ferromagnetism is included without SOC, the spin-up and spin-down bands become strongly separated, producing exchange splittings of about 0.93 eV and 1.11 eV, also shown in Fig. 19 [10]. These values are much larger than the SOC-only splittings, showing that intrinsic magnetism is the dominant $\mathcal{T}$ -symmetry-breaking ingredient. When both SOC and ferromagnetism are taken into account, the K and K' valleys become energetically inequivalent, and the direct band gaps at the two valleys differ by about 0.09 eV, indicating spontaneous valley polarization.

This valley asymmetry is further reflected in the Berry curvature. As shown in Fig. 20 [10], the Berry curvature retains opposite signs in the K and K' valleys, but its magnitude becomes unequal once spontaneous valley polarization appears. This imbalance underlies the anomalous valley Hall effect introduced prior. Tong *et al.* predicted a measurable transverse Hall voltage whose sign reverses when the valley polarization is reversed. On this basis, they further proposed a memory-device concept based on magnetic writing and electrical readout, shown in Fig. 21 [10].

Later in 2022, You *et al.* reported salt-assisted CVD growth of monolayer H-phase VSe₂ on mica [36]. Using aberration-corrected scanning transmission electron microscopy (STEM) and second-harmonic generation (SHG), they identified the broken-symmetry honeycomb structure characteristic of the H-phase, and transport measurements indicated apparent p-type behavior. The honeycomb crystal structure was directly observed by STEM, as suggested by the analysis of selected area electron diffraction [1]. Fig. 22 [36] shows the experimental preparation of H-phase monolayer VSe₂ and its corresponding transmission electron microscopy image. This work is important because it provides experimental progress toward realizing the H-phase VSe₂ TMD previously proposed as a ferrovalley candidate.

Furthermore, H-phase VS₂ and VTe₂ are structurally

similar to VSe₂, and have been likewise deemed ferrovalley materials [37, 38].

Shen *et al.* theoretically studied how spin orientation and strain influence the magnetic anisotropy, valley polarization, and magneto-optical Kerr effect in monolayer H-VS₂ [1, 38]. When the spin was aligned in the $+z$ direction, a valley splitting of 123 meV was determined. When the spin polarization, instead, was turned to the $-z$ direction, the valley splitting remained constant in magnitude but opposite in sign; therefore, the spin orientation can modulate the valley polarization.

Monolayer VTe₂ has a Curie temperature of 377 K and an enhanced valley splitting of 156.6 meV, making it, like VSe₂, favorable for room-temperature applications. Thus, the H-phase V-based TMDs, VS₂, VSe₂, and VTe₂, are reasonable candidates for valleytronic uses [1].

Beyond H-VSe₂, H-VS₂, and H-VTe₂, theoretical studies have proposed a broader family of H-MX₂ ferrovalley materials. These include NbX₂ ($X = S, Se$), FeCl₂, RuBr₂, OsBr₂, ScX₂ ($X = Br, Cl, I$), YX₂ ($X = I, Br, Cl$), LaX₂ ($X = H, I, Br$), GdX₂ ($X = F, Cl, Br, I$), LuX₂ ($X = Cl, Br, I$), and RI₂ ($R = Sc, Y, La-Lu$). These compounds share the same H-phase crystal structure and exhibit similar symmetry properties [1]. Adapted from Luo *et al.*, Table III summarizes key properties of representative ferrovalley TMDs, including magnetic moment, valley splitting, magnetic anisotropy, and Curie temperature [1]. Here, magnetic anisotropy describes the dependence of a material's magnetic behavior on the orientation of its magnetization relative to the crystal axes, while “Method” denotes the theoretical computational approach used to obtain the reported values, typically based on density functional theory (DFT).

Nearly all of the values in Table III come from DFT calculations; apart from the recent growth of H-phase VSe₂, few of these materials have been synthesized as monolayers, so experimental realization of 2D ferrovalley materials remains difficult. As of now, we therefore rely mainly on theoretical investigations to indicate what ferrovalley materials could offer in the field of valleytronics.

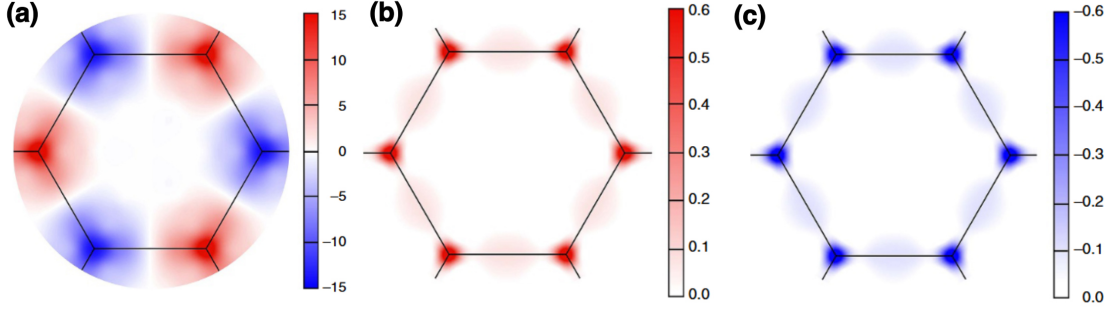


Figure 20. Berry-curvature distribution in ferrovalley monolayer 2H-VSe₂. (a) Contour map of the Berry curvature in k -space for the $d_{x^2-y^2}$ - and d_{xy} -dominated bands. (b) $\Omega(\mathbf{k}) + \Omega(-\mathbf{k})$ for positive valley polarization. (c) $\Omega(\mathbf{k}) + \Omega(-\mathbf{k})$ for negative valley polarization. The reversal from (b) to (c) reflects the reversal of valley polarization and the associated Berry-curvature imbalance. Reproduced from Ref. [35] with permission of the authors; peer-reviewed version in Ref. [10].

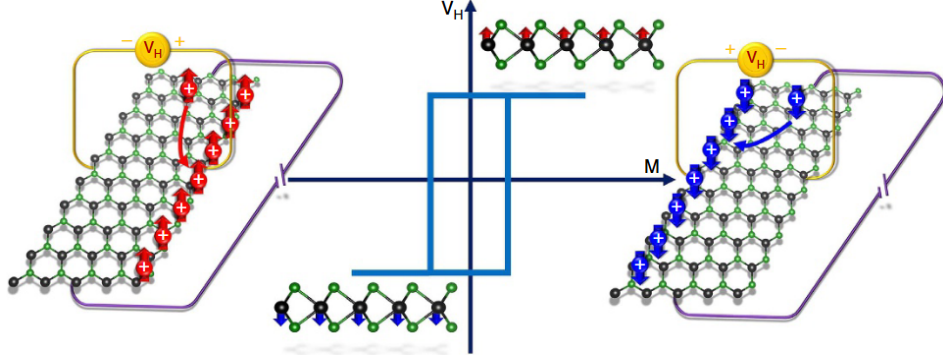


Figure 21. Schematic of data storage in hole-doped ferrovalley materials based on the anomalous valley Hall effect. The carriers denoted by white ‘+’ symbols are holes. Upward arrows in red and downward arrows in blue represent spin-up and spin-down carriers, respectively. Reproduced from Ref. [35] with permission of the authors; peer-reviewed version in Ref. [10].

IV. OUTLOOK

The field of valleytronics provides new possibilities for the development of emerging nanoelectronic devices. TMDs allow the valley degree of freedom to be accessed and controlled through electromagnetic radiation, electric fields, and magnetic fields.

The overarching goal of valleytronics is to enable new ways of encoding and processing information—functions traditionally handled by charge-based electronics. Valley polarization is the key enabling step: it gives rise to the valley Hall effect, generating a transverse voltage that can serve as an electrical readout of the valley state. As we have seen, the most accessible route to valley polarization between the K and K' valleys is optical, with σ^+ and σ^- circularly polarized light selectively exciting the K and K' states, respectively; the magnetic mechanisms surveyed here—and intrinsic ferrovalley materials in particular—offer complementary routes, though some have yet to be realized experimentally.

In intrinsically non-magnetic materials, valley polarization requires the K and K' states to occupy two different energy levels, breaking $\mathcal{T}$ -symmetry and providing a route to distinguishing the two inequivalent valleys. The valley index of these unequal valleys could represent “0” and

“1,” thereby integrating valleytronics into everyday electronic devices. To achieve this in non-magnetic materials, we have discussed three routes: external magnetic fields, magnetic proximity effects, and magnetic doping. External magnetic fields offer a readily available way to break valley degeneracy, but often provide only 0.1–0.2 meV/T of splitting [13, 17, 18]. Magnetic proximity effects utilizing magnetized heterostructures yield considerably larger valley splitting with promising tunability, though they are interface-sensitive and generally practical only at low temperatures [21, 23]. Combining an external field with a heterostructure has proven especially effective in amplifying and tuning the splitting [27]. Magnetic doping likewise produces significant exchange-driven splitting, but the impurities introduced through substitutional doping or adsorption can be disruptive [33, 34]. A common limitation across all three approaches is that the resulting energy imbalance is short-lived; thermal relaxation drives the system back to equilibrium, equally populating both valleys. Intrinsic ferrovalley materials circumvent this problem by exhibiting spontaneous valley polarization that maintains broken $\mathcal{T}$ -symmetry without external stimulus. Although such materials have been experimentally difficult to synthesize and identify, they remain a promising frontier for stable “0” and “1” encoding in valleytronic applications. The idea of encoding through

Table III. Magnetic moment, valley splitting, magnetic anisotropy, and Curie temperature reported for representative 2D ferrovalley materials. Data adapted from Ref. [1] (CC BY 4.0).

Ferrovalley materials	Method	Magnetic moment (μ_B)	Valley splitting (meV)	Magnetic anisotropy	Curie temperature (K)
VSe ₂ [10]	PBE	1	90	–	590 ^a
VSe ₂ [39]	HSE06	1	78	–	588
VSe ₂ [40]	HSE06	1	78	in-plane	590 ^a
VS ₂ [38]	HSE06	1	123	in-plane	292
NbX ₂ ($X = S, Se$) [41]	HSE06	1	156, 219	out-of-plane	–
MBr ₂ ($M = Ru, Os$) [42]	HSE06	4	354, 530	out-of-plane	–
FeCl ₂ [43]	HSE06	4	100	out-of-plane	96
RuBr ₂ [44]	PBE+ U	4	–	U -dependent	U -dependent
RuCl ₂ [45]	HSE06	4	240	out-of-plane	260
RuCl ₂ [46]	HSE06	4	223	out-of-plane	294
OsBr ₂ [47]	PBE+ U	1	–	U -dependent	U -dependent
LaBr ₂ [48]	HSE06	1	33	in-plane	–
GdX ₂ ($X = Cl, F$) [49]	PBE+ U	8	42, 47	out-of-plane	220
GdI ₂ [50]	HSE06	8	149	in-plane ^b	241
GdX ₂ ($X = Br, Cl$) [51]	PBE+ U	8	79, 35	in-plane	–
GdX ₂ ($X = F, Cl, Br$) [52]	HSE06	8	55, 38, 82	out-of-plane	300, 245, 225
CeI ₂ [53]	HSE06	2	208	in-plane	374
LaH ₂ [54]	PBE	1	166	out-of-plane	334
LaI ₂ , PrI ₂ [55]	PBE	1	28, 35	–	843, 77
LuX ₂ ($X = Cl, Br, I$) [56]	PBE	1	55, 93, 148	out-of-plane	89, 78, 124
RI ₂ ($R = Sc, Y, La-Lu$) [57]	SCAN meta-GGA	0–8	0–143	–	–
XI ₂ ($X = Sc, Y$) [58]	PBE+ U	1	97, 108	–	138, 58
ScX ₂ ($X = Br, Cl, I$) [59]	PBE+ U	1	50, 12, 93	in-plane	–
YI ₂ [60]	PBE+ U	1	105	–	–
YX ₂ ($X = I, Br, Cl$) [61]	HSE06	1	108, 57, 22	in-plane	220, 210, 200

^a Curie temperature quoted in the review table from Ref. [62].

^b Anisotropy as attributed in the review table to Ref. [63].

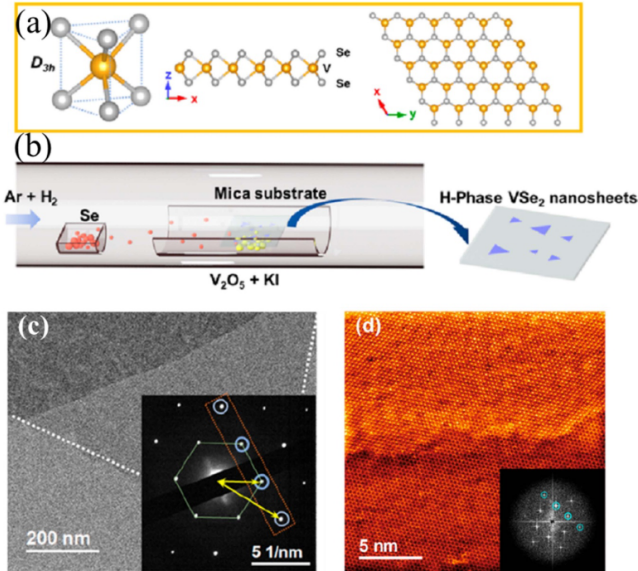


Figure 22. (a) Hexagonal crystal structure of monolayer H-phase VSe₂. (b) Schematic of the experimental preparation process for H-phase VSe₂ nanosheets. (c) Transmission electron microscopy (TEM) image of the synthesized VSe₂ nanosheets. (d) High-resolution transmission electron microscopy (HRTEM) image of monolayer VSe₂ [36]. Adapted from Ref. [1] under a CC BY 4.0 license.

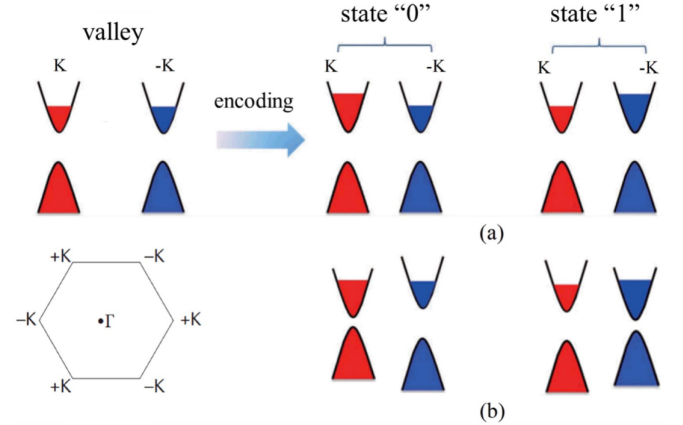


Figure 23. Schematic of encoding by valley polarization in (a) nonmagnetic materials and (b) magnetic valleytronic materials [64]. Adapted from Ref. [1] under a CC BY 4.0 license. Reproduced with permission of the authors of Ref. [64].

valley polarization is illustrated in Fig. 23.

Tong *et al.* proposed a schematic valleytronic device in which a transverse voltage generated by the valley Hall effect serves as the electrical signature of the valley state, as shown in Fig. 21 [10]. Although such devices remain largely conceptual, they highlight the broader technological promise of valley polarization for next-generation information processing.

STATEMENT ON THE USE OF AI

The original draft of this paper was written entirely by the author. Large language models—specifically Anthropic’s Claude (Claude Opus 4.7 and Claude Opus 5.5)—were used to help locate and track down relevant literature and to provide grammar, spelling, and copy-editing suggestions. Claude also helped write the Python code used to redraw Figs. 1, 4, 10, 17, and 18 from published structures, tight-binding parameters, and digitized data, and the author checked each redrawn figure against its source. The author reviewed every such suggestion and

made the final decision on all wording and content choices. No scientific claims, derivations, or interpretations were generated by AI; these remain entirely the author’s own.

ACKNOWLEDGEMENTS

I would like to thank Kathryn McGill for her guidance and for permission to reproduce Fig. 2 and Simon Rubinstein-Salzedo for directing Kaleidoscope Circles. I also thank K. F. Mak, E.-H. Yang, D. C. Ralph, T. Korn, X. Xu, X. Zuo, C.-G. Duan, X. Cui, and T. N. Narayanan for permission to reproduce figures from their work.

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- [1] C. Luo, Z. Huang, H. Qiao, X. Qi, and X. Peng, *Journal of Physics: Materials* **7**, 022006 (2024).
 - [2] G.-B. Liu, W.-Y. Shan, Y. Yao, W. Yao, and D. Xiao, *Physical Review B* **88**, 085433 (2013).
 - [3] K. F. Mak, K. L. McGill, J. Park, and P. L. McEuen, *Science* **344**, 1489 (2014).
 - [4] K. L. McGill, Two-dimensional materials: from Berry curvature to wrapping a microsphere, Ph.D. defense presentation, Cornell University (2018).
 - [5] K. F. Mak, K. He, J. Shan, and T. F. Heinz, *Nature Nanotechnology* **7**, 494 (2012).
 - [6] K. E. J. Goh, C. P. Y. Wong, and T. Wang, *Valleytronics in 2D Materials* (World Scientific, 2023).
 - [7] D. Xiao, Y. Yao, Z. Fang, and Q. Niu, *Physical Review Letters* **97**, 026603 (2006).
 - [8] P. Li, B. Liu, S. Chen, W.-X. Zhang, and Z.-X. Guo, *Chinese Physics B* **33**, 017505 (2024).
 - [9] K. F. Mak, K. L. McGill, J. Park, and P. L. McEuen, *The valley Hall effect in MoS₂ transistors* (2014), [arXiv:1403.5039 \[cond-mat.mes-hall\]](https://arxiv.org/abs/1403.5039).
 - [10] W.-Y. Tong, S.-J. Gong, X. Wan, and C.-G. Duan, *Nature Communications* **7**, 13612 (2016).
 - [11] S. Shrestha, M. Li, S. Park, X. Tong, D. DiMarzio, and M. Cotlet, *Nature Communications* **14**, 5234 (2023).
 - [12] S.-D. Guo, P. Li, and G. Wang, *Physical Review B* **111**, L140404 (2025).
 - [13] W. Liu, C. Luo, X. Tang, X. Peng, and J. Zhong, *AIP Advances* **9**, 045222 (2019).
 - [14] S.-D. Guo, G. Wang, and Y. S. Ang, *Applied Physics Letters* **123**, 171102 (2023).
 - [15] HandWiki, *Bloch wave*, HandWiki encyclopedia entry (2022).
 - [16] T. Zhou, L. Kou, Y. Ma, B. Wu, S.-D. Guo, X. Li, J. Zhang, L. Tang, and J. Feng, *Nano Letters* **15**, 5148 (2015).
 - [17] D. MacNeill, C. Heikes, K. F. Mak, Z. Anderson, A. Kormányos, V. Zólyomi, J. Park, and D. C. Ralph, *Physical Review Letters* **114**, 037401 (2015).
 - [18] A. Srivastava, M. Sidler, A. V. Allain, D. S. Lembke, A. Kis, and A. Imamoglu, *Nature Physics* **11**, 141 (2015).
 - [19] G. Aivazian, Z. Gong, A. M. Jones, R.-L. Chu, J. Yan, D. G. Mandrus, C. Zhang, D. Cobden, W. Yao, and X. Xu, *Nature Physics* **11**, 148 (2015).
 - [20] R. Luo, W. W. Xu, Y. Zhang, Z. Wang, X. Wang, Y. Gao, P. Liu, and M. Chen, *Nature Communications* **11**, 1011 (2020).
 - [21] J. Qi, X. Li, Q. Niu, and J. Feng, *Physical Review B* **92**, 121403 (2015).
 - [22] P. A. Lee, Lecture 22: The Kondo problem: Singlet ground state and kondo resonance, MIT Physics lecture notes (2004), [dSpace@MIT PDF](https://dspace.mit.edu/handle/1721.1/11100).
 - [23] D. Zhong, K. L. Seyler, X. Linpeng, R. Cheng, N. Sivadas, B. Huang, E. Schmidgall, T. Taniguchi, K. Watanabe, M. A. McGuire, W. Yao, D. Xiao, K.-M. C. Fu, and X. Xu, *Science Advances* **3**, e1603113 (2017).
 - [24] M. Ge, H. Wang, J. Wu, C. Si, J. Zhang, and S. Zhang, *npj Computational Materials* **8**, 32 (2022).
 - [25] M. Fang and E.-H. Yang, *Materials* **16**, 3701 (2023).
 - [26] D. MacNeill, C. Heikes, K. F. Mak, Z. Anderson, A. Kormányos, V. Zólyomi, J. Park, and D. C. Ralph, *Breaking of valley degeneracy by magnetic field in monolayer MoSe₂* (2014), [arXiv:1407.0686 \[cond-mat.mes-hall\]](https://arxiv.org/abs/1407.0686).
 - [27] P. Nagler, M. V. Ballottin, A. A. Mitoglu, F. Mooshammer, N. Paradiso, C. Strunk, R. Huber, A. Chernikov, P. C. M. Christianen, C. Schüller, and T. Korn, *Nature Communications* **8**, 1551 (2017).
 - [28] P. Nagler, M. V. Ballottin, A. A. Mitoglu, F. Mooshammer, N. Paradiso, C. Strunk, R. Huber, A. Chernikov, P. C. M. Christianen, C. Schüller, and T. Korn, *Giant Zeeman splitting inducing near-unity valley polarization in van der Waals heterostructures* (2017), [arXiv:1704.02208 \[cond-mat.mes-hall\]](https://arxiv.org/abs/1704.02208).
 - [29] W. Zhang, J. Wang, T. Zhang, B. Shao, and X. Zuo, *Journal of Materiomics* **11**, 100986 (2025).
 - [30] C. Zhao, T. Norden, P. Zhang, P. Zhao, Y. Cheng, F. Sun, J. P. Parry, P. Taheri, J. Wang, Y. Yang, T. Scrace, K. Kang, S. Yang, G.-X. Miao, R. Sabirianov, G. Kiioseoglou, W. Huang, A. Petrou, and H. Zeng, *Nature Nanotechnology* **12**, 757 (2017).
 - [31] D. Zhong, K. L. Seyler, X. Linpeng, R. Cheng, N. Sivadas, B. Huang, E. Schmidgall, T. Taniguchi, K. Watanabe, M. A. McGuire, W. Yao, D. Xiao, K.-M. C. Fu, and X. Xu, *Van der Waals engineering of ferromagnetic semiconductor heterostructures for spin and valleytronics* (2017), [arXiv:1704.00841 \[cond-mat.mes-hall\]](https://arxiv.org/abs/1704.00841).
 - [32] B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero, and X. Xu, *Nature* **546**, 270 (2017).

- [33] X. Chen, L. Zhong, X. Li, and J. Qi, [Valley splitting in the transition-metal dichalcogenides monolayer via atoms adsorption](#) (2016), [arXiv:1609.00880 \[cond-mat.mtrl-sci\]](#).
- [34] K. R. Sahoo, J. J. Panda, S. Bawari, R. Sharma, D. Maity, A. Lal, R. Arenal, G. Rajalaksmi, and T. N. Narayanan, [Physical Review Materials](#) **6**, 085202 (2022).
- [35] W.-Y. Tong, S.-J. Gong, X. Wan, and C.-G. Duan, [Concepts of ferrovalley material and anomalous valley Hall effect](#) (2016), [arXiv:1604.05833 \[cond-mat.mtrl-sci\]](#).
- [36] J. You, J. Pan, S.-L. Shang, X. Xu, Z. Liu, J. Li, H. Liu, T. Kang, M. Xu, S. Li, D. Kong, W. Wang, Z. Gao, X. Zhou, T. Zhai, Z.-K. Liu, J.-K. Kim, and Z. Luo, [Nano Letters](#) **22**, 10167 (2022).
- [37] C. Wang and Y. An, [Applied Surface Science](#) **538**, 148098 (2021).
- [38] C. Shen, G. Wang, T. Wang, C. Xia, and J. Li, [Applied Physics Letters](#) **117**, 042406 (2020).
- [39] J. Liu, W. Hou, C. Cheng, H. Fu, J. Sun, and S. Meng, [Journal of Physics: Condensed Matter](#) **29**, 255501 (2017).
- [40] S. Feng and W. Mi, [Applied Surface Science](#) **458**, 191 (2018).
- [41] Y. Zang, Y. Ma, R. Peng, H. Wang, B. Huang, and Y. Dai, [Nano Research](#) **14**, 834 (2021).
- [42] H. Huan, Y. Xue, B. Zhao, G. Gao, H. Bao, and Z. Yang, [Physical Review B](#) **104**, 165427 (2021).
- [43] P. Zhao, Y. Dai, H. Wang, B. Huang, and Y. Ma, [ChemPhysMater](#) **1**, 56 (2022).
- [44] S.-D. Guo, W.-Q. Mu, and B.-G. Liu, [2D Materials](#) **9**, 035011 (2022).
- [45] K. Sheng, B.-K. Zhang, H.-K. Yuan, and Z.-Y. Wang, [Physical Review B](#) **105**, 195312 (2022).
- [46] R. Li, N. Mao, X. Wu, B. Huang, Y. Dai, and C. Niu, [Nano Letters](#) **23**, 91 (2023).
- [47] S.-D. Guo, Y.-L. Tao, and B.-G. Liu, [Frontiers of Physics](#) **18**, 33304 (2023).
- [48] P. Zhao, Y. Ma, C. Lei, H. Wang, B. Huang, and Y. Dai, [Applied Physics Letters](#) **115**, 261605 (2019).
- [49] S.-D. Guo, J.-X. Zhu, W.-Q. Mu, and B.-G. Liu, [Physical Review B](#) **104**, 224428 (2021).
- [50] H. Cheng, J. Zhou, W. Ji, Y.-N. Zhang, and Y. P. Feng, [Physical Review B](#) **103**, 125121 (2021).
- [51] F. Ding, S. L. Ji, S. S. Li, L. Wang, H. Wu, Z. Hu, F. Li, and Y. Pu, [physica status solidi \(b\)](#) **258**, 2100356 (2021).
- [52] K. Sheng, H.-K. Yuan, and Z.-Y. Wang, [Physical Chemistry Chemical Physics](#) **24**, 3865 (2022).
- [53] K. Sheng, Q. Chen, H.-K. Yuan, and Z.-Y. Wang, [Physical Review B](#) **105**, 075304 (2022).
- [54] Y. Shi, N. Jia, J. Cai, Z. Lyu, and Z. Liu, [Journal of Physics: Condensed Matter](#) **34**, 475303 (2022).
- [55] A. Sharan and N. Singh, [Advanced Theory and Simulations](#) **5**, 2100476 (2022).
- [56] C.-S. Hu, Y.-J. Wu, Y.-S. Liu, S. Fu, X.-N. Cui, Y.-H. Wang, and C.-W. Zhang, [Chinese Physics B](#) **32**, 037306 (2023).
- [57] A. Sharan, S. Lany, and N. Singh, [2D Materials](#) **10**, 015021 (2023).
- [58] M. Lan, Y. Li, S. Qiao, T. Huang, S. Ma, F. Ding, H. Wu, F. Li, and Y. Pu, [Journal of Physics D: Applied Physics](#) **56**, 385103 (2023).
- [59] Y. Wu, J. Tong, L. Deng, F. Luo, F. Tian, G. Qin, and X. Zhang, [Acta Materialia](#) **246**, 118731 (2023).
- [60] Y. Wu, J. Tong, L. Deng, F. Luo, F. Tian, G. Qin, and X. Zhang, [Nano Letters](#) **23**, 6226 (2023).
- [61] B. Huang, W.-Y. Liu, X.-C. Wu, S.-Z. Li, H. Li, Z. Yang, and W.-B. Zhang, [Physical Review B](#) **107**, 045423 (2023).
- [62] H. Pan, [The Journal of Physical Chemistry C](#) **118**, 13248 (2014).
- [63] W. Liu, J. Tong, L. Deng, B. Yang, G. Xie, G. Qin, F. Tian, and X. Zhang, [Materials Today Physics](#) **21**, 100514 (2021).
- [64] H.-L. Zeng and X.-D. Cui, [Physics \(Wuli\)](#) **45**, 505 (2016).